The molecular structure of thiazyl chloride has been studied by electron diffraction at a nozzle temperature of 368°K and was found to be NSCl and not SNCl. The results for the interatomic distances, root-mean-square amplitudes, and the bond angle, respectively, are:

\[ r_{\text{N-S}} = 1.448 \pm 0.003 \text{ Å}, \quad r_{\text{S-Cl}} = 2.159 \pm 0.003 \text{ Å}, \quad r_{\text{N...Cl}} = 3.106 \pm 0.016 \text{ Å}, \quad \rho_{\text{N-S}} = 0.037 \pm 0.002 \text{ Å}, \quad \rho_{\text{S-Cl}} = 0.052 \pm 0.003 \text{ Å}, \quad \rho_{\text{N...Cl}} = 0.076 \pm 0.012 \text{ Å}, \text{ and } \angle_{\text{NSCl}} = 117.5^{\circ} \pm 1.0^{\circ}. \]

These values were determined from least-squares refinements of intensity curves. The estimated associated errors include standard errors derived from the least-squares procedure, estimates of correlation among the data, uncertainties in the experimental constants, and uncertainties in the factors employed in data reduction.

The infrared spectrum of gaseous NSCl has been studied from
The three fundamental vibrations, $\nu_1$ (NS stretch), $\nu_2$ (SCl stretch), and $\nu_3$ (bending) were directly observed at 1324.0, 417.8, and 271 cm$^{-1}$ respectively. Nitrogen and chlorine isotopes were used to verify the assignments.

The infrared spectrum of solid films of NSCl on various substrates cooled with liquid nitrogen has been studied from 95 to 4,000 cm$^{-1}$. The investigation showed the existence of two forms of the solid NSCl films: one form with main peaks at 1397.0, 205, and 140 cm$^{-1}$; and another form with main peaks at 1346, 350, and 256 cm$^{-1}$.

Using the fundamental frequencies, a set of force constants of a simple quadratic potential function, based on the symmetry co-ordinates $\Delta r_{NS}$, $\Delta r_{SCl}$, and $(r_{NS}r_{SCl})^{1/2}\Delta \alpha$, has been calculated. The values in millidynes per angstrom are $F_{NS} = 10.01$, $F_{SCl} = 1.58$, and $F_\alpha = 0.25$. Using these values, an equivalent set of force constants based on the symmetry co-ordinates $\Delta r_{NS}$, $\Delta r_{SCl}$, and $\Delta r_{NCl}$ has been calculated. The values in millidynes per angstrom for the three main force constants are $F'_{NS} = 10.62$, $F'_{SCl} = 2.40$, and $F'_{NCl} = 0.99$.

Using the mean-square-amplitudes of vibration obtained from the electron diffraction investigation together with the fundamental frequencies, a set of force constants of a simple quadratic potential function has been calculated. The values in millidynes per angstrom are $F''_{NS} = 9.33$, $F''_{SCl} = 2.22$, and $F''_{NCl} = 0.95$, which agree well
with those based on infrared data.

Using the force constants based on infrared data, root-mean-square amplitudes of vibration have been calculated. The values in angstroms are $l_{\text{NS}} = 0.0354$, $l_{\text{S-Cl}} = 0.0506$, and $l_{\text{N...Cl}} = 0.0749$, which agree well with those obtained from the electron diffraction investigation.
An Investigation of the Molecular Structure and Vibrational Potential Function of Thiazyl Chloride (NSCl) by Electron Diffraction and Infrared Spectroscopy

by

Walter Charles Emken

A THESIS

submitted to

Oregon State University

in partial fulfillment of the requirements for the degree of

Doctor of Philosophy

June 1971
This thesis is dedicated

To Monty
ACKNOWLEDGMENTS

I would like to thank Professors Kenneth W. Hedberg and John C. Decius for their supervision of this investigation.

In addition, I would like to thank the many graduate students, especially Mr. Frank Meserole, for their helpful discussions.

I would also like to thank Professor Kenneth Russell for his initial idea and encouragement of this investigation.

Finally, I am grateful to the National Aeronautics and Space Administration for financial support of this work (1964-67) in the form of a Graduate Fellowship.
# TABLE OF CONTENTS

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>INTRODUCTION</td>
<td>1</td>
</tr>
<tr>
<td>General</td>
<td>1</td>
</tr>
<tr>
<td>Thiazyl Fluoride</td>
<td>2</td>
</tr>
<tr>
<td>Thiazyl Chloride</td>
<td>6</td>
</tr>
<tr>
<td>SAMPLE PREPARATION</td>
<td>9</td>
</tr>
<tr>
<td>Introduction</td>
<td>9</td>
</tr>
<tr>
<td>Preparation of ((\text{NSCl})_3)</td>
<td>12</td>
</tr>
<tr>
<td>Preparation of (\text{^{15}N Enriched (NSCl)}_3)</td>
<td>17</td>
</tr>
<tr>
<td>ELECTRON DIFFRACTION</td>
<td>20</td>
</tr>
<tr>
<td>Introduction</td>
<td>20</td>
</tr>
<tr>
<td>Experimental and Data Reduction</td>
<td>26</td>
</tr>
<tr>
<td>Structure Determination, Refinement, Final Results</td>
<td>28</td>
</tr>
<tr>
<td>INFRARED SPECTROSCOPY</td>
<td>33</td>
</tr>
<tr>
<td>Introduction</td>
<td>33</td>
</tr>
<tr>
<td>Gas Phase</td>
<td>34</td>
</tr>
<tr>
<td>Solid Phase</td>
<td>38</td>
</tr>
<tr>
<td>FORCE CONSTANTS AND AMPLITUDES OF VIBRATION</td>
<td>45</td>
</tr>
<tr>
<td>Introduction</td>
<td>45</td>
</tr>
<tr>
<td>Force Constants Based on Infrared Data</td>
<td>46</td>
</tr>
<tr>
<td>Force Constants Based on Infrared and Electron Diffraction Data</td>
<td>53</td>
</tr>
<tr>
<td>Amplitudes of Vibration Based on Infrared Data</td>
<td>55</td>
</tr>
<tr>
<td>BIBLIOGRAPHY</td>
<td>89</td>
</tr>
<tr>
<td>Figure</td>
<td>Description</td>
</tr>
<tr>
<td>--------</td>
<td>------------------------------------------------------------------------------</td>
</tr>
<tr>
<td>1.</td>
<td>Experimental intensity curves for NSCl.</td>
</tr>
<tr>
<td>2.</td>
<td>Intensity curves for NSCl.</td>
</tr>
<tr>
<td>3.</td>
<td>Radial distribution curves for NSCl.</td>
</tr>
<tr>
<td>4.</td>
<td>Infrared spectrum of gaseous NSCl: $\nu_1$ (N-S stretching vibration) region of the spectrum.</td>
</tr>
<tr>
<td>5.</td>
<td>Infrared spectrum of gaseous NSCl: $\nu_2$ (S-Cl stretching vibration) region of the spectrum.</td>
</tr>
<tr>
<td>6.</td>
<td>Infrared spectrum of gaseous NSCl: $\nu_3$ (NSCl bending vibration) region of the spectrum.</td>
</tr>
<tr>
<td>7.</td>
<td>Infrared spectrum of a solid film of NSCl on KBr.</td>
</tr>
<tr>
<td>8.</td>
<td>Infrared spectrum of solid films of NSCl on CsI and polyethylene.</td>
</tr>
</tbody>
</table>
## LIST OF TABLES

<table>
<thead>
<tr>
<th>Table</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Molecular structure, fundamental frequencies and amplitudes of vibration for NSF.</td>
<td>57</td>
</tr>
<tr>
<td>2. Force constants of NSF.</td>
<td>57</td>
</tr>
<tr>
<td>3. Observed fundamental frequencies of NSCl.</td>
<td>58</td>
</tr>
<tr>
<td>4. Force constants and amplitudes of vibration for NSCl.</td>
<td>58</td>
</tr>
<tr>
<td>5. Data for electron diffraction photographs of NSCl.</td>
<td>59</td>
</tr>
<tr>
<td>6. Experimental intensity curve 1-231-02 for NSCl.</td>
<td>60</td>
</tr>
<tr>
<td>7. Experimental intensity curve 1-231-03 for NSCl.</td>
<td>60</td>
</tr>
<tr>
<td>8. Experimental intensity curve 1-230-09 for NSCl.</td>
<td>61</td>
</tr>
<tr>
<td>9. Experimental intensity curve 1-230-10 for NSCl.</td>
<td>62</td>
</tr>
<tr>
<td>10. Experimental intensity curve 1-233-06 for NSCl.</td>
<td>63</td>
</tr>
<tr>
<td>11. Experimental intensity curve 1-233-07 for NSCl.</td>
<td>64</td>
</tr>
<tr>
<td>12. Experimental intensity curve 1-233-08 for NSCl.</td>
<td>65</td>
</tr>
<tr>
<td>13. Composite experimental intensity curve for NSCl.</td>
<td>66</td>
</tr>
<tr>
<td>14. Experimental radial distribution curve for NSCl.</td>
<td>68</td>
</tr>
<tr>
<td>15. Theoretical intensity curve for NSCl.</td>
<td>70</td>
</tr>
<tr>
<td>16. Theoretical radial distribution curve for NSCl.</td>
<td>72</td>
</tr>
<tr>
<td>17. Comparison of weight factor ratios for the two possible models for NSCl and the observed peak areas.</td>
<td>74</td>
</tr>
<tr>
<td>18. Results of the least square refinements on NSCl.</td>
<td>74</td>
</tr>
<tr>
<td>19. Final parameter values for NSCl.</td>
<td>75</td>
</tr>
<tr>
<td>Table</td>
<td>Page</td>
</tr>
<tr>
<td>---------------</td>
<td>------</td>
</tr>
<tr>
<td>20. The symmetric error matrix for NSCl.</td>
<td>76</td>
</tr>
<tr>
<td>21. Observed vibrational frequencies for gaseous NSCl.</td>
<td>77</td>
</tr>
<tr>
<td>22. Observed frequencies for solid films of NSCl on KBr.</td>
<td>77</td>
</tr>
<tr>
<td>23. Observed frequencies for solid films of NSCl on CsI and on polyethylene.</td>
<td>78</td>
</tr>
<tr>
<td>24. List of constants used in calculations for NSCl.</td>
<td>79</td>
</tr>
<tr>
<td>25. Force constants of NSCl.</td>
<td>80</td>
</tr>
<tr>
<td>26. Amplitudes of vibration for NSCl.</td>
<td>80</td>
</tr>
</tbody>
</table>
AN INVESTIGATION OF THE MOLECULAR STRUCTURE AND VIBRATIONAL POTENTIAL FUNCTION OF THIAZYL CHLORIDE (NSCI) BY ELECTRON DIFFRACTION AND INFRARED SPECTROSCOPY

INTRODUCTION

General

The investigation of NSCI by electron diffraction and infrared spectroscopy was undertaken for several reasons. Most interesting, perhaps, is the geometry of the molecule, which at the beginning of this work was quite unknown, even to the arrangement of the atoms in the chain. The electron diffraction experiments could be expected to settle these questions, and in addition to provide amplitudes of molecular vibration which could be used in conjunction with the fundamental frequencies obtained from infrared work to help find the constants of the vibrational potential function. The solid state infrared spectrum of NSCI was of some interest in itself and it was decided to investigate it along with the gas phase work.

The early work done on NSCI was greatly influenced by the work that had been done and was being done on thiazyl fluoride, NSF. The conclusions concerning the geometry and related items of NSCI were based, to a large extent, on previous work done on NSF. This is to say that the history of the NSCI problem lies in the history of the NSF problem. Because of this close relationship between the two
compounds, the discussion of the early work done on NSCl will be preceded by a discussion of the work done on NSF.

**Thiazyl Fluoride**

A bent triatomic molecule composed of an atom each of nitrogen, sulfur and a halogen, has two possible isomers, SNX and NSX (X = halogen). A third isomer with the halogen in the center is chemically unlikely. The early work on thiazyl fluoride indicated the possible existence of the first two isomers. In 1955, Glemser, Schröder, and Haeseler reported the preparation of a mixture of SNF and SN₂F₂ by reacting AgF₂ with S₄N₄, but they were unable to obtain pure SNF (25). The preparation of the other isomer, NSF, was reported in 1956 by Glemser and Haeseler as resulting from the thermal decomposition of SN₂F₂ according to the following (20):

\[
2 \text{SN}_2\text{F}_2 \rightarrow 2 \text{SN} + \text{N}_2 + 2 \text{F}_2
\]

\[
2 \text{SN} + \text{F}_2 \rightarrow 2 \text{NSF}
\]

NSF was described as being quite stable with a boiling point of 4.8°C and a melting point of -79°C, and was compared with the isoelectronic compound, SO₂. They gave the following order of decreasing thermodynamic stability: NSF, NSF₃, SN₂F₂, and SNF. However, later work showed that NSF was quite unstable compared to NSF₃ (6, 19, 21, 23, 27, p. 1-30, 34, p. 227-237). Their hydrolysis experiment on NSF was interpreted as favoring the structure \( \text{N} \equiv \text{S-F} \).
In 1960, Gallup and Koenig reported the infrared spectra of \( \text{SN}_2\text{F}_2 \) and SNF (17). They prepared their sample of \( \text{SN}_2\text{F}_2 \) by the method of Glemser, Schröder and Haeseler (25). Along with the \( \text{SN}_2\text{F}_2 \), a compound with the formula SNF was formed which was considerably less stable than \( \text{SN}_2\text{F}_2 \) and decomposed over a period of a few hours. It was found difficult to remove the SNF from the \( \text{SN}_2\text{F}_2 \). Spectra were taken from both a freshly prepared sample of \( \text{SN}_2\text{F}_2 \) and from one 15 hours old. The freshly prepared sample showed three bands which had virtually disappeared at the end of 15 hours, which made easy the identification of the bands of \( \text{SN}_2\text{F}_2 \) and those of SNF. Frequency assignments for the sample, \( \text{SN}_2\text{F}_2 \) plus SNF, were based on a planar structure with \( C_{2v} \) symmetry for the \( \text{SN}_2\text{F}_2 \), a bent structure with \( C_5 \) symmetry for SNF, and the observed decrease in intensity with time of three of the bands in the spectrum. The three bands assigned to SNF were found at 1024, 941, and 391 \( \text{cm}^{-1} \).

In 1961, Glemser and Richert reported the isolation of SNF and gave 0.4 ± 2°C as its boiling point and -89°C as its melting point (23). It was described as an unstable, colorless and pungent gas. The SNF, along with NSF\(_3\), was prepared by reacting \( S_4\text{N}_4 \) with AgF\(_2\) in CCl\(_4\). Also, SNF was prepared by these authors by the thermal decomposition of \( S_4\text{N}_4\text{F}_4 \) and was described as being very unstable compared to NSF\(_3\). They obtained the infrared spectrum and the fluorine nuclear magnetic resonance spectrum of SNF and interpreted
their results in favor of the S=N-F structure (46). They gave the following frequencies (quite different from those of Gallup and Koenig) and force constants for their SNF model: $\nu_1 (SN) = 1372$, $\nu_2 (NF) = 640$, and $\nu_3 (\text{bend}) = 366 \text{ cm}^{-1}$; $f_{SN} = 10.4$ and $f_{NF} = 1.5$ millidyynes per angstrom. The method of sample preparation was the same as described earlier for $\text{SN}_2\text{F}_2$ by Glemser, Schröder and Haeseler (25), but they were unable to repeat the preparation of $\text{SN}_2\text{F}_2$ and were led to the conclusion that this compound did not exist. Also, since NSF was reported as arising from the thermal decomposition of $\text{SN}_2\text{F}_2$, they concluded that its existence was also in some doubt.

In 1961, Rogowski determined the structure of SNF by means of electron diffraction (48). He apparently worked with a sample supplied by Glemser and Richert (24). After considering over 100 theoretically feasible models (including both possible isomers, NSF and SNF), he too concluded that the molecule had a bent SNF structure with the following molecular parameters: $d_{SN} = 1.62 \pm 0.03 \text{ Å}$, $d_{NF} = 1.42 \pm 0.03 \text{ Å}$ and $\angle \text{SNF} = 122 \pm 3^\circ$.

In 1963, Kirchhoff and Wilson reported that they had investigated the microwave spectrum of the compound reported to be SNF and found, contrary to the electron diffraction report, that it had the bent structure $N=\hat{S}-F$ (37, 63). They pointed out that, if one uses Badger's rule (1) to estimate bond distances from the force constants that Glemser and Richert (46) had given for the SN and NF bonds in their
interpretation of the infrared spectrum of SNF based on the S=N-F model, the following bond distances are obtained: $d_{SN} = 1.45 \text{Å}$ and $u_{NF} = 1.63 \text{Å}$. The electron diffraction and infrared work thus appear not to be compatible. They also pointed out that Glemser and Richert had reported the preparation of SNF as also arising from the thermal decomposition of $S_4N_4F_4$ where the fluorine atoms were known to be bonded to the sulfur atoms (46, 60, 62). NSF and not SNF would be expected to result from such a decomposition unless some kind of molecular rearrangement occurs.

Kirchhoff and Wilson's values for the bond distances and bond angle of NSF are as follows: $d_{NS} = 1.446 \pm 0.01 \text{Å}$, $d_{SF} = 1.646 \pm 0.01 \text{Å}$, and $\angle NSF = 116^\circ 52' \pm 15'$. Their results are based upon the microwave spectra of the species $N^{32}SF$ and $N^{34}SF$, and agree well with distances calculated from Badger's rule using values of N-S and S-F stretching force constants obtained by Glemser and Richert from their infrared work using an NSF model (46). Because Kirchhoff and Wilson took care to prepare their sample according to the same procedure used by Glemser and Richert and, further, to repeat with similar results some of these authors' experiments on the chemistry of the compound, there seems no doubt that both groups were dealing with the same substance. Therefore, Kirchhoff and Wilson have shown that the compound claimed earlier to be SNF was in fact NSF, a bent triatomic molecule with sulfur in the center, and that the previous
infrared work by Glemser and Richert was actually compatible with this structure, whereas the electron diffraction work was apparently in error.

More recent work on NSF has resulted in a refinement of its structure by further investigation of its microwave spectrum (12), additional force constant calculations based on infrared and microwave data (12, 21, 43, 50), and an evaluation of its mean amplitudes of vibration by the Cyvin method (45). This information, along with the earlier results, is given in Tables 1 and 2.

Thiazyl Chloride

Early reports of research on thiazyl chloride contain mention of two isomers corresponding in type to those discussed above for the fluorine compound, but the existence of the form SNC1 is as yet unproven. In 1931 and 1932, Meuwsen mentioned the "yet unknown monomer," NSC1, as being connected with the preparation of tri-thiazyltrichloride, (NSCl)3 (41, 42). In 1957 and 1958, Becke-Goehring also mentioned NSCl as an unstable intermediate in one of the preparations of (NSCl)3 (5, p. 128-133, 8). However, neither of these authors isolated or characterized the compound.

Thiazyl chloride was first prepared by Glemser and Richert in 1961 by reacting chlorine gas with thiazyl fluoride, and was reported to have the structure SNC1 (23). The infrared spectrum of the gaseous
compound showed, in the NaCl region, only one band at 1322 cm\(^{-1}\) which was interpreted as the S-N stretching vibration. The N-Cl stretching vibration was presumed to lie below 460 cm\(^{-1}\). Since the infrared spectrum of this compound was very similar to that exhibited by what was thought to be at that time SNF (23, 24, 46, 48), Glemser and Richert concluded that the two gases must have similar structures. Also in 1961, Glemser and Perl reported the preparation of gaseous SNCl by heating the trimer, (NSCl)\(_3\), to 110°C in a high vacuum (22). They too reported that the infrared spectrum of this gaseous compound was similar to that of SNF and concluded that they had successfully established the existence of SNCl. It would seem more likely, however, that the isomer NSCl, and not SNCl, would have resulted from thermal decomposition of (NSCl)\(_3\), unless some kind of molecular rearrangement occurs, since at that time chemical evidence suggested that the chlorine atoms were attached to the sulfur (5, p. 63-68, 8, 42, 51). Later X-ray analysis of (NSCl)\(_3\) showed this certainly to be the case (59, 61).

After Kirchhoff and Wilson's microwave work on thiazyl fluoride (37, 63), which showed that the compound thought to be SNF was actually NSF, the situation concerning thiazyl chloride was also cleared up. The work on thiazyl chloride by Glemser and Richert and that by Glemser and Perl just described could now be interpreted as favoring the structure NSCl instead of SNCl using arguments based on
similarity of the infrared spectra (6, 19, 21, 27, p. 3-6). The preparation of NSCl from \((\text{NSCl}_3)\) was also put into the proper perspective from the structural point of view.

Between 1963 and 1966 several new methods of preparing NSCl were reported. These new methods, along with those briefly mentioned above, will be discussed in more detail in the sample preparation section that follows.

Towards the end of the investigation of NSCl described in this thesis, Müller reported the infrared spectrum, structure, force constants, mean amplitudes of vibration, thermodynamic functions and molecular polarizability of NSCl (44). The observed fundamental frequencies and their assignments are given in Table 3.

Müller calculated the force constants and mean amplitudes of vibration at room temperature for NSCl based upon the following assumed values of the molecular parameters: \(r_{\text{NS}} = 1.45 \text{ Å},\)
\(r_{\text{SCl}} = 2.00 \text{ Å}\) and angle NSCl = 116°. The results of these calculations are given in Table 4. On the basis of these results, especially the very high value of the force constant \(f_{\text{NS}} = 10.02\) millidyne per angstrom, the conclusion was that thiazyl chloride must have the NSCl, and not the SNCI, structure.
SAMPLE PREPARATION

Introduction

Thiazyl chloride, NSCl, has been described as a gaseous, pale yellow or green-yellow, unstable compound which rapidly trimerizes to form (NSCl)₃ at room temperature and which is very water reactive (19, 22, 27, p. 6). Ten preparations of NSCl have been described in the literature and are outlined in what follows.

Meuwsen has reported the preparation of (NSCl)₃ by passing chlorine gas through a carbon tetrachloride suspension of S₃N₂Cl₂. He postulated the formation of the intermediate, NSCl, according to the equation

\[(\text{NS})₂\text{SCl}_2 + \text{Cl}_2 \rightarrow \text{NSCl} + \text{NS} \cdot \text{SCl}_3\]

which then polymerized to the well known (NSCl)₃; however, he was unable to isolate the NS·SCl₃, which he thought probably decomposed into NSCl and SCl₂ (41).

Becke-Goehring also reported NSCl as an intermediate in a reaction of HOSN with thionylchloride in carbon tetrachloride that produced (NSCl)₃ as follows (5, p. 128-133, 8):

\[\text{HOSN} + \text{OSCl}_2 \rightarrow \text{ClSN} + \text{SO}_2 + \text{HCl}\]

\[3 \text{ClSN} \rightarrow (\text{NSCl})_3\]

She described the NSCl as being unstable due to a (postulated) negative
charge on the nitrogen and positive charge on the sulfur atom which
disappear upon polymerization (5, p. 133).

Glemser and Richert's preparation of NSCl has been mentioned (23). Later, Glemser and Perl prepared NSCl by heating (NSCl)$_3$ in a
high vacuum (22).

\[
\begin{align*}
110^\circ\text{C/vacuum} & \\
\text{(NSCl)}_3 & \rightarrow \text{3 NSCl.} \\
\text{room temp.} & \end{align*}
\]

In both cases the NSCl was characterized by its infrared spectrum (23).

In 1963, Maguire, Smith and Jolly reported four new methods
for the preparation of NSCl (40):

1. NSCl was obtained, along with $S_2Cl_2$, $SCl_2$ and $S_3N_2Cl_2$, when
$S_2Cl_2$ vapor was passed into a stream of active nitrogen and was
probably formed according to the reaction

\[2N + S_2Cl_2 \rightarrow 2\text{NSCl}.
\]

2. NSCl was also obtained when ammonium chloride was
refluxed in an excess amount of $S_2Cl_2$ for an extended period. The
probable reaction was

\[NH_4Cl + 2S_2Cl_2 \rightarrow 3S + \text{NSCl} + 4\text{HCl}.
\]

3. The compound was also formed when $S_3N_2Cl_2$ was heated to
80-100\(^\circ\)C in a vacuum:

\[3S_3N_2Cl_2 \rightarrow 80-100^\circ\text{C} \rightarrow 2S_3N_2Cl + 2\text{NSCl} + \text{SCl}_2.
\]
4. NSCl was not only formed by the above pyrolysis of $S_3N_2Cl_2$ but also by the reaction of chlorine gas with $S_3N_2Cl_2$ at room temperature in the absence of a solvent:

$$S_3N_2Cl_2 + Cl_2 \rightarrow 2 NSCl + SCl_2$$

$$3 NSCl \rightarrow (NSCl)_3.$$ 

This is the same reaction, essentially, as reported by Meuwsen if NS·SCl$_3$ is thought of as being NSCl + SCl$_2$ (41). In these four new methods, Maguire, Smith and Jolly characterized NSCl by its infrared spectrum (23), and allowed NSCl, in some cases, to polymerize to the trimer, which they then analysed for nitrogen and chlorine.

Also in 1963, Jolly reported that when $S_3N_2Cl$ was heated to 120-150°C in a vacuum it decomposed into a variety of products, including gaseous SCl$_2$ and NSCl, leaving a residue of pure $S_4N_3Cl$ behind (35, 36).

In 1965, Smith and Jolly reported one additional preparation of NSCl. They found that not only was NSCl formed by the reaction of active nitrogen with $S_2Cl_2$ but also by the reaction of active nitrogen with SCl$_2$ as follows (53, 54):

$$N + SCl_2 \rightarrow NSCl + 1/2 Cl_2$$

However, they concluded that neither of the reactions studied was really useful as a method of preparation for NSCl.

In 1966, Becke-Goehring reported yet another preparation of
NSCl that was very similar to that given earlier by Maguire, Smith and Jolly (40). She reported that NSCl resulted from the reaction of ammonia with $S_2Cl_2$ (and also with $SCl_2$) according to the reaction

$$4S_2Cl_2 + 2NH_3 \rightarrow 2NSCl + 6S + 6HCl,$$

and characterized the NSCl by its infrared spectrum (7).

Preparation of $(NSCl)_3$

The preparation of NSCl by heating $(NSCl)_3$ in a high vacuum as first described by Glemser and Perl (22) was chosen as the method of preparation for this investigation because it seemed to be the most suitable for both the infrared and electron diffraction work. $(NSCl)_3$ is a solid at room temperature and, even though it is highly sensitive to moisture (35, 42, 51), it can be stored in a dry atmosphere for long periods. This makes $(NSCl)_3$ a very convenient source of NSCl as the need arises.

The early directions for preparing $(NSCl)_3$ called for tetrasulfur tetranitride, $S_4N_4$, as the main starting material. However, since it is not easily obtained, it was decided to employ $S_3N_2Cl_2$ instead and to proceed with the preparation of $(NSCl)_3$ according to Maguire, Smith and Jolly's method 4 above. The compound $S_3N_2Cl_2$ is rather easily prepared by reaction of $NH_4Cl$ and $S_2Cl_2$ (35, 36, 40) in the presence of excess $NH_4Cl$. The overall reaction is
4S_2Cl_2 + 2NH_4Cl \rightarrow S_3N_2Cl_2 + 8HCl + 5S.

The procedure used here for the preparation of S_3N_2Cl_2 (and (NSCl)_3) was essentially the same as outlined by Jolly (35) with only minor variations as described in what follows.

S_3N_2Cl_2. One hundred grams (1.87 moles) of granular NH_4Cl (reagent grade), 20 grams of powdered sulfur (technical grade), and 100 milliliters (168 grams or 1.24 moles) of S_2Cl_2 were added in that order to a 500 milliliter round-bottomed flask with a 24/40 T joint. The S_2Cl_2 (practical grade) was purified by vacuum distillation (3 times) before being used. An air condenser (25 mm o. d., 30 cm long), made by joining an inner and an outer 24/40 T connection, was attached to the flask. Two drying tubes filled with anhydrous calcium sulfate were attached to the top of the air condenser and the entire setup was placed in a hood. All ground glass joints were lubricated with Kel-F No. 90 halocarbon grease. The yellow mixture was heated to boiling with a heating mantle and the heating then adjusted so that the upper level of the refluxing S_2Cl_2 was just inside the bottom joint of the air condenser. Thirty minutes after the start of the heating, brownish-yellow crystals of S_3N_2Cl_2 began to form in the air condenser. The heating was adjusted so that the crystals formed in the lower one-third of the air condenser but not inside the 500 milliliter flask. The heating was stopped after 12 hours of refluxing (before the S_2Cl_2 was entirely gone). The assembly was allowed to cool to
room temperature during which time most of the liquid adhering to the
S\textsubscript{3}N\textsubscript{2}Cl\textsubscript{2} crystals drained away. The air condenser containing the
crystals of S\textsubscript{3}N\textsubscript{2}Cl\textsubscript{2} was removed from the flask and the bottom of
the condenser was immediately closed off with a 24/40 T outer stopper.
The two calcium sulfate drying tubes were removed from the top of the
air condenser, a vacuum line connection was made, and the new
assembly was evacuated. The operation of removing the air condenser
from the reaction flask and making the vacuum connection was done in
less than 10 seconds, because the S\textsubscript{3}N\textsubscript{2}Cl\textsubscript{2} is very sensitive to water.
The air condenser was evacuated overnight to remove the volatile
impurities, hydrogen chloride and sulfur dichloride, which were col-
lected in a liquid nitrogen trap between the air condenser and the
vacuum pump.

\[ \text{(NSCl)}_3. \] The S\textsubscript{3}N\textsubscript{2}Cl\textsubscript{2} was converted into \((\text{NSCl})_3\) according to
the reaction (35)

\[ 3 \text{S}_3\text{N}_2\text{Cl}_2 + 3 \text{Cl}_2 \rightarrow 2(\text{NSCl})_3 + 3 \text{SCl}_2. \]

The air condenser containing the S\textsubscript{3}N\textsubscript{2}Cl\textsubscript{2} was placed in a dry bag
(Instruments for Industry and Research) together with a 200 milli-
liter three-necked (19/38 x 24/40 x 19/38 T) round-bottomed flask,
a 19/38 T vacuum connection with a 2 mm stopcock, a 19/38 T chlorine
gas connection with a 2 mm stopcock, two drying tubes filled with
anhydrous calcium sulfate, and a 2 mm stopcock. After flushing of the
dry bag and its contents with dry nitrogen gas, the original vacuum line connection and the bottom 24/40 T plug were removed from the air condenser. The two drying tubes (connected end-to-end themselves via a T joint) filled with calcium sulfate were attached to the top of the air condenser (via the 24/40 T joint), the bottom of which was then placed in the center neck of the 200 milliliter flask. The open end of the second drying tube had been previously fit with a one-hole rubber stopper into which the 2 mm stopcock was placed. The 19/38 T vacuum connection and chlorine gas connection were then placed in the remaining two necks of the flask. All ground glass joints were lubricated with Kel-F No. 90 grease.

With all three stopcocks closed, the apparatus was removed from the dry bag and placed in a hood where the vacuum connection was attached to a vacuum pump via a liquid nitrogen trap and the chlorine gas connection was attached to a tank of chlorine gas via tygon tubing. The 2 mm stopcocks between the flask and the tank of chlorine gas, between the flask and the vacuum pump, and in the end of the second drying tube will be referred to as stopcocks 1, 2 and 3 respectively. The system was first evacuated up to stopcock 2. With stopcocks 1 and 3 open and 2 closed, a slow stream of chlorine gas was passed over the $S_3N_2Cl_2$ through the air condenser. Within seconds after the start of the chlorine gas flow, the $S_3N_2Cl_2$ crystals turned into a dark red-brown slurry which eventually ran down the
sides of the air condenser and into the flask. The chlorine gas was passed for approximately 30 minutes with occasional shaking of the slurry. The chlorine gas flow was then stopped, stopcocks 1 and 3 were closed, stopcock 2 was opened, and the assembly was evacuated to less than $10^{-2}$ mm Hg. The pumping was continued for approximately 20 minutes during which time the sulfur dichloride ($S\text{Cl}_2$) produced in the reaction was removed and collected in the liquid nitrogen trap. After this first chlorination, the solid product in the flask was an orange-yellow color. Stopcock 2 was closed at the end of the pumping period, and dry air was admitted to the apparatus via the drying tubes by opening stopcock 3. Stopcock 1 was opened and a second 30 minute chlorination was done as described above, and the sulfur dichloride was again removed by evacuating the flask assembly. The solid product in the flask was more yellow and less orange in color at the end of this second chlorination. The chlorination procedure was repeated a third and a partial fourth (15 minutes) time until there was no darkening of the pale yellow product in the flask when the chlorine was passed over it. At the end of the chlorination procedure, the assembly was evacuated for approximately two hours at room temperature. Dry air was admitted to the flask assembly via stopcock 3, and the assembly was placed in the dry bag and flushed with dry nitrogen gas. The ($\text{NSCl}_3$) crystals were removed from the flask with a spatula and placed in a glass vial with an airtight polyethylene stopper.
This sample vial was then kept in a desiccator at room temperature when not being used.

Approximately ten grams of (NSCl)_3, a pale yellow crystalline solid at room temperature, were obtained from this preparation, corresponding to a 20% yield based on the S_2Cl_2. The (NSCl)_3 had a melting point of 74.1 - 75.9°C (with decomposition), which agreed well with the 75°C value reported by Jolly for 99% pure material (35, 40). The (NSCl)_3 was used with no further purification.

**Preparation of ^15_N Enriched (NSCl)_3**

The procedure described above for preparing the intermediate S_3N_2Cl_2 was designed to produce a maximum yield based on S_2Cl_2 and required an excess of NH_4Cl (35). Because of the high cost of ^15_NH_4Cl, a different procedure based on the same reactions but using a modest excess of S_2Cl_2 instead, was employed. The procedure was essentially that of Logan and Jolly (39), but incorporated small changes.

Seven grams of sulfur (technical grade), 3.30 grams of NH_4Cl and 20 milliliters of S_2Cl_2 (practical grade) were added to a 50 milliliter round-bottomed flask with a 24/40 T joint. The S_2Cl_2 was used without further purification. The NH_4Cl reagent was prepared by combining 2.97 grams of reagent grade ^14_NH_4Cl (99.64% ^14N) with 0.33 grams of ^15_NH_4Cl (Bio-Rad Laboratories; 99% ^15N) to give a
material corresponding to 10.7% $^{15}$N enrichment ($^{14}$N/$^{15}$N = 8.34). A 24/40 T water cooled West condenser (30 cm long and 12 mm i.d.), the top of which was fitted with two drying tubes filled with anhydrous calcium sulfate, was attached to the flask and the assembly was placed in a hood. All joints were again lubricated with Kel-F No. 90 halocarbon grease. The yellow slurry was heated with an electric heating mantle as before with the crystals of $S_3N_2Cl_2$ being made to form in the lower portion of the West condenser by proper adjustment of the heating. The heating was stopped after 18 hours of refluxing and the apparatus allowed to stand overnight in order to allow the liquid adhering to the crystals to drain off. There appeared to be very little $NH_4Cl$ unreacted. The two drying tubes on top of the West condenser were removed and quickly replaced with two others with fresh anhydrous calcium sulfate. The upper drying tube had a 2 mm stopcock (stopcock number 3) in its lower end as before. The chlorination apparatus described in the previous section was assembled, except for the central condenser section, in a hood, and the system was evacuated to stopcock 2 (the stopcock between the flask and the vacuum pump). The 50 milliliter flask was removed from the bottom of the West condenser, which was then immediately placed in the center neck of the three-necked 200 milliliter flask and the system was quickly evacuated up to stopcocks 3 and 1 (the stopcock between the flask and the tank of chlorine gas). The pumping was continued in this
manner for 10 minutes to remove any water vapor. Stopcock 2 was closed, dry air was admitted to the system via stopcock 3 and the drying tubes, and a stream of chlorine gas was passed over the $S_3N_2Cl_2$ as previously described. Two 30 minute chlorinations of the $S_3N_2Cl_2$, each followed with at least 15 minutes pumping to less than $10^{-2}$ mm Hg to rid the system of $SCl_2$, and one final 15 minute chlorination were carried out. After the final chlorination the apparatus was evacuated for two hours at room temperature. The flask assembly with all three stopcocks closed was transferred to the dry bag where the $^{15}$N enriched $(NSCl)_3$ was removed from the flask with a spatula and placed in a glass vial with an airtight polyethylene stopper. This sample vial was also kept in a desiccator at room temperature.

Only 0.623 grams of $^{15}$N enriched $(NSCl)_3$ were produced by this method—a yield of approximately 12% based on ammonium chloride. The $^{15}$N enriched $(NSCl)_3$ was used with no further purification.
ELECTRON DIFFRACTION

Introduction

Since the theory of electron diffraction and the general technique of molecular structure determination by electron diffraction have been described in numerous other works (3, p. 7-64, 10, p. 5-45, 11, 14, p. 5-44, 18, 49, p. 3-13, 58), only a brief outline of the method will be presented here.

When a beam of electrons accelerated by 40 kilovolts, which gives the electrons a wavelength of about 0.06 Å, is passed through a gaseous sample, diffraction effects are observed. The electrons are scattered by the potential field of the molecules and, since the molecules in the gaseous sample are randomly oriented, the diffraction pattern produced is radially symmetric. The diffraction pattern is normally recorded on a photographic plate. The total scattered intensity \( I_t \) of the electron beam is composed of four main parts; molecular scattering \( I_m \), atomic scattering \( I_a \), incoherent scattering \( I_i \), and extraneous scattering \( I_{ext} \), which is due to scattering from the diffraction apparatus:

\[
I_t = I_m + I_a + I_i + I_{ext}
\]  

The total scattered intensity \( I_t \) is related to the measured intensity striking the plate \( I_p \) by the equation
I_p = I_t \cdot a(s) \cdot \cos^3 \theta, \quad (2)

where \( a(s) \) is a function characteristic of the rotating sector (described below) and \( \theta \) is the scattering angle. A spiral shaped sector is used and rotated in a plane parallel to and just above the photographic plate during the exposure; the axis of the sector is coincident with the undiffracted electron beam. The sector opening is an increasing function of the scattering angle \( a(s) = \phi r^3 \), and is designed to alter the normal, rapid decrease of intensity with scattering angle in a way to give more uniform exposure over the full range of scattering angle. The \( \cos^3 \theta \) term is a geometric factor that takes into account the fact that the photographic plate is not everywhere equidistant from the scattering point.

The total scattered intensity can be represented by the equation

\[
I_t = k \sum_{i \neq j} \frac{|F_i|}{s} \frac{|F_j|}{s} e^{-\frac{r_{ij}^2}{2}} \cdot \cos(\eta_i - \eta_j) \cdot \sin(r_{ij})
\]

\[
+ k \sum_i \frac{|F_i|^2}{s^4} + k \sum_i \frac{Z_i S_i}{s^4} + I_{\text{ext}} \quad (3)
\]

where \( Z_i \) = atomic number of \( i^{\text{th}} \) atom

\( |F_i| \) = scattering amplitudes

\( r_{ij} \) = internuclear distance between atoms \( i \) and \( j \)

\( l_{ij} \) = root-mean-square amplitude of vibration between
atoms i and j

\[ \eta_i = \text{phase shift factors} \]

\[ S_i = \text{incoherent scattering factor} \]

\[ s = \frac{4\pi}{\lambda} \sin \theta \]

\[ \lambda = \text{wavelength of the electrons} \]

\[ 2\theta = \text{scattering angle}. \]

The four terms of the right-hand side of equation 3 may be respectively identified with those of equation 1. Since the last three terms are not sensitive to the molecular structure, they may be conveniently regarded as background, \( B_t \), and are subtracted from the total intensity (described in a later section) leaving the molecular structure sensitive part, \( I_m \). Thus

\[ B_t = I_a + I_i + I_{\text{ext}} \]

\[ I_m = I_t - B_t \]  \hspace{1cm} (4)

The molecular scattering part, \( I_m \), of equation 3 is normally multiplied by \( s^4 \), because the intensity as seen in equation 3, decreases rapidly with increasing \( s \). Thus the molecular scattering part becomes

\[ I_m' = s^4 I_m = k \sum_{i \neq j} \frac{|F_i||F_j|}{r_{ij}^4} \cdot e^{-\frac{1}{2} s^2/2} \cdot \cos(\eta_i - \eta_j) \sin r_{ij} \sin \theta \]

or

\[ sI_m' = k \sum_{i \neq j} \frac{|F_i||F_j|}{r_{ij}^2} \cdot e^{-\frac{1}{2} s^2/2} \cdot \cos(\eta_i - \eta_j) \sin r_{ij} \sin \theta . \]
The process of determining the molecular structure of the molecule in question now consists of analysing this molecular intensity curve. In its simplest form the procedure consists in the discovery of a trial structure based on the analysis of a radial distribution curve calculated from the experimental intensity curve followed by parameter refinement, usually by least squares adjustment of intensity curves. In practice, a preliminary radial distribution curve is obtained from the molecular intensity curve by a Fourier transformation. The initial values of the internuclear distances and root-mean-square amplitudes of vibration obtained from this radial distribution curve are then used to derive a model of the structure from which a theoretical intensity curve may be calculated for comparison with the experimental. This comparison may suggest corrections to the experimental background following which the process may be repeated, or the refinement stage may be initiated. Details of the procedure may vary, depending upon the complexity of the problem.

The most easily interpreted form of the radial distribution curve is generally held to be that represented by a sum of Gaussians, each corresponding to an interatomic distance of a weight determined by the scattering power of the atomic pair. Such a curve may be obtained for molecules composed of atoms of not-too-different atomic numbers \((\cos \Delta \eta_{ij} \approx 1)\) by multiplying the intensity represented by equation 5 by \(Z_r Z_S / |F_r| |F_S|\) to give
\[
\frac{sI^{\prime\prime\prime}}{m} = k \sum_{i \neq j} \frac{|F_i||F_j|}{|F_r||F_S|} \cdot \frac{Z_i Z_s}{r_{ij}} \cdot e^{-s^2/2} \cdot \cos(\eta_i - \eta_j) \sin r_{ij}.
\]

or

\[
\frac{sI^{\prime\prime\prime}}{m} = \sum_{i \neq j} A_{ij} \sin r_{ij}. \tag{6}
\]

Equation 6 can be rewritten as if the distribution of atoms is continuous over all space

\[
\frac{sI^{\prime\prime\prime}}{m} = \int_{0}^{\infty} \frac{P(r)}{r} \sin rsdr, \tag{7}
\]

where \(P(r)\) is a probability distribution. Use of the well-known Fourier integral theorem leads to the following form of the radial distribution curve:

\[
\frac{P(r)}{r} = \frac{2}{\pi} \int_{0}^{\infty} \frac{sI^{\prime\prime\prime}}{m} \sin rsds \tag{8}
\]

or

\[
\frac{P(r)}{r} = \frac{2}{\pi} \sum_{s_{\text{min}}}^{s_{\text{max}}} \frac{sI^{\prime\prime\prime}}{m} \sin rs \Delta s. \tag{9}
\]

In the approximation \(\cos \Delta \eta = 1\) and \(|F_i||F_j|/|F_r||F_S|\) equals a constant, the calculated radial distribution curve has the form

\[
\frac{P(r)}{r} = \frac{1}{\sqrt{2\pi}} \sum_{i \neq j} \frac{Z_i Z_s}{r_{ij}} \cdot e^{-s^2/2} \cdot \frac{1}{\Delta r_{ij}} \cdot \frac{1}{2}. \tag{10}
\]
where $\Delta r_{ij} = (r_{ij} - r_{ij})$. The intensity curve $sI''_m$ is normally multiplied by a factor $e^{-bs^2}$ to provide for better convergence of the series. Thus the radial distribution curve becomes

$$\frac{P(r)}{r} = \frac{2}{\pi} \sum_{s_{\text{min}}}^{s_{\text{max}}} sI''_m e^{-bs^2} \sin rs \Delta s$$

or

$$\frac{P(r)}{r} = \frac{1}{\sqrt{2\pi}} \sum_{i \neq j} \frac{Z_i Z_j}{r_{ij}^2} \cdot \frac{1}{\sqrt{1 + 2b}} \cdot e^{-\frac{(\Delta r_{ij})^2}{2(1 + 4b)}}$$

The damping factor $b$ is usually calculated from the relationship

$$e^{-bs^2} = 0.1 \text{ at } s = s_{\text{max}}.$$

The radial distribution function is related to the probability of finding a particular internuclear distance, $r_{ij}$, in a molecule. In the above (harmonic) approximation, the calculated function (curve) consists of a sum of curves, each one approximately a Gaussian, whose position represents an internuclear distance. The area of each peak is proportional to $nZ_i Z_j / r_{ij}$, where $n =$ multiplicity of the distance $r_{ij}$. The root-mean-square amplitude of vibration ($l_{1/2}$) may be calculated from the expression

$$\frac{1}{2} = \exp \left[ -\frac{(\Delta r_{1/2})^2}{2(1 + 4b)} \right]$$

where $\Delta r_{1/2}$ is the half-width of the peak at half-height and $b$ is the
The refinement of a structure derived as described above is ordinarily carried out by the method of least squares applied to intensity curves. The mathematical functions vary from laboratory to laboratory, and even from case to case; in this laboratory functions similar to (sometimes identical to) equation 5 are used. The principles of the method (non-linear least squares curve-fitting) are well known, and application of the method to electron diffraction is straightforward in theory but rather complicated in practice (4, 29, 33). It is sufficient for the purpose of this thesis to say only that the refined structure results from the satisfactory fit of a theoretical intensity curve to the experimental curve using as starting values of the parameters those of the suitable trial structure. The reader is referred to other accounts for detailed descriptions (4, 29, 31, 33).

**Experimental and Data Reduction**

A sample bulb with a 4 mm vacuum stopcock and a 10/30 T joint was charged with approximately 2 grams of trithiazyl chloride, (NSCl)$_3$, and attached to the new Oregon State University electron diffraction apparatus. All joints were lubricated with Kel-F No. 90 halocarbon grease. Electron diffraction photographs were taken of gaseous thiazyl chloride, NSCl, at each of the three nozzle-to-plate distances available using a rotating sector with an opening proportional
The NSC₁ was generated by heating the sample bulb containing the (NSC₁)₃ with a hot water bath held between 65 and 85°C. The nozzle section of the apparatus was heated to 90-100°C by passing hot nitrogen gas in and around the nozzle inset in order to minimize the rapid polymerization. A total of 25 photographs were taken (10 at the short, 10 at the middle, and 5 at the long camera distance) using the same sample of (NSC₁)₃. A total of 7 photographs (2 from each the long camera and middle camera distance and 3 from the short camera distance) were chosen to be analysed. Data for these 7 electron diffraction photographs are given in Table 5.

Each plate was scanned using a modified Joyce-Loebl microdensitometer which produces digital records of the plate density on punch tape. The plates were rotated about the axis of the rings during the scans in order to minimize the effect of emulsion grain. These data were reduced in the usual way (26, 28, 30) to give curves of a form corresponding to superposition of the molecular scattering (equation 5) superposed on a smooth background. These curves were plotted and a smooth background curve drawn through each. The difference curves (intensity curve minus background) were plotted and used for the structure determination. They are of the form of equation 5 and are shown in Figure 1 in their final form which includes small additional background corrections introduced in the course of the analysis. Tables 6-12 are the data for these curves.
Structure Determination, Refinement, Final Results

Determination of the structure of NSC1 began with the calculation of the experimental radial distribution curve as outlined in earlier paragraphs. Scattering amplitudes for sulfur and nitrogen (and chlorine for use in later calculations) corresponding to an accelerating voltage of 44.3 KV (the average used in the NSC1 experiments) were obtained from Cox's tables by an interpolation procedure (13, p. 41 and 46, 26). These amplitudes were multiplied by $s^2$ to put them into the form suitable for converting $s_{m}^{I}$ (equation 5) to $s_{m}^{II}$ (equation 6).

The calculation of the radial distribution curve is done on a single intensity curve which, for greatest general utility, should include the largest possible range of scattering angle. This curve (called a "composite intensity curve") was obtained by first averaging the curves obtained at each camera distance to give three average curves, and then scaling and averaging these curves in the overlap region to yield the composite curve having the mathematical form suggested by equation 5. A "constant coefficient" intensity curve of the form of equation 6 was calculated by multiplying the composite curve by the factor $Z_{N}Z_{S}/|F_{N}||F_{S}|$. A preliminary experimental radial distribution curve was calculated according to equation 11 with a damping factor $b = 0.0009 \, \text{Å}^2$ and an $s$ range of 1.00 to 48.75 $\, \text{Å}^{-1}$.

The radial distribution curve showed three peaks at 1.45, 2.16,
and 3.11 Å of which the two shorter correspond to bond distances and the longest to the non-bond distance. These distance values suggest immediately the structure NSCl: the sum of the single-bond radii for N and either S or Cl (1.78 and 1.73 Å respectively) is much too small to account for the 2.16 Å value, which is rather close to the value 1.99 Å corresponding to the S-Cl radius sum corrected for electronegativity. This conclusion about the structure finds further proof in the magnitudes (areas) of the peaks, which may be expressed by

\[ W_{ij} = \frac{nZ_i Z_j}{r_{ij}}. \]  

(14)

A comparison of the calculated areas of peaks corresponding to both models and the observed areas is given in Table 17. The agreement for the structure NSCl is excellent.

A (variable coefficient) theoretical intensity curve was calculated for NSCl according to equation 5 using the distance values shown by the preliminary experimental radial distribution curve and the root-mean-square amplitudes of vibration calculated from it according to equation 13. The theoretical intensity curve was compared to the composite experimental intensity curve. Small additional background corrections were made on the original individual curves and an improved composite intensity curve was then calculated. Theoretical intensity
data from this curve for the missing experimental data in the range 
$0 \leq s \leq 1.00$ were spliced to the experimental curve and the correspond-
ing experimental radial distribution curve was then calculated. The 
improved values of the distances and the amplitudes of vibration 
obtained from this new radial distribution curve were then used to 
calculate a better theoretical intensity curve which suggested additional 
small background corrections. This cyclic process was repeated until 
most of the background errors were eliminated. The final experimental 
intensity data are given in Tables 6-12; the curves are shown in 
Figure 1. The final experimental composite intensity data calculated 
from the individual data are given in Table 13; the curve is shown in 
Figure 2. The final experimental radial distribution data calculated 
from the composite intensity data are given in Table 14; the curve is 
shown in Figure 3.

Two different refinements of the molecular structure of NSCl 
were done using the method of least squares based on intensity curves 
(4, 29, 31, 33). Starting values of the parameters were $r_{NS} = 1.45 \text{ \AA}$, 
$r_{SCL} = 2.16 \text{ \AA}$, $r_{N...Cl} = 3.11 \text{ \AA}$, $l_{NS} = 0.0417 \text{ \AA}$, $l_{SCL} = 0.0509 \text{ \AA}$, 
and $l_{N...Cl} = 0.0755 \text{ \AA}$. In one of these refinements (Least Squares I) 
six individual intensity curves (all except I-233-07) were used; in the 
other (Least Squares II) the composite intensity curve was used. The
results of these two refinements are given in Table 18. The results of the more general refinement based on the individual curves was taken to be the best expression of the NSCl structure. The results of the refinement on the composite curve were used only for comparison purposes. As can be seen from Table 18, there is good agreement between the two calculations.

The final values for the parameters and the associated errors for the NSCl molecule are given in Table 19. The total associated errors $2\sigma$ were estimated from the following formulas (26, 28):

\[
2\sigma_1 = 2 \left[ 2\sigma_{LS}^2 + (0.0005 \sigma_r)^2 \right]^{1/2}
\]

\[
2\sigma_1 = 2 \left[ 2\sigma_{LS}^2 + (0.02 \overline{L})^2 \right]^{1/2}
\]

\[
2\sigma_0 = 2 \left[ 2\sigma_{LS}^2 \right]^{1/2}
\]

The $\sigma_{LS}^2$ values associated with each parameter were taken directly from the error matrix calculated during the course of the least squares refinement. The $\sigma_{LS}^2$ values were multiplied by a factor of 2 as a very rough estimate of the independency of the data. The factors 0.0005 and 0.02 reflect uncertainties in the measurement of the camera height, the wavelength of the electrons, the calibration of the rotating sector, the atomic form factors, and converting density to intensity (26, 28). The $r$ and $\bar{l}$ values were also taken from the least squares refinement.
The error matrix (4, 28, 29, 33) resulting from the refinement Least Squares I is given in Table 20 and reflects an important part of the analysis. It provides the information about the standard errors and error correlations. The diagonal elements are the square of the standard errors, \( \sigma^2_{LS} \), associated with each parameter, and the off-diagonal elements are the average products of the errors and give information about error correlations.

A final theoretical intensity curve and radial distribution curve, based on the final parameters as determined by the least squares refinement on the individual intensity curves, were calculated for NSCl according to equations 5 and 11 respectively. The theoretical intensity data is given in Table 15; the curve is shown in Figure 2 along with the experimental intensity curve and the difference curve (experimental minus theoretical). The theoretical radial distribution data is given in Table 16; the curve is shown in Figure 3 along with the experimental radial distribution curve and the difference curve (experimental minus theoretical). The agreement is excellent for both sets of curves.
The electron diffraction investigation of thiazyl chloride just described showed the molecule to have the bent NSCl structure. Such a structure (symmetry $C_s$) gives rise to three vibrational degrees of freedom according to the relevant symmetry considerations and selection rules (64, p. 77-168). These constitute three fundamental frequencies: $\nu_1$ and $\nu_2$, the frequencies corresponding approximately to the N-S and S-Cl stretching vibrations respectively; and $\nu_3$, the frequency corresponding approximately to the NSCl bending vibration. The normal modes of vibration for a molecule of this type have been described elsewhere (38, 52). All three vibrations are both infrared and Raman active and belong to the $A'$ symmetry species (64, p. 77-168).

This investigation of the infrared spectrum of NSCl was undertaken to measure the three fundamental frequencies of the gaseous molecule, and to study the previously unmeasured spectrum of NSCl in the solid state. It was planned to use the results of the gas phase work, along with the electron diffraction results, in a force constant analysis of the molecule.

At the beginning of this investigation of the infrared spectrum of
NSCl only one of the three fundamental frequencies of the gaseous molecule had been observed. Glemser and his co-workers had studied the infrared spectrum of gaseous NSCl in the NaCl region and reported seeing only one band at 1322-1330 cm\(^{-1}\), which was interpreted as the S-N stretching vibration (the compound was still thought to have the SNCl structure at that time as described earlier)(22, 23). Towards the end of my investigation, Müller reported all three fundamental frequencies for gaseous NSCl (44). However, the infrared spectrum was scanned very quickly after the preparation of NSCl and was only studied from 300-4000 cm\(^{-1}\). The two fundamentals \(\nu_1\) and \(\nu_2\) were directly observed, but the third one, \(\nu_3\), was calculated from overtone and combination bands. Their observed fundamental frequencies and assignments are given in Table 3.

**Gas Phase**

Gaseous NSCl was prepared by heating \((\text{NSCl})_3\) to 90-100°C in a high vacuum as first described by Glemser and Perl (22). A sample enriched with \(^{15}\)N was studied along with the one containing the naturally occurring isotopes. The samples of \((^{14}\text{NSCl})_3\) and \((^{15}\text{NSCl})_3\) were prepared as previously described under Sample Preparation. The infrared spectrum was studied from 200 to 4,000 cm\(^{-1}\) using Beckman IR 7 and IR 11 spectrophotometers.

The NSCl gas samples were contained in 10-cm glass infrared
cells with KBr and polyethylene windows at room temperature. These gas cells had two vacuum connections 180° opposite each other. The particular cell being used was attached to a vacuum line via one of these connections and evacuated. A small glass bulb charged with a small amount of (NSCl)₃ was attached directly to the other connection on the opposite side of the cell and immediately evacuated. The (NSCl)₃ had been transferred from the sample storage vial to the small glass bulb in a dry bag flushed with dry nitrogen gas. The gas cell with the sample bulb attached was pumped in this manner for a few hours to ensure proper removal of water vapor. A vacuum of 10⁻⁴ mm Hg could be produced in this system. After being pumped for a few hours, the cell was filled with NSCl gas by heating the small sample bulb with an oil bath held at 90-100°C. Pressures varying from a few mm to 50 mm Hg were used. The cell was then isolated and removed from the vacuum line and an infrared scan was made. The NSCl gas pressures (Pi) as read on the vacuum line manometer served only to designate upper limits of the actual pressures during the scans themselves since NSCl gas tends to polymerize at room temperature. At low pressures the polymerization problem was minimal. Repeated scans of the same sample at low pressure showed essentially the same band intensities, thus making it possible to use very slow scanning speeds and to position the bands as accurately as possible. The Beckman infrared spectrophotometers were calibrated
in the regions of interest to ensure accurate readings of the band positions.

Three main bands were observed in the infrared spectrum of gaseous NSCl at 1324.0, 417.8, and 271 cm\(^{-1}\), corresponding to the three fundamental frequencies \(\nu_1\), \(\nu_2\), and \(\nu_3\), respectively. These values given are for the frequencies corresponding to the most abundant isotopes present (\(^{14}\text{N}, \ ^{32}\text{S}, \ ^{35}\text{Cl}\)) and agree quite well with those reported by Müller (44). The relative intensities of the three bands were \(\nu_2 > \nu_1 > \nu_3\), with \(\nu_3\) being much weaker than the other two. No combination and overtone bands were observed for \(\nu_2 + \nu_3\) and \(2\nu_3\) as was reported by Müller (44), who apparently must have used much higher vapor pressures. Such a higher pressure would account for the fast scans used by Müller since the polymerization of NSCl becomes an important factor.

The N-S stretching vibration, \(\nu_1\), at 1324.0 cm\(^{-1}\) showed P-Q-R structure with a P-R splitting of 15.6 cm\(^{-1}\). The \(^{15}\text{N}\) enriched sample of NSCl showed a shift of this band to 1293.4 cm\(^{-1}\), but the R branch minimum was not measurable and a P-R splitting could not be calculated. The frequency shift expected for a \(^{14}\text{N}\) to \(^{15}\text{N}\) isotope change was calculated based on a harmonic oscillator-diatomic molecule (N-S) approximation. The calculated frequency for the \(^{15}\text{N} - ^{32}\text{S}\) species was 1293.0 cm\(^{-1}\) relative to 1324.0 cm\(^{-1}\) for the \(^{14}\text{N} - ^{32}\text{S}\) species. This is in excellent agreement with the observed value of
1293.4 cm\(^{-1}\) for the \(^{15}\)N enriched sample. The assignment of this fundamental frequency to the N-S stretching vibration was thus confirmed by the demonstrated nitrogen isotope dependence. The N-S stretching vibration portion of the spectrum is shown in Figure 4 and the observed frequencies are given in Table 21.

The S-Cl stretching vibration, \(\nu_2\), at 417.8 cm\(^{-1}\) showed P-Q-R structure with a P-R splitting of 17.0 cm\(^{-1}\). The \(^{15}\)N enriched sample of NSCl showed no measurable change in this fundamental frequency. The band did show a double Q branch at 417.8 cm\(^{-1}\) and 414.1 cm\(^{-1}\) corresponding to the presence of \(^{35}\)Cl and \(^{37}\)Cl isotopes respectively. The frequency shift expected for a \(^{35}\)Cl to \(^{37}\)Cl isotope change was calculated, as before, based on a harmonic oscillator-diatom molecule (S-Cl) approximation, although this approximation is normally not too good for intermediate frequencies. The calculated frequency for the \(^{32}\)S-\(^{37}\)Cl species was 412.4 cm\(^{-1}\) relative to 417.8 cm\(^{-1}\) for the \(^{32}\)S-\(^{35}\)Cl species, in fairly good agreement with the observed value of 414.1 cm\(^{-1}\). The band shape of this fundamental frequency is very similar to that of the C-Cl stretching vibration in methyl chloride in that both exhibit a P-Q-R structure with a double Q branch due to the two chlorine isotopes present (2, 9, 32, p. 312-314). The assignment of this fundamental frequency to the S-Cl stretching vibration was thus confirmed by this chlorine isotope dependence and lack of dependence on the nitrogen isotope. The S-Cl
stretching vibration portion of the spectrum is shown in Figure 5 and the observed frequencies are given in Table 21.

The NSCl bending vibration, \( \nu_3 \), at 271 cm\(^{-1}\) showed P-R structure with a P-R splitting of 17 cm\(^{-1}\). This portion of the spectrum is shown in Figure 6 and the observed frequencies are given in Table 21.

**Solid Phase**

Solid films of NSCl were prepared by heating \((\text{NSCl})_3\) to 90-100°C in a high vacuum and then immediately trapping the evolved NSCl gas onto substrate windows cooled by liquid nitrogen. A sample enriched with \(^{15}\text{N}\) was studied along with the one containing the naturally occurring isotopes. The samples of \((^{14}\text{NSCl})_3\) and \((^{15}\text{NSCl})_3\) were prepared as previously described under Sample Preparation. The infrared spectrum was studied from 95 to 4,000 cm\(^{-1}\) using the Beckman IR 7 and IR 11 spectrophotometers.

The NSCl solid films were deposited on NaCl, KBr, CsI, and polyethylene substrate windows in a pyrex low temperature infrared absorption cell similar to the one described by Wagner and Hornig (57), but differing in that aluminum end plates with O-ring grooves were made for attaching the infrared transparent windows, and a direct sublimation port, in a position perpendicular to the beam path, was added to the front of the vacuum jacket. The substrate windows,
imbedded in the copper cooling block attached to the cold finger of the
cell, could be rotated 90° to receive a film sample and then rotated
back into the position of the infrared beam path. A thin layer of Kel-F
No. 90 halocarbon vacuum grease was placed between the substrate
window and the small shoulder within the copper cooling block to
obtain adequate thermal contact between the window and the copper
block.

A direct sublimation technique was used in preparing the thin
films of NSCl. The low temperature cell was assembled and attached
to a vacuum line. A small glass bulb charged with a small amount of
(NSCI)$_3$ was attached to the sublimation port and immediately evacuated.
The cell and sample were pumped on for a few hours to ensure proper
removal of water vapor and then liquid nitrogen was added to the cold
finger to cool the substrate window. The substrate window was cooled
for at least two hours before heat was applied to the sample bulb by
means of an oil bath held at 90-100°C. The NSCl gas generated in this
manner was then trapped onto the substrate window in "puffs" by
opening and closing the stopcock between the sample bulb and the
vacuum jacket. In most cases, two to four "puffs" of NSCl gas gave
sufficient film thickness. The cell was isolated and removed from
the vacuum line. The substrate window was then rotated 90 degrees
into position to transmit the infrared beam, the cell was placed into
the appropriate spectrophotometer, and the spectrum was recorded.
The spectrophotometers were calibrated in the regions of interest to ensure accurate readings of the peak positions.

Only one main absorption at 1397.0 cm\(^{-1}\) was observed in the infrared spectrum of a solid film of NSCl on a KBr substrate window over the range 350 to 4,000 cm\(^{-1}\). The film was cloudy in appearance with a slight greenish-yellow color. The peak was very sharp, having a half-width of 1.5 cm\(^{-1}\) at half-height transmission. The spectral slit width was 0.8 cm\(^{-1}\) at 1400 cm\(^{-1}\). Also a sharp and much weaker peak was observed at 1384.8 cm\(^{-1}\) along with the main peak. These two absorptions at 1397.0 and 1384.8 cm\(^{-1}\) were tentatively assigned to N-S stretching vibrations of the \(^{14}\text{N}-^{32}\text{S}\) and \(^{14}\text{N}-^{34}\text{S}\) species respectively, because of their proximity to the gas phase value of 1324.0 cm\(^{-1}\). A \(^{15}\text{N}\) enriched sample of NSCl on KBr showed two additional peaks at 1365.0 and 1352.2 cm\(^{-1}\) corresponding to N-S stretching vibrations of the \(^{15}\text{N}-^{32}\text{S}\) and \(^{15}\text{N}-^{34}\text{S}\) species respectively. They, too, were very sharp with the 1365.0 cm\(^{-1}\) peak being much stronger than the 1352.2 cm\(^{-1}\) peak. The infrared spectrum of NSCl on NaCl was found to be identical to that of NSCl on KBr.

The frequency shifts expected for \(^{14}\text{N}\) to \(^{15}\text{N}\) and \(^{32}\text{S}\) to \(^{34}\text{S}\) isotope changes were calculated based on a harmonic oscillator-diatomic molecule (N-S) approximation. The calculated frequencies for the \(^{14}\text{N}-^{34}\text{S}\), \(^{15}\text{N}-^{32}\text{S}\), and \(^{15}\text{N}-^{34}\text{S}\) species were 1384.4, 1364.3, and 1351.5 cm\(^{-1}\), respectively, relative to 1397.0 cm\(^{-1}\) for the
$^{14}$N-$^{32}$S species. These frequencies are in excellent agreement with the observed values. The nitrogen and sulfur isotope dependence exhibited by the main peak at 1397.0 cm$^{-1}$ thus confirmed the assignment of this frequency to a N-S stretching vibration.

The large shift to higher wave-numbers and the sharpness of the peak of the N-S stretching vibration in the solid phase of NSCl on KBr compared to the gas phase tend to indicate an ionization of the NSCl molecule into (NS)$^+$, Cl$^-$ ions in the solid state. The large shift to lower wave-numbers (shown later) of the S-Cl stretching and NSCl bending vibrations in the solid phase compared to the gas phase can also be interpreted in terms of (NS)$^+$, Cl$^-$ ions. For this reason, I will refer to this solid form of NSCl as the "ionic" form. This N-S stretching vibration portion of the spectrum of NSCl on KBr is shown in Figure 7 and the observed frequencies are given in Table 22 along with those calculated frequencies mentioned above.

Additional qualitative evidence in favor of the interpretation of the 1397.0 cm$^{-1}$ absorption in terms of the (NS)$^+$ ion is found in the comparison of the fundamental frequency of the charged species with that of the neutral species. In 1955, Dressler made such comparisons for the NO, PO, and PS systems by reporting the ratio of the fundamental frequency of the charged species to that of the neutral species and found the ratios to be quite constant (16). (The calculation could not be made for the NS system because no value for the (NS)$^+$ frequency
He reported the following ratios: \((\text{NO})^+/\text{NO} = 2377/1904 = 1.25\); \((\text{PO})^+/\text{PO} = 1405/1232 = 1.14\); and \((\text{PS})^+/\text{PS} = 845/740 = 1.14\).

Using his value of 1220 cm\(^{-1}\) for the fundamental frequency of NS and the 1397.0 cm\(^{-1}\) value reported here for the \((\text{NS})^+\), I obtained the ratio \((\text{NS})^+/\text{NS} = 1.15\), which is in good agreement with the other ratios.

Three main peaks were observed at 1346, 350, and 256 cm\(^{-1}\) in the infrared spectrum of a solid film of NSCl on a CsI substrate window over the range 180 to 4,000 cm\(^{-1}\). In contrast to the cloudy appearance of the film of NSCl on KBr (and NaCl), this film was clear and glassy in appearance with a slight greenish-yellow color. The \(^{15}\text{N}\) enriched sample of NSCl on CsI showed one additional peak at 1315 cm\(^{-1}\). The spectrum of NSCl on CsI is shown as the upper curve in Figure 8 and the observed frequencies are given in Table 23. The peak at 1346 cm\(^{-1}\) corresponds to the N-S stretching vibration since it shows the nitrogen isotope dependence. The other two peaks at 350 and 256 cm\(^{-1}\) probably correspond to the S-Cl stretching vibration and to the NSCl bending vibration respectively. All three peaks are quite broad with the 1346 cm\(^{-1}\) peak having a half-width of 13 cm\(^{-1}\) at half-height transmission in comparison to the 1.5 cm\(^{-1}\) value for the "ionic" form peak at 1397.0 cm\(^{-1}\). For this reason, I will refer to this solid form of NSCl as the "molecular" form.

The "ionic" form could be produced on CsI by annealing the
"molecular" form. The "molecular" form was always formed with the initial deposit of NSCl on CsI cooled by liquid nitrogen. Once the presence of the "molecular" form was verified by its infrared spectrum, the cell was allowed to warm up very slowly while the peak at 1346 cm⁻¹ was being monitored. As the CsI substrate window warmed up, the peak at 1346 cm⁻¹ decreased in intensity. Once the intensity of the peak had decreased to about 5% of its original value, the cell was immediately cooled with liquid nitrogen, and the spectrum was re-scanned from 180 to 4,000 cm⁻¹. The three main peaks at 1346, 350, and 256 had disappeared and two others had developed at 1397.0 and around 205 cm⁻¹. The 1397.0 cm⁻¹ peak was very sharp and showed the same isotope dependence as the 1397.0 cm⁻¹ peak of the "ionic" form of NSCl on KBr. The peak around 205 cm⁻¹ was very strong and broad. The film had lost its clear glassy appearance and was now cloudy like the film on KBr. The middle curve in Figure 8 shows the spectrum of this "ionic" form of NSCl on CsI from 180 to 420 cm⁻¹, and the observed frequencies are given in Table 23. The region of the spectrum above 420 cm⁻¹ is the same as that shown in Figure 7 for the "ionic" form of NSCl on KBr.

Two peaks were observed at 205 and 140 cm⁻¹ in the infrared spectrum of a solid film of NSCl on a polyethylene substrate window over the range 95-640 cm⁻¹. Both peaks are quite broad and are attributed to the "ionic" form of NSCl since the 205 cm⁻¹ peak was
shown to be associated with the "ionic" form peak at 1397.0 cm$^{-1}$ by
the work done with the CsI substrate window. The absence of the two
peaks at 350 and 256 cm$^{-1}$, associated with the "molecular" form, also
substantiates this assignment. The film appearance was not observable
because of the cloudiness of the polyethylene substrate window itself.
The lower curve in Figure 8 shows the spectrum of NSCl on poly-
ethylene from 100 to 420 cm$^{-1}$, and the observed frequencies are given
in Table 23.
FORCE CONSTANTS AND AMPLITUDES OF VIBRATION

Introduction

The determination of all the force constants of a general quadratic potential function can be accomplished if one has enough information to solve the necessary equations. The general quadratic potential function for NSCl has six force constants, and one would need at least six fundamental frequencies to solve the problem completely (64, p. 169-207). The results of the infrared spectroscopic investigation of gaseous NSCl gave three (at best five) fundamental frequencies—not enough to determine all six force constants.

The amplitudes of vibration from the electron diffraction investigation could possibly be used along with the fundamental frequencies to determine all six force constants for NSCl, but the solution was too complicated to carry out without very much more additional work, so I was content with verifying that a simple quadratic potential function gave good agreement.

The fundamental frequencies were used to calculate the force constants of a simple quadratic potential function from the secular equation $|xG-\lambda E| = 0$ (64, p. 54-76). The amplitudes of vibration from the electron diffraction investigation and the fundamental frequencies were used to calculate force constants of a simple quadratic potential function from the secular equation $|xE-\epsilon E| = 0$. 

as developed by Cyvin (15, p. 89-100). The results of the two force constant calculations were then compared. Finally, amplitudes of vibration were calculated according to Cyvin's method and compared with those found by the electron diffraction investigation.

A general description of mean amplitudes of vibration and molecular force fields for bent XYZ molecules has been reported elsewhere (44, 45, 55, 56). The theory summarized below follows closely that presented in more detail by Wilson, Decius, and Cross (64, p. 54-71), and by Cyvin (15, p. 89-100).

**Force Constants Based on Infrared Data**

A bent asymmetrical NSCl molecule, belonging to the symmetry point group $C_s$, has three vibrational degrees of freedom, giving rise to three fundamental frequencies: $v_1$ and $v_2$, the frequencies corresponding approximately to the N-S and S-Cl stretching vibrations respectively; and $v_3$, the frequency corresponding approximately to the NSCl bending vibration. All three vibrations are both infrared and Raman active and belong to the $A^1$ symmetry species.

For the NSCl molecule, the number of symmetry co-ordinates equals the number of internal co-ordinates. Using the 1-3-2 molecular model to represent the NSCl molecule, the following symmetry co-ordinates under the symmetry species $A^1$ were constructed:
\[ S_1 = \Delta r_{13}, \quad S_2 = \Delta r_{23}, \quad S_3 = (r_{13} r_{23})^{1/2} \Delta \alpha \]  

(18)

where \( r_{13}, \ r_{23}, \) and \( \alpha \) correspond to the 1-3 bond, 2-3 bond, and 1-3-2 angle at the equilibrium configuration respectively. The angle displacement was multiplied by the equilibrium bond lengths in order to keep the units of quantities referring to the angle bending the same as those of the bonded atom pairs.

On the basis of the above symmetry co-ordinates, the \( G \) matrix elements are (64, p. 54-71):

\[
G_{11} = \mu_1 + \mu_3 \\
G_{12} = G_{21} = \mu_3 \cos \alpha \\
G_{13} = G_{31} = -(r_{13} r_{23})^{1/2} \rho_{32} \mu_3 \sin \alpha \\
G_{22} = \mu_2 + \mu_3 \\
G_{23} = G_{32} = -(r_{13} r_{23})^{1/2} \rho_{31} \mu_3 \sin \alpha \\
G_{33} = (r_{13} r_{23}) \left[ \mu_1 \rho_{31}^2 + \mu_2 \rho_{32}^2 + \mu_3 (\rho_{31}^2 + \rho_{32}^2 - 2 \rho_{31} \rho_{32} \cos \alpha) \right]
\]

(19)

where \( \mu_1 = \frac{1}{m_1} \) and \( \rho_{ij} = \frac{1}{r_{ij}} \).

In terms of the above symmetry coordinates, the general quadratic potential function is (64, p. 169-182):
\[ 2V = F_{r_{13}} (\Delta r_{13})^2 + F_{r_{23}} (\Delta r_{23})^2 + F_a (r_{13}r_{23})(\Delta a)^2 \]

\[ + 2F_{r_{13}r_{23}} (\Delta r_{13})(\Delta r_{23}) + 2F_{r_{13}a} (r_{13}r_{23})^{1/2}(\Delta r_{13})(\Delta a) \]

\[ + 2F_{r_{23}a} (r_{13}r_{23})^{1/2}(\Delta r_{23})(\Delta a), \]

where \( F_{r_{13}} \) and \( F_{r_{23}} \) are the 1-3 and 2-3 stretching force constants respectively, \( F_a \) is the 1-3-2 bending force constant, and all the others are the interaction force constants involving the respective parameters. Using the above potential function, the following \( F \) matrix, potential energy matrix, elements are obtained:

\[ F_{11} = F_{r_{13}}, \quad F_{12} = F_{r_{13}r_{23}}, \quad F_{13} = F_{r_{13}a} \]

\[ F_{22} = F_{r_{23}}, \quad F_{23} = F_{r_{23}a}, \quad F_{33} = F_a. \]  

The \( F \) and \( G \) matrix elements could now be used to write the secular equation in the form \( |FG-\lambda E| = 0 \), where \( E \) is the unit matrix and \( \lambda \) is a frequency parameter related to the fundamental frequencies, \( \nu_1 \) in wave-numbers, by \( \lambda_1 = 4\pi c^2 \nu_1^2 \), where \( c \) is the velocity of light. The force constants were calculated by solving the secular equation \( |FG-\lambda E| = 0 \) with the assumption of a diagonal \( F \) matrix, i.e. \( F_{ij} = 0 \) for \( i \neq j \). The values of the structural parameters for NSCl and other constants used in all the calculations are found in Tables 19 and 24 respectively. The fundamental frequencies used in the calculations were for the \( ^{14}N, \; ^{32}S, \; \text{and} \; ^{35}Cl \).
isotopes and are also included in Table 24. The calculated values of the force constants of NSCl are given in the second column of Table 25 and agree well with those calculated by Müller given earlier in Table 4 (44), and are in line with those for NSF given in Table 2.

A second set of symmetry co-ordinates was constructed for the NSCl molecule under the symmetry species A' as follows:

\[
S_1 = \Delta r_{13'}, \quad S_2 = \Delta r_{23'}, \quad S_3 = \Delta r_{12'},
\]

(22)

where \( r_{12} \) corresponds to the 1-2 nonbonding distance at the equilibrium configuration in the 1-3-2 molecular model representing the NSCl molecule, and the other terms are as defined in connection with equation 18. This set of symmetry co-ordinates is directly compatible with the electron diffraction investigation in which the amplitudes of vibration are given in terms of bonding and nonbonding atom pairs and not in terms of angle changes. A set of force constants based on these symmetry co-ordinates could then be directly used in evaluating a set of amplitudes of vibration based on infrared data.

On the basis of the above symmetry coordinates, the \( \tilde{G} \) matrix elements are (64, p. 54-71):
\[ G_{11} = \mu_1 + \mu_3 \]
\[ G_{12} = G_{21} = \mu_3 \cos \alpha \]
\[ G_{13} = G_{31} = \mu_1 \cos \beta \]
\[ G_{22} = \mu_2 + \mu_3 \]
\[ G_{23} = G_{32} = \mu_2 \cos \gamma \]
\[ G_{33} = \mu_1 + \mu_2 \]

where \( \alpha, \beta, \) and \( \gamma \) are the 1-3-2, 2-1-3, and 3-2-1 angles respectively, and the other terms are as defined in connection with equation 19.

In terms of the above symmetry coordinates, the general quadratic potential function is (64, p. 169-182):

\[
2V = F'_{r_1 r_3} (\Delta r_{13})^2 + F'_{r_2 r_3} (\Delta r_{23})^2 + F'_{r_1 r_2} (\Delta r_{12})^2 \\
+ 2F'_{r_1 r_3 r_2} (\Delta r_{13})(\Delta r_{23}) + 2F'_{r_1 r_2 r_3} (\Delta r_{13})(\Delta r_{12}) \\
+ 2F_{r_2 r_3 r_{12}} (\Delta r_{23})(\Delta r_{12}),
\]

where \( F'_{r_1 r_3} \), \( F'_{r_2 r_3} \), and \( F'_{r_1 r_2} \) are the 1-3, 2-3, and 1-2 stretching force constants respectively, and all the others are the interaction constants involving the respective parameters. Using the above potential function, the \( F' \) matrix elements are:
The $F$ and $G$ matrix elements were used to write the secular equation in the form $|FG-\lambda E| = 0$, and an attempt was made to solve the equation for the force constants with the assumption of a diagonal $F$ matrix as in the previous case. However, no solution was found, and since not enough frequency parameters were available to solve the secular equation for more than three force constants, the attempt to find a direct solution was abandoned. Apparently, the diagonal $F$ matrix was not a valid approximation in this case.

In order to obtain a set of force constants based on the symmetry co-ordinates of equation 22 equivalent to the set of diagonal force constants based on the symmetry co-ordinates of equation 18, a co-ordinate transformation was done. The two sets of force constants are connected through the following relationship based on the geometry of the NSCl molecule:

$$r_{12}(\Delta r_{12}) = r_{13}(\Delta r_{13}) + r_{23}(\Delta r_{23}) + r_{13}r_{23}\sin\alpha(\Delta\alpha) - \cos\alpha[r_{13}(\Delta r_{23}) + r_{23}(\Delta r_{13})].$$

(27)
which upon rearrangement gives

\[ \Delta a = \frac{A(\Delta r_{13})}{\sin a} + \frac{B(\Delta r_{23})}{\sin a} + \frac{C(\Delta r_{12})}{\sin a} \]  

(28)

where

\[
A = \begin{bmatrix}
\cos a & -1 \\
r_{13} & r_{23}
\end{bmatrix}
\]

\[
B = \begin{bmatrix}
\cos a & -1 \\
r_{23} & r_{13}
\end{bmatrix}
\]

\[
C = \begin{bmatrix}
r_{12} \\
r_{13}r_{23}
\end{bmatrix}
\]

Combining equations 20, 24, and 28, keeping in mind that the interaction force constants of equation 20 were set to zero, the following equations, relating one set of force constants to the other, can be derived:

\[
F'_{r_{13}} = F_{r_{13}} + F_a \left[ \frac{A^2r_{13}r_{23}}{\sin a} \right]
\]

\[
F'_{r_{23}} = F_{r_{23}} + F_a \left[ \frac{B^2r_{13}r_{23}}{\sin a} \right]
\]

\[
F_{r_{12}} = F_a \left[ \frac{C^2r_{13}r_{23}}{\sin a} \right]
\]

\[
F'_{r_{13}r_{23}} = F_a \left[ \frac{ABr_{13}r_{23}}{\sin a} \right]
\]

\[
F'_{r_{13}r_{12}} = F_a \left[ \frac{AGr_{13}r_{23}}{\sin a} \right]
\]

\[
F'_{r_{23}r_{12}} = F_a \left[ \frac{BG r_{13}r_{23}}{\sin a} \right]
\]  

(29)
The six force constants based on the symmetry co-ordinates of equation 22 were calculated using these above relationships and are given in column four of Table 25. They represent an equivalent set to the ones listed in column 2 of Table 25, which were based on the symmetry co-ordinates of equation 18.

**Force Constants Based on Infrared and Electron Diffraction Data**

Amplitudes of vibration were used in conjunction with the fundamental frequencies to calculate force constants for NSCl from the secular equation \[ \sum F - \epsilon F = 0 \] (15, p. 89-100). The symmetry co-ordinates used for the basis of the calculation are those given by equation 22. These are directly compatible with the amplitudes of vibration obtained from the electron diffraction investigation. The potential function and \( F \) matrix elements based on these symmetry co-ordinates are given by equations 24 and 25 respectively. Using those symmetry co-ordinates, the mean-square amplitude matrix elements are:
$$\Sigma_{11} = <S_1^2> = \frac{1^2}{r_{13}}$$

$$\Sigma_{22} = <S_2^2> = \frac{1^2}{r_{23}}$$

$$\Sigma_{33} = <S_3^2> = \frac{1^2}{r_{12}}$$

$$\Sigma_{12} = <S_1 S_2> = \frac{1}{r_{13} r_{23}}$$

$$\Sigma_{13} = <S_1 S_3> = \frac{1}{r_{13} r_{12}}$$

$$\Sigma_{23} = <S_2 S_3> = \frac{1}{r_{23} r_{12}}$$

The \( \Sigma \) and \( F \) matrix elements were then used to write the
secular equation in the form

$$|\Sigma F - \epsilon \Sigma| = 0,$$

where the values of \( \epsilon \)
are connected with the fundamental frequencies, \( \nu_i \) in wave-numbers,
by the relation

$$\epsilon_i = \frac{\hbar c \nu_i}{2} \coth\left( \frac{\hbar c \nu_i}{2kT} \right),$$

where \( \hbar \) is Plank's constant, \( k \) is Boltzmann's constant, and \( T \) is
absolute temperature. Force constants were calculated using ampli-
tudes of vibration from Least Squares I and II electron diffraction
results. The calculations were made with the assumption of diagonal
\( F \) and \( \Sigma \) matrices since only three amplitudes of vibration are
obtained from the electron diffraction investigation. The values of the
molecular parameters and other constants used in the calculations
are found in Tables 18 and 24 respectively. The calculated values of
the force constants of NSCl are given in the fifth and sixth columns of Table 25 and agree well with those calculated from infrared data via the co-ordinate transformation (column four).

**Amplitudes of Vibration Based on Infrared Data**

In the previous section, amplitudes of vibration obtained from the electron diffraction investigation were used to calculate force constants on the basis of the secular equation \( \sum F - \varepsilon E = 0 \). In this section, the reverse process is done, i.e., amplitudes of vibration are calculated from force constants based on infrared data and then compared to those obtained from the electron diffraction investigation.

The symmetry co-ordinates used as the basis for this calculation, the potential function and \( F \) matrix elements based on these symmetry co-ordinates, and the mean-square amplitude matrix elements are the same as those used in the previous section and are given by equations 22, 24, 25, and 30 respectively.

The \( \Sigma \) and \( F \) matrix elements were used to write the secular equation in the form \( \sum F - \varepsilon E = 0 \) as before, and the amplitudes of vibration were calculated using the force constants listed in column four of Table 25. The calculations were made with the assumption of diagonal \( F \) and \( \Sigma \) matrices. The calculated values of the root-mean-square amplitudes of vibration \((l_{ij})\) for NSCl at 368°K are given in column two of Table 26. The temperature value of 368°K was used in
the calculations to coincide with the temperature of the electron dif-
fraction experiment so that a direct comparison of the amplitudes of
vibration could be made. The amplitudes of vibration obtained from
the electron diffraction investigation are given in columns three and
four of Table 26 and agree well with those calculated by the above
method.
Table 1. Molecular structure, fundamental frequencies ($\nu$), and root-mean-square amplitudes of vibration ($l_{ij}$) for NSF.

<table>
<thead>
<tr>
<th>Fundamental Frequencies (cm$^{-1}$)</th>
<th>Molecular Structure (distances are in Å)</th>
<th>Root-Mean-Square Amplitudes of Vibration ($\bar{x}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\nu_1 = 1372$</td>
<td>$d_{NS} = 1.446 \pm 0.01$</td>
<td>$l_{NS} = 0.0379$</td>
</tr>
<tr>
<td>$\nu_2 = 640$</td>
<td>$d_{SF} = 1.646 \pm 0.01$</td>
<td>$l_{SF} = 0.0468$</td>
</tr>
<tr>
<td>$\nu_3 = 366$</td>
<td>$d_{NF} = 2.637^a$</td>
<td>$l_{NF} = 0.0704$</td>
</tr>
<tr>
<td></td>
<td>$\angle NSF = 116^052' \pm 15'$</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$\angle NSF = 116^055' \pm 2'$</td>
<td></td>
</tr>
</tbody>
</table>

Ref: (46) (37) (12) (45)

$^a$ Calculated from bond distances and bond angles given.

Table 2. Force constants of NSF.$^a$

<table>
<thead>
<tr>
<th>Force Constant</th>
<th>Microwave Data</th>
<th>Infrared Data</th>
<th>Microwave and Infrared Data</th>
<th>Fadini method$^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f_{NS}$</td>
<td>10.343</td>
<td>10.704</td>
<td>10.703</td>
<td>10.721</td>
</tr>
<tr>
<td>$f_{SF}$</td>
<td>3.017</td>
<td>2.853</td>
<td>2.872</td>
<td>2.884</td>
</tr>
<tr>
<td>$f_{a/d^c}$</td>
<td>0.416</td>
<td>0.414</td>
<td>0.411</td>
<td>0.410</td>
</tr>
<tr>
<td>Ref:</td>
<td>(12)</td>
<td>(12, 43, 46)</td>
<td>(12, 43)</td>
<td>(50)</td>
</tr>
</tbody>
</table>

$^a$ Force constants in millidynes per angstrom.

$^b$ A method which permits calculation of a complete set of force constants using only the vibrational frequencies and the geometry of the molecule.

$^c$ $d$ is the geometric mean of the NS and SF distances; thus $d^2 = (d_{NS})(d_{SF})$. $\alpha$ is the NSF bond angle.
Table 3. Observed fundamental frequencies of NSCl (44).

<table>
<thead>
<tr>
<th>Symmetry</th>
<th>Species</th>
<th>Number</th>
<th>Mode</th>
<th>Frequency (cm⁻¹)</th>
<th>P-R Separation (cm⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A'</td>
<td>ν₁</td>
<td></td>
<td>NS stretch</td>
<td>1325</td>
<td>15</td>
</tr>
<tr>
<td>A'</td>
<td>ν₂</td>
<td></td>
<td>SC1 stretch</td>
<td>414</td>
<td>16</td>
</tr>
<tr>
<td>A'</td>
<td>ν₃</td>
<td></td>
<td>NSCl bend</td>
<td>273ᵃ</td>
<td>--</td>
</tr>
</tbody>
</table>

ᵃThis frequency was not seen directly but was obtained from the observed combination band ν₂ + ν₃ = 687 cm⁻¹ and the overtone 2ν₃ = 545 cm⁻¹.

Table 4. Force constants in millidynes per angstrom and mean amplitudes of vibration in angstroms at room temperature for NSCl (44).

<table>
<thead>
<tr>
<th>Force Constant</th>
<th>(a)</th>
<th>(b)</th>
<th>Mean Amplitudes of Vibration</th>
</tr>
</thead>
<tbody>
<tr>
<td>fₙS</td>
<td>10.03</td>
<td>10.02</td>
<td>lₙS = 0.0392</td>
</tr>
<tr>
<td>fₚSCl</td>
<td>1.67</td>
<td>1.64</td>
<td>lₚSCl = 0.0539</td>
</tr>
<tr>
<td>fₚa</td>
<td>0.22</td>
<td>0.25</td>
<td>lₚNC1 = 0.0847</td>
</tr>
<tr>
<td>fₚNS/SCl</td>
<td>~0ᶜ</td>
<td>---</td>
<td></td>
</tr>
<tr>
<td>fₚNS/a</td>
<td>~0</td>
<td>---</td>
<td></td>
</tr>
<tr>
<td>fₚSCl/a</td>
<td>~0</td>
<td>---</td>
<td></td>
</tr>
</tbody>
</table>

ᵃFrom Fadini's method (50).
ᵇCalculated with a diagonal F-matrix.
ᶜThe calculated force constants were too small to be considered as significant.
Table 5. Data for electron diffraction photographs used for the structure determination of gaseous thiazyl chloride, NSCl.

<table>
<thead>
<tr>
<th>Plate Identification</th>
<th>Plate Size</th>
<th>Accelerating voltage (volts)</th>
<th>Wavelength (Å)</th>
<th>Exposure Times (sec)</th>
<th>Beam current (µ a)</th>
<th>Nozzle Temp (°C)</th>
<th>Bath Temp (°C)</th>
<th>Run-in pressure (mm Hg)</th>
<th>Nozzle-to-plate distance (mm)</th>
<th>s range</th>
</tr>
</thead>
<tbody>
<tr>
<td>I-231-02</td>
<td>8&quot;x10&quot;</td>
<td>44320</td>
<td>.057031</td>
<td>60</td>
<td>0.22</td>
<td>100</td>
<td>67</td>
<td>5.0x10^-6</td>
<td>749.38</td>
<td>1.00 - 12.75</td>
</tr>
<tr>
<td>I-231-03</td>
<td>8&quot;x10&quot;</td>
<td>44320</td>
<td>.057031</td>
<td>45</td>
<td>0.22</td>
<td>100</td>
<td>65</td>
<td>4.8x10^-6</td>
<td>749.38</td>
<td>1.00 - 12.75</td>
</tr>
<tr>
<td>I-230-09</td>
<td>8&quot;x10&quot;</td>
<td>44329</td>
<td>.057025</td>
<td>90</td>
<td>0.30</td>
<td>92</td>
<td>68</td>
<td>7.0x10^-6</td>
<td>299.40</td>
<td>7.00 - 29.75</td>
</tr>
<tr>
<td>I-230-10</td>
<td>8&quot;x10&quot;</td>
<td>44330</td>
<td>.057024</td>
<td>60</td>
<td>0.30</td>
<td>93</td>
<td>76</td>
<td>7.0x10^-6</td>
<td>299.40</td>
<td>7.00 - 29.75</td>
</tr>
<tr>
<td>I-233-06</td>
<td>5&quot;x7&quot;</td>
<td>44316</td>
<td>.057033</td>
<td>120</td>
<td>0.30</td>
<td>95</td>
<td>77</td>
<td>6.0x10^-6</td>
<td>120.19</td>
<td>24.00 - 48.75</td>
</tr>
<tr>
<td>I-233-07</td>
<td>5&quot;x7&quot;</td>
<td>44316</td>
<td>.057033</td>
<td>180</td>
<td>0.30</td>
<td>92</td>
<td>73</td>
<td>5.0x10^-6</td>
<td>120.19</td>
<td>24.00 - 48.75</td>
</tr>
<tr>
<td>I-233-08</td>
<td>5&quot;x7&quot;</td>
<td>44318</td>
<td>.057032</td>
<td>240</td>
<td>0.30</td>
<td>92</td>
<td>78</td>
<td>6.0x10^-6</td>
<td>120.19</td>
<td>24.00 - 48.75</td>
</tr>
</tbody>
</table>

*a Kodak process plates

*b Wave-lengths were determined from the accelerating voltage which was calibrated against diffraction patterns of gaseous CO₂ (26).
Table 6. Experimental intensity curve ($s_{\text{II}}^m$) for NSCl. Data from long camera distance (75 cm) plate I-231-02.

<table>
<thead>
<tr>
<th>$s$</th>
<th>$\Delta s$</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td>9.8363</td>
<td>12.2251</td>
<td>4.2155</td>
<td>-19.6725</td>
<td></td>
</tr>
<tr>
<td>2.00</td>
<td>-47.7761</td>
<td>-69.5564</td>
<td>-80.7978</td>
<td>-81.0789</td>
<td></td>
</tr>
<tr>
<td>3.00</td>
<td>-75.8797</td>
<td>-54.8020</td>
<td>-19.6725</td>
<td>26.2769</td>
<td></td>
</tr>
<tr>
<td>4.00</td>
<td>70.2590</td>
<td>101.4540</td>
<td>82.0300</td>
<td>3.2319</td>
<td></td>
</tr>
<tr>
<td>5.00</td>
<td>-77.2849</td>
<td>-125.3421</td>
<td>-69.5564</td>
<td>72.6478</td>
<td></td>
</tr>
<tr>
<td>6.00</td>
<td>244.5013</td>
<td>403.9892</td>
<td>438.4162</td>
<td>303.5189</td>
<td></td>
</tr>
<tr>
<td>7.00</td>
<td>9.8363</td>
<td>-346.3769</td>
<td>-674.4864</td>
<td>-816.6906</td>
<td></td>
</tr>
<tr>
<td>8.00</td>
<td>-753.1765</td>
<td>-510.0803</td>
<td>-179.1605</td>
<td>122.9532</td>
<td></td>
</tr>
<tr>
<td>9.00</td>
<td>404.6918</td>
<td>545.9124</td>
<td>587.3652</td>
<td>575.4212</td>
<td></td>
</tr>
<tr>
<td>10.00</td>
<td>435.6058</td>
<td>259.2557</td>
<td>44.2632</td>
<td>-226.5150</td>
<td></td>
</tr>
<tr>
<td>11.00</td>
<td>-432.7954</td>
<td>-537.4814</td>
<td>-484.7871</td>
<td>-231.1521</td>
<td></td>
</tr>
<tr>
<td>12.00</td>
<td>50.5865</td>
<td>326.9854</td>
<td>456.6835</td>
<td>376.1667</td>
<td></td>
</tr>
</tbody>
</table>

Table 7. Experimental intensity curve ($s_{\text{I}}^m$) for NSCl. Data from long camera distance (75 cm) plate I-231-03.

<table>
<thead>
<tr>
<th>$s$</th>
<th>$\Delta s$</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td>9.3095</td>
<td>11.6369</td>
<td>4.6548</td>
<td>-18.9294</td>
<td></td>
</tr>
<tr>
<td>2.00</td>
<td>-46.5477</td>
<td>-66.2529</td>
<td>-81.4585</td>
<td>-76.8037</td>
<td></td>
</tr>
<tr>
<td>3.00</td>
<td>-69.8216</td>
<td>-55.3918</td>
<td>-27.1528</td>
<td>29.0147</td>
<td></td>
</tr>
<tr>
<td>4.00</td>
<td>74.4763</td>
<td>105.5081</td>
<td>83.7859</td>
<td>3.5687</td>
<td></td>
</tr>
<tr>
<td>5.00</td>
<td>-69.8216</td>
<td>-114.0419</td>
<td>-72.4593</td>
<td>71.3731</td>
<td></td>
</tr>
<tr>
<td>6.00</td>
<td>251.3576</td>
<td>407.2924</td>
<td>433.6694</td>
<td>282.6997</td>
<td></td>
</tr>
<tr>
<td>7.00</td>
<td>0.0000</td>
<td>-371.1403</td>
<td>-663.3047</td>
<td>-817.6879</td>
<td></td>
</tr>
<tr>
<td>8.00</td>
<td>-744.7632</td>
<td>-524.7477</td>
<td>-211.0162</td>
<td>122.1101</td>
<td></td>
</tr>
<tr>
<td>9.00</td>
<td>363.0721</td>
<td>516.6795</td>
<td>574.8641</td>
<td>544.6081</td>
<td></td>
</tr>
<tr>
<td>10.00</td>
<td>434.4452</td>
<td>286.2684</td>
<td>40.6517</td>
<td>-233.5143</td>
<td></td>
</tr>
<tr>
<td>11.00</td>
<td>-443.7547</td>
<td>-575.9502</td>
<td>-517.4553</td>
<td>-273.3902</td>
<td></td>
</tr>
<tr>
<td>12.00</td>
<td>46.5477</td>
<td>323.0410</td>
<td>426.6873</td>
<td>375.7951</td>
<td></td>
</tr>
</tbody>
</table>
Table 8. Experimental intensity curve (\( s_{m}^{\lambda} \)) for NSCl. Data from middle camera distance (30 cm) plate I-230-09.

<table>
<thead>
<tr>
<th>s</th>
<th>( \Delta s )</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.00</td>
<td>216.2154</td>
<td>-103.4032</td>
<td>-411.4075</td>
<td>-602.2902</td>
<td></td>
</tr>
<tr>
<td>8.00</td>
<td>-600.1115</td>
<td>-418.3969</td>
<td>-131.0290</td>
<td>156.4264</td>
<td></td>
</tr>
<tr>
<td>9.00</td>
<td>392.6637</td>
<td>538.1130</td>
<td>584.9139</td>
<td>540.6252</td>
<td></td>
</tr>
<tr>
<td>10.00</td>
<td>364.5070</td>
<td>157.2874</td>
<td>-69.1348</td>
<td>-348.9097</td>
<td></td>
</tr>
<tr>
<td>11.00</td>
<td>-583.0614</td>
<td>-700.5636</td>
<td>-620.3649</td>
<td>-407.6146</td>
<td></td>
</tr>
<tr>
<td>12.00</td>
<td>-135.0286</td>
<td>132.5994</td>
<td>306.0927</td>
<td>258.7077</td>
<td></td>
</tr>
<tr>
<td>14.00</td>
<td>-233.8064</td>
<td>15.8696</td>
<td>287.8237</td>
<td>472.5022</td>
<td></td>
</tr>
<tr>
<td>15.00</td>
<td>506.9730</td>
<td>364.7399</td>
<td>105.7816</td>
<td>-213.8052</td>
<td></td>
</tr>
<tr>
<td>16.00</td>
<td>-491.6721</td>
<td>-677.8357</td>
<td>-754.2790</td>
<td>-684.8815</td>
<td></td>
</tr>
<tr>
<td>17.00</td>
<td>-534.4729</td>
<td>-252.9699</td>
<td>88.0238</td>
<td>489.2954</td>
<td></td>
</tr>
<tr>
<td>18.00</td>
<td>761.1943</td>
<td>968.4936</td>
<td>936.9073</td>
<td>673.1465</td>
<td></td>
</tr>
<tr>
<td>19.00</td>
<td>308.0240</td>
<td>-145.9294</td>
<td>-495.2751</td>
<td>-690.0198</td>
<td></td>
</tr>
<tr>
<td>20.00</td>
<td>-609.1937</td>
<td>-451.4150</td>
<td>-179.9782</td>
<td>50.3705</td>
<td></td>
</tr>
<tr>
<td>21.00</td>
<td>149.2576</td>
<td>184.3674</td>
<td>107.8142</td>
<td>-1.7468</td>
<td></td>
</tr>
<tr>
<td>22.00</td>
<td>-81.3379</td>
<td>-99.3948</td>
<td>-78.7105</td>
<td>-47.2689</td>
<td></td>
</tr>
<tr>
<td>23.00</td>
<td>24.9792</td>
<td>113.1655</td>
<td>218.5155</td>
<td>243.1521</td>
<td></td>
</tr>
<tr>
<td>24.00</td>
<td>111.3561</td>
<td>24.6086</td>
<td>-170.4234</td>
<td>-416.4847</td>
<td></td>
</tr>
<tr>
<td>25.00</td>
<td>-575.7204</td>
<td>-726.5292</td>
<td>-614.6915</td>
<td>-466.9343</td>
<td></td>
</tr>
<tr>
<td>26.00</td>
<td>-179.3923</td>
<td>116.1762</td>
<td>403.0398</td>
<td>558.4274</td>
<td></td>
</tr>
<tr>
<td>27.00</td>
<td>656.0200</td>
<td>565.0326</td>
<td>368.5350</td>
<td>144.1261</td>
<td></td>
</tr>
<tr>
<td>28.00</td>
<td>-83.8392</td>
<td>-307.9250</td>
<td>-315.7559</td>
<td>-370.8099</td>
<td></td>
</tr>
<tr>
<td>29.00</td>
<td>-261.3404</td>
<td>-147.8698</td>
<td>2.2118</td>
<td>73.9221</td>
<td></td>
</tr>
</tbody>
</table>
Table 9. Experimental intensity curve ($\text{sI}_m$) for NSCl. Data from middle camera distance (30 cm) plate 1-230-10.

<table>
<thead>
<tr>
<th>s</th>
<th>$\Delta s$</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.00</td>
<td></td>
<td>231.8241</td>
<td>-100.5844</td>
<td>-424.9727</td>
<td>-611.3612</td>
</tr>
<tr>
<td>8.00</td>
<td></td>
<td>-597.6898</td>
<td>-418.2957</td>
<td>-106.4612</td>
<td>178.9241</td>
</tr>
<tr>
<td>9.00</td>
<td></td>
<td>418.1216</td>
<td>578.5358</td>
<td>609.4167</td>
<td>565.9083</td>
</tr>
<tr>
<td>10.00</td>
<td></td>
<td>395.0657</td>
<td>194.8259</td>
<td>-33.8031</td>
<td>-326.6716</td>
</tr>
<tr>
<td>11.00</td>
<td></td>
<td>-547.6007</td>
<td>-628.4030</td>
<td>-549.4247</td>
<td>-366.4410</td>
</tr>
<tr>
<td>12.00</td>
<td></td>
<td>-115.1268</td>
<td>113.4977</td>
<td>269.1020</td>
<td>237.3419</td>
</tr>
<tr>
<td>13.00</td>
<td></td>
<td>49.2305</td>
<td>-175.5135</td>
<td>-372.5549</td>
<td>-364.4157</td>
</tr>
<tr>
<td>14.00</td>
<td></td>
<td>-233.0234</td>
<td>24.9584</td>
<td>290.1236</td>
<td>438.3141</td>
</tr>
<tr>
<td>15.00</td>
<td></td>
<td>497.1776</td>
<td>388.9995</td>
<td>105.9081</td>
<td>-197.5969</td>
</tr>
<tr>
<td>16.00</td>
<td></td>
<td>-456.8179</td>
<td>-633.0015</td>
<td>-730.7802</td>
<td>-692.4587</td>
</tr>
<tr>
<td>17.00</td>
<td></td>
<td>-499.9437</td>
<td>-254.3550</td>
<td>146.9183</td>
<td>505.2234</td>
</tr>
<tr>
<td>18.00</td>
<td></td>
<td>810.1884</td>
<td>989.8190</td>
<td>990.9933</td>
<td>737.2742</td>
</tr>
<tr>
<td>19.00</td>
<td></td>
<td>348.0379</td>
<td>-103.1428</td>
<td>-435.7809</td>
<td>-664.5599</td>
</tr>
<tr>
<td>20.00</td>
<td></td>
<td>-546.6328</td>
<td>-410.6701</td>
<td>-194.0339</td>
<td>41.3266</td>
</tr>
<tr>
<td>21.00</td>
<td></td>
<td>190.7635</td>
<td>148.9817</td>
<td>162.0653</td>
<td>0.2705</td>
</tr>
<tr>
<td>22.00</td>
<td></td>
<td>-7.5709</td>
<td>-20.4206</td>
<td>1.7644</td>
<td>8.2759</td>
</tr>
<tr>
<td>23.00</td>
<td></td>
<td>83.3774</td>
<td>219.6879</td>
<td>351.1880</td>
<td>313.8336</td>
</tr>
<tr>
<td>24.00</td>
<td></td>
<td>223.6315</td>
<td>119.8615</td>
<td>-30.9945</td>
<td>-257.2991</td>
</tr>
<tr>
<td>25.00</td>
<td></td>
<td>-429.4381</td>
<td>-513.6460</td>
<td>-549.1041</td>
<td>-347.4072</td>
</tr>
<tr>
<td>26.00</td>
<td></td>
<td>9.9524</td>
<td>346.0342</td>
<td>635.0112</td>
<td>775.5355</td>
</tr>
<tr>
<td>27.00</td>
<td></td>
<td>700.7578</td>
<td>628.6517</td>
<td>452.6084</td>
<td>211.6478</td>
</tr>
<tr>
<td>28.00</td>
<td></td>
<td>-15.0453</td>
<td>-93.4909</td>
<td>-238.6555</td>
<td>-314.2651</td>
</tr>
<tr>
<td>29.00</td>
<td></td>
<td>-210.3623</td>
<td>-97.4730</td>
<td>48.7912</td>
<td>121.8047</td>
</tr>
</tbody>
</table>
Table 10. Experimental intensity curve ($s_{m}^1$) for NSC1. Data from short camera distance (12 cm) plate 1-233-06.

<table>
<thead>
<tr>
<th>s</th>
<th>△s</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>24.00</td>
<td>288.4109</td>
<td>178.1105</td>
<td>-43.4139</td>
<td>-255.3512</td>
<td></td>
</tr>
<tr>
<td>25.00</td>
<td>-424.4020</td>
<td>-706.7280</td>
<td>-690.4963</td>
<td>-550.1820</td>
<td></td>
</tr>
<tr>
<td>26.00</td>
<td>-351.3539</td>
<td>-76.9126</td>
<td>279.3139</td>
<td>559.2963</td>
<td></td>
</tr>
<tr>
<td>27.00</td>
<td>504.0332</td>
<td>415.7299</td>
<td>267.1813</td>
<td>17.1987</td>
<td></td>
</tr>
<tr>
<td>28.00</td>
<td>-246.1102</td>
<td>-361.3254</td>
<td>-432.0044</td>
<td>-467.1991</td>
<td></td>
</tr>
<tr>
<td>29.00</td>
<td>-338.1775</td>
<td>-126.5955</td>
<td>23.6847</td>
<td>224.2233</td>
<td></td>
</tr>
<tr>
<td>30.00</td>
<td>273.2227</td>
<td>284.9574</td>
<td>234.3292</td>
<td>147.0351</td>
<td></td>
</tr>
<tr>
<td>31.00</td>
<td>91.1980</td>
<td>53.0879</td>
<td>165.4893</td>
<td>243.2991</td>
<td></td>
</tr>
<tr>
<td>32.00</td>
<td>263.5692</td>
<td>221.1635</td>
<td>240.3589</td>
<td>257.4066</td>
<td></td>
</tr>
<tr>
<td>33.00</td>
<td>68.6190</td>
<td>-132.8961</td>
<td>-327.7178</td>
<td>-361.2183</td>
<td></td>
</tr>
<tr>
<td>34.00</td>
<td>-370.7353</td>
<td>-427.8930</td>
<td>-399.3321</td>
<td>-125.4326</td>
<td></td>
</tr>
<tr>
<td>35.00</td>
<td>164.7882</td>
<td>295.1367</td>
<td>373.5674</td>
<td>433.3008</td>
<td></td>
</tr>
<tr>
<td>36.00</td>
<td>373.7748</td>
<td>259.1249</td>
<td>165.7297</td>
<td>43.1329</td>
<td></td>
</tr>
<tr>
<td>37.00</td>
<td>-74.1525</td>
<td>-115.0157</td>
<td>-312.3383</td>
<td>-42.7982</td>
<td></td>
</tr>
<tr>
<td>38.00</td>
<td>-15.1971</td>
<td>-90.5181</td>
<td>18.3465</td>
<td>58.0285</td>
<td></td>
</tr>
<tr>
<td>39.00</td>
<td>22.9598</td>
<td>102.7245</td>
<td>88.9907</td>
<td>181.4200</td>
<td></td>
</tr>
<tr>
<td>40.00</td>
<td>135.0730</td>
<td>108.1564</td>
<td>186.5925</td>
<td>132.5622</td>
<td></td>
</tr>
<tr>
<td>41.00</td>
<td>26.4369</td>
<td>8.1465</td>
<td>-37.5361</td>
<td>20.4683</td>
<td></td>
</tr>
<tr>
<td>42.00</td>
<td>-82.5941</td>
<td>-128.2143</td>
<td>-215.2743</td>
<td>-198.7076</td>
<td></td>
</tr>
<tr>
<td>43.00</td>
<td>-14.0231</td>
<td>-84.3722</td>
<td>-17.2583</td>
<td>153.2758</td>
<td></td>
</tr>
<tr>
<td>44.00</td>
<td>113.1448</td>
<td>119.3594</td>
<td>209.7966</td>
<td>101.4515</td>
<td></td>
</tr>
<tr>
<td>45.00</td>
<td>74.7594</td>
<td>86.2104</td>
<td>46.9652</td>
<td>-149.6009</td>
<td></td>
</tr>
<tr>
<td>46.00</td>
<td>-109.6216</td>
<td>-78.7242</td>
<td>-174.8336</td>
<td>-22.6331</td>
<td></td>
</tr>
<tr>
<td>47.00</td>
<td>-288.8509</td>
<td>-111.2679</td>
<td>-79.9546</td>
<td>-152.4170</td>
<td></td>
</tr>
<tr>
<td>48.00</td>
<td>-60.4279</td>
<td>59.1225</td>
<td>70.9315</td>
<td>-24.1003</td>
<td></td>
</tr>
</tbody>
</table>
Table 11. Experimental intensity curve (\(s_1^m\)) for NSCl. Data from short camera distance (12 cm) plate I-233-07.

<table>
<thead>
<tr>
<th>s</th>
<th>(\Delta s)</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>24.00</td>
<td>344.6322</td>
<td>176.1005</td>
<td>-73.3802</td>
<td>-272.8735</td>
<td></td>
</tr>
<tr>
<td>25.00</td>
<td>-460.2488</td>
<td>-558.0400</td>
<td>-508.7122</td>
<td>-346.8932</td>
<td></td>
</tr>
<tr>
<td>26.00</td>
<td>-135.0164</td>
<td>183.9502</td>
<td>486.9378</td>
<td>665.6710</td>
<td></td>
</tr>
<tr>
<td>27.00</td>
<td>719.6191</td>
<td>668.9443</td>
<td>443.5117</td>
<td>117.3905</td>
<td></td>
</tr>
<tr>
<td>28.00</td>
<td>-324.8574</td>
<td>-545.6838</td>
<td>-494.0370</td>
<td>-421.4402</td>
<td></td>
</tr>
<tr>
<td>29.00</td>
<td>-414.3676</td>
<td>-273.1786</td>
<td>-50.3600</td>
<td>137.0512</td>
<td></td>
</tr>
<tr>
<td>30.00</td>
<td>236.6428</td>
<td>213.0683</td>
<td>104.2190</td>
<td>111.3041</td>
<td></td>
</tr>
<tr>
<td>31.00</td>
<td>193.0744</td>
<td>219.3918</td>
<td>219.5816</td>
<td>298.3129</td>
<td></td>
</tr>
<tr>
<td>32.00</td>
<td>434.7935</td>
<td>439.9163</td>
<td>406.8714</td>
<td>369.6513</td>
<td></td>
</tr>
<tr>
<td>33.00</td>
<td>248.4417</td>
<td>-42.1930</td>
<td>-354.7495</td>
<td>-496.0766</td>
<td></td>
</tr>
<tr>
<td>34.00</td>
<td>-617.4645</td>
<td>-531.9165</td>
<td>-492.4744</td>
<td>-193.1120</td>
<td></td>
</tr>
<tr>
<td>35.00</td>
<td>59.9547</td>
<td>201.5346</td>
<td>336.3066</td>
<td>392.6258</td>
<td></td>
</tr>
<tr>
<td>36.00</td>
<td>597.3074</td>
<td>451.1116</td>
<td>273.7439</td>
<td>162.2780</td>
<td></td>
</tr>
<tr>
<td>37.00</td>
<td>-115.8698</td>
<td>-147.2189</td>
<td>-92.6857</td>
<td>-410.2557</td>
<td></td>
</tr>
<tr>
<td>38.00</td>
<td>-106.5596</td>
<td>-25.2976</td>
<td>14.1294</td>
<td>81.5062</td>
<td></td>
</tr>
<tr>
<td>39.00</td>
<td>155.6882</td>
<td>233.3519</td>
<td>249.9958</td>
<td>210.6395</td>
<td></td>
</tr>
<tr>
<td>40.00</td>
<td>251.9103</td>
<td>331.3491</td>
<td>291.5882</td>
<td>68.3871</td>
<td></td>
</tr>
<tr>
<td>41.00</td>
<td>-56.9282</td>
<td>56.9801</td>
<td>-1.0478</td>
<td>204.7855</td>
<td></td>
</tr>
<tr>
<td>42.00</td>
<td>45.4504</td>
<td>-212.2241</td>
<td>-195.9573</td>
<td>-275.5696</td>
<td></td>
</tr>
<tr>
<td>43.00</td>
<td>-258.9056</td>
<td>-237.5878</td>
<td>-253.3385</td>
<td>33.0714</td>
<td></td>
</tr>
<tr>
<td>44.00</td>
<td>209.5925</td>
<td>311.5369</td>
<td>52.4320</td>
<td>160.9913</td>
<td></td>
</tr>
<tr>
<td>45.00</td>
<td>292.4007</td>
<td>194.7757</td>
<td>76.7582</td>
<td>-5.8568</td>
<td></td>
</tr>
<tr>
<td>46.00</td>
<td>-134.6139</td>
<td>-57.8823</td>
<td>33.3127</td>
<td>3.1519</td>
<td></td>
</tr>
<tr>
<td>47.00</td>
<td>-3.7510</td>
<td>-274.7302</td>
<td>-141.8010</td>
<td>-28.1339</td>
<td></td>
</tr>
<tr>
<td>48.00</td>
<td>17.2418</td>
<td>-32.9166</td>
<td>56.6782</td>
<td>-135.5594</td>
<td></td>
</tr>
</tbody>
</table>
Table 12. Experimental intensity curve ($s^H_{\mathbf{m}}$) for NSCl. Data from short camera distance (12 cm) plate I-233-08.

<table>
<thead>
<tr>
<th>$s$</th>
<th>$\Delta s$</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>24.00</td>
<td>392.7662</td>
<td>186.1094</td>
<td>-94.3012</td>
<td>-376.4680</td>
<td></td>
</tr>
<tr>
<td>25.00</td>
<td>-474.5942</td>
<td>-567.6409</td>
<td>-626.9846</td>
<td>-428.4127</td>
<td></td>
</tr>
<tr>
<td>26.00</td>
<td>-214.5208</td>
<td>91.1857</td>
<td>343.0302</td>
<td>557.5592</td>
<td></td>
</tr>
<tr>
<td>27.00</td>
<td>678.6653</td>
<td>604.6993</td>
<td>278.0043</td>
<td>-80.1302</td>
<td></td>
</tr>
<tr>
<td>28.00</td>
<td>-359.0530</td>
<td>-542.9135</td>
<td>-539.3501</td>
<td>-397.9545</td>
<td></td>
</tr>
<tr>
<td>29.00</td>
<td>-308.5957</td>
<td>-168.1850</td>
<td>3.5466</td>
<td>132.9408</td>
<td></td>
</tr>
<tr>
<td>30.00</td>
<td>167.2860</td>
<td>128.9816</td>
<td>130.5923</td>
<td>0.7640</td>
<td></td>
</tr>
<tr>
<td>31.00</td>
<td>-1.4062</td>
<td>-10.9817</td>
<td>-63.8405</td>
<td>98.4500</td>
<td></td>
</tr>
<tr>
<td>32.00</td>
<td>269.0158</td>
<td>288.7713</td>
<td>265.3039</td>
<td>213.4436</td>
<td></td>
</tr>
<tr>
<td>33.00</td>
<td>34.5033</td>
<td>-70.2660</td>
<td>-236.2065</td>
<td>-195.4938</td>
<td></td>
</tr>
<tr>
<td>34.00</td>
<td>-268.7058</td>
<td>-351.0467</td>
<td>-202.1818</td>
<td>125.2778</td>
<td></td>
</tr>
<tr>
<td>35.00</td>
<td>163.4032</td>
<td>313.8668</td>
<td>472.7003</td>
<td>516.2609</td>
<td></td>
</tr>
<tr>
<td>36.00</td>
<td>514.4231</td>
<td>381.6925</td>
<td>151.6605</td>
<td>79.2488</td>
<td></td>
</tr>
<tr>
<td>37.00</td>
<td>-78.5297</td>
<td>-122.8198</td>
<td>-114.0525</td>
<td>-62.7220</td>
<td></td>
</tr>
<tr>
<td>38.00</td>
<td>-58.4979</td>
<td>18.4116</td>
<td>19.4282</td>
<td>-38.8483</td>
<td></td>
</tr>
<tr>
<td>39.00</td>
<td>24.6806</td>
<td>-15.4488</td>
<td>-58.0322</td>
<td>37.7306</td>
<td></td>
</tr>
<tr>
<td>40.00</td>
<td>58.4820</td>
<td>36.1345</td>
<td>82.6193</td>
<td>66.3639</td>
<td></td>
</tr>
<tr>
<td>41.00</td>
<td>172.9971</td>
<td>46.1424</td>
<td>-108.9073</td>
<td>-195.6412</td>
<td></td>
</tr>
<tr>
<td>42.00</td>
<td>-85.1787</td>
<td>-125.3392</td>
<td>-138.3580</td>
<td>-294.7576</td>
<td></td>
</tr>
<tr>
<td>43.00</td>
<td>-172.9711</td>
<td>-120.5832</td>
<td>-141.1939</td>
<td>11.8937</td>
<td></td>
</tr>
<tr>
<td>44.00</td>
<td>98.7640</td>
<td>142.2835</td>
<td>136.5052</td>
<td>138.1420</td>
<td></td>
</tr>
<tr>
<td>45.00</td>
<td>75.0927</td>
<td>84.9181</td>
<td>145.7389</td>
<td>-52.3250</td>
<td></td>
</tr>
<tr>
<td>46.00</td>
<td>-232.9122</td>
<td>-156.7744</td>
<td>-114.9432</td>
<td>-83.3681</td>
<td></td>
</tr>
<tr>
<td>47.00</td>
<td>-10.1663</td>
<td>-5.6424</td>
<td>57.1146</td>
<td>6.7584</td>
<td></td>
</tr>
<tr>
<td>48.00</td>
<td>-1.2453</td>
<td>18.4014</td>
<td>55.7991</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 13. Experimental intensity curve ($sI_m^1$) for NSC1. Composite curve of 12, 30, and 75 cm data.

<table>
<thead>
<tr>
<th>$s$</th>
<th>$\Delta s$</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.00</td>
<td></td>
<td>9.5730</td>
<td>11.9311</td>
<td>4.4352</td>
<td>-19.3012</td>
</tr>
<tr>
<td>2.00</td>
<td></td>
<td>-47.1624</td>
<td>-67.9054</td>
<td>-81.1291</td>
<td>-78.9421</td>
</tr>
<tr>
<td>3.00</td>
<td></td>
<td>-72.8514</td>
<td>-55.0975</td>
<td>-23.4130</td>
<td>27.6461</td>
</tr>
<tr>
<td>4.00</td>
<td></td>
<td>72.3685</td>
<td>103.4822</td>
<td>82.9954</td>
<td>3.4003</td>
</tr>
<tr>
<td>5.00</td>
<td></td>
<td>-73.5540</td>
<td>-119.6932</td>
<td>-71.0086</td>
<td>72.0113</td>
</tr>
<tr>
<td>6.00</td>
<td></td>
<td>247.9323</td>
<td>405.6454</td>
<td>436.0476</td>
<td>293.1124</td>
</tr>
<tr>
<td>7.00</td>
<td></td>
<td>114.4713</td>
<td>-230.3793</td>
<td>-543.5509</td>
<td>-712.0183</td>
</tr>
<tr>
<td>8.00</td>
<td></td>
<td>-673.9455</td>
<td>-467.8874</td>
<td>-156.9190</td>
<td>145.1060</td>
</tr>
<tr>
<td>9.00</td>
<td></td>
<td>394.6436</td>
<td>589.1494</td>
<td>556.6495</td>
<td>283.9073</td>
</tr>
<tr>
<td>10.00</td>
<td></td>
<td>407.4124</td>
<td>224.4130</td>
<td>-4.5059</td>
<td>-283.9073</td>
</tr>
<tr>
<td>11.00</td>
<td></td>
<td>-501.8112</td>
<td>-610.6092</td>
<td>-543.0165</td>
<td>-319.6548</td>
</tr>
<tr>
<td>12.00</td>
<td></td>
<td>-38.2562</td>
<td>364.6465</td>
<td>312.0074</td>
<td>312.0074</td>
</tr>
<tr>
<td>13.00</td>
<td></td>
<td>88.2153</td>
<td>-349.4466</td>
<td>-359.9727</td>
<td>145.1060</td>
</tr>
<tr>
<td>14.00</td>
<td></td>
<td>-233.4197</td>
<td>20.4145</td>
<td>288.9796</td>
<td>455.4172</td>
</tr>
<tr>
<td>15.00</td>
<td></td>
<td>502.0855</td>
<td>376.8777</td>
<td>105.8470</td>
<td>205.7051</td>
</tr>
<tr>
<td>16.00</td>
<td></td>
<td>-474.2544</td>
<td>-655.4316</td>
<td>-742.5446</td>
<td>-688.6843</td>
</tr>
<tr>
<td>17.00</td>
<td></td>
<td>-517.2186</td>
<td>-253.6677</td>
<td>117.4741</td>
<td>497.2698</td>
</tr>
<tr>
<td>18.00</td>
<td></td>
<td>785.7080</td>
<td>979.1766</td>
<td>963.9706</td>
<td>705.2255</td>
</tr>
<tr>
<td>19.00</td>
<td></td>
<td>328.0381</td>
<td>-124.5382</td>
<td>-465.5369</td>
<td>-677.3035</td>
</tr>
<tr>
<td>20.00</td>
<td></td>
<td>-577.9245</td>
<td>-431.0510</td>
<td>-187.0100</td>
<td>45.8494</td>
</tr>
<tr>
<td>21.00</td>
<td></td>
<td>170.0145</td>
<td>166.6776</td>
<td>134.9431</td>
<td>45.8494</td>
</tr>
<tr>
<td>22.00</td>
<td></td>
<td>-44.4545</td>
<td>-59.9081</td>
<td>38.4730</td>
<td>-19.4963</td>
</tr>
<tr>
<td>23.00</td>
<td></td>
<td>54.1800</td>
<td>166.4312</td>
<td>284.8590</td>
<td>278.4993</td>
</tr>
<tr>
<td>24.00</td>
<td></td>
<td>254.7690</td>
<td>126.1931</td>
<td>-85.5519</td>
<td>-319.2803</td>
</tr>
<tr>
<td>25.00</td>
<td></td>
<td>-477.8915</td>
<td>-615.4999</td>
<td>-595.3796</td>
<td>-424.5369</td>
</tr>
<tr>
<td>26.00</td>
<td></td>
<td>-159.1834</td>
<td>148.6221</td>
<td>444.4480</td>
<td>630.6512</td>
</tr>
<tr>
<td>27.00</td>
<td></td>
<td>656.3471</td>
<td>580.0762</td>
<td>370.1103</td>
<td>98.0105</td>
</tr>
<tr>
<td>28.00</td>
<td></td>
<td>-179.7738</td>
<td>-342.0857</td>
<td>-391.9054</td>
<td>-385.7425</td>
</tr>
<tr>
<td>29.00</td>
<td></td>
<td>-294.8193</td>
<td>-156.0223</td>
<td>8.8933</td>
<td>131.3084</td>
</tr>
<tr>
<td>30.00</td>
<td></td>
<td>225.7387</td>
<td>209.0071</td>
<td>156.3879</td>
<td>86.3475</td>
</tr>
<tr>
<td>31.00</td>
<td></td>
<td>94.2834</td>
<td>87.1662</td>
<td>107.0378</td>
<td>213.3613</td>
</tr>
<tr>
<td>32.00</td>
<td></td>
<td>322.5304</td>
<td>316.7030</td>
<td>304.2503</td>
<td>280.2159</td>
</tr>
<tr>
<td>33.00</td>
<td></td>
<td>117.2028</td>
<td>-81.7871</td>
<td>-306.2671</td>
<td>-350.9608</td>
</tr>
<tr>
<td>34.00</td>
<td></td>
<td>-419.0309</td>
<td>-437.0266</td>
<td>-364.6889</td>
<td>-64.3679</td>
</tr>
<tr>
<td>35.00</td>
<td></td>
<td>129.4113</td>
<td>270.2451</td>
<td>394.3057</td>
<td>447.5175</td>
</tr>
<tr>
<td>36.00</td>
<td></td>
<td>495.3136</td>
<td>364.0879</td>
<td>197.0833</td>
<td>94.9161</td>
</tr>
<tr>
<td>37.00</td>
<td></td>
<td>-89.5379</td>
<td>-128.3816</td>
<td>-173.0122</td>
<td>-171.9649</td>
</tr>
<tr>
<td>38.00</td>
<td></td>
<td>-60.1089</td>
<td>-32.4464</td>
<td>17.3055</td>
<td>33.5431</td>
</tr>
<tr>
<td>39.00</td>
<td></td>
<td>67.7903</td>
<td>106.8661</td>
<td>93.6306</td>
<td>143.2558</td>
</tr>
<tr>
<td>s</td>
<td>Δs</td>
<td>0.00</td>
<td>0.25</td>
<td>0.50</td>
<td>0.75</td>
</tr>
<tr>
<td>-----</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
<td>-------</td>
</tr>
<tr>
<td>40.00</td>
<td>148.4993</td>
<td>158.5599</td>
<td>186.9454</td>
<td>89.1068</td>
<td></td>
</tr>
<tr>
<td>41.00</td>
<td>47.5530</td>
<td>37.1077</td>
<td>-49.1943</td>
<td>9.8126</td>
<td></td>
</tr>
<tr>
<td>42.00</td>
<td>-40.7851</td>
<td>-155.2918</td>
<td>-183.2163</td>
<td>-256.4268</td>
<td></td>
</tr>
<tr>
<td>43.00</td>
<td>-148.7066</td>
<td>-147.5552</td>
<td>-137.3250</td>
<td>66.0579</td>
<td></td>
</tr>
<tr>
<td>44.00</td>
<td>140.5265</td>
<td>191.1063</td>
<td>132.9224</td>
<td>133.5671</td>
<td></td>
</tr>
<tr>
<td>45.00</td>
<td>147.4480</td>
<td>121.9935</td>
<td>89.8671</td>
<td>-69.2513</td>
<td></td>
</tr>
<tr>
<td>46.00</td>
<td>-159.1175</td>
<td>-97.8366</td>
<td>-85.4928</td>
<td>-34.3074</td>
<td></td>
</tr>
<tr>
<td>47.00</td>
<td>-100.8724</td>
<td>-130.5454</td>
<td>-54.8547</td>
<td>-57.9018</td>
<td></td>
</tr>
<tr>
<td>48.00</td>
<td>-14.7985</td>
<td>7.6308</td>
<td>48.6670</td>
<td>-34.6050</td>
<td></td>
</tr>
</tbody>
</table>
Table 14. Experimental radial distribution curve for NSCl. (Calculated from composite experimental intensity curve with damping factor = 0.0018)

<table>
<thead>
<tr>
<th>r</th>
<th>Δr</th>
<th>0.000</th>
<th>0.025</th>
<th>0.050</th>
<th>0.075</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.000</td>
<td>0</td>
<td>0.8350</td>
<td>0.2157</td>
<td>-2.2999</td>
<td></td>
</tr>
<tr>
<td>0.100</td>
<td>-5.7838</td>
<td>-8.4823</td>
<td>-8.9268</td>
<td>-6.8680</td>
<td></td>
</tr>
<tr>
<td>0.200</td>
<td>-3.3845</td>
<td>-0.0949</td>
<td>1.9348</td>
<td>2.7799</td>
<td></td>
</tr>
<tr>
<td>0.300</td>
<td>3.3564</td>
<td>4.5530</td>
<td>6.5136</td>
<td>8.5639</td>
<td></td>
</tr>
<tr>
<td>0.400</td>
<td>9.7018</td>
<td>9.1732</td>
<td>6.7465</td>
<td>2.6699</td>
<td></td>
</tr>
<tr>
<td>0.500</td>
<td>-2.4551</td>
<td>-7.7271</td>
<td>-12.0003</td>
<td>-14.1095</td>
<td></td>
</tr>
<tr>
<td>0.600</td>
<td>-13.2816</td>
<td>-9.5369</td>
<td>-3.8112</td>
<td>2.3357</td>
<td></td>
</tr>
<tr>
<td>0.700</td>
<td>7.3189</td>
<td>10.1404</td>
<td>10.6823</td>
<td>9.5087</td>
<td></td>
</tr>
<tr>
<td>0.800</td>
<td>7.3181</td>
<td>4.4500</td>
<td>2.7799</td>
<td>9.5087</td>
<td></td>
</tr>
<tr>
<td>0.900</td>
<td>-7.8030</td>
<td>-10.9011</td>
<td>-11.3760</td>
<td>-8.6809</td>
<td></td>
</tr>
<tr>
<td>1.000</td>
<td>-3.5139</td>
<td>2.5342</td>
<td>7.8162</td>
<td>11.3360</td>
<td></td>
</tr>
<tr>
<td>1.100</td>
<td>12.9471</td>
<td>13.1066</td>
<td>12.6442</td>
<td>12.2049</td>
<td></td>
</tr>
<tr>
<td>1.300</td>
<td>29.9494</td>
<td>51.4444</td>
<td>86.0959</td>
<td>131.1207</td>
<td></td>
</tr>
<tr>
<td>1.400</td>
<td>177.3975</td>
<td>212.1844</td>
<td>224.7327</td>
<td>211.5185</td>
<td></td>
</tr>
<tr>
<td>1.500</td>
<td>177.7677</td>
<td>134.4127</td>
<td>92.6710</td>
<td>59.7051</td>
<td></td>
</tr>
<tr>
<td>1.600</td>
<td>37.5004</td>
<td>24.5611</td>
<td>18.3702</td>
<td>16.8291</td>
<td></td>
</tr>
<tr>
<td>1.700</td>
<td>18.3377</td>
<td>21.3358</td>
<td>24.1750</td>
<td>25.4961</td>
<td></td>
</tr>
<tr>
<td>1.800</td>
<td>24.7057</td>
<td>22.1235</td>
<td>18.7097</td>
<td>15.5811</td>
<td></td>
</tr>
<tr>
<td>2.000</td>
<td>40.6533</td>
<td>69.4118</td>
<td>112.3726</td>
<td>166.6318</td>
<td></td>
</tr>
<tr>
<td>2.100</td>
<td>223.4361</td>
<td>270.0293</td>
<td>294.1445</td>
<td>289.0087</td>
<td></td>
</tr>
<tr>
<td>2.200</td>
<td>256.1739</td>
<td>204.6705</td>
<td>147.0877</td>
<td>94.7889</td>
<td></td>
</tr>
<tr>
<td>2.300</td>
<td>54.5893</td>
<td>28.0302</td>
<td>12.8368</td>
<td>5.2393</td>
<td></td>
</tr>
<tr>
<td>2.400</td>
<td>1.8770</td>
<td>6.6689</td>
<td>8.7317</td>
<td>1.8119</td>
<td></td>
</tr>
<tr>
<td>2.500</td>
<td>3.7166</td>
<td>6.0400</td>
<td>8.2318</td>
<td>9.8552</td>
<td></td>
</tr>
<tr>
<td>2.600</td>
<td>10.7965</td>
<td>11.2594</td>
<td>11.5569</td>
<td>11.8719</td>
<td></td>
</tr>
<tr>
<td>2.700</td>
<td>12.1718</td>
<td>12.3163</td>
<td>12.2250</td>
<td>11.9449</td>
<td></td>
</tr>
<tr>
<td>2.800</td>
<td>11.5867</td>
<td>11.2606</td>
<td>11.4883</td>
<td>11.6834</td>
<td></td>
</tr>
<tr>
<td>2.900</td>
<td>13.6284</td>
<td>17.8814</td>
<td>25.0331</td>
<td>34.9537</td>
<td></td>
</tr>
<tr>
<td>3.000</td>
<td>46.6888</td>
<td>58.7195</td>
<td>69.3877</td>
<td>77.2302</td>
<td></td>
</tr>
<tr>
<td>3.100</td>
<td>81.1313</td>
<td>80.4036</td>
<td>74.9263</td>
<td>65.3140</td>
<td></td>
</tr>
<tr>
<td>3.200</td>
<td>52.9369</td>
<td>39.6483</td>
<td>27.2759</td>
<td>17.1227</td>
<td></td>
</tr>
<tr>
<td>3.300</td>
<td>9.7277</td>
<td>4.9604</td>
<td>2.3169</td>
<td>1.2058</td>
<td></td>
</tr>
<tr>
<td>3.400</td>
<td>1.0879</td>
<td>1.4819</td>
<td>1.9460</td>
<td>2.1290</td>
<td></td>
</tr>
<tr>
<td>3.500</td>
<td>1.8838</td>
<td>1.3395</td>
<td>0.8290</td>
<td>0.6787</td>
<td></td>
</tr>
<tr>
<td>3.600</td>
<td>0.9991</td>
<td>1.6374</td>
<td>2.3259</td>
<td>2.8848</td>
<td></td>
</tr>
<tr>
<td>3.700</td>
<td>3.2880</td>
<td>3.5437</td>
<td>3.5401</td>
<td>3.0475</td>
<td></td>
</tr>
</tbody>
</table>
Table 14. (Continued)

<table>
<thead>
<tr>
<th>r</th>
<th>Δr</th>
<th>0.000</th>
<th>0.025</th>
<th>0.050</th>
<th>0.075</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.800</td>
<td></td>
<td>1.9104</td>
<td>.2446</td>
<td>-1.5649</td>
<td>-3.0852</td>
</tr>
<tr>
<td>3.900</td>
<td></td>
<td>-4.0774</td>
<td>-4.5592</td>
<td>-4.6797</td>
<td>-4.5769</td>
</tr>
<tr>
<td>4.000</td>
<td></td>
<td>-4.3557</td>
<td>-4.1460</td>
<td>-4.0974</td>
<td>-4.2653</td>
</tr>
<tr>
<td>4.100</td>
<td></td>
<td>-4.5147</td>
<td>-4.5888</td>
<td>-4.3176</td>
<td>-3.7683</td>
</tr>
<tr>
<td>4.200</td>
<td></td>
<td>-3.1831</td>
<td>-2.7624</td>
<td>-2.5150</td>
<td>-2.3113</td>
</tr>
<tr>
<td>4.300</td>
<td></td>
<td>-2.0559</td>
<td>-1.7790</td>
<td>-1.5596</td>
<td>-1.3944</td>
</tr>
<tr>
<td>4.400</td>
<td></td>
<td>-1.1768</td>
<td>-0.8072</td>
<td>-0.3011</td>
<td>.2285</td>
</tr>
<tr>
<td>4.500</td>
<td></td>
<td>.6844</td>
<td>1.0570</td>
<td>1.3815</td>
<td>1.6533</td>
</tr>
<tr>
<td>4.600</td>
<td></td>
<td>1.8121</td>
<td>1.8076</td>
<td>1.6515</td>
<td>1.3868</td>
</tr>
<tr>
<td>4.700</td>
<td></td>
<td>1.0252</td>
<td>.5544</td>
<td>.0195</td>
<td>-0.4366</td>
</tr>
<tr>
<td>4.800</td>
<td></td>
<td>-0.6713</td>
<td>-0.7096</td>
<td>-0.7918</td>
<td>-1.2009</td>
</tr>
<tr>
<td>4.900</td>
<td></td>
<td>-1.9992</td>
<td>-2.9225</td>
<td>-3.5520</td>
<td>-3.6196</td>
</tr>
<tr>
<td>5.000</td>
<td></td>
<td>-3.1780</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 15. Theoretical intensity curve ($sI_{m}$) for NSCl.

<table>
<thead>
<tr>
<th>s</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.00</td>
<td>0</td>
<td>0.0259</td>
<td>0.6752</td>
<td>3.5278</td>
</tr>
<tr>
<td>1.00</td>
<td>8.5493</td>
<td>11.0296</td>
<td>3.7084</td>
<td>-16.0968</td>
</tr>
<tr>
<td>2.00</td>
<td>-42.2102</td>
<td>-63.7577</td>
<td>-73.9143</td>
<td>-73.3372</td>
</tr>
<tr>
<td>3.00</td>
<td>-65.5638</td>
<td>-50.2970</td>
<td>-22.6329</td>
<td>18.8160</td>
</tr>
<tr>
<td>4.00</td>
<td>62.7965</td>
<td>86.0133</td>
<td>66.8407</td>
<td>4.4791</td>
</tr>
<tr>
<td>5.00</td>
<td>-70.9947</td>
<td>-108.1601</td>
<td>-61.6175</td>
<td>77.2685</td>
</tr>
<tr>
<td>6.00</td>
<td>265.2516</td>
<td>419.4621</td>
<td>453.7948</td>
<td>321.7660</td>
</tr>
<tr>
<td>7.00</td>
<td>43.7867</td>
<td>-296.4347</td>
<td>-585.0778</td>
<td>-725.1253</td>
</tr>
<tr>
<td>8.00</td>
<td>-674.3225</td>
<td>-457.0984</td>
<td>-147.0941</td>
<td>167.7731</td>
</tr>
<tr>
<td>9.00</td>
<td>417.9096</td>
<td>569.0641</td>
<td>618.1033</td>
<td>574.9385</td>
</tr>
<tr>
<td>10.00</td>
<td>449.1172</td>
<td>249.1358</td>
<td>-6.3928</td>
<td>-277.0549</td>
</tr>
<tr>
<td>11.00</td>
<td>-500.4561</td>
<td>-608.0365</td>
<td>-555.4419</td>
<td>-350.8042</td>
</tr>
<tr>
<td>12.00</td>
<td>-61.4481</td>
<td>207.2147</td>
<td>354.1836</td>
<td>327.8655</td>
</tr>
<tr>
<td>13.00</td>
<td>151.4835</td>
<td>-85.5625</td>
<td>-267.8093</td>
<td>-305.1697</td>
</tr>
<tr>
<td>14.00</td>
<td>-171.8374</td>
<td>84.2109</td>
<td>363.8154</td>
<td>560.0693</td>
</tr>
<tr>
<td>15.00</td>
<td>600.9500</td>
<td>471.7502</td>
<td>212.1705</td>
<td>-106.2873</td>
</tr>
<tr>
<td>16.00</td>
<td>-408.3334</td>
<td>-634.4662</td>
<td>-747.3832</td>
<td>-729.0877</td>
</tr>
<tr>
<td>17.00</td>
<td>-576.8136</td>
<td>-304.5688</td>
<td>50.6065</td>
<td>424.6157</td>
</tr>
<tr>
<td>18.00</td>
<td>733.0176</td>
<td>892.0105</td>
<td>848.0160</td>
<td>602.3001</td>
</tr>
<tr>
<td>19.00</td>
<td>218.0988</td>
<td>-196.1594</td>
<td>-523.0903</td>
<td>-677.7259</td>
</tr>
<tr>
<td>20.00</td>
<td>-636.4794</td>
<td>-441.4142</td>
<td>-178.9189</td>
<td>56.3950</td>
</tr>
<tr>
<td>21.00</td>
<td>196.6505</td>
<td>220.3407</td>
<td>151.4653</td>
<td>41.6762</td>
</tr>
<tr>
<td>22.00</td>
<td>-53.8927</td>
<td>-96.3102</td>
<td>-71.6659</td>
<td>10.8919</td>
</tr>
<tr>
<td>23.00</td>
<td>126.8410</td>
<td>244.6219</td>
<td>331.6269</td>
<td>358.5696</td>
</tr>
<tr>
<td>24.00</td>
<td>304.4498</td>
<td>163.3245</td>
<td>-48.9619</td>
<td>-292.1336</td>
</tr>
<tr>
<td>25.00</td>
<td>-507.3388</td>
<td>-632.0454</td>
<td>-620.3167</td>
<td>-460.7776</td>
</tr>
<tr>
<td>26.00</td>
<td>-184.5345</td>
<td>141.4812</td>
<td>434.4376</td>
<td>621.3540</td>
</tr>
<tr>
<td>27.00</td>
<td>661.0811</td>
<td>554.6674</td>
<td>341.3275</td>
<td>83.0475</td>
</tr>
<tr>
<td>28.00</td>
<td>-155.7455</td>
<td>-325.8439</td>
<td>-403.0782</td>
<td>-389.1823</td>
</tr>
<tr>
<td>29.00</td>
<td>-305.6912</td>
<td>-184.8613</td>
<td>-60.3642</td>
<td>39.8196</td>
</tr>
<tr>
<td>30.00</td>
<td>98.4704</td>
<td>111.6780</td>
<td>88.6257</td>
<td>48.4949</td>
</tr>
<tr>
<td>31.00</td>
<td>14.6118</td>
<td>7.0301</td>
<td>35.1261</td>
<td>93.1979</td>
</tr>
<tr>
<td>32.00</td>
<td>160.9397</td>
<td>209.6656</td>
<td>212.4590</td>
<td>154.5659</td>
</tr>
<tr>
<td>33.00</td>
<td>40.2821</td>
<td>-106.7595</td>
<td>-250.0078</td>
<td>-351.1709</td>
</tr>
<tr>
<td>34.00</td>
<td>-381.7556</td>
<td>-331.2895</td>
<td>-209.8832</td>
<td>-44.8495</td>
</tr>
<tr>
<td>35.00</td>
<td>127.0441</td>
<td>269.0859</td>
<td>353.2539</td>
<td>365.8536</td>
</tr>
<tr>
<td>36.00</td>
<td>309.2180</td>
<td>199.8979</td>
<td>63.8331</td>
<td>-69.7067</td>
</tr>
<tr>
<td>37.00</td>
<td>-174.7624</td>
<td>-234.0632</td>
<td>-242.1689</td>
<td>-205.6485</td>
</tr>
<tr>
<td>38.00</td>
<td>-140.2077</td>
<td>-65.8641</td>
<td>-1.1266</td>
<td>41.9490</td>
</tr>
<tr>
<td>39.00</td>
<td>60.4555</td>
<td>60.1151</td>
<td>51.8172</td>
<td>46.7316</td>
</tr>
</tbody>
</table>
Table 15. (Continued)

<table>
<thead>
<tr>
<th></th>
<th>Δs</th>
<th>0.00</th>
<th>0.25</th>
<th>0.50</th>
<th>0.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>40.00</td>
<td>51.8517</td>
<td>67.5906</td>
<td>87.9269</td>
<td>102.7342</td>
<td></td>
</tr>
<tr>
<td>41.00</td>
<td>101.6501</td>
<td>77.8954</td>
<td>31.1072</td>
<td>-31.9823</td>
<td></td>
</tr>
<tr>
<td>42.00</td>
<td>-99.0131</td>
<td>-155.0737</td>
<td>-186.6019</td>
<td>-184.8324</td>
<td></td>
</tr>
<tr>
<td>43.00</td>
<td>-148.1533</td>
<td>-82.6554</td>
<td>-0.8491</td>
<td>81.1143</td>
<td></td>
</tr>
<tr>
<td>44.00</td>
<td>147.0796</td>
<td>184.5401</td>
<td>187.2713</td>
<td>156.5997</td>
<td></td>
</tr>
<tr>
<td>45.00</td>
<td>100.6579</td>
<td>32.1851</td>
<td>-34.6837</td>
<td>-87.7218</td>
<td></td>
</tr>
<tr>
<td>46.00</td>
<td>-119.1273</td>
<td>-126.6634</td>
<td>-113.3278</td>
<td>-85.7335</td>
<td></td>
</tr>
<tr>
<td>47.00</td>
<td>-51.8890</td>
<td>-19.0328</td>
<td>7.9893</td>
<td>27.2926</td>
<td></td>
</tr>
<tr>
<td>48.00</td>
<td>39.5337</td>
<td>46.8543</td>
<td>51.5633</td>
<td>54.9991</td>
<td></td>
</tr>
</tbody>
</table>
Table 16. Theoretical radial distribution curve for NSCI. (Calculated from theoretical intensity curve with damping factor $= 0.0018$)

<table>
<thead>
<tr>
<th>$r$</th>
<th>$\Delta r$</th>
<th>0.000</th>
<th>0.025</th>
<th>0.050</th>
<th>0.075</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.000</td>
<td>0</td>
<td>0.0027</td>
<td>0.0065</td>
<td>0.0100</td>
<td></td>
</tr>
<tr>
<td>0.100</td>
<td>0.0097</td>
<td>0.0035</td>
<td>-0.0058</td>
<td>-0.0111</td>
<td></td>
</tr>
<tr>
<td>0.200</td>
<td>-0.0089</td>
<td>-0.0034</td>
<td>-0.0035</td>
<td>-0.0124</td>
<td></td>
</tr>
<tr>
<td>0.300</td>
<td>-0.0228</td>
<td>-0.0250</td>
<td>-0.0193</td>
<td>-0.0167</td>
<td></td>
</tr>
<tr>
<td>0.400</td>
<td>-0.0252</td>
<td>-0.0390</td>
<td>-0.0448</td>
<td>-0.0405</td>
<td></td>
</tr>
<tr>
<td>0.500</td>
<td>-0.0393</td>
<td>-0.0532</td>
<td>-0.0745</td>
<td>-0.0816</td>
<td></td>
</tr>
<tr>
<td>0.600</td>
<td>-0.0852</td>
<td>-0.0414</td>
<td>-0.0326</td>
<td>-0.0385</td>
<td></td>
</tr>
<tr>
<td>0.700</td>
<td>-0.0358</td>
<td>-0.0090</td>
<td>0.0269</td>
<td>0.0458</td>
<td></td>
</tr>
<tr>
<td>0.800</td>
<td>0.0481</td>
<td>0.0657</td>
<td>1.215</td>
<td>1.961</td>
<td></td>
</tr>
<tr>
<td>0.900</td>
<td>0.2493</td>
<td>0.2750</td>
<td>0.3152</td>
<td>0.4078</td>
<td></td>
</tr>
<tr>
<td>1.000</td>
<td>0.5324</td>
<td>0.6316</td>
<td>0.6861</td>
<td>0.7452</td>
<td></td>
</tr>
<tr>
<td>1.100</td>
<td>0.8614</td>
<td>1.0176</td>
<td>1.1669</td>
<td>1.3809</td>
<td></td>
</tr>
<tr>
<td>1.200</td>
<td>1.9833</td>
<td>3.6282</td>
<td>7.4607</td>
<td>15.4336</td>
<td></td>
</tr>
<tr>
<td>1.300</td>
<td>30.4297</td>
<td>55.4840</td>
<td>91.7752</td>
<td>136.2187</td>
<td></td>
</tr>
<tr>
<td>1.400</td>
<td>180.5238</td>
<td>213.2475</td>
<td>224.4800</td>
<td>210.6450</td>
<td></td>
</tr>
<tr>
<td>1.500</td>
<td>176.3602</td>
<td>132.0924</td>
<td>89.1491</td>
<td>55.1657</td>
<td></td>
</tr>
<tr>
<td>1.600</td>
<td>32.4723</td>
<td>19.4193</td>
<td>12.8913</td>
<td>10.1427</td>
<td></td>
</tr>
<tr>
<td>1.800</td>
<td>8.2489</td>
<td>7.1583</td>
<td>5.6361</td>
<td>3.6938</td>
<td></td>
</tr>
<tr>
<td>1.900</td>
<td>1.7302</td>
<td>0.8276</td>
<td>2.9219</td>
<td>10.8694</td>
<td></td>
</tr>
<tr>
<td>2.000</td>
<td>28.2434</td>
<td>58.4127</td>
<td>102.6443</td>
<td>157.8480</td>
<td></td>
</tr>
<tr>
<td>2.100</td>
<td>215.4870</td>
<td>263.0958</td>
<td>288.4336</td>
<td>284.4355</td>
<td></td>
</tr>
<tr>
<td>2.200</td>
<td>252.2705</td>
<td>200.7692</td>
<td>142.6152</td>
<td>89.4816</td>
<td></td>
</tr>
<tr>
<td>2.300</td>
<td>48.5702</td>
<td>21.7860</td>
<td>7.1330</td>
<td>0.9472</td>
<td></td>
</tr>
<tr>
<td>2.400</td>
<td>-0.3109</td>
<td>0.7741</td>
<td>2.6291</td>
<td>4.4233</td>
<td></td>
</tr>
<tr>
<td>2.500</td>
<td>5.7940</td>
<td>6.6579</td>
<td>7.0760</td>
<td>7.1506</td>
<td></td>
</tr>
<tr>
<td>2.600</td>
<td>6.9672</td>
<td>6.5902</td>
<td>6.0840</td>
<td>5.5221</td>
<td></td>
</tr>
<tr>
<td>2.700</td>
<td>4.9705</td>
<td>4.4708</td>
<td>4.0506</td>
<td>3.7610</td>
<td></td>
</tr>
<tr>
<td>2.800</td>
<td>3.7134</td>
<td>4.0966</td>
<td>5.1803</td>
<td>7.3182</td>
<td></td>
</tr>
<tr>
<td>2.900</td>
<td>10.9380</td>
<td>16.4748</td>
<td>24.2196</td>
<td>34.1119</td>
<td></td>
</tr>
<tr>
<td>3.000</td>
<td>45.5642</td>
<td>57.4201</td>
<td>68.1015</td>
<td>75.9286</td>
<td></td>
</tr>
<tr>
<td>3.100</td>
<td>79.5302</td>
<td>78.2212</td>
<td>72.2125</td>
<td>62.5609</td>
<td></td>
</tr>
<tr>
<td>3.200</td>
<td>50.8633</td>
<td>38.8121</td>
<td>27.7902</td>
<td>18.6454</td>
<td></td>
</tr>
<tr>
<td>3.300</td>
<td>11.6779</td>
<td>6.7686</td>
<td>3.5527</td>
<td>1.5756</td>
<td></td>
</tr>
<tr>
<td>3.400</td>
<td>-0.2774</td>
<td>-0.6924</td>
<td>-0.9504</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3.500</td>
<td>-1.1150</td>
<td>-1.2289</td>
<td>-1.3175</td>
<td>-1.3851</td>
<td></td>
</tr>
<tr>
<td>3.600</td>
<td>-1.4231</td>
<td>-1.4292</td>
<td>-1.4150</td>
<td>-1.3948</td>
<td></td>
</tr>
<tr>
<td>3.700</td>
<td>-1.3693</td>
<td>-1.3277</td>
<td>-1.2641</td>
<td>-1.1879</td>
<td></td>
</tr>
<tr>
<td>r</td>
<td>Δr</td>
<td>0.000</td>
<td>0.025</td>
<td>0.050</td>
<td>0.075</td>
</tr>
<tr>
<td>-----</td>
<td>--------</td>
<td>--------</td>
<td>--------</td>
<td>--------</td>
<td>--------</td>
</tr>
<tr>
<td>3.800</td>
<td>-1.1143</td>
<td>-1.0476</td>
<td>-0.9785</td>
<td>-0.8987</td>
<td></td>
</tr>
<tr>
<td>3.900</td>
<td>-0.8141</td>
<td>-0.7390</td>
<td>-0.6793</td>
<td>-0.6264</td>
<td></td>
</tr>
<tr>
<td>4.000</td>
<td>-0.5696</td>
<td>-0.5106</td>
<td>-0.4612</td>
<td>-0.4275</td>
<td></td>
</tr>
<tr>
<td>4.100</td>
<td>-0.4020</td>
<td>-0.3734</td>
<td>-0.3405</td>
<td>-0.3131</td>
<td></td>
</tr>
<tr>
<td>4.200</td>
<td>-0.2981</td>
<td>-0.2899</td>
<td>-0.2774</td>
<td>-0.2589</td>
<td></td>
</tr>
<tr>
<td>4.300</td>
<td>-0.2439</td>
<td>-0.2400</td>
<td>-0.2417</td>
<td>-0.2365</td>
<td></td>
</tr>
<tr>
<td>4.400</td>
<td>-0.2197</td>
<td>-0.2004</td>
<td>-0.1898</td>
<td>-0.1875</td>
<td></td>
</tr>
<tr>
<td>4.500</td>
<td>-0.1838</td>
<td>-0.1729</td>
<td>-0.1612</td>
<td>-0.1583</td>
<td></td>
</tr>
<tr>
<td>4.600</td>
<td>-0.1641</td>
<td>-0.1685</td>
<td>-0.1654</td>
<td>-0.1605</td>
<td></td>
</tr>
<tr>
<td>4.700</td>
<td>-0.1647</td>
<td>-0.1797</td>
<td>-0.1948</td>
<td>-0.2000</td>
<td></td>
</tr>
<tr>
<td>4.800</td>
<td>-0.1970</td>
<td>-0.1961</td>
<td>-0.2026</td>
<td>-0.2109</td>
<td></td>
</tr>
<tr>
<td>4.900</td>
<td>-0.2136</td>
<td>-0.2122</td>
<td>-0.2153</td>
<td>-0.2264</td>
<td></td>
</tr>
<tr>
<td>5.000</td>
<td>-0.2381</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 17. Comparison of weight factor ratios for the two possible models for thiazyl chloride and the observed peak area ratios from the experimental radial distribution curve.

<table>
<thead>
<tr>
<th>Peak (Å)</th>
<th>Weight ratios for</th>
<th>Observed peak area ratios</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>SNCl Model</td>
<td>NSCl Model</td>
</tr>
<tr>
<td>1.45</td>
<td>( W_{SN} : 0.88 )</td>
<td>( W_{NS} : 2.02 )</td>
</tr>
<tr>
<td>2.16</td>
<td>( W_{NCl} : 0.63 )</td>
<td>( W_{SCI} : 3.28 )</td>
</tr>
<tr>
<td>3.11</td>
<td>( W_{SCI} : 1.00 )</td>
<td>( W_{NCl} : 1.00 )</td>
</tr>
</tbody>
</table>

Table 18. Results of the least square refinements on NSCl: L. S. I. least squares refinement on individual intensity curves. L. S. II. least squares refinement on the composite intensity curve. Distances \( r_{ij} \) and root-mean-square amplitudes \( \sigma_{ij} \) in Å.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Least Squares I</th>
<th></th>
<th>Least Squares II</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Parameter</td>
<td>Standard( \sigma_{LS} )</td>
<td>Parameter</td>
<td>Standard( \sigma_{LS} )</td>
</tr>
<tr>
<td></td>
<td>Values</td>
<td>errors</td>
<td>Values</td>
<td>errors</td>
</tr>
<tr>
<td>( r_{NS} )</td>
<td>1.44827</td>
<td>±0.00084</td>
<td>1.44656</td>
<td>±0.00092</td>
</tr>
<tr>
<td>( r_{SCI} )</td>
<td>2.15928</td>
<td>±0.00079</td>
<td>2.15839</td>
<td>±0.00095</td>
</tr>
<tr>
<td>( r_{NCl} )</td>
<td>3.10633</td>
<td>±0.00549</td>
<td>3.11003</td>
<td>±0.00654</td>
</tr>
<tr>
<td>( l_{NS} )</td>
<td>0.03775</td>
<td>±0.00089</td>
<td>0.03489</td>
<td>±0.00104</td>
</tr>
<tr>
<td>( l_{SCI} )</td>
<td>0.05265</td>
<td>±0.00076</td>
<td>0.05198</td>
<td>±0.00094</td>
</tr>
<tr>
<td>( l_{NCl} )</td>
<td>0.07656</td>
<td>±0.00413</td>
<td>0.07496</td>
<td>±0.00488</td>
</tr>
<tr>
<td>( \angle_{NSCl} )</td>
<td>117.513°</td>
<td>±0.357°</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

\(^{a}\) Taken from the diagonal elements \((\sigma_{LS}^2)^{2}\) of the error matrix.
Table 19. Final parameter values for gaseous NSCl as determined by a least squares refinement on individual intensity curves. Distances ($r_{ij}$) and root-mean-square amplitudes of vibration ($l_{ij}$) in $\text{Å}$. Angle in degrees.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Parameter Value</th>
<th>Associated error $2\sigma^a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r_{NS}$</td>
<td>1.4483</td>
<td>0.0028</td>
</tr>
<tr>
<td>$r_{SCl}$</td>
<td>2.1593</td>
<td>0.0031</td>
</tr>
<tr>
<td>$r_{NCI}$</td>
<td>3.1063</td>
<td>0.0158</td>
</tr>
<tr>
<td>$l_{NS}$</td>
<td>0.0378</td>
<td>0.0029</td>
</tr>
<tr>
<td>$l_{SCl}$</td>
<td>0.0526</td>
<td>0.0030</td>
</tr>
<tr>
<td>$l_{NCI}$</td>
<td>0.0766</td>
<td>0.0121</td>
</tr>
<tr>
<td>$\angle_{NSCl}$</td>
<td>117.51</td>
<td>1.01</td>
</tr>
</tbody>
</table>

$^a$Values of $2\sigma$ calculated according to equations 15, 16 and 17.
Table 20. The symmetric error matrix for NSCl resulting from the least squares refinement on individual intensity curves. All values have been multiplied by $10^7$. Distances ($r_{ij}$) and amplitudes ($l_{ij}$) in Å, and angles in degrees.

<table>
<thead>
<tr>
<th></th>
<th>$k^a$</th>
<th>$l_{NS}$</th>
<th>$l_{SC1}$</th>
<th>$l_{NC1}$</th>
<th>$r_{NS}$</th>
<th>$r_{SC1}$</th>
<th>$r_{NC1}$</th>
<th>$\angle_{NSCl}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$k$</td>
<td>2850.78</td>
<td>85.197</td>
<td>83.960</td>
<td>116.902</td>
<td>-0.5760</td>
<td>3.950</td>
<td>-15.665</td>
<td>-1206.95</td>
</tr>
<tr>
<td>$l_{NS}$</td>
<td>7.898</td>
<td>2.309</td>
<td>4.010</td>
<td>-0.0823</td>
<td>-0.1034</td>
<td>-0.3534</td>
<td>23.438</td>
<td></td>
</tr>
<tr>
<td>$l_{SC1}$</td>
<td>5.843</td>
<td>2.800</td>
<td>0.1399</td>
<td>0.0247</td>
<td>0.4979</td>
<td>-283.858</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$l_{NC1}$</td>
<td>170.626</td>
<td>-0.0474</td>
<td>-1.014</td>
<td>-5.384</td>
<td>-270.421</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$r_{NS}$</td>
<td>7.016</td>
<td>-0.6182</td>
<td>0.7468</td>
<td>-348.048</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$r_{SC1}$</td>
<td>6.259</td>
<td>-0.2114</td>
<td>301.111</td>
<td>19,297.1</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$r_{NC1}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\angle_{NSCl}$</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$Scale parameter of no structural significance.
Table 21. Observed vibrational frequencies in cm\(^{-1}\) for gaseous NSCl. Spectrum scanned from 200-4,000 cm\(^{-1}\).

<table>
<thead>
<tr>
<th>(\nu_1) : N-S stretch</th>
<th>(\nu_2) : S-Cl stretch</th>
<th>(\nu_3) : NSCl bend</th>
</tr>
</thead>
<tbody>
<tr>
<td>Branch</td>
<td>Observed value</td>
<td>Branch</td>
</tr>
<tr>
<td>(^{14})N-(^{32})S P</td>
<td>1317.0</td>
<td>P</td>
</tr>
<tr>
<td>Q of (^{14})N-(^{32})S</td>
<td>1324.0</td>
<td>Q of (^{32})S-(^{37})Cl</td>
</tr>
<tr>
<td>R of (^{14})N-(^{32})S</td>
<td>1332.6</td>
<td>Q of (^{32})S-(^{35})Cl</td>
</tr>
<tr>
<td>(^{15})N-(^{32})S P</td>
<td>1286.6</td>
<td>R</td>
</tr>
<tr>
<td>Q of (^{15})N-(^{32})S</td>
<td>1293.4</td>
<td></td>
</tr>
<tr>
<td>R of (^{15})N-(^{32})S</td>
<td>not measurable</td>
<td></td>
</tr>
</tbody>
</table>

Table 22. Observed frequencies in cm\(^{-1}\) for solid films of NSCl on KBr. Spectrum scanned from 350-4,000 cm\(^{-1}\).

<table>
<thead>
<tr>
<th>N-S isotopes</th>
<th>Observed value</th>
<th>Calculated value(^a)</th>
</tr>
</thead>
<tbody>
<tr>
<td>14-32</td>
<td>1397.0</td>
<td>---------</td>
</tr>
<tr>
<td>14-34</td>
<td>1384.8</td>
<td>1384.4</td>
</tr>
<tr>
<td>15-32</td>
<td>1365.0</td>
<td>1364.3</td>
</tr>
<tr>
<td>15-34</td>
<td>1352.2</td>
<td>1351.5</td>
</tr>
</tbody>
</table>

\(^a\) Calculated values based on a harmonic oscillator-diatomic molecule (N-S) approximation starting with 1397.0 cm\(^{-1}\) for \(^{14}\)N-\(^{32}\)S.
Table 23. Observed frequencies in cm\(^{-1}\) for solid films of NSCl on CsI (spectrum scanned from 180-4,000 cm\(^{-1}\)) and on polyethylene (spectrum scanned from 95-640 cm\(^{-1}\)).

<table>
<thead>
<tr>
<th>Molecular Form</th>
<th>Ionic Form (^a)</th>
<th>Polyethylene Ionic Form</th>
</tr>
</thead>
<tbody>
<tr>
<td>1346, 1315(^b)</td>
<td>1397, 1365(^b)</td>
<td></td>
</tr>
<tr>
<td>350</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>256</td>
<td>---</td>
<td>---</td>
</tr>
<tr>
<td>~ 205</td>
<td>205</td>
<td>140</td>
</tr>
</tbody>
</table>

\(^a\) Observed frequencies are identical to those of NSCl on KBr in the region of common scan—see Table 22.

\(^b\) \(^{15}\)N enriched sample had this additional frequency. The other frequencies remained unchanged by this \(^{15}\)N isotope.
Table 24. List of constants used in calculations for thiazyl chloride.

<table>
<thead>
<tr>
<th>Constant</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m_{14N}$</td>
<td>14.00307</td>
<td>g/mole</td>
</tr>
<tr>
<td>$m_{15N}$</td>
<td>15.00011</td>
<td>g/mole</td>
</tr>
<tr>
<td>$m_{32S}$</td>
<td>31.97207</td>
<td>g/mole</td>
</tr>
<tr>
<td>$m_{34S}$</td>
<td>33.96786</td>
<td>g/mole</td>
</tr>
<tr>
<td>$m_{35Cl}$</td>
<td>34.96885</td>
<td>g/mole</td>
</tr>
<tr>
<td>$m_{37Cl}$</td>
<td>36.96590</td>
<td>g/mole</td>
</tr>
<tr>
<td>$h$</td>
<td>$6.62517 \times 10^{-27}$</td>
<td>erg sec</td>
</tr>
<tr>
<td>$k$</td>
<td>$1.38044 \times 10^{-16}$</td>
<td>erg/deg</td>
</tr>
<tr>
<td>$c$</td>
<td>$2.99793 \times 10^{10}$</td>
<td>cm/sec</td>
</tr>
<tr>
<td>$N_0$</td>
<td>$6.02322 \times 10^{23}$</td>
<td>atoms/g mole</td>
</tr>
<tr>
<td>$\nu_1$</td>
<td>1324.0</td>
<td>cm$^{-1}$</td>
</tr>
<tr>
<td>$\nu_2$</td>
<td>417.8</td>
<td>cm$^{-1}$</td>
</tr>
<tr>
<td>$\nu_3$</td>
<td>271.</td>
<td>cm$^{-1}$</td>
</tr>
<tr>
<td>$T$</td>
<td>368.</td>
<td>deg K</td>
</tr>
<tr>
<td>$\pi$</td>
<td>3.14159</td>
<td></td>
</tr>
</tbody>
</table>
Table 25. Force constants of NSC1 in millidynes per angstrom.

<table>
<thead>
<tr>
<th>Force Constant</th>
<th>Infrared data</th>
<th>Force Constant</th>
<th>Coordinate Transformation</th>
<th>Least Squares I Results</th>
<th>Least Squares II Results</th>
</tr>
</thead>
<tbody>
<tr>
<td>( F_{NS} )</td>
<td>10.01</td>
<td>( F'_{NS} )</td>
<td>10.62</td>
<td>9.33</td>
<td>10.92</td>
</tr>
<tr>
<td>( F_{SC1} )</td>
<td>1.58</td>
<td>( F'_{SC1} )</td>
<td>2.40</td>
<td>2.22</td>
<td>2.28</td>
</tr>
<tr>
<td>( F_a )</td>
<td>0.25</td>
<td>( F'_{NC1} )</td>
<td>0.99</td>
<td>0.95</td>
<td>0.99</td>
</tr>
<tr>
<td>( F_{NS/SC1} )</td>
<td>0.</td>
<td>( F'_{NS/SC1} )</td>
<td>0.71</td>
<td>0.</td>
<td>0.</td>
</tr>
<tr>
<td>( F_{NS/a} )</td>
<td>0.</td>
<td>( F'_{NS/NC1} )</td>
<td>-0.78</td>
<td>0.</td>
<td>0.</td>
</tr>
<tr>
<td>( F_{SC1/a} )</td>
<td>0.</td>
<td>( F'_{SC1/NC1} )</td>
<td>-0.90</td>
<td>0.</td>
<td>0.</td>
</tr>
</tbody>
</table>

- Calculated from \( | \sum \gamma - \lambda \bar{E} | = 0 \) assuming a diagonal \( \bar{E} \) matrix.
- Calculated for symmetry coordinates \( \Delta r_{NS}, \Delta r_{SC1}, \Delta r_{NC1} \) starting with the force constants for symmetry coordinate \( \Delta r_{NS}', \Delta r_{SC1}', \Delta r_{NC1}' \) listed in the second column.
- Calculated from \( | \sum E - \epsilon \bar{E} | = 0 \) assuming diagonal \( \bar{E} \) and \( \bar{E} \) matrices.

Table 26. Root-mean-square amplitudes of vibration \( (1_{ij}) \) in \( \overset{\text{A}}{\text{A}} \) for NSC1 at 368°K.

<table>
<thead>
<tr>
<th></th>
<th>Electron diffraction</th>
<th>Infrared Spectroscopy</th>
<th>Least squares I results</th>
<th>Least squares II results</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 1_{NS} )</td>
<td>0.0354</td>
<td>0.0378</td>
<td>0.0349</td>
<td></td>
</tr>
<tr>
<td>( 1_{SC1} )</td>
<td>0.0506</td>
<td>0.0526</td>
<td>0.0520</td>
<td></td>
</tr>
<tr>
<td>( 1_{NC1} )</td>
<td>0.0749</td>
<td>0.0766</td>
<td>0.0750</td>
<td></td>
</tr>
</tbody>
</table>

- Calculated from \( | \sum E - \epsilon \bar{E} | = 0 \) assuming diagonal \( \bar{E} \) and \( \bar{E} \) matrices.
Figure 1. Experimental intensity curves for NSCl.
Figure 2. Intensity curves for NSCl.
Figure 3. Radial distribution curves for NSCI.
Figure 4. Infrared spectrum of gaseous NSCI: $v_1$ (N-S stretching vibration) region of the spectrum. Initial vapor pressure (Pi) 8 mm, 10-cm path, KBr windows. Recorded on Beckman IR7. Note P-Q-R structure with $^{14}$N-$^{32}$S Q branch at 1324.0 cm$^{-1}$ and $^{15}$N-$^{32}$S Q branch at 1293.4 cm$^{-1}$.
Figure 5. Infrared spectrum of gaseous NSCl: $v_2$ (S-Cl stretching vibration) region of the spectrum. Initial vapor pressure (P) 5 mm, 10-cm path, KBr windows. Recorded on Beckman IR 7 with CsI interchange. Note P-Q-R structure with $^{32}$S-$^{35}$ Cl Q branch at 417.8 cm$^{-1}$ and $^{32}$S-$^{37}$ Cl Q branch at 414.1 cm$^{-1}$. 
Figure 6. Infrared spectrum of gaseous NSCl: $\nu_3$ (NSCl bending vibration) region of the spectrum. Initial vapor pressure ($P_i$) 30 mm, 10-cm path, polyethylene windows. Recorded on Beckman IR 11. Note P-R structure with missing Q branch. Highest point between P and R at 271 cm$^{-1}$ taken as bending frequency.
Figure 7. Infrared spectrum of a solid film of NSCl (ionic form) on KBr cooled with liquid nitrogen: N-S stretching vibration region of the spectrum. Recorded on a Beckman IR 7 using an infrared cold cell. Note $^{14}\text{N}-^{32}\text{S},^{14}\text{N}-^{34}\text{S},^{15}\text{N}-^{32}\text{S},$ and $^{15}\text{N}-^{34}\text{S}$ frequencies at 1397.0, 1384.8, 1365.0, and 1352.2 cm$^{-1}$ respectively.
Figure 8. Infrared spectrum of solid films of NSCl on CsI and polyethylene cooled with liquid nitrogen. Recorded on a Beckman IR 7 and IR 11 using an infrared cold cell. Upper curve is NSCl (molecular form) on CsI. Middle curve is NSCl (ionic form) on CsI (1300-1400 cm\(^{-1}\) region is identical to that shown in Figure 7). Lower curve is NSCl (ionic form) on polyethylene.
BIBLIOGRAPHY


64. Wilson, E. Bright, Jr., J. C. Decius and Paul C. Cross. 