

Hot electron luminescence in ZnS alternating-current thin-film electroluminescent devices

A. A. Douglas^{a)} and J. F. Wager

Department of Electrical and Computer Engineering, Center for Advanced Materials Research, Oregon State University, Corvallis, Oregon 97331-3211

D. C. Morton and J. B. Koh

U.S. Army Laboratory Command, Electronics Technology and Devices Laboratory, Fort Monmouth, New Jersey 07703-5000

C. P. Hough

Vitronics, Inc., Eatontown, New Jersey 07724

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Hot electron luminescence experiments are performed on ZnS alternating-current thin-film electroluminescent (ACTFEL) devices in order to determine the extent to which the electron distribution is heated. The luminescence spectrum is found to be broad and essentially featureless up to a high energy cutoff of approximately 3.7 eV, which is determined by optical absorption within the ZnS. This result indicates that under normal operating conditions in a ZnS ACTFEL device, a significant fraction of the electrons transported across the phosphor possess energies equal to or in excess of the ZnS band gap.

Electron transport in alternating-current thin-film electroluminescent (ACTFEL) devices is the source of some controversy. One point of view^{1,2} contends from Monte Carlo calculations and vacuum emission measurements that electrons travel ballistically through the ACTFEL device and reach exceedingly high energies. Other workers have concluded from Monte Carlo calculations^{3,4} and lucky-drift modeling⁵ that the average energy of the electron distribution is approximately 1–2 eV with high energy tails out to perhaps 3–4 eV for phosphor fields at which ACTFEL devices typically operate (≈ 1.5 – 2.0 MV/cm).

The purpose of this letter is to report the results of a study of hot electron luminescence in doped and undoped ZnS ACTFEL devices. Similar measurements to those discussed herein were reported previously⁶ using ZnS Schottky diodes subjected to dc bias. The hot electron luminescence spectrum observed under dc bias was interpreted⁶ as originating from hot electron radiative recombination within the conduction band; we assume that our ac hot electron luminescent spectrum arises from the same physical process.

The ACTFEL devices used in this work are fabricated by evaporation in the typical stack configuration where the phosphor layer, ZnS, is sandwiched between silicon oxynitride (SiON) insulators and aluminum and indium tin oxide (ITO) electrodes. The measurement is performed on doped and undoped ACTFEL devices.

The hot electron luminescence experiment consists of driving an ACTFEL device with a bipolar voltage waveform and monitoring the luminescence spectrum. The device is driven with alternating pulses with a voltage of approximately 20 V above the threshold for the onset of conduction at a frequency of 100 Hz. The phosphor electric field during conduction is calculated as

$$E_p = \frac{1}{d_p} \left(\frac{Q_{st}}{C_i} - V_{ext} \right), \quad (1)$$

where d_p is the phosphor thickness, V_{ext} is the external voltage, Q_{st} is the charge on a large sense capacitor (Sawyer–Tower configuration), and C_i is the total insulator capacitance. Note that Eq. (1) assumes that there is no space charge within the phosphor so that the electric field is constant across the phosphor layer. The hot electron luminescence is measured at three phosphor fields: $E_p = 1.2, 1.4,$ and 1.6 MV/cm. The largest phosphor field is the normal clamping field, whereas the two smaller phosphor fields are obtained using the field-control circuit shown in Fig. 1. The central idea behind the field-control circuit is to force the phosphor electric field to a constant value. This is accomplished by first rearranging Eq. (1) as follows:

$$V_{ext} = \frac{C_{st}}{C_i} \left(V_{st} - \frac{d_p C_i}{C_{st}} E_p \right), \quad (2)$$

where the following relation is used,

$$Q_{st} = C_{st} V_{st}. \quad (3)$$

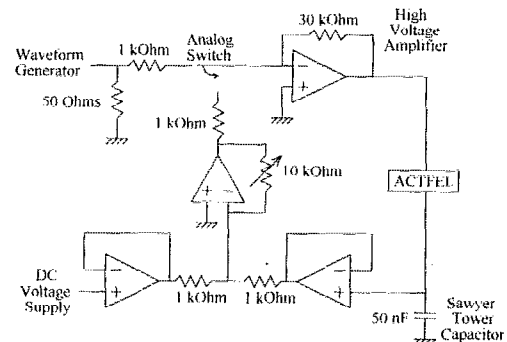


FIG. 1. Field-control circuit.

^{a)}Present address: Planar Systems, Inc., Beaverton, OR 97006.

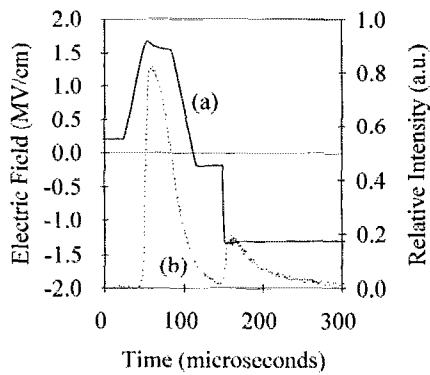


FIG. 2. (a) Phosphor field and (b) luminescence transient curves at 460 nm.

Then, field control is achieved by realizing Eq. (2) using the circuit shown in Fig. 1. Since the goal of the field-control circuit is to hold E_p constant, the last term in the parentheses is a constant; the magnitude of this constant is set by the dc bias, V_{dc} , indicated in Fig. 1. The prefactor multiplying the expression within the parentheses is the closed-loop gain of the field-control circuit which is equal to the ratio of C_{st} to C_f .

The hot electron luminescent spectra are measured using a SPEX 3/4 meter monochromator and a cooled, extended range photomultiplier tube (PMT). Spectra are obtained in a time-resolved manner by digitally acquiring the voltage across a 50 k Ω resistor between the PMT and ground. A single-system correction to the raw spectral data is performed which accounts for lens, grating, and detector response.

Optical absorption measurements of the ACTFEL stack are performed using a SPEX Fluorolog System, which includes a xenon lamp and a pair of 0.22 m double monochrometers.

Phosphor field and hot electron luminescence transient curves are shown in Fig. 2 for a wavelength of 460 nm. The first peak in the $E_p(t)$ curve corresponds to the application of an external voltage pulse; note that field clamping occurs at approximately 1.6 MV/cm. The 1.4 MV/cm constant field portion of the $E_p(t)$ curve occurs when the field-control circuit is operational. Note that although the du-

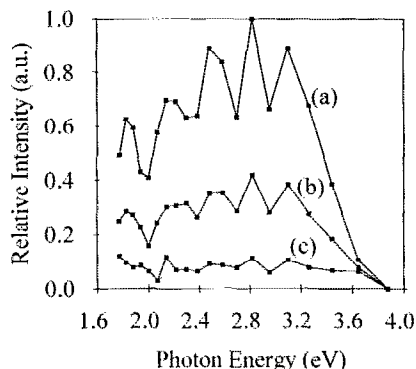


FIG. 3. Electron luminescence spectra for various phosphor fields: (a) 1.6 MV/cm, (b) 1.4 MV/cm, and (c) 1.2 MV/cm.

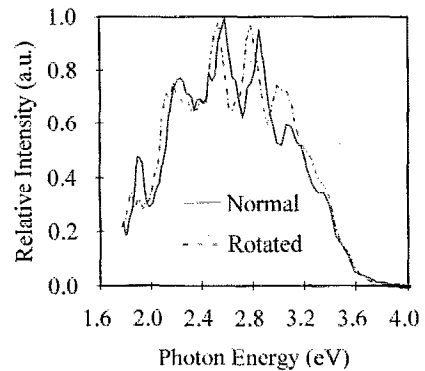


FIG. 4. Electron luminescence spectra at two emission angles at 1.6 MV/cm.

ration of the hot electron luminescence is very short (microseconds) and is concomitant with the flow of conduction current, the duration of the measured luminescence transient is approximately 100 μ s, which is determined by the RC time constant of the 50 k Ω resistor and the PMT. Also, notice that the 1.4 MV/cm luminescence signal is weak compared to that of the first peak; we have found 1.2 MV/cm to be the smallest phosphor field at which we can obtain adequate signal intensity with the present setup.

Hot electron luminescence spectra are plotted in Fig. 3 for $E_p=1.2, 1.4,$ and 1.6 MV/cm. The structure observed in Fig. 3 is attributed to optical interference since these peaks shift, as shown in Fig. 4, when the emission angle of the sample is rotated by approximately 45 $^\circ$ with respect to the monochromator. As apparent from Fig. 3, the high energy, hot electron luminescence cutoff occurs at about 3.7 eV. Optical absorption spectra for the ACTFEL stack, the glass substrate, and ITO on glass are given in Fig. 5. It is clear from Fig. 5 that the absorption edge at about 3.7 eV arises from absorption within the ZnS layer.

Thus, a comparison of the hot electron luminescence spectra of Fig. 3 and the optical absorption spectra of Fig. 5 indicates that the high energy, hot electron luminescence cutoff arises from optical absorption within the ZnS layer. Therefore, if we correct for optical interference and absorption effects, the hot electron luminescence spectrum is

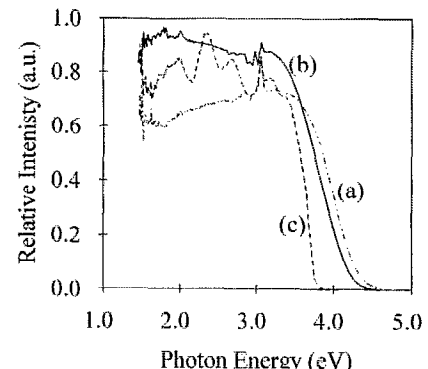


FIG. 5. Optical absorption spectra for (a) the glass substrate, (b) the glass substrate and the ITO layer, and (c) the full ACTFEL stack.

broad and rather structureless out to energies approaching 3.7 eV, the band gap of ZnS.

The hot electron luminescence spectrum arises from the radiative transition of energetic electrons within the ZnS conduction band⁹ and, therefore, is related to the hot electron distribution. Thus, we conclude that a significant fraction of the hot electrons transiting the ZnS possess kinetic energies near to, and most likely in excess of, the 3.7 eV band gap of ZnS. Moreover, although the intensity of the hot electron luminescence scales with the magnitude of the applied field, as indicated in Fig. 3, the shape of the hot electron luminescence spectrum is relatively independent of field. In particular, the high energy tail extends out to 3.7 eV for all of the fields investigated. This implies that for any field large enough to cause significant tunnel emission from interfaces states, a substantial number of the emitted electrons will reach *at least* band-gap energies. We cannot infer anything about the hot electron distribution for energies in excess of 3.7 eV since these energies are inaccessible to this experiment.

Further attempts were made to optically measure the high energy tail of the electron emission. A sample was broken and edge emission monitored. The edge emission spectrum contains the same 3.7 eV cutoff as before. This implies that the intensity of edge-emitted, high energy photons is so low that they are completely undetectable.

One implication of the high energy of the electron distribution inferred from the hot electron luminescence experiment is that it now seems possible that band to band impact ionization occurs in these devices. The threshold energy for impact ionization, E_{th} , is given by,⁷

$$E_{th} = \frac{2m_e + m_h}{m_e + m_h} E_G, \quad (4)$$

where⁸ $m_e = 0.34m_0$ is the electron effective mass, $m_h = 1.76m_0$ is the heavy-hole effective mass, and $E_G = 3.7$ eV is the band gap. This results in E_{th} of approximately 4.3 eV. Extrapolation of Fig. 3 suggests that band to band impact ionization would likely occur in ZnS ACTFEL devices under normal operating conditions.

A second implication of the work reported herein is that the achievement of efficient blue ACTFEL devices should not be impeded by the transport properties, if ZnS is employed as the host phosphor.

Finally, it should be noted that although all of the data shown in this letter are for undoped samples, the high energy portion of the hot electron luminescence spectrum is identical for Mn-doped samples, to within the accuracy of the experiment. Thus, the Mn luminescent impurities do not significantly cool the electron distribution. This result is consistent with the Monte Carlo simulation⁹ of Bhattacharyya *et al.*, who found the Mn impact excitation scattering rate to be several orders of magnitude smaller than that of polar optical phonon or intervalley scattering.

In conclusion, the hot electron luminescence spectrum is measured for an ACTFEL device in which the ZnS is grown by evaporation. The luminescence spectrum is broad and featureless and is cut off at the ZnS band gap due to optical absorption. This result provides evidence that a substantial fraction of the electrons transiting the ACTFEL device have energies near to or in excess of the ZnS band gap.

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