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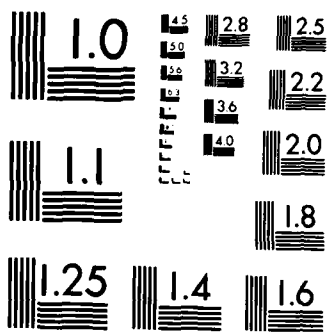

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THIN-FILM ELECTROLUMINESCENCE

FINAL REPORT

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C. B. COOPER, F. E. WILLIAMS

JULY 10, 1985

U. S. ARMY RESEARCH OFFICE

GRANT DAAG29-83-K-0060

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19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Electroluminescence, collision cross section, phosphors.

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

This project involved further theoretical and experimental work on thin film electroluminescence (TFEL) devices. A theoretical study was carried out to calculate the cross sections of collision excitation processes of the luminescence centers in TFEL devices. Cross sections for $ZnS:Mn^{2+}$, $ZnF_2:Mn^{2+}$, $ZnS:TbF_3$, $ZnF_2:TbF_3$, $Y_2O_3:Tb^{3+}$, and $Y_2O_3:Eu^{3+}$. Experiments were begun to fabricate EL cells designed for the purpose of measuring the energy distribution of the hot carriers in TFEL devices;

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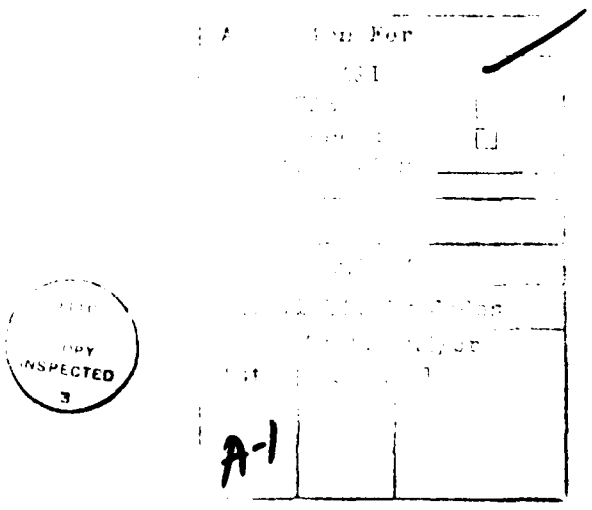
to get a better idea of the quantitative electrical behavior of TFEL cells; and to estimate the range of hot carriers in ZnF_2:Mn films.

FERD ELTON WILLIAMS

Professor Ferd E. Williams, Principal Investigator and long time moving spirit behind the Electroluminescent research program at the University of Delaware died suddenly and unexpectedly during the course of this project in June 1984.



(a) ***Statement of problem studied.***

The purpose of the work was to elucidate further the mechanism of high field electroluminescence (EL) and to develop new materials for EL devices, building on the experimental facilities and theoretical analyses developed here previously. This includes determination and use of collision excitation cross sections to guide the choice of structures and materials and attention will be directed to determining and modifying hot carrier distributions in order to improve EL performance.

(b) ***Summary of results***

In the theoretical program of the project, work has concluded on the calculation of cross-sections for the collision excitation process of luminescent centers in electroluminescent devices. The method involves exclusively the consideration of effects of polarization of the target by the electrostatic field of the incoming electron. Some of the effects of the host lattice on the target are included via the mechanism of expressing the polarizabilities of the target in different states in terms of the depths of the energy levels of those states in the crystal. Applications were made to manganese-doped zinc sulfide and zinc fluoride, to the same hosts doped with TbF_3, and to terbium and europium in yttrium oxide. Comparisons were made with previous estimates and with available experimental data.

Numerical results for several of the materials for which the calculation has been done appear plausable when compared with the experimental estimates available, but the accuracy of the experimental

results remains somewhat doubtful. The many approximations made in the calculation and the difficulty in determining the absolute positioning of the energy levels also leaves some doubt as to the accuracy of the calculated values, though it is expected that they are accurate to within a factor of two or three. The results for the cross section estimates for various materials are collected in the table.

Material	E_g(eV)	ΔE(eV)	$\sigma(cm^2)$
$ZnS:Mn^{2+}$	-9	2.2	2.2×10^{-16}
$ZnF_2:Mn^{2+}$	-11	2.2	1.7×10^{-16}
$ZnS:TbF_3$	-16	2.5	7.4×10^{-17}
$ZnF_2:TbF_3$	-16	2.5	7.4×10^{-17}
$Y_2O_3:Tb^{3+}$	-18	2.5	6.5×10^{-17}
$Y_2O_3:Eu^{3+}$	-21	2.1	6.2×10^{-17}

Cross sections for manganese and rare dopants in various hosts. E_g is the ground-state energy of the luminescent center in the crystal measured with respect to the vacuum, and ΔE is the excitation energy.

In conjunction with the collision excitation work, theoretical considerations of the effects of the hot carrier distribution on the efficiency of operating devices have been made. With the assumption of a Maxell-Boltzmann distribution of electron energies, the fraction of the total energy of the distribution contained in those electrons with energy above the excitation were estimated. It was further assumed that no re-equilibration occurs following each excitation process. It was found

that for reasonable values of the electron temperature and excitation threshold both the fractions mentioned above were lower than actual measured device efficiencies, contrary to expectation. While this casts doubt on the assumptions used, the calculations nevertheless show that the efficiency falls off rapidly with increasing threshold energy and decreasing hot-electron temperature, thus pointing up the need to emphasize research into methods of optimizing the hot-electron distribution. Complete details of this work are given in a Thesis by J. E. Bernard, referenced in part (c).

Experiments were started to measure the energy distribution of the hot carriers in a thin film electroluminescence (TFEL) device. The general approach was to change the TFEL cell by replacing aluminum back contacts with conducting transparent tin oxide (SnO_2). Then a differential optical absorption measurement between an operating and non-operating TFEL cell could be made to deduce the hot carrier distribution. With information on this distribution, the choice of optimum phosphors could be researched in order to gain: (1) an explanation of the loss mechanisms; and (2) basic improvements to the device. The experimental approach was to form a conducting layer of SnO_2 by using reactive sputtering. As this was a new technique in this lab, it required some development work. Essentially, a tin target was sputtered in an argon-oxygen atmosphere to produce Sn_xO_y. Depending upon the cathode voltage, sputtering gas pressure, and cathode-anode spacing, three phases of the compound are possible. This is very similar to the work of Perney et al (G. Perney, Vide, Compt. Rend., 263, 265 (1966)) on work on copper oxides. In that work it was found that variation of E (= cathode voltage/cathode-anode spacing x pressure) led to three distinct phases of copper oxide. Similarly the reactive sputtering

of tin, in an argon-oxygen atmosphere, can lead to similar results (producing Sn, SnO_2, and Sn_2O_4) with only one phase being desirable (SnO_2). With limited success, it was found possible to sputter semi-transparent films that were indeed conductive. Following this, work was done in order to produce a "clear" EL cell. Problems occurred with the final device, in terms of the lifetime of the cell. The technique of reactive sputtering leads to a broader choice of materials for the EL research. It is felt that it would be fruitful to continue with the program of measuring the hot carrier distribution to understand the mechanisms better.

An experimental project was started, of which the object was to get a better quantitative idea of the electrical behavior of thin film EL cells and obtain a better estimate of the range of hot carriers in ZnF_2:Mn thin films.

Samples were initially made by evaporating an SiO layer on a glass substrate coated with a transparent conducting layer (SnO_2). A layer of ZnF_2:Mn was then evaporated, followed by another layer of SnO_2, and a layer of Al contacts. It was found that the conductivity of ZnF_2 was stringly dependent on the doping levels. Hence the method of approach was changed and it was decided to do the experiment varying the thickness of the phosphor layers.

Thus some asymmetric EL cells (with structure SnO_2/ZnF_2:Mn/SiO/Al) were prepared with varying phosphor thickness (ZnF_2:Mn). Brightness vs voltage and I vs V curves were to measured for these films, from which the range of the hot carriers could be determined. Correction for interference effects would have to be made. This work was not completed by the time of expiration of the grant but appeared to be an interesting line of research.

(c) ***Publications***

(1) "Electrical and Optical Properties of a Thin-Film Electroluminescnce Device", by David C. Morton. Dissertation submitted to University of Delaware in partial fulfillment of the requirements for the Ph.D. degree. August 1983.

(2) "Theory of Collision Excitation in Electroluminescence", by James E. Bernard. Dissertation submitted to the University of Delaware in partial fulfillment of the requirements for the Ph.D. degree. June 1984.

(d) ***Participating Personnel.***

Ferd E. Williams, Principal Investigator (*Deceased*, June 1984)

David E. Berry, Part time Research Associate

Michael F. Martens, Part time Research Associate

David C. Morton, Graduate Student. (Received Ph.D. Degree, August 1983)

James E. Bernard, Graduate Student. (Received Ph.D. Degree, June 1984).

Gerald Negley, Graduate Student. (Received M.S. Degree in August 1984).

Sashank Sarwate, Graduate Student.

Charles B. Cooper, Substitute Principal Investigator.

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