Monte Carlo simulation of electron transport in alternating-current thin-film electroluminescent devices

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An ensemble Monte Carlo simulation of electron transport in bulk ZnS at different electric fields is presented. Scattering mechanisms associated with polar optical phonons, acoustic phonons (through deformation potential coupling), intervalley scattering, and impurities (neutral and ionized), are included in a nonparabolic multivalley model. Simulation indicates that the polar optical phonon and intervalley scattering mechanisms are dominant, whereas neutral and ionized impurity scattering are of no significance in determining the high-field electron transport in bulk ZnS. The simulated results show that approximately 26% of the electrons possess total energies exceeding 2.1 eV, the threshold energy for Mn impact excitation, at an electric field of 1 MV/cm. This fraction of electrons with energies exceeding 2.1 eV is estimated to be 50% and 65% at electric fields of 1.5 and 2.0 MV/cm, respectively. Transient overshoot effects are found to be of negligible importance in the operation of alternating-current thin-film electroluminescent (ACTFEL) devices. The steady-state electron distribution at high fields is sufficiently energetic to explain the observed efficiency of ACTFEL devices. No evidence for a significant electron population with energies in excess of 5 eV is found, even during the brief nonstationary regime, and thus very few carriers possess sufficient energy to induce band-to-band impact ionization.

I. INTRODUCTION

In achieving high-resolution, light-weight, compact video display panels for computer terminals or television screens, thin-film electroluminescent (EL) devices show promising performance in comparison with other existing technologies such as cathode ray tube, plasma, and liquid-crystal displays. Either ac or dc power may be used to accelerate majority carrier electrons to energies at which luminescent centers (intentionally introduced into the host material) can be impact excited. The performance deterioration with time of dc EL devices makes ac EL devices more attractive.

The operation of alternating-current thin-film electroluminescent (ACTFEL) devices depends on the physics of hot electrons in the active phosphor layer. There are currently two approaches used to describe ACTFEL electronic transport in the high-field regime. One relies on a mean free path for high-energy electrons determined by energy loss to phonons, known as the lucky-drift model, while the other is band-structure dependent and often based on Monte Carlo simulation. Bringuier used the lucky-drift model to calculate the impact excitation luminescence in ZnS. The lucky-drift model is correct when the mean free path of the electron, \( \lambda_E \), in the drift mode is much larger than the mean free path, \( \lambda \), in the ballistic mode, which is true for wide-gap semiconductors in the high-field regime.

Currently, two different views exist regarding high-field electron transport in ZnS:Mn ACTFEL devices based on Monte Carlo simulation. Brennan performed an ensemble Monte Carlo simulation of steady-state hot electron transport in bulk ZnS, including a full band structure calculation for the density of states in the first two conduction bands and a treatment of electron-phonon scattering including collision broadening. His results have been interpreted as suggesting that very few carriers are available with sufficient energy to excite Mn luminescent centers in ZnS at a field of 1 MV/cm. From this conclusion, it is difficult to explain how such high efficiencies and brightness are obtained in commercially manufactured ACTFEL devices. On the other hand, Müller and co-workers conclude from Monte Carlo calculations and vacuum emission experiments that the electrons in ZnS ACTFEL devices undergo ballistic or loss-free transport, resulting in extremely high-energy electrons. It should be mentioned that these latter Monte Carlo calculations are performed assuming a single, parabolic conduction band with scattering assumed to occur exclusively via polar optical phonons. Furthermore, the experimental electron distributions reported by Müller et al. may be unrealistic since work by Braunlich et al. indicated that the vacuum emission experiment is unreliable in assessing the high-energy tail of the distribution.

We believe that high-field transport in ACTFEL devices lies in between the two extremes suggested above. The intent of the present work is to help elucidate this apparent contradiction through an investigation of transient and steady-state electron transport in the active ZnS layer at high electric fields using an ensemble Monte Carlo simulation. A nonparabolic multivalley model is used, including scattering due to polar optical phonons, acoustic phonons (through deformation potential coupling), intervalley scattering, ionized, and neutral impurities, to ascen-
tain the influence of these mechanisms on the hot carrier transport characteristics.

II. MONTE CARLO MODEL

The Monte Carlo method as applied to semiconductor transport is a simulation of the trajectories of individual carriers as they move through a device under the influence of external forces and subject to random scattering events. The duration of the carrier free flights between successive collisions and the scattering events involved are selected stochastically in accordance with the given transition probabilities describing the microscopic processes. In our model, the conduction band is approximated by nonparabolic multivalley (gamma, L and X) bands, using the dispersion relation

\[ \gamma(E) = E(1 + \alpha_n E) = \frac{\hbar^2 k^2}{2m_n^*}, \]  

where \( \alpha_n \), \( m_n^* \), and \( k \) are the nonparabolicity parameter, effective mass of valley \( n \), and wave vector, respectively. Between collisions, the crystal momentum changes according to the local field, while the velocity of the particle is given by

\[ v = \frac{1}{\hbar} \frac{\partial E}{\partial k}. \]  

The duration of the free flight is given by

\[ t = -\frac{1}{\Gamma} \ln(r), \]  

where \( r \) is a random number uniformly distributed between 0 and 1 obtained from the computer random number generator.

The total scattering rate, \( \Gamma \), is given by

\[ \Gamma = \Gamma_{\text{pop}} + \Gamma_{\text{ac}} + \Gamma_{\text{iv}} + \Gamma_{\text{imp}} + \Gamma_{\text{self}}, \]  

where \( \Gamma_{\text{pop}}, \Gamma_{\text{ac}}, \Gamma_{\text{iv}}, \) and \( \Gamma_{\text{imp}} \) are the various scattering rates considered in the present model due to polar optical phonons, acoustic phonons, intervalley scattering, and impurities (both ionized and neutral), respectively. The self-scattering rate, \( \Gamma_{\text{self}} \), corresponds to a fictitious scattering mechanism that changes with time so that the total rate, \( \Gamma \), is constant. Self-scattering does not change the carrier momentum and energy and, thus, does not affect the carrier's trajectory. Its inclusion is necessary to simplify the random free flight time selection, which is central to the Monte Carlo technique.

The scattering probability for electrons with polar optical phonons is treated assuming the Fröhlich interaction where for nonparabolic bands, the total rate is given by

\[ \Gamma_{\text{pop}}(E) = \frac{e^2 \omega_{\text{op}} \sqrt{2m_n^*}}{\hbar \beta \pi \varepsilon_0} \left( \frac{1}{k_e} - \frac{1}{k_i} \right) \frac{N_n}{\sqrt{\gamma(E)}} \left[ 1 + 2\alpha_n(E + \hbar \omega_{\text{op}}) \right] \ln \left( \frac{\sqrt{\gamma(E) + \sqrt{\gamma(E + \hbar \omega_{\text{op})}}} + N_n + 1}{\sqrt{\gamma(E) - \sqrt{\gamma(E + \hbar \omega_{\text{op})}}} + \sqrt{\gamma(E)}} \right). \]  

where \( k_e \) and \( k_i \) are the high- and low-frequency dielectric constants of the phosphor, respectively, while \( \hbar \omega_{\text{op}} \) is the optical phonon energy.

The deformation potential Ansatz is employed for acoustic and intervalley scattering. For acoustic phonons, the rate is given by

\[ \Gamma_{\text{ac}}(E) = \frac{k_B T D_n^2}{\pi \hbar u_l \rho} \left( 1 + 2\alpha_n E \right) m_n^* \sqrt{2E(1 + \alpha_n E) m_n^*}, \]  

where \( D_n, u_l, \) and \( \rho \) are the acoustic deformation potential, longitudinal acoustic velocity, and mass density of the phosphor, respectively.

Intervalley scattering occurs between nonequivalent valleys (gamma to L, L to gamma, gamma to X, X to gamma, L to X, and X to L), and equivalent valleys (L to L, and X to X) with a scattering rate given by

\[ \Gamma_{\text{iv}}(E) = \frac{D_n^2 Z_{ij} m_n^* \sqrt{m_j^*}}{2\pi \hbar |\rho \omega_{ij}|} \left[ N_n \left[ 1 + 2\alpha_n \left( E + \hbar \omega_{ij} - \Delta E_{ij} \right) \right] \left( E + \hbar \omega_{ij} - \Delta E_{ij} \right) \left[ 1 + \alpha_n \left( E + \hbar \omega_{ij} - \Delta E_{ij} \right) \right] \right] \]  

\[ + \left( N_n + 1 \right) \left[ 1 + 2\alpha_n \left( E - \hbar \omega_{ij} - \Delta E_{ij} \right) \right] \left( E - \hbar \omega_{ij} - \Delta E_{ij} \right) \left[ 1 + \alpha_n \left( E - \hbar \omega_{ij} - \Delta E_{ij} \right) \right]. \]  

\( Z_{ij}, \Delta E_{ij}, \) and \( D_n \) are the number of equivalent valleys, the difference between the energies of the bottoms of the final and initial valleys, and the intervalley deformation potential, respectively, while \( \hbar \omega_{ij} \) is the intervalley phonon energy. For optical and intervalley scattering, the phonon occupation is assumed to follow the Bose–Einstein distribution.

We use the Conwell and Weiskof formula for ionizedimpurity scattering because the phosphor is essentially an insulator with a negligible carrier concentration and very few free carriers (\(< 1 \times 10^{11}/\text{cm}^3\)) are available from interface states in ACTFEL devices to screen the ionized impurity. The scattering rate for ionized impurity scattering is given by
TABLE I. Bulk material parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polar optical-phonon energy (eV)</td>
<td>0.044</td>
</tr>
<tr>
<td>Sound velocity (cm/s)</td>
<td>5.20 x 10^5</td>
</tr>
<tr>
<td>Low-frequency dielectric constant</td>
<td>8.32</td>
</tr>
<tr>
<td>High-frequency dielectric constant</td>
<td>3.60</td>
</tr>
<tr>
<td>Mass density (g/cm^3)</td>
<td>4.08</td>
</tr>
<tr>
<td>Neutral impurity density (cm^-3)</td>
<td>1 x 10^20</td>
</tr>
</tbody>
</table>

\[
\Gamma_{\text{imp}}(E) = \frac{\pi N_i Z^2 b^2}{(1 + 2\alpha_E E)^{3/2}} \sqrt{\frac{2\gamma(E)}{m^*_e}},
\]

where \( N_i \) and \( Z \) are the impurity density and charge of the impurity. In the Conwell and Weisskopf\(^7\) approach, an unscreened potential is assumed, which cuts off at the mean distance between impurities, \( b \), given by

\[
b = \left( \frac{3}{4\pi N_i} \right)^{1/3}.
\]

For scattering by neutral impurities (i.e., \( \text{Mn}^{+2} \)), we use Erginsoy's formula,\(^9\) which provides an energy-independent scattering probability given by

\[
\Gamma_{\text{imp(neutral)}} = \frac{20K N_i b^3}{m^*_e^2},
\]

where \( K = 4\pi\epsilon_0\epsilon_r \) and \( N_i \) are the free space permittivity and the neutral impurity density, respectively. In the literature, two different expressions\(^9,10\) exist for the scattering rate due to neutral impurities. It should be noted that Erginsoy's\(^9\) formula for a parabolic band predicts an energy-independent scattering probability, whereas according to Scarm's\(^10\) formula, the scattering probability should depend on the electron energy. The nature of neutral impurity scattering has not yet been analyzed in sufficient detail to allow us to decide which formula is more appropriate. The exact form of neutral impurity scattering is also expected to vary with the atomic nature of the impurity. We use Erginsoy's\(^9\) formula for the scattering rate to get an idea of its importance in determining the transport properties in ACTFEL devices.

TABLE II. Valley-dependent parameters.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Gamma</th>
<th>( L )</th>
<th>( X )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Effective mass ((m^*/m_0))</td>
<td>0.28</td>
<td>0.222</td>
<td>0.40</td>
</tr>
<tr>
<td>Nonparabolicity ((\text{eV}^{-1}))</td>
<td>0.690</td>
<td>0.650</td>
<td>0.360</td>
</tr>
<tr>
<td>Valley separation (eV)</td>
<td>--</td>
<td>1.449</td>
<td>1.454</td>
</tr>
<tr>
<td>Number of equivalent valleys</td>
<td>1</td>
<td>4</td>
<td>3</td>
</tr>
<tr>
<td>Intervalle deformation potential (eV/cm)</td>
<td>from gamma</td>
<td>0</td>
<td>( 1 \times 10^6 )</td>
</tr>
<tr>
<td>from ( X )</td>
<td>( 1 \times 10^8 )</td>
<td>( 9 \times 10^4 )</td>
<td>( 9 \times 10^8 )</td>
</tr>
<tr>
<td>from ( L )</td>
<td>( 1 \times 10^7 )</td>
<td>( 9 \times 10^4 )</td>
<td>( 9 \times 10^8 )</td>
</tr>
<tr>
<td>Intravalle deformation potential (eV)</td>
<td>4.9</td>
<td>4.9</td>
<td>4.9</td>
</tr>
<tr>
<td>Intravalle phonon energy (eV)</td>
<td>from gamma</td>
<td>0.0</td>
<td>0.0267</td>
</tr>
<tr>
<td>from ( X )</td>
<td>0.0279</td>
<td>0.0273</td>
<td>0.0279</td>
</tr>
<tr>
<td>from ( L )</td>
<td>0.0267</td>
<td>0.0267</td>
<td>0.0273</td>
</tr>
</tbody>
</table>

\(^a\)See Ref. 11.

The type of scattering terminating a free flight is chosen at random according to the relative scattering rates at the end of the flight. The final angle is then chosen randomly according to the differential scattering cross section. Several thousand particles are simulated simultaneously and ensemble averages are performed to obtain the quantities of interest such as the energy and velocity.

The material parameters used in the present Monte Carlo simulation are collected in Tables I and II.\(^3\) Table I deals with the valley-independent while Table II with the valley-dependent parameters of ZnS. In order to compare our simulation to that obtained\(^3\) by Brennan, we employed the same material and valley-dependent parameters as used by Brennan.

III. CALCULATED RESULTS

The scattering rates due to the various scattering mechanisms as a function of energy for the gamma valley are shown in Fig. 1 at 300 K. For electron energies less than about 1.7 eV, polar optical phonon scattering is the dominant scattering mechanism, whereas above 1.7 eV intervalley scattering is most important. As seen in Fig. 1, the gamma to \( X \) intervalley scattering rate is larger than the gamma to \( L \) rate; this is a consequence of the larger \( X \) valley density of states. Thus, the \( X \) valley is preferentially populated in comparison to the \( L \) valley. Figure 1 shows acoustic phonon scattering to be more important at higher electron energies but it is never a dominant factor in establishing the total scattering rate. Ionized impurity scattering is more important at low electron energies and is of negligible significance in determining the overall scattering rate of heated carriers relevant to ACTFEL performance. An ionized impurity concentration of \( 10^{19}/\text{cm}^3 \) is assumed in the calculated rate shown in Fig. 1. Even for this high concentration, the magnitude of the ionized impurity scattering rate is rather small and decreases with increasing electron energy. This result underscores the irrelevance of ionized impurity scattering for transport in ACTFEL devices that operate at high electric fields. Finally, note that the gamma valley scattering rate due to neutral impurities is too small (approximately \( 10^{11}/\text{s} \)) to be plotted on Fig. 1, even when a density of \( 10^{20}/\text{cm}^3 \) (corresponding to an
atomic doping concentration 0.5% in cubic ZnS, which is the approximate Mn concentration) is assumed. Clearly, neutral impurity scattering can be ignored.

Figure 2 shows the variation of the steady-state drift velocity as a function of the applied electric field for three different ionized impurity concentrations at 300 K. Brennan’s steady-state results are also included for comparison. The calculated steady-state velocity-field curve (Fig. 2) is in agreement with Brennan’s at high fields but deviates from his result at low fields. From the slope of the steady-state drift velocity versus electric field curve, the mobility at 1 kV/cm is found to be 250 cm²/V s with the inclusion of polar optic phonon, deformation potential acoustic phonon, and intervalley scattering mechanisms. The low-field mobility from Brennan’s result is 1700 cm²/V s at 1 kV/cm. The experimental low-field mobility data for the ZnS system lies in the range of 79 to 280 cm²/V s and is in much better agreement with our low-field mobility value. Inclusion of ionized impurity scattering with an impurity density of $1 \times 10^{19}$/cm³ brings the mobility to 59 cm²/V s. As shown in Fig. 2, ionized impurity scattering affects the slope of the velocity-field curve for fields less than 100 kV/cm and, thus, dominates the low-field mobility for impurity densities in excess of $10^{18}$/cm³. From Fig. 2 it is evident from the position of the peak of the velocity-field curve that the threshold field for intervalley transfer in the present study is twice that obtained by Brennan. This discrepancy in the threshold field is a consequence of differences in the low-field mobility (i.e., Brennan’s larger low-field mobility enables the electrons to heat up to higher energies at a given field and, thus, undergo intervalley scattering at a lower field). We believe that our estimate of the peak field is more accurate because of the unrealistically large low-field mobility reported by Brennan.

In Fig. 3 we plot the average velocity versus time for three different applied electric fields without ionized impurity scattering. From this figure it is clear that steady state is reached within a few tenths of a picosecond. Figure 4 gives the variation of the average electron energy with time for three different electric fields. Although there is a short time scale overshoot in the electron velocity before intervalley transfer to the L and X valleys occurs, there appears to be no significant overshoot in the energy of the particles during the traversal time in the phosphor layer, which is approximately one to two picoseconds based on the saturated velocity at high field.

Figures 5 and 6 show the steady-state electron energy distribution, $n(E)$, as a function of energy considering the total energy (kinetic plus potential) of the electrons. In Fig. 5 we plot the sum of the electron populations in the three different valleys for various electric fields. For comparison, we plot the calculated Mn impact excitation rate taken from the calculation of impact excitation cross section $\sigma(E)$ by Shen and Xu, which is based on the full band structure, rather than the nonparabolic model used in the present Monte Carlo simulation. To calculate the impact excitation rate, a value of 500 µs is used for the life...
IV. DISCUSSION

If we assume parabolic bands for the gamma, X, and L valleys, our Monte Carlo simulation exhibits electron run away, consistent with ballistic or loss-free transport, at a field 100 kV/cm and higher, in agreement with Müller and co-workers.4,5 However, if conduction band nonparabolicity is included in the Monte Carlo simulation, we no longer observe electron run away and the electron distribution is stable, i.e., nonparabolicity stabilizes the electron distribution by increasing the scattering rate because of the increased density of final states.

Ionized impurities only have an effect at low fields because the scattering rate decreases with increasing carrier energy. The low-field mobility is found to be dominated by ionized impurity scattering for impurity concentrations $10^{18}$/cm$^3$ and above. In contrast, acoustic phonon scattering is more important at higher electron energies. However, our Monte Carlo simulation indicates this mechanism to be of secondary importance in determining the operation of ACTFEL devices. Neutral impurity scattering does not play a role in determining the electron transport properties in ZnS. The calculation shows that the neutral impurities are ineffective in affecting transport due to their low scattering rate ($\approx 10^{11}$/s), compared to other scattering mechanisms, even for densities on the order of $1 \times 10^{20}$/cm$^3$. Therefore, at the fields necessary to excite luminescent transitions, both ionized and neutral impurities play little role in establishing the device performance and may be neglected in most cases. Thus, from a transport perspective this result implies that there is little incentive to go to advanced material growth technologies in order to obtain purer phosphor materials since impurity scattering does not appear to be an issue of importance.

As seen from the Figs. 5 and 6, at fields on the order of 1 MV/cm and larger a significant fraction of the total electron population, $n(E)$, resides at energies exceeding the threshold for Mn luminescence excitation (i.e., 2.1 eV). Figure 5 shows that the peak of the $n(E)$ versus energy curve shifts towards higher energy as the electric field increases. This trend implies that at higher phosphor fields, the carriers are heated to higher average energies. Note that there is a substantial overlap of the hot electron distribution with the Mn excitation cross section, a desirable situation for efficient excitation of luminescent impurities.

Our results differ quantitatively from those of Brennan1 (although the qualitative trends are not in serious disagreement) and are also in reasonable agreement with lucky-drift modeling.14 There has been some confusion in the interpretation of Brennan's Monte Carlo results be-

FIG. 4. Average electron energy vs time for three different ZnS electric fields. Impurity scattering is not included in these calculations.

FIG. 5. Steady-state electron energy distribution, $n(E)$, and Mn impact excitation rate, $R(E)$, as a function of energy. Impurity scattering is not included in these calculations.

FIG. 6. Steady-state electron energy distribution in each of the three valleys (gamma, L, and X) as a function of energy. Impurity scattering is not included in this calculation.
cause he plotted a normalized electron energy distribution (i.e., the hot electron distribution normalized by the ZnS density of states), \( f(E) \), instead of simply the electron energy distribution, \( n(E) \). Confusion arises in evaluating \( n(E) \) from \( f(E) \) because normalization by the ZnS density of states prevents one from assessing the shape of the original distribution. Our results show that at fields in the range of 1.5–2 MV/cm, corresponding to the field-clamping regime in ZnS ACTFEL devices, the average electron energy exceeds 2 eV, and thus there is an adequate supply of energetic carriers for efficient electroluminescence to occur. Brennan's Monte Carlo simulation shows that 1% of the electrons exceed 2.1 eV at an electric field of 1 MV/cm while lucky-drift modeling gives 19% at 1 and 66% at 2 MV/cm, respectively. This compares to our results of 26% at 1 MV/cm, 50% at 1.5 MV/cm, and 65% at 2 MV/cm, respectively. From a comparison of the Mn impact excitation rate with the electron energy distribution (i.e., Fig. 5), it is found that the percentage of electrons between 2.1 and 3.2 eV, for which the impact excitation cross section is significant, is 26%, 50%, and 54% for electric fields of 1, 1.5, and 2 MV/cm, respectively. Since the impact excitation rate decreases to zero above 3 eV, higher energy electrons cannot excite the luminescent centers. Note, however, that a more realistic calculation of the impact excitation cross section, which includes higher energy excited states, would result in a nonzero impact excitation cross section for energies above 3 eV. An important message from the impact excitation cross section is that electron heating above a certain energy does not lead to efficient electroluminescence.

From Fig. 5, we estimate that approximately 2% of the electrons in the distribution possess energies in excess of 2.75 eV, the approximate threshold energy for blue emission at a field of 1 MV/cm. Approximately 13% of the electrons have energies in excess of 2.75 eV at 1.5 MV/cm and 27% at 2 MV/cm.

No evidence for a significant electron population higher than 5 eV is found for fields up to 2 MV/cm, even during the nonstationary regime, in contrast to the results of Müller and co-workers. Note from Fig. 5 that very few electrons are available for band-to-band impact ionization via electron-hole pair generation since we estimate, the threshold for band-to-band impact ionization to occur at approximately 4.3 eV. Thus, we do not expect band-to-band impact ionization to play an important role in the device physics operation of ZnS ACTFEL devices.

V. CONCLUSIONS

Monte Carlo simulation indicates polar optical phonon and intervalley scattering mechanisms to be of dominant importance, whereas neutral and ionized impurity scattering are of no significance, in determining the high-field electronic transport properties of bulk ZnS. Nonstationary transport effects, such as velocity overshoot, are found to play no role in the operation of an ACTFEL device since the total scattering rate is found to be extremely large at the exceedingly large fields at which these devices operate. The average energy of an electron transiting a ZnS ACTFEL device exceeds 2 eV under normal field-clamping conditions such that the electron distribution is hot enough to account for the observed efficiency of commercial ACTFEL devices. The present Monte Carlo calculations show that the high energy tail of the hot electron distribution occurs below 4 eV; this would seem to preclude the occurrence of band-to-band impact ionization since the threshold energy is approximately 4.3 eV for this process.

ACKNOWLEDGMENT

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