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Transient acceleration process of electrons in ZnS-type thin film electroluminescence devices

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Abstract. The transient acceleration process of electrons in ZnS-type thin film electroluminescent devices is studied in detail for the first time through the Monte Carlo method. The transient time and drift length before the balance of acceleration and scattering are of the order of 200 fs and 30 nm respectively. The steady average kinetic energy of electrons increases with electric field. Field emission processes of electrons trapped at interface states only affect the transient process of electron transport and have no influence on the steady state. New explanations of the 'dead layer' phenomenon and the overshoot in the average drift velocity are proposed based on our calculation results.

1. Introduction

The technology of thin-film electroluminescent displays (TFELDs) has advanced rapidly since the 1980s. Yellow-emitting ZnS:Mn TFELDs are now commercially available in a variety of sizes and resolutions [1]. But the development of full colour displays has been postponed for several years due to the poor luminescence and efficiency of blue emission. The overcoming of this problem needs more detailed investigations on materials, devices and electroluminescence mechanisms.

In TFELD structures, a thin phosphor layer such as ZnS, which is sandwiched between two insulating layers, is subject to a large electric field (typically 1–3 MV cm^{-1}) when an ac voltage is applied. Due to the field emission into the conduction band from trapped interface states at these field strengths, electrons are accelerated to high enough energies to impact and excite luminescent centres, giving off visible light useful for display applications [2]. Since the light emission process depends sensitively on the fraction of carriers above the threshold energy for impact excitation (2–2.6 eV), the electron transport process in the phosphor layer is one of the key processes in electroluminescence. This transport process is composed of two sequential stages. As the electrons are emitted into the conduction band, their energies are lower. Then these electrons are accelerated by the electric field. This is the transient acceleration stage. Since several scattering mechanisms exist in materials and their rates increase with electron energy, this acceleration is influenced by scattering mechanisms more and more seriously as the average electron energy increased. Finally, a steady transport stage is reached in which the average energy is fixed at a stable value due to the balance between the field acceleration and the scattering of electrons.

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Although many studies have been done on the electron transport process, most of them are concerned with the steady transport process. Detailed reports on the transient acceleration process are not available to the authors' knowledge. In fact, this process is important since it determines the luminescent properties during the early stage of the applied voltage pulse and influences the steady transport process. In this article, the transient acceleration process of electrons in ZnS-type TFELD is investigated in detail by means of the Monte Carlo method.

2. Monte Carlo method

The Monte Carlo method is a simulation of the motion of individual carriers as they move under the influence of external fields and are scattered by various mechanisms [3]. This motion is composed by a series of 'free flight' processes terminated by the scattering events. During the 'free flight' process, the influence of scattering mechanisms is not considered and the motion could be described as

$$\frac{\mathrm{d}}{\mathrm{d}t}(\hbar\vec{k}) = -e\vec{E}$$

where $\hbar$ is the reduced Planck constant, e is the electronic charge, $\vec{E}$ is the electric field strength and $\vec{k}$ is the wave vector. The free flight time is chosen stochastically according to the total scattering rate,

$$t_r = -\frac{1}{\Gamma}\ln(r_1)$$

where Γ is the total scattering rate (the sum of rates of all scattering mechanisms considered) and r_1 is a random number generated by the program and is uniformly distributed between 0 and 1.

At the end of this free flight ($t = t_r$), another random number uniformly distributed between 0 and 1, r_2, is generated by the program to choose a certain kind of scattering from all of the scattering mechanisms. If there are n kinds of scattering mechanism and the rate of the ith is W_i, then the probability of occurrence of the mth scattering is

$$W_m\left(\sum_{i=1}^{n} W_i\right)^{-1}.$$

If

$$\sum_{i=1}^{m-1} W_i\left(\sum_{i=1}^{n} W_i\right)^{-1} < r_2 \leqslant \sum_{i=1}^{m} W_i\left(\sum_{i=1}^{n} W_i\right)^{-1}$$

the mth scattering mechanism is chosen.

Once a scattering mechanism has been chosen, the momentum and the energy of the electron are updated as determined by the specific mechanism that has been selected. Then the next 'free flight' process begins until it is again terminated by the next scattering event. This cycle is repeated many times. The motion of an electron in phosphor layer is thus simulated.

We have studied several important scattering mechanisms in ZnS, including acoustic phonon scattering, polar optical phonon scattering, ionized impurity scattering and intervalley scattering. The nonparabolic multivalley band model is adopted instead of the full band model for saving computation time. Although the difference between these two models is evident in the high energy region, i.e. above 2.0 eV, and this factor will affect the accuracy of the calculation results, they are coincident when the energy is lower. This is just the case for the transient acceleration process and this makes our calculation valid. We gained the rates of each kind of scattering at different energies and the relations between the final and initial states of

scattering. The calculation methods and the parameters used are the same as in [4]. Based on these calculations, the electron transport process is simulated using the Monte Carlo method described above. The number of simulated electrons is 200 000, which has been proved to be enough to assure the accuracy and repeatability of the results.

3. Results and discussion

3.1. Electron kinetic energy distribution

The electron kinetic energy distribution function (EKEDF) is one of the basic functions for the study of electroluminescent process, such as impact excitation, band-to-band ionization etc. The transient variation process of EKEDF is shown in figure 1. The shift of electrons from the low to high energy region is the result of acceleration. According to our calculations, the transient time of EKEDF is about 200 fs. After that, the function remains unchanged, which indicates that the steady stage is reached. We have studied this process under different electric fields (0.5–5.0 MV cm^{-1}). The results are similar and the transient times are all about 200 fs. Figure 1 shows only the case of 2.0 MV cm^{-1}. However, for the band model limitation, errors must exist in the high energy region in this figure. So this figure just gives the revolution of the EKEDF and could not be used in steady state analysis, such as impact excitation.

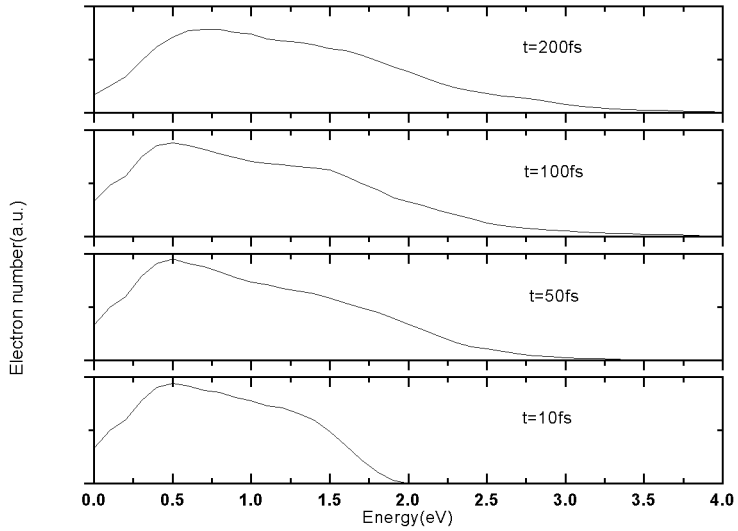

Figure 1. Transient process of electron kinetic energy distribution under 2.0 MV cm^{-1} electric field.

3.2. Average kinetic energy of electrons

Average kinetic energy (AKE) reflects the main characters of the EKEDF. By using the AKE, the variation of electronic states with other factors could be studied quantitatively. Figure 2 shows the variation of AKEs with time under different electric fields. The initial values of AKE are to be set as 0.5 eV. From this figure, although the steady value of AKE increases with electric field, the transient times are all about 200 fs under different fields. This coincides with the case of EKEDF. Under the fields of 0.5 and 1.0 MV cm^{-1}, AKEs decrease with time during

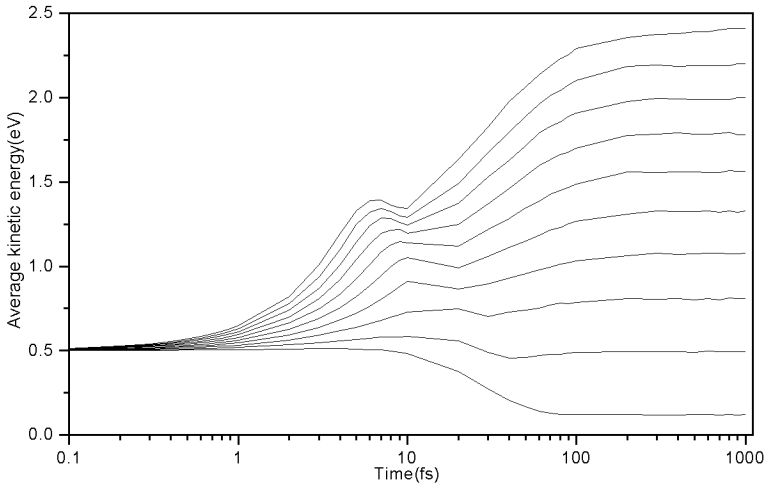

Figure 2. Transient process of average kinetic energy under different electric fields (curves from bottom to top correspond to the electric field of 0.5, 1.0, . . . , 5.0 MV cm^{-1}).

the transient process. This is due to the low field strength since under these fields the steady values of AKE are lower than 0.5 eV. It is worth noting that a peak appears around 5–10 fs in each curve when the electric field is above 1.5 MV cm^{-1}. The formation of these peaks will be discussed below.

The initial energy distribution is determined by the field emission process of electrons from trapped interface states to conduction band. This process has been investigated in detail, for example, by Bringuier [5]. But the quantitative description of this process is still very difficult because of the lack of material parameters. In order to investigate the influence of the field emission process on the electron transport process, we calculated the variation of AKE with time for different initial AKE values (0–1.6 eV). The results are shown in figure 3. According to this figure, the transient time and steady AKE do not vary with initial AKE. That is, the steady AKE is determined by the field strength and scattering mechanisms of the material. The field emission process has no influence on the steady AKE.

3.3. The average drift velocity of electrons

The motion of electrons in the phosphor layer is anisotropic when a voltage is applied and the privileged direction is the positive field direction. The average drift velocity (ADV), which is the algebraic average of velocity component in this direction of all electrons to be considered, describes the characters of the drift motions of electron under the electric field. This is also a basic quantity for the study of the electroluminescent process, especially when the spatial effects are considered. The transient processes of ADV under different electric fields are studied and the results are shown in figure 4. Although ADVs go through different transient processes under different electric fields, the transient times are almost the same, 200 fs.

As discussed above, the transient times of EKEDF, AKE and ADV are all about 200 fs in different fields and different initial conditions. It will be seen in the next section that the transient time of intervalley transfer is also 200 fs. That is, the transient acceleration time is roughly equal to the intervalley transfer time. It must be mentioned that, for lack of the experimental data, some parameters for intervalley scattering used in this calculation are taken

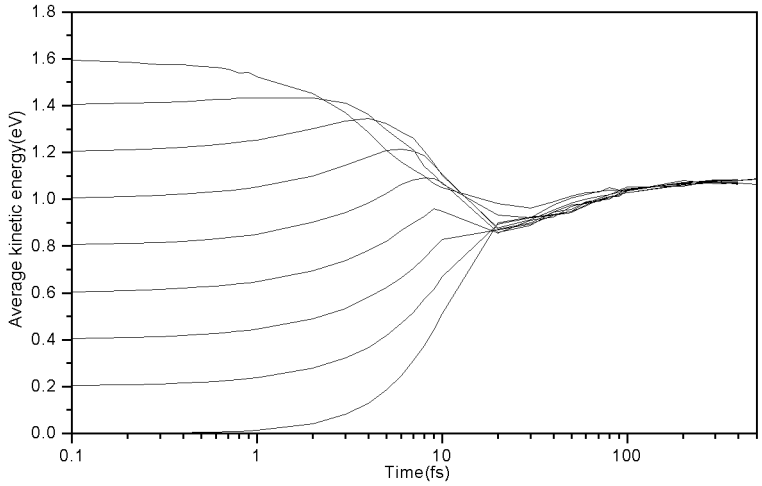

Figure 3. Transient process of average kinetic energy in different initial conditions (curves from bottom to top correspond to the initial energy of 0.0, 0.2, 0.4, . . . , 1.6 eV).

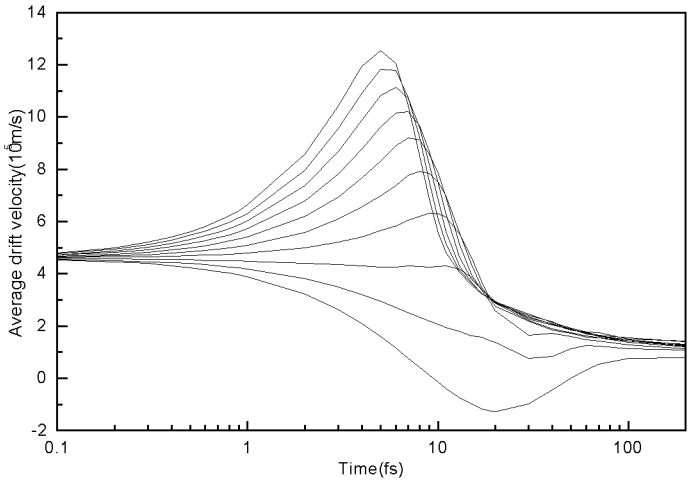

Figure 4. Transient process of average drift velocity in different electric fields (curves from bottom to top correspond to the electric field of 0.5, 1.0, . . . , 5.0 MV cm^{-1}).

from GaAs and InP, as done in [4] and [8]. This may introduce some uncertainty into the calculation result. So we can only say that the transient time of the electron transport process in a ZnS-type TFELD is of the order of 200 fs. According to the calculation results of ADV, the flight distance of electrons in this period is of the order of 30 nm. For a common TFELD, this is about one-tenth of the total phosphor layer thickness.

This result could be used to provide a possible explanation of the 'dead layer' phenomenon in TFELDs. In TFELDs, the part of the phosphor layer near the interface cannot emit light. This part is called the 'dead layer'. The thickness of this layer is 30 nm approximately [6]. This phenomenon has been ascribed to the poor crystalline of this part of phosphor layer [7]. According to our results, the transient length of electron transport is of the order of 30 nm, coinciding with the thickness of the 'dead layer'. We propose that electrons are accelerated and

do not reach the steady AKE in this region so that they cannot impact excite luminescent centres effectively. That is, even if this part of the phosphor layer is crystalline, luminescence also cannot be observed in the 'dead layer'. This phenomenon is an intrinsic property of TFELDs. We just provide a possible explanation for this phenomenon because of the limitation of the band model and material parameters and more experimental investigations are needed.

Just as in the case of AKE, a peak also appears around 5–10 fs in each curve of ADV. The appearance of this peak has been found by Brennan [8] and Fogarty *et al* [9]. Brennan made no explanation of it and Fogarty ascribed this to the anisotropy of polar optical phonon scattering [9]. To verify this explanation, we get rid of the polar optical scattering mechanism and carry out the same simulation. The peak also appears. Then we can say that Fogarty's explanation is not reasonable. In fact, since the peaks also appear in the curves of AKE, it cannot be the result of some anisotropy. In order to find out the real reason for this phenomenon, we studied the intervalley transfer process in TFELDs.

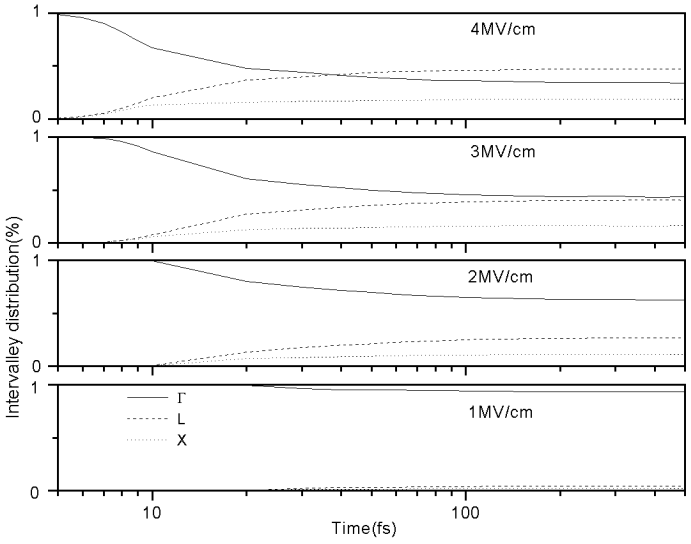

Figure 5. Intervalley transfer processes in different electric fields.

3.4. Intervalley transfer process

There are three valleys in the conduction band of ZnS, Γ, L and X valleys. In the field emission process, most of the electrons are emitted into the Γ valley (the lowest valley). Then the energy of electrons increases due to the acceleration effect of the electric field. Electrons begin to transfer to higher valleys (L and X valleys). Finally, the intervalley distribution of electron reaches a steady state in which electron transfer between these valleys keep a dynamic balance. Figure 5 shows the intervalley transfer processes under different electric fields. The transient times are all about 200 fs. The steady distribution shifts to higher valleys as the field increases. In the case of 1.0 MV cm^{-1}, the electron energies are too low. The intervalley transfer could not occur. In other cases, intervalley transfer is obviously. The beginning times of intervalley transfer are 10, 7 and 5 fs in the case of 2.0, 3.0 and 4.0 MV cm^{-1} field respectively. Comparing figure 2 with figure 4, we find that the appearance of the peak and the beginning time of intervalley transfer are all earlier under the higher field. Furthermore, under

a given field, these two times are almost equal. Based on these results, we proposed that the appearances of peaks in AKE and ADV curves are the results of electron intervalley transfer. In the early stage, most of the electrons are in the Γ valley. AKE and ADV increase with time. When the electrons become energetic enough to transfer to a higher valley, a part of the kinetic energies is converted to potential energies since the bottom of the Γ valley is lower than that of the others. So the AKE and ADV are lower and the peaks appear. To verify this explanation, we studied the transient process of total average energy (including kinetic and potential). In the curves of the total average energy, no peak appears. This confirms our explanation.

4. Conclusion

The transient acceleration process of electrons transport in ZnS-type TFELDs is studied by means of the Monte Carlo method. The transport curves of EKEDF, AKE, ADV and intervalley distributions in different conditions are gained. The transient time and length are of the order of 200 fs and 30 nm respectively. The steady AKE increases with field. The field emission process of electron only affects the transient process of electron transport and has no influence on the steady state. A new possible explanation is proposed for the 'dead layer' phenomenon based on the transient property of electron transport. The peaks appearing in AKE and ADV curves are also explained successfully based on the intervalley transfer process. To the authors' knowledge, this is the first detailed investigation of the electron transient acceleration process in TFELDs and the results could be used in the further study of the electroluminescent mechanism.

Acknowledgment

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References

[1] Ono Y A 1995 *Electroluminescent Displays* (Singapore: World Scientific) p 35
[2] Bringuier E 1997 High-field conduction in semi-insulating ZnS films *Phil. Mag.* B **75** 209–28
[3] Jacoboni C and Lugli P 1989 *The Monte Carlo Method for Semiconductor Device Simulation* (New York: Springer) pp 104–27
[4] Bhattacharyya K, Goodnick S M and Wager J F 1993 Monte Carlo simulation of electron transport in alternating-current thin-film electroluminescent devices *J. Appl. Phys.* **73** 3390–5
[5] Bringuier E 1989 Charge transfer in ZnS-type electroluminescence *J. Appl. Phys.* **66** 1314–25
[6] Sasakura H, Kobayashi H, Tanaka S, Mita J, Tanaka T and Nakayama H 1981 The dependence of electroluminescent characteristics of ZnS:Mn thin films upon their device parameters *J. Appl. Phys.* **52** 6901–6
[7] Ono Y A 1995 *Electroluminescent Displays* (Singapore: World Scientific) p 23
[8] Brennan K 1988 Theory of high-field electronic transport in bulk ZnS and ZnSe *J. Appl. Phys.* **64** 4024–31
[9] Fogarty J, Kong W and Solanki R 1995 Monte Carlo simulation of high field electron transport in ZnS *Solid-State Electron.* **38** 653–60