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The calculated low-energy side of the luminescence spectrum in zinc selenide

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ABSTRACT

In this paper we consider electron-hole recombination in wide gap semiconductor ZnSe (Zinc Selenide) under electric fields. The calculated low energy side of the luminescence spectrum displays the so-called Urbach's tail, which is characterized as resulting from the presence of side bands in the form of replicas of the main band, corresponding to recombination with accompanying emission of one, two, three, etc., LO-phonons. Through the numerical solution of associate quantum transport equations based on the Non Equilibrium Statistical Operator Method, the carrier drift velocity and the nonequilibrium temperatures of electrons and phonons were obtained and the dependence on the electric field strength was determined. The influence of the applied electric field on the luminescence spectrum is evidenced. Our results for electric fields intensities of 5–25 kV/cm points to 15.7–19.7 meV Urbach tail widths in ZnSe.

1. Introduction

The ZnSe (Zinc Selenide) is a wide-gap II-VI zincblende semiconductor. The ZnSe is an attractive semiconductor for various optoelectronic devices, for instance laser diodes [1–3]. The ZnSe semiconductor has received particular attention nowadays as a consequence of the large technological interest associated with their applications, and under the influence of high electric fields, the compound is driven to thermodynamic states far from equilibrium. Their main transport and optical properties are then strongly dependent on such nonequilibrium macrostates, and it is quite relevant to have a description of the latter and a knowledge of what is expected to be the behavior of the system in such conditions.

In this work, it is addressed an analysis of the low energy side of the luminescence spectrum displays the so-called Urbach's tail in the semiconductor ZnSe, subjected to electric fields up to 25 kV/cm. For this we used the *Nonequilibrium Statistical Operator Method* (NESOM) [4–7]. The NESOM is a powerful formalism that seems to offer an elegant and concise way for an analytical treatment in the theory of irreversible processes, adequate to deal with a large class of experimental situations [8], and physically clear picture of irreversible processes, as for example in far-from equilibrium semiconductors [9] which is the case considered here. This method (NESOM) was used in numerous works for studying transport processes and kinetic phenomena (see for example Refs. [10–12]).

The spectral Urbach's tail [13,14] consists of a nearly exponential side in the low energy region of the absorption and luminescence spectra extending inside the forbidden band gap below the bottom of the conduction band in the electron-hole recombination processes, or in the low frequency side of the exciton recombination line, depending on the carriers being or not, respectively, on the metallic side of Mott transition. It is the result of a superposition of effects corresponding to the presence of phonon-assisted side bands (of “hot” phonons in the treatment below) together with the influence of carriers’ (“hot” carriers here) lifetime (due to scattering by phonons and impurities, as well as carrier-carrier collisions) and self-energy corrections [13–17].

We address here the study of the recombination spectrum of electrons and heavy-holes in the semiconductor ZnSe subjected to electric fields, with particular attention to the low energy side of the spectrum which displays the so-called Urbach's tail. The optical response consisting of the luminescence spectrum of the “hot” carriers in ZnSe is analyzed considering their nonequilibrium state, to which are driven by the effect of energy and momentum transfer from an electric field of intensity $\mathscr{E}$. We characterize the Urbach's tail as resulting from the superposition of photoluminescense side bands in the form of replicas of the main band, corresponding to recombination with accompanying emission of one, two, three, etc. LO-phonons.

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2. Transport equations

We consider here the case of *n*-doped semiconductor ZnSe in contact with a reservoir at temperature $T_0 = 300$ K with an constant electric field $\mathcal{E}$ applied. The electric field drives the system out of equilibrium, and the time-dependent macroscopic state is described in terms of a statistical thermodynamics for irreversible systems. For the description of the nonequilibrium state of *n*-doped ZnSe under electric fields, we resort to a *Nonlinear Quantum Transport Theory* [18–20] derived from the NESOM [4–7]. According to the theory, the nonequilibrium macroscopic state of the system can be described in terms of a set of intensive nonequilibrium thermodynamic variables.

In the present case, the nonequilibrium thermodynamic state of the system is characterized by the *macrovariables*: carriers' linear momentum along the direction of the electric field $\mathcal{E}$: $\mathbf{P}(t)$; carriers' energy: $E_c(t)$; the energy of the longitudinal optical phonons: $E_{LO}(t)$; and the energy of the acoustic phonons: $E_{AC}(t)$. The transversal optical phonons have been ignored because the deformation potential interaction with electrons in the conduction band is negligible [21].

On the other hand, the *basic intensive nonequilibrium variables* are: the carrier drift velocity: $\mathbf{v}(t)$; the nonequilibrium temperature of carriers: $T_c^*(t)$; the nonequilibrium temperature of longitudinal optical phonons: $T_{LO}^*(t)$; and the nonequilibrium temperature of acoustic phonons: $T_{AC}^*(t)$. We emphasize that the *basic intensive nonequilibrium variables* $\{\mathbf{v}(t), T_c^*(t), T_{LO}^*(t), T_{AC}^*(t)\}$ are associated, respectively, to the *macrovariables*: $\{\mathbf{P}, E_c(t), E_{LO}(t), E_{AC}(t)\}$. [20]

We shall restrict the situation to the conditions of weak to moderate electric field intensities so that the parabolic band approximation is acceptable, i.e., we take $\epsilon_{\mathbf{k}} = \hbar^2\mathbf{k}\cdot\mathbf{k}/2\,m^*$. However, nonparabolicity and side valleys in a conduction band is significant in high-field transport processes (see, for example, Ref. [22]).

The equations of evolution for these basic macrovariables are derived in the nonlinear quantum kinetic theory described in Ref. [20]. They are

$$\frac{dE_c(t)}{dt} = -\frac{e\mathcal{E}}{m^*}\cdot\mathbf{P}(t) + J_{E,LO}^{(2)}(t)\,, \tag{1}$$

$$\frac{d\mathbf{P}(t)}{dt} = -nVe\mathcal{E} + \mathbf{J}_{\mathbf{P}}^{(2)}(t)\,, \tag{2}$$

$$\frac{dE_{LO}(t)}{dt} = J_{LO}^{(2)}(t) - J_{LO,an}^{(2)}(t)\,, \tag{3}$$

$$\frac{dE_{AC}(t)}{dt} = J_{AC}^{(2)}(t) + J_{LO,an}^{(2)}(t) - J_{AC,dif}^{(2)}(t)\,, \tag{4}$$

In Eq. (1), the first right hand term accounts for the rate of energy transferred from the electric field to the carriers, and the second term accounts for the transfer of the excess energy of the carriers to the phonons. In Eq. (2), the first right hand term is the driving force generated by the presence of the electric field. The second term is the rate of momentum transfer due to interaction with the phonons and ionized impurities. In Eqs. (3) and (4), the first right hand term describes the rate of change of the energy of the phonons due to interaction with the carriers. More precisely they account for the gain the energy transferred to then from the hot carriers and then the sum of contributions $J_{LO}^{(2)}(t)$ and $J_{AC}^{(2)}(t)$ is equal to the last term in Eq. (1), with change of sign. The second term in Eq. (3) accounts for the rate of transfer of energy from the optical phonons to the acoustic phonons via anharmonic interaction. The contribution $J_{LO,an}^{(2)}(t)$ is the same but with different sign in Eqs. (3) and (4). Finally, the last term in Eq. (4) is the diffusion of heat from the AC phonons to the reservoir. We notice that because the concentration n of electrons is fixed by doping $dn/dt = 0$. The detailed expressions for the collision operators are given in Ref. [20].

The set of coupled differential Eqs. (1)–(4) were resolved numerically for $n = 10^{17}$ cm^{-3}. The time evolution of the basic intensive nonequilibrium variables $\{T_e^*(t), T_{LO}^*(t), T_{AC}^*(t), \mathbf{v}(t)\}$ were obtained for different values of the electric field strength. To perform the numerical calculations, for the particular case of *n*-doped ZnSe, we have used the same parameters listed in Table 1 of Ref. [23]. In the presence of the constant electric field, after a very rapid (picosecond scale) transient has elapsed, a steady state sets in, when then the basic thermodynamic variables become constant in time. In this case: $dE_c(t)/dt = 0$, $d\mathbf{P}(t)/dt = 0$, $dE_{LO}(t)/dt = 0$, $dE_{AC}(t)/dt = 0$.

Table 1
Values of thermodynamic variables for two values of the electric field intensities.

Thermodynamic variables	5 kV/cm	25 kV/cm
T_e^* (K)	307.4	581.7
T_{LO}^* (K)	300.07	301.8
v_e (10^7cm/s)	0.28	1.42

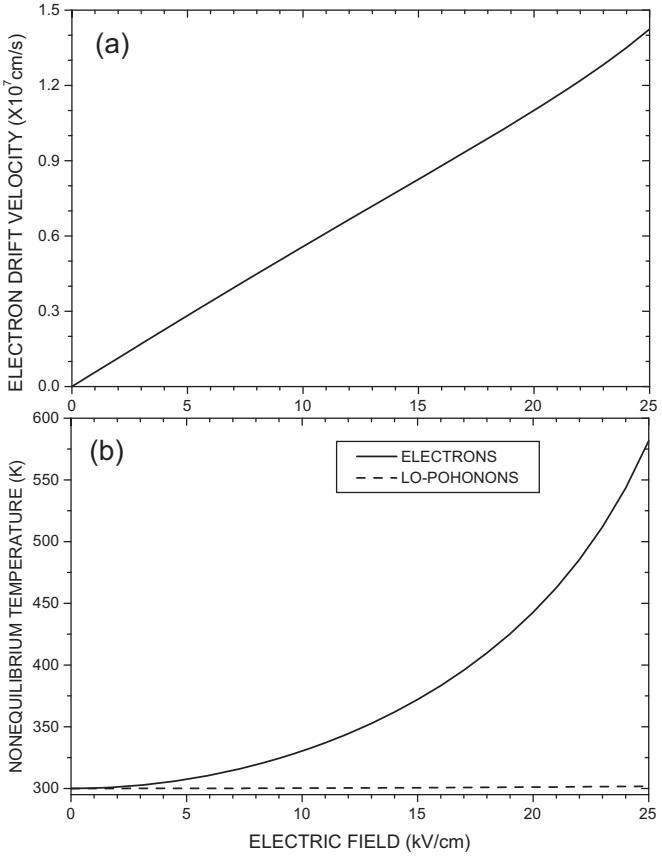

Fig. 1. The steady-state values of (a) the electron drift velocity, (b) nonequilibrium temperatures of electrons (solid) and LO-phonons (dashed) for ZnSe.

The dependence on $\mathcal{E}$ of the steady-state electron drift velocity, electron and LO-phonon nonequilibrium temperatures in ZnSe is depicted in Fig. 1. The AC-phonons are only slightly warmed up because of the rapid heat diffusion to the reservoir at room temperature; the LO-phonons are the warmed up less than 1% above room temperature. We noticed that the AC and LO phonons were considered to be internally thermalized, disregarding possible differentiated distribution of populations in reciprocal space as a result of what can be termed as Fröhlich-Cherenkov effect [24,25].

3. Luminescence spectrum in zinc selenide

Once it has been solved the question of characterization of the nonequilibrium thermodynamic state of the system, we proceed to calculate the luminescence spectrum $I(\omega)$ in the presence of the electric