Temperature-dependent exciton luminescence in quantum wells by computer simulation

S. D. Baranovskii
Institut für Physikalische Chemie und Wissenschaftliches Zentrum für Materialwissenschaften, Philipps-Universität Marburg, Hans-Meerwein-Straße, D-35032 Marburg, Germany

R. Eichmann and P. Thomas
Fachbereich Physik und Wissenschaftliches Zentrum für Materialwissenschaften, Philipps-Universität Marburg, Renthof 5, D-35032 Marburg, Germany

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Optical spectra in quantum wells are in many cases strongly influenced by disorder. In particular, energy relaxation of correlated electron-hole pairs through disorder-induced localized states determines the position and shape of photoluminescence lines. By a Monte Carlo simulation approach the energy relaxation is studied and the temperature dependence of the spectral peak and the linewidth are determined for a variety of model systems in the steady-state situation. [S0163-1829(98)05643-4]

I. INTRODUCTION

Semiconductor quantum-well (QW) structures are intensively studied in the last decade because of their potential for optoelectronic devices. In particular, QW's on the basis of semiconductor alloys are of high interest, since they allow us, using band-structure engineering, to fabricate structures with optimized optical properties for special devices. Therefore, the study of optical properties of alloy QW's is an indispensable task in semiconductor optics. The number of publications devoted to the optical properties of binary and alloy semiconductor QW's, both experimental and theoretical, is enormous. Most of the theoretical work is based on idealized systems where the influence of disorder is disregarded. However, all semiconductor heterostructures possess a certain degree of disorder due to their alloy structure and/or imperfect interfaces. In particular, in narrow QW's the interface roughness creates an essential disorder potential giving rise to band tails composed from localized states. These tails in general affect the dynamics of the Coulomb-correlated electrons and holes. While quite a number of experimental papers on the dynamics of optical excitations in a disordered environment exist, the theoretical literature on this subject is still comparatively scarce.

In this contribution we attempt to reduce this gap between experiment and theory and aim at a generally applicable description of relaxation processes in disordered QW structures on the basis of a phenomenological model. In many situations, depending on various microscopic parameters, the Coulomb-interaction can be treated by considering excitons moving in an effective disorder potential.[1] According to this approach the movement of a correlated electron-hole pair can be described by a disorder potential that acts solely on the center-of-mass coordinate of the pair. This approach is valid as long as the amplitude of the disorder potential does not exceed the exciton binding energy, otherwise the internal degrees of freedom of the electron-hole pair become severely affected and the notion of an exciton in the normal sense is questionable. Solving the corresponding Schrödinger equation for the center-of-mass motion in the effective disorder potential one obtains the energy spectrum of localized excitons.[1] This model has been successfully used to study the energy distribution of excitons in various two-dimensional (2D) systems.[2–4] Since localized excitons can still move in space and energy due to their coupling to the phonon system (hopping), the luminescence spectrum does not coincide with the absorption spectrum, i.e., with the energy distribution of localized states (DOS), but shows a more or less developed Stokes shift relative to the excitation spectrum. Such incoherent hopping processes build the central topic of our present discussion. The model is based on phenomenological microscopic parameters, since more detailed knowledge from microscopic theories is not available at present for the disordered systems studied here. Nevertheless, the predicted effects for the steady-state luminescence are robust against the choice of parameters.

Detailed experimental study of the photoluminescence (PL) Stokes shift at various temperatures has been first carried out by Skolnick *et al.*[5] for $In_xGa_{1-x}As/InP$ single quantum wells and by Davey *et al.*[6] for $In_xGa_{1-x}As/Al_xIn_{1-x}As$ multiple quantum wells. There were also many later studies of the temperature-dependent exciton PL in QW's (see, e.g., Daly *et al.*[7]), whose results confirm the most striking effects observed in Refs. 5 and 6. There are two of these effects that cause the most interest. One is the nonmonotonous temperature dependence of the Stokes shift even if a correction is made for the temperature-induced shift of the band gap. At low temperatures the Stokes shift increases with rising temperature T (i.e., a redshift of the PL spectrum) and at higher temperatures the shift decreases again with rising T (i.e., a blueshift of the PL spectrum). This effect was especially pronounced in Ref. 6. The other intriguing effect is the abrupt increase of the PL linewidth in a rather narrow temperature range.[5] A qualitative explanation of both effects has been already given by Skolnick *et al.*[5] If the emission were purely intrinsic then the PL peak energy would just follow the band-edge variation to lower energy with increasing T. Experimental results do not show this simple trend, and, hence, extrinsic processes, i.e., the motion of excitons via localized states, induced in QW's by disorder, should play a crucial

role for the exciton PL. With rising temperature, the excitons become more mobile and they are able to move over longer distances than at $T=0$. In such a movement the excitons can be trapped by centers with lower energy and, hence, the lower-energy states become increasingly populated with rising temperature. This provides a plausible explanation for the observed ''antithermalization'' behavior of the PL peak energy. With the further increase of temperature the motion of excitons over localized states becomes even faster and the thermal distribution of excitons can be achieved. Concerning the linewidth, its abrupt increase with T was also ascribed to the increase in the mobility of excitons. More mobile excitons can recombine from a broader energy distribution of the localized states than in the situation of lower temperature, where only lowest-lying localized states are occupied and a narrow PL line is observed.[5]

To verify these ideas, a more rigorous theory is necessary. The first demand to such a theory should be the ability to appropriately describe the motion of excitons over localized states. There is no lack in the attempts to develop such a theory; however, most of them fail to take into account explicitly the dependence of transition rates on the distances between the localized states involved. Because of the spatial localization of excitons, these dependencies are very strong. In the case of tunneling transitions, the dependence of the hopping rate on the length of the hop is exponential and it should be definitely taken into account. Unfortunately, most of the averaging procedures wash out this dependence. Moreover, the exponential dependence of the hopping rate on the hopping distance makes even the rates for hopping transitions downward in energy strongly dependent on the energy of localized states because the concentrations of available localized states and, hence, the distances to them are different at different energies. This fact is also not taken into account usually.

A simple model of exciton energy relaxation via hopping through localized states has been suggested by Abdukadyrov *et al.*[8] for bulk II-VI ternary alloys with common cation components. It is known for these materials that the compositional disorder due to a random substitution in the anionic sublattice leads to a pronounced valence-band tail, drastically modifying optical properties of excitons compared to those in ideal crystals.[9] In such systems, excitons are localized due to the localization of the holes and their transitions between localized states correspond to the hopping of the holes in the valence-band tail. In such a case it is reasonable to assume that transition rates of excitons have the same exponential dependence on the distances between localized states as the transition rates of single holes. To perform calculations in the framework of such a model, one first assumes some given DOS (usually a purely exponential or a Gaussian) and treats hopping of excitons as tunneling single-particle transitions between localized states. This model has been successfully applied to describe hopping of excitons in bulk II-VI alloys.[8,10,11]

It has also been applied to excitons in QW's (Refs. 12 and 13). However, application of such a model to QW's and also to bulk systems with large energy disorder acting on both electrons and holes is questionable. In these systems it is not possible, in general, to describe the influence of disorder on excitons by its influence just on holes. Moreover, even the dependencies of the transition rates on the distances between the localized states involved are not clear. They are either exponential if excitons hop via simultaneous tunnelling of electrons and holes or they are described by a power law if the excitons hop between localized states via exchange of virtual photons (dipole-dipole coupling[14]). Nevertheless, recent success of the model with exponential transition rates in the fitting of experimental data for $Zn_{0.8}Cd_{0.2}$/ZnSe QW's is striking.[12] Unfortunately, the analytical theory in Ref. 12 is valid only at extremely low temperatures because it can take into account only transitions downward in energy. There seems to be no way to extend this analytical theory for finite temperatures. An alternative approach to the theoretical description of the exciton hopping has been recently suggested by Zimmermann and co-workers.[15,16] They first performed an explicit calculation of the exciton DOS taking into account actual growth conditions of QW's and realistic wave functions. The exciton kinetics has been simulated as acoustic phonon scattering between disorder eigenstates, thus including bandlike as well as hoppinglike transitions. Nonmonotonous temperature dependencies of the Stokes shift and of the PL linewidth have been obtained. The interpretation given is very similar to that of Skolnick *et al.*[5] For the nonmonotonous dependence of the Stokes shift it has been suggested[15,16] that at finite but low temperatures excitons can overcome shallow barriers between localized states during their lifetime by thermal activation and fall down in still deeper states than at $T=0$.

In the present paper we study the temperature-dependent spectra of the steady-state exciton luminescence in the framework of a simple model with exciton hopping via uncorrelated localized states distributed in space and energy. As already mentioned above, it is not possible to extend the analytical quantitative theory developed for extremely low temperatures[8,12] to finite temperatures. Hence, we use a straightforward Monte Carlo computer simulation of the exciton hopping energy relaxation. In Sec. II, the numerical algorithm is described. In Sec. III the simulation results for exponential DOS are presented and compared with analytical theories. At zero temperature they are compared with the analytical theory of Ref. 12 and at finite temperatures the simulation results are compared with the qualitative analytical description on the basis of the transport-energy concept.[17] In Sec. IV, the simulation results are given for a more realistic Gaussian DOS and compared with experimental data available. Concluding remarks are gathered in Sec. V.

II. NUMERICAL ALGORITHM AND SIMULATION DETAILS

The simulation technique we use to study the hopping energy relaxation and luminescence of excitons is similar to that suggested by Silver *et al.*[18] and will, therefore, only briefly be described here. We assume that excitons behave like single particles in their hopping movement between localized states. However, contrary to single electrons and holes recombining via tunneling processes, excitons have some typical lifetime τ_0 with respect to their radiative recombination.

The simulation algorithm was the following. A 2D rectangle of the linear size $N_0^{1/2}$ containing N_0 sites is considered

and the periodic boundary conditions are employed. The sites within the rectangle are distributed in space at random. The energies of the sites were chosen in such a way that either a pure exponential or a Gaussian DOS was obtained. The energetic parameter of the DOS acts as a scaling parameter for all energies in the simulation. No correlation between energies of sites and their spatial positions were allowed. These sites simulate the set of localized states that an exciton can use for its hopping motion. The rate of a hopping transition from an occupied site i to an empty site j over a distance r_{ij} is determined by the Miller-Abrahams expression

$$\nu_{ij}=\nu_0\exp\left(-\frac{2r_{ij}}{\alpha}-\frac{\epsilon_j-\epsilon_i+|\epsilon_j-\epsilon_i|}{2kT}\right), \tag{1}$$

where ϵ_i and ϵ_j are the energies of the states i and j, respectively, α is the decay length of the exciton center-of-mass wave function in the localized states, and ν_0 is the attempt-to-escape frequency.

Hopping and recombination of a large number of excitons n has been simulated independently. The fate of each of them has been studied in the following way. First an exciton is situated at a random site i within the array of sites described above. Then the decay rate ν_i is calculated as

$$\nu_i=\tau_0^{-1}+\sum_j \nu_{ij}. \tag{2}$$

Because the hopping rates ν_{ij} depend exponentially on the distances and energy differences between localized states, only few of them determine the sum in the right-hand side of Eq. (2). Therefore, we restricted the number of ν_{ij} terms in this sum by M largest terms. Usually M was taken as 32 and it has been checked that the increase of M up to 64 does not change the simulation results.

Using a random number generator, the real time t_i of the next process, which is to occur with the chosen exciton, has been calculated as

$$t_i^{-1}=-\nu_i\ln\xi_1, \tag{3}$$

where ξ_1 is a random number from the uniform random-number distribution between 0 and 1. Then using the rates τ_0^{-1} and ν_{ij} and another random number ξ_2 the specific process is determined. If it is the hopping transition to some localized state j, the exciton is transferred to the site j, the term t_i is added to the whole time counter, and the algorithm is repeated. If it is the exciton recombination, the energy position of the exciton is stored along with the sum of times t_i, which the exciton has spent for the successive hops, and also for the last step. Then a new exciton is considered, and so on. The results of the simulation are the spectrum of the recombination energies and the distribution of the recombination times for all n excitons. Below we discuss only the stationary PL spectra, while the time-resolved results are reserved for a separate publication. It can be easily shown that the parameters of the system under consideration, i.e., τ_0, ν_0, α, and the concentration of the localized states N can be combined into a set of only two essential parameters $N\alpha^2$ and $\tau_0\nu_0$. The energies are always scaled by the typical energy scale of the DOS function (exponential or Gaussian).

The number of sites N_0 was chosen to be equal to 2500 and the number of exciton fates simulated for each set of parameters varied from 10^4 to 10^5, in order to get the reliable results after the averaging.

III. EXCITON LUMINESCENCE IN A SYSTEM WITH EXPONENTIAL DOS

In this section we consider a system with purely exponential density of localized states of the form

$$g(\epsilon)=\frac{N}{\epsilon_0}\exp\left(\frac{\epsilon}{\epsilon_0}\right), \tag{4}$$

where N is the concentration of localized states and ϵ_0 is the energy scale of the DOS energetic distribution. The localization energies ϵ are counted negative from some reference energy $\epsilon=0$.

Such a DOS being extremely simple allows us to use an analytical treatment of the hopping relaxation that is well developed quantitatively for the case of zero temperature[8,12] and has a very reasonable qualitative description for the case of finite temperatures.[17] This provides a nice opportunity to verify the numerical algorithm described above and the results of the analytical treatment. We start with the case of zero temperature.

At $T=0$ the hopping relaxation of particles via localized states can be identified with the energy-loss hopping, because thermally activated transitions are not possible at $T=0$. In such a case, the physical problem rather becomes a geometrical one, which allows us to perform the calculations to get the quantitative description of the exciton hopping relaxation.[8,12] In Ref. 8 such calculations have been performed for the bulk system. Recently, Golub *et al.*[12] have modified this approach for a 2D system and compared its results with the experimental PL data obtained on a 20 Å $Zn_{0.8}Cd_{0.2}Se/ZnSe$ quantum well. In Fig. 1 the measured steady-state PL spectrum from Ref. 12 is shown by the solid line. The dotted line in this figure shows the result of the analytical theory from Ref. 12 with parameters $\tau_0=170$ ps, $\nu_0=10^{13}$ s^{-1}, $N\alpha^2=0.0675$, and $\epsilon_0=8$ meV chosen for the best fit of the experimental data. Filled circles in Fig. 1 represent the results of our computer simulation for the same set of parameters. Agreement between the simulation and the analytical theory is really excellent. This agreement evidences that in the framework of the chosen model for the exciton relaxation the steady-state PL spectrum at very low temperatures is correctly described by both the analytical theory and the computer simulation. It is valuable to have such a check of the simulation results by the analytical calculations at least in this particular case of $T=0$. At finite temperatures there are no such explicit analytical calculations available and the straightforward computer simulation seems to be the only method to get a reliable information on the exciton PL spectra.

Let us now consider the case of finite temperatures. Rather curious nonmonotonous temperature dependence of the steady-state PL maximum and the PL linewidth has been observed experimentally.[5–7] We would first like to check whether such nonmonotonous temperature dependencies can arise in a system with the DOS described by Eq. (4) for

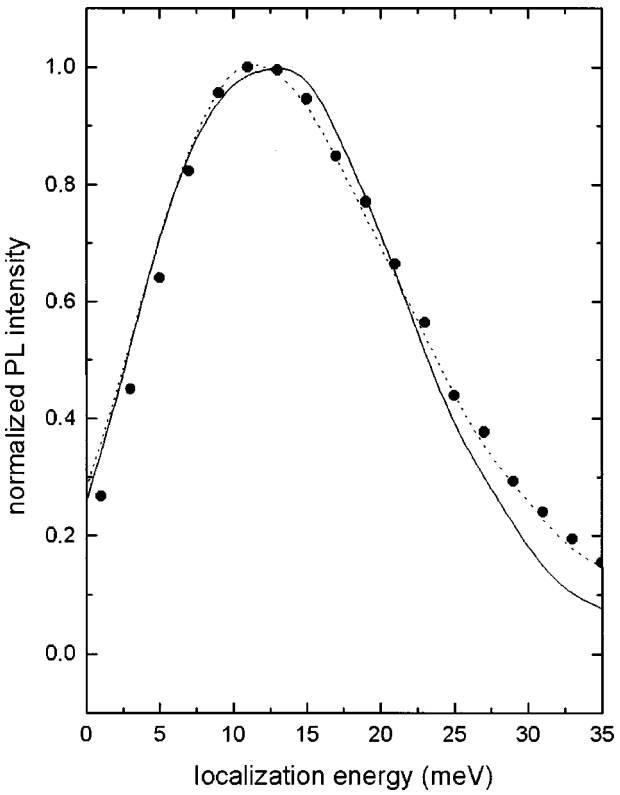

FIG. 1. PL spectrum under stationary excitation. Solid line, experimental results from Ref. 12; dotted line, analytical theory from Ref. 12; filled circles, simulation results.

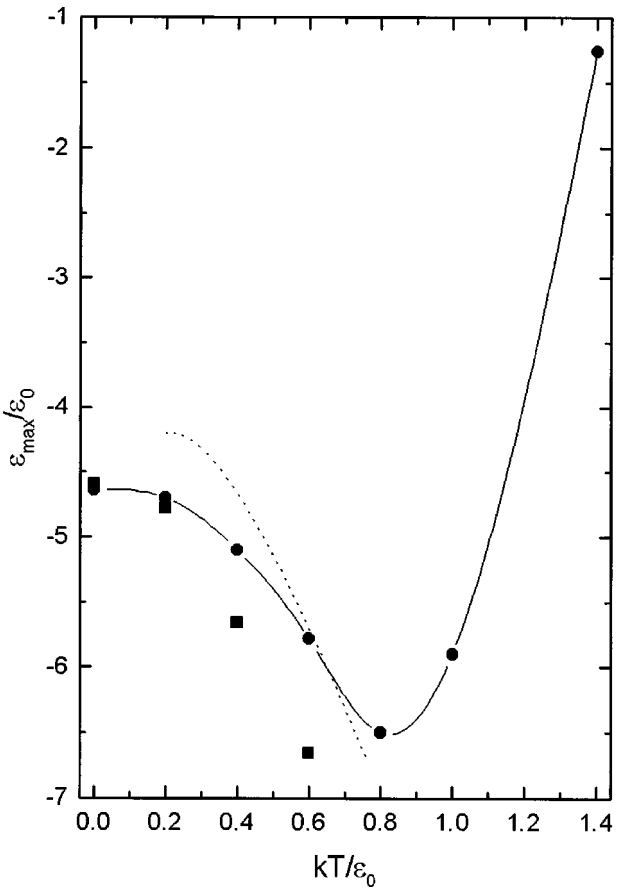

FIG. 2. Temperature dependence of the PL peak Stokes shift for the exponential DOS under stationary conditions for two sets of parameters: (filled circles) $N\alpha^2=1$, $\tau_0\nu_0=10^4$; (filled squares) $N\alpha^2=0.5$, $\tau_0\nu_0=10^{5.6}$. The dotted line corresponds to $\epsilon_d(\tau_0)/\epsilon_0$ for $N\alpha^2=1$ and $\tau_0\nu_0=10^4$.

which the semiquantitative theory has been developed to describe the hopping relaxation of particles at finite temperatures.[17] In Fig. 2 the result of our simulation for the steady-state PL peak is shown by filled circles connected for convenience by a solid line. The quantity ϵ_{max} shown in this figure is in fact the localization energy of excitons (in the units of ϵ_0) corresponding to the peak of the PL spectrum. The larger is $|\epsilon_{max}|$ the lower is the PL emission-peak energy. The data demonstrate that indeed at low temperatures the PL peak first shifts deeper in energy with respect to its position at $T=0$. Then with increasing temperature ϵ_{max} achieves a minimum and further at higher temperatures the energy of the PL peak increases with temperature. The challenging question is whether such a behavior can also be described analytically at least in a semiquantitative manner.

First, let us perform a simple estimate of ϵ_{max} at $T=0$. In the course of the energy-loss hopping, the excitons slow down their motion in the successive transitions because the number of available localized states, i.e., those with lower energies, decreases and the hopping distance to them and, correspondingly, the time of hops increases. When the time of the next hop becomes comparable to τ_0, the recombination comes in play, i.e., it becomes effective. From shallower states it is more favorable to perform an energy-loss hop, whereas for much deeper states, hops are so slow that recombination prevails. Therefore, one can estimate roughly the position of ϵ_{max} as the energy at which the hopping rate is equal to the inverse lifetime τ_0. If a particle is situated at some energy ϵ in the DOS distribution, the concentration of available states is

$$N(\epsilon)=\int_{-\infty}^{\epsilon} g(\epsilon)d\epsilon, \tag{5}$$

and the typical distance to the next available state is

$$R(\epsilon)\simeq[\pi N(\epsilon)]^{-1/2}. \tag{6}$$

The corresponding hopping rate is

$$\nu[R(\epsilon)]=\nu_0\exp\left\{-\frac{2R(\epsilon)}{\alpha}\right\}. \tag{7}$$

Using Eq. (4), we obtain from the equation

$$\nu[R(\epsilon_{max})]\simeq\tau_0^{-1} \tag{8}$$

the result

$$\epsilon_{max}\simeq-\epsilon_0\ln[(\pi N\alpha^2/4)\ln^2(\tau_0\nu_0)]. \tag{9}$$

In Ref. 12 it was shown that a more careful calculation leads to the expression

$$\epsilon_{max}\simeq-\epsilon_0\{\ln[(\pi N\alpha^2/4)\ln^2(\tau_0\nu_0)]+X\}. \tag{10}$$

For the constant X the value 0.567 has been suggested.[12] Our simulation shows that the value $X\simeq0.45$ is more suitable to fit the position of ϵ_{max} at $T=0$. It is very tempting to search for some universal combination of parameters that could

scale ϵ_{max}. Equations (9) and (10) show that at $T=0$ such a scaling parameter is $\zeta=(N\alpha^2)\ln^2(\tau_0\nu_0)$. Indeed, the computer simulation with different values of $N\alpha^2$ and $\tau_0\nu_0$ that keep ζ constant, lead to the same value of $\epsilon_{max}/\epsilon_0$ at $T=0$ as can be well seen in Fig. 2. It is also seen in this figure that at higher temperatures this scaling is violated. We are not aware of any scaling parameter at finite T.

The hopping relaxation of particles at finite temperatures via localized states with the DOS described in Eq. (4) has been considered for the bulk system by Monroe.[17] According to this consideration, a particle, starting from the mobility edge, most likely makes a series of hops downward in energy. This character of the relaxation process changes at some particular energy ϵ_t called the transport energy (TE). The hopping process near and below TE resembles the multiple trapping process, where ϵ_t plays the role of the mobility edge. Modifying the arguments of Ref. 17 for a 2D system one obtains

$$\epsilon_t/\epsilon_0=-2\ln[(\pi N\alpha^2)^{1/2}\epsilon_0/kT]. \tag{11}$$

Using the description of Orenstein and Kastner[19] for the relaxation of particles via the multiple trapping process with replacing the mobility edge by ϵ_t and modifying the approach for a 2D system, we obtain that at time t after the beginning of the relaxation, the peak in the energy distribution of the relaxing particles is situated near the so-called demarcation energy $\epsilon_d(t)$, determined as

$$\epsilon_d(t)/\epsilon_0=\epsilon_t/\epsilon_0-\frac{kT}{\epsilon_0}\ln(\nu_0 t)+2. \tag{12}$$

The assumptions used to derive Eq. (12) in the sense of Ref. 19 are the following: (i) $\epsilon_t/\epsilon_0-\epsilon_d(t)/\epsilon_0\gg 1$; (ii) $kT\ll\epsilon_0$.

It is reasonable to identify ϵ_{max} at finite temperatures with the quantity $\epsilon_d(\tau_0)$, provided conditions (i) and (ii) are fulfilled. For the simulation parameters used for the data in Fig. 2 one finds that condition (i) becomes fulfilled at $kT>0.5\epsilon_0$. Unfortunately, one cannot consider condition (ii) as fulfilled in such a case. Therefore, strictly speaking, Eq. (12) should not be applied to the results in Fig. 2. Nevertheless, using Eq. (12) formally, one obtains values for $\epsilon_d(\tau_0)/\epsilon_0$, which are rather close to the simulation results at $kT/\epsilon_0<0.8$. This shows that the quantity $\epsilon_d(\tau_0)$ may be a reasonable estimate for ϵ_{max} at finite temperatures in the restricted temperature range even if Eq. (12) is not rigorously applicable.

At very high temperatures $kT\gg\epsilon_0$ the excitons should reach thermal equilibrium rather fast, i.e., at times shorter than τ_0. The maximum of the energy distribution shifts in such a case to the edge of the DOS, and it becomes more convenient to discuss the average energy of particles instead of the maximum in the PL spectrum. It can be easily shown that the average energy of particles in thermal equilibrium in a system with the DOS described by Eq. (4) is equal to ϵ_0 at $kT\gg\epsilon_0$. We have checked that the average energy in the simulation is indeed equal to ϵ_0 at $kT\gg\epsilon_0$. Herewith, we have a reasonable semianalytical description of the system of relaxing excitons at very low temperatures, at intermediate temperatures ($kT\simeq 0.5\epsilon_0$), and at rather high temperatures $kT\gg\epsilon_0$. Simulation results agree with this description, although only a computer simulation provides the reliable results in the whole temperature range.

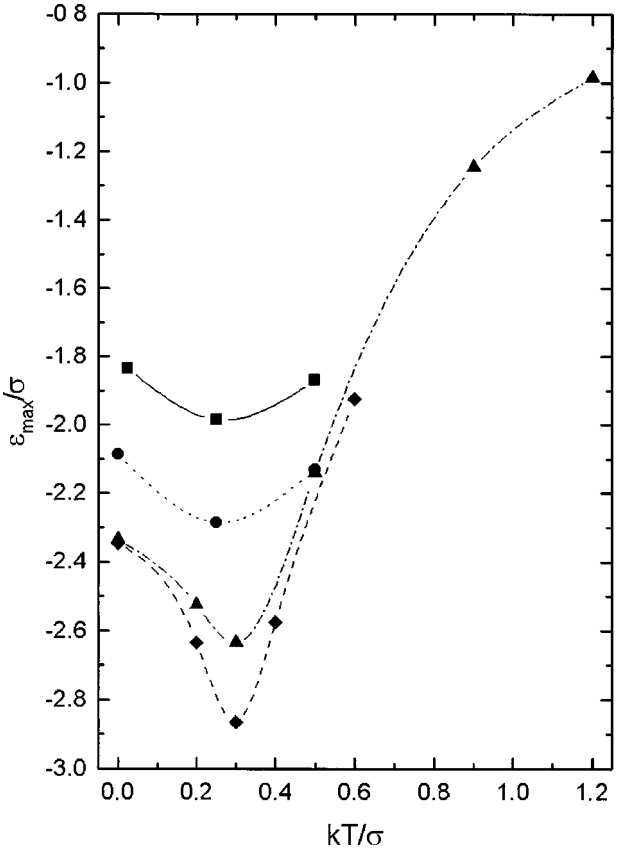

FIG. 3. Temperature dependence of the PL peak Stokes shift for a Gaussian DOS under the stationary conditions. The parameters are (filled triangles) $N\alpha^2=1$, $\tau_0\nu_0=10^4$; (filled circles) $N\alpha^2=1$, $\tau_0\nu_0=10^3$; (filled squares) $N\alpha^2=0.5$, $\tau_0\nu_0=10^3$; (filled diamonds) $N\alpha^2=0.5$, $\tau_0\nu_0=10^{5.6}$.

IV. EXCITON LUMINESCENCE IN A SYSTEM WITH GAUSSIAN DOS

Exponential DOS described by Eq. (4) is rather convenient for analytical treatments at least in the semiquantitative approach presented above. Therefore, the exponential DOS has been mostly used so far for analytical studies of exciton relaxation in quantum wells with disorder.[12] However, for such systems, a Gaussian DOS for optically active excitons seems more relevant[20]:

$$g(\epsilon)=\left(\frac{N^2}{2\pi\sigma^2}\right)^{1/2}\exp\left(-\frac{\epsilon^2}{2\sigma^2}\right), \tag{13}$$

where N is the concentration of localized states and σ is the energy scale of the distribution. The simulation results obtained with such DOS for $\epsilon_{max}(T)$ are shown in Fig. 3. In Fig. 4 the width of the steady-state PL spectrum $\Delta\epsilon$ [full width at half maximum (FWHM)] is shown as a function of temperature. In the case of a Gaussian DOS the analytical calculation is only possible for the limit of high temperatures $kT\gg\sigma$. It can be easily shown that in this limit $\epsilon_{max}\simeq-\sigma^2/kT$ and $\Delta\epsilon\simeq 2.4\sigma$.[21] These values agree well with the obtained simulation results. At intermediate and low tem-

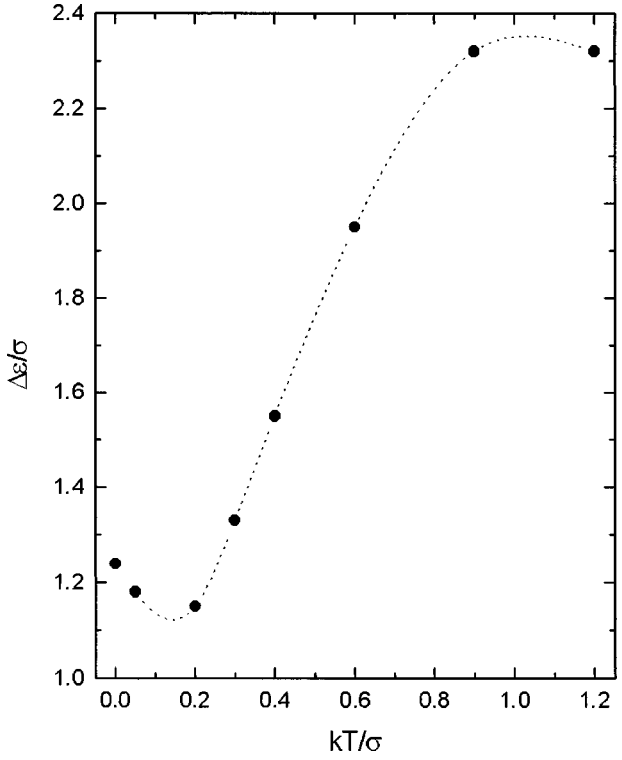

FIG. 4. Temperature dependence of the PL linewidth (FWHM) for a Gaussian DOS under the stationary conditions. The parameters are $N\alpha^2=1$, $\tau_0\nu_0=10^3$.

peratures, compared to σ, the only way to get reliable results is the computer simulation. It is well seen in Fig. 3 that the shape of the dependence $\epsilon_{max}(T)$ strongly depends on the material parameters $N\alpha^2$ and $\tau_0\nu_0$. The larger $N\alpha^2$, the easier the energy-loss hopping at low temperatures and, hence, particles can come to deeper energies during their lifetime $\simeq\tau_0$. Due to the same reason, the particles go deeper in energy for larger values of τ_0 if $N\alpha^2$ is unchanged. As mentioned above, there is a scaling parameter $\zeta=(N\alpha^2)\ln^2(\tau_0\nu_0)$ for the energy-loss hopping at $T=0$. In Fig. 3 it is seen that the value $\epsilon_{max}(0)$ does not change with changing $\tau_0\nu_0$ from 10^4 to $10^{5.6}$ with simultaneous changing of $N\alpha^2$ from 1 to 0.5 and keeping the value of ζ constant. At finite temperatures there is no universal scaling parameter of this kind. The simulation results in Fig. 3 confirm perfectly the qualitative ideas of Skolnick *et al.*[5] It is worth noting that in order to compare the simulated temperature dependencies of the PL peak energy with experimental results, one should take into account also the temperature-induced shift of the band gap that was out of our scope in the simulation. To make the corresponding correction, one assumes that the reference energy in the simulation is temperature dependent in accordance with the temperature dependence of the band gap.

The temperature dependence of the PL linewidth in Fig. 4 is rather curious. The main feature of this dependence is a relatively sharp transition from the low-temperature value of $\Delta\epsilon$ to the high-temperature value within the rather narrow temperature range. Such a dependence has been observed experimentally in Ref. 5 and it was also reported for the exciton PL from coupled quantum wells (CQW's) subjected to the influence of an external electric field.[22] The application of an electric field to CQW's leads to a spatial separation of electrons and holes in opposite wells. Correlated electron-hole pairs formed in CQW's have long lifetimes due to a small overlap between electron and hole wave functions and, hence, they are of particular interest to study the hopping relaxation processes of excitons via localized states. Studying the PL of excitons from 50-40-50 Å coupled $GaAs/Al_xGa_{1-x}As$ QW with an applied electric field of strength 28 kV/cm, Fukuzawa *et al.*[22] observed a sharp change in the PL linewidth from 3.4 meV (FWHM) at 5 K to 5.6 meV at 8.5 K for the e_1h_1 transition. This sharp temperature dependence of the PL linewidth was first attributed to a transition of the exciton system into some ordered state[22] and later to the equilibrium Fermi-Dirac distribution of excitons over localized states.[23] It is, however, clearly shown in Ref. 23 that there is no equilibrium distribution of excitons at 6 K, i.e., in the middle of the temperature range where the transition takes place. There is also no evidence for any ordered state of excitons under experimental conditions used in Ref. 22. Therefore, both the transition into some ordered state and the Fermi-Dirac distribution of excitons seem unlikely to account for the effect. As a possible alternative explanation, it has been suggested that the observed sharp temperature dependence of the linewidth can be caused by the temperature dependence of the DOS function itself provided the energy distribution of DOS is dominated by long-range Coulomb potentials of charged impurities.[24] It is, however, not clear whether or not such impurities were present in CQW's studied in Ref. 22. The sharp temperature dependence of $\Delta\epsilon$ in Fig. 4 shows that the puzzling feature is inherent for exciton hopping relaxation via localized states even in the simplest approach in which excitons are treated just as single particles. The same sharp dependence can be also shown for other parameters than those used for the curve in Fig. 4,[25] which just strengthens this conclusion. It is worth emphasizing that the sharp temperature dependence of $\Delta\epsilon$ should be observed for the exciton system being out of thermal equilibrium. In the equilibrium at high T, the linewidth does not depend essentially on temperature.

V. CONCLUSIONS

Computer simulation of the hopping energy relaxation of excitons via uncorrelated localized states has been carried out. Excitons were treated as single particles and their tunneling transitions with exponential dependence of the hopping rate on the hopping distance have been studied. Simulation results for the steady-state luminescence peak energy and for the linewidth show that experimental results on temperature dependencies of these quantities can be well understood in the framework of the considered model. In particular, the nonmonotonous temperature dependence of the peak energy and the abrupt increase of the linewidth in a narrow temperature range are inherent for excitons performing hopping energy relaxation. At very low temperatures the simulation results agree perfectly with those of the analytical theory.[12] At finite temperatures, a computer simulation seems to be the only reliable technique to obtain characteristics of the PL spectra of excitons in quantum wells.

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[1] S. D. Baranovskii and A. L. Efros, Fiz. Tekh. Poluprovodn. **12**, 2233 (1978) [Sov. Phys. Semicond. **12**, 1328 (1978)].
[2] R. Zimmermann, Phys. Status Solidi B **173**, 129 (1992).
[3] S. D. Baranovskii, U. Doerr, P. Thomas, A. Naumov, and W. Gebhardt, Phys. Rev. B **48**, 17 149 (1993).
[4] R. Zimmermann, Nuovo Cimento D **17**, 1801 (1995).
[5] M. S. Skolnick, P. R. Tapster, S. J. Bass, A. D. Pitt, N. Apsley, and S. P. Aldred, Semicond. Sci. Technol. **1**, 29 (1986).
[6] S. T. Davey, E. G. Scott, B. Wakefield, and G. J. Davies, Semicond. Sci. Technol. **3**, 365 (1988).
[7] E. M. Daly, T. J. Glynn, J. D. Lambkin, L. Considine, and S. Walsh, Phys. Rev. B **52**, 4696 (1995).
[8] A. G. Abdukadyrov, S. D. Baranovskii, S. Yu. Verbin, E. L. Ivchenko, A. Yu. Naumov, and A. N. Reznitskii, Zh. Eksp. Teor. Fiz. **98**, 2056 (1990) [Sov. Phys. JETP **71**, 1155 (1990)].
[9] S. Permogorov and A. Reznitsky, J. Lumin. **52**, 201 (1992).
[10] U. Siegner, D. Weber, E. O. Göbel, D. Bennhardt, V. Heuckeroth, R. Saleh, S. D. Baranovskii, P. Thomas, H. Schwab, C. Klingshirn, J. M. Hvam, and V. G. Lyssenko, Phys. Rev. B **46**, 4564 (1992).
[11] A. Reznitsky, S. D. Baranovskii, A. Tsekun, and C. Klingshirn, Phys. Status Solidi B **184**, 159 (1994).
[12] L. E. Golub, S. V. Ivanov, E. L. Ivchenko, T. V. Shubina, A. A. Toropov, J. P. Bergman, G. R. Pozina, B. Monemar, and M. Willander, Phys. Status Solidi B **205**, 203 (1998).
[13] H. Kalt, J. Collet, S. D. Baranovskii, R. Saleh, P. Thomas, Le Si Dang, and J. Cibert, Phys. Rev. B **45**, 4253 (1992).
[14] T. Takagahara, J. Lumin. **44**, 347 (1989).
[15] R. Zimmermann, E. Runge, and F. Grosse, *Proceedings of the 23rd International Conference on the Physics of Semiconductors, Berlin, 1996*, edited by M. Scheffler and R. Zimmermann (World Scientific, Singapore, 1996), Vol. 3, p. 1935.
[16] R. Zimmermann and E. Runge, Phys. Status Solidi A **164**, 511 (1997).
[17] D. Monroe, Phys. Rev. Lett. **54**, 146 (1985).
[18] M. Silver, G. Schönherr, and H. Bässler, Phys. Rev. Lett. **48**, 352 (1982).
[19] J. Orenstein and M. A. Kastner, Solid State Commun. **40**, 85 (1981).
[20] R. F. Schnabel, R. Zimmermann, D. Bimberg, H. Nickel, R. Lösch, and W. Schlapp, Phys. Rev. B **46**, 9873 (1992).
[21] R. Zimmermann, F. Grosse, and E. Runge, Pure Appl. Chem. **69**, 1179 (1997).
[22] T. Fukuzawa, E. E. Mendez, and J. M. Hong, Phys. Rev. Lett. **64**, 3066 (1990).
[23] J. A. Kash, M. Zachau, E. E. Mendez, J. M. Hong, and T. Fukuzawa, Phys. Rev. Lett. **66**, 2247 (1991).
[24] S. D. Baranovskii and P. Thomas, Phys. Rev. Lett. **69**, 993 (1992).
[25] S. D. Baranovskii, R. Eichmann, and P. Thomas, Phys. Status Solidi B **205**, R19 (1998).