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Hopping relaxation of excitons in GaInNAs/GaNAs quantum wells

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Exciton photoluminescence (PL) in a GaInNAs/GaNAs quantum well was measured in the temperature range from 15 K to 300 K. Two striking features of the PL were observed: the nonmonotoneous temperature dependence of the Stokes shift and the abrupt increase of the PL linewidth in a rather narrow temperature range. These features are known to be strong indications of the hopping relaxation of excitons via localized states distributed in space and energy. Computer simulations of the hopping relaxation of excitons were carried out. Comparison between the simulation results and the experimental data provides an important and reliable information on the energy shape of the density of states and also on the energy range, in which localized states for excitons are distributed.

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1 Introduction

Semiconductor quantum-well (QW) structures are intensively studied because of their potential for optoelectronic devices. QW's based on semiconductor alloys are of particular interest, since they allow one to fabricate structures with optimized optical properties. 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 a disorder potential giving rise to band tails composed from localized states. These tails affect the dynamics of the Coulomb-correlated electrons and holes and they influence essentially the optical properties of devices based on QW alloy heterostructures. In order to control the optical properties of devices, one should have a technique for characterization of the band tails caused by disorder. The crucial questions are: what is the energy scale of the band tails in QW's and what is the energy density of the tail states (DOS). We suggest such a characterization technique for quaternary GaInNAs-based QW's. The technique is based on the comparison between the experimental data and the results of the Monte Carlo computer simulation for the temperature dependences of the PL Stokes shift and that of the PL linewidth.

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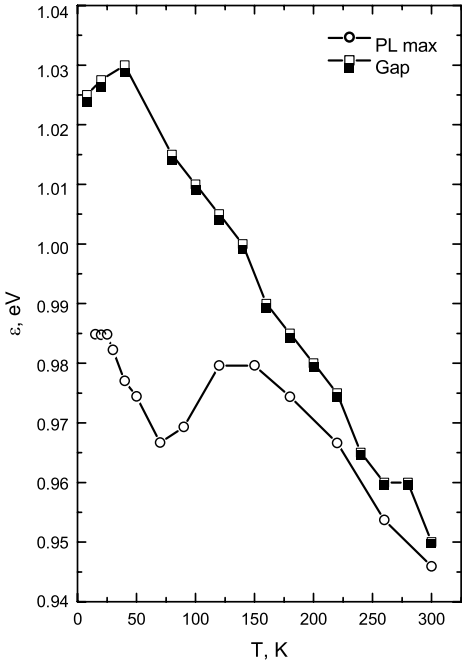

Fig. 1 Experimental results for both the PL peak and the gap as a function of temperature.

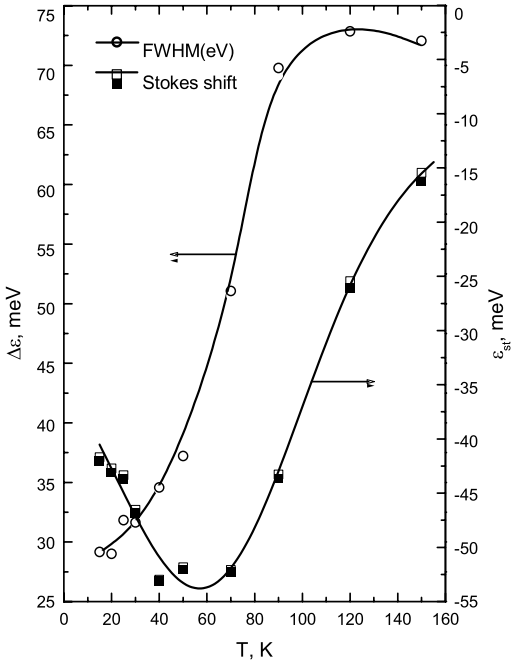

Fig. 2 Experimental results for the PL linewidth (FWHM) and Stokes shift as a function of temperature.

2 Results and discussion

The 6.2 nm wide $Ga_{0.63}In_{0.37}N_{0.01}As_{0.99}$ QW with $GaN_{0.017}As_{0.983}$ barriers was grown by metal-organic vapour-phase epitaxy on a (100) GaAs substrate. The temperature-dependent PL measurements were performed using a HeNe laser and low excitation densities on the sample. The corresponding band gap shift was determined by PL excitation spectroscopy at temperatures below 80 K and by photomodulated reflectance spectroscopy above 80 K using standard set-ups.

In Fig. 1 the results are given for the measured temperature dependence of the PL peak position and for the temperature dependence of the band gap. The anomalous increase of the gap with temperature below 40 K is within the experimental error. While the band gap decreases monotonously with raising temperature, the temperature dependence of the PL peak is essentially nonmonotonous. The temperature dependence of the Stokes shift is shown in Fig. 2 along with the temperature dependence of the PL linewidth [full width at half maximum (FWHM)]. One can see an abrupt increase of the linewidth in a rather narrow temperature range between 15 K and 90 K, while above 90 K the linewidth looks almost temperature-independent.

Both observed striking effects – the non-monotonous temperature dependence of the Stokes shift and the abrupt increase of the PL linewidth with temperature – are known in the literature for GaInNAs QW's [1, 2], as well as other alloy QW's, for example, for InGaAs/InP [3] and for InGaAs/AlInAs [4]. The qualitative explanation of both effects has already been given by Skolnick *et al.* [3]. They were attributed to the motion of excitons via localized states, induced in QW's by disorder. With rising temperature excitons become more mobile and they can be trapped by centers with lower energies and, hence, the lower-energy states become increasingly more populated with rising temperature. This enhances the Stokes shift. With the further increase of temperature the motion of excitons over localized states becomes even faster and their thermal equilibrium distribution can be achieved leading to higher PL energies and to the smaller

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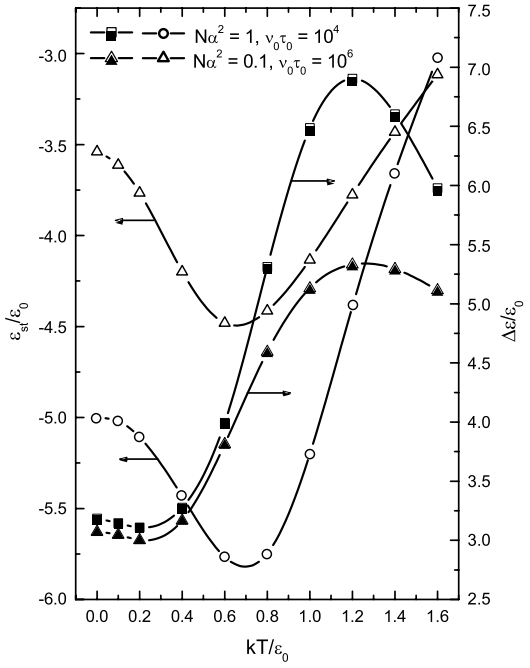

Fig. 3 Temperature dependence of the PL peak Stokes shift and PL linewidth (FWHM) for the exponential DOS (computer simulation).

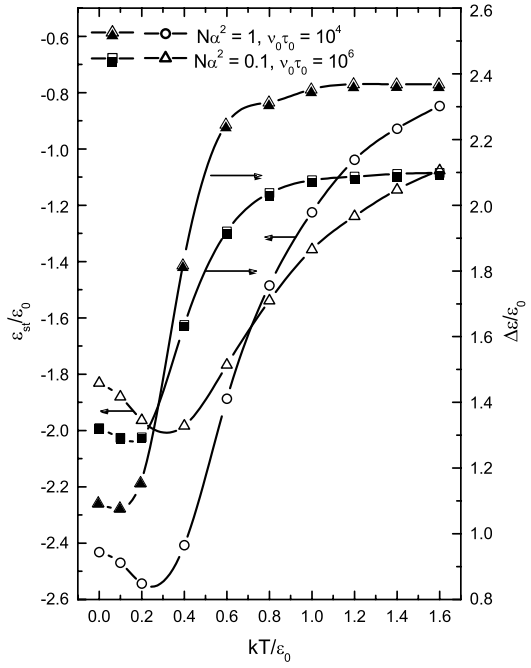

Fig. 4 Temperature dependence of the PL peak Stokes shift and PL linewidth (FWHM) for the Gaussian DOS (computer simulation).

Stokes shift. Concerning the linewidth, its abrupt increase with T was also attributed 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.

In our work we put this argumentation on a quantitative basis by carrying out Monte Carlo computer simulations of the exciton hopping motion via localized states. The simulation algorithm was similar to that described in detail in [5]. In this algorithm a hopping motion of excitons via localized states characterized by some energy distribution is simulated with using Miller-Abrahams transition rates. Besides the energy shape of the DOS, there are three essential parameters in the model: (kT/ϵ_0, $N\alpha^2$, and $\nu_0\tau_0$). Here ϵ_0 is the DOS energy scale, T is the temperature, k is the Bolzmann constant, N is the concentration of localized states, α is the decay length of the exciton wave function in the localized states, ν_0 is the attempt-to-escape frequency, and τ_0 is the typical exciton lifetime.

First we carried out the simulations of the PL with the exponential DOS

$$g(\epsilon_0) = \frac{N}{\epsilon_0} \exp\left(\frac{\epsilon}{\epsilon_0}\right). \tag{1}$$

The simulated results for the PL Stokes shift ϵ_{st} and for the FWHM $\Delta\epsilon$ are given in Fig. 3. The best way to identify the DOS energy scale, ϵ_0, from a comparison with experimental data is to find such a PL characteristic feature which depends on the ϵ_0 and does not depend (or very weakly depends) on other parameters. The best candidate is the PL linewidth $\Delta\epsilon/\epsilon_0$ at low T. As evident from Fig. 3, this quantity at low T is practically independent of $N\alpha^2$ and $\nu_0\tau_0$. In the experiment at low T, the FWHM is $\Delta\epsilon = 30$ meV (see Fig. 2). Computer simulations on the other hand give at low T $\Delta\epsilon \approx 3\epsilon_0$. Straightforward evaluation gives the DOS energy scale $\epsilon_0 \approx 10$ meV.

There is another characteristic feature of the simulated PL data which is also stable against the variation of parameters $N\alpha^3$ and $\nu_0\tau_0$. This is the temperature at which the Stokes shift has its minimum value. In

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the experiment this temperature is $T \approx 60$ K (see Fig. 2). The computer simulation gives $kT \approx 0.6\epsilon_0$. Hence, the DOS scale can be estimated as $\epsilon_0 \approx 9$ meV, which is consistent with the value $\epsilon_0 \approx 10$ meV obtained above from the FWHM data. Furthermore, the temperature kT/ϵ_0 at which the FWHM $\Delta\epsilon/\epsilon_0$ reaches its maximum value is also consistent with such an estimate $\epsilon_0 \approx 9$ meV. The simulation shows that the maximum of the FWHM corresponds to the temperature $kT \approx 1.2\epsilon_0$. In the experiment the FWHM maximum is observed for the temperature 120 K. From this comparison one comes to the energy scale $\epsilon_0 \approx 8 - 9$ meV. The next step is to check the possibility to describe with the exponential DOS the abrupt increase of the FWHM from 30 meV to 70 meV observed in the experiment for the temperature range from 15 K to 90 K. Since the DOS energy scale was roughly estimated as $\epsilon_0 \approx 10$ meV the simulation should provide the increase of the quantity $\Delta\epsilon/\epsilon_0$ from 3 to 7. As follows from Fig. 3 such increase is provided with very reasonable values of parameters $N\alpha^2 = 1$ and $\nu_0\tau_0 = 10^4$.

Computer simulations were also carried out with a Gaussian DOS

$$g(\epsilon) = \frac{N}{\epsilon_0\sqrt{2\pi}} \exp\left(-\frac{\epsilon^2}{2\epsilon_0^2}\right). \tag{2}$$

Corresponding results for the FWHM, $\Delta\epsilon$, and for the Stokes shift $\epsilon_{\max}$ are shown in Fig. 4. In contrast to the case of the exponential DOS, all PL characteristic features for the Gaussian DOS are sensitive to the choice of parameters $N\alpha^2$ and $\nu_0\tau_0$. Moreover, for the case of a Gaussian DOS, it appears not possible to describe consistently various observed PL features with a single set of model parameters. For example, analytical calculations for the Gaussian DOS show that the upper limit of the PL linewidth is $\Delta\epsilon \approx 2.4\epsilon_0$ [5]. In the experiment the upper limit of the FWHM is $\Delta\epsilon = 73$ meV. Hence, the DOS energy scale ϵ_0 can be estimated as $\epsilon_0 \approx 30$ meV. On the other hand, the simulation with a Gaussian DOS predicts the abrupt increase of $\Delta\epsilon$ at $kT \approx 0.6\epsilon_0$, i.e., $T \approx 210$ K for suggested $\epsilon_0 \approx 30$ meV. This temperature is approximately twice as large as the temperature $T \approx 90$ K observed in the experiment. Therefore we have to conclude that the Gaussian DOS is not suitable for description of the energy disorder in the studied QW's.

3 Conclusions

The PL linewidth and Stokes shift were measured in GaInNAs/GaAs quantum wells in the temperature range between 15 K and 300 K. The experimental results were compared with computer simulations of the exciton hopping relaxation via localized states. It is shown that the exponential energy distribution of localized states with a characteristic energy scale $\epsilon_0 \approx 9$ meV provides the best agreement with all pronounced experimentally observed features of the PL spectrum. In contrary, the Gaussian energy distribution fails to fit experimental data with any choice of model parameters. It is shown herewith that the study of the exciton luminescence in the hopping regime provides a strong tool to characterise the disorder-induced effects in III-V quantum wells.

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