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Influence of exciton localization on recombination line shapes: $In_xGa_{1-x}As/GaAs$ quantum wells as a model

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We present an approach for the optical interband density and for luminescence line shapes of quantum-well excitons by considering the localization of the excitonic center of mass due to potential fluctuations. The localization-induced violation of the $K=0$ selection rule effects considerably the high-energy side of the recombination line shape. Perfect agreement between line-shape simulations and photoluminescence spectra of $In_xGa_{1-x}As/GaAs$ quantum wells is obtained over the full temperature range from $T=2$ K up to 120 K. Furthermore, by introducing a quantitative measure of the degree of localization we find the exciton motion restricted to about 13 nm in the present case.

Currently, large efforts are made to study semiconductor heterointerface morphology playing a fundamental role for transport and optical properties of electronic and photonic devices.[1] The analysis of exciton localization being a consequence of interface roughness or composition variation-induced potential fluctuations[2,3] represents a powerful tool for characterization of interfaces. Most experiments concerning exciton localization concentrated in the past to lattice-matched GaAs- (Refs. 3 and 4) and InP-based[5,6] heterostructures. Little work was devoted to strained pseudomorphic heterostructures like $In_xGa_{1-x}As/GaAs$,[7,8] which are technologically particularly interesting and theoretically challenging.

Photoluminescence (PL) gives a direct proof of exciton localization and suppressed center-of-mass motion by, first, the redshift on its maximum energy compared to the absorption peak (Stokes shift),[3,9] second, by line splitting into different components corresponding to locally separated regions of different potential if their lateral extension is large enough,[10] and third, the *high-energy tail* of the spectrum as will be discussed in detail.

With few exceptions (e.g., Refs. 3 and 9) it is usually assumed that localization influences only the peak position and low-energy side of the PL spectrum, namely, below the absorption line center,[4,11] because the role of *momentum nonconservation* is ignored. Only the extreme but unrealistic cases of complete momentum conservation[3,12] *or* momentum nonconservation[3,13] corresponding to perfectly delocalized or perfectly localized excitons in real space, respectively, were discussed until now. Within this oversimplified picture good agreement between experiment and theory was obtained for low temperatures <10 K and room temperature[3] whereas at any intermediate temperatures attempts failed to fit line shapes.

The transition between both states implies the existence of an intermediate state of *partially localized* excitons. Recent experimental work[4,14] proving the existence of theoretically predicted[15] phonon-mediated migration of "localized excitons" between potential minima reducing their degree of localization supports this assumption.

In this paper a model considering the impact of *partial localization* of the excitonic center of mass (COM) on *partial violation of momentum conservation* in optical recombination influencing mainly the *high-energy tail* of the emission spectrum is presented. Spectra in the whole temperature range between 2 and 120 K are perfectly modeled. Experimental results are obtained for pseudomorphic $In_xGa_{1-x}As/GaAs$ quantum wells, a system which turns out to be particularly advantageous for such investigations because the heavy-hole–light-hole (hh-lh) splitting is sufficiently large to eliminate any lh contribution to the spectra.

In order to obtain an analytical expression for the recombination line shape we first consider the optical interband density (OD)—which is proportional to the absorption spectrum—of an ideal quantum well (QW) without any potential fluctuations. In this case the OD $\alpha(\omega)$ is locally invariant in two dimensions. Since exciton-exciton interaction can be neglected under low excitation the excitons move freely in the plane, being described by plane waves $\exp(iKR)$ [R and K are the two-dimensional (2D) center-of-mass coordinate and momentum, respectively]. Neglecting the small photon momentum, radiative recombination is only possible at $K=0$. The broadening mechanism in this ideal case is only homogeneous, that is, lifetime determined.

In realistic structures inhomogeneous broadening mechanisms caused by interface roughness and/or composition fluctuations are usually dominant. Especially in $In_xGa_{1-x}As/GaAs$ QW's, where the ternary component

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forms the well material, alloy fluctuations remain non-negligible up to large QW widths.[8] The amount to which width and alloy fluctuations contribute to line-shape broadening depends on whether their diameter is bigger or smaller as compared to the exciton diameter.

In the following, we restrict ourselves on deriving analytic expressions for broadening induced by QW width (L_z) fluctuations. The effect of alloy fluctuations is qualitatively identical. Furthermore, we consider only potential variations on a subexcitonic scale sensored by the relative part of the excitonic wave function. Consequently, the resulting exciton potential $V_X(R)$ differs slightly from place to place. In case the average diameter d_I of the growth island is much smaller than the exciton radius a_B, $V_X(R)$ is Gaussian distributed with standard deviation σ_V, given approximately by

$$\sigma_V^2 \approx \langle \Delta L_z^2 \rangle \left[\frac{dE_X}{dL_z} \right]^2 \frac{d_l^2}{\pi a_B^2} , \tag{1}$$

where E_X is the exciton transition energy.

Since each exciton averages the fluctuations over an area πa_B^2 the standard deviation of the confining potential due to L_z variations $\langle \Delta Lz \rangle^{1/2} l \, dE_X/dL_z l$ is attenuated as expressed in (1). Usually it is assumed that the optical density of the exciton coincides with the potential distribution.[12] In that picture, however, each exciton is supposed to be strongly localized, neglecting completely the exciton COM motion that is governed by the random potential $V_X(R)$. It has been shown[16] that $V_X(R)$ has a typical correlation length $l_{\text{corr}} \approx a_B$, much bigger than the interface roughness scale d_I. An exciton moving in $V_X(R)$ is called *partially localized.*

Figure 1 illustrates the effect on the exciton COM motion on the OD line shape. The solid curve results from a solution of the Schrödinger equation[16] and subsequent statistical averaging. For the latter we employ the exact method by Halperin,[17] which, however, works in one dimension only. The potential is assumed to be piecewise constant (length a_B) and uncorrelated outside. The potential distribution is shown as dashed curve in Fig. 1 (Gaussian with standard deviation σ_V). The shape

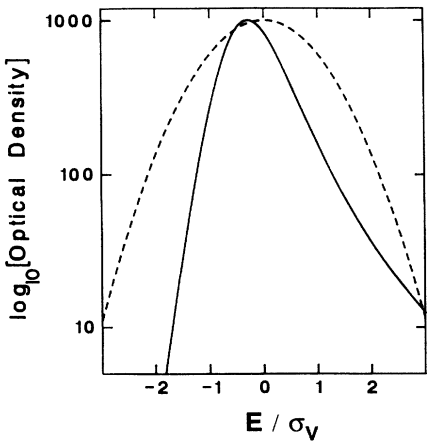

FIG. 1. Optical density of the ground-state exciton ($\sigma_V/E_{\text{ref}}=1$, solid line) moving in a random COM potential with distribution given by the dashed line.

of the OD depends only on the ratio σ_V/E_{ref}, where $E_{\text{ref}}=\hbar^2 a_B^{-2}/2M$ is a reference energy describing the quantized COM motion (M is the COM exciton mass). The solid curve in Fig. 1 displays the calculated OD for $\sigma_V/E_{\text{ref}}=1$, which is a quite realistic value, since E_{ref} is typically 1.8 meV for the samples under consideration.

As can be seen, the OD possesses a smaller full width at half maximum as compared to the potential distribution. This effect can be understood as an increase of the effective averaging volume due to the COM motion. Furthermore, the line is asymmetrically shaped, having Gaussian and nearly exponential slopes on the low- and high-energy side, respectively. Without disorder, the OD would reduce to a delta function since only the exciton ground state is considered here. For Frenkel excitons in a disordered chain, a similar "motional narrowing" has been calculated.[18]

Since the solution of the problem in two dimensions encounters fundamental difficulties, the results obtained for one dimension are taken as a motivation for a heuristic line-shape model of QW excitons. The key feature taken over from our 1D model is the asymmetric line shape (exponential versus Gaussian) which will appear in the recombination of partially localized excitons. Due to localization the exciton COM momentum K no longer represents a good quantum number, and the $K=0$ selection rule in optical recombination will be violated. Not only excitons with vanishing momentum will recombine, but a statistical distribution with a finite width around the $K=0$ state contributes to the emission spectrum. This violation of the strong K-selection rule is described by folding α in K space with a statistical distribution function. The momentum left from recombination of $K \neq 0$ excitons is transferred to the localizing potential. Additionally, the potential fluctuations give rise to a broadening in energy.

In our model both broadenings are described by Gaussians with standard deviations ΔK and σ_E, respectively, and are taken to be fixed throughout the entire spectrum. The OD is then written as

$$\bar{\alpha}(\hbar\omega) = \int \left[dE \frac{1}{\sqrt{\pi}\sigma_E} \exp\left[-\frac{(E-E_0)^2}{\sigma_E^2} \right] \right] \times \int \left[d^2K \frac{1}{\pi \Delta K^2} \exp\left[-\frac{K^2}{\Delta K^2} \right] \right] \times \alpha[\hbar\omega - E - E(K)] , \tag{2}$$

where E_0 is the average transition energy between the sublevels. The dispersion of the exciton COM energy is assumed to be $E(K)=\hbar^2K^2/2M$. Rearrangement of (2) leads to a folding function $\Delta(s)$ that becomes proportional to $\exp(-s^2/\sigma^2)$ and $\exp(-s/\eta)$ for $s \to -\infty$ and $s \to +\infty$, respectively, where the localization parameter η is defined as $\eta=\hbar^2\Delta K^2/2M$.

We use the approximation of the OD of bound and continuous excitonic states in QW's given by[19]

$$\alpha(E) = \sum_{n=1}^{\infty} \frac{2}{(n-q)^3} \delta\left[\frac{E}{R} + \frac{1}{(n-q)^2} \right] + \theta\left[\frac{E}{R} \right] , \tag{3}$$

where R is the bulk exciton Rydberg, and

$q=1-(R/E_B)^{1/2}$ designates the quantum defect which describes the reduced dimensionality of the system, E_B being the exciton binding energy in the QW.

The line shape of the PL spectrum is obtained from (1) simply by multiplication with the thermal occupation function, approximated by a Boltzmann function $\exp(-\hbar\omega/k_BT)$ in our case. This reflects the assumption that excitons can thermalize locally to regions of lower potential.

The effect of partial non-K-conservation of the OD and the PL spectrum is demonstrated in the calculated spectra of Fig. 2. For $\eta=0$ [complete K conservation, Fig. 2(a)] simple Gaussian line shapes for 1s and 2s excitons are observed in the OD and PL spectra while higher exciton lines merge into the broadened step function forming the 2D continuum. With increasing η [Fig. 2(b)] an exponential slope on the high-energy side of the 1s OD appears yielding a pseudotemperature Boltzmann tail in PL while the low-energy side has a Gaussian line shape determined by σ_E. The slope of the continuum tail still corresponds to the real carrier temperature. Only for $\eta\rightarrow\infty$ [Fig. 2(c)] does the excitonic OD become a steplike function and the high-energy tails of 1s excitons and continuum have roughly the same slope in PL. However, σ_E and η cannot be chosen independently since both depend on the same potential fluctuations. In Fig. 2, to illustrate clearly the effect of partial K conservation on PL line shape, we have set σ_E artificially to the small value of 0.4 meV. This is why the shift of the PL maximum to lower energies (Stokes shift) is quite small and is not resolved in Fig. 2(a).

We investigated a series of five $In_xGa_{1-x}As/GaAs$ quantum-well samples with In concentration varying from $x=0.13-0.19$. Each sample contains four single quantum wells of nominal width $L_z=10$, 6, 4, and 2 nm, being grown by molecular-beam epitaxy with a 190-nm GaAs barrier between each. The critical thickness for $x\leq 0.2$ is larger than 16 nm (Ref. 20). Thus the $In_xGa_{1-x}As$ layers are pseudomorphical and tetragonally strained. PL was excited by 500 mW/cm^{-2} of 514.5-nm Ar^+-laser light and measured by a Si avalanche diode with a spectral resolution of 0.08 nm.

A typical 5-K PL spectrum is shown in Fig. 3(a). The main peak results from the 1s heavy-hole (hh) exciton recombination. The smaller maximum can be identified as the 2s hh exciton emission followed by higher excitons and the continuum on its high-energy side. This identification is based mainly on two arguments: First, the temperature T_C deduced from the high-energy tail of the continuum is identical to the lattice temperature T_L, for T_L ranging from 50 K to room temperature, and is approximately 20 K for $T_L=2$ K. Thus, it represents a reasonable carrier temperature. Second, the exciton binding energies determined roughly from the peak differences and more accurately from line-shape evaluation are in perfect agreement with other data obtained for comparable samples by different methods[8] and with theory.

As a particularity of the investigated material system the light-hole band is shifted about 100 meV to higher energies due to the tetragonal distortion of the unit cell and is not superimposed to the hh PL. This favors the $In_xGa_{1-x}As/GaAs$ system for detailed investigation of the PL line shape compared to lattice-matched QW heterostructures where the lh exciton emission masks the continuum emission making line-shape evaluations impossible.

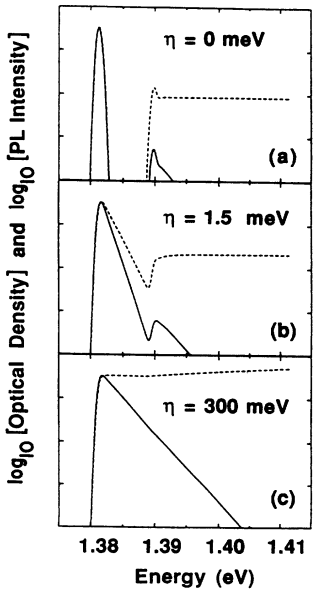

FIG. 2. Calculated hh exciton and continuum OD (dashed line) and PL (solid line, $T=30$ K) spectrum ($\sigma_E=0.4$). (a) Perfect K conservation, $\eta=0$ meV. (b) Partial K conservation, $\eta=1.5$ meV. (c) Complete violation of K selection rule, $\eta=300$ meV.

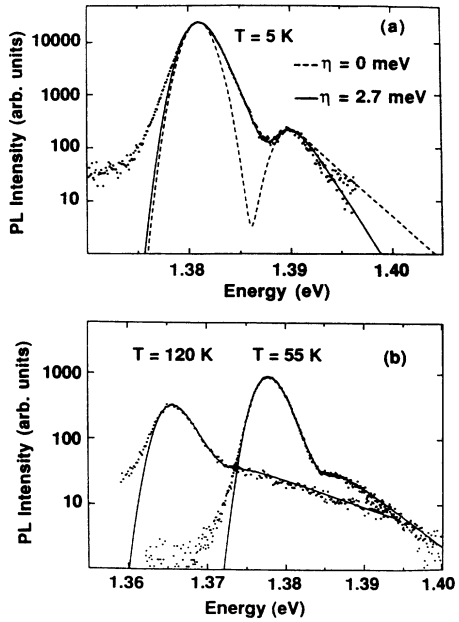

FIG. 3. (a) Low-temperature PL spectrum of an $In_xGa_{1-x}As/GaAs$ QW ($L_z=6$ nm, $x=0.16$). Experiment (points) and simulation assuming complete (dashed line) and partial (solid line) K conservation. (b) High-temperature PL spectra of the same QW as in (a). Experiment (points) and simulation with partial K conservation (solid line).

Figure 3 shows two fits to the experimental PL line shape. The dashed curve represents the best fit obtained with $\eta \approx 0$. This fit is quite bad for the $1s$ high-energy tail although the temperature used is even higher (30 K) than permitted by the continuum tail (≈ 16 K). The other fit parameters, like the exciton binding energy E_B, the sublevel energy E_X, and the sublevel broadening σ_E, are quite well defined by the peak difference, the peak position, and the curvature of both peak maxima, respectively. The solid curve is calculated using $\eta = 2.7$ meV. It represents a perfect fit to the whole high-energy side of the spectrum. The low-energy side is slightly influenced by impurity states not discussed here. All fit parameters being fixed by different features of the spectrum, it was found necessary to attenuate the $1s$ exciton oscillator strength by a factor $\beta = 0.08$ as compared to the $2s$ and continuum emission (see below).

The localization parameter η being related to the momentum uncertainty ΔK, a lower limit for the restriction of the exciton center-of-mass movement can be derived from Heisenberg's relation $\Delta K \Delta R \geq 1$. For the typical value of $\eta \approx 2.7$ meV we find $\Delta R \geq 10$ nm which is in reasonable agreement with the scale of the exciton Bohr radius.

Figure 3(b) gives two examples for PL line-shape fits at higher temperatures. Apart from slight differences at the low-energy side (two orders of magnitude below the peak intensity), perfect agreement is obtained for both spectra. Since the experimental PL intensity decreases considerably at still higher temperatures, it is found to be very difficult to obtain low excitation spectra allowing reasonable line-shape analysis. For $T > 120$ K, however, no fundamental change concerning the PL simulation is expected.

σ_E and η do not exhibit any important variations in the investigated temperature range. The factor β, however, which expresses the attenuation of the $1s$ exciton peak, shows an increase by one order of magnitude for $T > 5$ K for all samples. For temperatures $T \geq 50$ K we find $\beta \approx 0.9-1$. The surprising need to correct the theoretically determined relation of oscillator strength by a factor $\beta \simeq 0.08$ at low temperatures might be explained by assuming a nonequilibrium distribution between $1s$ and higher excited excitons. This hypothesis is supported by the temperature dependence, which—in this interpretation—shows that thermal equilibrium between ground and higher exciton states is established at higher temperatures due to enhanced exciton-exciton interaction. A calculation based on a simple rate equation yields a ratio $\tau_{rel}/\tau_{rec} \approx 0.18$ for $\beta \approx 0.08$, τ_{rel} and τ_{rec} being the relaxation time $2s \rightarrow 1s$ and the exciton recombination time, respectively. This ratio is in accordance with recent theoretical[14] and experimental[21,5] work that determines the relaxation time of localized excitons to be $10 < \tau_{rel} < 100$ ps in $In_xGa_{1-x}As$ QW's and the recombination time being of the order of several ns. It is also consistent with recent experiments[22] demonstrating the increase of the thermalization rate with temperature.

In summary, an approach for the optical density that takes into account the partial localization of the exciton center-of-mass motion in a fluctuating potential caused by interface roughness or alloy fluctuations is presented. Introducing the concept of *partial violation* of the K selection rule, a heuristic line-shape model is established. Line-shape simulations perfectly agree with experimental PL spectra over a large temperature range. Luminescence of pseudomorphic $In_xGa_{1-x}As/GaAs$ QW's is investigated. This tetragonally distorted system offers the advantage of large hh-lh splitting eliminating the masking of higher state ($n \geq 2$) hh exciton features by the $n = 1$ lh exciton. The localization parameter η deduced from the spectra is a quantitative measure of exciton localization, indicating a restriction of the center-of-mass motion to ≥ 13 nm independent of temperature.

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