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Linewidth of free excitons in quantum wells: Contribution by alloy disorder scattering

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A theory is developed for the luminescence linewidth in a quantum well made of ternaries when the two-dimensionally free excitons undergo alloy disorder scattering. The expression for linewidth shows a L^{-1} dependence on the well width L for infinite barrier height. For thin wells leakage of wave function into the barrier is considered. A comparison between the experimental data and the present values points out the dominant role of alloy disorder scattering. For wells thinner than 40 Å the calculated values decrease with decreasing well width, indicating the importance of surface roughness scattering.

The low-temperature optical properties in a quantum well (QW) are governed by the creation and recombination of two-dimensional (2D) excitons. Depending on the size of the island-like fluctuation in the width of the well relative to the Bohr radius, these 2D excitons may be either localized or free.[1,2] The luminescence linewidth (LW) of both the localized and free excitons has been the subject of intense theoretical and experimental investigations in recent years. With the progress in growth techniques, extremely high quality QWs are now available in which the LW and its temperature dependence may be solely governed by the scattering of free excitons by different agents. So far the values of LW have been calculated by considering acoustic phonon[3,4]: both deformation potential and piezoelectric, polar optic phonon,[5] impurity,[3] carrier-carrier,[6] and interface roughness scattering.[7]

Extensive work on the LW in InGaAs-based QWs has been performed by several workers.[8,9] The theoretical work reported so far [7,10,11] is based on the model of Schubert *et al.*,[12] in which the excitons are thought to be bound and the variance in the probability of having one of the cations within the exciton volume is calculated. On the other hand, if excitons are free, as in a high quality sample, the LW will be predominantly due to scattering by alloy disorder, much in the same manner as the mobility[13–15] and cyclotron resonance LW[16] in heterojunctions and QWs are determined by alloy scattering of 2D electrons. The relevant theory for LW is not, however, developed yet. The present letter gives an outline of the theory and provides an approximate expression for the LW. More refined expressions may, however, be obtained from a straightforward extension of the present work. The calculated values are compared with the available experimental data. The range of applicability and the limitations of the present model are then discussed.

The exciton wave function is written as[4]

$$|\mathbf{K}\rangle = \sum_{\mathbf{k},\mathbf{k}'} f(\mathbf{k},\mathbf{k}',\mathbf{K})\delta_{\mathbf{k}\ \ \mathbf{k}',\mathbf{K}} a_{c\mathbf{k}}^{+} a_{v\mathbf{k}'} |O\rangle, \tag{1}$$

with

$$f(\mathbf{k},\mathbf{k}',\mathbf{K}) = S^{-1}\int d^2r \int dz_e \int dz_h F(r,z_e,z_h) \times \exp[i(\alpha_e\mathbf{K}-\mathbf{k})\cdot\mathbf{r}],$$
$$\alpha_e = m_e/M, \alpha_h = m_h/M, \text{ and } M = m_e + m_h. \tag{2}$$

Here, $\mathbf{k}$ and $\mathbf{k}'$ are Bloch wave vectors, $\mathbf{K}$ is the exciton wave vector in the plane of free motion, a^+ and a are, respectively, the creation and annihilation operators in the Bloch representation, $c(v)$ refers to the subbands in the conduction (valence) band, $|O\rangle$ is the crystal ground state, S is the surface area of the crystal, $m_{e(h)}$ is the electron (heavy hole) effective mass, r is the relative coordinate between the electron and hole (in the x-y plane), and the quantization occurs in the z direction.

In the present work, the following variational wave function is employed for the 1s exciton state[3]:

$$F = A\exp(-\beta r/2)\cos(\pi z_e/L)\cos(\pi z_h/L), \tag{3}$$

where A is the normalization constant, β is the variational parameter, and L is the width of the well.

We assume that the scattering potential under virtual crystal approximation is a spherically symmetric square well of height ΔU_μ ($\mu = e,h$) and radius r_0. The potential at site $(\mathbf{r}_i,z_i)$ may be expanded in the following 2D Fourier series[13,14]:

$$V_i(\mathbf{r}_i,z_i) = \sum_{\mathbf{q}} 2\pi\Delta U_\mu\left(\frac{bJ_1(bq)}{q}\right) \times \exp(i\mathbf{q}\cdot|\mathbf{r}-\mathbf{r}_i|), \quad z_i - r_0 \leqslant z_\mu \leqslant z_i + r_0, \tag{4}$$

where $b^2 = r_0^2 - |z-z_i|^2$. Using this form of the potential, the matrix element for transition from a state $|\mathbf{K}\rangle$ to another state $|\mathbf{K}'\rangle$ may be expressed as

$$\langle\mathbf{K}'|V_i|\mathbf{K}\rangle = H_e(-\alpha_h\mathbf{q}) - H_h(-\alpha_e\mathbf{q}), \tag{5}$$

where

$$H_\mu(Q) = \int d^2r \int_{z_i-r_0}^{z_i+r_0} dz_\mu \int_{-L/2}^{L/2} dz_\nu \left(\frac{2\pi b\Delta U_\mu J_1(bq)}{q}\right) \times |F|^2 \exp[i(\mathbf{Q}\cdot\mathbf{r} - \mathbf{q}\cdot\mathbf{r}_i)]\delta_{\mathbf{K}'-\mathbf{K},\mathbf{q}}. \tag{6}$$

In order to evaluate $H_\mu(\mathbf{Q})$, it is assumed that r_0 is small, so that the approximation $J_1(x) = x/2$ is valid.[13] The integration over z_μ is performed by taking $\cos(\pi z_\mu/L) = \cos(\pi z_i/L)$ and then bringing this term outside the integral, obtaining thus a factor $(4r_0^3/3)$ after integration. The above approximation has been found to give negligible deviation of the values of mobility of 2D electrons from the values obtained by exact integration.[14,16] The r integration is straightforward and the matrix element is given by

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$$\langle \mathbf{K}'|V_i|\mathbf{K}\rangle = A^2 \sum_{\mathbf{K}'} \exp(-i\mathbf{q}\cdot\mathbf{r}_i)\,\frac{4\pi r_0^3}{3}\cos^2\left(\frac{\pi z_i}{L}\right)$$
$$\times\{\Delta U_e[\beta^2+(\alpha_h q)^2]^{-3/2}$$
$$-\Delta U_h[\beta^2+(\alpha_e q)^2]^{-3/2}\}. \quad (7)$$

The total scattering probability is calculated by considering the contributions from all the alloy sites, which are assumed to be randomly distributed. Converting summations over i and $\mathbf{K}'$ into integrations, the transition probability $W(\mathbf{K})$ for being scattered from the state $|\mathbf{K}\rangle$ may be calculated. However, the peak of the excitonic transition occurs at $\mathbf{K}=0$, and it is standard practice[3,5,6] to relate the linewidth Γ to $\hbar W(\mathbf{K}=0)$. In this approximation the LW becomes

$$\Gamma = [8\pi^2 Nx(1-x)r_0^6 M/(3\hbar^2 L)](\Delta U_e - \Delta U_h)^2, \quad (8)$$

where N is the number of alloy sites per unit volume. The LW decreases with increasing well width and is independent of temperature.

We have calculated the values of LW of excitons in InGaAs/InP QWs using Eq. (8). The values of parameters used in the calculation are $m_e = 0.042m_0$,[13] $m_h = 0.465m_0$,[17] $\Delta U_e = 0.53$ eV,[15,18] $\Delta U_h = 0.18$ eV,[19] $r_0 = \sqrt{3}a/4$, $a = 5.87$ Å.[13] The calculated values of LW are plotted against the well width in Fig. 1 as the dashed curve. The experimental values reported by Tsang[8] and by Skolnick *et al.*[9] are shown in the figure as squares and circles.

A comparison between the experimental data and the present calculations indicates that there is only qualitative agreement between the two. One must, however, take into consideration the fact that the impurity and surface roughness scattering have their contributions to the LW, the exact magnitudes of which are not known. In view of this one should not expect complete agreement between the experimental data and the present theoretical values due to alloy scattering alone. The only conclusion that may be drawn from the comparison is that the alloy scattering is the dominant process in determining the LW. A similar conclusion about the importance of the alloy scattering has been drawn in the study of mobility and cyclotron resonance of 2D electron gas.[13–16]

The theoretical values are quite large compared to the experimental ones for thin wells ($L < 50$ Å), for which the wave functions given by Eq. (3) valid for infinite barrier height are not acceptable. We have calculated the wave functions $\psi_e(\psi_h)$ and subband energies $E_e(E_h)$ of electrons (holes) by taking the value of $V_e(V_h)$: the conduction (valence) band discontinuities at the interface to be 220 (320) meV.[21] In the calculation, the effect of mismatch of the effective masses at the well and barrier layers is ignored. The wave function is given by[20,22]

$$\psi_e = B_e\cos(x_e z), \quad |z| < L/2$$
$$= C_e\exp(-y_e z), \quad z < -L/2, \quad z > L/2, \quad (9)$$

where $x_e^2 = 2m_e E_e/\hbar^2$, $y_e^2 = 2m_e(V_e - E_e)/\hbar^2$, $B_e^2 = [y_e/(1+y_e L/2)]$, and the subband energy E_e is obtained by solving[20]

$$[E_e/V_e]^{1/2} = \cos[(m_e E_e/2\hbar^2)^{1/2} L].$$

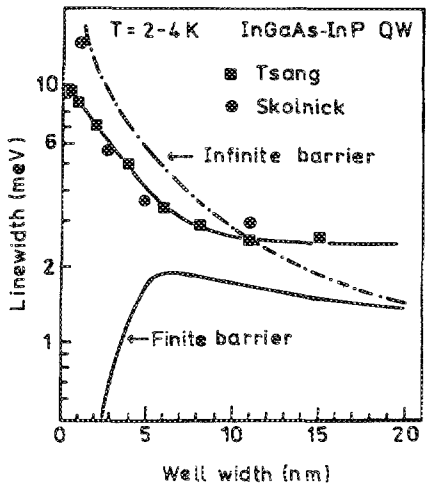

FIG. 1. Variation of photoluminescence linewidth with the thickness of the quantum well. Squares and circles: experimental data obtained by Tsang (Ref. 8) and Skolnick *et al.* (Ref. 9), respectively. Dash dotted curve: values obtained from Eq. (8). Solid curve: values of LW taking leakage of wave function into account.

The expression for the LW employing the previous approximation becomes

$$\Gamma = \left(16\pi^2 Nx(1-x)r_0^6\frac{M}{9\hbar^2}\right)\int_{L/2}^{L/2}[B_e^2\Delta U_e\cos^2(x_e z_i)$$
$$-B_h^2\Delta U_h\cos^2(x_h z_i)]^2 dz_i. \quad (10)$$

The integrals in Eq. (10) are lengthy but can be evaluated analytically.

The values of LW obtained by using Eq. (10) are plotted in Fig. 1 as the solid curve. It is found that there is negligible penetration of the wave functions for $L > 150$ Å. The curve shows that the LW has a peak at about 70 Å. The calculated values of LW are now lower than the experimental data, so that there is room for the contributions from other sources, especially from surface roughness scattering.

In conclusion, we have developed a theory for the excitonic LW assuming scattering of free excitons by alloy disorder, and have found that this scattering is the dominant mechanism in InGaAs/InP QWs for moderate values of well width.

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