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ANOMALOUS MAGNETOPHOTOLUMINESCENCE AS A RESULT OF LEVEL REPULSION IN ARRAYS OF QUANTUM DOTS

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Selectively excited photoluminescence (SPL) of an array of self-organized $In_{0.5}Ga_{0.5}As$ quantum dots has been measured in a magnetic field up to 11T. An anomalous magnetic field sensitivity of the SPL spectra has been observed under conditions for which the regular photoluminescence spectra are almost insensitive to the magnetic field due to large inhomogeneous broadening. The anomalous sensitivity is interpreted in terms of the repulsion of excited levels of the dots in a random potential. A theory presented to describe this phenomenon is in excellent agreement with the experimental data. The data estimated the correlation in the positions of excited levels of the dots to be 94%. The magnetic field dependence allows the determination of the reduced cyclotron effective mass in a dot. For our sample we have obtained $m_e m_h/(m_e + m_h) = 0.034m_0$.

An array of self-organized quantum dots (QDs) is a unique system consisting of very small atomic-like objects each with a few energy levels [1–4]. Studies of this system have shown the possibility to attain three-dimensional confinement of carriers within QDs. Such quantum dots are formed in highly strained semiconductor heterostructures by what is known as Stranski–Krastanow growth, where growth starts two-dimensionally, but after a certain critical thickness is reached, islands are formed spontaneously, and a thin wetting layer is left under the islands. In this process, the growth is interrupted immediately after the formation of the islands and before strain relaxation and misfit dislocations occur. Such in-situ formation of 0D quantum dots results in high quality defect-free materials. In addition, the coherent islanding and strain effects can produce QDs with a size uniformity within ±10% which is very promising for 0D quantum devices where the sharper density of states is exploited.

Photoluminescence (PL) spectrum of such an array has a broad line which is supposed to be mainly due to inhomogeneous broadening [5, 6]. Photoluminescence excitation (PLE) and selectively-excited photoluminescence (SPL) reveal a fine structure. This fine structure has been interpreted by one of us [7] as a result of splitting of the excited levels in quantum dots due to violation of cylindrical symmetry of the dots by a random potential.

In this work we have found an anomalous sensitivity of the SPL to a magnetic field in the region of this fine structure. The regular photoluminescence spectra are almost insensitive to such a small magnetic field due to a large inhomogeneous broadening. We show that this sensitivity is a direct result of the level repulsion. A preliminary discussion of this effect has been given earlier [8].

The dot layer studied here is pseudomorphically grown by MBE on (100) GaAs substrate, and the QDs are formed by the coherent relaxation into islands of a few monolayers of $In_{0.5}Ga_{0.5}As$ between GaAs buffer and cap layers. The actual amount of indium incor-

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porated in the dots can differ due to the complex dynamics of the adatoms during island formation. The growth and QD structural details have been reported earlier [9].

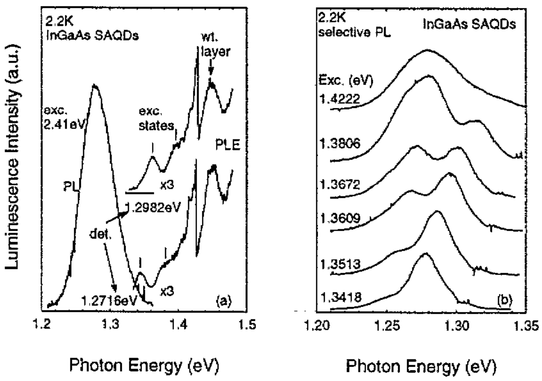

Fig. 1. PL and PLE (a) and SPL (b) spectra of InGaAs QDs with different excitation and detection energies.

Figure 1(a) shows PLE and regular PL spectra. Regular PL reveals a broad line with the FWHM=57 meV. The PLE spectra consist of significantly narrower lines. The PL spectrum is red-shifted with respect to the PLE by about 80 meV. This shift occurs since the PLE experiment detects the light emitted only by the levels below the level excited by the pumping light.

The SPL spectra for different excitation energies are presented in Fig. 1(b). The complicated character of the SPL spectra was analyzed earlier [7]. For our purposes it is important that at some energy of excitation (E_{ex} = 1.3672 eV) the SPL spectrum shows two symmetric peaks. At larger and smaller E_{ex} the intensities of these peaks become asymmetric.

For an intuitive picture it is helpful to assume that each dot has two optically active excited levels $E_\pm$ relatively close to each other (see Fig. 2). The dots are isolated from each other, so the light is emitted from the same dot it is absorbed in. The dots are excited into E_+ or E_- and, after thermalization, emit light from E_0. The red shift mentioned above originates from the difference $E_\pm - E_0$. In some dots the excitation energy $\hbar\omega_{ex}$ may be close either to E_- or to E_+. These two types of dots can be considered as two different subsets. These subsets should give two peaks in the SPL curve if we assume the correlation between the positions of $E_\pm$ and E_0. If this correlation is of such kind that the dots with larger $E_\pm$ have, in general, larger E_0, the excitation of E_+ will cause the lower peak in SPL. This is a natural proposal and it corresponds to our experimental data (see Fig. 1(b)). At some energy of excitation there are the same amounts of dots in each subset. However, if the energy of excitation is shifted upward, the number of the dots which have the lowest excited level at this energy is smaller, so the intensity of the higher-energy peak decreases. Correspondingly, the low-energy peak disappears as the excitation energy decreases.

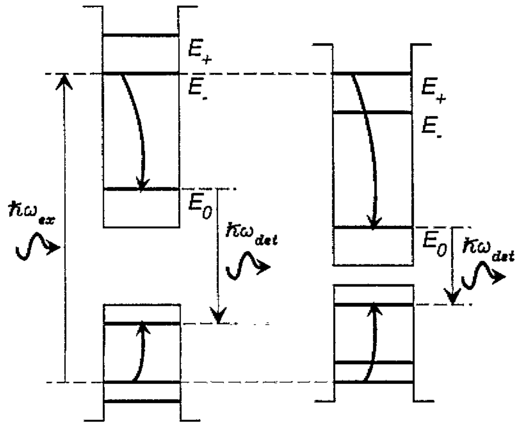

Fig. 2. An intuitive picture of the relevant energy levels in QDs. Note that excitation into the higher level gives lower-energy features in SPL

In our interpretation two close optically active excited levels originate from the doubly degenerate lowest excited level of a cylindrically symmetric dot. These levels are split by a random potential which may include a deviation of a dot shape from cylindrical symmetry. It is crucial that both the distance between the peaks and their width, as well as the linewidth of the nonselective PL have the same origin. They are determined by the random potential which splits degenerate levels and shifts randomly all energy levels in QDs.

At the excitation energy E_{ex} = 1.3672 eV, which gives two symmetric peaks in the SPL, we have studied the magnetic field dependence of the SPL spectrum. The results presented in Fig. 3 show anomalous sensitivity to the magnetic field. The relative intensity of the dip between two peaks decreases by 11% in the magnetic field of 2T. Note that the wide line of the regular (non-selective) PL is almost insensitive to a magnetic field up to ~10T [10, 11]. In the self-organized InP islands the effect of magnetic field on PL is larger [12], but still it is much weaker than the effect in the SPL we present here.

We show below that the two-level structure of SPL at

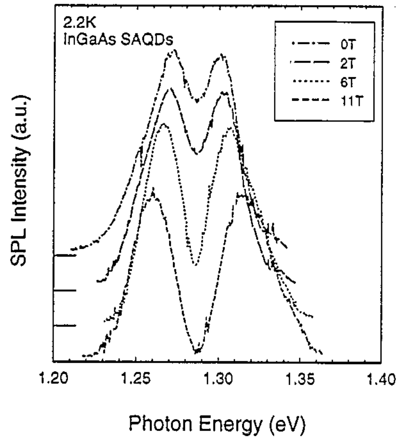

Fig. 3. SPL spectra under different magnetic fields. The excitation energy is $\hbar\omega_{ex}$=1.3672 eV. The spectra have been shifted upward for clarity and zero levels are indicated by the horizontal bars at the left.

zero field and its anomalous sensitivity to the magnetic field are both the results of the level repulsion.

For an axially symmetric dot two degenerate wave functions with an angular momentum $|m|$ have the form $\Psi_{m\pm}(\mathbf{r}) = \psi_m(r)e^{\pm im\phi}$, where ψ_m can be chosen real. For the first excited state m is equal to one. The positions of energy levels $\epsilon_\pm$, split and shifted by the random potential and the magnetic field, can be obtained as the eigenvalues of the secular matrix

$$\delta H = \begin{bmatrix} u + \frac{\hbar\omega}{2}, & x + iy \\ x - iy, & u - \frac{\hbar\omega}{2} \end{bmatrix}, \quad (1)$$

where the matrix elements u and $x + iy$ take random values in each quantum dot and are given by [7, 13]:

$$u = \int d^3r\, V(\mathbf{r})\psi_1^2(\mathbf{r}), \quad (2)$$

$$x + iy = \int d^3r\, V(\mathbf{r})\psi_1^2(\mathbf{r})e^{2i\phi}. \quad (3)$$

Here V(**r**) is an arbitrary Gaussian random potential which can be caused by alloy fluctuations. In principle, the same description is also valid in the case when the cylindrical symmetry is violated by strain fields or the shape of the dots [7].

The values u, x, and y are real independent Gaussian random variables with equal standard deviations σ_1. The eigenvalues of δH are $\epsilon_\pm = u \pm \Delta$, where Δ is the splitting of the excited level given by

$$\Delta = \sqrt{\Delta_0^2 + (\hbar\omega/2)^2}, \quad (4)$$

$\Delta_0 = \sqrt{x^2 + y^2}$ being the splitting in the absence of the magnetic field. The magnetic field provides an extra splitting of the excited level, which for an axially-symmetric dot is equal to the cyclotron energy $\hbar\omega$.

The distribution function for Δ is easy to calculate:

$$F(\Delta) = \frac{2\Delta}{\sigma_1^2}\exp\left(-\frac{\Delta^2 - (\hbar\omega/2)^2}{\sigma_1^2}\right) \times \theta\left(\Delta^2 - (\hbar\omega/2)^2\right), \quad (5)$$

where θ is the step function. At zero magnetic field $F(0) = 0$ due to the level repulsion. In a finite field $F(\Delta) = 0$ in the region $|\Delta| < \hbar\omega/2$ since the splitting cannot be less than $\hbar\omega$. Now we show that PLE and SPL lineshapes are closely related to the function $F(\Delta)$.

To find the lineshapes for both PLE and SPL, one should calculate the intensity as a function of two frequencies $I(E_{ex}, E_{det})$. The PLE and SPL lineshapes can be obtained from this function by fixing the corresponding variables. We assume that the matrix elements are energy independent, so the intensity is proportional to the distribution function $P(\epsilon_0, \epsilon_1)$ where ϵ_0 and ϵ_1 are the energies of the lowest and the next excited states respectively. To simplify the notation we measure energies from their average values, $\epsilon_\alpha = E_\alpha - \overline{E_\alpha}$.

The lowest state E_0 with the wave function $\psi_0(r)$ is also shifted by the random potential by

$$\epsilon_0 = \int d^3r\, V(\mathbf{r})\psi_0^2(\mathbf{r}), \quad (6)$$

The shift ϵ_0 is also a Gaussian random variable which is statistically independent of x and y, however, in general, it is correlated with u.

The general expression for $P(\epsilon_0, \epsilon_1)$ has the form:

$$P(\epsilon_0, \epsilon_1) = \sum_\pm \iiint du\,dx\,dy\; \delta(\epsilon_1 - u \mp \sqrt{x^2 + y^2 + (\hbar\omega/2)^2}) \times G_2(\epsilon_0, u; \sigma_0, \sigma_1, \rho) G_1(x; \sigma_1/\sqrt{2}) G_1(y; \sigma_1/\sqrt{2}) \quad (7)$$

Here σ_0 is the dispersion of ϵ_0.

The matrix element u determines the overall shift of the excited level due to the random potential. $G_1(\epsilon; \sigma)$ is the normal distribution, $G_1(\epsilon; \sigma) = \exp(-\epsilon^2/2\sigma^2)/\sigma\sqrt{2\pi}$.

The general form of the two-variable normal distribution of the variables ϵ_0 and u which takes into account the correlation between them is

$$G_2(\epsilon_0, u; \sigma_0, \sigma_1, \rho) = \frac{1}{2\pi\sigma_0\sigma_1\sqrt{1 - \rho^2}}$$

$$\times \exp\left\{-\frac{1}{2(1-\rho^2)}\left[\frac{\epsilon_0^2}{\sigma_0^2} - 2\rho\frac{\epsilon_0 u}{\sigma_0\sigma_1} + \frac{u^2}{\sigma_1^2}\right]\right\}, \quad (8)$$

Here ρ is the correlation coefficient, $|\rho| \leq 1$.

Performing the integrals in equation (7) one obtains

$$P(\epsilon_0, \epsilon_1) = G_1(\epsilon_0; \sigma_0) D_\rho(\epsilon_1 - \epsilon_0 \rho \frac{\sigma_1}{\sigma_0}; \sigma_1), \quad (9)$$

where the function $D_\rho(\epsilon; \sigma)$ is defined by

$$D_\rho(\epsilon;\sigma) = \frac{1}{\sigma}\sum_{(\pm\epsilon)}\left\{\frac{\sqrt{\mu-1}}{\mu\sqrt{\pi}}\exp\left(-\frac{(\hbar\omega/2-\epsilon)^2}{(\mu-1)\sigma^2}\right) + \frac{\epsilon}{\sigma\mu^{3/2}}\exp\left(\frac{\mu(\hbar\omega/2)^2-\epsilon^2}{\mu\sigma^2}\right)\times \left[1 - \operatorname{erf}\left(\frac{\mu\hbar\omega/2-\epsilon}{\sigma\sqrt{\mu(\mu-1)}}\right)\right]\right\}, \quad (10)$$

where $\mu = 3 - 2\rho^2$. The function $P(\epsilon_0, \epsilon_1)$ as determined by equations (9) and (10) is the generalization of the function introduced in [7] for zero magnetic field. Equation (10) yields equation (5) with $\epsilon = \Delta$ when $\rho \to 1$.

The effect of level repulsion manifests itself in SPL and in PLE in full scale if the overall shift of $E_\pm$ is proportional to the shift of E_0. This means $\rho = 1$. In this case we get

$$P(\epsilon_0, \epsilon_1) = \frac{1}{\sigma_0\sqrt{2\pi}}\exp\left(-\frac{\epsilon_0^2}{2\sigma_0^2}\right) F\left(\epsilon_1 - \epsilon_0\frac{\sigma_0}{\sigma_1}\right), \quad (11)$$

where F is the distribution function of splittings given by equation (5).

The PLE lineshape can be obtained from equation (11) by fixing the detection energy ϵ_0. One can see that it is just given by the function F. To get the SPL lineshape one should fix ϵ_1. The lineshape of the SPL also reproduces the features of the function F. Because of the level repulsion both SPL and PLE are zero when $\epsilon_1 = \epsilon_0\sigma_1/\sigma_0$. In the magnetic field they are zero in the range $|\epsilon_1 - \epsilon_0\sigma_1/\sigma_0| < \hbar\omega/2$.

If the fluctuations of the lowest and the next excited states were uncorrelated, the structure will be substantially smeared. However, the correlation occurs to be strong. In the model of alloy disorder the correlation coefficient ρ is calculated to be $\rho = 0.795$ [7]. The experimental value we obtain below is $\rho = 0.945$. This coefficient describes the correlation in positions of the lowest excited level E_0 and the next split excited level $E_\pm$. The value of ρ close to 1 suggests that a substantial part of the fluctuations originates from the change of the size of the dots which shifts energy levels proportionally. That is why such a tiny effect as the repulsion of split levels makes SPL spectra very sensitive to a relatively weak magnetic field.

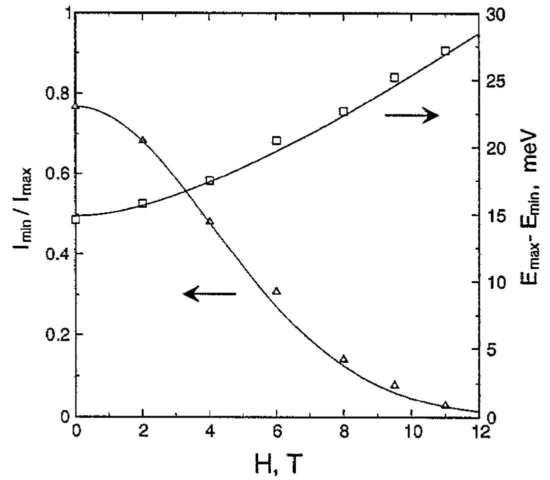

Fig. 4. The relative valley intensity (squares) and peak position (triangles) as a function of magnetic field. The solid lines represent the best fit by equations (9), (10), which yields $m_{\text{eff}} = 0.034m_0$. The parameters of the dots obtained from the zero-field data are $\rho = 0.945$, $\sigma_0 = 25.5$meV, and $\sigma_1 = 21.4$meV.

Figure 4 shows the relative peak position and the relative valley intensity as a function of magnetic field. Solid lines represent the best fit to the experimental data with equations (9), (10). The fit has been performed in the following way. The parameters ρ, σ_0, and σ_1 can be determined from zero-field data. Two dimensionless parameters, ρ and σ_0/σ_1, are obtained from the relative depth of the dip in SPL and the shift of PLE maxima with detection energy as provided by equation (9). This gives $\rho = 0.945$ and $\sigma_0/\sigma_1 = \rho/(dE_{PLE}/d\hbar\omega_{det}) = 1.189$. The absolute value of σ_1 can be found from the relative peak position at zero field, which gives $\sigma_1 = 21.4$ meV and $\sigma_0 = 25.5$ meV. To check the consistency, we can obtain σ_0 from the width of the non-selective PL in Fig. 1(a). Assuming the Gaussian shape of PL, the FWHM=57 meV gives $\sigma_0 = 57/(2\sqrt{2\log 2}) = 24.2$ meV.

With these parameters given, the magnetic-field dependencies of both the relative depth of the dip and the relative peak position are determined by the value of effective mass only. The value $m_{\text{eff}} = 0.034m_0$ gives a perfect fit for both quantities as shown in Fig. 4.

The value of the effective mass we obtained can be understood by taking into account that the cyclotron frequency that enters equation (1) contains the reduced effective mass for the electron and hole, $m_{\text{eff}} =$

$m_e m_h/(m_e + m_h)$. If we assume $m_e = m_h$, our result would imply $m_e = 0.068 m_0$. If we take for the effective mass of the electron the value for bulk $Ga_{0.5}In_{0.5}As$, $m_e = 0.045$ [14], the value of $m_{eff} = 0.034 m_0$ would give for the mass of the hole $m_h = 0.14 m_0$. The small value of the hole mass is reasonable taking into account the two-dimensional nature of the QDs. On the other hand, the effects of strain and confinement are believed to increase the effective mass of the carriers with respect to the bulk material [15, 16].

In conclusion, we have observed anomalous sensitivity of SPL to magnetic fields which are too low to affect the regular PL spectrum. We interpret such sensitivity in terms of the repulsion of energy levels in QDs. We present a theory which describes the phenomenon strikingly well. The data allows to determine the reduced cyclotron effective mass for the carriers in the dots and the correlation in the positions of excited levels.

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