PHYSICAL REVIEW B VOLUME 57, NUMBER 11 15 MARCH 1998-I

Electric-field effects on excitons in quantum dots

W. Heller
Walter Schottky Institut, Technische Universität München, Am Coulombwall, 85748 Garching, Germany

U. Bockelmann
Laboratoire de Physique de la Matière Condensée, Ecole Normale Supérieure, 24 rue Lhomond, 75005 Paris, France

G. Abstreiter
Walter Schottky Institut, Technische Universität München, Am Coulombwall, 85748 Garching, Germany

(Received 7 July 1997)

Using low-temperature microphotoluminescence spectroscopy we have investigated the effect of a lateral electric field on quantum dot excitons trapped by monolayer width fluctuations in a narrow quantum well. We observe a redshift of the ground state and several excited states, when a lateral field is applied. This Stark shift is reduced when the dot is populated with several excitons that screen the electric field. The lines broaden increasingly at high fields, which is attributed to field-induced lateral tunneling out of the dot. [S0163-1829(98)07908-9]

Space-resolved photoluminescence (PL) spectroscopy is an interesting tool for the investigation of intrinsic properties of semiconductor nanostructures. Inhomogeneous broadening, which is a consequence of sample inhomogeneities, is avoided and effects that are normally obscured by inhomogeneously broadened lines become observable. Quantum dots, formed by monolayer width fluctuations in narrow quantum wells, have attracted a lot of interest in recent years.[1–4] The narrow lines found in these systems open the possibility to investigate even very small changes of the peak positions in external fields. Recently, we have observed the Zeeman spin splitting and diamagnetic shift of the ground state and two excited states of such a quantum dot in a magnetic field and found strong differences between the g factors and shifts of the three states.[7] Brown *et al.*[5] found that the spin splitting slightly changes when the polarization of the exciting laser beam is changed from σ^- to σ^+ and attributed this to a hyperfine interaction.

In this paper we study the optical properties of such single quantum dots in an external electric field. The sample is a 35-Å-wide GaAs/Ga$_{0.65}$Al$_{0.35}$As quantum well, where monolayer width fluctuations give rise to a localization of excitons. We have processed interdigital gate electrodes with 10-μm gold stripes and 10-μm spacings on top of the sample, which are used to induce an electric field in the plane of the quantum well, located 160 nm below the sample surface. The beam of a titanium sapphire laser is focused by a microscope objective through a thin window into the cryostat ($T \approx 5$ K). The spot size at the sample surface is about 1.5 μm. The luminescence signal is collected by the same objective, passes through a pinhole, placed in an image plane of two lenses, which defines the spatial resolution of the detection and is then dispersed by a triple grating spectrometer. An *xyz* piezo table is used to move the microscope objective and enables us to search for positions where luminescence from one single quantum dot arises. For symmetry reasons we try to find dots located in the center between two gate fingers.

In Fig. 1(a) we show the PL and PLE spectra of such a quantum dot. The excitation power is only 2 μW, which according to the estimation presented in Ref. 6 means that the dot is occupied by less than one exciton on the time average. The line at about 1657 meV corresponds to the localized exciton. The sharp peak at 1662 meV in the PLE spectrum is an excited state of this localized exciton. This is supported by experiments in a magnetic field[7] where strongly different exciton g factors and diamagnetic shifts were found for ground and excited states. At 1670 meV the quantum-well luminescence is observed. When a lateral field is applied and the sample is excited above the quantum-well energy the intensity of this line drops because the electron and hole become separated before they condense into an exciton. The drop in intensity allows one to roughly estimate the strength of the lateral electric field. It means that the field is already in a range where the drop in energy across one exciton diameter is of the order of the exciton binding energy $E \approx 10^4$ V/cm. We experimentally find a strong decrease of the PL intensity at voltages of about 10 V. Numerical calcu-

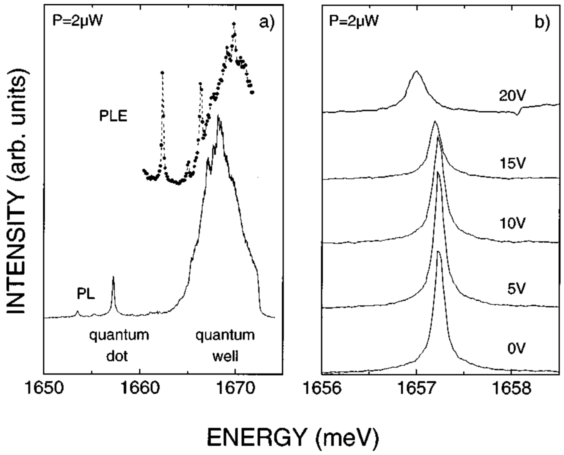

FIG. 1. (a) PL and PLE spectra of a single quantum dot. (b) PL spectra of the ground state at different electric fields.

lations for our sample geometry yield an electric field of about 0.6×10^4 V/cm in the center between two gate fingers, at this external voltage, which is consistent with the above estimate.

Regarding the narrow luminescence line that is originating from excitons localized in a quantum dot we observe a shift to lower energy with increasing electric field. This is shown in Fig. 1(b). The dot is pumped resonantly into the excited state to avoid the above-mentioned field ionization of the exciton, which takes place when excitation is done in the continuum. The shift is mainly quadratic in E, at an exteral voltage of 25 V we find a shift of about 0.5 meV. This shift is explained by the Stark effect. In the electric field, valence and conduction bands become tilted and the energetic difference between the electron and the hole state is reduced, accompanied by a spatial separation of electron and hole. This separation is limited by the lateral potential barrier. In the picture of independent electron-hole pairs, the recombination at this reduced energy is allowed because the wave functions of electron and hole have evanescent tails in the forbidden gap with some overlap. The transition, however, becomes more and more spatially indirect. This leads to a reduction of the oscillator strength and the luminescence intensity drops with increasing field. In addition excitonic effects have to be considered in the quantum dot case. The electric field changes the exciton binding energy and thus influences the transition energy. This effect may even cause a blueshift of the PL line as was recently shown by Arakawa *et al.*[8] in a study on quantum wires. Detailed theoretical calculations on the Stark effect in quantum dots are found in Refs. 9 and 10.

In Fig. 2 the peak positions of the ground state and three excited states of a different dot are shown as a function of the electric field. The ground state of this dot is shifted by about 0.4 meV at an external voltage of 23 V. The solid line is a fit curve where a quadratic dependence was assumed. This dependence is expected as a first approximation in the regime where the exciton becomes increasingly polarized due to the separation of electron and hole. For the excited states we also observe a quadratic shift to lower energies. The intensity of these lines decreases with increasing field due to ionization effects. Therefore we cannot trace the excited states up to the same fields as the ground state. Within the observable range we find that the Stark shift is stronger for the excited states and increases with the energy of the corresponding state. This tendency is expected if a parabolic potential is assumed for the quantum dot. In this case higher states are more extended than the ground state and the electron hole pair can be separated more easily. This may lead to a stronger shift. It has been reported in papers[11,12] on the quantum confined Stark effect in quantum wells that shifts up to several zero-field exciton binding energies have been observed. We do not observe such strong shifts in our case because the height of the lateral barrier is only of the order of 15 meV compared to a barrier of several hundred meV, when the field is applied along the growth direction of a quantum well. In our sample tunneling out of the dot becomes important, which is discussed in the following.

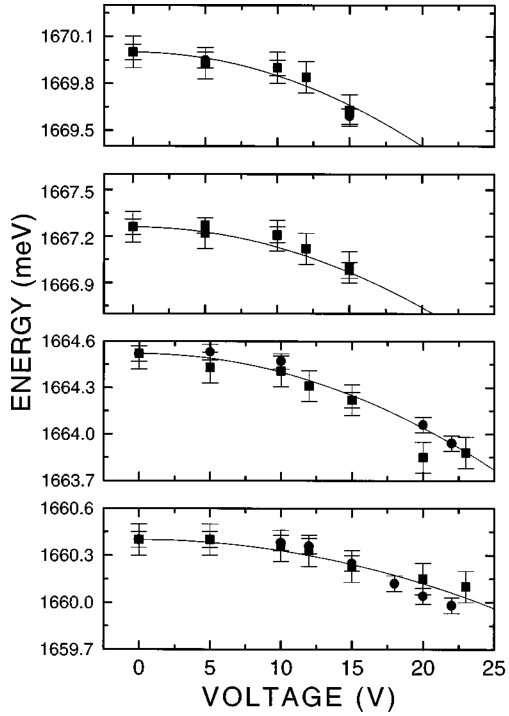

FIG. 2. Voltage dependence of the peak positions of the ground state and three excited states for a different quantum dot. The solid lines are fit curves based on a quadratic dependence. Each plot shows two sets of data measured with different integration times. The error bars indicate the precision to which the peak positions could be determined from the data ($P=2$ μW).

Figure 3 shows the dependence of the linewidth of the ground state as a function of the applied external voltage at fixed excitation density. The experiment is done on the dot shown in Fig. 1. The dot is pumped resonantly into the first excited state. We find that the linewidth increases from 0.14 meV at zero field to about 0.35 meV at an external voltage of 25 V, which corresponds to an electric field E of about 1.5×10^4 V/cm. The instrumental linewidth is about 70 μeV. This increase of the linewidth is attributed to tunneling of carriers out of the dot. The tunneling process leads to a reduced lifetime of the exciton in the dot and thus leads to an increase of the linewidth Γ. In a semiclassical model we have estimated the broadening of the line. We consider a particle in a rectangular potential with a lateral extension a and depth V_0. In an electric field this potential becomes tilted as shown in the inset of Fig. 3. Within the WKB approximation, the

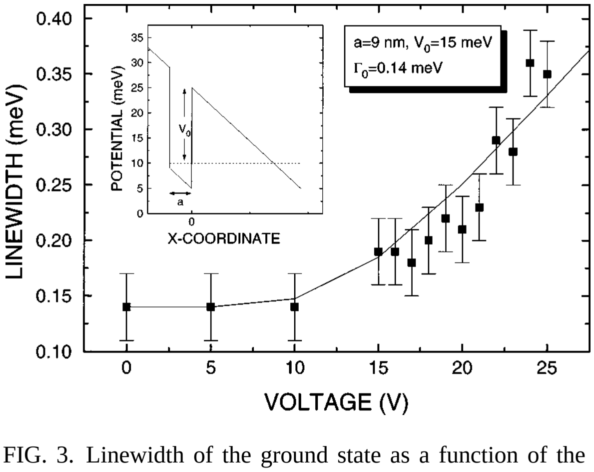

FIG. 3. Linewidth of the ground state as a function of the applied external voltage. The solid line is a fit curve calculated in the model described in the text ($P=2$ μW).

decay probability (τ^{-1}) is calculated as the frequency of collisions with the wall at $x=0$ multiplied with the transmission coefficient through the barrier V_0-eEx. The frequency of wall collisions is connected with the velocity v of the particle, which is estimated via the uncertainty principle as $v \geqslant \hbar/(2ma)$. From the decay time τ we obtain the contribution of this tunneling process to the linewidth by the uncertainty relation $\Gamma \times \tau \geqslant \hbar/2$. This leads to the following expression:

$$\Gamma \geqslant \frac{\hbar^2}{8ma^2}\exp\left[-\frac{4}{3}\frac{\sqrt{2m}}{eE\hbar}V_0^{3/2}\right]+\Gamma_0.$$

Γ_0 is the linewidth at zero electric field, m is the electron effective mass in GaAs since the contribution of the electron dominates the contribution of the hole due to its lower mass. In this case V_0 includes the lateral confinement and the interaction of the electron with the remaining hole. We have tried to fit our data with this model and find the best agreement (see Fig. 3) with the parameters $V_0 = 15$ meV and $a = 90$ Å. These values are in a reasonable range, V_0 and a being of the order of the lateral barrier height and the exciton Bohr radius, respectively. We therefore believe that lateral field-induced tunneling out of the dot leads to the observed broadening of the ground state. For the excited states strong changes of the linewidth are also expected. However, especially at high fields where the broadening should be seen, the PLE signal is too small for a precise determination of the linewidth.

We now want to discuss the positions of the peaks when the excitation power is increased at a fixed external voltage of 14 V. In Fig. 4 we show spectra where the excitation intensity is increased by about two orders of magnitude. The left side shows the PL spectra of the ground state. The dot is pumped resonantly into the first excited state at 1664.3 meV. On the right, PLE spectra for the excited states are depicted. When the excitation power is raised from 2 to 185 μW, the ground state shifts by about 0.15 meV to higher energies, which is approximately the position at zero electric field (dashed line in Fig. 4). This means that the field is screened when the dot is occupied by several excitons. For the excited states we also find a blueshift, which is, however, different for each state. While the first excited state almost reaches its zero-field position the shift for the higher states is much weaker. This is explained by the fact that the PLE intensity of the higher states, which is a measure of the population of the dot, is weaker. Therefore the number of excitons in the dot on the time average is smaller, which leads to a reduced screening and a smaller shift. This variation shows that the screening is due to carriers within the dot and is not caused by carriers in the barriers around the dot, because then the absorption should increase with the energy and therefore lead to a stronger shift for the excited states.

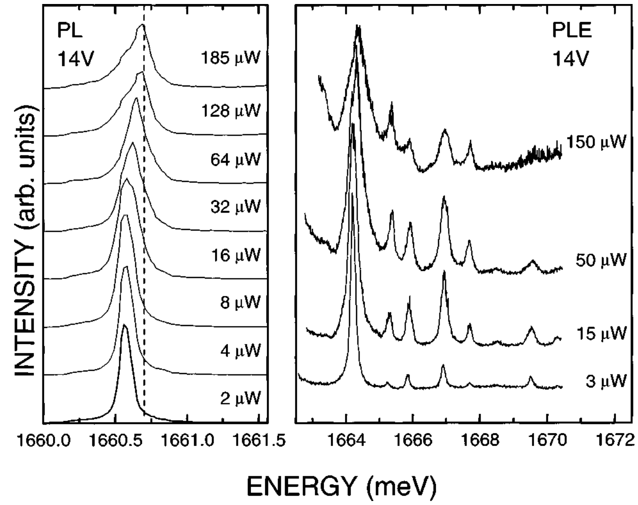

FIG. 4. Power dependence of the peak positions at a fixed external voltage of 14 V. The dashed line indicates the peak position at zero electric field.

At high excitation densities the luminescence line of the ground state broadens and becomes asymmetric. A reason for this may be the statistical variation of the number of excitons in the dot during the integration time of the spectrum because the position of the line depends on the number of excitons in the dot. Therefore periods where the occupation of the dot is weak contribute to the luminescence on the low-energy side of the peak leading to the observed asymmetry. If no electric field is applied and the pumping power is increased the spectrum essentially remains unchanged. The line is only weakly broadened from 70 to 90 μeV, but we do not find a noticeable shift of the line within the experimental resolution.

In conclusion we have presented a study of single quantum dots in an external electric field E. We investigate the ground state and several excited states and find a shift to lower energy that is quadratic in E. The higher states reveal a stronger shift than the ground state. At high-power excitation the dot is populated by several excitons. The lines then shift towards their zero field energies because the field becomes increasingly screened. At high electric fields the linewidth of the ground state increases, which indicates field-induced lateral tunneling out of the dot.

We thank G. Böhm and G. Weimann for providing the samples and acknowledge the financial support by the Deutsche Forschungsgemeinschaft (SFB 348).

---

[1] K. Brunner, G. Abstreiter, G. Böhm, G. Tränkle, and G. Weimann, Appl. Phys. Lett. **64**, 3320 (1994); Phys. Rev. Lett. **73**, 1138 (1994).

[2] A. Zrenner, L. V. Butov, M. Hagn, G. Abstreiter, G. Böhm, and G. Weimann, Phys. Rev. Lett. **72**, 3382 (1994).

[3] H. F. Hess, E. Betzig, T. D. Harris, L. N. Pfeiffer, and K. W. West, Science **264**, 1740 (1994).

[4] D. Gammon, E. S. Snow, B. V. Shanabrook, D. S. Katzer, and D. Park, Phys. Rev. Lett. **76**, 3005 (1996); Science **273**, 87 (1996).

[5] S. W. Brown, T. A. Kennedy, D. Gammon, and E. S. Snow, Phys. Rev. B **54**, 17 339 (1996).

[6] U. Bockelmann, W. Heller, A. Filoramo, and Ph. Roussignol, Phys. Rev. B **55**, 4456 (1997).

[7] W. Heller and U. Bockelmann, Phys. Rev. B **55**, 4871 (1997).

[8] T. Arakawa, Y. Kato, F. Sogawa, and Y. Arakawa, Appl. Phys. Lett. **70**, 646 (1997).

[9] G. W. Wen, J. Y. Lin, H. X. Jiang, and Z. Chen, Phys. Rev. B **52**, 5913 (1995).

[10] S. Jaziri, G. Bastard, and R. Bennaceur, J. Phys. IV **3**, 367 (1993).

[11] D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Phys. Rev. B **32**, 1043 (1985).

[12] H.-J. Polland, L. Schultheis, J. Kuhl, E. O. Göbel, and C. W. Tu, Phys. Rev. Lett. **55**, 2610 (1985).