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Electron localization by self-assembled GaSb/GaAs quantum dots

M. Hayne,[a] J. Maes, S. Bersier, and V. V. Moshchalkov
Laboratorium voor Vaste-Stoffysica en Magnetisme, Katholieke Universiteit Leuven, Celestijnenlaan 200D, B-3001 Leuven, Belgium

A. Schliwa, L. Müller-Kirsch, C. Kapteyn, R. Heitz, and D. Bimberg
Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstrasse 36, 10623 Berlin, Germany

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We have studied the photoluminescence from type-II GaSb/GaAs self-assembled quantum dots in magnetic fields up to 50 T. Our results show that at low laser power, electrons are more weakly bound to the dots than to the wetting layer, but that at high laser power, the situation is reversed. We attribute this effect to an enhanced Coulomb interaction between a single electron and dots that are multiply charged with holes. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1583853]

Self-assembled quantum dots[1] (QDs) are well known for their suitability for the study of fundamental phenomena such as confinement effects, and for their strong potential for device applications. Type-II QDs, in which either the electron or the hole is confined in the dot, are expected to reveal novel phenomena with respect to type-I QDs, and may prove to be useful in carrier storage devices. In the GaSb/GaAs system, it is known that the holes are strongly confined to the dots by the valence-band (VB) offset (the hole activation energy is about 450 meV),[2] but relatively little is known about the electrons in these structures. In optical experiments, such as those studying the blueshift of the photoluminescence (PL) with increasing laser excitation,[3,4] it is quite reasonably assumed that since the electron and hole are spatially separated the exciton binding energy is low, and that intrinsic electron localization is negligible. In this letter, we report magnetophotoluminescence measurements of self-assembled GaSb QDs in a GaAs matrix. Our data show that at low incident laser power, the QD excitons are more weakly bound than the wetting layer (WL) excitons. However, at high laser power, the QD binding energy increases such that it becomes larger than the WL exciton binding energy. We attribute this effect to the increased Coulomb potential of dots that are multiply charged with holes as a result of optical pumping.[3–5]

The samples were grown by low-pressure metalorganic chemical vapor deposition on semi-insulating GaAs (001) substrates. The growth procedure is similar to that described in Ref. 6. After the growth of a 300-nm GaAs/$Al_{0.7}Ga_{0.3}As$/GaAs buffer layer, about 3 monolayers of GaSb were deposited, leading to the formation of self-assembled QDs. An interruption of 2 s preceded the growth of 30 nm of GaAs, 20 nm of $Al_{0.7}Ga_{0.3}As$, and then 10 nm of GaAs to cap the structure. PL experiments were performed at 4.2 K in a He bath cryostat placed in the bore of a pulsed-field magnet. The field was applied perpendicular to the plane of the sample, which was excited with the 532-nm line of a solid-state laser or the 514.5-nm line of an Ar^+ laser via a 200-μm-core optical fiber. The incident laser power density P was varied between 0.01 and 100 $W\,cm^{-2}$.[7] Six other fibers were used to collect the PL and transmit it to a 0.25-m spectrometer with a 300-grooves/mm grating. Photon integration was performed by an InGaAs diode array with count times varying between 0.5 and 3.8 ms in field, and up to 3 s at zero field. The field resolution for data taken during the 25-ms field pulse was at worst $\pm 5\%$, and typically much better.

Figure 1 shows a typical zero-field spectrum for the GaSb/GaAs QD sample taken at $P = 30\ W\,cm^{-2}$. It is dominated by the WL peak at about 1.38 eV. A small group of peaks centered around 1.5 eV arise from the GaAs matrix, while the QD PL is seen as a low-intensity line at 1.15 eV with a width of about 80 meV. The inset to Fig. 1 shows the dependence of the WL luminescence peak energy on magnetic field at the same laser power density, which we now discuss. It should be noted that we have verified that the field dependence of the WL PL is the same at low power ($0.5\ W\,cm^{-2}$) as it is at high laser power ($30\ W\,cm^{-2}$). The data show behavior that is typical of an exciton, with a parabolic shift at low field, and a linear field-dependence at high

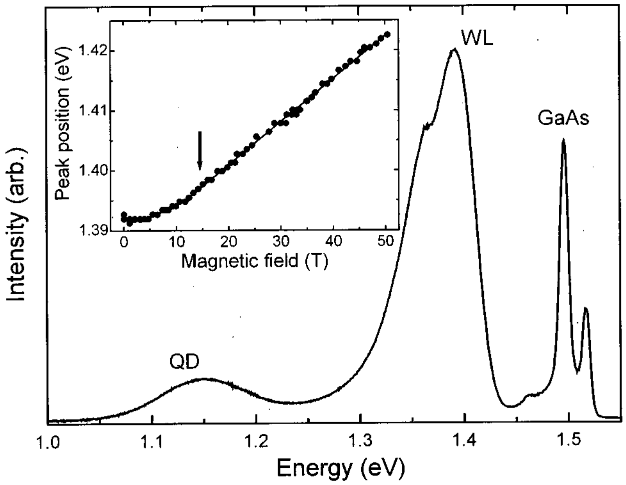

FIG. 1. Low-temperature PL spectrum at $P = 30\ W\,cm^{-2}$. The shoulder on the low-energy side of the WL peak is an artifact due to a step-like increase in the quantum efficiency of the detector from high to low energy. The inset shows the field dependence of the peak of the WL PL. The solid line is a fit to the data, while the arrow indicates the transition between high- and low-field limits.

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[a] Electronic mail: manus.hayne@fys.kuleuven.ac.be

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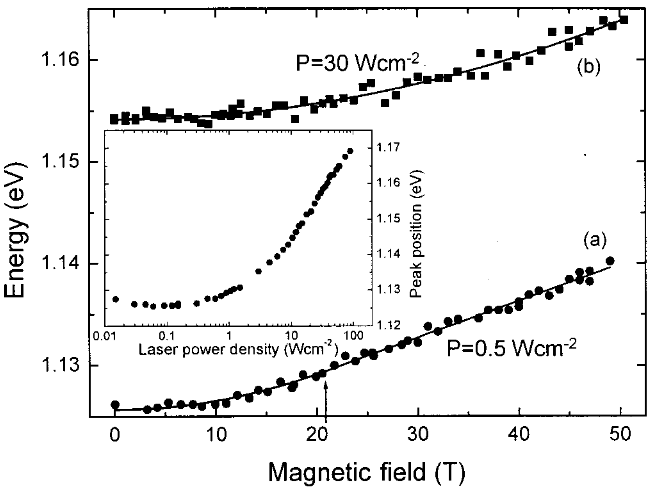

FIG. 2. Peak position of the QD PL as determined by Gaussian fits as a function of magnetic field for incident laser powers of (a) 0.5 W cm^{-2}, and (b) 30 W cm^{-2}. The solid lines are fits to the data. The arrow in (a) indicates the transition between high- and low-field limits. The inset shows the dependence of the PL peak energy on the incident laser power density.

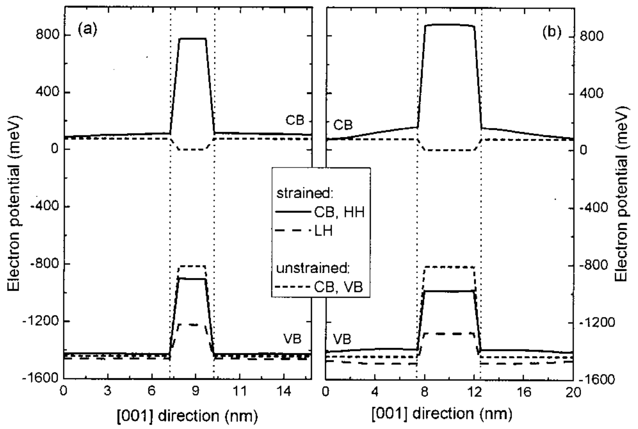

FIG. 3. Calculated structure of the minimum of the CB and maximum of the VB for GaSb/GaAs QDs in the growth direction through the center of (a) a 2.2-nm-high "thin" dot and (b) a 4.4-nm-high "thick" dot. Both dots are truncated pyramids with base widths of 25 and 22 nm, respectively. The vertical dashed lines show the upper and lower interfaces of the dot. Note that heavy-hole (HH) light-hole (LH) degeneracy is lifted by the strain.

field. The field at which the transition between the two regimes occurs is reached when the magnetic length is $1/\sqrt{2}$ times the exciton Bohr radius a_B, while the high-field slope gives the reduced exciton mass μ. Fitting the data[8] reveals that these parameters are 9.5 nm and $0.081m_0$, respectively, where m_0 is the free-electron mass. In both GaSb and GaAs, the electron effective mass, m_e, is about 10 times smaller than the hole effective mass; therefore, in either material the electron will, to a good approximation, determine the exciton effective mass and effective Bohr radius. Since GaSb has a large dielectric constant ε (15.7) and small m_e ($0.039m_0$),[9] the donor effective Bohr radius is relatively large at 21 nm, and is twice that of GaAs. The Bohr radius we measure is thus comparable to that of an electron in GaAs, and the mass is also closer to that of an electron in GaAs than in GaSb (m_e in GaAs is $0.067m_0$).[9] In the hydrogen model of the exciton, the Bohr radius $a_B = a_B^0 \varepsilon/(\mu/m_0)$, where $a_B^0 = 0.529$ Å is the Bohr radius of the hydrogen atom. Substituting in our experimental values for a_B and μ, we find that $\varepsilon = 14.5$, which is exactly the value given by the average of ε for GaSb and GaAs, and therefore indicates that such a model is reasonable.

We now turn to the QDs. Figure 2(a) shows the QD PL peak position as a function of magnetic field at a much lower P of 0.5 W cm^{-2}. Using a QD density of 3×10^{10} cm^{-2} (Ref. 6), a ground-state recombination time of 27 ns (Ref. 5), and assuming that all the photoexcited holes are captured by the dots, we find that the average hole occupancy of the dots at this laser power is ~1. The magneto-PL data show a similar form to that of the inset of Fig. 1, but the size of the diamagnetic shift between zero and 50 T is reduced by a factor of 2 (14 meV instead of 31 meV). Fitting the curve yields $a_B = 8.0$ nm and $\mu = 0.16m_0$. The size of the diamagnetic shift is inversely proportional to μ, so the strong decrease in the size of the shift for these data compared to those of the WL is due to the increase in μ, which is itself almost certainly a result of a strain-induced enhancement of the electron mass due to the deformation of the GaAs lattice by the GaSb QDs.[10] A larger μ should also reduce the Bohr radius. Specifically, in the hydrogen model of the exciton described earlier, doubling μ should halve a_B, but the reduction we see is considerably weaker. Thus, even though a_B for the dot exciton is smaller than for the WL exciton, the former is actually *more* extended than expected from the exciton mass, indicating a strong reduction in the Coulomb interaction due to the separation of the electron and the hole. We can parameterize this in the approximation of the hydrogenic binding energy, $E_B = \hbar/2\mu a_B^2$, which shows a reduction from 5.2 meV for the WL exciton to 3.7 meV for the QD exciton.

In order to understand how this additional electron–hole separation arises, we have calculated the form of the conduction-band (CB) minimum and the VB maximum in the growth direction for the GaSb/GaAs system using eight-band **k.p** calculations that include the strain distribution.[10] Figure 3(a) shows the result for a thin dot, which, in the limiting case, can be considered to describe the WL. Electrons will be expelled from the GaSb by the large strain-induced maximum in the CB, but can be bound either above or below the WL by the Coulomb interaction with a hole that is confined within the WL, forming an indirect exciton. For a thick dot [Fig. 3(b)] the CB offset also excludes the electrons from the GaSb, but the strain in the GaAs matrix surrounding the dot varies the local band gap, warping the CB and *further repelling the electrons*. The net effect is that the exciton binding energy for the QD should be lower than for the WL, as we have observed.

We now go on to discuss the magnetic field dependence of the QD PL at $P = 30$ W cm^{-2} [Fig. 2(b)], the *same laser power* as was used for the investigation of the WL PL. Using the same assumptions as at low power, but a recombination time of 5 ns to account for the charging of the dots at high power,[5] we estimate the hole occupancy to be 13. It is immediately observed that at zero-field, the QD PL peak position in Fig. 2(b) is 28 meV higher in energy than for Fig. 2(a); a result of the widely reported laser-induced PL blueshift (inset of Fig. 2) in type-II dots.[3,4,11,12] At this laser power, the diamagnetic shift between zero and 50 T is reduced to 10 meV. Indeed, the lateral confinement is now so strong that the data can be described by a parabolic field dependence up to the highest fields. Fitting the data with a parabolic and linear field dependence gives a curve indistinguishable from the entirely parabolic one, with a transition to the high-field limit at 47 T, and a μ and a_B of $0.16m_0$ and 5.3 nm, respectively. It is worth noting that since the slope of the data can only increase (or stay the same) at higher fields, these values are both upper limits. Indeed, given that any decrease in these parameters is restricted by the fact that the ratio a_B^2/μ should remain constant, and given that μ is the same as at lower power, it is reasonable to assume that these values are close to the correct ones. Doing so, we find a binding energy of 8.5 meV, which is more than twice as large than it was at low power, and 60% larger than the WL binding energy. Since a_B and μ are upper limits, this is a *lower* limit.

We believe that the enhanced binding energy is the result of the dots becoming multiply charged with holes as the laser power is increased. This strengthens the Coulomb interaction between each dot and a single electron, making the latter strongly bound. In this model, the dot is analogous to the nucleus of an atom, with the electron responsible for the PL playing the role of a strongly bound ''inner'' electron. Such a mechanism is excluded for the WL, where the holes are free to drift apart. This effect is closely related to the laser-induced blueshift of the QD PL shown in the inset of Fig. 2, and our explanation is entirely consistent with recent work on the subject. Müller-Kirsch *et al.*[3] proposed that the laser-induced blueshift is the combined result of Coulomb charging and state filling of the dots, rather than a deformation of the confining potential due to charge separation, as was previously suggested.[4] This conclusion was subsequently supported by micro-PL measurements on type-II InP/GaAs self-assembled QDs, which showed that the blueshift was the result of the occupation of higher energy levels.[12] Recent time-resolved PL measurements on a sample similar to that studied here showed that the PL decay after intense excitation has a fast component, corresponding to the recombination of multi-exciton states, and a slow component, due to recombination of the ground state.[5] From deep-level transient spectroscopy and capacitance measurements, we know that the dots may be charged with up to 15 holes,[2] while, as mentioned earlier, we estimate a hole occupancy of 13 at $30\,\mathrm{W\,cm^{-2}}$. A more detailed analysis with a rate equation model will be presented elsewhere.[13]

In summary, we have measured the diamagnetic shift of the photoluminescence from self-assembled GaSb/GaAs quantum dots in magnetic fields up to 50 T. At low laser power, we find that the QD excitons are more weakly bound than wetting-layer excitons. We attribute this to the spatial separation of electrons and holes by strain in the GaAs surrounding the dots. At high laser power, the dots become multiply charged with holes, and the QD binding energy exceeds that of the WL excitons.

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