AIP | Journal of Applied Physics

Anomalous optical magnetic shift of self-assembled GaSb/GaAs quantum dots

Ta-Chun Lin, Liang-Chen Li, Sheng-Di Lin, Yuen-Wuu Suen, and Chien-Ping Lee

Citation: Journal of Applied Physics **110**, 013522 (2011); doi: 10.1063/1.3607973
View online: http://dx.doi.org/10.1063/1.3607973
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/110/1?ver=pdfcov
Published by the AIP Publishing

JOURNAL OF APPLIED PHYSICS 110, 013522 (2011)

Anomalous optical magnetic shift of self-assembled GaSb/GaAs quantum dots

Ta-Chun Lin,[1,a)] Liang-Chen Li,[2] Sheng-Di Lin,[1] Yuen-Wuu Suen,[3] and Chien-Ping Lee[1,2]
[1]Department of Electronics Engineering and Institute of Electronics, National Chiao Tung University, Hsinchu, Taiwan
[2]Center for Nano Science and Technology, National Chiao Tung University, Hsinchu, Taiwan
[3]National Nano Device Laboratories, Hsinchu, Taiwan and Institute of Nanoscience and Department of Physics, National Chung Hsing University, Taichung, Taiwan

(Received 17 April 2011; accepted 31 May 2011; published online 11 July 2011)

We report the magneto-photoluminescence (PL) measurement results on type-II self-assembled GaSb/GaAs quantum dots with the magnetic field applied in Faraday and Voigt configurations. The emission of the quantum dots exhibited a typical diamagnetic blueshift when the magnetic field was applied in a Faraday configuration. However, when the magnetic field was in the Voigt configuration, an unusual redshift in the emission peak accompanied with a rapid increase of the PL intensity was observed. Guided by numerical calculations, the magnetic field applied in the Voigt configuration is found to provide an additional vertical confinement to electrons, and therefore, substantially enhance the radiative electron-hole recombination. The resulting decrease of the steady-state hole concentration gives rise to the observed anomalous magnetic redshift.
 [doi:10.1063/1.3607973]

I. INTRODUCTION

Type-II self-assembled quantum dots (QDs), where the staggered band lineup spatially separates the electrons and holes, have attracted considerable interests for the exhibition of novel physical phenomena, such as optical Aharonov-Bohm effect in magnetic fields,[1–3] and the potential application for carrier storage devices.[4] In the self-assembled GaSb QDs embedded in GaAs matrix, holes are strongly localized inside the QDs with a large activation energy of about 450 meV,[4] and the electrons are weakly bound in GaAs regions next to the GaSb QDs by the Coulomb attraction. The magneto-photoluminescence (PL) measurements for this material system have been carried out in the past to investigate the extent of the electron wave function. However, it was usually done in a Faraday configuration, where only the lateral electron-hole (e-h) spatial separation was revealed.[5–8] Because of the shape of the QDs, where the height is much smaller than the width, electrons are mostly confined in GaAs regions above and below the QDs.[3] Therefore, the optical properties of the excitons in these QDs have stronger dependence on the vertical e-h separation. This separation, however, can only be studied with the magnetic field applied in the Voigt configuration.

In this work, we compared the magneto-PL results of type-II GaSb/GaAs QDs with the magnetic field applied in both the horizontal and the vertical directions. When the magnetic field was applied in a Faraday configuration, the QD emission revealed a typical diamagnetic blueshift along with a slightly decreased PL intensity. However, when the magnetic field was in a Voigt configuration, an unusual redshift in the emission peak accompanied with a rapidly increased intensity was observed. By means of numerical model calculations, this anomalous optical magnetic response is attributed to the reduction of the vertical e-h separation and the resulting increase of the radiative e-h recombination rate in the magnetic fields applied in the Voigt configuration.

II. EXPERIMENT

The GaSb QD sample (Rn283) was fabricated on a GaAs (001) substrate by a Veeco Gen-II molecular beam epitaxy (MBE) system equipped with a valve cracker of Sb_2 source. Nominal 3 monolayers (MLs) of GaSb were deposited at 500 °C to form self-assembled QDs. Further details of sample growth have been described in Ref. 9. The surface topography of the uncapped QDs was measured by an atomic force microscope (AFM) and is shown in Fig. 1(a). The areal density of QDs was estimated to be about 7×10^9 cm^{-2} with the average dot height of about 12 nm ($\pm$2 nm) and the base width of about 55 nm ($\pm$8 nm). However, the realistic dimensions of the embedded QDs are expected to be much smaller due to the overestimation of AFM.[10] Conventional PL and magneto-PL measurements were performed at 1.4 K with a 14 T superconducting magnet. The sample was excited by Nd:yttrium aluminum garnet (YAG) laser at 532 nm via an optical fiber. The PL signal was collected by a fiber bundle, dispersed by a 320 mm monochromator and detected by an InGaAs photodiode.

III. PHOTOLUMINESCENCE

A. Zero-field photoluminescence

Figure 1(b) presents the zero-field PL spectra for the GaSb/GaAs QD sample taken at varied pumping powers from 10 mWcm^{-2} to 200 mWcm^{-2}. The observed two prominent emission peaks are originated from the WL with a peak energy around 1.28 eV and from the QD ensemble with

a)Electronic mail: dajin6@yahoo.com.tw.

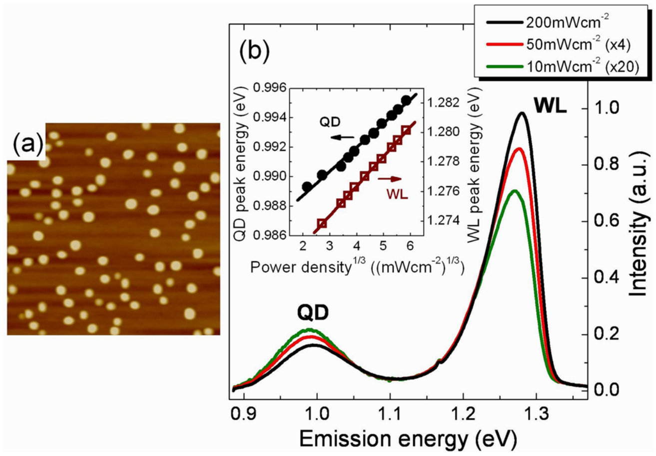

FIG. 1. (Color online) (a) AFM surface image of GaSb QD sample in 1 μm square image area. (b) The zero-field PL spectra taken at varied excitation powers from 10 mWcm^{-2} to 200 mWcm^{-2}. The inset depicts the QD and the WL emission peak energies as a function of 1/3 power of the excitation power density.

a peak around 1.0 eV. When the excitation power increases, both the emission peaks reveal energy blue shifts in proportion to the 1/3 power of excitation power density as seen in the inset. This blueshift is a signature of type-II structures and is attributed to the capacitive Coulomb charging and the excited state filling effects by the photoexcited holes.[11,12]

B. Magnetophotoluminescence in a Faraday configuration

The PL spectra have been measured with the magnetic field applied along the growth direction (in a Faraday configuration). Figure 2 displays the emission peak energy shifts of the QDs and the WL as a function of the magnetic field. At low fields, both QD and WL emissions reveal a typical diamagnetic blueshift, which has a quadratic dependence on the magnetic field. The diamagnetic coefficients of the WL emission measured at different excitation powers are almost the same and are about two times those of the QD emission. The much smaller diamagnetic coefficients of the QDs are associated to the increase of the effective mass of the electrons near the QDs due to the strained GaAs lattice by the GaSb QDs.[5,13] At higher excitation power densities, the GaSb QDs show a somewhat larger diamagnetic shift. Similar results were also observed in Ref. 7. Besides, as shown in the inset, the integrated PL intensity of the WL increases with the magnetic field due to the additional magnetic confinement to the electrons and the holes. However, the intensity of the QD emission decreases with the field. The cause could be that the magnetic confinement also suppresses the hole relaxation from the WL to the QDs, and hence the carrier concentration in the QDs decreases with the magnetic field.

C. Magnetophotoluminescence in a Voigt configuration

The inset in Fig. 3 presents the PL spectra of the GaSb QDs measured at a laser pumping power density of 20 mWcm^{-2} in various magnetic fields applied along the in-plane direction (Voigt configuration). An unusual PL redshift accompanied with a rapidly increased intensity is clearly seen. The peak energy shifts of the QDs and the WL measured at different pumping powers are plotted against the magnetic field in Fig. 3. Very different behaviors were observed between the energy shifts from the QDs and the WL. The QD luminescence exhibits a rapid redshift at low magnetic fields followed by a slow blueshift. The slow blueshift indicates that the diamagnetic increase of the PL energy overcomes this redshift effect at high magnetic fields. On the other hand, the observed redshift of the WL is much smaller. This is due to a larger diamagnetic effect caused by a smaller exciton effective mass in the WL. This redshift effect gets stronger at lower pumping powers for both the QD and WL emissions. At the lowest power density of 10 mWcm^{-2}, the maximum of the energy redshift of the QD emission is found to be about -6.5 meV at a magnetic field of 7 T. We marked this value with an arrow in Fig. 3 to be the upper limit of this redshift effect. The anomalous effect seems to have an onset point at low magnetic fields. Figure 4(a) shows the low field peak energy at different pumping powers. The onset of the redshift appears to be at 1 T and is independent of the pumping power.

Figure 4(b) depicts the integrated intensity normalized to that at a zero field of the QD and the WL emissions. The intensities of both emissions increase rapidly at low magnetic fields and slow down at high fields. The intensity increase of the WL PL is larger than that of the QD PL, probably due to the same reason as that in the Faraday configuration, i.e., the suppression of the hole relaxation from the WL to the QDs by the magnetic field. Similar to the PL redshift effect, this intensity enhancement is stronger at lower pumping powers for both the QD and WL PL. For QDs, the onset point and the upper limit of the rapid intensity increase are

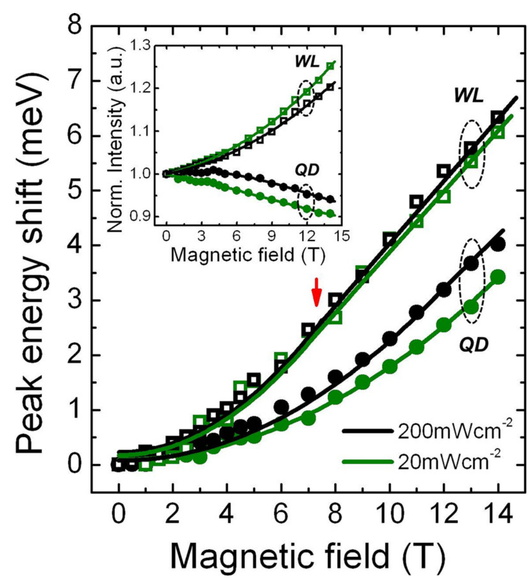

FIG. 2. (Color online) The peak energy shifts of the GaSb QDs and the WL as a function of the magnetic field at different excitation powers in a Faraday configuration. The arrow indicates the low field region where the energy shift has a quadratic dependence on the magnetic field. The inset reveals the integrated PL intensity normalized to that at a zero field of the GaSb QDs and the WL vs the magnetic field.

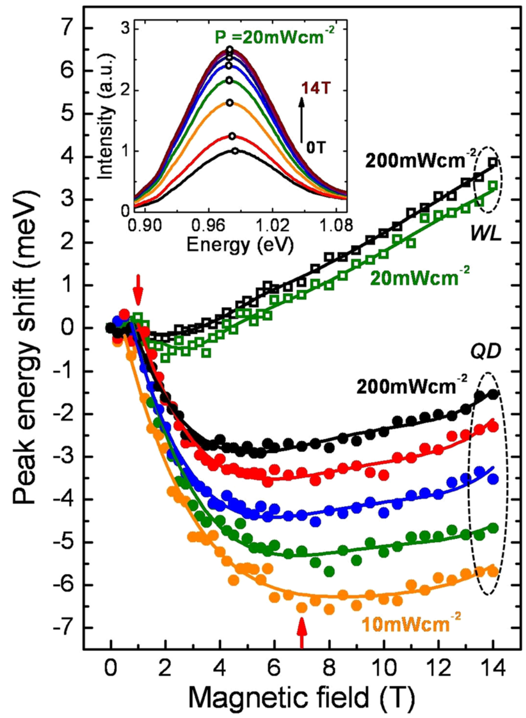

FIG. 3. (Color online) The PL peak energy shifts of the QDs and the WL as a function of the magnetic field at different excitation powers in a Voigt configuration. The excitation power densities are 10, 20, 40, 100, and 200 mWcm^{-2}. The arrows indicate the onset point and the upper limit of the energy redshift effect. The inset shows the PL spectra of the GaSb QDs in magnetic fields.

estimated to be 1 T and 7 T, respectively, and marked with arrows in Fig. 4(b). Obviously, there is a correlation between the anomalous energy redshift and the intensity enhancement in the presence of a magnetic field applied in the Voigt configuration.

IV. SIMULATION AND DISCUSSION

In order to interpret the different magneto-optical responses observed in the Faraday and Voigt configurations, a theoretical analysis considering the electron-hole Coulomb interaction in the presence of a magnetic field was carried out. A disk-shaped QD with a layer of WL in a GaAs matrix was used in the calculation. The dot width was 40 nm, the

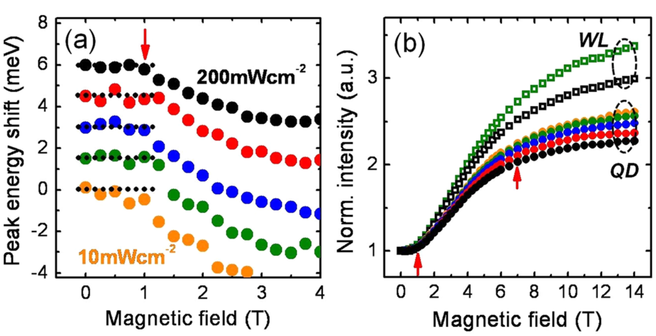

FIG. 4. (Color online) (a) The PL shifts of the QDs vs the magnetic field in the low field region. (b) The integrated PL intensity normalized to that at a zero field of the GaSb QDs and the WL vs the magnetic field. The arrows indicate the onset points and the upper limit of the energy redshift and the rapid intensity increase effects.

dot height was 8 nm, and the thickness of the WL was 1 nm. The composition of the dot was assumed as $GaAs_{0.3}Sb_{0.7}$ to account for the partial intermixing of As and Sb atoms during growth.[14] The electron effective mass in the GaAs matrix was taken as 0.12 m_0 owing to the strain-induced enhancement.[13] Other parameters used in the calculation were taken from Ref. 15. The magnetic response was calculated by superimposing a magnetic confining potential to the Hamiltonian. This confining potential energy can be written as

$$V_{B(e,h)} = \frac{e^2 R_{e,h}^2 B^2}{e m_{e,h}},$$

where R is the projection of $\overline{R}$ onto the plane perpendicular to the magnetic field.

The calculated ground-state electron and hole wave functions at a zero magnetic field are illustrated in Fig. 5(a). Holes are found to be localized inside the QD, and electrons are weakly confined in GaAs regions above and below the QD by Coulomb attraction. When the magnetic field is applied, the electron wave function is strongly altered due to the large $V_{B(e)}$ arising from the small effective mass and the large wave function extent. The magnetic force acted on the electrons, which is the derivative of $V_{B(e)}$, is marked in Fig. 5(a). It is found that the magnetic field applied in a Voigt configuration can provide a vertical magnetic confinement to the electrons and hence pushes them to the localized holes.

The vertical e-h spatial separation is calculated as $\sqrt{\langle z_e^2 \rangle}$ (since the hole is localized in the center of the dot, the origin) and is plotted versus the magnetic field in Fig. 5(b). N_h is the hole occupancy of the dot, which determines the magnitude of the Coulomb potential to electrons. At a zero field, the increase of the holes leads to the stronger Coulomb attraction and the resulting decrease of the e-h separation. As the magnetic field is applied in a Voigt configuration, the electron is pushed to the holes by the vertical magnetic confinement, and the vertical e-h separation is hence dramatically reduced. Note that the separation for different N_h is almost the same at the high field of 14 T. This is because the confinement from the magnetic potential for the electrons in high fields is much stronger than that from the Coulomb potential. However, in the Faraday configuration, the reduction of the e-h separation is much weaker.

The reduction of the e-h separation is expected to enhance the oscillator strength of the radiative e-h recombination, which is proportional to the square of the wave function overlap between electrons and holes. Figure 5(c) presents the oscillator strength normalized to that at a zero field as a function of the magnetic field. As the magnetic field is applied in a Faraday configuration, the oscillator strength is found to be slightly increased. On the other hand, as the magnetic field applied in a Voigt configuration is larger than 1 T, the oscillator strength gets considerably enhanced. Now, we turn to the explanation of this anomalous magnetic redshift. The power-dependent PL spectra indicate that the exciton energy has a strong dependence on the hole concentration. Since the magnetic field applied in the Voigt configuration enhances the radiative e-h recombination, the holes are depleted by the optical transition and the steady-

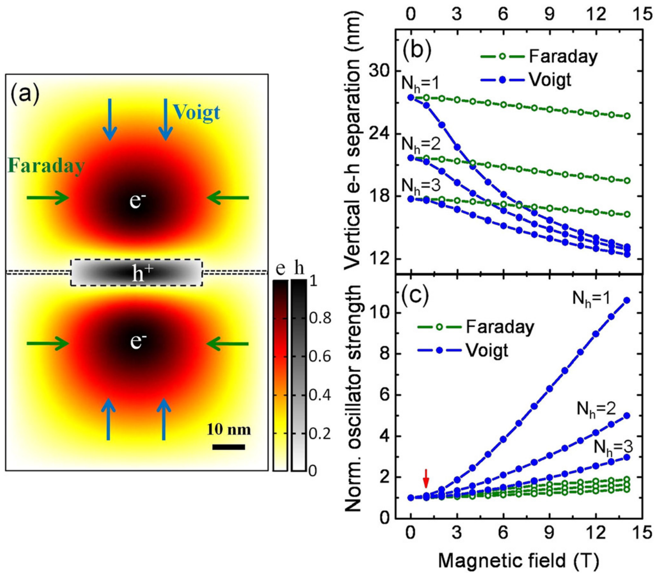

FIG. 5. (Color online) (a) The calculated electron and hole wave functions at a zero magnetic field. The QD and WL are placed in the center of the matrix to simplify the calculation. The arrows indicate the direction of the magnetic force acted on electrons. (b) The vertical e-h spatial separation vs the magnetic field for different hole occupancy in the QD. (c) The oscillator strength normalized to that at a zero field as a function of the magnetic field. The arrow represents the onset point of the oscillator strength enhancement.

state hole concentration is therefore decreased. The resulting decrease of the exciton energy of the QDs gives rise to the observed magnetic PL redshift. The calculated Coulomb charging energy for ground-state holes is about 9.3 meV (which is of the same order of the experimental data for the GaSb QDs grown by metal-organic chemical vapor deposition (MOCVD) of 13 meV (Ref. 4)). Therefore, when the average hole number per dot is decreased by about 0.7, the average exciton energy will redshift by 6.5 meV, which is the maximum of the observed redshift. Besides, the calculated oscillator strength enhancement is much stronger for smaller N_h. This is because the higher hole occupancy of the QDs strengthens the Coulomb interaction and therefore weakens the effect of the additional magnetic confinement to electrons. The simulation results agree with the experimental finding shown in the Figs. 3 and 4(b), where the energy redshift and the PL intensity enhancement get stronger at lower pumping power.

V. CONCLUSIONS

In summary, we have studied the magneto-optical responses of type-II self-assembled GaSb/GaAs QDs in both Faraday and Voigt configurations. When the magnetic field is applied in a Faraday configuration, a typical diamagnetic response is observed. However, when the field is in a Voigt configuration, the QDs exhibit an anomalous magnetic redshift (with a maximum of the redshift of −6.5 meV) together with a rapid increase of the PL intensity. Besides, both the redshift and the intensity enhancement are stronger at lower excitation power. Based on simulation results, the magnetic field in the Voigt configuration is found to provide an additional vertical confinement and push the weakly-bound electrons to the localized holes. The decrease of the e-h spatial separation leads to the considerable increase of the radiative e-h recombination rate. The resulting decrease of the steady-state hole concentration in the QDs gives rise to the decrease of the exciton energy and the observed anomalous magnetic redshift. Finally, the simulation results also indicate that a higher hole occupancy in the dots induces a stronger Coulomb interaction and therefore weakens the effect of the additional magnetic confinement, which explains the power-dependence of this anomalous redshift.

ACKNOWLEDGMENTS

We acknowledge support from the National Science Council under Contract Nos. NSC99-2221-E-009-079-MY3, NSC99-2120-M-009-009, and NSC98-2120-M-009-002; Center for Nano Science and Technology of National Chiao Tung University; National Nano Device Laboratories; and "Aim for the Top University Plan" of the National Chiao Tung University and Ministry of Education, Taiwan. We would like to thank M. C. Lo for the sample growth.

[1]E. Ribeiro, A. O. Govorov, W. Carvalho, Jr., and G. Medeiros-Ribeiro, Phys. Rev. Lett. **92**, 126402 (2004).

[2]I. R. Sellers, V. R. Whiteside, I. L. Kuskovsky, A. O. Govorov, and B. D. McCombe, Phys. Rev. Lett. **100**, 136405 (2008).

[3]K. L. Janssens, B. Partoens, and F. M. Peeters, Phys. Rev. B **66**, 075314 (2002).

[4]M. Geller, C. Kapteyn, L. Müller-Kirsch, R. Heitz, and D. Bimberg, Appl. Phys. Lett. **82**, 2706 (2003).

[5]M. Hayne, J. Maes, S. Bersier, V. V. Moshchalkov, A. Schliwa, L. Müller-Kirsch, C. Kapteyn, R. Heitz, and D. Bimberg, Appl. Phys. Lett. **82**, 4355 (2003).

[6]M. Hayne, O. Razinkova, S. Bersier, R. Heitz, L. Müller-Kirsch, M. Geller, D. Bimberg, and V. V. Moshchalkov, Phys. Rev. B 70, 081302(R) (2004).

[7]B. Bansal, M. Hayne, M. Geller, D. Bimberg, and V. V. Moshchalkov, Phys. Rev. B 77, 241304(R) (2008).

[8]T. Sato, T. Nakaoka, M. Kudo, and Y. Arakawa, Physica E **32**, 152 (2006).

[9]M.-C. Lo, S.-J. Huang, C.-P. Lee, S.-D. Lin, and S.-T. Yen, Appl. Phys. Lett. **90**, 243102 (2007).

[10]L. Müller-Kirsch, R. Heitz, U. W. Pohl, D. Bimberg, I. Häusler, H. Kirmse, and W. Neumann, Appl. Phys. Lett. **79**, 1027 (2001).

[11]N. N. Ledentsov, J. Böhrer, M. Beer, F. Heinrichsdorff, M. Grundmann, D. Bimberg, S. V. Ivanov, B. Ya. Meltser, S. V. Shaposhnikov, I. N. Yassievich, N. N. Faleev, P. S. Kop'ev, and Zh. I. Alferov, Phys. Rev. B **52**, 14058 (1995).

[12]L. Müller-Kirsch, R. Heitz, A. Schliwa, D. Bimberg, H. Kirmse, and W. Neumann, Appl. Phys. Lett. **78**, 1418 (2001).

[13]O. Stier, M. Grundmann, and D. Bimberg, Phys. Rev. B **59**, 5688 (1999).

[14]R. Timm, H. Eisele, A. Lenz, S. K. Becker, J. Grabowski, T.-Y. Kim, L. Müller-Kirsch, K. Pötschke, U. W. Pohl, D. Bimberg, and M. Dähne, Appl. Phys. Lett. **85**, 5890 (2004).

[15]I. Vurgaftman, J. R. Meyer, and L. R. Ram-Mohan, J. Appl. Phys. **89**, 5815 (2001).