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Anomalous diamagnetic shift for negative trions in single semiconductor quantum dots

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We report on the magnetic response of negative trions X^- in single self-assembled InAs/GaAs quantum dots. Unlike the conventional quadratic diamagnetic shift for neutral excitons, the observed X^- diamagnetic shifts are small and nonquadratic. In particular, we also observed a reversal in sign of the conventional diamagnetic shift. A theoretical analysis indicates that such anomalous behaviors for X^- arise from an apparent change in the electron wave function extent after photon emission due to the strong Coulomb attraction induced by the hole in its initial state. This effect can be very pronounced in small quantum dots, where the electron wave function becomes weakly confined and extended much into the barrier region. When the electrons gradually lose confinement, the magnetic response of X^- will transit gradually from the usual quadratic diamagnetic shift to a quartic dependence, and finally into a special *paramagnetic* regime with an overall negative energy shift.

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The diamagnetic shift of confined excitons has long been used as a measure of the spatial extent of the excitonic wave functions in various semiconductor nanostructures such as quantum wires,[1–4] quantum dots (QDs),[4–6] as well as quantum rings.[7] In a weak magnetic field B, the exciton diamagnetic shift is expected to exhibit a quadratic B dependence, i.e., $\Delta E=\gamma B^2$, with a diamagnetic coefficient γ proportional to the area of the excitonic wave function, which reflects both the spatial confinements and interparticle Coulomb interactions.[8] For excitons (X) strongly confined in QDs, where the single-particle energies dominate over the Coulomb energies, γ is a measure of the spatial confinement of the QDs, while the magnetic responses of interparticle Coulomb energies only appear as correction terms to the overall diamagnetism. This simple picture is also true for biexcitons (XX) and trions (X^+ or X^-) strongly confined in QDs, except for a slight but systematic difference in γ due to the different magnetic responses of interparticle Coulomb energies.[6] On the other hand, when excitons are weakly confined in QDs such that Coulomb energies dominate over the single-particle energies, γ becomes a measure of the magnetic response of Coulomb energies. For exciton complexes weakly confined in QDs, the diamagnetic behaviors are expected to be more complicated because of the more elaborate Coulomb interactions. In particular, it has been theoretically predicted that a weakly confined negative trion (X^-) in large-size QDs would exhibit a negative magnetic dispersion,[5] based on the interpretation of the reported weak negative magnetic dispersion for X^- in two-dimensional systems.[9,10] However, such an unusual behavior of X^- has not yet been observed in past works focused on charged trions in QD system,[11,12] probably because the condition to observe this anomalous behavior is critical as we shall explain in this Brief Report.

In this Brief Report, we report on the magnetic response of negative trions (X^-) in single InAs/GaAs self-assembled QDs. We show that X^- fell into a special regime where the conventional quadratic diamagnetic shift failed to describe its magnetic response. Our measurements show that the X^- diamagnetic shifts in most QDs investigated are relatively small and nonquadratic or even exhibit a negative energy shift. A theoretical analysis indicates that such anomalous behaviors for X^- arise from an apparent change in the electron wave function extent after photon emission due to the strong Coulomb attraction induced by the hole in its initial state. We point out that when the electrons gradually lose confinement, the magnetic response of X^- will transit gradually from the usual quadratic diamagnetic shift to a quartic B dependence, and finally into a special *paramagnetic* regime with an overall negative energy dispersion.

The InAs QDs (LM4596) were grown on a GaAs (100) substrate using the Stranski-Krastanow mode by a Varian Gen II molecular beam epitaxy system. The sample structure and growth condition were detailed previously.[6] The InAs QDs were grown without substrate rotations yielding a gradient in area dot density ranging from 10^8 to 10^{10} cm^{-2}. To isolate individual QDs, a 100-nm-thick aluminum metal mask was fabricated on the sample surface with arrays of 0.3 μm apertures using electron-beam lithography. Single QD spectroscopies were carried out at 5–8 K in a specially designed microphotoluminescence (μ-PL) setup, where the sample was mounted in a low-temperature stage and inserted in the bore of 6 T superconducting magnet for magneto-PL measurement. A He-Ne laser beam was focused onto the aperture through a microscope objective (NA=0.5). The PL signals were collected by the same objective, analyzed by a 0.75 m grating monochromator, and detected by a liquid-nitrogen-cooled charged-coupled device camera, which yield a resolution limited spectral linewidth of about 60 μeV. Several apertures containing only one QD have been investigated and all of which showed similar spectral features, which in general consist of four emission lines associated with the recombination of neutral excitons (X), biexcitons (XX), and positive and negative trions (X^+ and X^-). These excitonic spectra have been unambiguously identified according to power-dependent and polarization-resolved PL measurements.[6]

Magneto-PL measurements have been performed on a total of seven QDs with X energy distributed over the range of

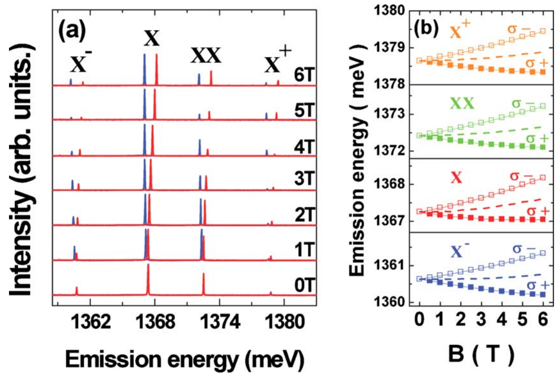

FIG. 1. (Color online) (a) The magneto-PL spectra for QD1 under a magnetic field $B=0-6$ T. (b) The corresponding peak energies of different excitonic species as a function of B for QD1, where $\sigma-$ and $\sigma+$ in each form a Zeeman doublet. The dashed line is the average energy of $\sigma-$ and $\sigma+$.

1349–1385 meV. Representative spectra selected from one particular dot (QD1) are shown in Fig. 1. When a magnetic field B was applied along the QD growth direction (Faraday geometry), each line splits into a doublet through the Zeeman effect. As shown in Fig. 1(b), the average energy of each Zeeman doublet increases with the increasing B, known as the diamagnetic shift. In Fig. 2, the measured diamagnetic shifts for the four excitonic emission lines are plotted as a function of B^2. For X, X^+, and XX, the measured diamagnetic shifts show a quadratic dependence $\Delta E=\gamma B^2$, with a clear trend of $\gamma_X > \gamma_{XX} \cong \gamma_{X^+}$. This trend holds for all investigated dots and is a consequence of different magnetic responses of interparticle Coulomb energies, as has been discussed previously.[6] In very strong contrast, the diamagnetic shift for the X^- does not follow the quadratic dependence. If we still use a quadratic dependence to fit the anomalous diamagnetic shift, the diamagnetic coefficient γ_{X^-} was found to be the smallest one among the four excitonic species as depicted in Fig. 3.

The reduced optical diamagnetic coefficient of X^- could be caused either by relatively localized particle wave functions of the initial X^- state or the extended electron wave function in the final $1e$ state. In the latter, the electron wave function is free of interparticle interactions and determined solely by the confining potential of QD. By contrast, a particle in a few-particle X^- complex is subjected to additional interparticle (e-e and e-h) interactions. The extents of the

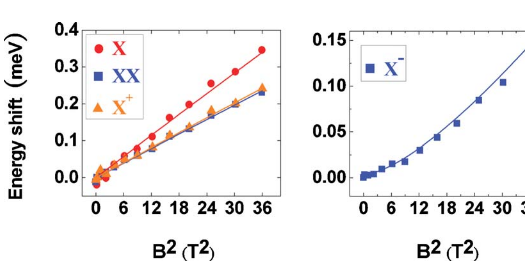

FIG. 2. (Color online) Emission energy of exciton complexes shift with B^2 for QD1. The simulated QD is 11.6 nm in diameter and 1.1 nm in height. Points are the measured data and lines are the simulated result.

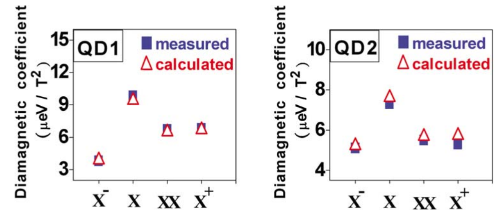

FIG. 3. (Color online) The diamagnetic coefficient of the exciton complexes for the two typical QDs (QD1 and QD2). The simulated parameters are diameter of 11.6 nm and height of 1.1 nm (QD1); diameter of 11 nm and height of 1.4 nm (QD2).

particle wave functions of the X^- and $1e$ states could become substantially different if the interparticle interactions are comparable to or even stronger than the confining strength of QD. As shown by Ref. 6, the hole wave functions are much more localized than those of electrons in such small QDs. The imbalanced e-h and e-e interactions yield a net Coulomb attraction to electrons in X^- and thus make the extent of the electron wave function of the X^- state smaller than that of the $1e$ state. To confirm such a scenario for the explanation of the observed anomalous diamagnetic behavior, we perform the numerical simulation for the wave functions of interacting particles in X complexes implemented by using the finite element method within the Hartree approximation.[13,14]

In the Hartree approximation, the Schrodinger equation of a particle in an interacting X complex confined in QD is written as

$$[H_{sp}(\vec{r}_i) + V_H(\vec{r}_i)]\varphi_n(\vec{r}_i) = \varepsilon_n^i \varphi_n(\vec{r}_i), \quad (1)$$

where $\vec{r}_i$ denotes the position coordinate of the ith particle, $H_{sp}(\vec{r}_i)$ stands for the Hamiltonian of the (noninteraction) electron or hole, ε_n^i the eigenenergy, $\varphi_n(\vec{r}_i)$ the particle wave function of the eigenstate $|n\rangle$, and $V_H(\vec{r}_i)$ is the Hartree potential, a sum of the electrostatic potentials induced by other charged particles besides the considered particle itself.

In the calculation, cone-shaped QDs of various sizes sitting on a 0.4-nm-thick wetting layer are considered. The material parameters including the effective mass, dielectric constant, band offset, and band gap with the consideration of strain effect are taken from Ref. 15. For an N-particle X complex, the N-coupled equations for each particle according to Eq. (1) are self-consistently solved by using an iterative approach. The total energy of the N-particle X complex is then determined by the sum of single-particle energies $\{\varepsilon^i\}$ but subtracted by the doubly counted interparticle interaction energy. The simulated results of diamagnetic coefficients for four different exciton complexes are plotted in Fig. 3. The diamagnetic coefficients are obtained by taking the second derivative of magnetoenergy spectra with respect to B using three-point numerical differentiation.

We first discuss the QD's size effect on the diamagnetic shifts. The calculated diamagnetic coefficients for the four exciton complexes in QDs with a diameter ranging from 12 to 26 nm are shown in Fig. 4(a). For large-sized QDs ($D > 16$ nm), the diamagnetic coefficients of the four exciton complexes show similar increasing trends with the QD size. Because the diamagnetic coefficient is proportional to the

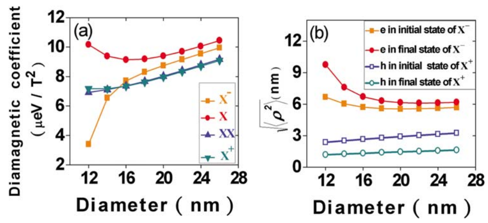

FIG. 4. (Color online) The simulation for QDs with various diameters, the diamagnetic coefficients (a), and the mean radiuses of electron and hole wave functions in their initial and final states (b).

area of the carrier's wave function, it will reflect the dot diameter for large-size QDs.[4,16] However, for smaller QD sizes ($D<16$ nm), the calculated values of γ_X increase with the decreasing dot size, while those of γ_{XX} and γ_{X^+} remain nearly unchanged. The most striking feature is that the calculated γ_{X^-} drops rapidly with the decreasing dot size. It is worth to mention that our self-consistent calculations reproduce the experimental finding of $\gamma_X > \gamma_{XX} \cong \gamma_{X^+}$ very well, consistent with previous calculations based on configuration-interaction methods.

For the emission of negative trion X^-, the initial state consists of two electrons and one hole leaving one electron in its final state after recombination. The X^- diamagnetic shift thus reflects the diamagnetic responses of both the initial and final states. To understand the anomalous behavior for the X^-, it is necessary to take a closer look at the lateral extent $\ell_e \equiv \sqrt{\langle \rho_e^2 \rangle}$ of the electron wave functions before and after photon emission. Figure 4(b) shows the calculated wave function extents $\ell_{e,i}$ and $\ell_{e,f}$ for the initial-state and the final-state electrons of X^-, respectively. One can see that the $\ell_{e,f}$ is always more or less larger than $\ell_{e,i}$. This can be realized from the presence of the hole in its initial state, which contracts the electron wave function by the Coulomb attraction. When the sizes of QDs are larger than about 16 nm, the differences between $\ell_{e,i}$ and $\ell_{e,f}$ are small; i.e., the presence of the hole does not change the electron wave function significantly. However, as the QD sizes reduces, $\ell_{e,f}$ increases rapidly, with a rate even faster than $\ell_{e,i}$. Such an increasing trend for $\ell_{e,f}$ indicates that the electron gradually loses confinement as the dot size reduces, which pushes the electron level toward the wetting-layer continuum, resulting in a very extended electron wave function penetrating into the barrier material. In such a case of weak confinement regimes, the very extended initial-state electron becomes sensitive to the long-range Coulomb attractive potential produced by the hole, by which $\ell_{e,i}$ will be contracted and become apparently smaller than $\ell_{e,f}$. As a result, the final-state diamagnetic shift increases, so that the overall diamagnetic shift in X^- is reduced. This explains why the X^- diamagnetic shift decreases rapidly for small-sized QDs shown in Fig. 4(a).

Likewise, for the emission of positive trion X^+, the initial state consists of one electron and two holes leaving one hole in its final state after recombination. As shown in Fig. 4(b), unlike X^-, the lateral extents of hole wave functions $\ell_h \equiv \sqrt{\langle \rho_h^2 \rangle}$ in the initial and final states are almost identical for all QD sizes. Due to the larger effective mass of holes, their wave functions are well confined even in such small QDs. In this case, the Coulomb attractive potential produced by the weakly confined electron becomes less important, so that the size dependence of γ_{X^+} behaves as usual.

Now one may ask the question why the X^- diamagnetic shift exhibits a nonquadratic B dependence. In general, a quadratic diamagnetic shift holds only in the weak-field limit, i.e., when the magnetic length $\ell_M = \sqrt{\hbar/eB}$ is large compared to the lateral extents of the carrier's wave functions $\ell = \sqrt{\langle \rho^2 \rangle}$. We noted that $\ell_M = 15$ nm at $B=3$ T, which becomes comparable with $\ell_{e,f}=10$ nm for the final-state electron in a QD with a base diameter of 12 nm. In this regime, the diamagnetic shift would deviate from the typical B^2 dependence. To illustrate this behavior, we consider the diamagnetic shift in the carrier's single-particle energy and expand it in powers of B as $\Delta\varepsilon_\alpha^{\mathrm{SP}}(B) = \gamma_\alpha B^2 + \kappa_\alpha B^4 + \cdots$, where the quadratic and quartic coefficients are $\gamma_\alpha = e^2 \ell_\alpha^2 / 8 m_\alpha$ and $\kappa_\alpha = -e^4 \ell_\alpha^6 / 128 m_\alpha \hbar^2$, in which $\alpha = e$ or h denotes the electron or hole, and m_α represents the effective mass of the electron or hole. Because $|\kappa/\gamma|$ varies as $\sim \ell^4$, the contribution from the B^4 term becomes increasingly important as $\ell \sim \ell_M$. By taking into account the difference between $\ell_{e,i}$ and $\ell_{e,f}$, a simple algebraic analysis for the X^- diamagnetic shift $\Delta E_{X^-}(B)$ gives the following expression:

$$\Delta E_{X^-}(B) \approx \gamma_{X^-} B^2 + \kappa_{X^-} B^4 + \cdots, \qquad (2)$$

where $\gamma_{X^-} = (2\gamma_{e,i} + \gamma_{h,i}) - \gamma_{e,f}$ and $\kappa_{X^-} = (2\kappa_{e,i} + \kappa_{h,i}) - \kappa_{e,f}$. Because of $\ell_h < \ell_e$ and $m_h \gg m_e$, the $\gamma_{h,i}$ and $\kappa_{h,i}$ for the hole only have minor influences on the overall diamagnetism. Accordingly, we obtain $\gamma_{X^-} \approx 2\gamma_{e,i} - \gamma_{e,f} = e^2(2\ell_{e,i}^2 - \ell_{e,f}^2)/8m_e$ and $\kappa_{X^-} \approx 2\kappa_{e,i} - \kappa_{e,f} = -e^4(2\ell_{e,i}^6 - \ell_{e,f}^6)/128 m_e \hbar^2$. Equation (2) makes clear how the difference in $\ell_{e,i}$ and $\ell_{e,f}$ can lead to anomalous diamagnetic behaviors for the emission energy of X^-. We first consider a normal case of $\ell_{e,i} \approx \ell_{e,f} = \ell_e$; we have $\gamma_{X^-} \approx \gamma_e \approx \gamma_X$ and $|\gamma_{X^-}| \gg |\kappa_{X^-}|$, as long as $\ell_e < \ell_M$. That is, the X^- diamagnetic shift behaves as the usual quadratic dependence with a coefficient similar to that of X, which is just the case for large-size QDs shown in Fig. 4(a). A very interesting case occurs when $\sqrt{2}\ell_{e,i} = \ell_{e,f}$; i.e., $\ell_{e,i}$ of the initial-state electrons were contracted to $\sim 70.7\%$ of its final-state extension $\ell_{e,f}$ by the hole. In this special case, the condition $2\gamma_{e,i} = \gamma_{e,f}$ cancels out the B^2 term leading to a dominant quartic dependence on B. As the difference between $\ell_{e,i}$ and $\ell_{e,f}$ becomes even larger ($\sqrt{2}\ell_{e,i} < \ell_{e,f}$), the magnetic response of X^- goes into a new regime where the quadratic coefficient γ_{X^-} is negative; i.e., the energy shift is *paramagnetic*. This anomalous behavior can be best seen from the calculated results shown in Fig. 5(a), where we keep $\ell_{e,f} = 10$ nm but varying $\ell_{e,i}$ from 7.6 to 6.6 nm. The magnetic response of X^- emissions transited gradually from the usual quadratic diamagnetic shift to quartic dependences, and finally into an overall negative energy shift, resembling paramagnetic behaviors. In Fig. 5(b), we selected four typical QDs that could represent the behaviors in different regimes in qualitative agreement with our calculations.

The magnetic responses of negative trions X^- in single self-assembled InAs/GaAs quantum dots have been investigated. Unlike the conventional quadratic diamagnetic shift

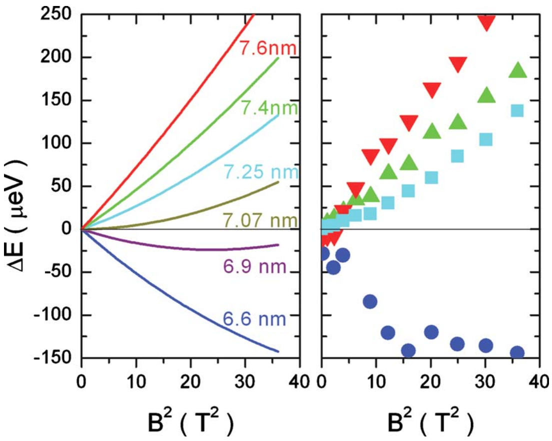

FIG. 5. (Color online) The theoretical (left) and experimental (right) anomalous diamagnetic shifts in small QDs.

for neutral excitons, the X^{-} diamagnetic shifts in most of the investigated dots were found to be considerably small and nonquadratic. In particular, we also observed a reversal in sign of the conventional diamagnetic shift. A theoretical analysis indicates that such anomalous behaviors for X^{-} arise from an apparent change in the electron wave function extent after photon emission due to the strong Coulomb attraction induced by the hole in its initial state. This effect can be very pronounced in small quantum dots, where the electron wave function becomes weakly confined and extended much into the barrier region. When the electrons gradually lose confinement, the magnetic response of X^{-} will transit gradually from the usual quadratic diamagnetic shift to a quartic dependence, and finally into a special paramagnetic regime with an overall negative energy shift.

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