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Coherent Aharonov-Bohm oscillations in type-II (Zn,Mn)Te/ZnSe quantum dots

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The magneto-photoluminescence of type-II (Zn,Mn)Te quantum dots is presented. As a result of the type-II band alignment, Aharonov-Bohm (AB) oscillations in the photoluminescence intensity are evident. In addition, an interesting interplay between the AB effect and the spin polarization in these diluted magnetic semiconductor quantum dots is observed. The intensity of the AB oscillations increases with both magnetic field and the degree of optical polarization, indicating that the suppression of spin fluctuations improves the coherence of the system.

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The Aharonov-Bohm (AB) effect describes a phase shift induced upon a charged particle as it moves in a closed trajectory in the presence of a magnetic field.[1] Despite the fact that the AB effect is a property of charged particles, it has been shown that for neutral excitons in semiconductor nanorings, a nonzero electric dipole moment exists,[2–5] which is adequate to create AB oscillations. Such behavior results from the differences in confinement potential and effective masses of the electron and hole, resulting in a different trajectory for each of the charge carriers and, thus, a measurable AB effect. Type-II quantum dots (QDs) have been predicted to be particularly amenable to exhibiting such AB effects because of the enhanced polarization of the exciton due to the spatial separation of the carriers in such systems.[2,4,5]

In this work, we present experimental evidence of AB oscillations in the magneto-photoluminescence (MPL) of type-II (Zn,Mn)Te/ZnSe QDs. In this system, the hole is strongly confined within the (Zn,Mn)Te QD, while the electron resides in the ZnSe matrix, confined only through coulomb attraction to the hole. Although the AB effect has been observed optically for charged excitons in In(Ga)As QDs,[6] for neutral excitons in both Type-II GaAs/InP[7] and Zn(SeTe) QDs[8,9] and also in capacitance[10] and magnetization measurements[11] in InAs quantum rings, we report such effects in a diluted magnetic semiconductor (DMS) system. In DMS materials, the application of a magnetic field strongly aligns the Mn spins, thus, polarizing the optical emission through the carrier-Mn exchange interaction.[12] At saturation polarization, the carrier spins will also be preferentially aligned, and the spin disorder in the system is significantly reduced.[12,13]

The samples studied were grown by molecular beam epitaxy (MBE) on (001) GaAs substrates. The GaAs buffer layer was planarized at 580 °C before the temperature was reduced to 300 °C to deposit a ZnSe buffer. The (Zn,Mn)Te QDs were formed through conventional self-assembled growth, resulting from the 7% lattice mismatch between the ZnSe and (ZnMnTe) layers. In the sample described here, five 2.6 ML (Zn,Mn)Te QD layers were grown, separated by narrow (5 nm) ZnSe spacer layers. The Mn composition in the dot containing layers was estimated to be ~5% by x ray diffraction techniques. The growth optimization and material characterization of this sample are described fully elsewhere.[13]

Figure 1 shows the photoluminescence (PL) at 4.2 K. The QD emission peaks at ~1.92 eV, which is much lower in energy than the peak emission from bulk $Zn_{0.95}Mn_{0.05}Te$ at low temperatures (~2.4 eV).[12,14] This large redshift of the energy gap observed for the (Zn,Mn)Te/ZnSe QDs is a direct result of the type-II band alignment and confinement potential of the dots.

The incorporation of Mn into the QDs forms the paramagnetic alloy (Zn,Mn)Te, with large *g*-factors for both the electrons and holes due to the large exchange interaction that exists between the localized Mn spins and the charge carriers in this material.[12] These properties result in a large optical polarization of the PL with applied magnetic field. This behavior is illustrated in the inset (a) to Fig. 1, which shows the integrated PL intensity of the σ^+ and σ^- emission measured in the Faraday geometry. As the magnetic field increases, the electron ($m_s=\pm 1/2$) and heavy hole states ($m_j=\pm 3/2$) split, creating a situation where the carriers preferentially occupy their lowest energy states ($m_s=-1/2$, $m_j=3/2$). The emitted polarization of the PL then directly represents the

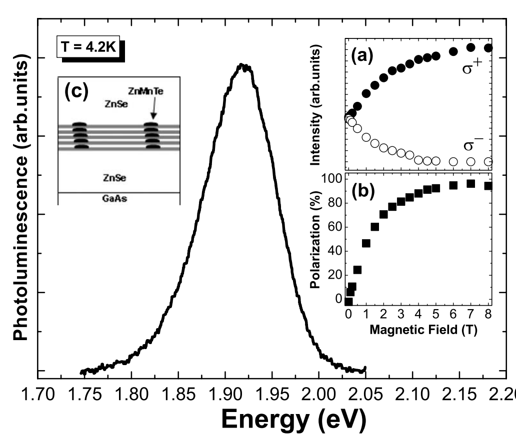

FIG. 1. PL of the ZnSe/(Zn,Mn)Te quantum dots at 4.2 K. The inset (a) shows the intensity of the σ^+ (full circles) and σ^- (open circles) photoluminescence component versus magnetic field. The inset (b) shows the degree of optical polarization while inset (c) shows a schematic of the structure.

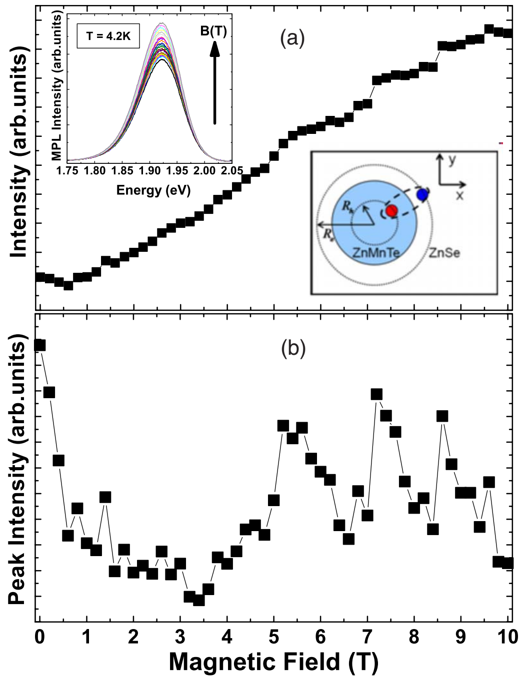

FIG. 2. (Color online) (a) Integrated PL intensity versus magnetic field for the ZnSe/(Zn,Mn)Te QDs at 4.2 K. The upper inset (left) shows the magneto-photoluminescence spectra with increasing magnetic field. The lower inset (right) represents a simple model created by the electron and hole in this type-II columnar geometry. (b) Peak PL intensity versus magnetic field for the QDs with the rising background observed in (a) removed.

total carrier spin orientation due to the selection rules of the transition.[14] Here, since the dominant transition is associated with an exciton formed between (spin down) electrons ($m_s=-1/2$) and (spin up) holes ($m_J=+3/2$), with total angular momentum projection equal to $+1$,[15] the emission of photons is dominated by left-circularly polarized (σ^+) light.

Figure 1(b) shows the optical polarization $P=(I_+-I_-)/(I_++I)$, where $I^+(I^-)$ represents the intensity of the $\sigma^+(\sigma^-)$ luminescence, respectively. Despite the inhomogeneity of the QDs, the optical polarization degree is greater than 95% at magnetic fields above 4 T.

As a result of the relatively narrow ZnSe spacer layers in these structures, the ZnMnTe QDs form columns due to strain interaction between adjacent dot-containing layers.[16,17] This geometry, which was confirmed by cross-sectional transmission electron microscopy,[13] coupled with the type-II band alignment, has previously been shown to be particularly appropriate for observing the optical AB effect.[8,9] Not only is the electron *bound* to the *confined* hole through coulomb attraction, but because of the narrow ZnSe barrier layer, it is constrained to move in-plane, thus, defining a ringlike geometry.

The main panel in Fig. 2(a) shows the magnetic field dependence of the PL intensity for an ensemble of (Zn,Mn)Te/ZnSe QD columns. The overall PL intensity increases with magnetic field consistent with the field, "squeezing" the electron wave function at the QD interface, which increases the electron-hole wave function overlap and, thus, the oscillator strength. We also note here that an unusually large increase in the PL intensity of II-VI DMS systems has been reported previously, although the origin of this behavior is still under discussion.[18] In the main panel of Fig. 2(a), periodic oscillations are visible, superimposed upon the rising intensity background of the emission. These oscillations are clear evidence of a coherent AB effect between the constituent particles of the exciton —the electron and hole— and are consistent with the behavior observed in similar Zn(SeTe) QDs.[8,9,19]

The specific origin of oscillatory behavior in the PL intensity is related to the cylindrical symmetry of the QD column and the optical AB effect. The radial spatial separation in the type-II QD creates a rotating dipole with the two charged particles of the exciton orbiting over different areas. This behavior is described qualitatively by a simple model of a rotating dipole [shown as inset of Fig. 2(a)] with a magnetic field normal to the plane,

$$E_{\text{exc}} = E_g + \frac{\hbar^2}{2MR_0^2}\left(L + \frac{\Delta\Phi}{\Phi_0}\right)^2, \tag{1}$$

where L is the angular momentum quantum number, E_g is the confined hole-to-electron ground state energy, $\Phi_0=hc/|e|$ is the magnetic flux quantum, $R_0=(R_e+R_h)/2$, and $M=(m_eR_e^2+m_hR_h^2)/R_0^2$; R_h and R_e are the averaged radii of orbits of the hole and electron, respectively, and m_h and m_e are their masses. Here, since the hole is strongly localized, $R_h\approx 0$. The quantity $\Delta\Phi$ is the magnetic flux through the area between electron and hole trajectories.

With increasing magnetic field, the L value characterizing the ground state angular momentum projection changes from zero to successively larger (in magnitude) nonzero values due to the cylindrical symmetry of the system. Such behavior should have significant impact upon the PL intensity since the selection rules for the optical transition are modified, and in the simplest case, with increasing magnetic field when $L\neq 0$, the intensity should be strongly suppressed.[4] Interestingly, the oscillations in the PL intensity shown in Fig. 2(a) are not consistent with this picture, with *bright* nonzero L states evident with increasing magnetic field. This unusual behavior is likely related to a relaxation of the selection rules due to symmetry breaking in the nonideal QDs studied here. In addition, impurity scattering and localization effects at the QD perimeter[20] can enhance these effects, as was observed in the nonmagnetic ZnSeTe QDs formed through Te clustering.[9] In those particular nonmagnetic dots, although oscillations in the PL intensity were evident for higher order $L\neq 0$ states, the overall intensity of the emission actually decreased.[9] For the DMS dots described in this Rapid Communication, spin-orbit interaction may also play a role. However, since spin-orbit coupling is relatively weak for electrons, particularly in wide gap ZnSe, these effects are probably weak.

The effective radius ($R_e=\sqrt{\Phi_0/\pi\Delta B}$) of the electron orbit can be estimated from the period of the AB oscillations and in this case, is determined to be $\sim$28 nm. This is larger than the 20 nm lateral extent of the QDs determined by structural analysis[13] and, as such, is consistent with the picture of the electron orbiting the QD perimeter bound to the hole.[2,4,5]

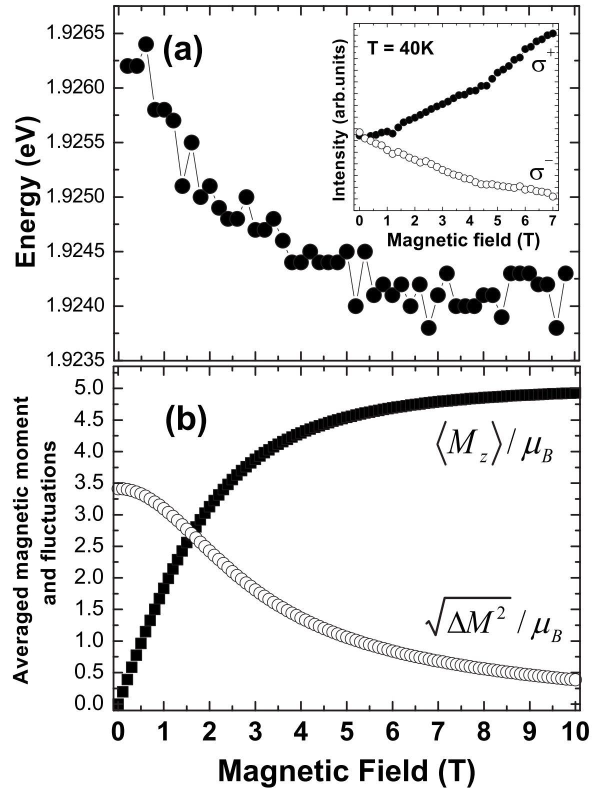

FIG. 3. (a) PL energy versus magnetic field for the ZnSe/(Zn,Mn)Te QDs. The inset shows the intensity of the σ^+ (full circles) and σ^- (open ciricles) PL at 40K. (b) Calculated average magnetic moment (closed squares) and fluctuations of a single Mn spin (open circles) in the units of the Bohr magneton.

Importantly, the estimated AB oscillations in the energy of the lowest state of the exciton, determined from Eq. (1), is ~ 0.04 meV due to the relatively large lateral radius (R_e) of the electron orbit (28 nm). As a result, we do not expect to observe these weak energy oscillations experimentally in the peak PL energy due to the strong inhomogeneous broadening (~ 80 meV) of the excitonic peak. This is indeed shown to be the case in Fig. 3(a), where the position of the PL peak does not exhibit any visible oscillations but rather a slight reduction in the transition energy with magnetic field. The origin of this behavior is related to the Zeeman contribution to the emission, which, interestingly, is much smaller (~ 7 meV) than expected for bulk $Zn_{0.95}Mn_{0.05}Te$ (~ 50 meV). This unusual behavior is currently under investigation, the details of which will be discussed elsewhere.

Turning back to the oscillations in the emission intensity, it can be seen from the data of Fig. 1(a) that the strength of the oscillations increases with magnetic field. This is more obvious in Fig. 2(b), which shows the peak intensity of the oscillations with the PL normalized to the featureless background luminescence. In this figure, three oscillations are clearly evident with peaks at ~ 5.4, 7.4, and 9 T. This enhancement of the AB effect at higher magnetic fields appears to be strongly related to the spin polarization of the carriers in the DMS QDs (and, thus, to the magnetization of the dots) and can be correlated with the optical polarization [see inset (b) of Fig. 1]. At low magnetic fields, the Mn spins are randomly orientated and fluctuating[14] due to the finite temperature of the system (~ 4 K) and low transition temperature of dilute ZnMnTe.[12,14]

Since the *exciton* and the Mn spins are coupled through an exchange interaction, fluctuations of Mn spins create a fluctuating exchange potential for the electron orbiting a QD column. In addition, the finite penetration of the electron wave function into the Mn-containing QD will further enhance these effects.[8] Such fluctuations may have a destructive effect upon the AB phase, which may not survive. The behavior of the spectra in Fig. 2(b) at $B<4$ T is consistent with this idea. As the magnetic field aligns the Mn spins, magnetic fluctuations in the system are reduced and the AB oscillations increase in strength. This is also consistent with the increase in the optical polarization degree with the largest AB oscillations observed at magnetic fields above polarization saturation (>4 T).

The average magnetic moment of a single Mn impurity and the strength of spin fluctuations can be written as

$$\bar{M}_z = \langle M_z \rangle = S \cdot g_{\mathrm{Mn}} \cdot \mu_B \cdot B_{5/2}(x) \quad \text{and} \quad \sqrt{\Delta M^2}$$
$$= \sqrt{\langle (M_z - \bar{M}_z)^2 \rangle} = \sqrt{k_B T \frac{\partial \bar{M}_z}{\partial B}}, \tag{2}$$

where $\langle M_z \rangle$ is the thermal average over the states of Mn spin with $S=5/2$, $\boldsymbol{B}_{5/2}(\boldsymbol{x})$ is the Brillouin function $x = g_{\mathrm{Mn}} \cdot S \cdot \mu_B \cdot B / k_B T$, and B is the magnetic field. We take $T=4.2$ K and $g_{\mathrm{Mn}}=2$. The results of these calculations are shown in Fig. 3(b) and clearly illustrate the suppression of spin fluctuations for $B>4$ T. Consequently, these predictions support the assumption that the increase in AB oscillations at higher B is related to the reduction of the fluctuations of the exchange potential. This behavior is also supported by temperature-dependent measurements, which are shown in the inset to Fig. 3(a). With increasing temperature the AB oscillations damp and, although weak, remain evident at 40 K. Above 50 K, which is the temperature at which thermal effects completely randomize the system, the coherence is lost.[21] In Mn-doped systems, some amount of Mn ions may form antiferromagnetic clusters with frozen spin configurations; these clusters also create a scattering potential for electrons and can affect the AB oscillations.

In summary, we have presented clear evidence of the Aharonov-Bohm effect in the PL intensity of type-II (Zn,Mn)Te/ZnSe QDs. Simultaneously, we have observed an interesting interplay between the AB oscillations and the magnetization. The strength of the AB effect appears to be correlated with the degree of optical polarization of the system. We believe that with increasing magnetic field and, therefore, increased spin polarization, the AB oscillations become enhanced due to the suppression of spin fluctuations and related decoherence. The appearance of such oscillations in the columnar QDs presented here appears to confirm previous predictions of the suitability of this particular geometry[8,9] for the creation and control of AB effects in semiconductor systems.

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