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Modulation of the Aharonov–Bohm effect in type-II II–V ZnSe: Te quantum dots by a far-infrared laser

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Abstract

We present magneto-photoluminescence of a ZnTe/ZnSe multilayer structure in which the clustering of Te atoms results in the formation of columns of type-II ZnSe:Te quantum dots. This unique geometry permits the observation of strong Aharonov–Bohm oscillations in *both* the emission energy and peak intensity with increasing magnetic field. Furthermore, far-infrared optically detected resonance techniques exhibit resonances that are ascribed to the activation and recapture of electrons by different subsets of quantum dots.

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The Aharonov–Bohm effect (ABE) describes a phase shift induced upon a charged particle when it traverses a closed orbit in a magnetic field [1]. The ABE on an exciton in such geometry is predicted to create an oscillation in the band edge emission energy and a "blinking" of the intensity in photoluminescence (PL) [2].

Although oscillations in the energy have now been observed in the charged exciton emission from single In(Ga)As quantum rings [3] and in neutral exciton spectra of ensembles of type-II InP quantum dots (QDs) [4] to our knowledge multiple-oscillation variation in the luminescence intensity of excitonic emission has not been demonstrated experimentally. In the following article, clear experimental evidence of the optical ABE in type-II Zn(Te)Se QDs as a function of magnetic field is presented. Furthermore, we show that it is possible to modulate these AB oscillations with a far-infrared (FIR) laser beam and measure resonances using optically detected resonance spectroscopy (ODR) [5].

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The sample consists of a 240-layer ZnSe/ZnTe superlattice structure grown by molecular beam migration enhanced epitaxy upon a GaAs substrate. The growth technique is described in detail elsewhere [6], however, some important details of the dynamics of this process are described here for clarity. Upon deposition, Te atoms substitute for Se along specific crystal orientations. This process results in a clustering of the Te atoms in multilayer structures and has been shown recently to result in the formation of ZnTe QDs [6–8]. It is QDs formed in this manner that are described in this work.

PL measurements were performed between 0 and 300 K in a 10 T Oxford superconducting magnet using a HeCd laser at 3.81 eV. The resulting PL at 4 K for the ZnSe/ZnTe structure is shown in Fig. 1. The emission is dominated by two features: one at 2.66 eV (A) and another at 2.496 eV (B). A weak shoulder is also evident at 2.35 eV (C), the contribution of which increases significantly with temperature (not shown here) due to carrier redistribution. The free excitons of the alloyed ZnSe:Te matrix can also be observed at 2.79 eV (D).

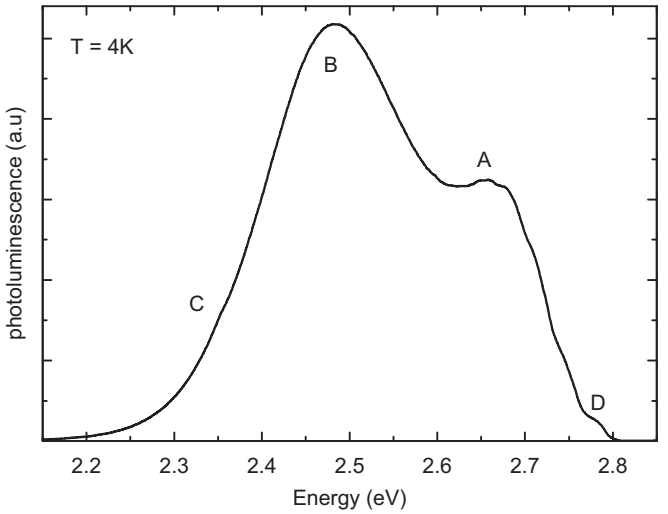

Fig. 1. Photoluminescence of a 240-layer ZnSe/ZnTe superlattice structure at 4 K. The emission is the result of the various contributions of excitons bound to isoelectronic centers (A), a combination of clusters of such excitons and type-II ZnSe:Te QDs (B) and Te-rich ZnTe QDs (C). The free excitonic emission of the alloyed ZnSeTe matrix is also evident at 2.79 eV (D).

The emission properties of the Zn(Te)Se system have been studied extensively [8–12], and it is generally considered that the PL results from the emission of excitons bound to pairs of Te atoms, which substitute onto Se sites isoelectronically [12]. The two broad bands in Fig. 1 are a characteristic of the emission of the Zn(Te)Se system and are generally attributed to isoelectronic centers of various configurations. The *higher* energy band (A) has historically been interpreted as emission from interacting excitons bound at two Te atoms, while the *lower* energy feature (B) is usually attributed to "clusters" of such complexes. However, recently it has been shown that in multilayer structures these "clusters" *evolve* into type-II quantum QDs [6,7], in which the hole is confined within the Te-rich QD and the electron sits in the ZnSe matrix. Soon after, it was also shown that such QDs also form in single ZnSe/ZnTe layers if the ZnTe deposition is >3 MLs [8].

In the structure described in this article, the formation of such QDs is supported by intensity dependent and time-resolved photoluminescence (TRPL) [7,11] measurements, which indicate that the emission changes from that dominated by bound excitons at high energy, to that of type-II QDs at lower energies. Furthermore, the AB oscillations presented below can only occur for the type-II QDs and should be absent for the excitonic complexes and quantum wells (QWs), because these structures do not exhibit the correct geometry [2,13]. This analysis is consistent with the magneto-photoluminescence (MPL) measurements discussed where AB oscillations were not observed for feature (A). Based upon these considerations, we believe that peak (B) in Fig. 1 is the result of the combined contribution of isoelectronic clusters and ZnSe:Te QDs, while peak (C) results from a *smaller* number of pure Te QDs.

The MPL of the ZnSe/ZnTe structure at 4 K is shown in Fig. 2. Fig. 2(b) and (d) show the peak intensity for the ZnSe:Te (B) and ZnTe (C) QDs, respectively, which are obtained by considering that the overall emission results from the inhomogeneously broadened contribution of the various centers involved and fitting each feature (A–C) with a Gaussian function. Similar analysis is used in Fig. 2(a) and (c) to determine the emission energy of the ZnSe:Te and ZnTe QDs, respectively.

Considering first the energy, both the ZnSe:Te (Fig. 2(a)) and the ZnTe QDs (Fig. 2(c)) display clear oscillations, indicating the presence of the ABE for both type of QD. The strength of this ABE also appears strongly related to the *inhomogeneity* of the QDs because the size of the oscillations for ZnTe is much greater than that of the ZnSe:Te QDs. Further evidence for the effect of inhomogeneity, which is probably predominantly due to composition fluctuations, of the QDs can be observed in the intensity measurements. In Fig. 2(b), the ZnSe:Te QDs display clear oscillations, but the overall intensity decreases with increasing magnetic field. This behavior is not evident for the ZnTe QDs shown in Fig. 2(d). We believe this behavior is related to potential fluctuations at the ZnSe/ZnTe interface inducing localization of the charge carriers. Such localization effects have been observed previously in type-II AlGaAs/AlAs quantum wells [14]. The attribution of the decrease in intensity to effects of localization is further supported by temperature-dependent measurements (not shown here) which show an overall increase in the emission intensity at $T>60$ K.

To understand the origin of the oscillations in the optical properties, the symmetry of the system must be considered. As a function of applied magnetic field, the electron will orbit the QD periphery with an energy, which, to first order, is described by

$$E_e = \frac{\hbar^2}{2mR^2}\left[l_e + \frac{\Phi}{\Phi_0}\right]^2,$$

where, $\hbar$ and m are Planck's constant and the electron effective mass, respectively, R is the radius of the electron orbit, $\Phi = \pi R^2 B$, $\Phi_0 = \hbar c/e$ and $l_e = 0, \pm 1, \pm 2, \pm 3$ is the orbital angular momentum projection quantum number.

The cylindrical symmetry causes the lowest electron energy (ground state) to change from a state with an initial orbital angular momentum projection of $l_e = 0$ to states having increasing orbital angular momentum projection with increasing magnetic field. It is the crossing of the initial ground states to those having increasing orbital angular momentum projection that results in the oscillations in the emission energy and "blinking" of the intensity as predicted previously [2] and shown experimentally above.

Although similar effects have been observed previously in type-I In(Ga)As [3] and type-II InP/GaAs rings, [4] there are two important differences here. Foremost, this is the *first time* such effects have been demonstrated in both the

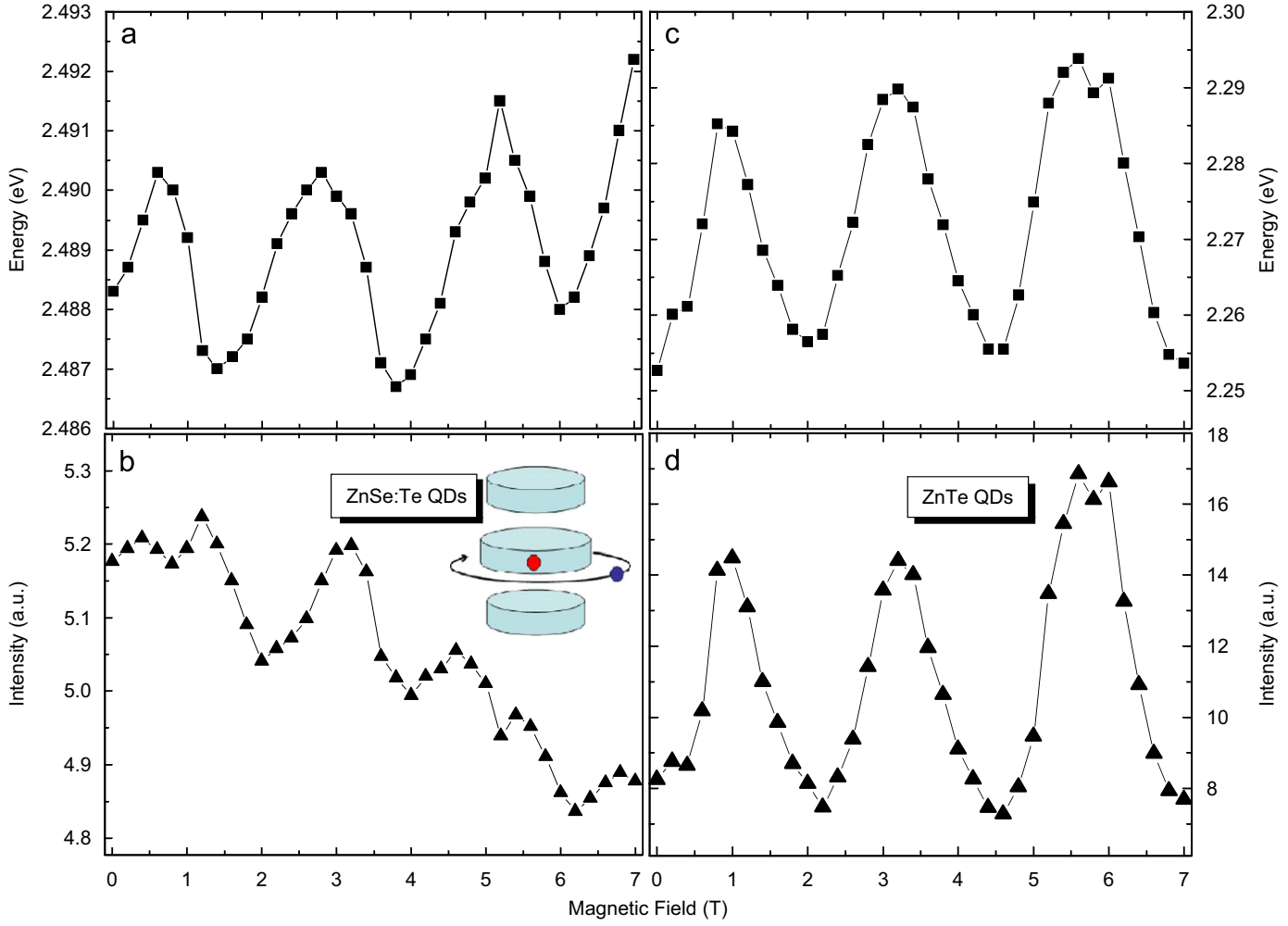

Fig. 2. Peak intensity versus magnetic field for ZnSe:Te (a) and ZnTe (b) QDs at 4 K, respectively. The emission energies for ZnSe:Te and ZnTe QDs are also shown at 4 K in parts (b) and (d). The inset in part (b) shows a schematic representation of the ring-like geometry of the columnar QDs [13] produced due to clustering of Te atoms in the multilayer structure.

emission energy and PL intensity, and secondly, the effects here are considerably stronger, with no significant damping of the oscillations evident within the range of magnetic field studied. We believe that this is strongly related to the geometry of the system presented, which "creates" a ring-like geometry, due to the combined contribution of the type-II offset and the coulomb attraction between the charge particles. Furthermore, it has been shown that in the particular multilayer structure described, the clustering of Te results in the formation of QDs that are stacked vertically creating a column of QDs [13]. This geometry (shown schematically in the inset of Fig. 2(b)) will *force* the electron to orbit the confined hole in the plane of the layer; a geometry particularly suitable for traversal of the electron around the applied magnetic field. We should also note that the same sample was investigated in Ref. [13] and the observed MPL spectra were interpreted in terms of the AB effect. In Ref. [13], however, mainly one pronounced AB-related maximum in the PL intensity was recorded, probably due to the collection of the PL from a large area. Here, by reducing the laser spot, we were able to see several strong oscillations in PL intensity and peak position, associated with the AB effect.

In addition to the oscillatory behavior of the *interband* transitions described above, it is also of interest to study the *intraband* properties of the system. Since l_e, which labels the projection of angular momentum along the magnetic field, is a good quantum number, the selection rule for transitions between angular momentum states is $\Delta l_e = \pm 1$. Such transitions are allowed and are predicted in the present system to be of the order of 3 meV in energy [13]. As such, it is possible to probe these states with FIR spectroscopy [15] and directly deduce the energy of these transitions.

Far-infrared ODR spectroscopy is a two-color PL technique which allows the observation of *subtle* changes in the emission after the excitation of the sample with a FIR laser. The ODR signal represents the difference in the emission when the FIR laser is ON and OFF and has been successfully used to study the nature of charged excitons in QWs [16] and more recently QDs [17]. Here, we use a CO_2-pumped FIR molecular gas. In the following, methanol gas

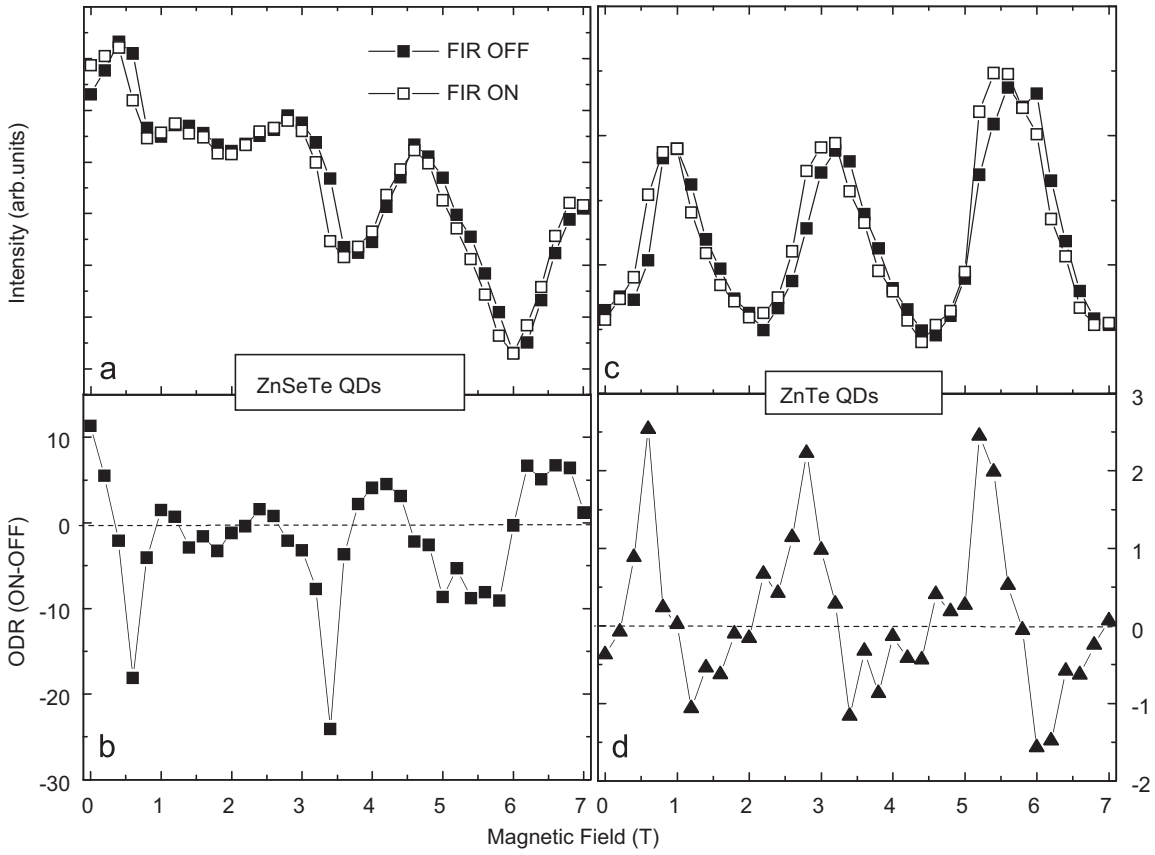

Fig. 3. Photoluminescence intensity versus magnetic field for ZnSe:Te (a) and ZnTe (c) QDs, respectively. The closed squares (■) represent the intensity with the FIR laser OFF and the open squares (□) with the laser ON. The ODR resonances for an FIR energy of 2.1 meV are shown for the ZnSe:Te and ZnTe QDs in parts (b) and (d), respectively. These resonances represent the difference in PL intensity with the FIR laser ON and OFF.

is used as the lasing medium with emission at 2.1 meV (570 μm).

Fig. 3(a) and (c) show the effect of FIR excitation upon the Aharonov–Bohm oscillations in the intensity for the ZnSe:Te QDs (~2.5 eV) and the ZnTe QDs (~2.35 eV), respectively. The closed (■) and open (□) squares represent the intensity with the FIR laser OFF and ON, respectively. For both the ZnSe:Te and ZnTe QDs, a shift in the overall oscillations to lower magnetic field is evident with a small effect upon their general shape; the largest shift occurs on the falling edge of the oscillations for the ZnSe:Te QDs and the rising edge for the ZnTe QDs. This overall behavior seems to be independent of the FIR laser energy (2–10 meV) used and it is thus attributed to non-resonant heating effects. Despite these generic non-resonant heating effects, evidence also exists for *resonant* effects upon FIR excitation.

The ODR signal (difference between FIR laser ON and OFF) for the ZnSe:Te and ZnTe QDs for an FIR energy of 2.1 meV is shown in Fig. 3(b) and (d), respectively. The behavior of both types of QDs is qualitatively similar with three well-defined resonances apparent around 0.6, 3.4 and 5.2 T. However, the resonances for the higher energy ZnSe:Te QDs are negative, while those of the lower energy ZnTe QDs are positive. These resonances are specific to excitation at 2.1 meV and were not evident for any other FIR energies used, unlike the thermal behavior previously described. A positive ODR signal suggests the activation of a carrier, and has been observed previously for charged excitons in QWs [16]. In such excitonic complexes, the negative ODR due to trion emission is followed by a subsequent increase in the neutral exciton emission, observed as a positive ODR [16]. Fig. 3(b) and (d) appear to indicate that the reduction of the ZnSeTe emission induced by the FIR at 2.1 meV has a direct consequence upon the ZnTe QDs; it enhances their signal.

To explain the origin of the resonances observed in Fig. 3(b) and (d) we consider that the separation of the $\Delta l_e = \pm 1$ states for an *idealized* picture of the QDs studied in this work [13] was predicted to be of the order of ~3 meV. In a realistic picture, the lateral size, composition and strain will impact this energy, which describes the *intraband* state separation of the QDs. Since three Aharonov–Bohm oscillations are evident over the magnetic field range studied, and these transitions are resonant with an FIR energy of 2.1 meV, we suggest that the ODR observed is the result of the resonant activation and subsequent capture of electrons from intraband transitions of subsets of different QDs.

In summary, we have shown it is possible to observe strong ABE in *both* the intensity and energy of type-II ZnSe:Te QDs due to the unique geometry of this system. Furthermore, using FIR excitation resonances are evident

at 2.1 meV in the AB oscillations, which appear to result from activation of electrons from smaller ZnSe:Te QDs and their subsequent capture by larger ZnTe QDs.

References

[1] Y. Aharonov, D. Bohm, Phys. Rev. 115 (1959) 485.
[2] A.O. Govorov, et al., Phys. Rev. B. 66 (2002) 081309.
[3] M. Bayer, et al., Phys. Rev. Lett. 90 (2003) 186801.
[4] E. Ribeiro, et al., Phys. Rev. Lett. 92 (2004) 126402.
[5] M.G. Wright, et al., Semicond. Sci. Technol. 5 (1990) 438.
[6] I.L. Kuskovsky, et al., Phys. Rev. B 63 (2001) 155205.
[7] M. C-K. Cheung, et al., Appl. Phys. Lett., submitted for publication.
[8] M. Jo, et al., J.C.G. 301 (2007) 277.
[9] Q. Fu, et al., Phys. Rev. B 39 (1989) 3173.
[10] A. Muller, et al., Phys. Rev. B 73 (2006) 081306.
[11] Y. Gu, et al., Phys. Rev. B 71 (2005) 045340.
[12] V. Akimova, et al., Sov. Phys. Solid State 27 (1985) 1041.
[13] I.L. Kuskovsky, et al., Phys. Rev. B 76 (2007) 035342.
[14] J. Haetty, et al., Phys. Rev. B 56 (1997) 12364.
[15] I.R. Sellers, et al., Appl. Phys. Lett. 88 (2006) 081108.
[16] H.A. Nickel, et al., Phys. Rev. Lett. 88 (2002) 056801.
[17] C.J. Meining, et al., Phys. Rev. B 75 (2007) 165301.