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Determination of excitonic size with sub-nanometer precision via excitonic Aharonov-Bohm effect in type-II quantum dots

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A spectral analysis of the Aharonov-Bohm (AB) oscillation in magneto-photoluminescence intensity was performed for stacked type-II ZnTe/ZnSe quantum dots (QDs). Very narrow AB oscillations (~0.3 T) allowed for probing of both the lateral size distribution in the stack ensemble of QDs and the size of type-II excitons as determined by the electronic orbit with sub-nanometer precision. Two sets of stacks with excitonic size of 18.2 and 17.5 nm are determined to be present in the sample. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4721489]

In type-II semiconductor heterostructures spatial separation of the photo-generated electrons and holes occurs due to preferable band alignment. Knowledge of the size of such spatially indirect excitons is of fundamental importance. In type-I systems, for example, the excitonic size is often determined via magneto-photoluminescence (PL) studies. When excitons are subjected to external magnetic fields, for relatively weak fields, parabolic (diamagnetic) dependence of emission energy is observed and for strong fields that become linear (Landau level transitions).[1,2] One can then extract the excitonic size from the crossover of the two different field regimes (see, e.g., Refs. 3–5 and references therein).

In this letter, we show that for type-II quantum dots (QDs) with cylindrical symmetry (stacked in our case), the excitonic radius can be determined with very high accuracy via the Aharonov-Bohm (AB) effect.[6] Specifically we report the AB signature of two distinct sets of QDs stacks present in the sample and determined the excitonic size to be 18.2 and 17.5 nm, respectively.

The AB effect has been predicted[7,8] to manifest itself in optical emission of radially polarized excitons in nanorings and disk like type-II QDs, followed by experimental verification for both systems.[9–12] Although, in type-II QDs, excitons are particularly sensitive to the AB effects due to relatively larger spatial separation of the charged particles, in general, more reports are available for quantum ring systems[12–18] than for type-II QDs of suitable geometry.[9–11,19,20]

The AB phase reveals itself in magneto-PL of cylindrical type-II QDs via the change of the exciton ground state from a zero orbital angular momentum state ($|L=0\rangle$) to $|L\neq 0\rangle$ with increasing magnetic flux. This transition of the angular momentum to a non-zero value influences the optical properties in two ways:[7,8,10] (i) The ground state energy oscillates as the orbital angular momentum states cross and (ii) the PL intensity changes due to optical selection rules. Experimentally, the latter is often observed as one or more oscillations ("peaks"), which can arise due to such factors as QD shape anisotropy, e.g., elongation,[9] presence of impurities,[19,21] and/or built-in electric field.[15] In these cases, the excitonic states do not possess a definite value of the angular momentum, and the selection rules for optical transitions are relaxed.[9,19,21,22] Also, a "peak" in magneto-PL of disk-like type-II QDs (with one carrier strongly confined inside the dot, as the hole in our case) can appear in real experiments where the whole system is in the magnetic field, and the electron wavefunction is "squeezed" closer to the QD boundary,[20] leading to an increase in the electron-hole overlap, and thus increased PL intensity, which abruptly decreases when the electron changes its state from $|L=0\rangle$ to $|L\neq 0\rangle$ with increasing magnetic flux (field). In all cases[9,13,15,21] the maximum of the first oscillation occurs at the magnetic flux value $\Phi=\Phi_0/2$, where $\Phi_0=h/e$ is the flux quantum and h and e are the Planck's constant and the electron charge, respectively. Therefore, since in actual measurements one determines the magnetic field rather than the flux, the characteristic area enclosed by the orbiting exciton (dipole) can be obtained.

Considering that the stacking of QDs results in averaging out shape and size variations[9] and that at low temperature $|L=0\rangle$ dominates the optical properties,[21] we can estimate the excitonic size, using the following expression:

$$\pi R_e^2 B_{AB} = \Phi_0/2, \tag{1}$$

where B_{AB} is the magnetic field corresponding to the first oscillation peak and R_e is the radius of electronic orbit that gives the size of the type-II polarized exciton.

To determine the excitonic size for various QD stacks in the system, we spectrally resolved the AB oscillations in the optical emission of the stacked ZnTe/ZnSe QDs. Previously,[9,11,15,19,22] oscillations in the integrated PL intensity have been commonly demonstrated; this can be thought as an overall averaged interpretation of the emission from all the emission centers that contribute to the PL spectra. A spectral study, on the other hand, can reveal the properties at particular spectral positions and thus can be used as a fine probe to study contributions from different centers present in ensemble systems.

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High resolution PL measurements (at zero magnetic fields) were done with a TriVista SP2 500i Triple monochromator coupled with a thermoelectrically cooled CCD camera. The optical excitation was achieved with the 351 nm line of an Ar-ion laser. The magneto-PL experiments were performed with a Cryo Industries of America 9 T superconducting magnet outfitted with fiber optic probe, used to excite and collect the PL. The detection for magneto-PL experiments was done by a portable high resolution ocean optics solid state spectrometer.

The samples studied are stacked type-II ZnTe/ZnSe QDs grown via migration enhanced epitaxy using three submonolayer deposition cycles of Zn-Te-Zn sandwiched between nominally undoped ZnSe barriers. Details of growth and optical analysis of similar samples were reported elsewhere (see Refs. 23–25 and references therein). In these systems the PL of the QDs (generally seen as a broad "green band" with peak energy of ~2.5 eV, sometimes with a low energy shoulder[11] and as low as 2.3 eV (Refs. 25 and 26) is convoluted with the emission from excitons bound to isoelectronic centers (ICs) of various sizes ($Te_{n\geq 2}$).[23] The samples studied here exhibit a high degree of separation of QD containing layers from the barriers as observed by the presence of the sharp band edge emission (Fig. 1), which previously was seen *only* in samples grown with a single Zn-Te-Zn MEE cycle.[27] As expected, this sample exhibited the AB oscillation in the integrated PL intensity, as shown in the inset of Fig. 1. The full width at the half maximum (FWHM) of the integrated intensity oscillation in the magnetic field is ~0.3 T.

In Figs. 2(a) and 2(b), we show the spectral analysis of the magneto-PL in this sample. We traced the values of B_{AB} at energies across the PL spectrum. The PL spectrum for the sample taken under experimental condition of magneto-PL is shown in Fig. 2(b), which is shown to relate the spectral behavior of B_{AB} (Fig. 2(a)). The observed B_{AB} changes from a lower value of ~1.98 T at the lower energy side (below ~2.50 eV) to a higher value of ~2.15 T at the higher energy side (above ~2.54 eV), and these variations are measurable beyond the experimental error. The transition in B_{AB} is very clear and suggests an existence of two sets of QDs. Using the expression for the transition field (Eq. (1)) we estimate the size of the type-II exciton (which coincides with radius of the electronic orbit, as discussed above) for each set of QDs from the value of B_{AB} to be ~18.2 and ~17.5 nm for the lower and higher energy sides of the PL spectrum, respectively. We, thus, have measured spatial component of the excitonic wavefunction with sub-nanometer accuracy.

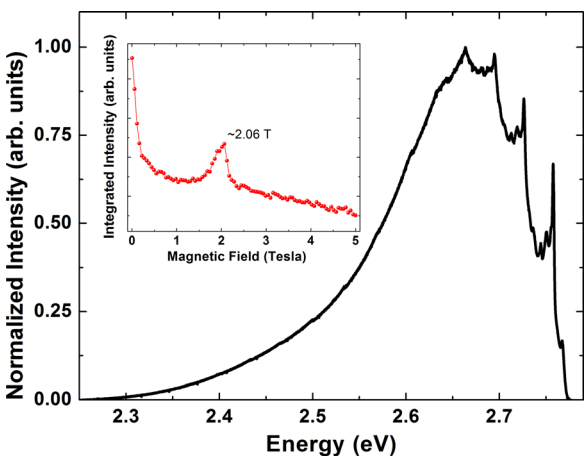

FIG. 1. Low temperature (T = 10 K) high resolution PL of type-II QD sample grown with three Zn-Te-Zn sequences. Inset: integrated PL intensity as a function of magnetic field; the peak at 2.06 T is due to the excitonic AB effect.

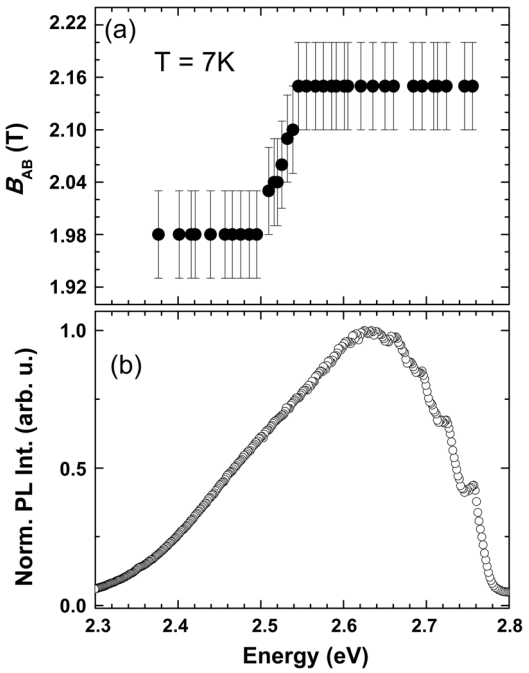

FIG. 2. (a) Spectral dependence of B_{AB}; (b) PL spectrum. Note the high energy shoulders in (b) are seen as sharp lines in higher resolution measurements shown in Fig. 1.

In summary, we gained insight into the lateral size distribution of QDs in stacked type-II ZnTe/ZnSe system via a spectral study of the excitonic AB oscillation in PL intensity that could distinguish the presence of two sets of QDs stacks. We also determined the characteristic size of type-II polarized exciton for each set with sub-nanometer precision. The robust AB oscillations in our material system arising due to stacked nature of cylindrical QDs suggest that such a system can be a good candidate for quantum information related applications.

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[1]R. Rinaldi, P. V. Giugno, R. Cingolani, H. Lipsanen, M. Sopanen, J. Tulkki, and J. Ahopelto, Phys. Rev. Lett. **77**, 342 (1996).
[2]T. Wimbauer, K. Oettinger, A. L. Efros, B. K. Meyer, and H. Brugger, Phys. Rev. B **50**, 8889 (1994).
[3]R. Provoost, M. Hayne, V. V. Moshchalkov, M. K. Zundel, and K. Ebert, Appl. Phys. Lett. **75**, 799 (1999).
[4]A. Polimeni, S. T. Stoddart, M. Henini, L. Eaves, P. C. Main, K. Uchida, R. K. Hayden, and N. Miura, Physica E **2**, 662 (1998).

[5]B. Bansal, S. Godefroo, M. Hayne, G. Medeiros-Ribeiro, and V. V. Moshchalkov, Phys. Rev. B **80**, 205317 (2009).
[6]Y. Aharonov and D. Bohm, Phys. Rev. **115**, 485 (1959).
[7]A. O. Govorov, S. E. Ulloa, K. Karrai, and R. J. Warburton, Phys. Rev. B **RC66**, 081309 (2002).
[8]A. B. Kalametsev, V. M. Kovalev, and A. O. Govorov, JETP Lett. **68**, 669 (1998).
[9]I. L. Kuskovsky, W. MacDonald, A. O. Govorov, L. Muroukh, X. Wei, M. C. Tamargo, M. Tadic, and F. M. Peeters, Phys. Rev. B **76**, 035342 (2007).
[10]E. Ribeiro, A. O. Govorov, W. Carvalho, Jr., and G. Medeiros-Ribeiro, Phys. Rev. Lett. **92**, 126402 (2004).
[11]I. R. Sellers, V. R. Whitesides, I. L. Kuskovsky, A. O. Govorov, and B. D. McCombe, Phys. Rev. Lett. **100**, 136405 (2008).
[12]M. Bayer, M. Korkusinski, P. Hawrylak, T. Gutbrod, M. Michael, and A. Forchel, Phys. Rev. Lett. **90**, 186801 (2003).
[13]A. M. Fischer, J. V. L. Campo, M. E. Portnoi, and R. A. Romer, Phys. Rev. Lett. **102**, 096405 (2009).
[14]B. Li and F. M. Peeters, Phys. Rev. B **83**, 115448 (2011).
[15]M. D. Teodoro, V. L. Campo, V. Lopez-Richard, E. Marega, G. E. Marques, Y. G. Gobato, F. Iikawa, M. J. S. P. Brasil, Z. Y. AbuWaar, V. G. Dorogan, Y. I. Mazur, M. Benamara, and G. J. Salamo, Phys. Rev. Lett. **104**, 086401 (2010).
[16]V. M. Fomin, V. N. Gladilin, S. N. Klimin, J. T. Devreese, N. A. J. M. Kleemans, and P. M. Koenraad, Phys. Rev. B **76**, 235320 (2007).
[17]F. Palmero, J. Dorignac, J. C. Eilbeck, and R. A. Römer, Phys. Rev. B **72**, 075343 (2005).
[18]A. V. Maslov and D. S. Citrin, Phys. Rev. B **67**, 121304 (2003).
[19]M. H. Degani, M. Z. Maialle, G. Medeiros-Ribeiro, and E. Ribeiro, Phys. Rev. B **78**, 075322 (2008).
[20]K. L. Janssens, B. Partoens, and F. M. Peeters, Phys. Rev. B **64**, 155324 (2001).
[21]L. G. G. V. Dias da Silva, S. E. Ulloa, and A. O. Govorov, Phys. Rev. B **70**, 155318 (2004).
[22]S. Miyamoto, O. Moutanabbir, T. Ishikawa, M. Eto, E. E. Haller, K. Sawano, Y. Shiraki, and K. M. Itoh, Phys. Rev. B **82**, 073306 (2010).
[23]Y. Gu, I. L. Kuskovsky, M. van der Voort, G. F. Neumark, X. Zhou, and M. C. Tamargo, Phys. Rev. B **71**, 045340 (2005).
[24]I. L. Kuskovsky, Y. Gu, M. van der Voort, G. F. Neumark, X. Zhou, M. Munoz, and M. C. Tamargo, Phys. Stat. Sol. **241**, 527 (2004).
[25]B. Roy, A. Shen, M. Tamargo, and I. Kuskovsky, J. Electron. Mater. **40**, 1775 (2011).
[26]M. C. K. Cheung, A. N. Cartwright, I. R. Sellers, B. D. McCombe, and I. L. Kuskovsky, Appl. Phys. Lett. **92**, 032106 (2008).
[27]I. L. Kuskovsky, Y. Gong, G. F. Neumark, and M. C. Tamargo, Superlattices Microstruct. **47**, 87 (2010).