Biplob Barman

Time-resolved magnetophotoluminescence studies of magnetic polaron dynamics in type-II quantum dots

B. Barman,[1] R. Oszwałdowski,[1,2] L. Schweidenback,[1] A. H. Russ,[1] J. M. Pientka,[1,3] Y. Tsai,[1] W-C. Chou,[4] W. C. Fan,[4] J. R. Murphy,[1] A. N. Cartwright,[5] I. R. Sellers,[6] A. G. Petukhov,[2] I. Žutić,[1] B. D. McCombe,[1] and A. Petrou[1]

[1]*Department of Physics, University at Buffalo SUNY, Buffalo, New York 14260, USA*

[2]*Department of Physics, South Dakota School of Mines & Technology, Rapid City, South Dakota 57701, USA*

[3]*Department of Physics, St. Bonaventure University, St. Bonaventure, New York 14778, USA*

[4]*Department of Electro-physics, National Chiao Tung University, Hsinchu 300, Taiwan*

[5]*Department of Electrical & Electronic Engineering, University at Buffalo SUNY, Buffalo, New York 14260, USA*

[6]*Department of Physics and Astronomy, University of Oklahoma, Norman, Oklahoma 73019, USA*

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We used continuous wave photoluminescence (cw-PL) and time-resolved photoluminescence (TR-PL) spectroscopy to compare the properties of magnetic polarons (MP) in two related spatially indirect II-VI epitaxially grown quantum dot systems. In the ZnTe/(Zn,Mn)Se system the holes are confined in the nonmagnetic ZnTe quantum dots (QDs), and the electrons reside in the magnetic (Zn,Mn)Se matrix. On the other hand, in the (Zn,Mn)Te/ZnSe system, the holes are confined in the magnetic (Zn,Mn)Te QDs, while the electrons remain in the surrounding nonmagnetic ZnSe matrix. The magnetic polaron formation energies E_{MP} in both systems were measured from the temporal redshift of the band-edge emission. The magnetic polaron exhibits distinct characteristics depending on the location of the Mn ions. In the ZnTe/(Zn,Mn)Se system the magnetic polaron shows conventional behavior with E_{MP} decreasing with increasing temperature T and increasing magnetic field B. In contrast, E_{MP} in the (Zn,Mn)Te/ZnSe system has unconventional dependence on temperature T and magnetic field B; E_{MP} is weakly dependent on T as well as on B. We discuss a possible origin for such a striking difference in the MP properties in two closely related QD systems.

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I. INTRODUCTION

Quantum dots (QDs), also known as artificial atoms, can allow versatile control of the number of carriers, their spins, Coulomb interactions, and quantum confinement [1–4]. Compared to their bulk counterparts [5–9], magnetically doped semiconductor QDs could provide control of the magnetic ordering [10–16] with the onset of magnetization at substantially higher temperatures [17–21]. Experiments typically focus on Mn-doped II-VI and III-V QDs, in which it is possible to include both single [22–25] and several magnetic impurities [17–21,26–40], having similarities with nuclear spins [41,42]. In the first case (single magnetic ion), such systems could be considered as potential quantum bits, quantum memories, or probes to detect an unconventional orbital ordering [17,23–25,43]. In the second case, the presence of several magnetic ions can lead to the formation of a magnetic polaron (MP), a long-standing research topic in magnetic semiconductors [5–8,44].

MP can be viewed as a cluster of localized magnetic ion spins, aligned through an exchange interaction with the spin of a confined carrier. Initial studies in bulk systems involved MPs with a carrier spin bound to an impurity center (donor or acceptor) [8]. In contrast, in semiconductor nanostructures with reduced dimensionality, the confinement removes the need for the presence of impurities and enhances the stability of the MPs [7]. As depicted in Fig. 1, after a sufficiently long time interval after photoexcitation (comparable to MP formation time), Mn spins in II-VI systems typically couple ferromagnetically with electron spins and antiferromagnetically with hole spins. The simultaneous presence of carriers and Mn ions in QDs result in the formation of MP through lowering of the exciton energy by an amount as shown in Fig. 1. Two main classes of magnetic QDs have been investigated; those grown using molecular beam epitaxy (MBE) [7,17,20–25,29–35,39] and those that are solution processed, known as colloidal QDs [12,26–28,36,37,40]. Despite entirely different growth procedures, in both classes of QDs the MP formation is associated with the observed magnetic ordering [12,20–22,30,37]. Several interesting effects have been attributed to MPs in nanostructures, such as the long "spin memory" times in (Cd,Mn)Te QDs [30], giant magnetoresistance in ErAs:GaAs nanocomposites [45], and room-temperature ferromagnetic ordering in MnGe QDs [21]. The temporal evolution of the MP (Fig. 1) can be studied using time-resolved photoluminescence spectroscopy (TR-PL) [39]. In these experiments large (tens of meV) redshifts of the photoluminescence (PL) peak energy are observed as a function of time delay between laser excitation and PL detection.

The majority of published work describe studies of magnetic QDs with type-I band alignment [7,39], where the location of electrons and holes coincide spatially. In this work we investigate TR-PL measurements in QD structures with type-II band alignment where the holes are confined in the QDs while the electrons reside in the surrounding matrix. Schematic diagrams of type-I and type-II alignment are shown in Figs. 2(a) and 2(b), respectively. We have studied two closely related ZnTe/ZnSe QD systems, grown using the same MBE process. In sample 1, Mn^{2+} ions are incorporated in the matrix: ZnTe/(Zn,Mn)Se, while in sample 3, Mn^{2+} ions are in the QDs region: (Zn,Mn)Te/ZnSe.

These type-II structures offer two clear advantages for the study of MP dynamics over type-I QDs: (i) In type-II QDs, the photoexcited electrons and holes are spatially

