Remanent Zero Field Spin Splitting of Self-Assembled Quantum Dots in a Paramagnetic Host

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We report on the observation of a finite spin splitting at zero magnetic field in resonant tunneling experiments on CdSe self-assembled quantum dots in a (Zn,Be,Mn)Se barrier. This is remarkable since bulk II-VI dilute magnetic semiconductors are paramagnets. Our experiment may be viewed as tunneling through a single magnetic polaron, where the carriers contained inside the dot act to mediate an effective ferromagnetic interaction between Mn ions in their vicinity. The effect is observable up to relatively high temperatures, which we tentatively ascribe to a feedback mechanism with the electrical current, previously predicted theoretically.

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Nanomagnetics has, over the past few years, produced a series of fascinating and often unanticipated phenomena. To name a few, molecular magnets exhibit quantum tunneling of the magnetization [1], magnetic atoms on a surface exhibit giant magnetic anisotropies [2], and the harnessing of magnetic domain walls as data carriers [3]. Here, we report on another remarkable phenomenon: self-assembled quantum dots fabricated from II-VI semiconductor possess a remanent magnetization at zero external field due to the local interaction of free carriers in the dots with nearby Mn in the surrounding dilute magnetic semiconductor (DMS) barrier to form a magneto-polaron type complex, which would not be stable in bulk material, but is stabilized by zero dimensional impurities such as quantum dots [4]. This novel magnetic behavior is both scientifically intriguing, and potentially technologically relevant, as the effect may eventually allow the operation of dots as voltage-controlled spin filters, capable of spin-selective carrier injection and detection in a semiconductor spintronics device. We describe here the experimental observation of this behavior in a sample based on the incorporation of nonmagnetic CdSe self-assembled quantum dots (SADs) in paramagnetic (Zn,Be,Mn)Se and present a qualitative model which may explain the effect.

We previously demonstrated a prototype of a voltage-controlled spin filter using a II-VI DMS-based resonant tunneling diode (RTD) [5]. Since II-VI DMS are paramagnets, the spin filtering in this device required an external magnetic field. RTDs based on ferromagnetic III-V semiconductors like (Ga,Mn)As would not require an external field. However, this material is not suitable for resonant tunneling devices due to the short mean free path of holes [6]. Recent theoretical works [4,7–9] have suggested that spin selection may be achievable in II-VI DMS without any external magnetic field by creating localized carriers that might mediate a local ferromagnetic interaction between nearby Mn atoms.

Our sample is an MBE-grown all-II-VI RTD structure consisting of a single 9 nm thick semimagnetic $Zn_{0.64}Be_{0.3}Mn_{0.06}Se$ tunnel barrier, sandwiched between gradient doped $Zn_{0.97}Be_{0.03}Se$ injector and collector. Embedded within the barrier are 1.3 monolayers of CdSe. The lattice mismatch between the CdSe and the $Zn_{0.64}Be_{0.3}Mn_{0.06}Se$ induces a strain in the CdSe material, which is relaxed by the formation of isolated CdSe dots [10]. The full layer stack is given in Fig. 1. Standard optical lithography techniques were used to pattern the structure into 100 μm square pillars, and contacts were applied to the top and bottom ZnSe layers in order to perform transport measurements vertically through the layer stack. More details of the fabrication procedure are given in Ref. [5]. From the size of the pillars, and the typical dot density, one would expect some million dots within our device. However, despite this number, transport through similar III-V SAD-RTDs is usually dominated by only a few dots that come into resonance at lower bias voltages [11–13]. We therefore interpret the low bias transport through our sample as corresponding to electrons tunneling from the injector through a single quantum dot (QD) as schemati-

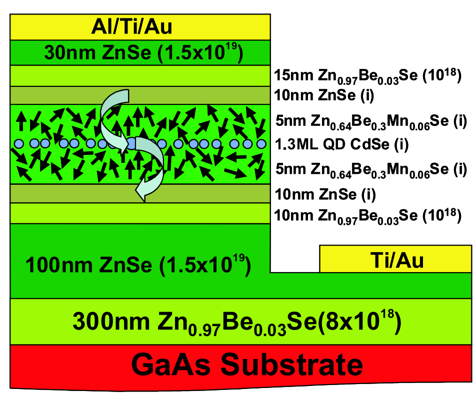

FIG. 1 (color online). Layer structure of the device, and schematic of the transport mechanism. Electrons tunneling through the dot mediate a local magnetic interaction aligning nearby Mn ions.

cally depicted in Fig. 1. From calculations of energy levels of strained QD, we find several populated QD levels at zero bias. Hence the electrons tunnel through excited states of QD containing a finite number of electrons.

Figure 2 shows a full current-voltage curve up to a bias voltage of 170 mV. A first feature is observed at a bias of 55 mV, associated with the first dot coming into resonance. At bias voltages above 100 mV, several resonances due to the ensemble of QD can also be observed. We will first focus on the low bias feature which is shown in the inset to the figure. These more detailed curves taken at 0 and 4 T clearly show a complex structure consisting of four distinct peaks, which evolve with the magnitude of the applied magnetic field. We verified that the evolution of the features does not appreciably depend on the direction of the magnetic field, indicating that the magnetic response of the system cannot be associated with artefacts such as two-dimensional states in the injector or wetting layer [5,11–13], and that it must be a property of the dot or the barrier. We also verified that the sample does not exhibit any magnetic hysteresis and that the main characteristics of the data are robust under multiple thermal cycling.

A better understanding of the evolution of the features with magnetic field can be obtained from Fig. 3. In Fig. 3(a), we plot the current through the device as a color-scale surface with respect to bias voltage and magnetic field. This puts into evidence two very important features of the data. First, that as the magnetic field is increased, features split apart with a behavior reminiscent of the Brillouin function, and second, that the splitting remains finite in zero external magnetic field. The same behavior can be seen for many of the higher bias resonances presented in Fig. 3(b) at 1.3, 4.2, and 10 K. Note that while the Brillouin-like behavior shows the expected temperature dependence, the $B = 0$ splitting is temperature independent. The first of these effects, that the levels should split following a Brillouin function, is not all that surprising as it was previously observed in III-V devices that resonances split in a magnetic field following the Landé g factor of the dot material [13].

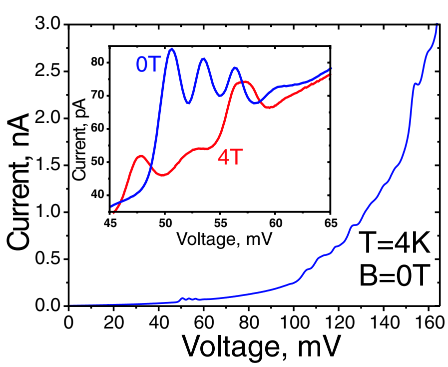

FIG. 2 (color online). Current-voltage characteristic of the device, with a high resolution view of the first resonance feature in the inset.

The main difference here is that the barrier, and not the dot, is magnetic. Since the effect of giant Zeeman splitting on the height of the barriers is negligible, the presence of Mn has little effect on the barrier properties. However, given that electrons are not perfectly localized in the dots, but rather have wave functions extending into the barrier, it is not surprising that the levels in the QD spin split following the magnetization of the Mn in the barriers, yielding results reminiscent of those observed [5] for tunneling through a dilute magnetic quantum well. We note in passing that experience has shown a strong out-diffusion mechanism when attempting to incorporate Mn into CdSe dots in a ZnSe lattice. We therefore believe that diffusion of Mn into our dots is unlikely, and thus that the Mn is located exclusively on the outskirts of the dot. Direct verification

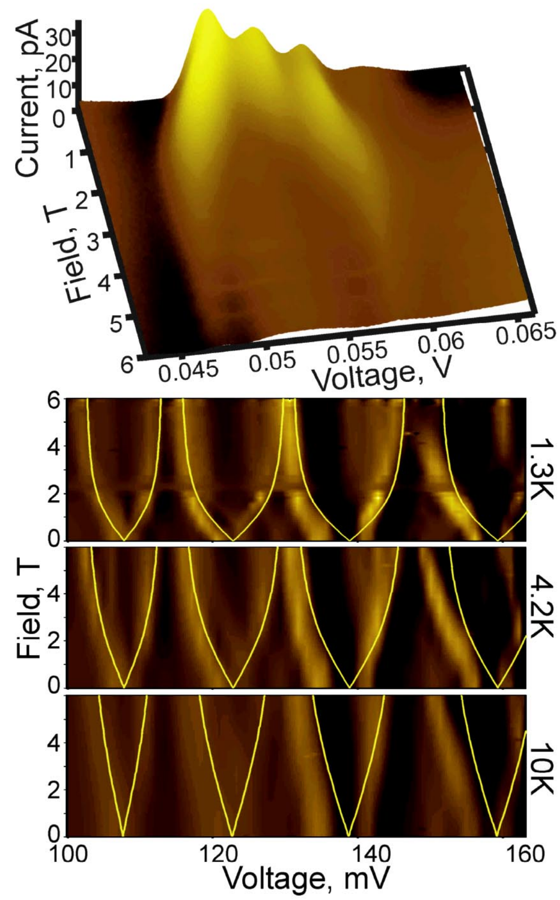

FIG. 3 (color online). (a) Surface plot of the current through the device near the first resonance feature, as a function of magnetic field and bias voltage. (b) Color scale image as a function of magnetic field and voltage for higher bias resonances. Since these higher resonances are weaker and on a significant background, the color scale in (b) is proportional to the voltage derivative of the current in order to better resolve the position of the resonances. In both (a) and (b), the data at higher magnetic fields clearly is Brillouin-like, as evidenced by the lines plotting Brillouin functions in (b). However, at fields below 500 mT, the behavior departs from a Brillouin function, with the splitting becoming constant and remaining finite even at zero field.

of this hypothesis is, however, impossible, so Mn in the dots cannot be fully ruled out. Within our picture, the exact location of the Mn is secondary, changing the coupling constants quantitatively, but not affecting the qualitative model.

The *splitting remaining finite at* $B = 0$ is, however, very surprising, since there is *a priori* nothing ferromagnetic in the sample. This observation is the central point of this Letter. It can be qualitatively understood by considering the effect of interactions between electrons in the dot and the Mn atoms in the vicinity of the dot [14].

Electrons populate QD levels following the Pauli principle and Hunds rules [15] whenever there is orbital degeneracy. For a parabolic dot, the total electron spin follows the sequence $S = \{1/2, 0, 1/2, 1, 1/2, 0, 1/2, 1, 3/2, \ldots\}$ with increasing electron number. Hence, the total spin of the dot is finite for almost all electron numbers. The interaction of this total net spin with the spin of Mn ions induces an effective ferromagnetic Mn-Mn interaction. Consider the total Hamiltonian of the electronic and Mn system [4,7,15,16]:

$$H = H_e + g^* \mu_B \vec{B} \cdot \sum_i \vec{S}_i - J_C \sum_{\vec{R},i} \vec{M}_{\vec{R}} \cdot \vec{S}_i \delta(\vec{r}_i - \vec{R}) + \sum_{\vec{R}} g_{Mn} \mu_B \vec{B} \cdot \vec{M}_{\vec{R}} + \frac{1}{2} \sum_{\vec{R},\vec{R}'} J_{\vec{R},\vec{R}'} \vec{M}_{\vec{R}} \cdot \vec{M}_{\vec{R}'}.$$

Here $\vec{M}_{\vec{R}}$ is the spin of Mn ions ($M = 5/2$) at position $\vec{R}$; S_i is the spin of the ith electron ($S = 1/2$). J_c is the s-d exchange constant between the conduction electrons and the Mn d electrons and $J_{RR'}$ is the antiferromagnetic Mn-Mn interaction. The first term is the spin independent Hamiltonian of electrons in a QD and a magnetic field, interacting via a pairwise potential. The full interaction between electron spins and Mn ions in the barrier is an extremely complicated problem. We restrict ourselves here to a demonstration that the electron spin is capable of compensating the antiferromagnetic interaction among Mn ions and lead to their ferromagnetic arrangement. We consider only a single electron in the ground state and in the absence of external magnetic field. The effective spin Hamiltonian reads:

$$H = E_0 - J_C \sum_{\vec{R}} |\Phi(R)|^2 \vec{M}_{\vec{R}} \cdot \vec{S} + \frac{1}{2} \sum_{\vec{R},\vec{R}'} J_{\vec{R},\vec{R}'} \vec{M}_{\vec{R}} \cdot \vec{M}_{\vec{R}'}, \quad (1)$$

where E_0 is the electron energy and $|\Phi(R)|^2$ is the probability of finding an electron at position $\vec{R}$ of a Mn ion. Even for such a simplified Hamiltonian the number of configurations is very large in the number of Mn ions.

The physics of the Mn-Mn interactions mediated by an electron spin can, however, be understood by examining an exactly solvable problem of two antiferromagnetically coupled Mn ions [17]. The energy spectrum of the coupled Mn-spin system is characterized by the total spin $J = M \pm 1/2$ where M is the total Mn spin and the $\pm 1/2$ corresponds to the directions of the electron spin. The evolution of the energy of the system as a function of the total Mn spin depends on the direction of the electron spin as follows:

$$E(M, +) = -\left(\frac{\hat{J}_C}{2}\right) M + \left(\frac{J_{NN'}}{2}\right)\left[M(M+1) - \frac{35}{2}\right]$$

$$E(M, -) = \left(\frac{\hat{J}_C}{2}\right)(M+1) + \left(\frac{J_{NN'}}{2}\right)\left[M(M+1) - \frac{35}{2}\right]$$

as shown in Fig. 4(a). In the absence of coupling to the electron spin ($J_c = 0$), it is obvious that the minimum energy state for either electron spin corresponds to the total Mn spin $M = 0$, i.e., an antiferromagnetic arrangement. However, as shown in Fig. 4(a), with coupling to the electron spin, the $E(M, +)$ ground state of the combined system has finite total Mn spin $M^* = (\frac{\hat{J}_C}{J_{NN'}} - 1)/2$. To estimate the value of M^* we approximate our QD by a sphere of radius $R = 4$ nm and a barrier potential of 1 eV estimated from strain and the Bir-Pikus Hamiltonian. The effective electron-Mn exchange interaction for Mn on the surface of the sphere is then $\hat{J}_C = J_C|\Phi(R)|^2 = 4.5$ μeV. For a typical Mn separation in the barrier of $R_{12} = 1.2$ nm, we estimate the antiferromagnetic interaction strength

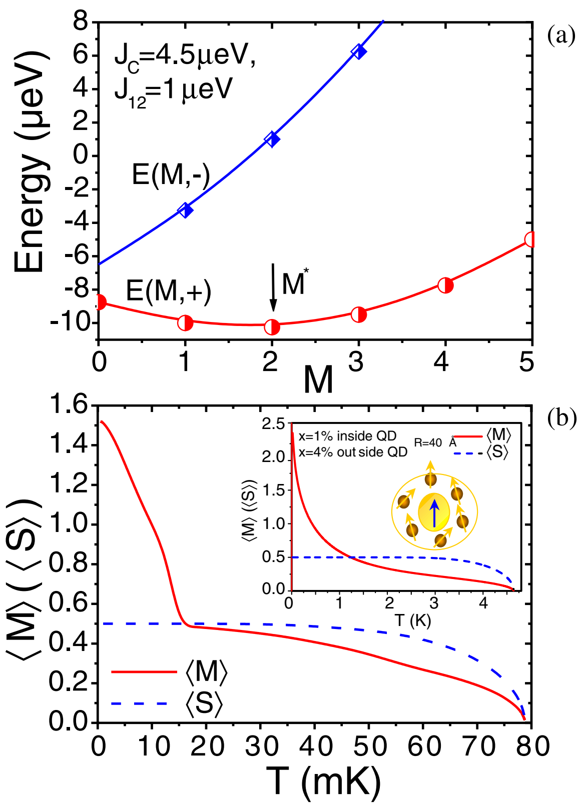

FIG. 4 (color online). (a) The energy levels $E[+, M]$, $E[-, M]$ for two different electron spin orientation as a function of total spin of two Mn ions M localized on the surface of a spherical quantum dot and (b) average magnetization as a function of temperature of a samples with 4% Mn ions randomly distributed on a surface of a 4 nm radius spherical quantum dot. The inset shows the same with 1% Mn ions inside the dot.

$J_{12} = 1\ \mu$eV. Hence for our model system we find $M^* = 2$ and the coupling to the electron spin aligns spins of nearest neighbor Mn ions. Independent mean field calculations of some 100 Mn ions randomly distributed in a QD confirm the existence of ferromagnetic ordering of Mn in the vicinity of the QD [8]. In Fig. 4(b), we show the calculated averaged Mn and electron spin magnetization as a function of temperature for Mn ions localized in the barrier surrounding a spherical CdSe QD. We find the existence of the magnetic polaron, with the Mn magnetization decaying as one moves away from the quantum dot. These findings are in agreement with previous calculations of magnetic polarons [4,7,8,10], and for reasonable parameters for our system show that the presence of electrons in the dot will mediate a local ferromagnetic interaction between Mn atoms near this dot.

The model allows a plausible interpretation of our experimental observations: electrons localized in the dot mediate a local ferromagnetic interaction which causes a finite spin splitting even in the absence of an external applied field. Our experiment is therefore tantamount to measuring transport through a single magnetic polaron. The local interaction has a strength corresponding to an effective field of the order of some hundreds of mT, and can be randomly oriented. When an external magnetic field is applied, the ferromagnetic order will first rotate towards the direction of the applied field, but this will have no effect on the transport, which explains why the resonance positions are independent of magnetic field for fields below ~500 mT. However, as the field is further increased, it will start to dominate, and the spin splitting will grow following the normal paramagnetic interaction of the dilute Mn system [5]. A question remains as to why the zero magnetic field splitting is observed in our transport experiment at relatively high temperatures while theory predicts only a low temperature effect. Moreover, the optical measurements of Ref. [18,19] did not show the effect. This remains an open issue which will require further investigations to clarify. One possible explanation for the lack of temperature sensitivity could result from interdiffusion of Mn into the dots. As shown in the inset of Fig. 4, this would enhance the strength of the alignment due to the greater wave function overlap. As mentioned previously, we view such interdiffusion as very unlikely, and this, coupled with the fact that we observe similar finite splitting for the higher bias peaks as well, leads us to believe that this is not a likely explanation. As an alternative, we tentatively suggest that a second effect, which was predicted in Ref. [4,7], may be acting here. Once current begins flowing through the dot, a feedback mechanism sets in where polarization of the Mn spins leads to polarization of the current which in turn enhances the polarization of Mn spins which in turn enhances the polarization of the current. While the extension of this feedback mechanism from the single spin considered in Ref. [7], to a magneto-polaron involving a finite number of electrons and an aggregate of Mn spins is certainly nontrivial, certain aspects of the modeling can be intuitively applied. It is clear that the Mn ions will be subject to an alignment mechanism stemming from the current traversing the dot, and that this will introduce an additional energy scale into the problem which can also compete against the randomizing effect of temperature. Moreover, the strength of this effect increases with increasing spin, and thus could be very significant in our case where the aggregate of Mn ions can reach a large total spin. The results of Ref. [4] are supportive of this assumption. This dynamical effect also explains why spin polarization is observed at relatively high temperature.

In conclusion, we have shown that electrons in a quantum dot can mediate a local ferromagnetic interaction in a surrounding dilute Mn system, leading to a finite energy splitting of spin levels in the dot in the absence of an external magnetic field. While there are still some outstanding issues in fully understanding this observation, particularly with regards to the temperature behavior, we believe that our results are an important first step in understanding a new transport phenomena.

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