Self Assembled II-VI Magnetic Quantum Dot as a Voltage-Controlled Spin-Filter.

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A key element in the emergence of a full spintronics technology is the development of voltage controlled spin filters to selectively inject carriers of desired spin into semiconductors. We previously demonstrated a prototype of such a device using a II-VI dilute-magnetic semiconductor quantum well which, however, still required an external magnetic field to generate the level splitting. Recent theory suggests that spin selection may be achievable in II-VI paramagnetic semiconductors without external magnetic field through local carrier mediated ferromagnetic interactions. We present the first experimental observation of such an effect using non-magnetic CdSe self-assembled quantum dots in a paramagnetic (Zn,Be,Mn)Se barrier.

Nanomagnetics has over the past few years produced a series of fascinating and often unanticipated phenomena. To name a few, molecular magnets exhibit quantum tunnelling of the magnetization\textsuperscript{1}, magnetic atoms on a surface exhibit giant magnetic anisotropies\textsuperscript{2}, and magnetic domain walls are being harnessed as data carriers\textsuperscript{3}. Here, we report on another remarkable phenomenon: self-assembled quantum dots, fabricated from II-VI dilute magnetic semiconductors (DMS) that macroscopically exhibit paramagnetism, possess a remanent magnetization at zero external field. This allows us to operate the dots as voltage controlled spin filters, capable of spin-selective carrier injection and detection in semiconductors. Such spin filter devices could provide a key element in the emergence of a full spintronics technology\textsuperscript{4}. We present the first experimental observation of such a device using an approach based on the incorporation of non-magnetic CdSe self-assembled quantum dots (SADs) in paramagnetic (Zn,Be,Mn)Se.

We previously demonstrated a prototype of such a spin filter using a II-VI DMS-based resonant tunnelling diode\textsuperscript{5}. However, while that device was tuned by a bias voltage, the spin filtering mechanism still required an external magnetic field. Moreover, ferromagnetic III-V semiconductors like (Ga,Mn)As are not suitable for resonant tunnelling devices due to the short mean free path of holes\textsuperscript{6}. Recent theoretical works\textsuperscript{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 resonant tunnelling diode (RTD) structure consisting of a single 9 nm thick semi-magnetic Zn\textsubscript{0.64}Be\textsubscript{0.3}Mn\textsubscript{0.06}Se tunnel barrier, sandwiched between gradient doped Zn\textsubscript{0.97}Be\textsubscript{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\textsubscript{0.64}Be\textsubscript{0.3}Mn\textsubscript{0.06}Se induces a strain in the CdSe material, which is relaxed by the formation of isolated CdSe dots\textsuperscript{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. \textsuperscript{11}. From the size of the pillars, and the typical density of the dots, 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\textsuperscript{11-13}. We therefore interpret the low bias transport through our sample as corresponding to electrons tunnelling from the injector into a single quantum dot and out of the dot into the collector as schematically depicted in Fig. 1. Based on calculations of energy levels of strained quantum dots, we find several quantum dot levels populated by electrons at zero bias. Hence the electrons tunnel through excited states of quantum dots containing a finite number of electrons.

The experiments were carried out in a magnetocryostat and studied at temperatures down to 1.3 K and in fields from 0 to 6 T. Fig. 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 voltage above 100 mV, several resonances due to the ensemble of dots 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 4T clearly show that the feature actually has 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\textsuperscript{6,11-13}, 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.
A better understanding of the evolution of the features with magnetic field can be obtained from Fig. 3(a). In Fig. 3(a), we plot the current through the device as a colour-scale surface with respect to bias voltage and magnetic field. This puts into evidence two very important features of the data. Firstly, that as the magnetic field is increased, features split apart with a behaviour reminiscent of the Brillouin function, and secondly, that the splitting remains finite in zero external magnetic field. The same behaviour can be seen for many of the higher bias resonances presented in Fig. 3(b). The first of these effects, that the levels should split following a Brillouin function is not all that surprising. It was previously observed in III-V devices that resonances split in a magnetic field following the Landé g factor of the material in the dots [13].

The main difference here is that in the present experiment, it is the barrier, and not the dots that are mag-
netic. Since the effect of the giant Zeeman splitting on the height of the barriers is negligible, the presence of Mn should have little effect on the barrier properties. However, given that electrons are not perfectly localized in the dots, but rather have wave functions which extend into the barrier, it is not surprising that the quantum levels in the dots spin-split following the magnetization of the Mn in the barriers, yielding results reminiscent of those previously observed [2] for the case of tunnelling through a dilute magnetic quantum well.

The observation that the splitting remains finite at $B=0$ is however more surprising since there is a priori nothing ferromagnetic in the sample. This observation can be understood by considering the effect of interactions between electrons in the dot and the Mn atoms in the vicinity of the dot.

Electrons populate quantum dot levels according to the Pauli exclusion principle, and Hund’s rules [12] whenever there is orbital degeneracy. For a parabolic dot, the total electron spin follows the sequence $S = \{1/2,1/2,1/2,1/2,1/2,1/2,1/2\}$ with increasing electron numbers. Hence, for almost all electron numbers, the total spin of the dot is finite. The interaction of this total net spin with the spin of Mn ions induces an effective ferromagnetic Mn-Mn interaction. This can be seen by considering the total Hamiltonian of the electronic and Mn system [6, 7, 12, 16]:

$$H = H_e + g^\ast \mu_B \vec{B} \cdot \sum_i \vec{S}_i - J_{C} \sum_{\vec{R}, \vec{R}'} \vec{M}_{\vec{R}} \cdot \vec{S}_i \delta(\vec{r}_i - \vec{R}) + \sum_{\vec{R}} g_{MN} \mu_B \vec{B} \cdot \vec{M}_{\vec{R}} + \frac{1}{2} \sum_{\vec{R}, \vec{R}'} J_{RR'} \vec{M}_{\vec{R}} \cdot \vec{M}_{\vec{R}'}$$

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 $i$-th electron ($S=1/2$), $J_C$ is the sp-d exchange constant between the conduction electrons and the d-electrons of the Mn shell and $J_{RR'}$ is the antiferromagnetic Mn-Mn interaction. The first term is the spin independent Hamiltonian of electrons confined to a quantum dot in a magnetic field, and interacting via a pair wise 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 anti-ferromagnetic 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, leaving the problem of interacting many-electron dots for future analysis.

The effective spin Hamiltonian now reads:

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

where $E_0$ is the electron energy and $|\Phi(\vec{R})|^2$ is the probability of finding an electron at the 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 Mn-Mn interactions mediated by electron spin can however be understood by examining an exactly solvable problem of two anti-ferromagnetically coupled Mn ions. The energy spectrum of the coupled Mn-spin system is characterized by the total spin $J=\text{M}+1/2$ where $\text{M}$ 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 in the following way:

$$E(M, +) = -(\frac{J_C}{2})M + (\frac{J_{NN'}}{2})[M(M+1) - \frac{35}{2}]$$

$$E(M, -) = -(\frac{J_C}{2})(M+1) + (\frac{J_{NN'}}{2})[M(M+1) - \frac{35}{2}]$$

as shown in Fig. 4(a). In the absence of coupling to the electron spin ($J_e=0$), 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 $M^\ast = (\frac{J_{NN'}}{2\text{M}} - 1)/2$. To estimate the value of $M^\ast$ we approximate our quantum dot by a spherical CdSe dot with radius $R=4\text{ nm}$ and a barrier potential of $1\text{ eV}$ estimated from strain and the Bir-Pikus Hamiltonian. The effective electron-Mn exchange interaction for Mn ions on the surface of the sphere is then given by $J_C = J_{C}[\Phi(\vec{R})]^2 = 4.5\text{ eV}$. For a typical Mn separation in the barrier of $R_{12}=1.2\text{ nm}$, we estimate the antiferromagnetic interaction strength $J_{12}=1\text{ eV}$. Hence for our model system we find $M^\ast = 2$ and the coupling to electron spin aligns spins of nearest neighbour Mn ions. Independent mean field calculations involving tens of Mn ions randomly distributed in the barrier around a spherical or disk shaped quantum dots confirm the existence of ferromagnetic ordering of Mn ions in the vicinity of quantum dots [9]. 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 quantum dot with radius of $4\text{ nm}$ and Mn concentration of $4\%$. We find the existence of the magnetic polaron, with the Mn magnetization decaying as one moves away from the quantum dot. These finding are in agreement with previous calculations of magnetic polarons [2, 3, 4], and for reasonable parameters for our system shows that the presence of electrons in the dot will mediate a local ferromagnetic interaction between Mn atoms near this dot.

The interpretation of our experimental observations is therefore clear. 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 effec-
randomly oriented. When an external magnetic field is then 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 in the experimental data, the resonance positions are independent of magnetic field for fields below \( \sim 500 \) mT. However, as the magnetic 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. A question remains as to why the zero magnetic field splitting is observed here while it was not seen in the optical measurements of Ref. [16, 17]. This however can be understood by the fact that once current begins flowing through the dot, a feedback mechanism sets in where spin polarization of the current enhances the polarization of Mn spins which in turn enhances the polarization of the current. This dynamical effect also explains why spin polarization is observed at much higher temperature than the predicted temperature dependence of magneto-polaron of Fig. 4(b).

In conclusion, we have shown that electrons in a quantum dot can mediate a local ferromagnetic interaction in a surrounding dilute Mn system, and that this leads to a finite energy splitting of spin levels in the dot in the absence of an external magnetic field. Coupled with the resonant tunnelling scheme which allows the bias controlled selection of which dot level is used in tunnelling, our results open up exciting new possibilities of a voltage controlled spin filter which can operate in absence of any external magnetic field, without relying on an inherent ferromagnetism of the component materials.

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