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Enhancement of the electron spin memory by localization on donors in a CdTe quantum well

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We present easily reproducible experimental conditions giving rise to a long electron spin memory at low temperature. The proposed system consists of an electron localized by a donor potential, and immerged in a quantum well. We have measured, by using photoinduced Faraday rotation technique, the spin relaxation time of electrons localized on iodine donors placed at the middle of a 80 Å CdTe quantum well, and we have obtained 20 ns; this spin relaxation time is two orders of magnitude longer than for free electrons in a similar CdTe quantum well [J. Tribollet *et al.*, Phys. Rev. B **68**, 235316 (2003)].

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INTRODUCTION

In the last years, interest in spin physics has been renewed due to its potential application in spintronics and quantum information.[1] In this framework, the main required property is the presence of long spin memory. Several studies in III-V bulk *n*-doped semiconductors performed in the past, demonstrated a long electron spin relaxation time at low temperature.[2–4] That is why the electron spin is a promising candidate to form a "qubit" in future quantum computers. Recent studies of the spin relaxation time as a function of donors concentration, in *n*-doped GaAs crystals, have revealed a maximum of about 100 ns for a concentration near 10^{16} cm^{-3}, and this behavior has been explained as an interplay of different relaxation mechanisms.[5]

In bulk materials, doping atoms and electrons coexist in the same sample, and at low temperature the electrons are mainly localized on the donors. Epitaxy techniques allow to spatially separate the electrons from the donors. Indeed, the donors can be introduced in the material which forms barriers, in such a way that the electrons migrate to the quantum well (QW) material, creating a two-dimensional (2D) free electron gas. There have been some studies of spin relaxation in doped QWs,[6–14] which have mainly measured a spin relaxation time of the order of several hundreds of picoseconds; when a longer time was observed, in general the authors claimed about the localization of electrons in the QW potential fluctuations, associated to spatial variations of the density of donors in the barrier.[13,14]

In this paper, we present easily reproducible experimental conditions giving rise to long electron spin relaxation and decoherence times at low temperature in a QW. We focus our study on electrons localized on donors which have been inserted at the middle of a 80 Å QW. This system is known to increase the localization of the electron wave function, with respect to its localization on donors in three-dimensional (3D) crystals.[15] We have chosen, here, a CdTe QW for a comparison with previous results on free electrons[9] be possible. To our knowledge there is no study of spin relaxation or decoherence times in such a system. We will discuss advantages and disadvantages, when a long spin memory is wanted, of the CdTe compound *versus* the most currently studied GaAs compound.

We use a pump-probe technique, the photoinduced Faraday rotation (PFR), which is well adapted for studying spin relaxation and decoherence times of resident electrons. Comparing the experimental results with previous results obtained for free electrons in a similar CdTe QW, we show that the localization of electrons enhances their spin memory by two orders of magnitude.

SAMPLE CHARACTERIZATION

The studied sample consists of a CdTe/CdMgTe heterostructure grown by molecular-beam epitaxy on a (100)-oriented GaAs substrate, and containing a 80 Å CdTe QW. A donor layer of iodine atoms was placed at the center of the QW. The donors concentration is approximately 10^{11} cm^{-2}. In order to perform transmission and PFR measurements, we have chemically suppressed the GaAs substrate.

Figure 1 shows the low-temperature transmission and photoluminescence (PL) spectra of the studied sample. The transmission spectrum is dominated by a broad band with minimum at 1.622 eV, and a shoulder at lower energy, 1.6175 eV. Kheng *et al.*[16] have shown that the introduction of donors in a CdTe QW leads to the observation, in absorption spectra, of a band associated to the formation of an exciton bound to a neutral donor, called D^0X. In Fig. 1, the vertical arrow indicates the energy of the transmission minimum of a 80 Å QW without doping layer, i.e., the energy necessary to form a free exciton (1.6265 eV). The energy difference between the transmission minima of an empty QW and the doped QW, allows to determine the binding energy of the donor-bound exciton D^0X: 4.5 meV. The shoulder at lower energy is assigned to the formation of an exciton bound to a neutral acceptor; indeed, the introduction of donor impurities in the QW creates compensation sites, then follows the presence of acceptor sites which, in our case, are probably cadmium vacancies. The PL spectrum, obtained after excitation with a 5 mW 633 nm He-Ne laser, is also shown in Fig. 1. It is dominated by the recombination of excitons bound to acceptors, with a long tail at low energy

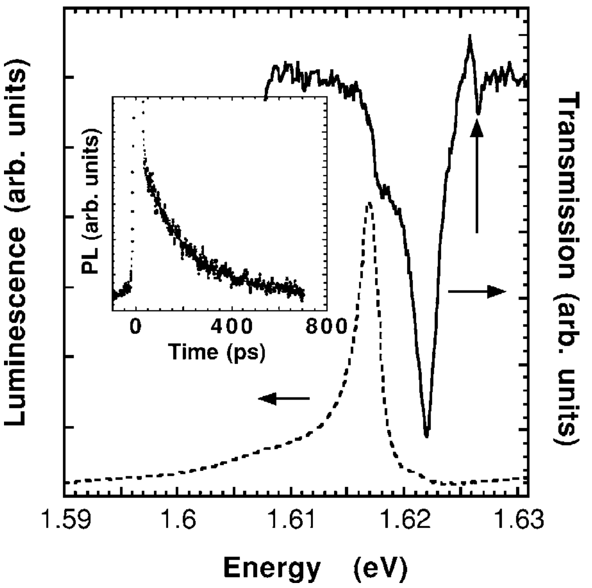

FIG. 1. Transmission (full line) and PL (dashed line) spectra obtained at 2 K for a 80 Å CdTe/CdMgTe QW with a layer of iodine atoms (10^{11} cm^{-2}) placed at its center. The vertical arrow indicates the energy of the transmission minimum observed for the QW without doping layer. The inset represents the 10 K PL decay observed at 1.622 eV after resonant excitation; the black line corresponds to an exponential fit of characteristic time 175 ps.

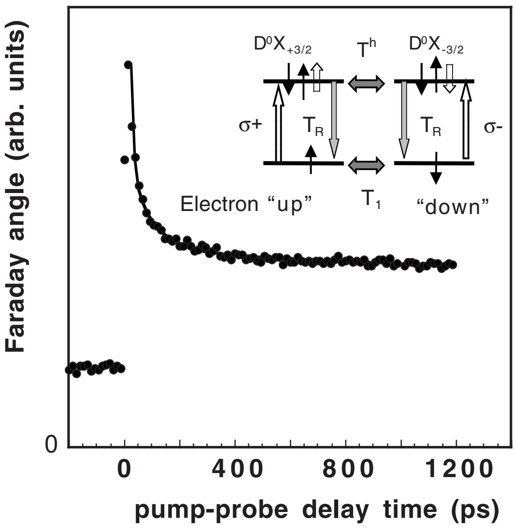

FIG. 2. PFR signal as a function of pump-probe delay, obtained at 2 K for degenerated pump and probe beams tuned to 1.622 eV. The laser repetition period is $\tau_L = 13$ ns. The full line is a fit to a three-exponential decay with characteristic times 10 ps, 80 ps, and 15 ns. The inset shows a diagram of the different optical transitions and relaxation times involved in the formation and evolution of a D^0X complex (see text).

and a very small shoulder at high energy corresponding to the recombination of donor-bound excitons.

Picosecond pulses from a Ti:sapphire laser are used to excite the PL of D^0X, while a streak camera placed after a double monochromator records its time-resolved decay. The inset in Fig. 1 shows the time-resolved PL obtained under resonant excitation of D^0X (at 1.622 eV). Due to this resonant excitation, the first 40 ps are affected by laser diffusion, even when a linearly polarized light is used in the excitation and the cross-polarized emission is detected. The signal shows a monoexponential decay with a characteristic time of 175 ps. Similar single exponential decays are also observed for nonresonant excitations.

PHOTOINDUCED FARADAY ROTATION MEASUREMENTS AND DISCUSSION

The light source of our PFR experimental setup is a Ti:sapphire laser beam with a 2 ps pulse duration and a repetition rate of 76 MHz, which is split into pump and probe beams. The pump average intensity is of the order of 1 $\mathrm{W/cm}^2$, and the probe average intensity is ten times less. The pump beam polarization is $\sigma+/\sigma-$ modulated at 42 KHz with a photoelastic modulator; the probe beam is linearly polarized, and its intensity is modulated with an optical chopper at 1 KHz. After transmission through the sample, the rotation angle of the probe beam polarization is analyzed in an optical bridge.[9] To improve signal-to-noise ratio, a double lock-in amplifier analysis of this rotation angle is performed.

Spin relaxation measurements

Figure 2 shows the PFR signal obtained at low temperature, 2 K, when the pump and probe beams are tuned to the D^0X transition, at 1.622 eV. The band width of the used mode-locked Ti:sapphire laser is less than 1 meV, allowing the pump to create, mainly, excitons bound to neutral donors. We have to note a nonzero PFR signal at negative pump-probe delay times, indicating that the electron spins are not fully relaxed within the 13 ns repetition period of the laser. Assuming an exponential decay of the long-living PFR signal, and taking its extrapolated value at delay $\Delta t = 0^+$ and its value at $\Delta t < 0$, we have estimated the decay time of this long-living signal to be 15 ns. According to the already discussed results of time-resolved PL, the long-living signal has to be related to the net spin polarization of the donor-bound electrons, which are the only species present in the sample when the recombination of the D^0X complexes is finished. The spin polarization of the resident electrons is built *via* the polarization of the D^0X complexes, resonantly excited by a circularly polarized pump pulse, and the subsequent transfer of this polarization to the electrons. The mechanisms involved in this polarization transfer are very similar to those present in the polarization of a 2D electron gas *via* the resonant excitation of trions.[9] The essential condition for this spin transfer to occur, is that the spin relaxation time of the photocreated holes should remain in the same order of magnitude or be smaller than the recombination time of D^0X.

The inset in Fig. 2 gives a diagram of the different optical transitions and relaxation times involved in the formation and evolution of a D^0X complex. A $\sigma+$ ($\sigma-$) circularly polarized pump photocreates D^0X complexes with +3/2 (−3/2) holes, from spin up (down) electrons bound to donors; the spin of a D^0X is the spin of the photocreated hole, because its two electrons, one photocreated and the other associated to the initial neutral donor, are in a singlet state in the lowest energy state. The corresponding localized D^0X complexes are denoted $D^0X_{+3/2}$ ($D^0X_{-3/2}$). Immediately after the pump pulse, the sample contains more donors with down (up) electrons than with up (down) electrons, and also +3/2 (−3/2) polarized D^0X complexes. Let us denote T^h the spin relaxation time of holes, T_1 the spin relaxation time of electrons, and T_R the recombination time of D^0X. If $T^h \ll T_R \ll T_1$, during the presence in the sample of the D^0X complexes, the populations of $D^0X_{+3/2}$ and $D^0X_{-3/2}$ tend to balance, and the recombination of the D^0X complexes gives localized electrons with spins up or down with almost the same probability: Once all D^0X have recombined, the number of resident electrons having spin down (up) is larger than the number of electrons having spin up (down). The same electronic polarization is reached, though with a smaller efficiency, when the less stringent condition $T^h \leqslant T_R \ll T_1$ is fulfilled. This spin polarization is detected in a PFR signal. Thus, the key condition to obtain a spin polarization of the donor-bound electrons is the spin relaxation of holes contained in D^0X complexes be faster than, or at most in the same order of, the D^0X lifetime.

The PFR decay curve given in Fig. 2 can be fitted by three exponential decays: a very fast decay with a short characteristic time of 10 ps, a middle one with an associated time of 80 ps, and a very long third one with a time of 15 ns. Both shortest times are smaller than the D^0X recombination time, and then they have to be associated to the presence of D^0X complexes. The decay of the difference between the $D^0X_{+3/2}$ and $D^0X_{-3/2}$ populations is expected to follow a single exponential with decay rate $1/T=1/T_R+1/T^h$. This rate comes from the two possible channels through which this population difference diminishes, one being the recombination of D^0X, and the other one the hole spin flip at constant total population. Taking $T=80$ ps we deduce a hole spin relaxation time of $T^h=147$ ps, which is slightly smaller but of the same order of magnitude than the experimentally determined recombination time $T_R=175$ ps. However, if we take $T=10$ ps, the associated hole spin relaxation time is equal to $T^h \approx 10$ ps, clearly shorter than the recombination time. We have no model for the presence in the same sample of these two short times (10 ps and 80 ps), but we remark that assuming a unique recombination time of 175 ps, in both cases the condition $T^h \leqslant T_R$ for the realization of the polarization of donor-bound electrons is fulfilled. Thus, we are able to polarize the spins of electrons localized on donors, and we have determined the characteristic damping time of this polarization, $T_1=15$ ns. Remarkably, this time is larger by two orders of magnitude than the spin relaxation time 180 ps obtained for free electrons of a 2D gas (1.7 10^{11} cm^{-2}) in a 100 Å CdTe/CdMgTe QW.[9]

Actually, as this first estimate $T_1=15$ ns is very close to the repetition period $\tau_L=13$ ns of the pump pulses, we worked to obtain a more precise T_1 value using a pulse picker in our experimental setup, which allowed us to change the laser repetition rate from τ_L to $N\tau_L$, with $N=2,3,\ldots,6$. We have carefully tuned the peak powers of the pump and probe pulses to the same values in each experiment $N=2$ or $3\cdots$ or 6. We obtained curves similar to the one in Fig. 2, the PFR signal "at $\Delta t<0$," $S(\Delta t=\tau_L)$, becoming $S(\Delta t=N\tau_L)$. In Fig. 3, we have plotted the ratio $S(\Delta t=N\tau_L)/S(\Delta t=\tau_{\text{ref}})$ as a function of $N\tau_L-\tau_{\text{ref}}$, with $\tau_{\text{ref}}=1$ ns. Assuming a monoexponential decay, we extract a spin relaxation time $T_1=20\pm1$ ns, a value slightly greater than the previous first estimate.

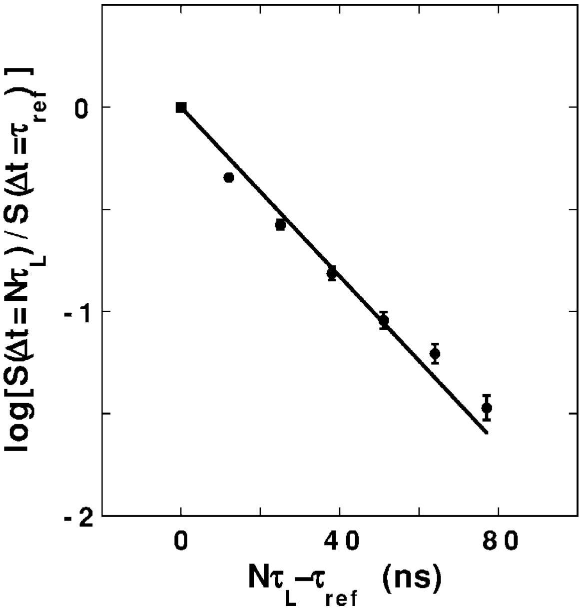

FIG. 3. Semilogarithmic plot of the ratio $S(\Delta t=N\tau_L)/S(\Delta t=\tau_{\text{ref}})$, S denoting the PFR signal, *versus* time $N\tau_L-\tau_{\text{ref}}$, $N=2,3,\ldots,6$, with $\tau_{\text{ref}}=1$ ns. The data, obtained in experiments (2 K, 1.622 eV) with different laser periods $N\tau_L$ leading to curves similar to the one in Fig. 2, are collected for identical peak powers of the pump and probe pulses from one experiment to another. The line crossing the origin (marked by a full square) is a fit of the experimental data which gives $T_1=20\pm1$ ns, a value slightly greater than the previous first estimate extracted from Fig. 2.

Spin decoherence measurements

We have also measured the PFR signal in presence of a transverse magnetic field. The magnetic field is applied along the x direction, and the pump and probe beams are aligned to the z direction, which is also the growth direction of the sample. As already explained, a pump pulse initially creates polarized D^0X complexes, and polarizes the ensemble of donor-bound electrons. When a transverse magnetic field is applied in the QW plane, this field induces a precession of the net spin polarization of the electrons, but not for the D^0X complexes. We have already mentioned that a D^0X contributes to the spin dynamics by its hole only. The precession frequency is the Larmor frequency $\Omega_{e,h}=\frac{g_{e,h}^{\perp}\mu_B}{\hbar}B$, and is almost zero for holes because $g_h^{\perp}\approx 0$, which is not the case for electrons. Then the electronic spin dynamics after initialization by a short pump pulse, can be described by the following equation:

$$\frac{d\vec{S}_{\perp}}{dt}=\vec{\Omega}_e \wedge \vec{S}_{\perp}-\frac{\vec{S}_{\perp}}{T_2^*}+\frac{J(t)}{T_R}\vec{e}_z, \tag{1}$$

where $\vec{S}_{\perp}$ is the transverse component (to the applied magnetic field) of the total electronic spin, $\vec{\Omega}_e=\Omega_e\vec{e}_x$, T_2^* is the decoherence time of the electronic spin, and $J(t)=J_0e^{-t/T}$ is

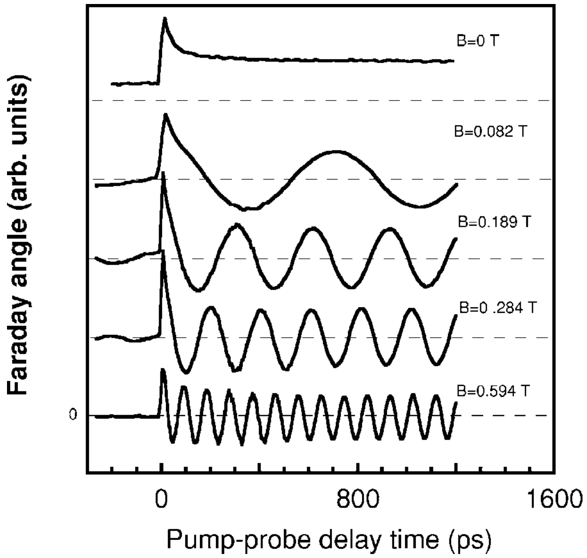

FIG. 4. PFR signals as a function of pump-probe delay obtained at 2 K, 1.622 eV, for several values of a transverse magnetic field. The laser repetition period is $\tau_L = 13$ ns. The curves have been shifted for clarity; the horizontal dashed lines fix the zero of each curve.

one third of the average kinetic momentum of the ensemble of D^0X complexes, with J_0 its value at $t=0$. The source term in Eq. (1) involves the D^0X complexes recombining with rate $1/T_R$ and producing donor spins aligned with z axis. The solution to Eq. (1) gives a simple expression for $S_z(t)$ when $t \gg T$:

$$S_z(t) \propto e^{-t/T_2^*} \cos \Omega_e t. \tag{2}$$

Figure 4 shows that the PFR signal is dominated by an oscillatory behavior, which is associated to the polarization of the electronic spins. We have fitted all PFR curves to expression (2). Figure 5(a) shows the dependence of Ω_e on the applied magnetic field. A linear fit of the data gives an electron Landé factor $|g_e^{\perp}| = 1.3$, which is comparable to the value obtained for free electrons in similar CdTe QWs.[9,11]

For low magnetic fields, less than 0.59 T, the oscillatory behavior of the PFR signals is also observed at negative delay times. That means, first, that the damping time of the oscillations is comparable to the repetition period 13 ns of the laser, and, second, that this damping time decreases for increasing magnetic fields. Figure 5(b) shows $1/T_2^*$ as a function of magnetic field. We have observed, for fields ranging from 0.082 T to 1.18 T, a linear dependence of $1/T^*{}_2$ on magnetic field, leading to a T_2^* ranging from 9.4 ns to 1.5 ns. The used PFR technique gives information on an ensemble of electronic spins, and it is then sensitive to inhomogeneities, such as local magnetic fields or local variations in the electron $g_e^{\perp}$ factor; the measured decoherence time T_2^* thus results from dephasing of the individual contributions of the neutral donors. Assuming a Lorentzian distribution of $g_e^{\perp}$ factors with half width at half maximum $\Delta g_e^{\perp}$, the experimentally determined T_2^* is related to the decoherence time T_2 unaffected by $g_e^{\perp}$ inhomogeneities, as follows:

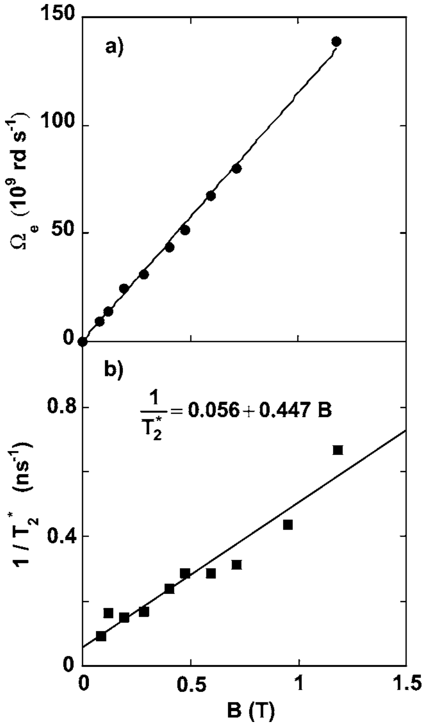

FIG. 5. (a) Measured Larmor frequency *versus* applied transverse magnetic field. A linear fit gives the Landé factor $|g_e^{\perp}| = 1.3$; (b) Inverse of the spin decoherence time T_2^* *versus* applied transverse magnetic field. A linear fit according to Eq. (3) gives the decoherence time T_2 free from $g_e^{\perp}$ inhomogenities, and $\Delta g_e^{\perp}$ (see text).

$$\frac{1}{T_2^*} = \frac{1}{T_2} + \Delta g_e^{\perp} \frac{\mu_B}{\hbar} B. \tag{3}$$

From a linear fit to the expression (3) of the experimentally determined spin dephasing rates $1/T^*{}_2$, shown in Fig. 5(b), we obtain $T_2 \approx 18$ ns and $\Delta g_e^{\perp} \approx 0.005$. The latter value leads to $\frac{\Delta g_e^{\perp}}{g_e^{\perp}} \approx 0.4\%$, which is similar to the value obtained for free electrons in other QWs,[11] but smaller than in GaAs quantum dots[12] ($\Delta g/g = 8\%$), which indicates that donor-bound electrons is a very homogeneous system. As expected, the former value $T_2 \approx 18$ ns, obtained for a vanishing magnetic field, is equal to $T_1 = 20$ ns within experimental uncertainty. It is worth noting that the measured spin coherence time of neutral donors, is longer than the spin coherence time of 2.5 ns reported for localized trions in GaAs,[13] and shorter than 100 ns reported in bulk GaAs.[4,5]

Discussion

In bulk materials, it is well established[5] that for concentrations below Mott transition there are two main mechanisms responsible for the electronic spin relaxation at low temperature, their relative importance depending on donors concentration: the hyperfine interaction with nuclei is the most important at low donors concentration, and the anisotropic spin exchange interaction dominates at high donors concentration. At this point, it is worth making a comparison of the importance of these spin relaxation mechanisms in GaAs and CdTe. On the one hand, according to the previously referenced theoretical works[5] on bulk materials, the strength of the anisotropic exchange interaction is proportional to α^2, with α a dimensionless factor appearing in a term proportional to the third power of k in the conduction-band Hamiltonian.[17] This factor has been experimentally determined for GaAs, but no experimental data can be found for CdTe. To obtain an α estimate for both compounds, we have used the following expression:[18]

$$\alpha \approx \frac{4\Delta_{SO}}{\sqrt{(E_g+\Delta_{SO})(3E_g+2\Delta_{SO})}}\frac{m^*}{m_0}, \tag{4}$$

where Δ_{SO} is the energy difference between the valence band (Γ_8) and the spin-orbit band (Γ_7), E_g is the energy gap of the compound, and m^* is the electron effective mass. Taking values from Ref. 19 for GaAs and from Ref. 20 for CdTe, we obtain that the anisotropic interaction is stronger in CdTe than in GaAs by a factor of nine, leading to expected shorter spin relaxation times in CdTe.

On the other hand, the most important spin relaxation mechanism for isolated localized electrons in semiconductors is the hyperfine interaction. In an ensemble of localized electrons, two regimes have been predicted[21] for the spin polarization decay: the first regime consists of an initial fall of the spin polarization, which makes it reach 10% of its initial value within a characteristic time T_Δ; the second regime is a plateau of the spin polarization, at 1/3 of its initial value, reached from a typical time of $2T_\Delta$—this plateau decaying at a very long time scale, of the μs order. The essential features of this two-regime decay for the electronic spin polarization have been recently observed in p-doped InAs quantum dots.[22] The spin relaxation time T_Δ is fixed[21] by the hyperfine constants A^i, the nuclear spin quantum numbers I^i, and the total number N_L of nuclei (magnetic or not) within the wave function envelope of an electron

$$\frac{\hbar}{T_\Delta} = \sqrt{\frac{2n}{3N_L}\sum_i I^i(I^i+1)(A^i)^2P_i}. \tag{5}$$

n is the number of nuclei inside a unit cell. The sum runs over all nuclei of the crystal basis, and for each such nucleus over its isotopes i which possess a nonzero nuclear spin I^i with probability P_i. In the CdTe compound, one Cd and one Te nuclei are found inside a unit cell ($n=2$), and nonzero nuclear spins are only held by Cd^{111}, Cd^{113}, Te^{123}, and Te^{125}, with $I^{\mathrm{Cd}}=I^{\mathrm{Te}}=1/2$. Both magnetic Cd isotopes have almost the same hyperfine constant:[21,23] $A^{\mathrm{Cd}}=31\ \mu\mathrm{eV}$, and can be found together among all the Cd isotopes with natural abundance $P_{\mathrm{Cd}}=25\%$. The hyperfine constant of Te^{123} and Te^{125}, which have together a $P_{\mathrm{Te}}=8\%$ abundance, is estimated to be $A^{\mathrm{Te}}=45\ \mu\mathrm{eV}$. To calculate T_Δ, we take the localization of a donor-bound electron in bulk CdTe, which is a volume of radius 5.3 nm; we then obtain $N_L=1.8\times10^4$, and finally get $T_\Delta=4.4$ ns. This value is one order of magnitude greater than 500 ps observed in InAs quantum dots of similar volumes;[22] the difference essentially arises from scarce magnetic isotopes in CdTe with low (1/2) spins, whereas all As and In nuclei are magnetic with high spins ($I^{\mathrm{As}}=3/2$, $I^{\mathrm{In}}=9/2$). In fact, we are facing here a general trend: all isotopes in III-V compounds have nonzero nuclear spins, while magnetic nuclei are hardly found in II-VI compounds.

We can conclude that the electron spin relaxation in our sample is not governed by the hyperfine interaction of isolated electrons with their surrounding magnetic nuclei, because the predicted spin polarization plateau is not observed (see Fig. 3). Moreover the technological donors concentration of $10^{11}\ \mathrm{cm}^{-2}$ seems consistent with an important contribution to the electron spin relaxation of the anisotropic exchange interaction between donor-bound electrons.[5,24] Nevertheless Fig. 3 could let appear a short characteristic time 4 ± 1 ns: at short times, the hyperfine interaction could contribute to the spin relaxation. In the future, it would then be interesting to study lower concentrations of donors inside a CdTe QW, to enter a regime where the hyperfine interaction dominates and leads a quasiconstant partial spin polarization, after a fast decay has occurred. Furthermore, this initial fast decay could be suppressed[21] using an applied longitudinal magnetic field larger than $\Delta_B=\hbar/|g_e^\perp|\mu_B T_\Delta=2$ mT, several ten times smaller than the one necessary for InAs quantum dots of comparable sizes.[22]

CONCLUSION

In conclusion, we have obtained the enhancement, by two orders of magnitude, of the spin memory of electrons when they are localized by the Coulombic potential of donors immerged inside a QW: the spin relaxation time of neutral iodine donors located at the middle of a 80 Å CdTe QW is measured to be 20 ns. Electrons localized on donors are a reproducible and very homogeneous system of n-doped quantum dots. They also give the possibility to study the electronic spin relaxation time as a function of their concentration, and of their localization controlled by the QW thickness. In the framework of quantum computation, the major interest of the location of neutral donors inside QWs, is the possibility for them to be spectrally addressed. At low concentrations, isolated donors in a CdTe QW could be promising to reach long-time spin memory, while freezing the spin relaxation process in a very small magnetic field.

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[24] In the absence of theory for the 2D Mott transition, we consider that our donors concentration 10^{11} cm^{-2} corresponds to a 3D concentration $(10^{11})^{3/2}=3\times10^{16}$ cm^{-3}, which is slightly inside the metallic phase for bulk GaAs (Ref. 5); nevertheless, as in CdTe the donor binding energy is higher than in GaAs, we can reasonably hope that our sample is in an insulating phase (for a matter of fact, the sharp D^0X absorption line of Fig. 1 seems consistent with this assumption).