Optically induced multispin entanglement in a semiconductor quantum well

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According to quantum mechanics, a many-particle system is allowed to exhibit non-local behaviour, in that measurements performed on one of the particles can affect a second one that is far away. These so-called entangled states are crucial for the implementation of most quantum information protocols and, in particular, gates for quantum computation. Here we use ultrafast optical pulses and coherent techniques to create and control spin-entangled states in an ensemble of non-interacting electrons bound to donors (at least three) and at least two Mn²⁺ ions in a CdTe quantum well. Our method, relying on the exchange interaction between localized excitons and paramagnetic impurities, can in principle be applied to entangle an arbitrarily large number of spins. The problem of quantum entanglement has attracted much attention since the early days of quantum mechanics. One of the most intriguing features of this phenomenon is that noninteracting parts of a system can show non-local correlations reflecting interactions that occurred in the past (see, for example, ref. 1). Although techniques to entangle two particles and, in particular, two photons² have been known for some time, it is only recently that many (up to four)-particle entanglements have been demonstrated experimentally^{3,4}. Following developments in quantum cryptography, together with the discovery of quantum algorithms that outperform those of classical computation, research on the foundations of quantum mechanics has now moved to the centre of the new field of quantum information⁵. As a result, the question of entanglement has gained practical significance.

In this work, we propose and demonstrate experimentally a novel method for many-particle entanglement involving Zeeman-split spin states of non-interacting paramagnetic impurities (Mn²⁺ and donors) in a CdTe quantum well. Our approach relies on the exchange interaction between the impurity spin and excitons that are generated and controlled by optical means (for related mechanisms, see refs 6,7). As such, our scheme belongs to the category of entanglement mediated by an auxiliary quantum particle (for example, the centre-of-mass phonon in ion-trap quantum computers)⁴, as opposed to that resulting from the variation of parameters such as external fields or exchange constants^{8,9}. Because the entangled spins do not interact with each other unless an exciton is present, our method distinguishes itself from other approaches where the sources of entanglement are interactions that cannot be controlled by the experimenter (for example, excitons in quantum dots10 and NMR quantum computers^{5,11}). Although our approach is, in principle, suitable for controlling the collective state of a single exciton-impurity system, the present study involves an inhomogeneous ensemble of excitons, each one mediating the interaction for the set of impurities that falls within its localization range.

Our entanglement proposal exploits the level diagram shown in Fig. 1, which was originally proposed^{12,13} to explain multiple spin-flip Raman scattering in the same material system we use to demonstrate the method (mechanisms for multiple spin-flip Raman scattering are closely related to those for overtones of longitudinal optical phonons¹⁴). $\vec{S} = (S_x, S_y, S_z)$ is the total spin of Nbound electrons and \mathbf{B}_e is an effective

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Figure 1 Generic level structure of a system of paramagnetic impurities coupled to a single localized exciton in a quantum-well. Large (small) blue circles represent the hole (electron) in the exciton, of energy E_e . Bound electrons are represented by green circles. Optical transitions are depicted by red arrows and spins by arrows attached to the circles. *S* is the total spin quantum number. The diagram shows two exciton states differing in the orientation of the electron spin. Parabolas represent the vibrational analog for S >> 1.

magnetic field describing the exchange coupling between the exciton and the impurities^{12,13,15}. Central to our scheme is the requirement that B_e should not be parallel to the external field **B**, so that the quantization axes in the ground (x axis) and in the optically excited state (w axis) should not be the same, and that $\langle S_z \rangle = \Delta_s \neq 0$ when the exciton is present. This is made possible by a combination of quantum confinement and spin-orbit coupling leading to heavy-hole states for which the hole-spin is parallel to the quantum-well zaxis, irrespective of the orientation of **B**. (The heavy-hole states are the $m_1 = \pm 3/2$ eigenstates associated with the pseudo total angular momentum J with J = 3/2. For the heavy-hole spin to be oriented along the zaxis, the splitting between the heavy and the $m_l = \pm 1/2$ light-hole states must be large compared with the Zeeman splitting; this also applies to bulk wurtzite crystals and zinc-blende materials under strain)¹⁶. Because the eigenstates of S_x and S_w are not orthogonal to each other, this opens optical paths, such as the one shown by the red arrows in Fig. 1, that can be used to establish Raman coherences between an arbitrary pair of eigenstates of S_x and, independently, of S_w . Focusing for the moment on the S_x ladder, we then expect that a single, properly tailored optical pulse will be able to create entangled superposition states of the form



Figure 2 Spontaneous resonant Raman scattering data. The spectra show multiples of the Mn²⁺-paramagnetic resonance (PR). SF and PL denote, respectively, the spin-flip of a donor electron and photoluminescence due to localized excitons. The laser energy is 1.685 eV and 1.659 eV for the top and bottom spectra, respectively.

$$\mathbf{P} = \sum_{k=0}^{2} C_k e^{-ik\Omega_0 t} \left| S - k \right\rangle \tag{1}$$

with predetermined values for the coefficients C_k , where k is an integer and t is time. Here, $|S - k\rangle$ is the eigenstate of S_x of eigenvalue (S - k) and $\Omega_0 = g\mu_{\rm B}B/\hbar$, where g is the gyromagnetic factor and $\mu_{\rm B}$ is the Bohr magneton. Note that $|\vec{S}|^2\Psi = S(S + 1)\Psi$ and that states with different values of S are orthogonal to each other. We also emphasize the fact that, although Ψ contains 2S independent coefficients C_k , the associated product states have only one independent variable, because the coherent superposition is symmetric under spin exchange. Therefore, Ψ is entangled except for a set of C_k values of measure zero.

We stress the fact that a single pulse can create simultaneously S_x and S_w Raman coherences. In time-domain measurements, the signature for a Raman coherence involving states $|S - j\rangle$ and $|S - l\rangle$ is the observation of oscillations of frequency $|j-l|\Omega_0$. Note that, for donors (spin $\sigma = l/_2$), the *m*th harmonic of Ω_0 is associated with an entanglement involving at least *m* impurities. For S >> 1, we can map the problem into that of harmonic oscillators displaced by Δ_S with respect to each other, as represented by the parabolas in Fig. 1. Thus, the Hamiltonian becomes identical to that of a molecule with electrons that couple to a single vibrational mode. The associated Huang–Rhys factor is given by $\Delta_S^2/2 = S(1+B^2/B_c^2)^{-1}/2$. This mapping is important, for it allows us to use the vast literature on coherent molecular spectroscopy (particularly time-domain studies^{17,18}) to analyse our experimental results.

The measurements were performed on a multiple quantum well structure consisting of 100 periods of $Cd_{0.997}Mn_{0.003}$ Te wells of thickness

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Figure 3 Time-domain experiments. a, Differential reflectivity data at $\hbar\omega_c = 1.687 \text{ eV}$ (open circles) for two different external magnetic field strengths (*B*). Curves are fits using the linear prediction method^{25,26}. The mode parameters gained from the fits were used to generate the Fourier transform (FT) spectra. The dominant SF peak is a doublet associated with electron spin-flips. Its lower-frequency component, due to electrons bound to donors, exhibits harmonics at twice (2SF) and three times (3SF) the fundamental frequency (data has been multiplied by ten for clarity. b, Data at 3.4T showing SF-overtones. These results were obtained by subtracting the fitted contributions of all other oscillations.

58 Å, and MnTe barriers of thickness 19 Å grown in the [001] z direction (for a review of diluted magnetic semiconductors, see ref. 19). The Mn²⁺ ions were not intentionally introduced in the wells during growth. The presence of Mn²⁺ ions there is due to diffusion from the MnTe barriers with an interface broadening length of, typically, 1-2 monolayers²⁰. The sample is nominally undoped. Consistent with other reports^{12,13,21,22}, however, our Raman scattering experiments reveal the presence of isolated donors in the wells. We used a continuous wave Ti-sapphire laser to acquire Raman scattering, photoluminescence, and photoluminescence excitation spectra, as well as a mode-locked Tisapphire laser (emitting subpicosecond pulses) in a standard pumpprobe set-up to obtain time-domain differential reflectivity data. Changes in the intensity of the reflected probe pulse, ΔR , give a measure of the dependence of the optical constants on the coefficients C_k characterizing the particular entangled states created by the pump. Following the molecular nomenclature, the entangled states with and without excitons will be referred to in the following as 'excited-state' and 'ground-state' coherences. Processes by which ground-state coherences modulate the probe signal are impulsive stimulated Raman scattering and its associated imaginary component, nonlinear absorption. Excitedstate coherences modulate the spectrum of stimulated emission^{17,18}.

Figure 2 shows resonant Raman results obtained in the Voigt geometry. Depending on the laser energy, the spectrum shows up to

~10 lines at multiples of the Mn^{2+} paramagnetic resonant (PR) frequency and, in addition, a peak at ~13 cm⁻¹ due to the spin-flip of an electron bound to a donor. The PR scattering is strongly enhanced when the energy of the scattered photon resonates with the main photoluminescence feature at the exciton energy $E_{e} \sim 1.677$ eV, associated with dipole-allowed transitions involving heavy-hole states. Our Raman scattering results are in excellent agreement with work reported12,13 on similar samples. In particular, we find that the position of the photoluminescence is red-shifted (by $\sim 6 \text{ meV}$ at B=0) with respect to the maximum of the photoluminescence excitation spectrum, indicating that the relevant intermediate states of the Raman process are localized excitons. Consistent with the model depicted in Fig. 1, the multiple PRscattering is not observed in the Faraday geometry where the hole quantization axis, B and Be are all along z. We recall that the peaks involving an odd (even) number of Mn2+ spin-flips are observed mainly when the polarizations of the incident and scattered light are perpendicular (parallel) to each other^{12,13}. This agrees with the fact that the corresponding excitations are odd (even) under time-reversal and, hence, that they belong to the antisymmetric A_2 (symmetric A_1) representation of the D_{2d} point group of the quantum well (however, a departure from this symmetry is observed at very low temperatures)^{12,13}.

Figure 3 shows time-domain data obtained in the Voigt geometry using a circularly polarized pump, which couples to only one of the two

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Figure 4 Frequency versus magnetic field. Black (red) symbols represent spin-flips of donor (exciton) electrons. Associated curves are fits using the Brillouin function; see Methods. Mn^{2+} -transitions are shown in blue. Lines are linear fits to the data. $\hbar\omega_c = 1.687$ eV.

 $m_1 = \pm 3/2$ states, and a probe detection scheme for which only antisymmetric A_2 excitations are allowed (we used the differential Faraday rotation method²³; it can be shown that the corresponding geometry allows for A_2 - but not A_1 -symmetry excitations). We worked at sufficiently low laser fluences so that the normalized differential reflectivity, $\Delta R/R$, depends linearly on the pump, and is independent of the probe intensity. The long-lived oscillations are due to the Mn2+ PRtransition. At short times, the signal is dominated by electron Zeeman quantum beats, that is, oscillations showing field and temperature behaviour consistent with that of spin-flips of electrons. The corresponding feature in the Fourier transform (FT) spectrum is labelled SF. These results bear a close resemblance to those reported for (Zn,Cd,Mn)Se heterostructures for which the rapidly decaying oscillations were ascribed to free photoexcited electrons²⁴. However, close inspection of the data brings out important differences concerning the nature of the oscillations as well as the source of the coherence. As shown in Fig. 3, linear prediction fits^{25,26} reveal that the dominant SF feature is actually a doublet at ~12 and 13 cm⁻¹ (B = 7 T), and that there are additional contributions due to a mode labelled PR(e) whose frequency depends weakly on the field and, most strikingly, peaks 2SF and 3SF that appear at nearly twice and three times the frequency of SF. These modes do not appear in the Raman scattering spectra but, like the Raman features, their amplitudes exhibit a large enhancement when the central energy of the pulses, $\hbar\omega_c$, is tuned to resonate with the photoluminescence associated with localized states.

For reasons discussed in the following, we assign all but the PR(*e*)feature and one of the components of the SF doublet to spin-flip ground-state coherences. Explicitly, we ascribe the low- (high-) frequency component of the SF doublet to the spin-flip of the electron that belongs to the donor (exciton); the PR(*e*) mode to the Mn²⁺ transition in the excited state; and the 2SF and 3SF modes to multiple spin-flips of donors. Because the electron spin is $\sigma = 1/2$, the observation of the SF-overtones indicates the establishment of Raman coherences and, hence, entanglement involving at least three donor impurities. Because the spectrum of localized excitations is very susceptible to Mn²⁺ density fluctuations, and also depends on the positions of the localization centres in the quantum well, we believe that the fast decay of all but the PR-mode is caused by inhomogeneous broadening. We further note that, in terms of the vibrational analog¹⁵, the excited quantum oscillator in the impulsive limit is in a coherent state^{17,18}. This, and the fact that the two-ion spin system (S = 5) is well described by a harmonic oscillator whereas the three-donor case (S = 1.5) is strongly anharmonic, offers an explanation as to why overtones occur for the donors but not the Mn²⁺ ions.

To further substantiate our assignments, consider the heavy-hole component of the exchange interaction $V_{\text{HH}} = (\beta/3)\Sigma_i \delta(\mathbf{r} - \mathbf{r}_i) S(\mathbf{r}_i) \mathbf{J}$ between a quantum well exciton and Mn²⁺ ions of spin S at sites r_i, Here, δ is the Dirac function, β is a constant, and r is the coordinate¹⁹. As mentioned earlier, the effect of $V_{\rm HH}$ on the ions can be expressed in terms of an effective field $B_e(\mathbf{r}) = (\beta / 3\mu_B g_{Mn}) |\Psi_{HH}(\mathbf{r})|^2 J$ where $g_{Mn} \approx 2$ and Ψ_{HH} is the hole wavefunction^{12,13}. Hence, in the presence of the exciton the paramagnetic transition energy evolves from $\mu_B g_{Mn} B_e$ at B = 0 to $\mu_B g_{Mn} B$ for $B >> B_e$. The latter is consistent with the asymptotic behaviour and the basis for assigning the PR(e) peak to the Mn^{2+} spin-flip transition in the excited state. From the PR(e) frequency at zero field and using for CdTe $n_0\beta = 0.88$ eV, where n_0 is the number density of unit cells (ref. 19), we obtain the very reasonable estimate of ~40 Å for the hole localization length (the bulk exciton radius in CdTe is 50 Å) leading on average to 2.5 Mn²⁺ ions per hole. From this number, the Huang–Rhys factor we infer is ~1.06 (B = 7 T) which is consistent with the observation of ~10 overtones in the Raman spectrum. Albeit indirect, the combination of the Raman scattering and coherent results gives strong evidence for exciton-mediated entanglement involving at least two Mn²⁺ ions.

Our assignment of the SF doublet, 2SF and 3SF, as being due to the spin-flips of donor-bound electrons is supported, first, by the observation that these features resonate at $\hbar\omega_{\rm C} \approx E_{\rm e}$ and, second, by their dependence on temperature and field, which show excellent agreement with theoretical predictions. The curves in Fig. 4 are fits using the standard Brillouin function to account for <S·B> in the expression for the effective electron parameter g_c (see Methods). The resonant behaviour is a clear indication that the relevant states are localized. This, and the linear dependence of the signal with the pump intensity are consistent with the donor interpretation for 2SF and 3SF, because linearity excludes the possibility that the overtones could be due to multiple spin-flips of bound excitons. Additional proof is given by the fact that 2SF is usually much weaker than 3SF; see Fig. 3. This supports our assignment because double-flip excitations transform like A₁ and, therefore, are nominally forbidden in the geometry we used (all other modes, being single or triple flips, belong to the A_2 representation). On the basis of the value of the frequencies at large fields, the fundamental mode from which 2SF and 3SF derive is ascribed to the lower-frequency component of the SF doublet. This mode and the overtones exhibit a similar $\hbar \omega_{\rm C}$ behaviour, different to that of the higherfrequency component associated with the spin-flip of the electron in the bound exciton (see Fig. 1). The fact that the energy of the latter is slightly larger is attributed to exchange effects in the excited-state. From the measured splitting, we obtain an upper limit of ~600 µeV for the electron-hole exchange that is consistent with the value 270 µeV from the literature²⁷. This estimate ignores electron-electron exchange, which is large for exciton-donor complexes²⁸ and may provide an additional longer-range mechanism for donor entanglement.

To provide a quantitative estimate of the entanglement, we neglect absorption and write the effective interaction of light with bound electrons as $\sum_{k,l} H_{S}(k, l)$ where

$$H_{\mathcal{S}}(k, l) = E^{2}(t) \times \left\{ \sum_{\mathrm{uv}, \eta} e_{\mathrm{u}} e_{\mathrm{v}} R_{\mathrm{uv}}^{\eta}(k, l) | \eta, \mathcal{S} - k \rangle \langle \eta, \mathcal{S} - l | + c.c. \right\}. (2)$$

The sum is over all sets of $N \equiv 2S$ electrons, each set denoted by η . E is the electric field, $\mathbf{e} = \mathbf{E}/E$ is a unit vector of components e_u , $R_{uv}^{\eta}(k, l)$ is the Raman tensor for the corresponding transition of the particular set (this tensor vanishes unless all the impurities in the set interact with a single exciton), and *c.c.* is the complex conjugate. Consider a weak impulsive excitation and zero temperature. Then, if $|0\rangle$ is the wave function



immediately before the pulse strikes (the ground state with all spins aligned along B), integration of Schrödinger equation gives for t > 0 the coherent superposition state

$$\Phi \approx \left| 0 \right\rangle + i I_0 \sum_{\mathcal{S} \ge k > 0, \eta} \Xi_{\eta k} \left| \eta, \mathcal{S} - k \right\rangle \exp\left(-i k \Omega_0 t \right) \tag{3}$$

which is entangled for most values of the integrated intensity of the pump pulse, I_0 . Here, $\Xi_{\eta k} = (4 \pi / n_R c\hbar) \Sigma_{uv} e_u e_v R_{uv}^{\eta} (k, 0)$ and n_R is the refractive index, and *c* is the speed of light. A secondary effect of the interaction is that Φ leads to time-varying optical constants (see, for example, ref. 29) with concomitant oscillations in the intensity of the reflected probe pulse. Explicitly,

$$\Delta R/R \propto I_0 \sum_{\eta k} \left| \Xi_{\eta k} \right|^2 \exp\left(-ik \,\Omega_0 \, t\right) \,. \tag{4}$$

For a given set of impurities, and to lowest order in I_0 , the only nonzero components of the density matrix operator, $\hat{\rho}_{\eta}(k,l) = |\eta, S-k\rangle\langle \eta, S-l|$, are $\langle \Phi | \hat{\rho}_{\eta}(0, 0) | \Phi \rangle \approx 1$ and $\langle \Phi | \hat{\rho}_{\eta}(k, 0) | \Phi \rangle = \langle \Phi | \hat{\rho}_{\eta}(0, k) | \Phi \rangle^* =$ $-i I_0 \Xi_{\eta k} \exp(-i k \Omega_0 t)$. Let \mathcal{N}_m be the number of sets of *m* impurities. Defining the ensemble average

$$\rho(m) = \sqrt{\sum_{\eta} \left| \left\langle \Phi \right| \hat{\rho}_{\eta}(m, 0) \right| \Phi} \right\rangle |^2 / N_m \quad , \qquad (5)$$

it follows that the amplitude of the *m*th-harmonic oscillations gives a direct measure of the entanglement because it is proportional to $|\rho(m)|^2$. From the results for donors at 3.4 T (Fig. 3), we get $\langle \rho(2) \rangle / \langle \rho(1) \rangle \approx 0.014$ and $\langle \rho(3) \rangle / \langle \rho(1) \rangle \approx 0.46$ (because the evidence for entanglement is indirect, values for Mn²⁺ cannot be directly inferred from the experiments). It is important to realize that the strength of the *m*th harmonic also measures the probability of finding *m* donors in the region where the coupling between the localized exciton and the impurities is significant. Assuming an interaction length of ~100 Å (in CdTe, the donor radius is ~50 Å), the ratio between the frequency-integrated intensities of the first (SF) and third harmonic (3SF) gives the crude estimate of 5×10^{16} cm⁻³ for the density of donors in our sample. Using this density, we obtain $\langle \rho(1) \rangle / I_0 \approx 30$ m² J⁻¹, which is ~10³-10⁴ larger than the value calculated for off-resonance excitation.

In conclusion, our results confirm that there is a system of localized excitons coupled to paramagnetic impurities in a CdTe quantum well that is well described by the level structure of Fig. 1. We have shown that such a system can be optically excited to generate many-spin Raman coherences for an ensemble. Because the exciton localization centres (due to, for example, surface roughness) and the impurities do not necessarily overlap, our scheme can in principle be applied to entangle a particular group and, ultimately, a large number of spins by using one exciton at a time to address different sets of impurities. Hence, the procedure is potentially set-specific and scalable, thereby holding promise for quantum computing applications.

METHODS

The mode-locked Ti-sapphire laser (Spectra Physics, Tsunami), pumped by a continuous wave 532 nm, 5 W, diode-pumped solid-state laser (Spectra Physics Millennia), provided ~130 fs pulses at a repetition rate of 82 MHz, which were focused to a spot of 400 μ m diameter using an average power of 3–4 mW. The home-built, continuous wave Ti-sapphire source operated at power densities of ~10⁻² W cm⁻². The laser range covers the fundamental gap of the wells, at ~1.68 eV. Data were obtained either in the Voigt (B \pm z) or the Faraday (B//z) geometry with the photon wavevector along z.

MAGNETIC FIELD BEHAVIOUR

The dominant features of both spontaneous Raman and coherent time-domain spectra are intra-well excitations involving the ${}^{6}S_{\alpha 2}$ -multiplet of Mn^{2*} and spin-flips associated with states derived from the conduction band. The field dependence of these transitions is given by the Zeeman coupling $\mu_{\rm B} {}^{6}C_{\alpha}\sigma + g_{\rm Mn} S$) where σ and S are, respectively, the electron spin of the donor ($\sigma = l_2$) and the Mn^{2*} ions (S = 5/2) with $g_{\rm Mn} \approx 2$ (ref. 19). The parameter $g_e = \bar{g}_e -n_{\theta} \alpha x \langle S \cdot B \rangle / \mu_B B^2$ is an effective factor that accounts

for the exchange interaction of the donor electron with the Mn^{2+} -spins. Here $\bar{g}_c \approx -1.64$ is the bare *g*-factor, x is the Mn^{2+} concentration and n_0 is the density of unit cells; for CdTe, $n_0 \alpha \approx 0.22$ eV (ref. 19). Coupling to the Mn^{2+} ions leads to a strongly nonlinear dependence of the Zeeman levels with B, characterized by saturation, that can be used to determine the Mn^{2+} concentration as well as to distinguish electron spin-flip from other features (note that the *g*-factor has approximately the same value for free electrons, electrons bound to donors and electrons in free and bound excitons)¹⁹.

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Competing financial interests

The authors declare that they have no competing financial interests.