

Optical Stark Effect and Dressed Exciton States in a Mn-Doped CdTe Quantum Dot

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We report on the observation of spin-dependent optically dressed states and the optical Stark effect on an individual Mn spin in a semiconductor quantum dot. The vacuum-to-exciton or the exciton-to-biexciton transitions in a Mn-doped quantum dot are optically dressed by a strong laser field, and the resulting spectral signature is measured in photoluminescence. We demonstrate that the energy of any spin state of a Mn atom can be independently tuned by using the optical Stark effect induced by a control laser. High resolution spectroscopy reveals a power-, polarization-, and detuning-dependent Autler-Townes splitting of each optical transition of the Mn-doped quantum dot. This experiment demonstrates an optical resonant control of the exciton-Mn system.

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Semiconductor quantum dots (QDs) exhibit discrete excitonic energy levels with an atomlike light-matter interaction. Resonant optical excitation of QDs has allowed us to observe the absorption of a single QD [1] and the efficient preparation of the quantum state of a single confined carrier [2,3] or Mn spin [4]. So far, only a few experiments have demonstrated the possibility to use a strong continuous wave laser field to create hybrid matter-field systems and manipulate QDs states in their solid environment [5]. The optical dressing of an exciton via the biexciton transition using absorption spectroscopy [6], the Autler-Townes effect in the fine structure of the ground state of a neutral [7] or charged [8] QD, the Mollow absorption spectrum of an individual QD [9], and the emission of an optically dressed exciton-biexciton complex for a QD in a planar microcavity [10] have been reported. It has also been demonstrated that the optical Stark effect can be used to compensate the exchange splitting in anisotropic QDs to produce entangled photon pairs [11].

We show in this Letter that the energy of any spin state of an individual Mn atom embedded in a II–VI semiconductor QD can be tuned by using the optical Stark effect induced by a strong laser field. Under resonant excitation, hybrid light-matter states are created independently for all Mn spin states. The corresponding Rabi energy measured through the Autler-Townes splitting can reach 250 μeV . At low temperature, the energies that control the dynamics of an isolated Mn spin in a CdTe QD, like the strain-induced magnetic anisotropy or hyperfine coupling to the nuclei, are weaker than the observed optical Stark shifts. This opens a way to control the dynamics of a single Mn spin in its solid state environment. We also report optical Stark shifts and optically dressed states of the Mn exchange coupled with the exciton or biexciton. Finally, we discuss a particular situation where two Mn spin states are mixed by the coupling between bright and dark excitons. In

spite of the Mn spin mixing, we show that an optical manipulation of individual spin states can be performed.

The sample used in this study is grown on a ZnTe substrate and contains CdTe QDs. A 6.5 monolayer thick CdTe layer is deposited at 280 °C by atomic layer epitaxy on a ZnTe barrier grown by molecular beam epitaxy at 360 °C. The dots are formed by a tellurium deposition or desorption process [12] and protected by a 100 nm thick ZnTe top barrier. The QDs are 10–20 nm wide and a few nanometers high. Mn atoms are introduced during the growth of the CdTe layer. The Mn concentration is adjusted to optimize the probability to detect one Mn per dot.

Optical addressing of QDs containing a single magnetic atom is achieved by using microspectroscopy techniques. A high refractive index hemispherical solid immersion lens is mounted on the surface of the sample to enhance the spatial resolution and the collection efficiency of single dot emission in a low-temperature ($T = 5$ K) scanning optical microscope [13]. This technique also reduces the reflected and scattered light at the sample surface allowing the measurement of spin-flip scattered photons from a Mn-doped QD [4]. Single QD photoluminescence (PL) is excited with a continuous wave dye laser tuned to an excited state of the QD [14]. Simultaneously, a tunable continuous wave single-mode dye ring laser, named control laser in the following, is used to resonantly excite the excitonic transitions. The resulting circularly polarized collected PL is dispersed and filtered by a 1 m double monochromator before being detected by a cooled CCD camera.

When a Mn atom is included in a II–VI semiconductor QD (CdTe in ZnTe), the spin of the optically created electron-hole pair (exciton) interacts with the five d electrons of the Mn (total spin $S = 5/2$). This leads to a splitting of the once simple PL spectrum of an individual QD into six $(2S + 1)$ components [15]. This splitting results from the spin structure of the confined heavy holes which are quantized along the QDs' growth axis with their spin component taking only the

values $J_z = \pm 3/2$. In first approximation, the hole-Mn exchange interaction reduces to an Ising term $J_z S_z$ and shifts the emission energy of the QD, depending on the relative orientation of the spin state of the Mn (S_z) and hole (J_z). As the spin state of the Mn atom fluctuates during the optical measurements, the six lines are observed simultaneously in time-averaged PL spectra: The PL (emission energy and polarization) is a probe of the spin state of the Mn when the exciton recombines [16].

Only one spin state of the Mn is addressed when a control laser is circularly polarized ($\sigma^\pm$) and tuned on resonance with an emission line of the exciton-Mn (XMn) complex. As illustrated in Fig. 1, the splitting induced by the control laser tuned to the high energy line of XMn in σ^+ polarization can be detected in σ^- polarization on the low energy line of XMn. This is the equivalent of the Autler-Townes splitting observed in atomic physics [17]. The control laser field mixes the states with a Mn spin component $S_z = +5/2$ in the presence (XMn) or absence (Mn alone) of the exciton. At the resonance, the unperturbed states $|Mn\rangle \otimes |n\rangle$ and $|XMn\rangle \otimes |n-1\rangle$ can be dressed into pairs of hybrid matter-field states $|I, n\rangle$ and $|II, n\rangle$, where $|n\rangle$ is a n -photons state of the control laser (see Fig. 1). These states can be written as [18]

$$\begin{aligned} |I, n\rangle &= c|Mn\rangle \otimes |n\rangle - s|XMn\rangle \otimes |n-1\rangle, \\ |II, n\rangle &= s|Mn\rangle \otimes |n\rangle + c|XMn\rangle \otimes |n-1\rangle \end{aligned} \quad (1)$$

with corresponding energies $E_\pm = \frac{\hbar}{2}(\omega_c + \omega_0) \pm \frac{\hbar}{2}\Omega'_r$. Here, $c = \sqrt{(1/2)[1 - (\delta/\Omega')]}$ and $s = \sqrt{(1/2)[1 + (\delta/\Omega')]}$. $\delta = \omega_c - \omega_0$ is the laser detuning with ω_0 the resonance frequency of the unperturbed transition and ω_c the frequency of the control laser. $\hbar\Omega'_r = \hbar\sqrt{\Omega_r^2 + \delta^2}$ defines the energy splitting of the dressed states, where $\Omega_r = \mathcal{P}\mathcal{E}/\hbar$ is the Rabi frequency with $\mathcal{P}$ the dipolar moment of the QD transition and $\mathcal{E}$ the amplitude of the electric field of the control laser. A

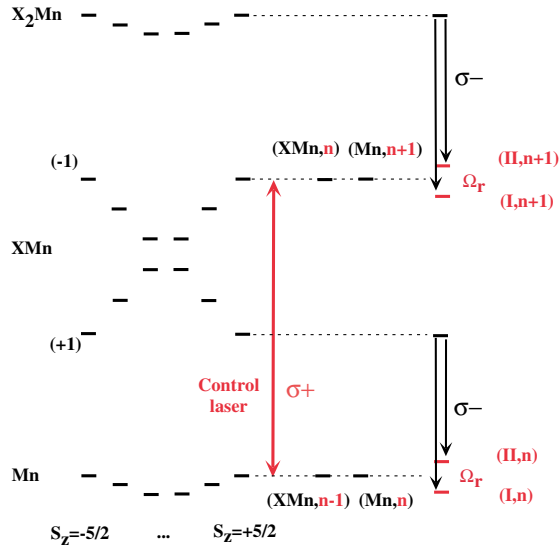


FIG. 1 (color online). Energy scheme of a Mn-doped QD and formation of light-matter hybrid states by a laser field. In the absence of carriers, the Mn fine structure is dominated by the strain-induced magnetic anisotropy, which also splits the biexciton states (X_2Mn). The bright exciton levels (X , with kinetic momentum ± 1) are split by the exchange interaction with the Mn (XMn levels). A pump laser tuned to a QD excited state is used to produce PL of any exciton and biexciton states. The Rabi splitting $\hbar\Omega_r$ induced on the Mn state by the control laser (circularly polarized σ^+) can be probed in the PL of the exciton, while the splitting of XMn is observed in the PL of the biexciton. $|I, n\rangle$, $|II, n\rangle$, $|I, n+1\rangle$, and $|II, n+1\rangle$ are the optically dressed states produced by the mixing of the uncoupled states $(XMn, n-1)$, (Mn, n) , (XMn, n) , and $(Mn, n+1)$, respectively, where n is the number of photons in the control laser.

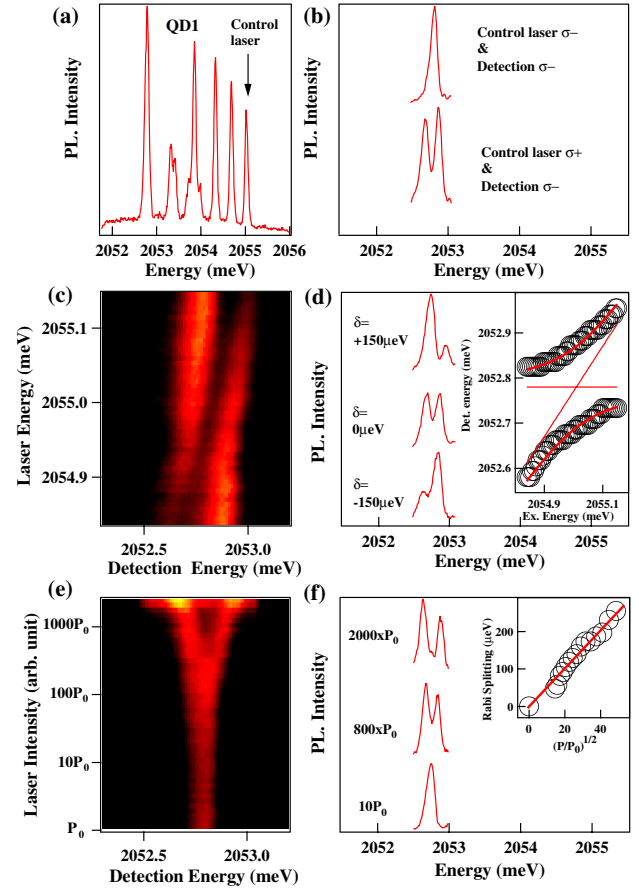


FIG. 2 (color online). Autler-Townes splitting of the emission of $| -1, +5/2 \rangle$ in a Mn-doped QD (QD1) resonantly excited on $| +1, +5/2 \rangle$. (a) shows the nonresonant photoluminescence of the QD. The intensity map (c) shows the excitation energy dependence of the Rabi splitting. The corresponding emission line shape is presented in (d). The inset shows the spectral position of the Autler-Townes doublet as a function of the pump detuning. The fit is obtained with a Rabi energy $\hbar\Omega_r = 180 \mu\text{eV}$. The straight lines correspond to the uncoupled exciton and laser energy. The excitation intensity dependence of the Autler-Townes doublet is presented in the intensity map (e). The corresponding emission line shape are presented in (f). The inset shows the evolution of the Rabi splitting as a function of the square root of the pump intensity. A linear increase is observed. (b) presents the circular polarization dependence of the Rabi splitting obtained under resonant excitation.

power-dependent Autler-Townes type splitting is then expected for all transitions that share such an optically dressed state [17,19].

Experimental data corresponding to a control laser tuned on $|+1, +5/2\rangle$ and the observation of an Autler-Townes splitting in the PL of the state $|-1, +5/2\rangle$ are presented in Fig. 2. Particular care is given to the effect of the detuning of the control laser from the XMn resonance [Figs. 2(c) and 2(d)] and its intensity [Figs. 2(e) and 2(f)]. At large laser detuning, the optically active transitions asymptotically approach the original excitonic transitions where the remaining energy offset is the optical Stark shift. At the resonance, an anticrossing is observed showing that the strong coupling between the laser field and the exciton creates hybrid light-matter states. As presented in the inset in Fig. 2(d), a good agreement with the simple dressed atom model is obtained with a Rabi energy of $\hbar\Omega_r = 180 \mu\text{eV}$. On resonance, the emission from the $|-1, +5/2\rangle$ state splits into a doublet when the power of the control laser is increased, as expected from the Autler-Townes model. The splitting is plotted as a function of the square root of the control laser intensity in Fig. 2(f),

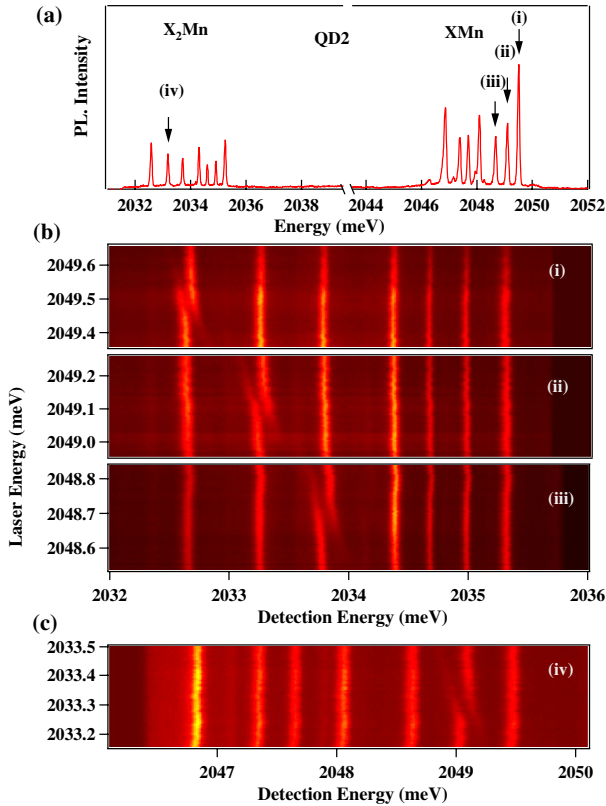


FIG. 3 (color online). (a) PL of the exciton and biexciton in a Mn-doped QD (QD2). (b) Autler-Townes splitting of the exciton in QD2 detected on the biexciton PL under resonant excitation of the ground-to-exciton transition for the spin state of the Mn $S_z = +5/2$ (i), $S_z = +3/2$ (ii), and $S_z = +1/2$ (iii) (arrows in the PL of QD2). (c) Emission of the exciton for a dressed exciton-to-biexciton transition. The excitation is tuned around the state $S_z = +3/2$ of the biexciton (iv).

showing that the splitting linearly depends on the laser field strength. A Rabi splitting larger than $250 \mu\text{eV}$ is obtained at high excitation intensity. It is worth noting that these energy shifts can be easily larger than the magnetic anisotropy of an isolated Mn spin created by the strain in the QD plane ($\approx 40 \mu\text{eV}$) [20–22]. This optical tuning of the fine structure may lead to a control of the coherent dynamics of the isolated Mn spin.

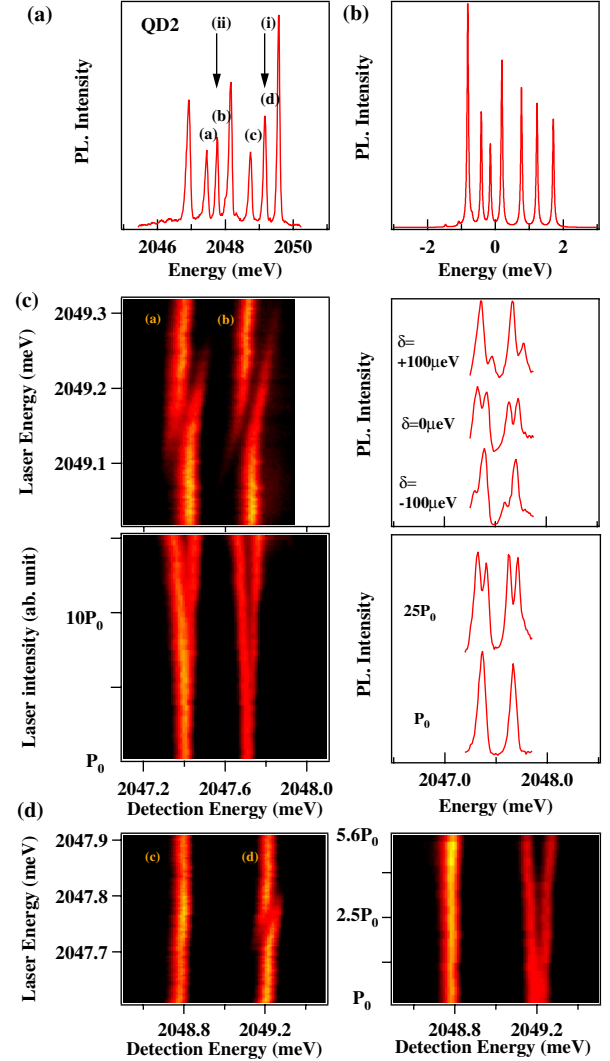


FIG. 4 (color online). Rabi splitting obtained on mixed bright-dark exciton states in QD2. (a) presents the PL of QD2. The intensity of the lines is influenced by the absorption selectivity of the excited state (see Ref. [14]). The corresponding theoretical emission spectra is presented in (b). It is calculated with $J_{eMn} = -0.095 \text{ meV}$, $J_{hMn} = 0.3 \text{ meV}$, $J_{eh} = -0.6 \text{ meV}$, $\epsilon_{VBM} = 0.1$, $\epsilon_{VBM} = 0.1$, $\delta_2 = 0.05 \text{ meV}$, and $T = 25 \text{ K}$ (see Ref. [23] for a detail of the model). (c) presents the laser detuning dependence and the excitation power dependence of the Rabi splitting obtained on mixed bright-dark exciton under excitation on (i). (d) presents the detuning and excitation intensity dependence measured on the high energy transitions associated with the Mn spin states $S_z = +1/2$ and $S_z = +3/2$ under excitation on the mixed dark-bright exciton (ii).

The high energy transition of the XMn complex is twice degenerated. The corresponding optical transitions differ by the polarization of the absorbed or emitted photons. The polarization dependence of the laser-induced splitting shown in Fig. 2(b) confirms the Mn spin selectivity of the strong coupling with the laser field: σ^+ photons couple with the state $|+5/2\rangle$ of the Mn to create two hybrid light-matter states, while no splitting of the σ^- PL line is observed with σ^- control photons.

The strong coupling with the control laser is also observed in optical transitions that involve the biexciton exchanged coupled to a single Mn (X_2 Mn). This is illustrated in Fig. 3 in the case of successive resonant excitations on the XMn levels with a Mn spin state $S_z = +1/2$, $+3/2$, and $+5/2$. In these cases, the recombination of X_2 Mn probes the laser-induced splitting of XMn for given spin states of the Mn. It is shown here that any XMn transition, and consequently any Mn spin state, can be optically shifted by a control laser tuned on resonance. As illustrated in Fig. 3(c), by coherently driving the X_2 Mn-to-XMn transition, one can also tune the energy of any state of the XMn complex. This set of experiments demonstrates that a complete optical control of the exciton-Mn system is possible.

It is also demonstrated here that the use of a resonant strong laser field allows one to individually address any spin state of the Mn even if they are coupled by the exciton through the valence band mixing (VBM) [23]. The particular situation where the Mn spin states $|+1/2\rangle$ and $|+3/2\rangle$ are significantly mixed is presented in Fig. 4. The spectrum of this QD [Fig. 4(a)] differs clearly from the one expected in the pure heavy-hole approximation and presents seven peaks. Such a PL spectrum appears when the high energy lines of the dark exciton states overlap the low energy lines of the bright exciton states. When dark and bright exciton states are close in energy in the heavy-hole approximation, simultaneous hole-Mn spin flips allowed by the strain-induced VBM mix the dark and bright states: From one dark and one bright state, one gets two split states with a bright component [23]. This is the case for QD2: The VBM couples $| -1, +3/2\rangle$ with $| +2, +1/2\rangle$, and the new eigenstates share the oscillator strength of the bright state $| -1, +3/2\rangle$. The two lines on the right of the low energy state can be attributed to the bright part of mixed bright-dark excitons. This attribution is confirmed by the calculation of the energy levels presented in Fig. 4(b) [23,24].

As shown in Figs. 4(c) and 4(d), it is possible to optically address selectively one state (and one only) of the Mn spin in the mixed bright-dark XMn levels. When the σ^+ control laser is tuned on the state $| +1, +3/2\rangle$ [line (d)], an Autler-Townes splitting is observed in σ^- polarization for both components of the emission of the dark-bright excitonic complexes [lines (a) and (b) in Fig. 4(c)]. This arises from the control laser-induced splitting of their common final state with a Mn spin $S_z = +3/2$. With a resonant

excitation on the mixed bright-dark states, only the states which share a $S_z = +3/2$ Mn spin are split: The dark part with Mn spin $S_z = +1/2$ is not affected. This is demonstrated in Fig. 4(d). The σ^- control laser splits the Mn state $| +3/2\rangle$, which leads to the doublet formation in the σ^+ PL from the state $| +1, +3/2\rangle$ [line (d)], while no splitting of the state $| +1, +1/2\rangle$ [line (c)] is observed. The Mn spin state $S_z = +1/2$ is not affected by the control laser tuned on the dark-bright mixed exciton. This experiment suggests the possibility to optically control the exciton-induced coupling between two spin states of the Mn atom, an important step towards coding quantum information on an individual magnetic atom [25].

In summary, we have demonstrated that the ground, exciton, and biexciton states in a Mn-doped QD can be coherently manipulated by applying a strong resonant laser field on the optical transitions. At the resonance, hybrid matter-field states are created that should significantly influence the Mn spin dynamics. Our results demonstrate that, even under a strong optical field, the transition in a Mn-doped QD behaves like isolated two-level quantum systems well described by the dressed atom picture. In the ground state, the laser-induced shift of the Mn spin could be used for a fast optical manipulation of the spin degree of freedom of the Mn atom.

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