Observation of Dressed Excitonic States in a Single Quantum Dot

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We report the observation of dressed states of a quantum dot. The optically excited exciton and biexciton states of the quantum dot are coupled by a strong laser field and the resulting spectral signatures are measured using differential transmission of a probe field. We demonstrate that the anisotropic electron-hole exchange interaction induced splitting between the x- and y-polarized excitonic states can be completely erased by using the ac-Stark effect induced by the coupling field, without causing any appreciable broadening of the spectral lines. We also show that by varying the polarization and strength of a resonant coupling field, we can effectively change the polarization axis of the quantum dot.

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Interaction of semiconductor quantum dots (QD) with laser fields has emerged as a new paradigm in quantum optics. Various experimental achievements based on nonresonant laser excitation of QDs include the demonstration of single-photon sources [1,2], strong-coupling cavity QED [3–7], and polarization-entangled photon generation [8,9]. In parallel, resonant excitation of QDs has enabled the first observation of absorption [10,11] and high-fidelity electron spin preparation [12] in self-assembled QDs. Signatures of nonperturbative laser coupling on QD resonances, such as saturation of absorption contrast [11], Rabi oscillations in photocurrent [13], or ac-Stark shifts in fs-pump-probe spectroscopy [14] have also been demonstrated. In contrast, the success of pump-probe-type experiments where the effect of a strong laser field on the QD is studied using resonance fluorescence has been limited, due to background scattered light arising from the (imperfect) solid-state environment. Two-color experiments where a weak laser probes the signatures of strong field coupling are in turn limited by the shot-noise of the strong laser impinging on the same detector as the probe laser. Only a few experiments have successfully overcome these difficulties: resonant fluorescence was detected in waveguide geometry [15,16] which avoids scattered laser background, and transmission of a weak probe laser was detected using polarization selective rejection of the strong laser [17,18]. The latter experiments led to the observation of a power-dependent Autler-Townes splitting.

Here, we report an observation of dressed states of a QD, by coherently driving the biexciton transition with a strong laser field and monitoring the transmission of a weak probe field applied on the fundamental exciton transition. The energy difference of the two lasers is substantial enough to allow for spectral filtering which in turn provides access to the polarization degrees of freedom. When the strong field is polarized along one of the QD axes and is tuned off-resonance, its principal effect is to induce an ac-Stark shift on the copolarized excitonic state: we demonstrate that this ac-Stark shift can be used to eliminate the exciton fine-structure splitting (FSS)—an all-optical alternative to other postgrowth manipulation techniques for eliminating the FSS [19–22] with the goal of deterministic polarization-entangled photon-pair generation. Alternatively, when the strong laser is not parallel to either of the two QD axes, both fine-structure split excitonic states can be mixed with the biexciton state in a fully coherent way: when the Rabi frequency of a resonant strong field exceeds the FSS, the QD axes are determined by the polarization of the field. The absorption spectrum in this case consists of an Autler-Townes doublet formed due to coherent mixing of the biexciton with the bright exciton that is copolarized with the strong field, and an orthogonally polarized (uncoupled) ''dark exciton'' [23].

The energy scheme of a neutral QD and its optically excited states is presented in Fig. 1(a). The two near-degenerate neutral-exciton states, $|X\rangle$ and $|Y\rangle$, are split by the anisotropic electron-hole exchange energy which gives rise to the FSS $\hbar\delta_{xy}$ [25,26]. The exciton states couple via linearly polarized optical transitions to the QD ground state $|0\rangle$ (empty QD) and the biexciton state $|XX\rangle$ (doubly excited QD). The corresponding transition energies differ by ~3.5 meV due to Coulomb interaction of the electrons and holes confined in the QD: this anharmonicity allows us to probe and manipulate the exciton and biexciton states independently and profit from spectral filtering through diffraction on a reflective holographic grating and subsequent spatial filtering. We choose the energy $(E_x + E_y)/2$ of a QD without FSS as the reference for energy detunings.

Optical control is established with two narrow-band diode lasers with adjustable frequencies and polarization vectors $\hat{u}_c$ and $\hat{u}_p$. The corresponding laser intensities determine the Rabi frequencies Ω_c and Ω_p through the electric field $E\hat{u}_i$ along the exciton dipole moment $\mu\hat{u}_j$ $(j = x,\ y)$ through $\Omega_i = (\hat{u}_j \cdot \hat{u}_i)\mu E/\hbar$ $(i = c,\ p)$. The charging state of the QD and the electrostatic dc-Stark field are set using a field-effect heterostructure where the QDs are sandwiched between a semitransparent Schottky gate electrode and a highly n-doped Ohmic back contact [27]. The sample was operated in an optical helium bath

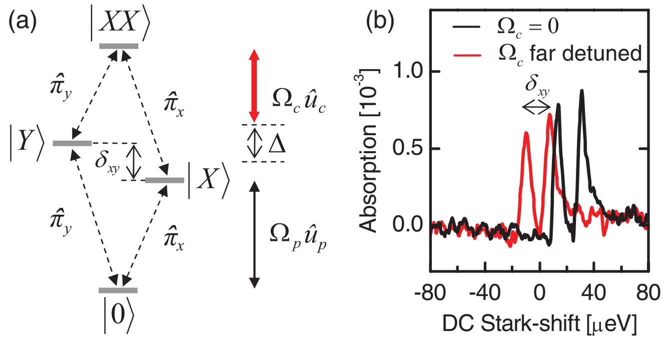

FIG. 1 (color). (a) Energy scheme of the quantum dot optical transitions coupled by polarized laser fields of Rabi frequency Ω_p (weak probe laser) and Ω_c (strong coupling laser) with the corresponding polarization vectors $\hat{u}_p$ and $\hat{u}_c$. The exciton states $|X\rangle$ and $|Y\rangle$, split by fine-structure δ_{xy} due to anisotropic exchange interaction, couple through linearly polarized transitions $\hat{\pi}_x$ and $\hat{\pi}_y$ to the biexciton state $|XX\rangle$ and the empty quantum dot ground state $|0\rangle$. The detuning Δ of the coupling laser is measured with respect to the energy $E = \frac{1}{2}(E_x + E_y)$ of a neutral exciton without fine-structure splitting. (b) Differential transmission spectra of the neutral-exciton transitions in a single quantum dot probed with $\hat{\pi}_{+45}$-polarized laser at 4.2 K: The two spectra were measured for the same dot with the coupling laser on but far detuned (red line) and in the absence of a coupling laser (black line).

cryostat at 4.2 K base temperature and excited through a diffraction limited spot with 1.5 μm full-width at half-maximum.

We identify the neutral-exciton state of a single QD either from gate-controlled charging plateaus in photoluminescence [27] or differential absorption spectra which show characteristic fine-structure split resonances [11]. Differential absorption was measured using dc-Stark shift modulation spectroscopy [10] with a fixed probe laser frequency. Figure 1(b) shows the corresponding spectrum (black solid line) at 4.2 K with the probe field applied at the exciton transition: both $|X\rangle$ and $|Y\rangle$ exciton resonances separated by the FSS $\hbar\delta_{xy} = 20$ μeV are detected since the probe laser polarization was chosen to have equal overlap with the polarization axes of the QD, $\hat{\pi}_x$ and $\hat{\pi}_y$ ($\hat{u}_p = \hat{\pi}_{+45}$).

When we turn the coupling laser on, we observe a blueshift of the exciton energies by 20 to 30 μeV [Fig. 1(b) red curve] [28]. This shift is independent of the coupling laser frequency, provided that the detuning of the coupling laser from the QD resonances is much larger than the width of the lines and the Rabi frequencies. Similar order-of-magnitude blueshifts in absorption lines, most likely induced by creation and trapping of carriers in deep defects within the QD environment, have been observed in other experiments based on strong laser excitation of QDs [17]. For the QD that we studied in our experiments, we measured a QD exciton transition linewidth of $\hbar\gamma_0 = 5$ μeV for probe laser intensities well below saturation. This linewidth, which remained unchanged when the off-resonance coupling field was turned on, is not lifetime limited ($\hbar\gamma_{\mathrm{rad}} = 1$ μeV) and is most likely to be induced by the spectral fluctuations of the exciton energy [11]. All of the experiments reported here were carried out in the limit where the excitonic resonances were weakly power-broadened to $\hbar\gamma \simeq 9$ μeV by the probe laser with $\Omega_p \simeq 1.1\gamma_0$, in order to maximize the signal-to-noise ratio of differential transmission measurements [29].

Figures 2(a) and 2(b) present experimental data where a coupling field with $\hbar\Omega_c = \hbar\Omega_{c,x} = 45$ μeV polarized along one of the QD axes ($\hat{\pi}_x$) is tuned across the biexciton-exciton transition: the gray-scale plots depict experimental absorption spectra measured as a function of the coupling laser detuning from the biexciton resonance (horizontal axis) and the dc-Stark shift controlled detuning with respect to the fixed frequency probe laser

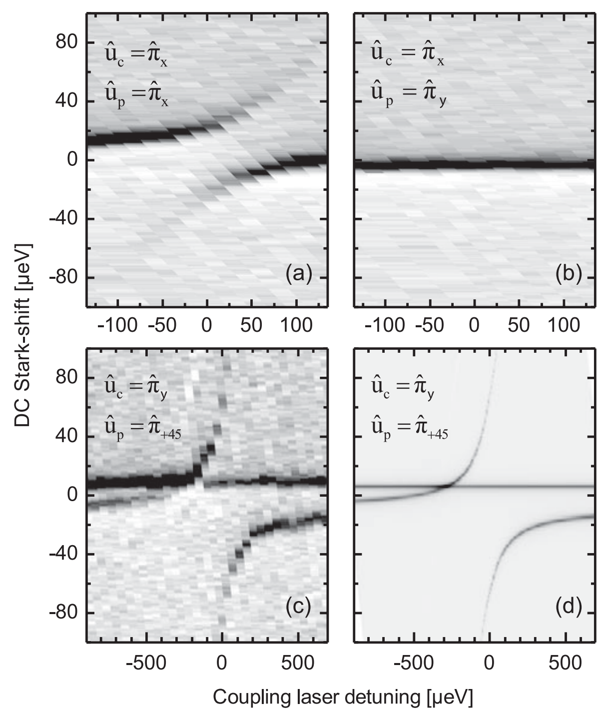

FIG. 2. Neutral exciton states in the presence of a strong exciton-biexciton coupling field in gray scale representation (black and white corresponding to a relative absorption of 0.7×10^{-3} and 0, respectively). Copolarized probe and coupling in (a) reveal the Autler-Townes doublet and the ac-Stark shift of the neutral exciton $|X\rangle$ that is parallel to the coupling laser polarization. The exciton state $|Y\rangle$ orthogonal to the coupling laser remains unaffected, as shown for cross-polarized probe and coupling in (b). Experimental data (c) showing that the exchange splitting can be canceled by exclusively shifting one transition (note different abscissa scales in the upper and lower panel). The corresponding calculation is plotted in (d). Parameters: $\hbar\Omega_c = 45$ μeV in Figs. 2(a) and 2(b) and $\hbar\Omega_c = 120$ μeV in Figs. 2(c) and 2(d); $\hbar\Omega_p = 5.3$ μeV in all graphs.

(vertical axis). The experiment is performed as a sequence of gate-voltage sweeps with stepwise increasing coupling laser energy. Since exciton and biexciton both exhibit a dc-Stark shift, the detuning of the coupling field from the biexciton transition varies as we change the gate voltage. The measured dc-Stark shifts are of equal magnitude for exciton and biexciton, such that each gate sweep lies on a diagonal line of slope -1. For a probe laser that is polarized parallel to the coupling laser, we observe an Autler-Townes doublet, typical for ladder-type atomic systems [Fig. 2(a)]. In contrast, an orthogonally polarized probe laser coupling to the $\hat{\pi}_y$-polarized exciton transition shows no dependence on the coupling laser detuning [Fig. 2(b)]: these results demonstrate that it is possible to realize highly selective coherent manipulation of QD resonances.

Figure 2(c) shows the absorption spectrum of a probe laser that is $\hat{\pi}_{+45}$ polarized and hence couples to both excitonic lines simultaneously. For a $\hat{\pi}_y$-polarized coupling field with a Rabi frequency $\hbar\Omega_{c,x} = 120\ \mu$eV, we find that the dressed $|Y\rangle$-polarized QD state $|D\rangle$ becomes degenerate with the $|X\rangle$ exciton for a detuning of 260 μeV. The vanishing absorption of the other (higher energy) dressed state indicates that $|D\rangle$ is predominantly exciton-like, with vanishing probability for biexciton excitation. In this limit, the effect of the coupling laser can be understood as creating an ac-Stark shift of the $|Y\rangle$ state that exactly cancels the FSS. The experiment depicted in Fig. 2(c) agrees very well with calculations shown in Fig. 2(d), which are in turn based on the Hamiltonian (expressed in the basis $|X\rangle$, $|Y\rangle$, $|XX\rangle$):

$$H = -\frac{\hbar}{2}\begin{pmatrix} \delta_{xy} & 0 & \Omega_{c,x} \\ 0 & -\delta_{xy} & \Omega_{c,y} \\ \Omega_{c,x} & \Omega_{c,y} & -2\cdot\Delta \end{pmatrix}. \qquad (1)$$

Here, $\hbar\Delta$ is the detuning between the coupling laser and the exciton-biexciton transition, and $\Omega_{c,x}$, $\Omega_{c,y}$ are the Rabi frequencies of the coupling laser along the exciton dipole moment axes $\hat{\pi}_x$ and $\hat{\pi}_y$. The calculations depicted in Fig. 2(d) use Rabi frequencies derived from the experimentally measured coupling laser intensity and previously measured values of the excitonic oscillator strength ($f \sim 10$) [30]. The Hamiltonian of Eq. (1), derived using the electric-dipole and rotating-wave approximations, accurately describes the dressed excited states of the QD in the limit of a perturbative probe field. Since the experimental results are obtained using a probe field with $\Omega_p \simeq 1.1\gamma_0$, we could only expect to have a qualitative match with the theoretical model. The effect of dissipation and line broadening can be included using a master equation in the Lindblad form. We would expect the nonperturbative coherent coupling of the QD transition to lead to interference effects that alter the observed absorption line shapes [31].

When the coupling laser frequency is not polarized along one of the QD axes, it leads to the formation of three dressed states which are coherent superpositions of the biexciton state and the two exchange-split excitonic states; in this regime, laser polarization and the competition between the anisotropic exchange interaction and laser Rabi coupling determine the optical response [see Eq. (1)]. In the limiting case of $\Omega_{c,x}, \Omega_{c,y} \gg \delta_{xy}, \Delta$, we would expect the exchange interaction to be insignificant. Among the three dressed eigenstates, one will be a superposition of the two bare excitonic states that lead to absorption or emission polarized orthogonal to the coupling laser; this dark eigenstate [23] will have an energy that is equal to the bare exciton emission energy (i.e., the energy when $\delta_{xy} = 0 = \Omega_c$). The other two eigenstates will be superpositions of the exciton and biexciton states that are split by Ω_c, much like in the case depicted in Fig. 2(a).

Figure 3 shows an experiment confirming these predictions. By applying a coupling laser polarized at 45° with respect to the QD axis and of 1.05 kW/cm^2 intensity, we achieve $\hbar\Omega_{c,x} = \hbar\Omega_{c,y} = 40\ \mu\text{eV} > \hbar\delta_{xy}$. Figure 3 shows the evolution of the QD excitonic absorption as the coupling field frequency is swept through the biexciton-exciton resonance. Unlike Fig. 2, we observe that all three states are coupled; in particular, the state that is Y polarized for large red-detuning maps on to the X-polarized exciton state for large blue detuning [Fig. 3(b)]. For $\Delta = 0$, this

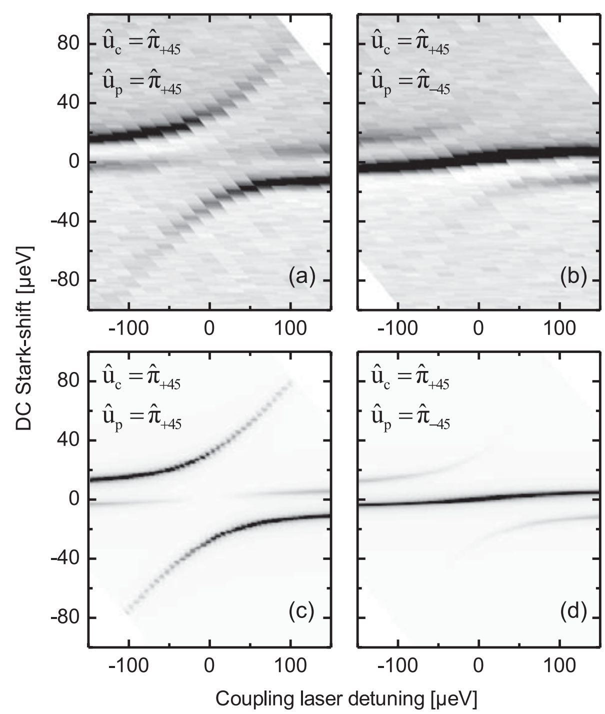

FIG. 3. The strong laser is coupled equally to the two biexcitonic transitions. The upper panel shows experimental data for (a) copolarized lasers and (b) cross-polarized lasers. The lower panel shows the corresponding simulation with $\hbar\Omega_{c,x} = \hbar\Omega_{c,y} = 40\ \mu$eV and $\hbar\Omega_p = 5.3\ \mu$eV which follow from experimental settings. The gray scales are the same as in Fig. 2.

eigenstate can be written as $|X\rangle$-$|Y\rangle$, i.e., its polarization axis is orthogonal to that of the coupling field. In this regime, the QD response can be characterized as consisting of an Autler-Townes doublet polarized parallel to the coupling laser and an orthogonally polarized dark-excitonic state.

An important question that arises for emitters embedded in a solid-state matrix is whether strong laser excitation leads to unwanted coupling or broadening effects that cannot be captured by the simple 4-level model depicted in Fig. 1(a). To address this question, we have measured the total area under the absorption resonances and the transition linewidths, as a function of the coupling laser intensity. While the former tells us whether there is any appreciable excitation of other QD states, the latter would point out to decoherence effects mediated by the strong field itself. We find that even for the strongest coupling field intensities used in our experiments ($\hbar\Omega_{c,x} = 120\ \mu$eV), the changes in the total area under the resonances were well within the error bar of our experiments. We did observe a slight increase of ~10% in the average transition linewidths when the coupling laser was on resonance. More importantly, we observed that the fluctuations in the measured linewidths (from one scan to another) was enhanced; for example, the measured linewidths of the two Autler-Townes split lines could differ by as much as ~50%. We tentatively conclude that these fluctuations as well as the slight linewidth enhancement is due to intensity fluctuations of the coupling laser, and is not a result of a deviation from the simple 4-level model.

In summary, we have demonstrated that QD states can be coherently manipulated, by applying a nonperturbative resonant laser field on the biexciton-exciton transition. The observation of polarization-selective ac-Stark shift of excitonic resonances could be considered a key step towards laser-induced Zeeman-like shift of QD spin states, which would in turn allow for fast manipulation of spin degrees of freedom [32]. The excitonic dark states that are immune to strong field excitation suggest that phenomena such as coherent population trapping are within reach in these solid-state emitters.

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