Photon Beats from a Single Semiconductor Quantum Dot

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Single-photon interference is observed on the ultranarrow long-term stable exciton resonance of an individual semiconductor quantum dot. This interference is related to the fine-structure splitting and allows direct conclusions about the coherence properties of the exciton. When selectively addressing a particular dot by quasiresonant phonon-assisted excitation, despite a rapid orientation relaxation on a 1-ps time scale, coherence is partly maintained. No significant further decoherence occurs when the ground state is reached until the exciton recombines radiatively ($\approx$300 ps).

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Mesoscopic superpositions of quantum states and their coherence is a fundamental issue in quantum physics. Semiconductor quantum dots (QDs) have attracted considerable interest in this regard, motivated, e.g., by using them in quantum information processing [1]. A general obstacle is, however, that decoherence caused by coupling to external degrees of freedom such as phonons destroys the unitary quantum evolution. The band continua of bulk semiconductors allow efficient elastic and inelastic scattering processes, giving rise to decoherence times on the fs-time scale. The atomlike exciton ground state of an ideal and isolated QD may be, however, predominantly damped by its interaction with the radiation field, implying that coherence is maintained during the exciton lifetime. Recent work has focused on the coherent control of individual excitons localized by interface fluctuations in quantum wells [2] or has investigated ensemble averages [3], where single-particle features are hidden by the large inhomogeneous broadening.

In this Letter, we report on a coherence study of single self-assembled CdSe QDs by analyzing their time-resolved secondary emission under quasiresonant optical excitation. We observe distinct photon quantum beats that allow direct conclusions about the degree of electronic coherence and how coherence is maintained during relaxation.

The ultrapure CdSe/ZnSe QD structures are grown by molecular beam epitaxy using a thermal activation procedure [4], enabling maximum control and reproducibility of the formation process. The QDs are on top of a 2 monolayer (ML) thick wetting layer, their height is 1.6 ± 0.2 nm, the lateral extension is below 10 nm, and the density is $10^3\ \mu\text{m}^{-2}$. Single-QD photoluminescence (PL) and PL excitation (PLE) spectra were measured using a micro-optical setup. Both excitation and detection are along the heterostructure growth axis. In order to ensure selectivity also with respect to the excited QD states, islands with lateral extensions of 50–500 nm were prepared by lithographic etching. For detection, a double monochromator with a spectral resolution of 70 μeV was used in combination with a charge-coupled device matrix or a single-photon-counting readout system. Spectrally sharp and tunable excitation is achieved by a dye laser with a three plate Lyot filter. In time-resolved measurements, the PL was excited with 3.5 ps pulses (repetition rate 76 MHz), dispersed in a subtractive double monochromator and detected with time-correlated single-photon counting, providing an overall time resolution of 35 ps. For maximum noise reduction, typical counting rates include 10^{11} excitation shots per PL transient. The absence of any biexcitonic emission affirms low-density conditions (one exciton per QD per shot). Linear polarization control of both excitation and secondary emission with better than 100:1 is achieved by properly adjusted $\lambda/2$ plates. All measurements were carried out at temperatures below 10 K.

Figure 1 summarizes cw-PL spectra of single QDs from a $150 \times 150\ \text{nm}^2$ island. The width of the single PL line in the lower part is resolution limited, demonstrating an exceedingly small homogeneous broadening of the optically allowed ground state exciton, referred to as $|X_0\rangle$ in what follows. We emphasize that there is no resolvable spectral line drift or blinking effect within typical detection periods of $\leq$30 min. This excludes any defect-related environmental coupling, e.g., due to short-term recharging processes. Even when extending the observation over several hours, the long-term line shift is below 150 μeV (see inset of Fig. 1). This ultrahigh stability of the QD emission is paramount for the time-resolved measurements presented below. For symmetry below D_{2d}, $|X_0\rangle$ is split by the electron-hole exchange interaction in two levels ($\alpha = x, y$) with dipole moments oriented along the two nonequivalent in-plane QD axes [5]. The upper panel of Fig. 1 represents data for QDs with a higher degree of in-plane anisotropy, where this splitting is indeed directly manifested by the doublet character of the PL line. Variation of the energy splitting ($40 < \Delta E_{\text{ex}} \leq 450$ μeV) signifies considerable fluctuations of the QD symmetry.

Figure 2 depicts excitation spectra of three individual QDs with different exciton ground state energy E_0. We note that the spectra are background-free, ensuring that indeed signals from QDs other than the selected one are not involved. In the energy range displayed in Fig. 2, the excitation spectra exhibit three characteristic features. First, there is a sharp line (FWHM = 0.5 meV) on

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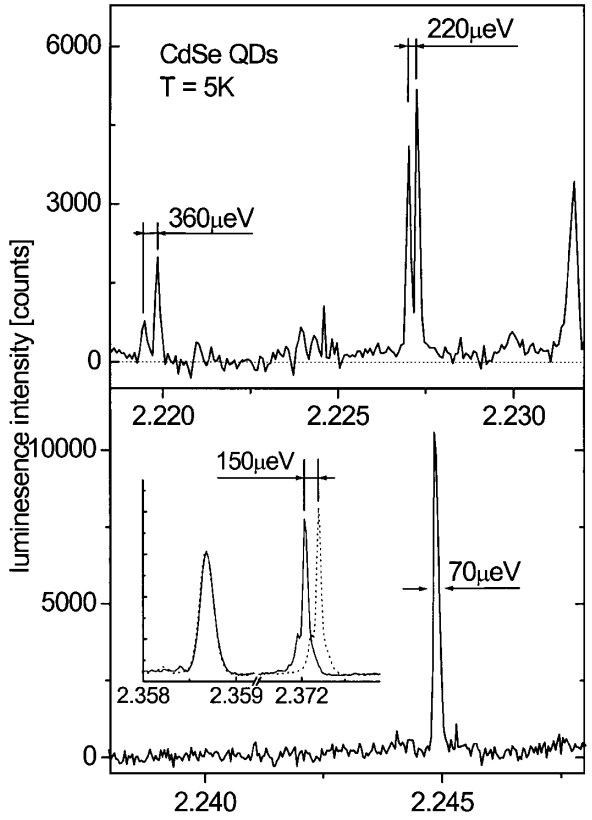

FIG. 1. PL spectra of single CdSe/ZnSe QDs from a 150 × 150 nm island at $T = 5$ K. Inset: PL lines of two QDs, right after starting the measurement (full line) and 5 h later (dotted line) under cw excitation. Note that the left-hand QD exhibits even no measurable long-term shift.

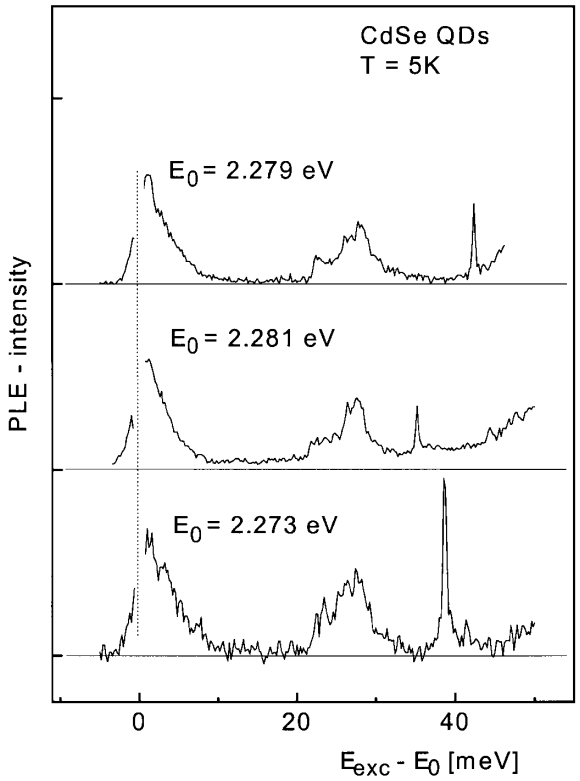

FIG. 2. Excitation spectra of three different CdSe/ZnSe QDs. The energy zero corresponds to the respective ground state PL position E_0.

the high-energy side, associated with the first excited optically allowed state $|X_1\rangle$. While its overall position is consistent with the above size data, fluctuations in the E_1-E_0 separation of 35–45 meV indicate a complex internal QD structure. In addition, there are two broad bands, one in direct proximity of the ground state and the other 25 to 30 meV above, with practically no variation of their position relative to the corresponding E_0. They are due to acoustical and optical phonons (bulk CdSe: $\hbar\omega_{\mathrm{LO}} = 26.3$ meV [6]), respectively. We do not observe any signature of Brillouin or Raman scattering, affirming that the excitation spectra are due to the real population of $|X_0\rangle$. There are also no inverse phonon replica in emission beyond the noise level of our measurements, implying that the phonon coupling of $|X_0\rangle$ is weak. A detailed discussion of shape and magnitude of the phonon contributions is beyond the scope of this Letter and is found elsewhere [7]. In the present context, it is merely of relevance that these features allow quasiresonant and selective excitation of one particular QD.

The energy doublet of the $|X_0\rangle$ exciton with its radiative coupling to the total QD ground state represents a V-type system, where quantum beats in the spontaneous emission may occur [8]. These beats, associated with single-photon interference, carry direct information about the coherence of the electronic state behind the emission. Previous observations on semiconductors were based on the exciton spin splitting in an external magnetic field [9]. In the present case, the doublet consists of two linearly cross-polarized components, so that interference is possible only by projecting the polarizations on a common axis before detection. This suggests an experimental geometry as schematized in Fig. 3. Two cross-aligned polarizers for excitation ($\vec{e}_0$) and detection ($\vec{e}_A$) can be tuned with variable angle φ_0 relative to the fundamental QD axes, oriented along [1,1,0] and $[1,\overline{1},0]$, and denoted by $\vec{x}$ and $\vec{y}$, respectively. The far-field detector signal is then, in Wigner-Weisskopf approximation,

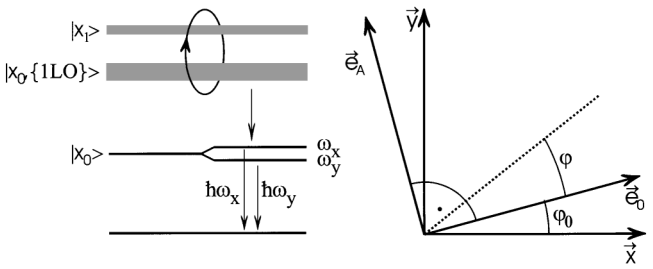

FIG. 3. Energy level scheme (left part) and geometry (right part) of the measurements.

$$I(t) \propto \langle \Psi(t)|\vec{e}_A\vec{E}^{(-)}(t)\vec{e}_A\vec{E}^{(+)}(t)|\Psi(t)\rangle \propto |(\vec{e}_A\vec{x})c_x e^{-(i\omega_x+\Gamma/2)t} + (x \rightarrow y)|^2.$$

Here, $\Psi(t)$ is the exciton-photon wave function, and ω_α denotes the transition frequency. We have assumed equal squares of both dipole moments and, hence, radiative decay rates Γ. The initial wave function $\Psi(0) = c_x|x\rangle + c_y|y\rangle$ prepared by the optical excitation is a superposition of both fine-structure components with coefficients c_α (no photon).

Figure 4 summarizes typical time-resolved PL transients of a single QD for different excitation conditions. For excitation in the continuum states of the wetting layer (inset), the PL decay is monotonous with a single-exponential time constant of $1/\Gamma = 320$ ps. This value is in the range of the anticipated radiative lifetime. However, tuning the excitation laser at the 1-LO-phonon excitation feature and addressing, hence, the QD quasiresonantly, clear modulations of the signal appear for $\varphi_0 = 45°$ (upper curve). The beat period of 320 ps is consistent with a fine-structure splitting

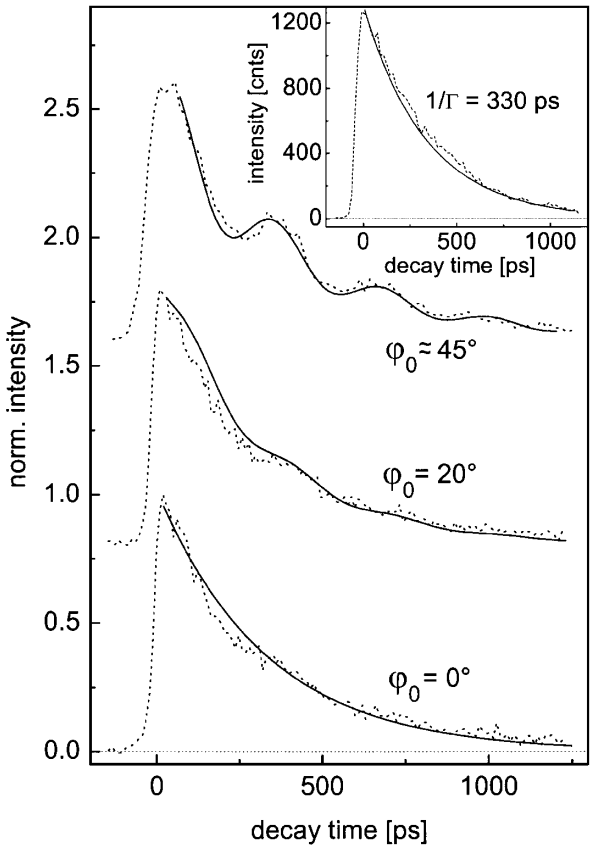

FIG. 4. PL transients of a single CdSe/ZnSe QD for excitation at the 1-LO-phonon resonance for three polarization configurations. Dotted curves: experiment; lines: fits with $I(t)$ given in the text ($u = 0.37, \nu = 0.09, \frac{1}{\Gamma} = 330$ ps, $\frac{1}{\Delta\omega_{ex}} = 322$ ps). Inset: wetting layer excitation for comparison. Note that the 35 ps apparatus function is not deconvoluted in the data fits.

of $\Delta E_{ex} = \hbar(\omega_x - \omega_y) = 13\mu$eV, not resolvable in the spectral domain. On the other hand, when analyzing parallel to the excitonic dipole moments ($\varphi_0 = 0°$), the beats disappear in accord with the above formula for $I(t)$, while the single-exponentially decaying background persists despite $\vec{e}_A \perp \vec{e}_0$.

In none of the measurements in Fig. 4 is a signal rise observable, so that relaxation and subsequent radiative decay are well-separated processes in time. However, during relaxation, coupling with the macroscopic reservoir of phonon modes randomizes the $\Psi(0)$ from which the emission emerges. Since integrating over many excitation spots, our data represent, thus, time averages of this randomization. These averages enter the signal by $\langle|c_x|^2\rangle = 1 - \langle|c_y|^2\rangle$ and $\langle c_x c_y^*\rangle$, where the latter displays the degree of coherence. For wetting layer excitation many phonon energies above E_0, any correlation between c_x and c_y is washed out, yielding $\langle c_x c_y^*\rangle = 0$. The beats occurring under excitation via the 1-LO-phonon band do not exhibit a visible Hanle-type $\sin(\Delta\omega_{ex}t)$ component. The phase shift between c_x and c_y is, hence, small, meaning that linear polarization is practically maintained, however, with a statistically distributed orientation. Defining $c_x = \cos(\varphi_0 + \varphi)$ and $c_y = \sin(\varphi_0 + \varphi)$, the signal reads as $I(t) \propto e^{-\Gamma t}[A + B\cos(\Delta\omega_{ex}t)]$, $A = 2x^2y^2 + (x^2 - y^2)^2u - xy(x^2 - y^2)\nu$, $B = 2x^2y^2(2u - 1) + xy(x^2 - y^2)\nu$, $x = \sin(\varphi_0)$, $y = \cos(\varphi_0)$, with the two statistical parameters,

$$u = \int_0^\pi d\varphi\, P(\varphi)\sin^2(\varphi),$$

$$\nu = 2\int_0^\pi d\varphi\, P(\varphi)\sin(\varphi)\cos(\varphi),$$

where $P(\varphi)$ is the angular distribution—measured relative to $\vec{e}_0$—established during relaxation (cf. Fig. 3). Good agreement with the experimental transients is achieved for $u = 0.37$ and $\nu = 0.09$. The beat data uncover, thus, a rapid orientational relaxation of the initially imprinted dipole moment. We ascribe this to the fact that excitation at $E_0 + \hbar\omega_{LO}$ creates an exciton-phonon complex, comprising components of both $|X_0, \{1_{LO}\}\rangle$ and $|X_1, \{0_{LO}\}\rangle$ (cf. Fig. 3) [10]. Though we have at the moment no details about the internal structure of $|X_1\rangle$, it is very unlikely that $|X_0\rangle$ and $|X_1\rangle$ have identical dipole moments. Consequently, the total moment is not conserved during succeeding evolution. The lifetime of the exciton-phonon complex is limited by the decay of the LO in acoustical phonons, escaping from the QD, by which the current polarization is projected onto the $|X_0\rangle$ subspace. The homogeneous width of $|X_1\rangle$ in PLE yields an upper limit for the LO-phonon lifetime of $\tau_{LO} = 1.5$ ps, which is in fact close to the inverse level spacing $\hbar/(E_1 - E_0 - \hbar\omega_{LO})$. Note that the lifetime of the electron-(LO)phonon oscillation in bulk semiconductors is about 100 fs or shorter [11].

The assumption behind the Wigner-Weisskopf approximation for $I(t)$ is that the electronic state is purely radiatively damped and stays, hence, entirely coherent during its lifetime. To allow for decoherence, extra dampings have to be introduced: $1/\tau_1$ for the total occupation $\langle |c_x|^2 + |c_y|^2 \rangle$, $1/\tau_2$ for the occupation difference $\langle |c_x|^2 - |c_y|^2 \rangle$, and $1/\tau_{\mathrm{coh}}$ for $\langle c_x c_y^* \rangle$ [12]. However, the fact that the beat amplitude decays with the same time constant as the overall signal clearly demonstrates $\Gamma \gg 1/\tau_1,\ 1/\tau_2,\ 1/\tau_{\mathrm{coh}}$. Therefore, another important finding revealed by the single-dot PL transients is that, once the exciton has reached the $|X_0\rangle$ ground state, no further decay of coherence takes place. We conclude that coupling with acoustical phonons or imperfections plays indeed no role during the $\approx$300 ps exciton lifetime. The absence of relaxation among the fine-structure levels of $|X_0\rangle$ is also confirmed by single-dot magneto-PL data, where both Zeeman components occur with virtually equal intensity up to highest fields, even when the line splitting is an order of magnitude larger than kT [7].

In conclusion, we have observed single-photon interference from the ultranarrow long-term stable exciton resonance of an individual semiconductor QD. This observation is a direct reflection of the which-path problem for quantum objects. The random interaction with the macroscopic phonon environment during the relaxation pathway of the exciton spoils the coherence only partly. For the ground state itself, there occurs no measurable further decoherence during the exciton lifetime. For example, using fs-laser sources, this allows a reasonable number of qbit operations per QD. Our data suggest that the ground state exciton is indeed purely radiatively damped, enabling many other interesting quantum optical experiments on these man-made "atoms."

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