Transient nonlinear spectroscopy of excitons and biexcitons in single quantum dots

T. H. Stievater,* Xiaoqin Li, and D. G. Steel
Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan 48109

D. Gammon, D. S. Katzer, and D. Park
Naval Research Laboratory, Washington, DC 20375
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Transient nonlinear spectroscopy is used to study excitons and biexcitons confined to single interface fluctuation quantum dots (QD's). Exciton lifetimes measured by this technique are found to be between 25 and 50 ps, depending on the QD studied. Biexciton binding energies and polarization selection rules agree with previous measurements from cw nonlinear spectroscopy, and biexciton lifetimes are found to be half of the exciton lifetime for the same quantum dot. The ability to perform transient nonlinear spectroscopy of excitons and biexcitons in single quantum dots is an important step towards the development of solid state quantum computing based on interacting excitons in quantum dots.

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The atomlike properties of semiconductor quantum dots (QD's) have made these structures attractive candidates for use in many quantum devices, including solid-state quantum logic gates,[1,2] sources of nonclassical light,[3,4] and QD-cavity strong coupling.[5–8] The development of such QD-based devices has emphasized the need to understand decoherence times and lifetimes in quantum-confined semiconductor structures. Time-resolved photoluminescence (PL) spectroscopy has previously been used for measurements of radiative lifetimes of both excitons[9,10] and biexcitons[11–13] in single quantum dots. Other optical properties, such as coherences, nonradiative decay rates, or transition strengths, can be studied using nonlinear optical spectroscopy.[14] Lifetime measurements based on transient nonlinear spectroscopy include both radiative and nonradiative decay and the temporal resolution is limited only by the pulse width. An understanding of the creation and subsequent decay of biexcitons in single QD's has become increasingly important not only for insight into higher-order Coulomb correlations in zero-dimensional semiconductor systems,[15,16] but also for an improved understanding of decoherence in two-qubit quantum logic gates.[2,17,18]

In this paper, we demonstrate transient differential transmission (DT) spectroscopy of single QD excitonic and biexcitonic states. This technique is used to measure lifetimes of both excitons and biexcitons in single QD's. The measurements are made possible by the use of submicron apertures that restrict the optical excitation to only a few QD states[19,20] and the enhanced optical absorption by excitons confined to single interface fluctuation QD's.[6,21]

The experiments are performed on QD's formed naturally in a thin (4.2 nm) GaAs layer grown between two 25-nm $Al_{0.3}Ga_{0.7}As$ layers. Two-minute growth interruptions at each $GaAs/Al_{0.3}Ga_{0.7}As$ interface lead to the formation of monolayer-high islands which tend to localize the heavy-hole excitons in the GaAs layer.[20,22] Excitons in isolated QD's are probed through ~ 0.5-μm apertures in an aluminum mask laid directly onto the sample's surface. The GaAs substrate is removed to allow for transmission studies and the experiments are performed at 6 K.

The monolayer islands are elongated in the $[\bar{1}10]$ direction, leading to linear polarization selection rules for the excitonic and biexcitonic transitions, denoted by Π_x and Π_y.[19,23,24] The heavy-hole excitonic and biexcitonic levels for a single asymmetric quantum dot are shown in Fig. 1. The density matrix equations for the four-level system used to describe the relevant Π_y excitonic and biexcitonic transitions for the third-order pump-probe experiments in this paper are given in Eqs. (1)–(5). Π_y-polarized fields were used for this paper, though Π_x fields could have been used to obtain similar results:

$$\dot{\rho_{gg}} = \Gamma_{yg}\rho_{yy} + \left(\frac{i\mu_{gy}}{2\hbar}\rho_{yg}\sum_k E_k^*(t)e^{i\Omega_k t} + \text{c.c.}\right), \quad (1)$$

$$\dot{\rho_{yg}} = -(\gamma_{yg} + i\omega_{yg})\rho_{yg} + \frac{i\mu_{yg}}{2\hbar}(\rho_{gg} - \rho_{yy})\sum_k E_k(t)e^{-i\Omega_k t}, \quad (2)$$

$$\dot{\rho_{yy}} = -\Gamma_{yg}\rho_{yy} + \Gamma_b/2\rho_{bb} - \left(\frac{i\mu_{gy}}{2\hbar}\rho_{yg}\sum_k E_k^*(t)e^{i\Omega_k t} + \text{c.c.}\right) + \left(\frac{i\mu_{yb}}{2\hbar}\rho_{by}\sum_k E_k^*(t)e^{i\Omega_k t} + \text{c.c.}\right), \quad (3)$$

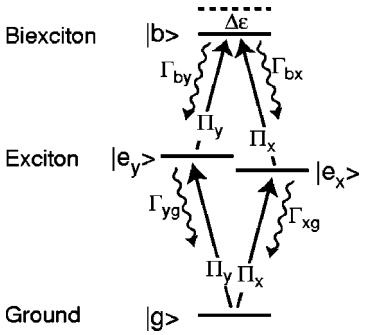

FIG. 1. The energy-level structure and polarization selection rules for heavy-hole quantum dot excitonic and biexcitonic transitions.

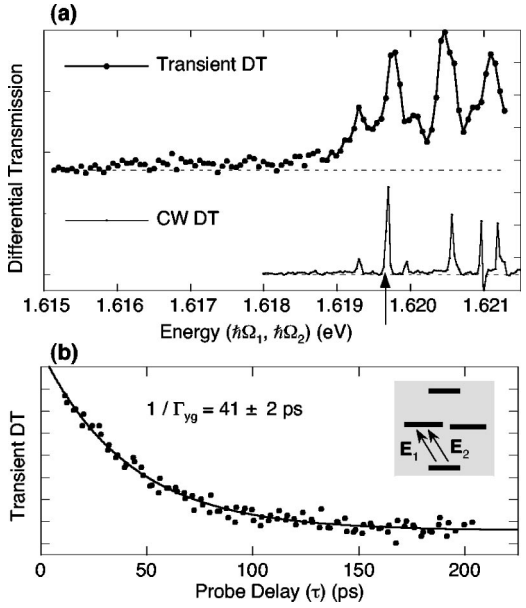

FIG. 2. Degenerate Π_y copolarized single QD DT spectrum obtained with pulsed fields (top) and cw fields (bottom) at a probe delay of about 6 ps. The dashed line denotes the zero signal level. DT vs probe delay for transient excitation tuned to the state denoted by the arrow in (a), fit to an exponential decay.

$$\dot{\rho_{by}} = -(\gamma_{by} + i\omega_{by})\rho_{by} + \frac{i\mu_{by}}{2\hbar}(\rho_{yy} - \rho_{bb})\sum_k E_k(t)e^{-i\Omega_k t} \tag{4}$$

$$\dot{\rho_{bb}} = -\Gamma_b \rho_{bb} - \left(\frac{i\mu_{yb}}{2\hbar}\rho_{by}\sum_k E_k^*(t)e^{i\Omega_k t} + \text{c.c.}\right). \tag{5}$$

The fields are written $\mathbf{E}_k(t) = 1/2(E_k(t)e^{-i\Omega_k t} + \text{c.c.})\hat{\epsilon}_k$, ω_{ij} is the frequency of the $|i\rangle \rightarrow |j\rangle$ transition, Γ_{ij} is the relaxation rate of population from $|i\rangle \rightarrow |j\rangle$, γ_{ij} is the dephasing rate of the coherence ρ_{ij}, and μ_{ij} is the magnitude of the dipole moment $\boldsymbol{\mu}_{ij}$ for the $|i\rangle \rightarrow |j\rangle$ transition. The biexciton lifetime is simply Γ_b^{-1}, where $\Gamma_b = \Gamma_{bx} + \Gamma_{by}$. For simplicity, the near symmetry of the system is used to assume $\Gamma_{bx} \approx \Gamma_{by}$, so that $\Gamma_b \approx 2\Gamma_{by}$.

Two 76-MHz synchronously pumped dye lasers with pulsewidths of about 6 ps are used to perform degenerate and nondegenerate pump-probe measurements. A picosecond pulse has both a sufficiently narrow temporal resolution to measure lifetimes of tens of picoseconds, as well as sufficiently narrow bandwidth (~ 300 μeV) so as not to excite spectrally adjacent QD states [see Fig. 2(a)]. In addition, the laser bandwidth is much larger than the fine-structure exchange splitting [$\hbar(\omega_{yg} - \omega_{xg}) \approx 20\mu$eV], but much smaller than the biexciton binding energy [$\Delta\varepsilon = \hbar(\omega_{yg} - \omega_{by}) \approx 3.5$ meV], important for the measurements discussed below.

The pump ($\mathbf{E}_1$) and probe ($\mathbf{E}_2$, delayed by a time τ with respect to the pump) are focused to a waist at the aperture. Absorption of the probe at the excitonic (or biexcitonic) resonance creates an induced third order polarization field $\mathbf{P}^{(3)}(t)$. Time-integrated homodyne detection of this polarization field with the transmitted probe represents a differential transmission (DT) signal that is proportional to the imaginary part of the amplitude of $\mathbf{P}^{(3)}(t)$. An amplitude modulation at ~ 1 MHz of both the pump and probe allows for the use of phase-sensitive lock-in detection at the difference of the two modulation frequencies.

For excitation and detection of single QD excitons, both the Π_y-polarized pump and π-polarized probe fields are tuned to a single exciton transition, $|g\rangle \rightarrow |e_y\rangle$ ($\Omega_1 = \Omega_2 = \omega_{yg}$). The pump excites the excitonic population $\rho_{yy}(t)$ to second order, which then decays back to the crystal ground state ρ_{gg} at the relaxation rate Γ_{yg}. The polarization induced by the probe is given by $\mathbf{P}^{(3)}(t) \propto [\boldsymbol{\mu}_{yg}\rho_{gy}^{(3)}(t) + \text{c.c.}]$. Other terms that could in principle contribute to the DT signal but do not involve $\rho_{yy}^{(2)}(t)$ created by the pump are either not detected because of the double chopping used, or are zero delay terms (present only during probe delays within a pulsewidth of zero delay) that are omitted in the analysis of the data. In the limit of δ-function pulses this time-integrated DT is given by

$$DT \propto \theta(\tau)e^{-\Gamma_{yg}\tau} \tag{6}$$

The step function $\theta(\tau)$ is zero for $\tau < 0$ and 1 for $\tau > 0$. This equation assumes that the QD is initially in equilibrium such that $\rho_{gg}^{(0)} \gg \rho_{yy}^{(0)}$. A nonequilibrium initial population difference created by a prepulse can be represented by an additional term $\rho_{gg}^{(0)} - \rho_{yy}^{(0)}$ in Eq. (6). This is valid for prepulse-pump delays much smaller than the decay time of the exciton population, Γ_{yg}^{-1}. Equation (6) shows that the decay of the DT signal is a direct measure of the relaxation rate of the excitonic state. This relaxation rate contains both radiative and nonradiative contributions to the exciton lifetime.

The degenerate DT obtained through a submicron aperture with Π_y-copolarized fields is shown in Fig. 2(a). The top spectrum is obtained with picosecond pulses near zero probe delay. A cw DT spectrum is shown for comparison, indicating that the resonances in the transient DT spectrum correspond to single QD states. The linewidths in the transient spectrum are broadened by the laser bandwidth, whereas the resonances in the cw spectrum are homogeneously broadened. With $\mathbf{E}_1$ and $\mathbf{E}_2$ tuned to the excitonic resonance shown by the arrow in Fig. 2(a), the transient DT/T (where T is the probe transmission at the probe modulation frequency) is approximately 2.9×10^{-4}. The decay of the DT as a function of probe delay is a direct measure of Γ_{yg} for that QD state. This data is shown in Fig. 2(b), giving $\Gamma_{yg}^{-1} = 41 \pm 2$ ps. As discussed above, data within a pulse width of zero delay could involve excitation not related to a second-order excitonic population created by the pump and is therefore excluded from the fit.

Relaxation rates were measured in this way for many other single interface fluctuation QD's, giving lifetimes between 25 and 50 ps. Similar values were observed for Π_x excitation. These numbers are comparable to dephasing rates (γ_{xg} and γ_{yg}) obtained from CW DT line shapes, consistent with previous observations that show an absence of pure dephasing processes in the decoherence of interface fluctuation QD's.[14] Calculations of the radiative lifetimes for interface fluctuation QD's are of order 100 ps[25–27], though none

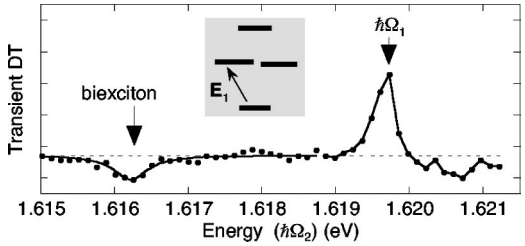

FIG. 3. Nondegenerate Π_y copolarized single QD DT spectrum obtained with a probe delay of about 6 ps with the pump ($\mathbf{E}_1$) fixed at the excitonic resonance. The dashed line denotes the zero signal level.

of these calculations are specific to the geometry of the QD's investigated in this paper. Recent measurements[21] and calculations[6] found an oscillator strength of about 90 for excitons confined to single interface fluctuation QD's of the same size as those used for the measurements in this paper. This oscillator strength can be used to directly calculate a radiative lifetime,[28] estimated to be about 80 ps for our QD's. The discrepancy between the measured lifetimes and the calculated radiative lifetimes suggests that nonradiative decay may play a role in the overall relaxation of confined excitons in our structures. It should be emphasized, however, that the DT technique used in this paper simply measures an overall relaxation rate, and cannot distinguish between radiative and nonradiative decay. Recent four-wave-mixing measurements[29] and corresponding calculations based on a relaxation model that is dominated by nonradiative decay[25] found that the exciton lifetime in interface fluctuation QD's in 3-nm-thick GaAS layers is about 20 ps. In that model, the nonradiative decay arises from phonon-assisted scattering into other exciton states.

Because the laser bandwidth is much larger than the exchange splitting of the $|e_y\rangle$ and $|e_x\rangle$ states, a cross-linearly polarized pump and probe (Π_y and Π_x) can be used to study relaxation between these two states. The detailed results of this study were reported elsewhere,[30] but showed that these spin relaxation times are on the order of hundreds of picoseconds. Within the noise level of the copolarized data shown above, such times are too large to be observable.

The resonant excitation of a biexcitonic state in a single QD requires the absorption of two photons, whose energies sum to the ground to biexciton transition energy $[\hbar(\omega_{yg}+\omega_{by})]$. Normal DT (such as that in Fig. 2) corresponds to increased probe transmission induced by a saturating pump. However, a pump tuned to the excitonic transition will induce absorption (not transmission) of a probe tuned to the biexcitonic transition; in the absence of the pump, the excitonic state is unoccupied, and the probe is unaffected by the biexcitonic transition. Induced absorption appears as negative signal in a DT measurement.

This biexcitonic resonance is shown in the copolarized nondegenerate transient DT spectrum in Fig. 3, obtained at a probe delay of about 6 ps. The experiment is identical to that of Fig. 2(a), except here the pump field ($\mathbf{E}_1$) is fixed at the excitonic transition ($\Omega_1=\omega_{yg}$) while the energy of the probe is tuned. About 3.4 meV lower in energy, a dip in the DT is evidence of induced probe absorption due to the creation of

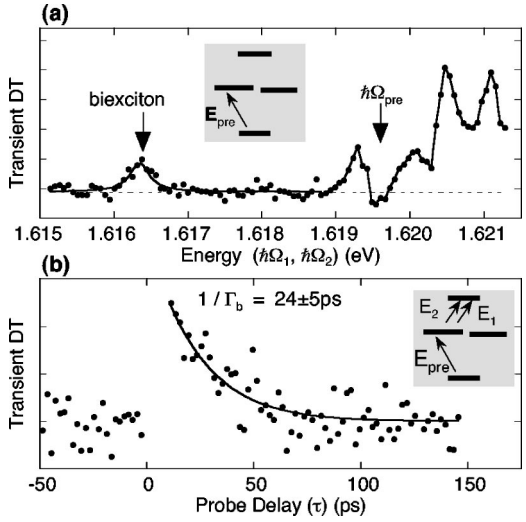

FIG. 4. (a) Same as Fig. 2(a), but with a Π_y prepulse tuned to the excitonic state shown with the arrow. The dashed line denotes the zero signal level. (b) DT vs probe delay with both the pump and probe tuned to the biexcitonic resonance for a system initially excited by the prepulse. The line is a fit to Eq. (7), with Γ_{yg}^{-1} set to 41 ps.

the biexciton. This biexciton binding energy is in excellent agreement with recent measurements using cw nondegenerate DT.[31] By leaving the probe energy fixed at the biexciton transition, ($\Omega_2=\omega_{by}$), the decay of the induced absorption can be measured by varying the probe delay. Since this third-order measurement does not probe the biexciton population ($\rho_{bb}^{(4)}$ is the lowest-order biexciton population term) the induced absorption signal decays at the single exciton decay rate, Γ_{yg}. The measured decay time is found to be consistent with that measured for Γ_{yg}^{-1} in Fig. 2(b). The induced absorption signal would be expected to disappear in the case that (1) the probe arrived before the pump, and (2) the pump and probe were oppositely linearly polarized (see Fig. 1). In both cases, no induced absorption signal was observed at the biexcitonic transition.

A measurement of the biexciton lifetime Γ_b^{-1} is accomplished with a strong (unchopped) prepulse ($\mathbf{E}_{pre}$) tuned to the excitonic transition, followed by excitation by the pump and probe at the biexcitonic transition. The purpose of the prepulse is to populate the $|e_y\rangle$ state for subsequent pump-probe excitation of the biexcitonic transition. Then, the two-level system $|e_y\rangle \rightarrow |b\rangle$ is analogous to the $|g\rangle \rightarrow |e_y\rangle$ two-level system, with the important difference that $|g\rangle$ does not decay whereas $|e_y\rangle$ does. The measured degenerate transient DT spectrum for a $-6\text{-}ps$ prepulse tuned to the same excitonic transition as that studied in Figs. 2 and 3 ($\Omega_{pre}=\omega_{yg}$) is shown in Fig. 4(a). The effects of the prepulse are purely incoherent, due to the lack of mutual coherence between the prepulse laser and the pump/probe laser. The DT from the excitonic transition is now negative, a consequence of the strong prepulse that has slightly inverted the excitonic transition ($\rho_{gg}^{(0)}-\rho_{yy}^{(0)}<0$). The ability to fully invert the excitonic system with a π pulse has important implications for quantum logic gates based on single QD's, and is discussed

in detail elsewhere.[32] Figure 4(a) also indicates that the DT at the biexcitonic transition is positive in the presence of a prepulse at the excitonic transition. This is expected because the saturating pump induces increased probe transmission, similar to the excitonic case.

Figure 4(b) shows the decay of the DT as the probe delay is increased with both the pump and probe fixed at the biexcitonic resonance ($\Omega_1=\Omega_2=\omega_{by}$). In this case, $\mathbf{P}^{(3)}(t) \propto[\vec{\mu}_{by}\rho_{yb}^{(3)}(t)+\text{c.c}]$, similar to the case of the nondegenerate DT of Fig. 3(b). The time-integrated homodyne detected DT is then (again assuming δ-function pulses)

$$DT\propto(\rho_{yy}^{(0)}-\rho_{bb}^{(0)})\,\theta(\tau)\left[e^{-\Gamma_b\tau}\left(1+\frac{\Gamma_b}{2(\Gamma_b-\Gamma_{yg})}\right)+e^{-\Gamma_{yg}\tau}\left(1-\frac{\Gamma_b}{2(\Gamma_b-\Gamma_{yg})}\right)\right]. \quad (7)$$

Note that the signal depends on the population difference $(\rho_{yy}^{(0)}-\rho_{bb}^{(0)})$ which is zero without the Π_y prepulse tuned to the excitonic transition. Like Eq. (6), this formulation neglects the DT signal near zero delay, which is also ignored in the fitting to the data shown below. The DT obtained in this way can be biexponential due to decay of both the $|b\rangle$ and $|e_y\rangle$ states. In the case that the biexciton relaxation rate is determined by the relaxation of individual excitons, $\Gamma_b \approx 2\Gamma_{yg}$. In this case, $\Gamma_b/2(\Gamma_b-\Gamma_{yg})\approx 1$, and the decay of the DT is a single exponential with decay rate Γ_b.

The data in Fig. 4(b) is fit to Eq. (7), where Γ_{yg}^{-1} is set to 41 ps. The fit is shown in the plot and yields $\Gamma_b^{-1}=24\pm 5$ ps. Thus the decay of the DT is consistent with a biexciton lifetime that is one-half the single exciton lifetime. Based on the above discussion, this would imply that the data can be fit by a single exponential of decay time Γ_b^{-1}. Within the noise level of the data, this is true, yielding a biexciton lifetime of $\Gamma_b^{-1}=22\pm 4$ ps, consistent with the above result. Measurements of exciton and biexciton decay rates in a similar QD are also consistent with a biexciton lifetime that is one-half the exciton lifetime, with $\Gamma_{yg}^{-1}=29\pm 2$ ps and $\Gamma_b^{-1}=16.8\pm 4$ ps [from fitting Eq. (7)] and $\Gamma_b^{-1}=14.5\pm 4$ ps (from fitting a single exponential).

The lifetimes of biexcitons in GaAs quantum wells have been shown to be one-half that of excitons,[33] which is consistent with the point of view that since the biexciton contains two excitons, the probability per unit time for decay is twice that of a single exciton. Other studies, however, have contradicted these findings; for example, low-temperature four-wave-mixing measurements of dephasing rates in a single GaAs QW found $\gamma_{bg}\approx\gamma_{yg}$.[34,35] These measurements imply that the biexciton lifetime Γ_b^{-1} [equal to $(2\gamma_{bg})^{-1}$ in the absence of pure dephasing] is approximately equal to the exciton lifetime Γ_{yg}^{-1} [equal to $(2\gamma_{yg})^{-1}$ in the absence of pure dephasing]. Note that this would be true for a lifetime limited by either radiative or nonradiative decay.

Biexciton decay in a QW can be complicated by dissociation into a two-exciton state without radiative recombination. Excitons localized in all three dimensions, however, should be free from such effects because of the complete discretization of states and the enhanced biexciton binding energy. Nevertheless, previously reported theory and measurements regarding the ratio of the exciton lifetime to biexciton lifetime in QD's are not in agreement. In a strongly localized QD the Coulomb correlation between the carriers has been shown theoretically to lead to a biexciton lifetime that is larger than the exciton lifetime.[36] Previous measurements using time-resolved PL spectroscopy of single II-VI QD's have found the biexcitonic and excitonic radiative decay rates for the same quantum dot to be nearly equal.[11] The model used in that work, though, does not include both bright exciton states in the decay channels of the biexciton. On the other hand, measurements in InAs self-assembled QD's have found an exciton to biexciton radiative lifetime ratio of 2,[37] consistent with theoretical predictions[13] and in agreement with the reasoning discussed above. To the authors' knowledge, no specific theoretical prediction exists for the ratio of exciton to biexciton lifetime in a QD system where nonradiative decay may be significant.

Biexcitonic dephasing rates (the decay rate of the coherences ρ_{by} and ρ_{bg}) of similar interface fluctuation QD's have been measured using nondegenerate CW DT,[31] and found to be of the same order as the relaxation rates reported here. In addition, the biexciton signal strength near zero delay can be used to estimate the ratio of the biexciton dipole moment to the exciton dipole moment, μ_{by}/μ_{yg}. The level of inversion of the exciton shown by Fig. 4(a) indicates that the prepulse creates the ρ_{yy} exciton with a probability of approximately 0.56. These data, coupled with the ratio of the biexciton signal strength in Fig. 4 to the exciton signal strength in Fig. 2 (and assuming that the measured lifetimes are purely radiative), implies that $\mu_{by}/\mu_{yg}=0.85\pm 0.08$. This ratio is consistent with preliminary estimates of the biexciton dipole moment from Rabi oscillation experiments (data not shown), though it is in slight disagreement with a predicted ratio of approximately 1.4 for spherical GaAs quantum dots of similar volume.[38,39]

Current proposals to implement quantum information processing using two interacting excitons in QD's[2,17,18] are based on the assumption of biexciton relaxation and dephasing rates on the same order as (or smaller than) single exciton rates. The measurements described in this work support this assumption. In addition, nonlinear pump-probe measurements are an important part of the proposals of Chen *et al.*,[17,18] due to their ability to resonantly probe only the QD state under investigation. In particular, the three-pulse technique described in this work, combined with other recent results demonstrating single qubit rotations in single QD's,[32] are important steps toward the eventual implementation of a quantum logic gate using interacting excitons in quantum dots.

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*Current address: Naval Research Laboratory, Washington, DC 20375

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