Comment on "coherent nonlinear optical response of single quantum dots studied by ultrafast near-field spectroscopy"

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Comment on "Coherent Nonlinear Optical Response of Single Quantum Dots Studied by Ultrafast Near-Field Spectroscopy"

In a recent Letter [1,2], Guenther *et al.* report on spectrally resolved pump-probe femtosecond reflectivity measurements in single quantum dots. For negative pump-probe delays, they observe spectral oscillations due to perturbed free induction decay (FID) of the excitonic polarization. Using the experimental observation that the spectrally integrated differential reflectivity vanishes at negative delays, they rule out the contribution from pump-induced changes in the oscillator strength and thus conclude that excitation-induced dephasing (EID) is the leading mechanism to the perturbation of the FID. In this Comment, I will show that this reasoning is incorrect.

Spectral oscillations in negative-delay pump-probe spectra have been reported previously in a great variety of femtosecond experiments, for example, spectral hole burning [3,4], exciton bleaching [3,5], exciton optical Stark shift [3,6,7], and infrared spectroscopy [8]. In all of these cases, the frequency-integrated differential spectra vanish for negative delays. Indeed, using the notations of Ref. [1] [see Eq. (1)], the differential reflectivity reads

$$\Delta R(\omega, \Delta t) \propto \mathrm{Re}\tilde{E}_R^*(\omega)\Delta\tilde{E}_{QD}(\omega, \Delta t), \qquad (1)$$

where Δt is the pump-probe time delay. $\tilde{E}_R(\omega)$ is the reflected probe and $\Delta\tilde{E}_{QD}(\omega, \Delta t) = \tilde{E}_{QD}(\omega, \Delta t) - \tilde{E}_{QD,0}(\omega)$ is the pump-induced change in the radiated field. Figure 1 shows these fields in time domain. The integral of $\Delta R(\omega, \Delta t)$ over frequency can be computed using the Parseval-Plancherel theorem, which yields the time-domain integral of $E_R^*(t)\Delta E_{QD}(t, \Delta t)$. Because of the causality principle, $\Delta E_{QD}(t, \Delta t)$ takes nonzero values only after the sample has been excited by the pump, which means that $\Delta E_{QD}(t, \Delta t)$ and $E_R(t)$ do not overlap in the case of negative Δt (see Fig. 1). Hence, although they do produce frequency-resolved interferences, these two fields cannot produce frequency-integrated inteferences and the integral always vanishes [4,5]. Therefore, it is incorrect to rule out the contribution of oscillator strength change (i.e., exciton bleaching) on the grounds of a vanishing integrated reflectivity.

Using EID only, Guenther *et al.* obtain an excellent fit of negative-Δt spectra by assuming that the dephasing time changes from an initial value of 15 ps down to 3 ps after the pump pulse is absorbed (dashed line in Fig. 1). However, one can also interpret this pump-induced damping in a very different way: the damping can be due to the buildup of exciton bleaching with a time constant of 3 ps. Since exciton bleaching is indeed mentioned in Ref. [1] in order to explain the spectra recorded for positive Δt, there is no reason it should not contribute to the negative-Δt spectra as well.

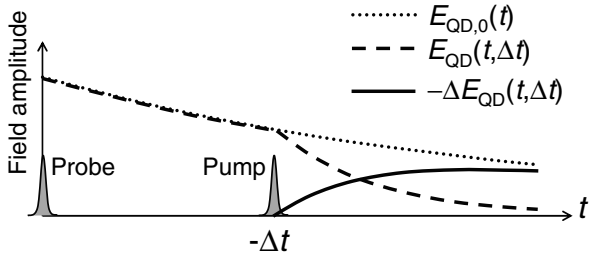

FIG. 1. Time-domain representation of the fields radiated by the quantum dot for a negative pump-probe delay.

To summarize, *any* perturbation to the FID, including an exciton bleaching, results in a vanishing spectrally integrated signal for negative Δt. Furthermore, two processes as different as EID and a slow buildup of exciton bleaching can result in identical negative-Δt spectra, while they do correspond to different positive-Δt spectra. Consequently, although many-body effects (including EID) undoubtedly play an important role in quantum dot excitonic dynamics, the coherent oscillations reported in Ref. [1] do not provide on their own a direct evidence that EID is the dominant mechanism.

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