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Mechanism for enhanced optical nonlinearities and bistability by combined dielectric–electronic confinement in semiconductor microcrystallites

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We propose a new type of mechanism for enhanced optical nonlinearities and intrinsic optical bistability that relies on the combination of intrinsic feedback due to local field effects and excitonic resonances in semiconductor crystallites. These effects will be further enhanced by quantum confinement in small crystallites.

Lately the prospect of digital optical processing has excited much interest in the study of optical switches and logic elements based on optical bistability and related effects. The operation of these devices generally requires the combination of a strong optical nonlinearity and a feedback mechanism.[1,2] Most often optically bistable devices use an external cavity to provide the necessary feedback. However, internal feedback based on light-induced modification of the intrinsic properties of nonlinear media has been considered,[2,3] and some examples of this class have been demonstrated.[2]

In this Letter we propose a novel approach to optical bistability that exploits the combination of intrinsic feedback and enhanced nonlinearities that we anticipate for semiconductor quantum dots (QD's), i.e., microcrystallites with dimensions of the order of the carrier's de Broglie wavelengths.

Optical bistability due to local field effects in bulk material was considered previously.[4–6] Local field effects in small particles arise from dielectric confinement; they are qualitatively different from those seen in bulk material and are responsible for surface-enhanced Raman scattering[7] as well as for the large nonlinearities of metal- and semiconductor-doped glasses.[8,9] Excitonic behavior observable in many semiconductors at low temperature is enhanced by quantum size effects in microstructures with dimensions of the order of or smaller than the bulk exciton Bohr diameter. For example, large room-temperature nonlinearities have been observed in semiconductor quantum-well structures in which, because of the confinement in ultrathin layers, excitons become quasi two dimensional.[10] Even larger enhancement of excitonic nonlinearities is expected in one- and zero-dimensional quantum confined systems.

The field inside a small particle whose dimensions are small compared with the optical wavelength but large compared with atomic dimensions is related to the external field by $E_{in} = fE_{out}$, where the local field factor is given by[11] $f = [1 + A(\epsilon - 1)]^{-1}$, $\epsilon = \epsilon_1 + i\epsilon_2$ is the bulk dielectric constant relative to the surrounding medium, and A is the so-called demagnetization factor, which depends only on the geometry of the particle. For a small sphere, $A = \frac{1}{3}$. Thus the intensity inside the particle is $I_{in} = FI_{out}$, with

$$F = |f|^2 = \frac{A^{-2}}{(\epsilon_1 - 1 + A^{-1})^2 + \epsilon_2{}^2}, \tag{1}$$

and the particle exhibits an absorption governed by the imaginary part of effective dielectric constant; $\tilde{\epsilon}_2 = F\epsilon_2$.

The structure of the electronic levels of microscopic semiconductor particles (compared with that of the bulk material) is radically modified by both dielectric and electronic confinement. For particles that consist of more than a few tens of atoms, the effective mass approximation should hold, and quantum size effects appear whenever the dimension of the particle becomes comparable with the de Broglie wavelength of the carriers. In this case extended electronic states are forbidden by the quantum confinement, and electronic bands transform into a set of discrete levels.[12] Excitonic interaction will produce shifts in the optical transitions, but it will not affect the discrete level structure of the optical spectrum.

Excitonic optical nonlinearities in bulk semiconductors arise from many-body effects.[13] However, in two-dimensional systems, photocarrier-induced direct Coulomb screening is already strongly reduced, and the mechanisms governing the optical nonlinearities are the Pauli-principle-related phase-space filling (or -state filling) and exchange interaction.[17] As the dimensionality is further reduced to total confinement in QD's, photocarrier screening becomes completely negligible and only the Pauli-principle state filling effect remains active. Consequently, the absorption saturation of a given interband transition, although excitonic, will behave as a simple two-level absorber, with the absorption saturated when the upper state is (half) occupied. Further details on the theory of saturation of a QD will be published elsewhere.[15]

In summary, we can expect the absorption spectrum of QD's to consist of a set of discrete lines with enhanced oscillator strength and saturation governed by energy-level occupation. Consequently it is legitimate to model the dielectric constant close to an exci-

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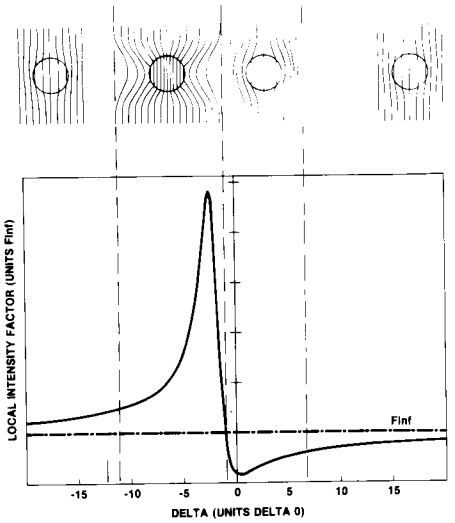

Fig. 1. Universal shape of the local intensity factor profile near a Lorentzian resonance. As shown in the upper part of the figure, far from resonance (on both sides) the field in the particle is smaller than outside; near the resonance on one side the field is concentrated in the particle, whereas on the other side the field cannot penetrate the particle.

tonic resonance by a background plus a saturable two-level resonance.[16]

In what follows we examine the effect of dielectric confinement on a small particle with a dielectric constant of the form

$$\epsilon = \epsilon_\infty + \beta \frac{\delta + i}{1 + \delta^2 + J}, \tag{2}$$

where ϵ_∞ is the dielectric constant of the background; δ is the normalized detuning, which can be written as $\delta = (\Omega - \omega)/\Gamma$ in terms of the optical frequency ω and of the resonance frequency Ω and width Γ; β is the resonance contribution to the dielectric constant; and $J = I/I_s$ is the intensity normalized to the saturation intensity.

Let us first consider the behavior of F for ϵ given by Eq. (2) at very small excitations, i.e., $J = 0$. When expressed in terms of the shape-renormalized nonresonant susceptibility, $\kappa = \epsilon_\infty - 1 + A^{-1}$, F is found to have the well-known Fano–Beutler profile[17]

$$F = F_\infty \frac{1 + \delta^2}{1 + (\delta - 2\delta_0)^2}, \tag{3}$$

where $F_\infty = A^{-2}/\kappa^2$ is the local intensity factor far from the resonance. The characteristic detuning $\delta_0 = -\beta/\kappa$ measures the resonance oscillator strength relative to the nonresonant contribution. Cast in this form, the profile of F is universal; it is plotted in Fig. 1. Far from resonance ($\epsilon_1 > 1$, $\epsilon_2 \sim 0$), the field inside the particle is smaller than outside ($F < 1$). However, large changes occur in a small range around the resonance. For small negative values of δ (photon energies above the resonance), the field is concentrated inside the particle, whereas immediately on the other side (i.e., photon energies below the resonance), the field is actually expelled from the particle.

The mechanism for intrinsic optical feedback can now be described. In the presence of saturation the dielectric constant inside the particle depends on the intensity. If the intensity is changed the local field factor is modified; this in turn changes the intensity inside the particle, which changes the dielectric constant, and so on. First this increases the dependence of ϵ on the incident intensity, hence giving enhanced optical nonlinearities. Furthermore, depending on the value of the detuning, the feedback can be positive or negative; hence the appearance of regions of optical bistability. Although we consider here the specific case of Lorenzian resonance, it is most likely that this mechanism would hold for other functional forms of ϵ.[18]

The correct description of saturable crystallites requires some care. The intensity inside the particle is the local intensity, so in the expression of the dielectric constant, Eq. (2), J should be replaced by FJ, where F is given by Eq. (3) and, in $J = I/I_s$, I is now the external intensity. This determines the self-consistent condition that the dielectric constant must satisfy:

$$\epsilon = \epsilon_\infty + \beta \frac{\delta + i}{1 + \delta^2 \dfrac{A^{-2}J}{(\epsilon_1 - 1 + A^{-1})^2 + \epsilon_2^2}}. \tag{4}$$

In the case of Lorentzian functional form, Eq. (2), an analytical solution of this equation can be found.

In order to study the behavior of the QD response, we have solved Eq. (5a) by using parameters approximately equal to those of CdS (a compound that has been shown to form microcrystallites with evidence of enhanced excitonic behavior from quantum size effects,[19,20] although we will use only the bulk parameters here as a worst case. We assume spherical QD's. In Fig. 2 we have plotted the frequency dependence of ϵ_2 and ϵ_1 for $J = 20$ (heavy lines) and for comparison for $J = 0$ (light lines). Away from resonance as well as for small positive δ, little saturation occurs. Far from resonance this straightforward behavior is enhanced

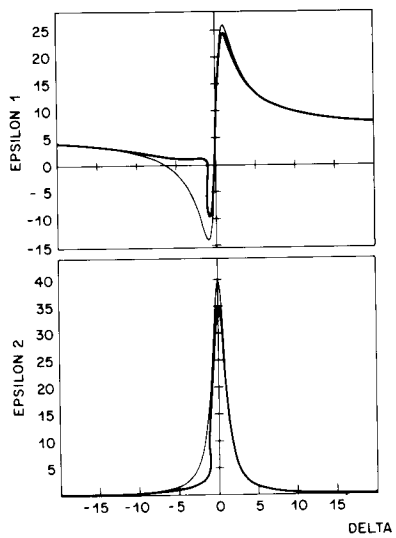

Fig. 2. Plots of the frequency dependence of the real and imaginary parts of the dielectric constant for $J = 20$ (heavy lines) and for $J = 0$ (light lines). The material parameters have the rounded-up values of CdS: $\epsilon_\infty = 6$, $\Gamma = 0.4$ meV, $\Omega = 2.555$ eV, and $\beta = 40$ (corresponding to an oscillator strength $\Gamma\beta = 1.6 \times 10^{-1}$).

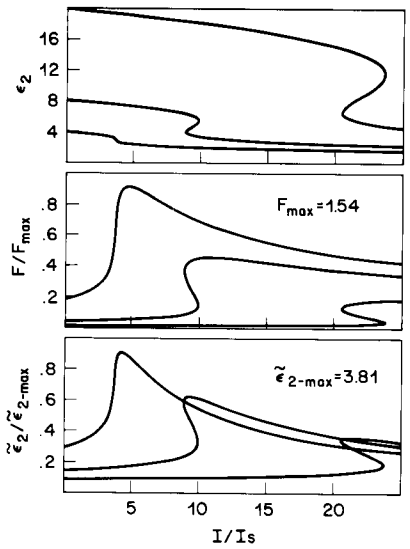

Fig. 3. Plots of (a) the imaginary part of the dielectric constant, of (b) the local intensity factor, and of (c) the imaginary part of the effective dielectric constant in a CdS spherical QD as a function of the light intensity normalized to the saturation intensity.

only by the value of $F < 1$. However, at resonance and immediately close to it, for $\delta > 0$ the absence of saturation is due to the fact that the field does not penetrate the QD's. For small $\delta < 0$, for which the field is attracted into the QD's, the strong effect of dielectric confinement is apparent; both ϵ_1 and ϵ_2 exhibit strong saturation and even become multivalued functions. We have plotted in Fig. 3 the quantities ϵ_2, $\tilde{\epsilon}_2$, and F versus intensity for three values of the detunings $\delta = -1, -2, -3$. The characteristic bistability S shape is seen on the curves for $\delta = -1, -2$, whereas for $\delta = -3$ the detuning is too large and the curve shows only a kink. Close to resonance the local field factor is strongly dependent on both δ and J; thus $\tilde{\epsilon}_2$ can show enhanced or reduced variations compared with ϵ_2. Because of the reversal of sign in ϵ_1, the orientation of the sense of the S in the ϵ_2 curve is opposed to that of the two other ones. Finally, even at detunings where intrinsic bistability is not possible strong absorptive nonlinearity can occur, an interesting aspect in its own right. To evaluate the magnitude of these effects we use the CdS parameters and assume a 10^{-3} filling factor. We find that, without assuming further quantum confinement enhancement of the oscillator strength over that of the bulk, a change of several hundreds of inverse centimeters in the absorption coefficient at the bistability transition or in differential gain are easily obtained.

In conclusion, we have identified a new type of enhancement of optical nonlinearity and intrinsic optical bistability that arises from the combination of local field effects from dielectric confinement and saturable excitonic resonances in semiconductor particles. Furthermore, we predict that these effects will be greatly enhanced when the particles are so small that they also exhibit quantum confinement. This mechanism presents a number of novel features. It should be observable in single crystallite, thus avoiding inhomogeneous resonance broadening due to size fluctuation. It should be also observable if the particles are arranged in a matrix. In this case the interaction among particles can become large enough to produce coherent effects. However, the main conclusions of this Letter should remain valid since the principal consequence of particle interaction is to renormalize the geometrical factor A.[21]

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Note added in proof: Since the original submission of the manuscript of this Letter to another journal a paper has appeared[22] that discusses a similar optical bistability occurring close to plasmon resonances in microparticles.

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