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A Lossless Negative Dielectric Constant from Quantum Dot Exciton Polaritons

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ABSTRACT

Prospects for a lossless negative dielectric constant material for optical devices are studied. Simulations show that with sufficient gain, a mixture of two semiconductor quantum dots (QDs) can produce an effective dielectric constant that is lossless and negative. This permits, in concept, arbitrarily small scaling of the optical mode volume, a major goal in the field of nanophotonics. The proposed implementation of a lossless negative dielectric constant material based on colloidal QDs opens a tractable path.

Achieving a negative dielectric constant at an (optical) frequency ω where the loss goes to zero, i.e., $\varepsilon = \varepsilon' + i\varepsilon''$ with $\varepsilon' < 0$ and $\varepsilon'' = 0$, is of fundamental importance in nanophotonics. A negative refractive index requires both negative dielectric constant and permeability.[1] Imaging with a negative index slab has a resolution that is limited only by the loss.[2,3] A plasmon−polariton surface wave exists at a metal–insulator interface because of the negative dielectric constant.[4] With two interacting surfaces, a metal–insulator–metal waveguide mode exists, and this propagates for arbitrarily small gaps. As the gap reduces, the wavelength reduces for a fixed vacuum wavelength. It is thus possible in principle to shrink the size of optical components. Unfortunately, the loss increases as the waveguide mode wavelength reduces. A means to offset this loss would offer the prospect of achieving arbitrarily small mode volumes and hence high density optical elements and circuits.

Resonant states in semiconductors provide a source for negative dielectric constant provided that the dipole strength and oscillator density are adequate to offset the background. Furthermore, semiconductors offer the prospect of pumping, either optically or electrically, to achieve a gain mechanism that can offset the loss. We address the dielectric constant of exciton polaritons in a mixture of semiconductor quantum dots (QDs) and then present the influence of gain on the homogenized dielectric constant in order to describe the required QD density and pumping to achieve $\varepsilon' < 0$ and $\varepsilon'' = 0$.

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There is a large body of work describing the scattering of light from low-dimensional quantum structures,[5–9] and many-particle excitonic effects.[10–15] Here we calculate the polarization of the QD and the energy dissipation of the incident light due to the exciton excitation in the QD by scattering theory and the generalized Golden rule[16] in order to obtain QD's effective dielectric constant ε_{QD}. QDs will then be immersed into a background material, either a solid-state material or a polymer matrix. The effective ε of the composite system (QDs plus the background matrix) will be estimated using the dielectric theory of Maxwell-Garnet.[17,18]

For an optical absorption process induced by an external electromagnetic field $E(r, t)$, an allowed electric dipole transition creates an exciton in an intrinsic QD, $\Psi_n(r_e, r_h)$, from the filled valence band; The wave function of the initial state is simply unity, normally denoted by Ψ_0 in the formalism of second quantization.[11,19] Here r_e and r_h denote the electron and the hole, respectively. In the following, H_0 denotes the Hamiltonian of the exciton in the absence of $E(r, t)$. $\langle\Psi_0|H_0|\Psi_0\rangle = 0$ is the energy of Ψ_0, $\langle\Psi_n|H_0|\Psi_n\rangle = \hbar\omega_n$ is the energy of the exciton state n. The first-order perturbation Hamiltonian induced by $E(r, t)$ is $H_1 = \int \mathrm{d}(r)\cdot E(r, t)\, \mathrm{d}r$, where $\mathrm{d}(r) = -er_e\, \delta(r - r_e) + er_h\, \delta(r - r_h)$is the dipole-moment operator. We express the time-dependent wave function

$$|r_e, r_h, t\rangle = a_0(t)|\psi_0(r_e, r_h)\rangle + \sum_n c_n(t)|\psi_n(r_e, r_h, t)\rangle \quad (1)$$

where $\Psi_n(r_e, r_h, t) = \Psi_n(r_e, r_h)e^{-i\omega_n t}$. In the above equation, $a_0(t) = a_0^{(0)}$ for $t < 0$, which denotes the occupation probability of the QD ground state before we switch on $E(r, t)$ at $t = 0$. By inserting eq 1 into the time-dependent Schrödinger equation,

$$i\hbar\frac{dc_n(t)}{dt} = a_0(t)\langle \Psi_n(r_e, r_h)| \int d(r)\cdot E(r, \omega)\, dr|\Psi_0(r_e, r_h)\rangle\ e^{i\omega_n t} \quad (2)$$

By expressing the optical radiation as

$$E(r, t) = E(r, \omega)\, e^{-i\omega t} + \text{ c.c.} \quad (3)$$

where "c.c." denotes "complex conjugate", the first-order perturbation results in

$$i\hbar\frac{dc_n(t)}{dt} = a_0(t)\langle \Psi_n(r_e, r_h)| \int d(r)\cdot E(r, \omega)\, dr|\Psi_0(r_e, r_h)\rangle\ e^{i(\omega_n-\omega)t} \quad (4)$$

Here we have neglected the term corresponding to the less-likely photon emission during the creation process of the exciton. By the scattering theory and the generalized Golden rule,[16] we obtain the following steady-state solution

$$c_n(t) = a_0^{(0)}\frac{e^{i(\omega_n-\omega)t}}{\hbar(\omega_n-\omega-i\gamma)}\langle \Psi_n(r_e, r_h)| \int d(r)\cdot E(r, \omega)\, dr|\Psi_0(r_e, r_h)\rangle \quad (5)$$

where γ denotes the damping rate of the excitonic state.

We consider colloidal II−VI QDs[20] (see more discussions below about their unique properties for lossless negative dielectric constant applications). Because of the large electron–phonon interaction and thus the unltrafast relaxation from excited exciton states to the exciton ground state,[21] we only need to consider the dominant exciton ground state. Furthermore, the radius of the QDs under investigation, R, about 10 nm, is on the order of the exciton Bohr radius a_B (about 5 nm in II−V and III−V semiconductors); one can neglect the internal motion of the exciton in the QD and the exciton ground state is described as

$$\Psi_n(r_e, r_h) = \frac{1}{\sqrt{2\pi R}|r-a|}\sin\left(\frac{\pi|r-a|}{R}\right)\frac{1}{\sqrt{\pi a_B^3}}e^{-\frac{|r_e-r_h|}{a_B}} \quad (6)$$

for $|r - a| \le R$, where a is the center of the QD, $r = (m_e^* r_e + m_h^* r_h)/(m_e^* + m_h^*)$, m_e^* and m_h^* are effective masses of electron and hole. For such an exciton wave function and for the electromagnetic field of eq 3, the excitonic contribution to the dielectric polarization $P(r, t) = \langle r_e, r_h, t| d(r)|r_e, r_h, t\rangle$ becomes

$$P(r, \omega) = \frac{e^2 p_{cv}}{\hbar(\omega_n-\omega-i\gamma)\omega_n^2 m_0^2}\Psi_n(r, r)\int \Psi_n(r', r')p_{cv}\cdot E(r', \omega)\, dr' + \text{c.c.} \quad (7)$$

where p_{cv} is the optical dipole momentum between conduction and valence band edge states and m_0 the electron rest mass.[13]

An effective permittivity is defined for the QD exciton by writing $D(r, \omega) = \varepsilon_\infty E(r, \omega) + P(r, \omega) = \varepsilon_{QD}(r, \omega)E(r, \omega)$, where ε_∞ is the background dielectric constant of the QD material. By eqs 6 and 7 (neglecting the "c.c." term),

$$\epsilon_{QD}(r, \omega) = \epsilon_\infty\left[1 + \frac{2(2|a_0^{(0)}|^2-1)\omega_{LT}}{\omega_n-\omega-i\gamma}\frac{\sin\alpha}{\alpha}\right] \quad (8)$$

where $\alpha = \pi|r - a|/R$. $\varepsilon_\infty\omega_{LT}a_B^3 = e^2p_{cv}^2/\pi\hbar\omega_n^2m_0^2$. A large effective permittivity is expected in the vicinity of ω_n, which led to the proposal of a subwavelength QD waveguide.[22] In obtaining eq 8, it is assumed that the electric field inside the QD is uniform because the wavelength of interest is about 1 μm, whereas the QD radius is about 10 nm or less. Furthermore we have neglected the "c.c." term in eq 7 when obtaining eq 8. By doing so we have introduced an imaginary part to the effective permittivity. It is easy to show that the energy loss of $E(r, \omega)$ due to the QD exciton excitation is exactly expressed by the imaginary part of eq 8. This of course is nothing more than the Kramer−Kronig relationship. Notice that pseudopotential calculations of dielectric constants were performed for small Si QDs (radius < 2 nm),[23] whereas we adopt here the effective medium approximation to describe the exciton state confined in the QD that is relatively large (10 nm).[11]

By averaging $\varepsilon_{QD}(r, \omega)$ over the QD space, we obtain the expression for the effective permittivity of the QD due to the ground-state exciton excited by $E(r, \omega)$

$$\varepsilon_{QD}(\omega) = \varepsilon_\infty\left[1 + \frac{6(2|a_0^{(0)}|^2-1)\omega_{LT}}{\pi^2(\omega_n-\omega-i\gamma)}\right] \quad (9)$$

Note that $|a_0^{(0)}|^2 > 0.5$ indicates an optical loss and $|a_0^{(0)}|^2 < 0.5$ optical gain for $E(r, \omega)$.

A desirable QD should have a large value of ω_{LT} in order to achieve $\varepsilon' < 0$. GaAs and InAs are commonly used as QD materials grown on semiconductor substrates, but their ω_{LT} are rather small.[24] II−VI materials have large ω_{LT}.[25] The size of colloidal II−VI QDs is very well controlled during the chemical synthetic processes.[26] Many colloidal II−VI QDs used in bioimaging applications are core−multishell formed[20] so that possible effects of the surface and external environment on the exciton in the central core region are minimal. For such QD assemblies, the widths of photoluminescence spectra match with the size distributions so that $\hbar\gamma$ is on the order of 1.0 meV.[21] In this work, II−VI PbSe/ZnSe QDs are assumed. Figure 1 shows the calculated dielectric constant for a set of QDs before and after some fraction of the exciton states become populated. From here and throughout, ε' and ε'' are expressed in units of ε_0.

We require $\varepsilon'' = 0$ at some frequencies in order to achieve lossless waveguide modes. To investigate prospects for obtaining isotropic material, we consider distributions of two types of QDs, one providing an absorptive resonance and the other gain through either optical or electrical pumping. Consider a PbSe/ZnSe QD assembly immersed in a medium of dielectric constant ε_i. The macroscopic dielectric constant ε for the ensemble of the QDs can be described by the dielectric theory of Maxwell-Garnet (MG),[17,18] which for two QD species can be written as

$$\frac{\varepsilon-\varepsilon_i}{\varepsilon+2\varepsilon_i} = x_1\frac{\varepsilon_{QD1}-\varepsilon_i}{\varepsilon_{QD1}+2\varepsilon_i} + x_2\frac{\varepsilon_{QD2}-\varepsilon_i}{\varepsilon_{QD2}+2\varepsilon_i} \quad (10)$$

where x_i is the volume fraction of the ith QD species. While eq 10 works best for x_i less than 0.4, there is evidence that useful information can be available with higher concentrations.[27]

We consider two types of the background medium, semiconductor material with ε_i about 10 and polymer around

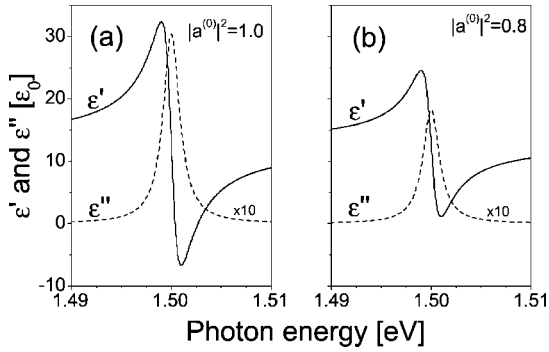

Figure 1. Dielectric coefficients of the QD before and after the QD becomes populated. $\varepsilon_\infty = 12.8$, $\hbar\omega_n = 1.5$ eV, $\hbar\omega_{LT} = 5$ meV, and $\hbar\gamma = 1$ meV for typical II−VI semiconductor QDs, e.g., colloidal PbSe/ZnSe QDs.

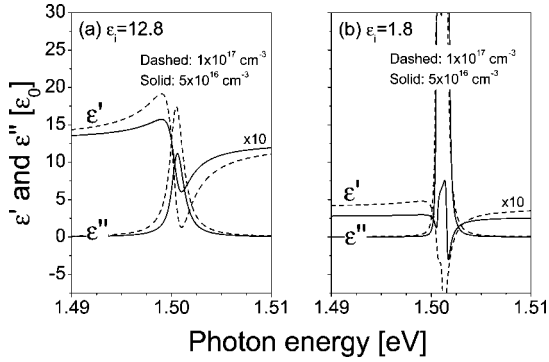

Figure 2. Effective dielectric constant for an ensemble of QDs immersed in a medium of dielectric constant ε_i. The QD density are 5×10^{16} and 1×10^{17} cm^{-3}, respectively, and correspond to volume percentages of 20.5% and 41%. (a) QDs embedded in semiconductor substrate $\varepsilon_i = 12.8$. (b) QDs immersed in a medium with $\varepsilon_i = 1.8$, e.g., a conducting polymer.

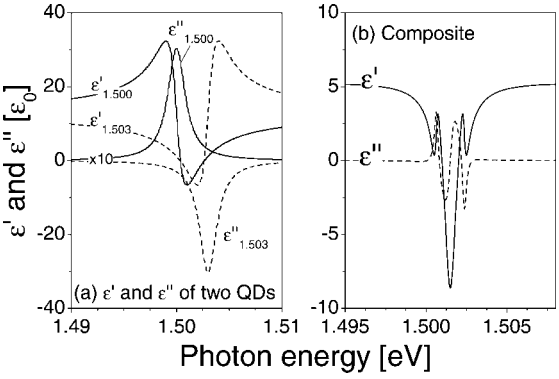

Figure 3. (a) One type of QDs at 1.50 eV and $|a^{(0)}|^2 = 1.0$ (solid black lines), the other type of QDs at 1.503 eV and $|a^{(0)}|^2 = 0.0$ (dashed lines). (b) The two types of QDs are immersed in $\varepsilon_i = 1.8$. The densities of two type of QDs are 7×10^{16} cm^{-3}.

2. Furthermore, we assume a QD radius of 10 nm. Consider first the distribution of one type of QDs (i.e., $x_1 \neq 0$ and $x_2 = 0$), the spectra of ε for two QD densities and two background dielectric constants are shown in Figure 2. Both ε' and ε'' increase with increasing QD density and decreasing ε_i. A low background dielectric constant is thus important in reducing the QD concentration.

For the combination of two types of QDs, one type is lossy, and the other has been pumped for optical gain, immersed in $\varepsilon_i = 1.8$, the optical spectrum is presented in Figure 3. Figure 3a depicts the spectra of ε'_{QD} and ε''_{QD} of the two QDs. Note that gain is at the higher energy, a condition we have found necessary in order to achieve $\varepsilon' < 0$ and $\varepsilon'' = 0$, which are given in Figure 3b. Note that by combining lossy QDs with optically pumped QDs, the macroscopic permittivity of our structure is approximately in the form of

$$\frac{i}{\omega - \omega_r - i\gamma} = \frac{-\gamma}{(\omega - \omega_r)^2 + \gamma^2} + i\frac{\omega - \omega_r}{(\omega - \omega_r)^2 + \gamma^2} \quad (11)$$

where ω_r denotes the macroscopic resonance frequency of the dimer system and γ is the damping rate. The loss is minimal when ω approaches ω_r. The expression follows exactly the Kramers–Kronig relations as required by the stability of the system. Such a spectrum is fundamentally different from

$$\frac{1}{\omega_r - \omega - i\gamma} = \frac{\omega_r - \omega}{(\omega_r - \omega)^2 + \gamma^2} + i\frac{\gamma}{(\omega_r - \omega)^2 + \gamma^2} \quad (12)$$

for which γ is to be reduced in the form of $\gamma = 0$, $\partial\gamma/\partial\omega = 0$, and $\partial^2\gamma/\partial\omega^2 > 0$ in order to become lossless, which results in a pole in the upper half-plane and hence violates causality.[28] $\varepsilon' = -8.5$ and $\varepsilon'' = 0$ is achieved in Figure 3b at $\hbar\omega_r = 1.5015$ eV with a total QD density of 1.4×10^{17} cm^{-3}, which is 58% in terms of volume fraction. The QD concentration can be reduced, see Figure 5c. Notice that eq 10 neglects dipole interactions among excitons in QDs. The dipole interactions have been included in the calculation of dispersion relation of periodic QD photonic crystal, which increase the reflection peak width.[29] For the distributed QDs, we make the following estimation: the electric field strength of a dipole is approximately $E_{QD} = P_{QD}/4\pi\varepsilon_i r^3$, where r is the distance measured from the center of the dipole. $P_{QD} = 4\pi R^3 \varepsilon_{QD} E_{in}/3$, where E_{in} is the transmitted electric field inside the QD from ε_i. $E_{in}/E_{ex} \approx 2\sqrt{\epsilon_i/\epsilon_{QD}}/(\sqrt{\epsilon_i/\epsilon_{QD}} + 1) = 0.39$ for $\varepsilon_i = 1.8$ and $\varepsilon_{QD} = 30$, where E_{ex} is the electric field of the incident radiation. However, the averaged value of E_{in}/E_{ex} inside the QD is only about 0.02, obtained numerically from standard FDTD calculation.[30] This gives $E_{QD}/E_{ex} = 0.4$ at the surface of the QD, which falls off as the cube of the distance r. We thus can conclude that the dipole interaction is not negligible. It however will not modify qualitatively the numerical results from the MG expression of eq 10.

In Figures 4 and 5, we show results for an ensemble of two types of QDs immersed in a medium as a function of the QD concentrations and the difference between the exciton energies of the two types of QDs. In Figure 4a, with $\delta = 1$ meV, even though the higher energy level is pumped, the MG expression produces gain at the lower frequency, while for larger gaps (such as $\delta = 4$ meV), the gain reverts back to the higher frequency. The basis for this is the weighting in the denominator of the MG expression, which also produces ripples in Figure 3.

To realize the structure described here, significant levels of amplification are required, which are around 1 dB/μm in the region of negative ε' in Figure 3 (which can be much smaller after reducing the amplitude of the negative ε'). Compared to ordinary semiconductor laser amplifiers of around 0.1 dB/μm, this implies that there should be no feedback mechanisms at the peak gain wavelength such that

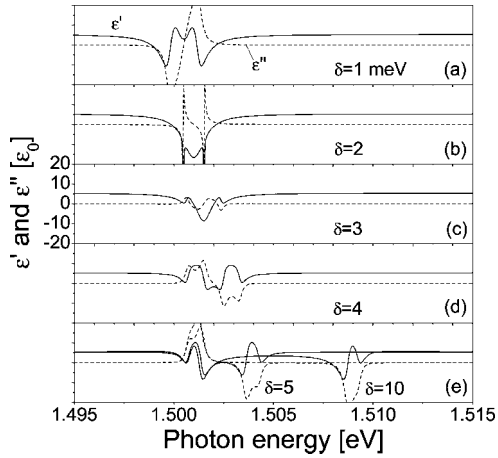

Figure 4. Effective dielectric constant for an ensemble of two types of QDs immersed in a medium of dielectric constant ε_i. δ is the difference between the exciton energies of the two types of QDs. The higher-energy QDs are pumped, $\varepsilon_i = 1.8$. The densities of two types of QDs are 7×10^{16} cm^{-3}.

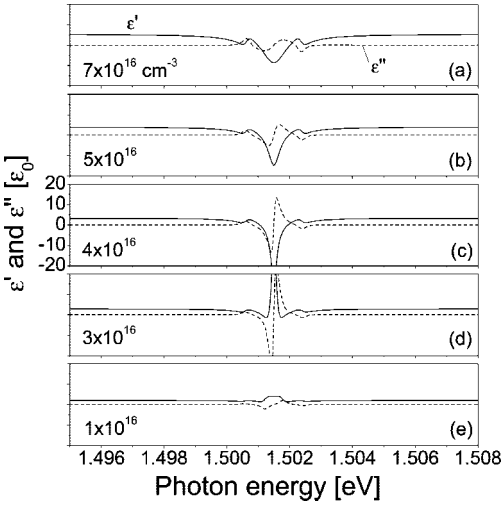

Figure 5. Same as Figure 4. $\delta = 3$ meV, higher-energy QDs are pumped, and $\varepsilon_i = 1.8$.

lasing occurs. For waveguide amplifiers, this implies rather short waveguides of several tens of μm. Because of the small $\hbar\omega$ assumed here, saturation for such short waveguides will not be an issue. It is outside the scope of this paper to discuss more practical details of the various structures made possible by the arrangements described here. Furthermore, to maintain N QDs in excited states, the minimum optical power is $P = N\hbar\omega/2\tau$,[31] where τ is associated with QD's spontaneous emission decay. For colloidal II−VI QDs of ref 20, the emission decay time was 3.6 ns, with a standard deviation of 6.19% measured by using a Sarastro 2000 confocal microscope.[21] The required optical power thus becomes 1.5×10^6 W/cm^3 when the QD density is $N = 5 \times 10^{16}$ cm^{-3}, $\hbar\omega = 1.5$ eV. For a film of thickness of 10 μm and a cross-section of 1 mm^2, this corresponds to an average optical power of 15 W, which is readily achievable using even continuous-wave laser systems. As another example, in a waveguide with a cross section of 10 μm^2 and 100 μm in length, the absorbed power needs to be only 1.5 mW. Various efficiencies must be applied that will make the pumping task more difficult, but it would appear not beyond reach. We further have noticed the fast experimental development of optical gain in colloidal QDs, e.g., colloidal CdSe/ZnS core−shell QD microcavities (colloidal QDs in PMMA polymer matrix)[32] and single-exciton optical gain in CdS/ZnSe QDs.[33]

Our study suggests that achieving a lossless isotropic material at a single frequency is possible. It further indicates that fabrication using colloidal II−VI QDs that are mixed in a polymer background and properly optical pumped should be a tractable path.

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