Polarization splitting of the gain band in quantum wire and quantum dot arrays

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We theoretically predict and experimentally confirm that arrays of resonantly amplifying quantum wires and quantum dots (QD's) exhibit splitting of the gain band into separate bands for *E*- and *H*-polarized field. This effect originates from the diffraction at single anisotropically shaped QD's and the collective effect of the anisotropy of the QD array both described by an effective medium approach. Experimental results for multiple sheets of $Zn_xCd_{1-x}Se$ QD's in a ZnSe matrix confirm the theoretical predictions and signal the discovery of a new class of ''active'' composites. [S0163-1829(99)01120-0]

Transparent and dissipative heterogeneous media with electrically small inclusions (of dimension much smaller than the wavelength) of one material in another material, conventionally referred to as composite materials, have found numerous applications in solid-state physics, device physics, and optics. Artificial dielectrics and metalo-dielectrics,[1,2] chiral composites,[2] nonlinear composite films,[3] and nanotube films[4] are well-known examples of such structures. Composites exhibit, in general, mechanical, electronic as well as optical properties that are not inherent to each individual component. Electromagnetic properties of composites are usually modeled in the framework of the effective-medium approach,[5] which implies that the electromagnetic field averages over material inhomogeneities. Thus, a homogeneous medium with effective constitutive parameters instead a composite is being considered. The effective parameters are expressed in terms of the generic and the geometrical parameters of the inclusions and the host medium.

Recent development in semiconductor epitaxial growth techniques[6] has made possible fabrication of dense arrays of low-dimensional nanosized structures such as quantum wires and quantum dots (QD's). Because of the exceptionally strong modification of the electronic spectrum and optical properties due to the two- or three-dimensional confinement of the charge carriers, respectively, these structures are presently in the center of numerous fundamental research activities. Ultrahigh material gain, drastically reduced threshold current density, improved temperature stability of the threshold current, and improved dynamic characteristics were recently demonstrated for QD-based lasers.[7]

The active medium of such a laser consists of a three-dimensional ensemble of resonantly amplifying nanostructures forming under certain growth conditions a quasiregular lattice in the growth plane, which can be periodically stacked perpendicularly to this plane.[6,8] The characteristic geometrical size of a QD is much smaller than the emission wavelength in the host material. Thus, a system of QD's can be considered as a composite. The idea of the present letter is to extend the effective-medium approach to arrays of resonantly amplifying semiconductor QD's as *active composites*.

Previous theoretical treatments of QD arrays were based on simple summing over the microscopic gain $\tilde{g}(\omega)=-(\omega/c)\mathrm{Im}\{\sqrt{\varepsilon(\omega)}\}$ of individual QD's. Here, $\varepsilon(\omega)=\varepsilon'(\omega)+i\varepsilon''(\omega)$ is the complex permittivity of the QD, an $\exp(-i\omega t)$ time dependence of the fields is implicit and $\varepsilon''(\omega)<0$ for active media. The imaginary part of the resonant dielectric response of an individual QD is found on the basis of a charge-carrier level-structure being calculated accounting for the size quantization in two or three dimensions[9,10] and realistic geometry, strain distribution, and material properties of the QD's.[11] As a result, in this conventional approach the macroscopic gain g_{con} of an ensemble of QD's is introduced as[9,10]

$$g_{con}(\omega)=f\tilde{g}(\omega), \tag{1}$$

where $\tilde{g}(\omega)=-\omega\varepsilon''(\omega)/2c\sqrt{\varepsilon_h}$, f is the volume fraction of QD's and ε_h is the permittivity of the host medium. This

simple relation appears as a result of averaging of the local electromagnetic field over material inhomogeneities under the assumption that the field inside and outside the QD's is the same. It presents a first approximation of the QD array gain spectrum. Owing to the ultrahigh gain of QD's, the imaginary part of the permittivity of a QD throughout the gain band can be as large as the real part. It results in a strong frequency dependence of $\varepsilon'(\omega)$. Thus, inside the gain band the difference between dielectric constants of the QD's and the host becomes significant, resulting in a strong depolarizing field.[12] This field is neglected deriving Eq. (1). Moreover, the interaction between QD's contributes to the depolarization field.

For modeling the electromagnetic response of a QD ensemble, we make use of the Maxwell-Garnett model with the Clausius-Mossotti correction as it is based on a rigorous solution of Maxwell equations under the assumption of a small inclusion density.[2] The effective permittivity tensor $\hat{\varepsilon}_{eff}$ of a composite can be expressed in terms of a Cartesian basis diadics by $\hat{\varepsilon}_{eff}(\omega)=\varepsilon_H(\omega)(\mathbf{xx}+\mathbf{yy})+\varepsilon_E(\omega)\mathbf{zz}$ (see, for example, Ref. 4), where

$$\varepsilon_\sigma(\omega)=\varepsilon_h\left[1+\frac{f\alpha_\sigma(\omega)}{1+fL_\sigma\alpha_\sigma(\omega)}\right], \tag{2}$$

$\sigma=(E,H)$ refers to light polarized along (E polarization) or normal (H polarization) to the z axis, $\alpha_\sigma(\omega)$ is the polarizability of a single QD in σ-polarized field, and L_σ is a frequency-independent coefficient being defined by the geometry of the array. On the basis of the considerations given in Ref. 12 the polarizability of a QD can be written as

$$\alpha_\sigma(\omega)=\left[\frac{\varepsilon_h}{\varepsilon(\omega)-\varepsilon_h}+N_\sigma\right]^{-1}, \tag{3}$$

where N_σ is the depolarization coefficient of a single QD. Note that the denominator in the second term of Eq. (2) is the Clausius-Mossotti correction responsible for the electromagnetic interaction of QD's. Using Eqs. (2) and (3) allows us to define the anisotropic macroscopic gain

$$g_\sigma(\omega)=-\frac{\omega\varepsilon''_\sigma(\omega)}{c\sqrt{2[\varepsilon'_\sigma(\omega)+|\varepsilon_\sigma(\omega)|]}}, \tag{4}$$

which is obviously different from that given by Eq. (1). Here, $\varepsilon_\sigma=\varepsilon'_\sigma+i\varepsilon''_\sigma$.

Now, let us estimate how the difference between Eqs. (1) and (4) manifests itself physically. For that aim, as a first step, we choose a single-resonance Lorentz model for the microscopic permittivity of one QD

$$\varepsilon(\omega)=\varepsilon_h+\frac{g_0}{\omega-\omega_0+i/\tau}, \tag{5}$$

where $g_0>0$ is the parameter defined the maximum gain of the QD, τ is the QD exciton dephasing time, and ω_0 is the resonance frequency. The gain of an individual QD can be isotropic or anisotropic depending on the wave functions of the electronic states involved and the particular QD geometry. We assume first the gain of QD to be isotropic. The dielectric constant ε_h of the host medium is assumed to be a real, isotropic and frequency-independent quantity.

Further, we restrict the analysis to two different geometrical models of arrays: (i) a quadratic lattice composed of infinitely long parallel cylinders [Fig. 1(a)]; (ii) a tetragonal lattice composed of ellipsoids [Fig. 1(b)]. The first geometry represents an ensemble of quantum wires or closely stacked coupled QD's while the second one can serve as a model for an ensemble of separated QD's vertically stacked with finite period.[6] In the framework of the Maxwell-Garnett approach, the depolarization factors N_σ and lattice geometrical coefficients L_σ for model (i) are as follows:

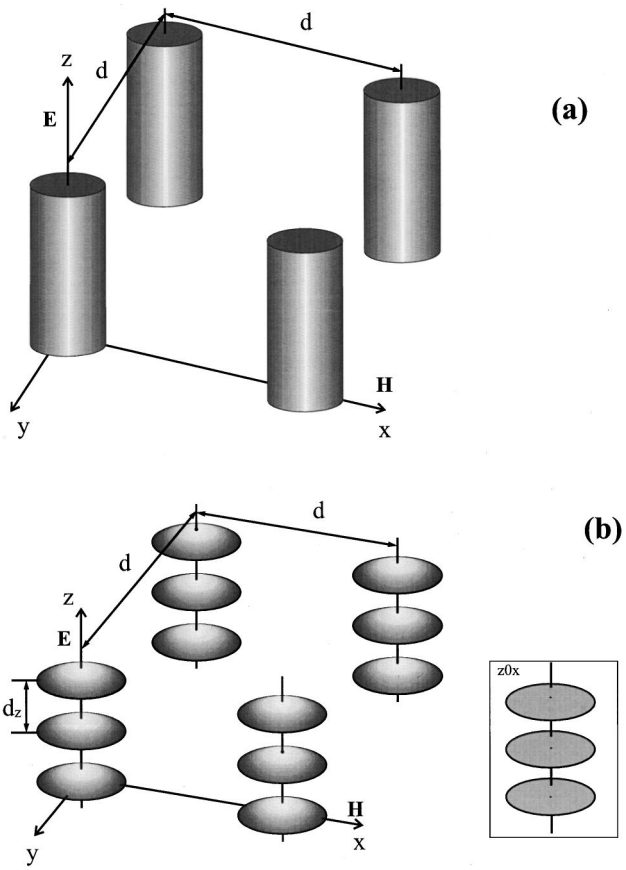

FIG. 1. (a) Array of infinitely long cylindrical QD's arranged on a quadratic lattice and (b) array of vertically stacked disc QD's arranged on a tetragonal lattice.

$$N_E=L_E=0, \quad N_H=L_H=\tfrac{1}{2}. \tag{6}$$

For model (ii) in accordance with Ref. 12

$$N_E=\frac{e^2+1}{e^3}(e-\arctan e), \quad N_H=\tfrac{1}{2}(1-N_E), \tag{7}$$

where $e=\sqrt{a^2/b^2-1}$ is the ellipsoid eccentricity, a and b are the ellipsoid semiaxes in the $x0y$ plane and the z direction, respectively. These formulas hold true for both disclike ($a>b$) and cigarlike ellipsoids ($a<b$). Infinite stretching of the ellipsoids ($a/b\to 0$) results in $N_E\to 0$, $N_H\to 1/2$ and Eq. (7) reproduce the polarizabilities of the cylinders given by Eq. (6). The geometrical coefficients L_σ for a tetragonal lattice have been derived in Ref. 13 and have the following form:

$$L_E=\frac{1}{4\pi}\left[\left(18-\frac{10}{\beta^2}\right)\arctan\frac{Q}{\beta^2}+30\frac{1-\beta^2}{\beta^4}\times\left(2\arctan Q-\ln\frac{1+Q}{1-Q}\right)\right], \tag{8}$$

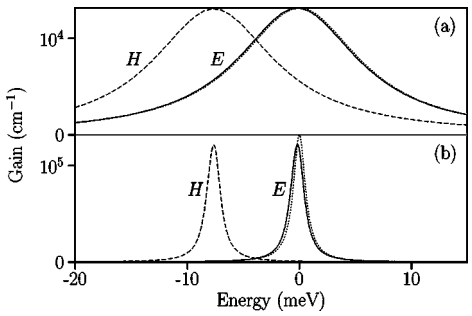

FIG. 2. Gain bands of a cylindric QD array for *E*- and *H*-polarized fields, dephasing time $\tau=0.1$ ps (a); 1 ps (b), volume fraction $f=0.05$, host dielectric constant $\varepsilon_h=12.25$, wavelength $\lambda=1\mu$m. Also shown by dotted curves are gain bands of a QD ensemble defined by Eq. (1).

$$L_H=\frac{1}{4\pi}\left[\left(18+\frac{20}{\beta^2}-\frac{30}{\beta^4}\right)\arctan Q+15\frac{1-\beta^2}{\beta^4}\ln\frac{1+Q}{1-Q}\right], \tag{9}$$

where $\beta=d_z/d$ is the ratio of lattice periods in z direction and in x or y direction, $Q=\beta/\sqrt{2+\beta^2}$. The above expressions are not valid for $\beta\ll 1$ or $\beta\gg 1$. Therefore, close stacking of QD's is considered here in the framework of model (i). For a cubic lattice $\beta=1$ and the depolarizing field induced by QD interaction becomes isotropic: $L_E=L_H=\frac{1}{3}$.

Substitution of Eqs. (5)–(9) into Eq. (2) allows us to evaluate the macroscopic gain (4) for QD arrays for both polarizations. The maximum gain of an individual QD is found using the data in Refs. 9,10, and 14. For example, it accounts to 10^6 cm^{-1} for a spherical GaAs QD of diameter 5 nm and homogeneous broadening of about 7 meV ($\tau=0.1$ ps). Figure 2 demonstrates the results for model (i) at different dephasing times while Fig. 3 corresponds to model (ii) for cubic and tetragonal lattice.

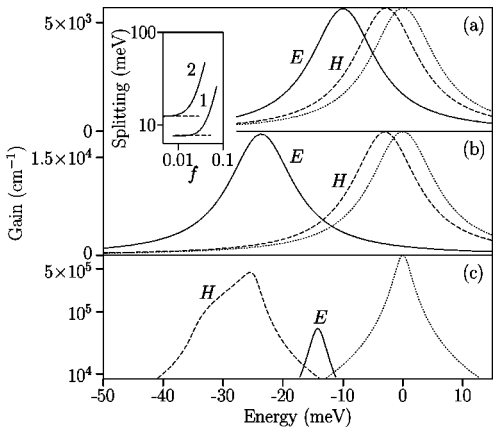

FIG. 3. Macroscopic gain bands for *E*- and *H*-polarized fields for an array of disc QD's with $b/a=0.33$, $a/d=0.25$ arranged on a cubic lattice with $\beta=1$ (a) and a tetragonal lattice with vertical stacking period $\beta=0.3$ (b), $f=0.02$ (a), 0.07 (b), $\tau=0.1$ ps, $\varepsilon_h=12.25$, $\lambda=1\mu$m. (c) Gain bands as in (b) but for QD peak gain two times smaller for *E* polarization, 10 times larger for *H* polarization and $\tau=0.5$ ps. Also shown by dotted curves are the gain bands of QD ensembles defined by Eq. (1). Inset, polarization splitting of an array of disc QD's versus fraction f for decreasing vertical stacking period of the tetragonal lattice, $a/d=0.25$, $b/a=0.3$ (curve 1), 0.1 (2). The dashed lines are given by Eq. (10).

As one can see, the resonance energy of the macroscopic gain is shifted with respect to the microscopic resonance energy $\hbar\omega_0$ and the shift is different for *E* and *H* polarizations. Equation (4) incorporates two different mechanisms responsible for this shift and splitting. The first mechanism is related to diffraction at individual QD's of anisotropic shape. Although QD's are assumed to be electrically small and, by this reason, the diffraction can be described within the dipole approximation, its role turns out to be essential owing to the resonant nature of the gain. The diffraction induces a depolarizing field that shifts the resonance of α_σ with respect to the resonance of the dielectric function $\varepsilon(\omega)$ of an individual QD. It is obvious that the diffraction induced shift does not depend on the dephasing time τ and the volume fraction f of the QD's. In the low-concentration limit this shift can be found from Eqs. (2) and (3) for a Lorenzian gain band

$$\Delta\omega_\sigma=-N_\sigma\frac{g_0}{\varepsilon_h}. \tag{10}$$

For the cylindrical structures the shift of the resonance is absent for an *E*-polarized field and equal to $\Delta\omega_H=-g_0/2\varepsilon_h$ for an *H*-polarized field. For *E* polarization, the ensemble of infinitely long cylinders in z direction produces the same average field as the incident field with no depolarizing field and no shift of the gain band. Similarly, in the case of the disclike QD's the gain band for an *H*-polarized field directed towards the long axis of the disc possesses the smaller shift if the gain of the single QD is not polarization dependent. Note that this diffraction induced shift is analogous to that which occurs in ferromagnetic resonance where the resonance frequencies of small-sized grains and infinite continuous media are different because of the effect of demagnetization.[12,15]

The second mechanism inducing a frequency shift is a collective effect and is defined by interaction between QD's in the ensemble. It results, as was stated above, from the Clausius-Mossotti contribution to the permittivity tensor components defined by Eq. (2). This contribution shifts the resonance frequency for both field polarizations. The shift, however, depends on lattice type as one can see in Figs. 3(a) and 3(b) and becomes significant at large concentrations of QD's in the host medium (Fig. 3, inset).

The polarization dependence of the shift results in an appreciable polarization splitting of the gain bands for *E* and *H* polarizations, which become completely resolved for sufficiently large dephasing times (Figs. 2 and 3). Due to fluctuations of the QD sizes, shapes, and lattice period, typically an inhomogeneous broadening of the gain bandwidth occurs. However, this effect becomes significant only if the inhomogeneous broadening approaches values comparable to the value of the polarization splitting. Thus, radiation from an active composite must be linearly polarized if spontaneous emission does not play a significant role.

In general, also the anysotropic shape of QD causes different oscillator strengths for transitions involving *E*- and *H*-polarized field.[9] To estimate the influence of the different gain for the different polarizations we considered the case when the gain for *E* polarization is lower and for *H* polarization is higher [Fig. 3(c)] as compared to the case of equal gain in Figs. 3(a) and 3(b) [parameter g_0 in Eq. (5) is the diagonal tensor]. To make the effect more clear we select

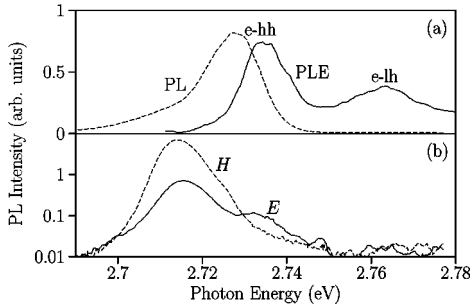

FIG. 4. Photoluminescence (PL) and photoluminescence excitation (PLE) spectra (a) and *E*- and *H*-polarized spectra of stimulated emission in edge geometry (b) for the observation temperature 7 K, excitation density 1 MW/cm^2, energy of exciting photon $E_{ex} = 2.88$ eV.

also a longer exciton dephasing time. The remarkable result is that the gain peak of *E* polarization locates closer to the QD resonance energy, since in accordance with Eq. (10) the decrease in the gain peak dominates over the corresponding depolarization coefficient and results in smaller shift.

To verify the predicted effects experimentally we fabricated a structure with twelve stacks of dense arrays of $Zn_xCd_{1-x}Se$ disclike QD's with $\sim 10^{12}$ cm^{-2} surface density, ~ 40 Å lateral size, and ~ 12 Å height in a ZnSe matrix separated with 50 Å ZnSe spaces.[16] At low-excitation density in edge geometry the QD photoluminescence (PL) is linearly polarized throughout the PL band with a degree of polarization of 50% of the *H* component (alignment as in Fig. 1). This effect is related to the anisotropy in optical transition matrix elements for *H* and *E* polarizations originated from the disclike shape of the QD's and the heavy-holelike nature of the QD exciton. The light-hole-like exciton QD state is also seen in the PL excitation spectrum and is shifted significantly away from the spectral range of interest. With increase in excitation density gain develops and causes a strong superlinear growth of the PL intensity. Spectra of the *H* and *E* components of stimulated emission are shown in Fig. 4(b). The predominantly *H*-polarized component is shifted from the QD resonance revealed in the PL excitation and optical reflectance spectra [Fig. 4(a)]. The degree of polarization of this component strongly increases with excitation density. At the same time, we distinctly observed an appearance of a *second* separate peak in the *E* component at energies closer to the QD resonance. The intensity of the *E*-polarized peak is much smaller pointing to a smaller gain coefficient as compared to that for the *H*-polarized peak at lower photon energy. Therefore, the peak location of the *E* component corresponds to the predictions of the calculations for the case of different gain coefficients for *E* and *H* polarizations [Fig. 3(c)]. As the photoluminescence excitation (PLE) spectrum was recorded in the direction perpendicular to the surface, and there exists no strong anysotropy for the in-plane arrangement of the islands, also no pronounced polarization of the signal or polarization splitting effect has been observed in this case.

To conclude, in the present letter we have theoretically predicted an effect of a polarization-dependent frequency shift of the macroscopic gain band of QD-based active composites with respect to the microscopic gain. We have shown that this effect results from the diffraction of the electromagnetic wave on an array of anisotropically shaped QD's and the interaction between QD's arranged on an anisotropic lattice. The theoretical predictions are confirmed by the experimentally observed polarization splitting of the gain band of a multiple sheet of $Zn_xCd_{1-x}Se$ QD's.

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