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OPTICS

Resonance Reflection of Light by a Thin Layer of a Dense Nonlinear Medium

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Abstract—The possibility of hysteretic behavior of resonant reflection spectral curves is determined analytically for the conditions of the dipole–dipole interaction and spectral shift of the field. The phase shift of the acting field is associated with the variation of the energy state of a dense resonant medium forming a thin boundary layer. The problem is considered for the parameters of quantum-size structures based on semiconductors used in optics.

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Analysis of optical properties of size-limited planar structures (in particular, thin semiconducting films) makes it possible to search for perspective elements for effective control over laser radiation. Nonlinear coupling between the field of an electromagnetic wave passing through a thin film of the material formed by resonant atoms and the polarizability of the medium may lead to a number of interesting physical effects [1, 2]. Dense resonant media, viz., materials with a high concentration of active centers (atoms, molecules, ions, excitons, etc.) have been actively studied recently. These media clearly exhibit a number of nonlinear phase effects associated with short-range dipole–dipole interactions, which can be manifested, for example, in intrinsic optical bistability or instability leading to the development of self-sustained oscillations in transmitted radiation. The objects in which such phenomena can be observed include, in particular, thin films of dense materials based on semiconductor nanostructures formed by quantum dots [3]. In this case, it is also important that the elements of nanostructures possess high dipole moments associated with exciton transitions (the values are at least 1×10^{-28} C m) [4]. As a result of grouping of ten or more strata, a submicrometer planar film with a clearly manifested nonlinear response to the resonant radiation field can be formed from nanostructural elements [5]. A characteristic feature of the response of such objects is that an additional component relative to the Fresnel reflection (refraction), which is associated with resonant surface polarizability, must be present in reflected (transmitted) beams in the case of a resonant action of light [6]. For relatively large dipole moments, the polarizabilities of active particles in the ground and excited states differ significantly (the quasi-resonance component of the polarizability turns out to be significant). In accordance with the variation of the concentration of active particles, the polarizability of the medium must change. Then, nonlinear refraction is observed for frequencies of the acting field, which correspond to the optical resonance region; a noticeable manifestation of this polarizability in semiconductors is a spectral shift (broadening) of the acting field [7]. Analysis of the features of resonant reflection under the conditions of phase effects caused by the dipole–dipole interaction and quasi-resonant polarizability is undoubtedly of interest. In the work forming the basis of this communication, we tried to demonstrate the effectiveness of both factors of the phase shift in determining the peculiarities of the dispersion dependence of nonlinear reflection of light by a thin layer of a dense resonant medium.

In the formulation of the model, we assumed that optical media with linear permittivities ε_1 and ε_2 are separated by such a layer. We assumed that the spacing between resonant particles in the thin layer is large enough to avoid overlapping of their electron orbitals; in this case, a traditional description of the interaction of atomic dipoles in a quantum-size structure can be preserved [3]. The field with amplitude E_i incident along the normal to the boundary layer is assumed to be quasi-stationary and generated by a plane wave. The interaction of the substance of the boundary layer with the laser radiation field (with carrier frequency $\omega = 2\pi c/\lambda$) is described analogously (see, for example, [8]) by a modified system of the Maxwell–Bloch equations for quasi-stationary complex amplitudes of the transmitted (E) and reflected (E_r) waves and probabilistic variables of the resonant response of the medium (polarizability ρ and difference in populations n of the exciton transition levels per atom):

$$E = \frac{2\sqrt{\varepsilon_1}}{\sqrt{\varepsilon_1}+\sqrt{\varepsilon_2}}E_i(t)$$
$$-\frac{\omega Nl}{(\sqrt{\varepsilon_1}+\sqrt{\varepsilon_2})c}\left[\frac{\mu}{\varepsilon_0}\rho + i2\pi\Delta\alpha(n_0-n)E\right],$$
$$\dot{\rho} + \frac{1}{T_2}(1+i\Delta)\rho = \frac{\mu}{\hbar}nE,$$
$$\dot{n} + \frac{1}{T_1}(n+n_0) = -\frac{\mu}{2\hbar}(\rho^* E + \rho E), \quad (1)$$
$$E = \frac{1}{1-2\pi\Delta\alpha N(n_0-\Delta n)/3}\left(E + i\frac{\mu N}{3\varepsilon_0}\rho\right),$$
$$E_r = E - E_i, \quad \Delta = (\omega-\omega_0)T_2.$$

Here, μ is the average dipole moment of active particles, N is their volume density, n_0 is the initial value of the population difference, Δ is the normalized detuning of the probe field frequency from center ω_0 of the resonance spectral absorption line, T_1 and T_2 are the longitudinal and transverse relaxation times (corresponding to the interband relaxation time and homogeneous line width reciprocal to T_2), and l is the layer thickness (in the computational algorithm used here, thickness l is naturally smaller than wavelength λ). The field coupling equations in system (1) are written analogously to [6] on the basis of the conditions imposed on fields E, E_r, and E_i at the interface between the media, which are used for solving the Maxwell equations. The inclusion of the polarizability variable into the coupling equations indicates the allowance for the component of the nonlinear response of the boundary layer substance, which is associated with superradiance of the ensemble of active particles forming the medium. The expression for the polarizability contains the component that usually takes into account the quasi-resonant polarizability component, which is significant in the presence of transitions excited by an external field of frequency ω and close to the resonance transition. It is found that parameter $\Delta\alpha$ (the difference between the polarizabilities in the ground and excited states of the atom) is significant. Its value determines the parameter of refractive nonlinearity with manifestations observed and measured in the structures made of superconductors that are used in optics [5], which are estimated as "giant." The variation of refraction is proportional to the resonant variation $n_0 - n(t)$ of the populations, which experiences saturation under the action of laser fields. It was emphasized above that the effect associated with the short-range dipole–dipole interactions is significant in dense resonant media. For this reason, system of equations (1) is formulated with allowance for local fields produced by dipole atoms: the optical field $E'(t)$ acting on the atoms of the layer contains the Lorentz correction calculated in the mean field approximation, which includes resonant components.

The distinguishing regularity of the response of an especially thin layer to resonant radiation, which is usually considered in such models, is optical bistability. It is analyzed for the time-independent problem when the intensity of radiation probing the boundary layer varies extremely slowly as compared to the relaxation times of the two-level system and can therefore be treated as continuous in time. This means that for the given value of the applied field amplitude $E_i(t) = E_0$, equilibrium characterized by a certain value of the material variables is established in the system. It is convenient to normalize the intensities of the applied (Y) and transmitted (X) fields to the power of the field saturating the absorption: $Y = \mu^2 T_1 T_2 E_0^2/\hbar^2$ and $X = \mu^2 T_1 T_2 |E_S|^2/\hbar^2$; here, E_S is the equilibrium amplitude of the field transmitted into the layer. Using these concepts (and considering the time-independent approximation of system (1)), we can express the relations connecting the dimensionless intensities of transmitted field (X) and the field acting in the medium (X') with intensity Y of the applied field as follows:

$$\frac{4\eta X}{(1+\eta)^2} = X'\left[1-\gamma G(\Delta-\beta X') + \frac{G}{1+\eta}\right]^2$$
$$+ G^2 X'\left(\frac{\Delta-\beta X'}{1+\eta}+\gamma\right)^2, \quad (2)$$
$$Y = X'[1-\gamma G(\Delta-\beta X')]^2 + (G\gamma)^2 X',$$
$$G = \frac{\kappa}{1+\Delta^2+X'}, \quad \kappa = \frac{\mu^2\omega_0 Nl}{\varepsilon_0 c\hbar}T_2.$$

Here, $\beta = 2\pi\Delta\alpha\varepsilon_0\hbar/\mu^2 T_2$ is the resonant nonlinear refraction parameter, $\gamma = c/3\omega(1+\eta)l$ is the normalized coefficient in the local Lorentz correction, κ is the unsaturated absorption coefficient, G is the saturated absorption taking into account the form factor of the line, and $\eta = \sqrt{\varepsilon_2/\varepsilon_1}$ is the relative refractive index.

Physically, relations (2) can be treated as the system of equations describing nonlinear and dispersion properties of a layer of an optical medium in the mean field approximation for the conditions under which the absorption and the position of its resonance on the frequency scale exhibit a peculiar dependence on the intensity of the transmitted field. In the definition of resonant absorption, a special role is played by the dispersion component describing phase modulation of radiation, which indicates "pulling" of its carrier frequency to the shifted center of the line. Using relations (2), we can directly express the dependence of transmittance $X(Y)$ of the thin active layer as well as its effective reflectance $R(Y)$ on the intensity using the following expression (where $r = (\eta-1)/(\eta+1)$ is the Fresnel reflectance):

$$R = \frac{\{r + G[1/(\eta + 1) - r\gamma(\Delta - \beta X)]\}^2 + G^2[(\Delta - \beta X)/(\eta + 1) + r\gamma]^2}{\{1 + G[1/(\eta + 1) - \gamma(\Delta - \beta X)]\}^2 + G^2[(\Delta - \beta X)/(\eta + 1) + \gamma]^2}. \quad (3)$$

It is convenient to estimate the behavior of these dependences using parametric calculations (i.e., assuming that one of the variables (X) is a linear, nonnegative, and continuously increasing parameter). The results of calculations shown in Figs. 1 and 2 were obtained using the values of model coefficients, which mainly corresponded to the range of parameters of quantum-size structures in the InAs/(Al)GaAs systems. Judging from the curves in Fig. 1, we can conclude that the values of the reflectance, which are also determined by the resonant polarizability in the surface layer, can be much higher than the Fresnel value of r^2; however, these values decrease upon saturation of absorption in the layer and approach exactly the Fresnel value. In a certain range of Y values in which the phase contributions from the short-range dipole–dipole interaction and the factor of spectral line broadening are comparable, the nonlinear characteristics of reflection for the absorption coefficient exceeding the threshold value (κ] ~ 1.5) exhibits bistability. In this case, for a cyclic variation of the external signal intensity, the steady-state response of the struc-

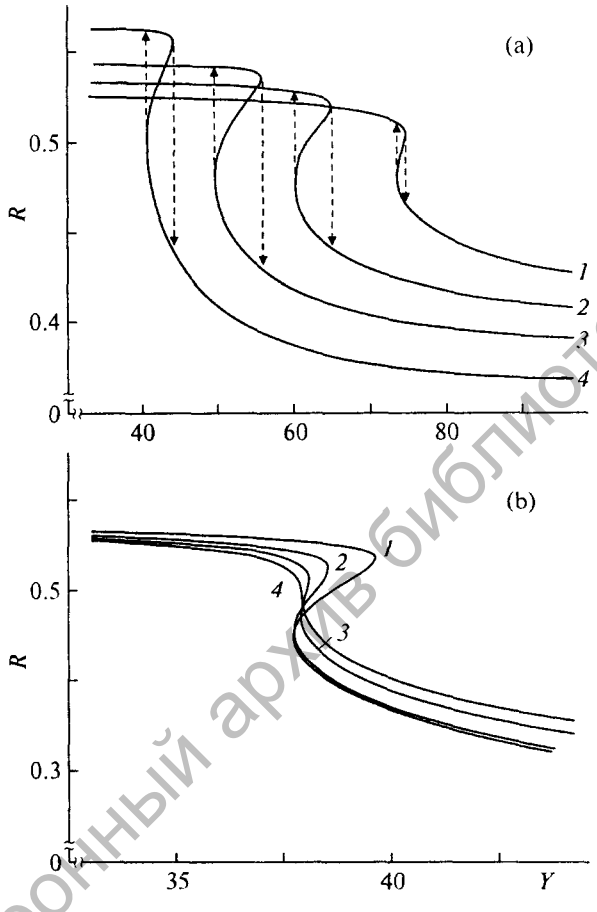

Fig. 1. Dependence of resonant reflectance on the level of normalized intensity of the applied field (dashed arrows in (a) indicate the directions of hysteretic jumps); (a) $\kappa = 2.0$, $\beta = 0.1$; $\Delta = 0.1$ (*1*), 0.5 (*2*), 1.0 (*3*), and 2.0 (*4*); (b) $\kappa = 1.7$, $\beta = 0$ (*1*), 0.05 (*2*), 0.08 (*3*), and 0.10 (*4*); $\Delta = 0.5$; $\gamma = 0.15$, $\lambda = 1.25 \times 10^{-6}$ m; $\eta = 3.6$, $T_1 = 1 \times 10^{-9}$ s, and $T_2 = 1 \times 10^{-12}$ s.

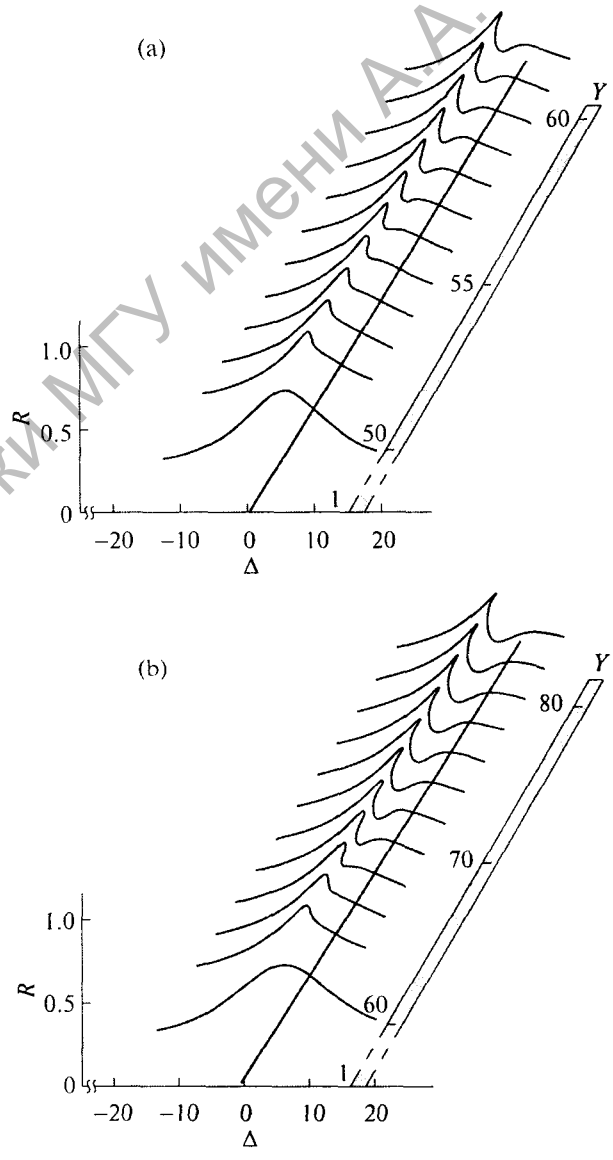

Fig. 2. The shape of spectral resonant reflection curves as functions of the normalized intensity of the applied field: (a) $\kappa = 2.0$, (b) 2.4; $\beta = 0.5$, $\gamma = 0.14$, $\lambda = 1.25 \times 10^{-6}$ m; $\eta = 3.6$, $T_1 = 1 \times 10^{-9}$ s, and $T_2 = 1 \times 10^{-12}$ s.

ture must exhibit a hysteresis. The distance between the turning points on the characteristic curves, at which hysteretic variations of reflectance are possible, depends on the frequency detuning (Fig. 1a) and on the nonlinear refraction parameter (Fig. 1b).

Using relations (2) and (3), we can determine the shape of the resonance curves describing the dependence of reflectance on detuning, $R(\Delta)$, for a fixed excitation level Y on the entire frequency scale. The first expression in (2) should be solved in this case as a nonlinear algebraic equation in X'. Determining X' from its solution and calculating R by formula (3) for increasing values of detuning Δ, we can construct the dispersion curve $R(\Delta, Y)$. Thus, we can determine the behavior of the reflectance of the layer as a function of frequency provided that the spectral width of the signal with uniform intensity Y is much larger than the spectral width of the resonant absorption line. Figure 2 illustrates the typical results of calculation of normalized spectral curves $R(\Delta, y)$ for various saturated absorption levels. For a low saturation, the spectral curves are the resonance lines typical of uniform broadening, which are almost symmetric about the central frequency that is displaced, however, due to the short-range dipole–dipole interaction. In both fragments in Fig. 2, such a behavior of the dependence is represented by front curves (for $Y = 1$). The frequency shift due to the dipole–dipole interaction is compensated by the nonlinear displacement, and the resultant shift of the central frequency decreases with increasing saturation of absorption. This can be seen from the shape of the curves corresponding to the values of incident field intensity close to the range in which the bistability of nonlinear characteristics of reflection is manifested (this range of intensities will be referred to as critical). Here, the increase in intensity leads primarily to the development of asymmetry of the curves due to the opposite action of frequency pulling to the line center and nonlinear spectral line broadening. With increasing excitation level, the deformation of the curves associated with the contribution of phase effects is manifested in the formation of a dip (local minimum) at the line center which corresponds to a decrease in the reflectance in this spectral range. On the segment adjoining the region of this minimum, the resonance curves for the critical values of intensity can acquire an inflection typical of bistability (which subsequently disappears upon an increase in the saturation level). Thus, bistable dependence $R(\Delta, Y)$ can be observed on the frequency scale also. In the bistability frequency band, during the intense signal frequency tuning, sharper hysteretic jumps of the effective reflection of the optical field by the nonlinear layer must take place. Comparison of the behaviors of the curves on fragments (a) and (b) in Fig. 2 shows that the jump is larger for films with a higher unsaturated absorption index κ. The values of critical intensity in this case are higher; however, its range is found to be wider, and hence it is easier to detect the hysteretic properties in the reflectance of resonant films with a relatively higher absorbance.

The interrelation between finer nonlinearity effects accompanying resonant absorption in thin layers based on media with a high density of active particles noticeably changes the conditions in which bistability and optical hysteresis must be manifested. The estimates of the possibility of the hysteretic behavior of the nonlinear and dispersion dependences of resonant reflection of light considered here can be useful for the development of nonlinear reflectors, active coatings, and inertialess frequency filters in passive devices controlling coherent radiation flows.

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