NONLINEAR OPTICAL PHENOMENA

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Coherent propagation of an optical pulse in a Bragg plasmon grating

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Abstract. The propagation of ultrashort pulses in a dielectric medium with periodically arranged metal nanoparticles is considered theoretically. Plasmon oscillations in these particles are described by the model of an anharmonic oscillator with the driving force proportional to the electric-field strength of an electromagnetic pulse. A system of equations determining the behaviour of electromagnetic waves is obtained in the approximation of slowly varying envelopes of ultrashort pulses and medium polarisation. Under the assumption that the frequencies of the carrier wave and oscillators coincide and the Bragg resonance condition is fulfilled, the solution of the obtained equations is found, which corresponds to the solitary wave of the ultrashort-pulse field and the medium polarisation (Bragg soliton). The numerical simulation shows the formation of a Bragg soliton (from the initial Gaussian pulse of the sufficient energy) and a nonstationary solitary wave with the vanishing group velocity.

Keywords: Bragg solitons, resonance Bragg grating, coherent propagation of ultrashort pulses, solitary wave.

1. Introduction

Optical phenomena in the media with the linear refractive index, which periodically changes along the propagation direction of an electromagnetic wave, have attracted attention of researchers for the last fifty years. One-dimensional periodic structures, in which the wave vectors of counterpropagating waves are related by the Bragg condition, are called Bragg gratings [1–3]. In nonlinear media with a periodically changing linear part of the refractive index, localised waves can be formed, which are called Bragg solitons. These phenomena are considered in detail in [3].

Not only the linear refractive index can change periodically. Thus, a homogeneous linear dielectric medium was

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Received 23 October 2006; revision received 29 January 2007 Kvantovaya Elektronika 37 (6) 549-553 (2007) Translated by I.A. Ulitkin considered in papers [4-7], in which thin films containing resonance impurities were arranged parallel to each other with a step a. This medium was called the resonance Bragg grating (RBG). The results of numerous studies of optical phenomena in RBGs are presented in review [8]. It has been shown recently with the help of numerical simulation [9] that a nonstationary pulse exists in RBGs, which looks like a soliton with a periodically changing propagation velocity—the so-called optical zumeron for which an approximate analytic expression has been also obtained.

An important achievement in modern optics is the fabrication of holey fibres based on a photonic crystal and hollow waveguides [10–13]. Such waveguides can be used to produce new quasi-one-dimensional nonlinear media by embedding various nonlinear impurities in their hollow core, for example molecules or molecular aggregates, semiconductor microcrystals or metal nanoparticles. The dynamics of acoustic waves generated by the electromagnetic field in a RBG containing resonance impurities in the form of molecular aggregates and the effect of exciton—phonon and exciton—phonon—photon interaction on the parameters of the photonic band structure have been investigated recently [14].

The development of nanotechnologies lead to the creation of composite materials doped with nanoparticles, nanowires, carbon nanotubes, nanomagnets and photonic crystals with metal structure elements. To emphasise their artificial nature, they are called metamaterials [15, 16]. In many cases, metamaterials are produced in the form of films and their electrodynamic properties are determined by plasmon oscillations in nanoparticles. Thus, a Bragg grating can be formed by a periodic sequence of metamaterial films embedded into a usual dielectric (linear or nonlinear) medium.

In this paper we consider a simple model of such a RBG, whose nonlinear properties are determined by the plasmons of the structural elements of a film.

2. Reduced two-wave Maxwell's equations

Let an infinite sequence of thin films of a polarising material be located with a step a normally to the x axis, along which electromagnetic waves with the frequency ω_0 propagate in the forward and backward directions. It is assumed that the film thickness $l_{\rm f}$ is smaller than the radiation wavelength propagating through this medium. The passage from complete Maxwell's equations to the equations for slowly varying pulse envelopes propagating in the forward and backward directions in the RBG is described in many papers and reviews devoted to the

electrodynamics of periodic media [1-3]. By assuming that the total electric-field strength is determined by the expression

$$E(x,t) = [\mathscr{A}(x,t) \exp(iq_0 x) + \mathscr{B}(x,t) \exp(-iq_0 x)]$$

$$\times \exp(-\mathrm{i}\omega_0 t),$$
 (1)

and following papers [4-8, 14, 17], we can write the system of equations, which in the long-wavelength approximations gives the envelope evolution of the forward $[\mathcal{A}(x, t)]$ and backward $[\mathcal{B}(x, t)]$ waves:

$$i\left(\frac{\partial}{\partial x} + \frac{1}{v_g}\frac{\partial}{\partial t}\right) \mathcal{A} - \frac{q_2}{2}\frac{\partial^2}{\partial t^2} \mathcal{A} + \Delta q_0 \mathcal{A} = -\frac{2\pi\omega_0}{c\sqrt{\varepsilon}}\mathcal{P},$$

$$i\left(\frac{\partial}{\partial x} - \frac{1}{v_g}\frac{\partial}{\partial t}\right) \mathcal{B} + \frac{q_2}{2}\frac{\partial^2}{\partial t^2} \mathcal{B} - \Delta q_0 \mathcal{B} = +\frac{2\pi\omega_0}{c\sqrt{\varepsilon}}\mathcal{P},$$
(2)

where $\Delta q_0 = q_0 - 2\pi/a$; $q_0 = \omega_0 \sqrt{\epsilon}/c$ is the wave number in the medium with the dielectric constant ϵ . The parameter q_2 takes into account the group-velocity dispersion v_g of the second order. To determine polarisation $\mathscr P$ in (2), it is necessary to choose a model describing the response of thin films to the external electromagnetic field.

Dielectric properties of the metamaterial are often considered based on Lorenz oscillators for plasmon oscillations, and the magnetic properties are described by a system of oscillating circuits [18-22]. The simplest generalisation of this model is achieved by taking into account anharmonicity of plasmon oscillations [23, 24] or the inclusion of the nonlinear capacity in the oscillating circuit [25]. We consider here thin films of nonmagnetic materials, whose microscopic polarisation P is determined by the expression [23]

$$\frac{\partial^2 P}{\partial t^2} + \omega_d^2 P + \Gamma_e \frac{\partial P}{\partial t} + \kappa P^3 = \frac{\omega_p^2}{4\pi} E,$$
 (3)

where ω_p is the plasma frequency; ω_d is the frequency of the dimensional nanoparticle quantisation; and κ is the anharmonicity constant. The parameter Γ_e takes into account losses caused by the decay of plasmon oscillations. It is assumed that the duration of electromagnetic pulses can be chosen so small that the losses can be neglected. If the constant κ is zero, expression (3) corresponds to the Lorenz model used in [15, 18–21, 26–29] for studying the propagation and refraction of electromagnetic waves in metamaterials.

Because electromagnetic pulses are described in quasiharmonic approximation (1), expression (3) can be reduced in a standard way in the approximation of slowly varying pulse envelopes:

$$i\frac{\partial \mathcal{P}}{\partial t} + (\omega_{\rm d} - \omega_0)\mathcal{P} + \frac{3\kappa}{2\omega_0}|\mathcal{P}|^2\mathcal{P} = -\frac{\omega_{\rm p}^2}{8\pi\omega_0}\mathcal{E}_{\rm tr}.\tag{4}$$

Here, ω_0 is the frequency of the carrier wave; $\mathscr{E}_{tr} = \mathscr{A}(x,t) + \mathscr{B}(x,t)$ is the slowly varying pulse envelope of an electromagnetic field inside a thin film equal to the sum of amplitudes of the forward and backward waves.

Let us pass to dimensionless variables by introducing the following notation:

$$e_1 = \frac{\mathscr{A}}{A_0}, \ e_2 = \frac{\mathscr{B}}{A_0}, \ p = \frac{4\pi\omega_0}{\sqrt{\varepsilon}\,\omega_\mathrm{p}A_0}\mathscr{P}, \ \tau = \frac{t}{t_0}, \ \zeta = \frac{\omega_\mathrm{p}}{2c}\,x.$$
 (5)

The quantity of $t_0 = 2\sqrt{\epsilon}/\omega_p$ is the time interval determining the characteristic scale in the problem under study. For $\delta = 2(c/\omega_p)\Delta q_0$ and by neglecting the second-order group-velocity dispersion, systems (2) and (4) take the form:

$$i\left(\frac{\partial}{\partial \zeta} + \frac{\partial}{\partial \tau}\right)e_1 + \delta e_1 = -p,$$

$$i\left(\frac{\partial}{\partial \zeta} - \frac{\partial}{\partial \tau}\right)e_2 - \delta e_2 = +p,$$

$$i\left(\frac{\partial p}{\partial \tau} + \Delta p + \mu|p|^2 p = -(e_1 + e_2),$$
(6)

where $\Delta=2\sqrt{\varepsilon}(\omega_{\rm d}-\omega_0)/\omega_{\rm p}$ and the parameter $\mu=(3\kappa\sqrt{\varepsilon}/\omega_0\omega_{\rm p})(\sqrt{\varepsilon}\,\omega_{\rm p}/4\pi\omega_0)^2A_0^2$ depends on the normalisation amplitude A_0 .

System of equations (6) can be rewritten in another from by introducing functions

$$f_{s} = -(e_{1} + e_{2}) \exp(-i\delta\tau), f_{a} = (e_{1} - e_{2}) \exp(-i\delta\tau),$$

$$q = p \exp(-i\delta\tau).$$
(7)

Then equations of system (6) take the same form as equations used in [5, 9]:

$$\frac{\partial f_{s}}{\partial \zeta} - \frac{\partial f_{a}}{\partial \tau} = 0, \quad \frac{\partial f_{a}}{\partial \zeta} - \frac{\partial f_{s}}{\partial \tau} = 2iq,
i \frac{\partial q}{\partial \tau} + (\Delta - \delta)q + |q|^{2}q = f_{s},$$
(8)

where the parameter μ determining the nonlinear response of the metamaterial is chosen equal to unity. By excluding f_a , equations (8) can be rewritten in the form:

$$\frac{\partial^2 f_s}{\partial \zeta^2} - \frac{\partial^2 f_s}{\partial \tau^2} = 2i \frac{\partial q}{\partial \tau},$$

$$i \frac{\partial q}{\partial \tau} + (\Delta - \delta)q + |q|^2 q = f_s.$$
(9)

The obtained system of equations describes the propagation of ultrashort pulses in a Bragg grating from nanoparticles, in which plasmon oscillations are induced by the electric field of ultrashort pulses propagating in forward and backward directions.

3. Steady-state solutions describing a solitary wave

Steady-state solutions of system (9) correspond to travelling waves, whose envelopes depend only on the variable $\eta = t - az$ (α is an arbitrary parameter). The boundary conditions for system (9), which have the form

$$f_{s}(\tau,\zeta) = \frac{\partial f_{s}(\tau,\zeta)}{\partial \zeta} = \frac{\partial f_{s}(\tau,\zeta)}{\partial \tau} = q(\tau,\zeta) = 0$$
(for $\tau,\zeta \to \pm \infty$), (10)

single out solitary waves from this set of solutions. We will not consider the solutions with nonzero asymptotics and periodic waves because they do not satisfy boundary conditions (10). The condition for the strict resonance $(\omega_{\rm d} - \omega_0) = 0$ and the condition for the Bragg resonance $q_0 = 2\pi/a$ are also assumed fulfilled [4–9].

By integrating the first equation in (9), under the resonance conditions we obtain from system (9) two ordinary differential equations:

$$\frac{\partial f_{s}}{\partial \eta} = i\beta q, \qquad i \frac{\partial q}{\partial \eta} + |q|^{2} q = f_{s}, \tag{11}$$

where $\beta = 2/(\alpha^2 - 1)$. The passage to the real variables u, r, φ and ψ [$f_s = u \exp(i\phi)$, $q = r \exp(i\psi)$] yields the system of equations

$$\frac{\partial u}{\partial \eta} = \beta r \sin \Phi, \quad \frac{\partial r}{\partial \eta} = u \sin \Phi, \tag{12}$$

$$u\frac{\partial\varphi}{\partial\eta} = \beta r\cos\Phi, \quad r\frac{\partial\psi}{\partial\eta} = r^3 - u\cos\Phi, \tag{13}$$

where $\Phi = \varphi - \psi$. The relation (the first integral of motion) $u^2 - \beta r^2 = \text{const} = 0$ follows from the equations for real amplitudes taking boundary conditions (10) into account. The equations for the phase difference Φ

$$\frac{\partial \Phi}{\partial \eta} = -r^2 + \left(\frac{u}{r} + \frac{\beta r}{u}\right) \cos \Phi \tag{14}$$

and amplitude u [from(12)], taking the first integral of motion into account, yield the system of equations

$$\frac{\partial u}{\partial \eta} = \sqrt{\beta} u \sin \Phi, \quad \frac{\partial \Phi}{\partial \eta} = -\beta^{-1} u^2 + 2\sqrt{\beta} \cos \Phi. \tag{15}$$

Equations (15) lead to the second integral of motion, which, taking boundary conditions (10) into account, can be written in the form

$$\cos \Phi = \left(4\beta\sqrt{\beta}\right)^{-1}u^2. \tag{16}$$

The substitution of (16) into (12) gives

$$\left(\frac{\partial u}{\partial \eta}\right)^2 = \beta u^2 \left[1 - \left(\frac{u^2}{4\beta\sqrt{\beta}}\right)^2\right]. \tag{17}$$

By replacing u by $\omega^{-1/2}$, expression (17) yields

$$\left(\frac{\partial w}{\partial \eta}\right)^2 = 4\beta \left[w^2 - \left(4\beta\sqrt{\beta}\right)^{-2}\right],$$

whose solution is

$$w = \left(4\beta\sqrt{\beta}\right)^{-1}\cosh\left[2\sqrt{\beta}(\eta - \eta_0)\right].$$

The integration constant η_0 determines the position of the pulse maximum. Thus, the solution of amplitude equations (12) has the form:

$$u^{2}(\eta) = \frac{4\beta\sqrt{\beta}}{\cosh\left[2\sqrt{\beta}(\eta - \eta_{0})\right]},$$

$$r^{2}(\eta) = \frac{4\sqrt{\beta}}{\cosh\left[2\sqrt{\beta}(\eta - \eta_{0})\right]}.$$
(18)

In addition, it follows from (14) and (18) that

$$\sin \Phi(\eta) = -\tanh \left[2\sqrt{\beta}(\eta - \eta_0)\right]. \tag{19}$$

By using (18), the phases

$$\varphi(\eta) = \varphi_0 \pm \arctan \tanh \left[\sqrt{\beta} (\eta - \eta_0) \right], \tag{20}$$

$$\psi(\eta) = \psi_0 \pm 3 \arctan anh \left[\sqrt{\beta} (\eta - \eta_0) \right]$$

are determined from (13). The initial values of phases are chosen so that the phase difference for $\eta \to -\infty$ was determined by the condition $\Phi_0 = \varphi_0 - \psi_0 = \pi/2$. It follows from (15) that when the field strength of the electromagnetic pulse is small, this relation between the initial phases provides an increase in the amplitude of plasmon oscillations with increasing the amplitude of the driving force, i.e. the field amplitude.

By using (7) we can obtain the normalised envelopes of the waves, which form a coupled state – split soliton:

$$e_1(\eta) = -0.5(1+\alpha)f_s(\eta)\exp(i\delta\tau),$$

$$e_2(\eta) = -0.5(1-\alpha)f_s(\eta)\exp(i\delta\tau)$$
(21)

in the medium. In the linear approximation, these waves are counterpropagating.

4. Nonstationary waves

To study the nonstationary behaviour of nonlinear solitary waves, we solved numerically system of equations (8) in the range $(-10 < \zeta < 10)$ with the initial conditions

$$e_1(-10, \tau) = e_{10} \exp\{-0.5[(\tau - 3)/1.5]^2$$

$$+i \arctan \tanh[1.5(\tau-3)], e_2(10,\tau) = 0,$$
 (22)

and $q(-10,\tau) = q(10,\tau) = 0$ for $\tau \to -\infty$. The results of calculations showed that a stationary pulse corresponding to (18)-(20) propagates, as was expected, without any distortions. Initial pulses (22) for $e_{10} > 3$ exhibit a complex evolution. Because two counterpropagating electromagnetic waves are coupled with plasmon oscillations in the Bragg grating, it is convenient to present the results of calculations as the dependence $|q(\zeta,\tau)|$ on the normalised spatiotemporal variables of the plasmon-wave envelope. One can see in Fig. 1 that the input pulse close to the Bragg soliton in amplitude acquires amplitude modulation and emits linear waves in the forward and backward directions. An increase in the amplitude of the input pulse leads to the formation of a Bragg soliton (Fig. 2). We can distinguish here three evolution stages. At the first stage $(-10 < \zeta < -7)$, the initial pulse is split into the fast and slow waves. The slow wave is coupled with plasmon oscillations in the grating, which are excited by the electromagnetic pulse. At the next stage $(-7 < \zeta < 0)$, the fast solitary wave transforms into a Bragg soliton and the amplitude and the propagation velocity of slow waves decrease. In essence, they transform into localised oscillations in the Bragg grating. At the third stage $(0 < \zeta < 9)$, a quasi-stationary pulse with a small emission of the linear wave remains. A further increase in the initial-pulse amplitude is accompanied by the formation of a Bragg soliton and a nonstationary pulse whose

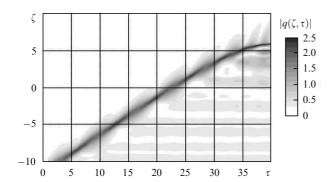


Figure 1. Nonstationary pulse of plasmon oscillations close in amplitude to the Bragg soliton ($e_{10} = 2$).

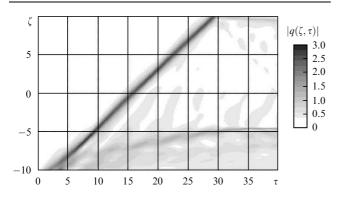


Figure 2. Nonstationary pulse of plasmon oscillations close in amplitude to the Bragg soliton ($e_{10} = 3.5$).

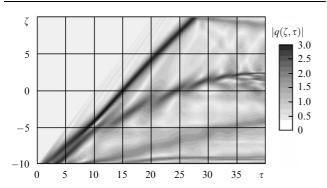


Figure 3. Propagation of a nonstationary pulse of plasmon oscillations $(e_{10} = 4)$ exhibiting a slowing down of an electromagnetic wave.

propagation velocity almost vanishes. One can see in Fig. 3 that the slow solitary wave stops near the point $\zeta \approx 0$.

5. Conclusions

We have considered the propagation of ultrashort pulses in the Bragg grating which is formed by thin films from metal nanoparticles embedded into a dielectric matrix. Plasmon oscillations in these particles are described by the model of an anharmonic oscillator with the driving force proportional to the electric-field strength of an electromagnetic pulse [23]. It was assumed that the matrix was made of a linear nondispersive material. In the long-wavelength approximation equations have been obtained for slowly varying envelopes of counterpropagating ultrashort pulses and for nonlinear polarisation of thin films. If the fields are weak, so that nonlinear properties of the medium are

insignificant, one can find the spectrum of electromagnetic waves in this system and make sure that it contains forbidden bands, as a one-dimensional photonic crystal should

The nonlinear effects are most pronounced upon the resonance interaction of radiation with the medium. Due to a scatter in the nanoparticle sizes (in the general case, for any other structural units of the metamaterial), the oscillator eigenfrequencies differ. If it is necessary to take into account the effect of the inhomogeneous resonance line broadening, equations (9) should be modified:

$$\frac{\partial^2 f_s}{\partial \zeta^2} - \frac{\partial^2 f_s}{\partial \tau^2} = -2i \frac{\partial \langle q \rangle}{\partial \tau},$$

$$i \frac{\partial q}{\partial \tau} + (\Delta - \delta)q + |q|^2 q = f_s,$$
(23)

where angle brackets show the averaging of the normalised polarisation amplitude over all frequency detunings within the inhomogeneously broadened resonance line. The deviation from the Bragg resonance can occur due to fluctuations of the grating step. Therefore, averaging in (23) assumes averaging of Bragg detunings. The role of inhomogeneous broadening is significant for such effects as photon echo and free induction decay [30]. By restricting ourselves to the consideration of nonlinear solitary waves, we can assume for simplicity that the inhomogeneous broadening is absent. In this case under the assumption that the conditions for the Bragg resonance are fulfilled and the frequencies of the carrier wave and oscillators coincide, we have obtained an expression for the envelopes of solitary waves of the ultrashort pulse field and polarisation. This solution describes the propagation of a coupled pair of the forward and backward waves of the electromagnetic radiation. The polarisation repeats the envelope of the stationary ultrashort pulse. Instantaneous values of the phase of each of the pulses change inside the ultrashort pulse, which means a slow change in the carrier-wave frequency.

Stationary ultrashort pulses from a continuous family of pulses, which is numbered by the positive parameter β = $2/(\alpha^2-1)$ determining both the pulse width and its group velocity. If we express the independent variable η by physical variables $[\eta = (\omega_p/2\sqrt{\epsilon})(t - \alpha x\sqrt{\epsilon}/c)]$, one can see from the obtained expression that all terms of the family of stationary ultrashort pulses propagate slower that the linear wave in the matrix. Therefore, during the decay of the initial nonstationary pulse we can expect the formation of slow Bragg solitons. Either the collision of two stationary pulses or interaction of the stationary ultrashort pulses with a weak linear wave can lead to this effect. The numerical solution of the corresponding system of equations has shown that during the decay of the initial pulse, apart from a soliton a slow nonstationary wave is formed, which finally stops. This phenomenon is related to the excitation of localised modes of plasmon oscillation in thin films, which form the grating. If we take into account the interaction between the films, we can expect that the slow plasmon wave evolves into the nonlinear wave, whose propagation velocity will be determined by the interaction between the films. Equation for polarisation envelope (4) takes the form of a nonlinear Schrödinger equation with a driving force. In the future we will study this process in more detail.

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