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NONLINEAR LIGHT SCATTERING

Nonlinear Induced Reflection of Light Waves in Semiconductors

A. F. Bunkin*, V. G. Mikhalevich, S. M. Pershin***, and V. N. Streltsov******

Wave Research Center, Prokhorov General Physics Institute, Russian Academy of Sciences, ul. Vavilova 38, Moscow, 119991 Russia

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Abstract—A scheme of nonlinear optical four-wave mixing of two counterpropagating laser beams on the surface of a semiconductor is proposed and analyzed. It is shown that the density modulation of the electron-hole plasma current carriers in the light-induced grating manifests itself in the probe beam depolarization signal after the reflection from the surface, which is sensitive to the state of the surface and presence of complex molecules on it.

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1. INTRODUCTION

In particular cases, multiwave active spectroscopy of condensed matters is based on scattering or absorption of the probing electromagnetic wave on collective light-induced excitations of the medium. In semiconductors or metals these excitations can be perturbations in the electron-hole solid plasma. In our recent work [1], we paid attention to the fact that longitudinal density oscillations can be excited in the field of two counterpropagating quite intensive transverse electromagnetic beams in semiconductor plasma, which leads to partial nonlinear reflection of these beams. In experiments on four-wave light scattering [2–4], this reflection can effectively manifest itself as rotation of the polarization vector of the detected signal.

2. THEORY

In this work we consider another possible physical mechanism for stationary modulation of conduction electron density in a semiconductor, which leads to similar nonlinear reflection. Modulation arises from interband optical transitions in a semiconductor in the field of the counterpropagating electromagnetic waves $E^{(1)}$ and $E^{(2)}$ with equal frequency ω:

$$\mathbf{E}^{(1)} = \frac{1}{2}\mathbf{E}_1 \exp\left[-i(\omega t - kz)\right] + \text{c.c.},$$

$$\mathbf{E}^{(2)} = \frac{1}{2}\mathbf{E}_2 \exp\left[-i(\omega t + kz)\right] + \text{c.c.}$$

It is assumed that frequency ω exceeds the width of the bandgap and the frequency of the plasma oscillation corresponding to the equilibrium concentration of the conduction electrons in the sample, N_0. Vertical interband optical transitions will occur in the medium under the effect of illumination. The free electron generation rate G is determined by the optical absorption coefficient of the medium, γ, and the illumination intensity: $G = \gamma|\mathbf{E}^{(1)} + \mathbf{E}^{(2)}|^2$. For semiconductors with spherical energy bands and extrema lying at one point,

$$\gamma = \frac{\sqrt{2}}{3}\frac{e^2}{\omega c m_{\text{p}}^2 \hbar^2}(m_{\text{r}})^{5/2}(\hbar\omega - \Delta E)^{3/2}, \quad (1)$$

where c is the light velocity in vacuum, m_{p} is the effective mass of the holes, m_{r} is the reduced mass of the electron and the hole, and ΔE is the width of the bandgap.

To avoid overcomplication in what follows, we assume that the mass of holes is much larger than the effective mass m^* of electrons, and the latter is scalar. Holes thus make up a fixed neutralizing background.

To describe kinetics of electron plasma, we will consider the zero moment of the distribution function f of electrons, that is, the continuity equation with a collision term corresponding to the particle generation and recombination processes:

$$\partial n/\partial t + \operatorname{div}\mathbf{j} = \gamma\left|\mathbf{E}^{(1)}+\mathbf{E}^{(2)}\right|^2 - (n-n_0)/\tau_{\text{rel}}. \quad (2)$$

Here n is the density of free electrons, n_0 is the dark (in the absence of illumination) concentration of electrons, $\mathbf{j} = \int \mathbf{V} f(\mathbf{r}, \mathbf{V}, t)\, d\mathbf{V}$, τ_{rel} is the longitudinal time of interband relaxation in the system.

In the one-dimensional model for the stationary regime, we have

*E-mail: abunkin@kapella.gpi.ru
**E-mail: mikhal@kapella.gpi.ru
***E-mail: pershin@kapella.gpi.ru
****E-mail: mpvstrel@mail.ru

$$\frac{dj}{dz} = \frac{\gamma}{4}\left[\mathbf{E}_1\mathbf{E}_2^*\exp(-2ikz) + \text{c.c.}\right] - \frac{n(z) - N_0}{\tau_{\text{rel}}}, \quad (3)$$

where N_0 is the equilibrium density of the electrons in the field,

$$N_0 = n_0 + \frac{\gamma}{4}\tau_{\text{rel}}\left(|\mathbf{E}_1|^2 + |\mathbf{E}_2|^2\right). \quad (4)$$

Spatial inhomogeneity of illumination leads to inhomogeneity of the distribution function f. A diffusion current occurs in the system due to the gradient of the effective chemical potential proportional to dn/dz. The diffusion shift in turn gives rise to an uncompensated volume charge and an internal electric field with strength $\mathbf{E}$:

$$\operatorname{div}\mathbf{E} = 4\pi e(n - N_0), \quad (5)$$

and also to an ohmic current proportional to the total density of the free electrons. Equation (3) takes the form

$$\frac{\langle\tau\rangle}{m^*}\frac{d}{dz}\left[e(N_0 + y)E - kT\frac{dy}{dz}\right] = \frac{\gamma}{4}\left[\mathbf{E}_1\mathbf{E}_2^*\exp(-2ikz) + \text{c.c.}\right] - \frac{y}{\tau_{\text{rel}}}. \quad (6)$$

Here $y = n(z) - N_0$ is the electron density deviation of interest, and $\langle\tau\rangle$ is the average (with energy weight) relaxation time of the distribution function.

We intentionally left in (6) the microscopic kinetic parameters of the semiconductor that determine the mobility of electrons $\mu = e\langle\tau\rangle/m^*$.

In what follows we assume that the intensity of one of the pump waves is substantially higher than the intensity of the other wave. Then $y < N_0$, and considering (5), we ultimately obtain from (6)

$$\frac{d^2y}{dz^2} - \frac{m^*}{kT}\left[\omega_{\text{p}}^2 + \frac{1}{\langle\tau\rangle\tau_{\text{rel}}}\right]y = -\frac{m^*}{kT\langle\tau\rangle}\frac{\gamma}{4}\mathbf{E}_1\mathbf{E}_2^*\exp(-2ikz) + \text{c.c.}, \quad (7)$$

where $\omega_{\text{p}}^2 = 4\pi N_0 e^2/m^*$ is the induced plasma frequency.

Note that the condition $y < N_0$ is also fulfilled at comparable intensities of the pump waves when the dark electron density is rather high, $n_0 \approx N_0$.

It is very simple to find the solution to (5):

$$y = \chi\mathbf{E}_1\mathbf{E}_2^*\exp(-2ikz) + \text{c.c.}, \quad (8)$$

$$\chi = \frac{m^*}{kT\langle\tau\rangle}\frac{\gamma}{4}\frac{1}{4k^2 + \dfrac{m^*}{kT}\left(\omega_{\text{p}}^2 + \dfrac{1}{\langle\tau\rangle\tau_{\text{rel}}}\right)}.$$

Thus, the electron-hole plasma density suffers periodic spatial modulation governed by the optical pump field of the sample.

To describe dynamics of a transverse electromagnetic wave (e.g., $\mathbf{E}_2$) in a medium, one can proceed from the Maxwell equations with the transverse current $j_\perp$ and the vector of polarization $P_\perp$ in the form

$$j_\perp = neV_\perp(t), \quad P_\perp = ner_\perp(t), \quad (9)$$

where $V_\perp(t)$ and $r_\perp(t)$ are, respectively, the classical velocity and displacement of the electron in the electric field of the wave under consideration. Far from exciton resonances in the weak-collision limit for $\mathbf{P}_2$ we obtain

$$\mathbf{P}_2 = -\frac{1}{2}\frac{e^2}{\omega^2}\mathbf{E}_2\exp\left[i(\omega t + kz)\right](N_0 + y). \quad (10)$$

As could be expected, in the system there arises a reflected wave $\mathbf{E}_{\text{ref}}$ with polarization vector $\mathbf{E}_2$ corresponding to the light-modulated nonlinear part $\mathbf{P}_{2\text{nl}}$,

$$\mathbf{P}_{2\text{nl}} = -\frac{1}{2}\frac{e^2}{\omega^2}\chi\mathbf{E}_2(\mathbf{E}_1\mathbf{E}_2^*)\exp\left[i(\omega t - kz)\right]. \quad (11)$$

For $\mathbf{E}_{\text{ref}}$ we have an ordinary wave equation

$$\Delta\mathbf{E}_{\text{ref}} - \frac{1}{c^2}\frac{\partial^2 E_{\text{ref}}}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2\mathbf{P}_{2\text{nl}}}{\partial t^2}. \quad (12)$$

Going in (12) to the slow stationary amplitude $\mathbf{E}_{\text{ref}}(z)$,

$$\mathbf{E}_{\text{ref}} = \frac{1}{2}\mathbf{E}_{\text{ref}}(z)\exp\left[i(\omega t - kz)\right] + \text{c.c.},$$

we arrive at the equation

$$-ik\frac{d\mathbf{E}_{\text{ref}}}{dz} = -\frac{\pi e^2}{c^2}\chi\mathbf{E}_2(\mathbf{E}_1\mathbf{E}_2^*). \quad (13)$$

From the above we ultimately obtain the obvious result for the exit amplitude in the approximation of the given pump field:

$$\mathbf{E}_{\text{ref}} = i\pi\frac{e^2}{kc^2}\chi\mathbf{E}_2(\mathbf{E}_1\mathbf{E}_2^*)L, \quad (14)$$

where L is the effective mixing length of the counterpropagating pump waves.

3. CONCLUSIONS

It is worth noting that all the results of this work are valid (with necessary quantitative corrections) for any types of semiconductors and optical transitions mechanisms. The above-mentioned mechanism for nonlinear optical mixing of laser waves on the surface of a semiconductor can be used for developing a new generation of optical metamaterials sensitive to the presence of complex molecules [5].

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