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1) This approximation is of course valid in the time interval $\tau < a/u$, where a is the space scale.

2) The introduction of model-dependent sound damping into Eq. (2) leads to a weakening of this breakup.

3) To estimate the upper bound of the results of one-dimensional self-focusing, we assume $n = -|A^2(\tau)|$.

[1] V. E. Zakharov, Zh. Eksp. Teor. Fiz. 62, 1745 (1972) [Sov. Phys.-JETP 35, 908 (1972)].

[2] V. I. Bespalov, A. G. Litvak, and V. I. Balanov, in: Nelineinaya optika (Nonlinear Optics), Nauka, 1968, p. 428.

RAMAN SCATTERING OF LIGHT BY SURFACE POLARITONS IN MEDIA WITH INVERSION CENTERS

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A broken symmetry method is proposed and makes it possible to use Raman scattering of light (RS) of first order by surface polariton to study the dispersion of the dielectric constant in media (crystals, glasses) having an inversion center. A formula is obtained for the intensity of the considered process, and the dependence of the line width of the RS by surface polaritons on the scattering angle is determined.

1. It is known that the Raman scattering (RS) of light by polaritons, in conjunction with lasers, has yielded valuable information on the dispersion of light in crystals. In centrosymmetrical media (CSM) such as crystals or glasses, however, this method cannot be used, for in these media the nonlinear susceptibility tensor χ_{ijl} , which determines the intensity of the process, vanishes identically. The situation changes, however, if one resorts to RS of light by surface polaritons under conditions when at least one of the surfaces of the CSM borders on a medium that is transparent¹⁾ to the laser radiation, but one having no inversion center. Since the electromagnetic field in the surface polariton differs from zero over distances on the order of its wavelength ($\lambda \approx 10 \mu$) on both sides of the interface, and consequently also in the region where $\chi_{ijl} \neq 0$, the intensity of the RS by the surface polariton turns out to be different from zero and, as will be shown by the calculation below, is strong enough to be experimentally observable²⁾. This conclusion agrees also with the results of [1], where RS of light by surface polaritons was first observed in a medium without an inversion center (GaAs on a sapphire substrate). Naturally, in the RS spectrum obtained in [1] the most intense peaks corresponded to excitation of volume polaritons, and this hindered the observation of the effect. In the "inverted" situation proposed by us, where absence of an inversion center is ensured by the choice of the substrate, the volume polaritons of the CSM should not be excited at all.

2. We proceed to calculate the effectiveness of the process. Neglecting the possible anisotropy of the media, and also the dispersion of light in the substrate, we assume its dielectric constant ϵ_1 to be positive, and assume $\chi_{ijl} = \chi |e_{ijl}|$, where e_{ijl} is a fully antisymmetric tensor of third rank. The measured dispersion of the surface polariton, $\omega = \omega(\vec{k})$, is determined (see [2]) by the relation $k^2 = \omega^2 \epsilon_1 \epsilon / c^2 (\epsilon + \epsilon_1)$, where $\epsilon = \epsilon(\omega) < 0$ is the dielectric constant of the CSM. By way of the perturbation we use the operator

$$\hat{H} = -\frac{1}{2} \int_{z>0} \chi_{ijl} \hat{E}_i \hat{E}_j \hat{E}_l^p dr,$$

where \hat{E} and \hat{E}^p are the electric intensity operators of the high-frequency field and of the field in the surface polariton. Neglecting the difference between the refractive indices of the laser

light in the CSM and in the substrate, we disregard, for the sake of simplicity, the possible reflection of the high-frequency field from the interface, since this reflection is immaterial for the estimate of the effect. We assume for concreteness that the CSM and the substrate occupy the half-spaces $z \leq 0$ and $z > 0$, respectively, and that the laser beam propagating along the z axis (wave vector \vec{k}^a) is polarized along the x axis, while the scattered light (wave vector $(\vec{k}^b \equiv (k_x^b, 0, k_z^b))$ is polarized along y . In the polariton in question (wave vector $\vec{k} = \vec{k}^a - \vec{k}^b$ parallel to the x axis) the relations between the nonzero amplitudes of the electromagnetic field are

$$E_1^p = -i \frac{\kappa_0}{k} E_3^p = -i \frac{c \kappa_0}{\omega \epsilon_1} H_2^p, \quad \kappa_0 = (k^2 - \frac{\omega^2}{c^2} \epsilon_1)^{1/2}, \quad k^2 > \frac{\omega^2}{c^2} \epsilon_1$$

if $z > 0$

and

$$E_1^p = -i \frac{\kappa}{k} E_3^p = -i \frac{c \kappa}{\omega \epsilon} H_2^p, \quad \kappa = (k^2 - \frac{\omega^2}{c^2} \epsilon)^{1/2}, \quad \epsilon < 0 \quad \text{if } z < 0$$

On the other hand, the absolute values of these amplitudes can be determined from the condition that $\hbar\omega$ be equal to the energy of the electromagnetic field in the polariton. Calculation shows that the only polariton electric-field component in the matrix element of the perturbation operator is the z component and is given at $z > 0$ by the relation $|E_3^p|^2 = (4\pi\hbar\omega/S)\Phi(k)$, where S is the area of the interface and

$$\Phi(k) = \frac{k^2}{\kappa_0^2} \left[k^2 \left(\frac{\epsilon_1}{\kappa_0^3} + \frac{\epsilon}{\kappa^3} \right) + \frac{\omega}{2\kappa} \left(1 + \frac{k^2}{\kappa^2} \frac{d\epsilon}{d\omega} \right) \right]^{-1}.$$

Using this relation and also the known values of the amplitudes of the high-frequency field, we can easily prove within the framework of ordinary perturbation theory that the differential effectiveness of the spontaneous RS of light by a surface polariton is determined in the considered system by the relation (at a crystal temperature $T = 0$)

$$\frac{\partial^2 I}{\partial \Omega \partial \omega} = \frac{4\pi\hbar\omega_a^4 \omega}{c^4} \chi^2 \Phi(k) [\kappa_0^2 + (k_z^a - k_z^b)^2]^{-1} \delta(\omega_a - \omega_b - \omega).$$

This relation enables us to trace the dependence of the RS intensity on the scattering angle. It is minimal in the nonrelativistic limit

$$(k \gg \frac{\omega}{c} |\epsilon|, \frac{\omega}{c} \epsilon_1), \text{ where } \Phi(k) = \frac{2k}{\omega} \left(\frac{d\epsilon}{d\omega} \right)^{-1} \text{ and at } \epsilon(\omega) = \epsilon_\infty + \frac{(\epsilon_0 - \epsilon_\infty)}{\omega^2 - \omega_1^2} \omega_1^2$$

(ω_1 is the frequency of the transverse phonon in the CSM) we have

$$\Phi(k) \approx k \frac{\omega_1^2}{\omega^2} \frac{(\epsilon_0 - \epsilon_\infty)}{(\epsilon_1 + \epsilon_\infty)^2}.$$

The intensity integrated over the width of the polariton line is then $dI/d\Omega = 4\pi\hbar\omega_a^4 \omega_1^2 (\epsilon_0 - \epsilon_\infty)^2 \chi^2 / c^4 k \omega (\epsilon_1 + \epsilon_\infty)^2$. Assuming³⁾ $\omega_a/c = 10^5$, $k = 10^3$, $\omega_1 = 10^{13}$, $\epsilon_1 + \epsilon_\infty = 10$, $\epsilon_0 - \epsilon_\infty = 3$, and $\chi = 10^{-6}$, we obtain $dI/d\Omega \approx 4 \times 10^{-11}$, which, as already indicated, is quite sufficient to observe the effect. The value of $dI/d\Omega$ increases appreciably on going to the relativistic region ($k \approx (\omega/c) |\epsilon|$).

3. The line broadening in RS by the surface polariton, due to the damping of the light, can be easily obtained from the condition that determines the pole of the electromagnetic Green's function in the presence of an interface (for a similar analysis for volume polaritons see [3, 4]). This condition takes the form $\epsilon = \epsilon_1 k^2 / (k^2 - \epsilon_1 \omega^2 / c^2)$. At $\epsilon(\omega) = \epsilon_\infty + \omega_1^2 (\epsilon_0 - \epsilon_\infty) / (\omega_1^2 - \omega^2 + 2i\omega\pi_0)$ we obtain for the width of the line of RS by a surface polariton the relation $\Gamma(k) = \Gamma_0 k^2 [k^2 + (\epsilon_0 - \epsilon_\infty) (\omega_1^2 / c^2)]^{-1}$. Thus, at large k , we get $\Gamma(k) \approx \Gamma_0$. On the other hand, if k^2 equals its limiting value $\omega_1^2 \epsilon_1 / c^2$, then $\Gamma = \Gamma_0 \epsilon_1 / (\epsilon_1 + \epsilon_0 - \epsilon_\infty)$, so that at $\epsilon_0 - \epsilon_\infty \gg \epsilon_1$ there should be an appreciable narrowing of the RS lines with increasing scattering angle. We can consider analogously the $\Gamma(k)$ as a function of k for multilayered media.

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1) If this medium (i.e., the substrate) is opaque to the laser then Raman scattering of light in reflection can be used.

2) Allowance for the effect of the substrate on the dispersion of surface polaritons does not raise any difficulties (see [2] and below).

3) For concreteness, we use the data for GaP in the He-Ne emission region (in cgs esu).

[1] D. J. Evens, S. Ushieda, and J. D. McMullen, Phys. Rev. Lett. 31, 369 (1973).

[2] L. D. Landau and E. M. Lifshitz, Elektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Gostekhizdat, 1957 [Pergamon, 1965].

[3] H. J. Benson and D. J. Mills, Phys. Rev. B1, 4835 (1970).

[4] V. M. Agranovich and V. L. Ginzburg, Zh. Eksp. Teor. Fiz. 61, 1243 (1971) [Sov. Phys.-JETP 34, 662 (1972)].

USE OF PROTON CHANNELING TO DETERMINE THE LOCATION OF OXYGEN DISSOLVED IN A NIOBIUM SINGLE CRYSTAL

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Using the fact that the flux density of charged particles becomes redistributed in a channel, we determine the location of oxygen introduced into a niobium single crystal. The oxygen is identified by the nuclear reaction $O^{18}(p, \alpha)N^{15}$. It is shown that the oxygen occupies octahedral interstices.

The dynamic redistribution of the flux density of self-trapping particles in a single crystal [1 - 3] uncovers prospects for investigating the positions occupied in the lattice of a single crystal by as few foreign atoms as several tenths or hundredths of an atomic percent of the impurity.

To determine the positions occupied by the oxygen in the single-crystal niobium, we used the nuclear reaction $O^{18}(p, \alpha)N^{15}$. The differential cross section of the reaction is ~ 50 mb at a proton energy $E_p = 700$ keV. The niobium single crystal was oriented relative to a proton beam with a divergence angle not exceeding 0.05° by using a goniometer having three degrees of freedom [4]. The elastically scattered protons and α particles of the reactions were registered with charged-particle spectrometers based on semiconductor detectors.

Niobium single-crystal samples measuring $10 \times 5 \times 1$ mm were cut perpendicular to the $\langle 111 \rangle$, $\langle 110 \rangle$, and $\langle 100 \rangle$ axes and were saturated with oxygen enriched with O^{18} to 42% at a pressure 4×10^{-4} mm Hg and a temperature 1150°C . The homogenization was at 1750°C and 5×10^{-6} mm Hg for 20 minutes. The oxygen concentration in the samples was 0.1 to 0.2 at. %.

Figure 1 shows the spectrum of the protons elastically scattered by the niobium atoms (a) and of the α particles of the reaction (b), with the proton direction coinciding with the $\langle 100 \rangle$ axis. It shows the spectrum of the protons (A) and α particles (B) when there is no channeling. The yield of the scattered protons is one-ninth that for the channeled beam, thus indicating that the original single crystal was of good quality.

To determine the positions occupied by the oxygen, we performed angle scanning in steps of 0.05° in the vicinities of the axes $\langle 111 \rangle$, $\langle 100 \rangle$ and the planes $\{100\}$, $\{110\}$, and $\{122\}$ with simultaneous registration of the yields of protons elastically scattered by the niobium atoms, and of the α particles from the reaction $O^{18}(p, \alpha)N^{15}$. The results of the scanning are shown in Fig. 2. An appreciable increase in the yield of the α particles is observed when the direction of the proton beam coincides with the crystallographic axes $\langle 111 \rangle$ and $\langle 100 \rangle$ and with the planes $\{110\}$ and $\{122\}$. This indicates that oxygen is an interstitial impurity occupying regular interstices [5].