The nature of new spectral lines in the spectrum of scattered light detected by means of acousto-electronic amplification in piezoelectric semiconductors

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A new line and a continuous background in the spectrum of light scattered by ZnO and CdS crystals in a constant electric field have been observed experimentally. The observed phenomenon is described theoretically.

In the letter of Velichkina *et al.*,[1] the spectrum of light scattered by a piezoelectric semiconductor crystal ZnO, situated in a static electric field greater than the critical field,[1)] was found to exhibit new spectral lines between the unshifted line and that Mandel'shtam-Brillouin (MB) component which increases in intensity with increasing strength of the electric field.

In this letter we continue the theoretical and experimental studies which have enabled us to determine the nature of these lines. We used ZnO and CdS crystals in the experiments. The spectra of light scattered by CdS crystals were also found to exhibit new lines. In addition to these lines, the spectrum of light scattered by ZnO and CdS crystals revealed the presence of a continuous nonuniform background which extended from the MB component, amplified in the electric field, to the unshifted line.

Figure la is a trace of the spectrum of molecular scattering of light by a CdS crystal and Fig. 1b is a trace of the spectrum of light scattered by the same crystal after the application of an electric field. We see that the intensity of the new line may be greater than the dominant Mandel'shtam-Brillouin component.

The continuous background and the additional lines can be explained in a natural way. In piezoelectric semiconductors in a static electric field, the piezoelectrically active, elastic heat waves increase markedly in strength within a certain bandwidth. These waves are responsible for the scattering of light observed by us. In experiments in which the new spectral lines and the continuous background were observed, the frequency of an elastic heat wave, which accounts for the Mandel'shtam-Brillouin component, was found to differ appreciably from the frequency at which the sound reached its maximum intensity.[2] The wave vectors of the incident light and of the light scattered by an elastic wave satisfy the Bragg condition. The rest of the elastic waves do not satisfy the Bragg condition in the chosen direction of observation of the scattered light. The amplitudes of these waves (over a certain frequency range) can, however, be much larger than the amplitude of the wave responsible for the Mandel'shtam-Brillouin component. The diffraction of light by these waves will accordingly contribute significantly to the intensity of the region of the spectrum being studied.

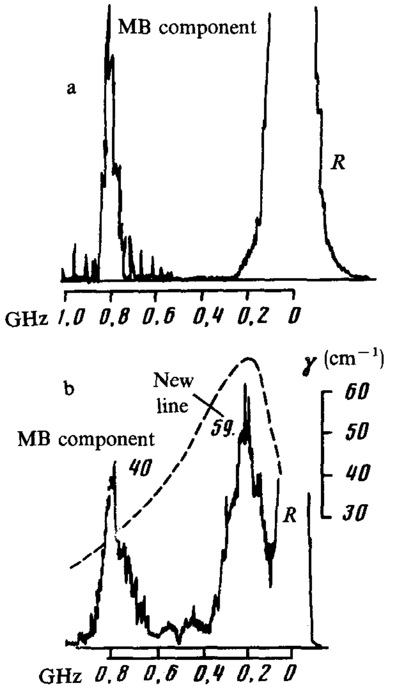

FIG. 1. a—The spectrum of light scattered by a CdS crystal; the Mandel'shtam-Brillouin (MB) component is the spectral component due to the scattering by a piezoelectrically active, transverse heat wave, and R is the Rayleigh line; b—the same as in (a), except that an electric field E is applied to the crystal: $E/E_c = 1.5$, $x = 2.4$ mm, and the conductivity is $\sigma = 2.2 \times 10^{-8}\ \mathrm{s}^{-1}$. The intensities of the MB component and of the new line are given relative to the intensity of the thermal MB component. The dashed curve represents the frequency dependence of the amplification factor of sound in CdS crystal, calculated from Eq. (17) in Ref. 2 for the electron mobility $\mu = 200\ \mathrm{cm}^2/(\mathrm{V}\cdot\mathrm{s})$.

This contribution can be determined by calculating the intensity of light, I, diffracted by the acoustic wave of arbitrary frequency ν in the chosen direction of observation. The intensity $I(\nu)$, divided by the intensity of the MB component, $I_1(\nu_1)$, can be written in the form

$$\frac{I(\nu)}{I_1(\nu_1)} = F(\Delta k\ R)\exp\{x[\gamma(\nu, E) - \gamma_1(\nu_1, E)]\}. \tag{1}$$

Here $\Delta k = |k - k_1|$; ν_1, $k_1(\nu_1)$, and $\gamma_1(\nu_1)$ are the frequency, the wave number, and the gain of the acoustic wave for which the Bragg condition is satisfied, and ν, $k(\nu)$, and $\gamma(\nu)$ are the same quantities for the wave of frequency ν under consideration. We now introduce x, the coordinate of the point at which the scattering in the bulk of the crystal occurs (this point is reckoned from the cathode in the direction of the applied electric field), and R is the radius of the laser beam. The function $F(y)$, where $y = \Delta kR$, describes the scattering of light by acoustic waves of arbitrary frequencies. At $y < 1$ we always have

$$F = 1 - Cy^2, \quad (2)$$

where $C \sim 1$. Hence we can see that the scattering peaks at $y = 0$, i.e., at $k = k_1$ when the Bragg condition holds. The half width of the line, as we can infer from (2), is

$$\Delta k / k \approx (k_1 R)^{-1}.$$ [2]

In our experiments we used a lens to focus the light into the crystal. The radius of the laser beam, R, in the caustic of the lens was no greater than 5×10^{-3} cm in air.[3] The Mandel'shtam-Brillouin spectra were recorded at different scattering angles ranging from 2° to 7°. The Bragg condition was satisfied for acoutic waves with wave vectors k_1 in the range $(0.6\text{–}2) \times 10^4$ cm^{-1}, respectively. In the case of large values of y, i.e., at $|\Delta k| \gg 1/R$, the decay of the function $F(y)$ depends essentially on the distribution of the intensity along the laser-beam profile $W(r)$, where r is the distance to the beam axis. If $W(r)$ has the shape of a step, for example, we would have $F(y) \sim 1/y^3$. If the intensity has a parabolic profile $[W(r) \simeq 1 - r^2/R^2]$, the function $F(y)$ would behave approximately as $1/y^4$ and if the beam has a Gaussian shape $[W(r) = \exp(-r^2/R^2)]$, we would have $F(y) \sim \exp(-y)^2$.

The factor in front of the exponent in Eq. (1) describes the ratio of the intensities of the scattering of the laser beam by elastic sinusoidal waves with the wave vectors k and k_1 in the chosen direction and the exponential function takes into account the difference between the amplitudes of these waves which lie within the sound amplification band. This exponential function leads to a deformation of the line and to the appearance of a background—the scattering increases toward the frequencies ν at which $\gamma(\nu) > \gamma_1(\nu_1)$. In contrast, the scattering terminates abruptly in the opposite direction. This behavior is clearly illustrated in Fig. 1b. In the case of reasonably large values of x and a reasonably large difference $\gamma(\nu) - \gamma_1(\nu_1)$ (in the example shown in Fig. 1b, the exponential factor is on the order of 2×10^5), an increase in the scattering due to an increase in the amplitude of the elastic wave may exceed the decrease in $F(y)$. In this case we see the appearance of an additional line, whose frequency is approximately equal to the frequency ν_m at which the amplification factor of sound is maximum, $\gamma(\nu_m) = \gamma_{\max}$. Furthermore, the new line should be slightly shifted relative to the frequency ν_m toward the MB component. The line shift increases as the decay of the function $F(y)$ near the new line accelerates. To check the validity of our explanation of the background and of the new single lines, we carried out the following experiments. By increasing the electrical conductivity and decreasing the scattering angle of light of the CdS crystal we were able to match the frequency of the MB component with the frequency at which the amplification band of sound is maximum. As a result, the background and the new lines vanish in the scattered-light spectrum (Fig. 2b). A further decrease of the scattering angle and of the frequency of the MB component shifts the background to the rf region (with respect to the MB component) of the spectrum, as predicted by the theory. The relative intensity of the new line increases with increasing scattering-volume coordinate x and with increasing strength of the electric field E, in complete agreement with Eq. (1).

We conclude on the basis of these observations that the results of the experiments on Bragg diffraction by Debye waves are strongly affected by the intensity distribution in the laser beam. Accordingly, we cannot generally draw any conclusions concerning

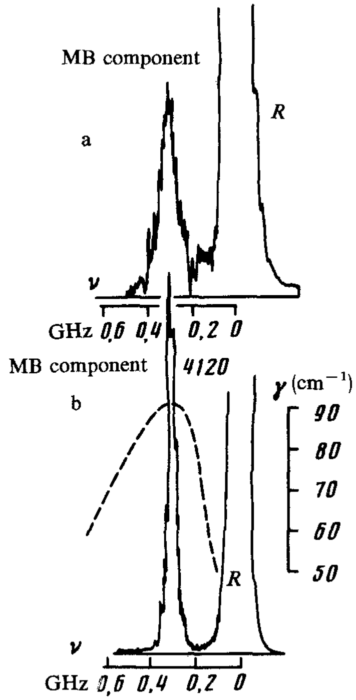

FIG. 2. a, b—The same as in Figs. 1a and 1b, but the scattering angle is smaller and the field is $E/E_c = 1.6$.

the shape and width of the sound amplification band, without a spectral decomposition of light, since the intensity of the new spectral lines is comparable to the intensity of the MB component.

A reduction in the width of the focused laser beam strengthens the effects we have considered here.

We wish to thank I. L. Fabelinskiĭ for a useful discussion.

[1)]The critical electric field E_c is that field at which the drift velocity of electrons is equal to the velocity of sound, v.
[2)]The width and shape of the Mandel'shtam-Brillouin component was discussed by Gal'perin *et al.*[3]
[3)]This value is apparently even smaller in a crystal.

[1]T. S. Velichkina, A. M. D'yakonov, O. I. Vasil'eva, *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **37**, 571 (1983) [JETP Lett. **37**, 571 (1983)].
[2]D. L. White, J. Appl. Phys. **33**, 2547 (1962).
[3]Yu. M. Gal'perin, V. L. Gurevich, and É. A. Rzaev, Fiz. Tverd. Tela **25**, 2386 (1983) [Sov. Phys. Solid State **25**, 1369 (1983)].

Translated by S. J. Amoretty