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MANDEL'SHTAM-BRILLOUIN SCATTERING IN CALCITE

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Mandel'shtam-Brillouin scattering along the principal crystallographic directions in single crystal calcite is investigated in detail. It is found that, in accord with theoretical predictions,[1] double refraction leads to the splitting of the Mandel'shtam-Brillouin components.

It has been shown by Vladimirskiĭ[1] that each Mandel'shtam-Brillouin (MB) component is a crystal with double refraction is split in the general case into four lines, depending on the polarization of the incident and scattered light.

A light wave propagating in similar crystals is a superposition of two waves, mutually perpendicularly polarized, with different wave vectors. In MB scattering, two values of the wave vector $\mathbf{k}_1$ and $\mathbf{k}_2$ of the incident light and the two values of the vector $\mathbf{k}_1'$ and $\mathbf{k}_2'$ of the light scattered at a given angle determine the four different wave vectors of the phonons, q_{ij}, which satisfy the scattering condition

$$\mathbf{q}_{ij} = \mathbf{k}_i' - \mathbf{k}_j \quad (i, j = 1, 2). \tag{1}$$

Changing only the polarization of the incident and scattered beams, we can observe the scattering by any of these four phonons. Here the shifts $\Delta\nu$ of the MB components, will be different in the general case, as follows from the condition

$$\Delta\nu = v\mathbf{q}, \tag{2}$$

where v is the sound velocity in the crystal in the direction of the vector q.

For scattering at 90°, we have

$$|\mathbf{q}_{ij}| = \lambda^{-1}\sqrt{n_i^2 + n_j'^2} \quad (i, j = 1, 2). \tag{3}$$

Here n_i and n_j' are the indices of refraction of the incident and scattered light, λ the wavelength of the incident radiation. The wave vectors of the phonons corresponding to this case are shown in Fig. 1.

The observation of MB scattering in calcite and other doubly refracting crystals has not, up to the present time, revealed any splitting, because of the small anisotropy of the index of refraction. A very suitable object for the observation of the described effect is calcite, the double refraction in which is sufficiently large. Up to the present time, only Raman scattering has been sufficiently discussed for calcite, and no published results on MB scattering are known to us. The object of the present work was the study of MB scattering in single crystal calcite.

Natural single crystal calcite $CaCO_3$ was used in the research.1) The specimens were cut in the shape of parallelepipeds with edges perpendicular to the threefold symmetry axis C_3 (the z axis), the twofold symmetry axis C_2 (the x axis), and the y axis, respectively (x ⊥ y ⊥ z). All the experiments were carried out at room temperature.

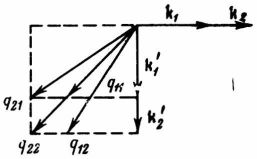

FIG. 1

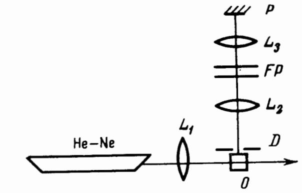

FIG. 2

FIG. 1. Diagram of Mandel'shtam-Brillouin scattering.

FIG. 2. Apparatus for the observation of the spectral composition of scattered light; L_1, L_2, L_3 are lenses, O—specimen, D—diaphragm, FP—fabri-Perot Interferometer, P—photographic film.

The research was conducted on two identical sets of apparatus, shown in Fig. 2. The schemes of similar setups for the study of the spectral composition of the scattered light have been described in sufficient detail, for example, by Fabelinskiĭ in[2]. As a light source, we used a He-Ne laser with λ = 6328 Å. The light scattered at an angle of 90° was analyzed by a Fabry-Perot interferometer. The interferograms (Fig. 3a) were recorded on A660 film. Spatial separation of MB components of different polarization was achieved by a Wollaston prism.

The interferograms were analyzed on a microphotometer with photographic and chart recording (Fig. 3b). The reduction of the microphotograms was with an IZA-2 comparator. The relative error of measurement of the distance with account of scatter of the data did not

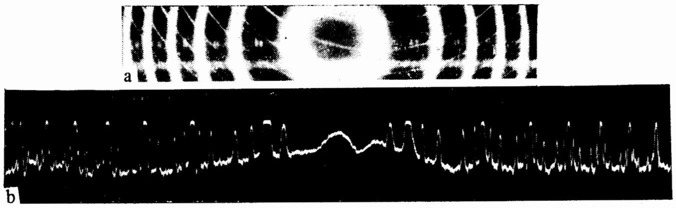

FIG. 3a. Interferogram of scattered light; the incident light is directed along the z axis and is polarized along the y axis; the scattered light is observed along the y axis. The MB components in the upper light filament were polarized along the x axis and in the lower along the z. Fig. 3b. Microphotograph of the spectrum.

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1) The authors express their gratitude to their fellowworker of the Institute of Crystallography, V. A. Shamburov, for kindly providing the single crystal calcite.

	Direction of incident light	Direction of scattered light	Direction of polarization of scattered light	Direction of polarization of incident light	Shift of MB components (in cm^{-1}) computed from elastic constants	Shift of MB components (in cm^{-1}) obtained experimentally
I	y	z	x	x	0.415 0.592 0.740	0,416 ± 0,004 — 0,750 ± 0,008
II	y	z	z	x	0,381 0.546 0.701	— — 0.723 ± 0,008
III	$-y$	z	x	x	0.342 0.376 0,902	0.340 ± 0,004 0,369 ± 0,004 0,903 ± 0.009
IV	$-y$	z	z	x	0.319 0,367 0.843	— 0.364 ± 0.004 —
V	x	y	z	z	0,326 0.626 0,831	— — 0,822 ± 0.008
VI	x	y	z	x	0.346 0.541 0,872	0,323 ± 0,003 0,531 ± 0,005 —
VII	z	x	y	y	0.390 0,455 0.845	— 0.445 ± 0,005 0,845 ± 0.009
VIII	z	x	y	z	0.359 0.438 0,789	— 0,421 ± 0.004 —

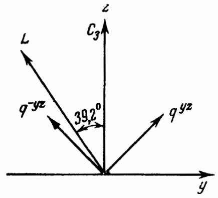

FIG. 4. Diagram of light scattering in the plane of symmetry of the crystal: L—direction of the acoustical axis, q^{yz}—direction of the phonon in the case in which the light is incident along the y axis and is observed along the z; q^{-yz}—direction of the phonon in the case in which the light is incident along −y and is observed along z.

exceed 1% in order of magnitude. The divergence of the data averaged over all orders, obtained on the different apparatus, was no more than ±0.5%. The reproducibility on each apparatus was within ±0.2%.

Experiments were carried out in which the incident light was directed along each of the three axes x, y, and z. In each chosen direction of the incident light, the scattered light was observed along the two other axes. The wave vector and the direction of propagation of the phonon from which scattering occurred were determined from (1).

The measured MB shifts are given in the table. For fixed directions of propagation and polarization of the incident and scattered beams of light, six MB components should be observed, which correspond to scattering by three phonons with the same q but with different polarization of the oscillations. However, in most cases, one can record only four of these or two because of the weak intensity of the others. In the table, the absence of similar components is indicated by a line.

It is seen from the results that a change in the polarization of one or both beams of light leads to a shift in the MB components. In the table, one should compare the MB components, which correspond in the sense of the polarization of the phonons, in the lines I and II, VII and VIII, III and IV. In the first two cases, the value of the splitting amounts to about 5% of the same MB displacements, thus exceeding the error of the experiment, and in the last case, it lies within the experimental limits of error. This same effect is seen directly in the photographs of Fig. 3a.

For comparison, values of the MB displacements computed from the elastic constants of calcite are given in the table. These constants are taken from the work of Dandekar.[3] The calculation was made by the usual method (see, for example,[4]), i.e., the eigenvalues of the tensor $\Gamma_{ik} = c_{ijkl}q_j q_l$ are found, where c_{ijkl} is the tensor of the elastic constants, and q_m are the components of the vector q. The solution of the cubic equations $|\Gamma_{ik} - \rho v^2| = 0$ (ρ is the density, v the velocity of the ultrasonic waves in the crystal) were carried out on the Ural-2 computer. In most cases, the divergence of the computed values from the experimental does not exceed 3%, with the exception of the upper line in VI.

In the general case, there can be ten acoustical axes in a trigonal crystal, i.e., such directions along which purely longitudinal and transverse waves propagate, and the transverse velocities are identical. From symmetry considerations, it follows that one of the axes is parallel to C_3 and the others lie in planes of symmetry. General equations were obtained by Khatkevich[5] for finding the acoustical axes. For calcite (space group D^6_{3d}) they have the form

$$\sin\theta = 0, \quad \mathrm{tg}^3\,\theta + 0.499\,\mathrm{tg}^2\theta - 0.458\,\mathrm{tg}\,\theta - 0.502 = 0,$$

where θ is the angle between the acoustical axis and C_3 in the plane of symmetry. The first equation defines the acoustical axis parallel to C_3. The second equation has one real root $\tan\theta = 0.8156$, i.e., $\theta = 39.2°$, which defines the acoustical axis that does not coincide with the crystallographic axes. Thus, there are four acoustical axes in calcite. One is parallel to C_3 and the other three lie in planes of symmetry and make an angle of 39.2° with C_3.

In the study of MB scattering in the zy plane, an important difference is observed in the location of the components in cases when the incident light is directed along y and the scattered along z (y, z) or, keeping the direction of the scattered light fixed, we direct the incident light along −y (−y, z). It is seen from Fig. 4 that different orientations of q relative to the acoustical axes correspond to these two positions. In the case (−y, z), the direction of q is close to the direction of the acoustical axis. Therefore, the difference in the shifts of the MB components, corresponding to the transverse waves, is insignificant. In the case (y, z), the difference in the position of the MB components corresponding to the quasi-transverse waves is of the usual order (see lines I and II in the table).

Thus, the investigations that have been carried out show that the observed positions of the MB components in calcite are in excellent agreement with those computed from the elastic constants. It is found that the MB components change their position as a function of the direction of polarization of the incident and scattered light. The change in the position in the two cases exceeds the experimental error and in the third lies within the error limits. The calculation of the position of the acoustical axes in calcite has been carried out. The experiments confirm qualitatively the presence of the acoustical axes in the planes of symmetry.

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