SPONTANEOUS PARAMETRIC LIGHT SCATTERING IN $Gd_2(MoO_4)_3$

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Results are presented on a gadolinium molybdate crystal examined in spontaneous parametric scattering at polaritons; the frequency-angle spectra indicate the dispersion of the ordinary refractive index in the range 350-5000 cm^{-1}. The frequencies of the type B_1 lattice modes have been determined. Two-particle state bands are observed, which determine the absorption of the crystal in the region around 1800 cm^{-1}.

Results are presented on a gadolinium molybdate GMO crystal examined by spontaneous parametric scattering at polaritons SPS. SPS spectroscopy enables one to examine the lattice dynamics and the optical parameters of the scattering medium in the infrared region. SPS spectra are reported for GMO over the range 280-5000 cm^{-1}, with measurement of the natural frequencies and determination of the dispersion of the ordinary refractive index in the range 350-5000 cm^{-1}.

A GMO crystal is biaxial at room temperature; it has an orthorhombic unit cell and point symmetry mm2. However, the longitudinal spontaneous birefringent is only $\Delta n_{xy} \sim 4 \cdot 10^{-4}$ [1], so there is no need in a good approximation to draw a distinction between n_x and n_y, so the crystal can be considered as uniaxial, i.e., $n_x \approx n_y = n^0$, $n_z = n^e$.

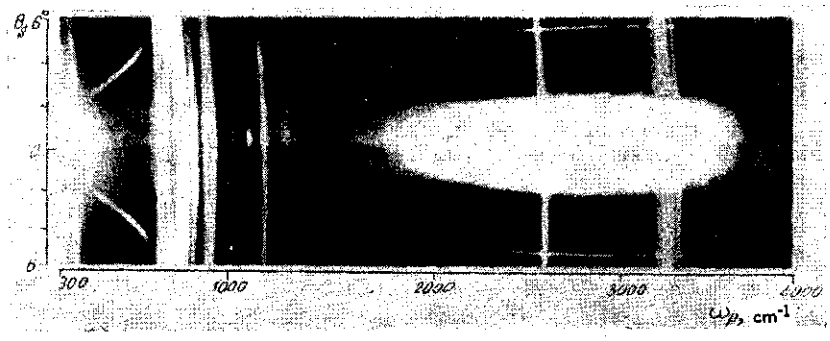

Fig. 1. Frequency-angle SPS spectrum of a GMO crystal, where θ_s is the scattering angle outside the crystal and ω_p is the polariton frequency.

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Frequency-angle SPS spectra were recorded for gadolinium molybdate with a standard experimental system with photographic recording [2]. Figure 1 shows the general form of the spectrum with the following scattering geometry: the triangle of wave vectors $\mathbf{k}_L = \mathbf{k}_s + \mathbf{k}_p$ lies in the XY plane ($\mathbf{k}_L$, $\mathbf{k}_s$, $\mathbf{k}_p$ are the wave vectors for the pumping wave, signal, and polariton), with the pumping wave directed along the X axis and representing the ordinary wave ($\mathbf{k}_L \| X$, $\mathbf{e}_L \| Y$), while the signal wave was the extraordinary wave ($\mathbf{e}_s \| Z$). Here there was scattering at ordinary polaritons.

The natural lattice frequencies appear in this spectrum in the region 280-1000 cm^{-1}; the transmission region begins at 1300 cm^{-1}. The lines in that part of the spectrum are due to crystal luminescence, and the corresponding wavelengths lie in the range 0.50-0.62 μm. To distinguish the natural-frequency lines from the luminescent lines we compared the spectra obtained by pumping at different wavelengths: λ_L = 457.9, 476.5, 488.0, and 514.5 nm. The luminescence lines do not alter in position, whereas the natural-vibration lines move along with the pumping in such a way that the frequency shift $\omega_p = \omega_L - \omega_s$ remains constant. The pumping source was an argon laser.

Table 1

SPS ω, cm^{-1}	RS [3] ω, cm^{-1}
282	284
302	306
330	327
348	345
407	407
718	727
749	748
816	821
828	828
849	848
881	882
931	930
960	965

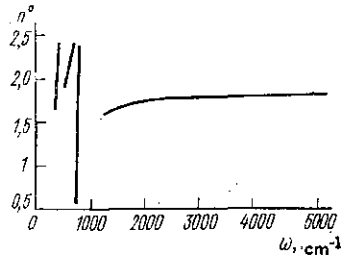

Fig. 2. Dispersion of the ordinary refractive index n^0 in the range 350-5000 cm^{-1}.

The SPS spectra allowed us to measure natural lattice frequencies in the range 280-1000 cm^{-1} with an error of ±10 cm^{-1}. Table 1 gives the results. This gives frequencies obtained by Raman scattering [3] for type B_1 modes. Table 1 shows that the frequencies measured by the various methods are in good agreement. Also, the wavelengths of the luminescence lines were determined. The strongest lines had wavelengths of 540; 543,3; 549; 564,6; 567; 570; 576,8; 589; 594; 599,6; 599,6; 604; 607; 611 nm.

The integral luminescence intensity throughout the spectrum is substantially independent on the pumping wavelength, as are the relative intensities of the individual lines. This luminescence cannot be related to the trivalent gadolinium ion, since all the Gd^{3+} lines lie in the ultraviolet region [4], and it is due to impurities in the crystal, e.g., other rare-earth ions or divalent gadolinium. On the other hand, the luminescence intensity is very much dependent on the particular crystal (although the spectrum is the same for a given pumping), and the intensity may vary from specimen to specimen by a factor 5, which is clearly governed by the impurity concentration.

One of the features of these spectra is that there are two broad lines at 1756 and 1800 cm^{-1}, which interact with the SPS curve and cause a deviation for the monotonic form, and therefore they have nonzero dipole moments. The SPS method is highly sensitive to the dipole moments of the vibrations; it provides

Table 2

λ, Å	n^o	n^e
4078	1,885	1,946
5780	1,847	1,901
6123	1,841	1,894
5461	1,852	1,907

Table 3

ω, cm^{-1}	n^o
1356	1,607
1515	1,659
1665	1,691
1849	1,707
2066	1,739
2298	1,752
2697	1,774
2825	1,777
3375	1,790
4786	1,814
4933	1,814

reliable recording of vibrations of dipole moment about 10^{-4} [5]. The highest-frequency first-order vibration [3] has a frequency of 965 cm^{-1}. Therefore, the 1756 and 1800 cm^{-1} lines are probably second-order lines formed for example by the pairs $1756 \approx 931+828$ cm^{-1}, $1800 \approx 960+849$ cm^{-1}. The interaction between the electromagnetic wave and the second-order vibrations increases the damping at these frequencies and thus results in an additional absorption band, which was observed in the range 5.5-6 μm [6].

The infrared refractive index may be measured (including in the region of the lattice modes) from the SPS frequency-angle spectra if one knows the refractive indices for the pumping wave and the scattered wave, whose frequencies lie in the visible region [7]. Table 2 gives the corresponding refractive indices as measured by the prism method with a GS-30 goniometer with an error of $\pm 5 \cdot 10^{-4}$. The SPS spectra were used to determine the refractive index for the ordinary wave n^0 in the range 350-5000 cm^{-1}. Figure 2 and Table 3 gives the dispersion of n^0 in this range.

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REFERENCES

1. A. W. Smith and G. Burns, Phys. Lett., vol. 28A, p. 501, 1968.
2. O. A. Aktsipetrov, G. M. Georgiev, A. G. Mikhailovskii, and A. N. Penin, Fiz. Tverd. Tela, vol. 18, p. 665, 1976.
3. F. G. Ullman, B. T. Holden, and B. N. Ganguly, Phys. Rev., vol. B8, p. 2991, 1973.
4. Laser Handbook [in Russian], Moscow, 1978.
5. K. D. Kneipp, G. E. Ponat, V. L. Strizhevskii, and Yu. N. Yashkir, Pis'ma ZhETF, vol. 18, p. 89, 1973.
6. K. J. Nassau, Phys. Chem. Sol., vol. 26, no. 12, 1965.
7. G. M. Georgiev, Phys. Stat. Sol., vol. B86, p. 431, 1978.

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