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ACTIVE SPECTROSCOPY OF RAMAN SCATTERING OF LIGHT WITH THE AID OF A QUASICONTIN-UOUSLY TUNABLE PARAMETRIC GENERATOR

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1. The purpose of this article is to describe the principles and certain results of an experimental realization of a method of active Raman-scattering (RS) spectroscopy with the aid of a quasicontinuously tunable parametric light generator (PLG).

The use of a pair of intense oscillations with tunable frequencies (ν_0 and $\nu_{1,2}$ – see Fig. 1) makes it possible to excite a selected vibrational transition (optical mode) of frequency Ω_0. By observing the scattering of a probing light beam (frequency ν_{pr} – see Fig. 1) from this mode one can greatly improve the signal/noise ratio at the output of the RS spectrometer and to measure the scattering line shape (especially in the study of broad lines) and the scattering cross section, with an accuracy exceeding the accuracy attained in the investigation of spontaneous Raman scattering. Strong mode excitation uncovered new possibilities in the investigation of anharmonic effects (combination modes, etc.). With the aid of a quasicontinuous PLG it is convenient also to carry out measurements in accordance with the inverted-scattering scheme – a promising scheme which so far has not found wide use only because there are no suitable tunable radiation sources [10].

2. The spectroscopic information obtained with stimulated RS is scanty. The principal reason for this is the strong competition of the lines, the broadening of the spectrum due to nonlinear mechanisms, etc. It is therefore of interest to search for a method combining the extensive spectroscopic capabiltiies of the spontaneous Raman scattering with the advantages of the method of stimulated Raman scattering, such as obtaining high-intensity scattered light, coherent excitation of optical phonons in a considerable volume, etc.

This problem was discussed in a number of papers (see, e.g., [1 - 6]). A procedure for scattering a probing beam by coherent optical phonons excited during the course of the stimulated Raman scattering was proposed and realized in [1, 7, 8] (see also [9]).

The spectroscopic possibilities are greater when the phonons are excited by a biharmonic light signal below the threshold of stimulated Raman scattering. Specially selected line doublets were previously used for this purpose [2,
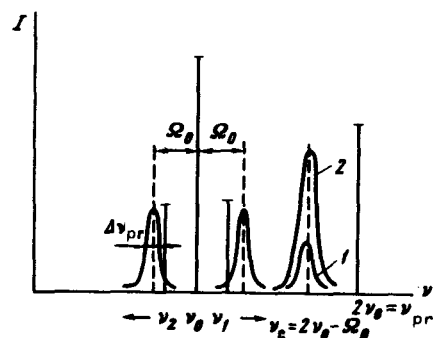

Fig. 1. Diagram explaining the principle of active scattered-light spectroscopy with the aid of a parametric generator. Here ν_0 and $2\nu_0 = \nu_{pr}$ are the frequencies of the laser and of its second harmonic; ν_1 and ν_2 ($\nu_1 + \nu_2 = 2\nu_0$) are the smoothly tunable frequencies of the PLG; Ω_0 is the Raman shift. Curve 1 - Stokes signal of probing scattering at $|\nu_0 - \nu_{1,2}| \neq \Omega_0$; curve 2 - signal at $|\nu_0 - \nu_{1,2}| \approx \Omega_0$.
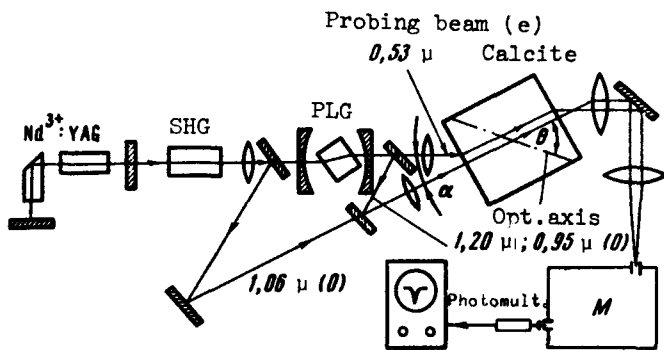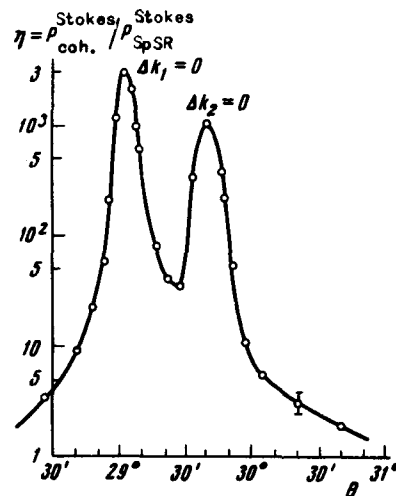
Fig. 2

Fig. 3

Fig. 2. Block diagram of experimental setup. The multi-frequency generator for the active-spectroscopy scheme is a quasi-cw laser using aluminum-yttrium garnet, its second harmonic, and a parametric laser using a lithium-niobate crystal, excited by this second harmonic. M - triple monochromator, SHG - second-harmonic generator.

Fig. 3. Experimental plot of the normalized intensity of Stokes scattering of the probing beam at the mode A_{1g} ($\Omega_0 = 1086$ cm^{-1}) in a calcite crystal at T = 300°K on the angle θ between the optical axes of the crystal and the pump beams.

3, 5, 6]. It would be ideal, of course, to have a smoothly tunable multi-frequency generator operating in the cw or quasi-cw regime.

3. These are precisely the properties possessed by the multi-frequency radiation source employed by us. It consists of a quasi-cw laser based on YAG with Nd^{3+} ($\lambda_0 = 1064$ nm, pulse power $P_0 \simeq 1$ MW, frequency up to 100 Hz), a second-harmonic generator (SHG) using an LiNbO$_3$ crystal ($P \simeq 100$ kW), which excites a two-resonator PLG using a LiNbO$_3$ crystal ($P_{1,2} \simeq 2$ kW); the tuning region is 0.9 - 1.25 μ. The second harmonic of the laser is used also as the probing signal.

When, for example, the frequency detuning $\nu_1 - \nu_0$ ($\nu_0 - \nu_2$) is close to Ω_0, the chosen mode is excited; if at the same time the momentum conservation law is also satisfied

$$\Delta k_{1,2} = (k_o - k_{1,2}) - (k_{pr} - k_{s,pr}) = 0 \tag{1}$$

then the intensity of the scattering of the probing beam increases sharply (see [1]). If the widths of the lines that build up the oscillations are smaller than the width of the spontaneous Raman-scattering line, it is possible to measure the shape of the spontaneous line directly. In the scheme in which the frequencies ν_1 and ν_0 are used, at $I_0 \gg I_1$, the buildup of the molecular oscillations is accompanied by a resonant increase of the intensity of the wave ν_1, and it becomes possible to measure simultaneously the parameters of the RS line also in accordance with the inverted-scattering scheme.

4. Figure 2 shows the experimental setup. The measurements were performed at the line $\Omega_0 = 1086$ cm^{-1} of type A_{1g} in calcite. The probing radiation and the pump radiation were introduced into the crystal at an angle corresponding to

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satisfaction of the condition (1). (The probing wave in the calcite is extraordinary, the pump waves are ordinary; $\alpha = 8°10'$, $\theta \simeq 29°$.)

Figure 3 shows the experimental dependence of the scattering power normalized to the spontaneous Raman scattering power, $\eta = P_{coh}/P_{spont}$, on the angle θ between the optical axis of the crystal and the pump beam; this dependence was compared with the calculated one:

$$\eta_{theor} = \frac{9\pi^3}{4}(\sigma N z)\frac{c^4}{\omega_c^6}I_0/n_c^2 n_0^2(\hbar\Delta\omega_{sp})^2\left[I_1\left(\frac{\Delta\omega_{sp}}{\Delta\omega_1}\right)\frac{\sin^2\frac{\Delta k_1(\theta)z}{2}}{(\Delta k_1(\theta)z/2)^2} + \right.$$

$$\left. + I_2\left(\frac{\Delta\omega_{sp}}{\Delta\omega_2}\right)\frac{\sin^2(\Delta k_2(\theta)z/2)}{(\Delta k_2(\theta)z/2)^2}\right]\frac{4\pi}{\delta\Omega/n_c^2}\frac{S_0}{S_{pr}}; \qquad (2)$$

Here σ is the scattering cross section, N the molecular-number density, z the effective interaction length, I_0 and $I_{1,2}$ the wave intensities, $\Delta\omega_{1,2}$ the widths of the PLG lines, $\Delta\omega_{sp}$ the width of the spontaneous Raman scattering line, $\delta\Omega$ the solid angle within which the spontaneous Raman scattering is registered, and S_0 and S_{pr} – the areas of the beams.

Substance RS line (Ω_0)	Pump radiation		Probe & scattered radiation		Coherent length
	λ_{H1}, nm	λ_{H2}, nm	λ_{pr}, nm	$\lambda_s,\lambda_{antis}$, nm	$L_{coh} = (\Delta k)^{-1}$ cm
Calcite	1048 ordin.	1080.3 ord.	1064.0 ordin.	1097 ordin.	0.23
284 cm^{-1}	1064.0 ordin.	1097 ord.	532.0 ordin.	540.2 ordin.	0.083
Quartz	1034.4 ordin.	1086.8 ord.	1064.0 ordin.	1115.2 ordin.	0.84
466 cm^{-1}	1034.4 extraord.	1086.8 extr.	1064.0 extraord.	1115.2 extraord.	0.52
Benzene	962.4	1064	532.0	561.6	0.0025
992 cm^{-1}	1010.6	1123.3	1064.0	962.4	0.064
Carbon disulfide	994.6	1064.0	532.0	551.2	0.002
656 cm^{-1}	1028	1102.5	1064	994.6	0.081
Hydrogen	868.0	1360.4	1064.0	736.3	$\geqslant 10$
4155 cm^{-1} ρ = 100 atm	1064.0	736.3	532.0	682.9	3
Methane 2915 cm^{-1}	921.2	1260.6	1064.0	812.2	$\geqslant 10$
ρ = 30 atm	812.2	1064.0	532.0	631.8	5.6

At $I_0 = 5.4 \times 10^8$ W/cm^2, $I_1 = 0.8 \times 10^5$ W/cm^2, $\Delta k_1 = 0$, $(\Delta\omega_{sp}/\Delta\omega_1) = 0.1$, $S_0/S_{pr} = 0.25$, $\delta\Omega = 0.20$ sr and z = 1.4 cm, formula (2) yields $\eta_{theor} = 2.7 \times 10^3$ (the values of σ and N were taken from [11]). The experimental value is $\eta_{exp} = 3 \times 10^3$. A special check was made on the nonlinearity of the dependence of the intensity of the probing scattering on the intensity of each of the three interacting waves.

Under our experimental conditions it is possible to observe coherent scattering on lines for which $\sigma/\Delta\omega_{sp}$ is smaller by 2 – 3 orders of magnitude than

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for the investigated calcite line. The sensitivity can be increased further by narrowing down the line and increasing the power of the PLG; this also increases the resolution.

5. The maximum effect is reached along the synchronism direction ($\Delta k = 0$). However, the requirements with respect to the accuracy with which the beams are set are in many cases not too stringent. The table lists the values of the coherent scattering length $L_{coh} = (\Delta k)^{-1}$ far from the exact synchronism for different media and different frequencies of the pump and of the probing signal. It is seen that in gases $L_{coh} \gtrsim 10$ cm and in general there are no problems connected with the exact synchronism. In strongly dispersive media, $L_{coh} \sim 1$ cm is reached by having the pump and probing-signal frequencies close. Of course, the possibility of using exact synchronism remains in all cases.

The choice of a green probing beam with wavelength $\lambda_{pr} = 532$ nm is convenient because it makes it possible to use the recording apparatus of the commercial Raman spectrograph with the argon laser serving for the excitation of the RS spectra.

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FAST-NEUTRON-COMPENSATED n-GERMANIUM AS A MODEL OF AMORPHOUS SEMICONDUCTORS

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As is well known, transport phenomena in amorphous (vitreous) semiconductors differ significantly from those in crystals: in particular, they exhibit an anomalously small "Hall" mobility, a nonlinear S-shaped current-voltage characteristic, and a conductivity activation energy that decreases continuously with decreasing temperature [1]. According to recently developed concepts, the physical properties of amorphous semiconductors are determined by the presence in them of random potential fluctuations whose scale is comparable with the width of the forbidden band [2, 3]. A similar potential relief exists, as is well known, also in crystalline semiconductors if they are strongly compensated by randomly distributed charged impurities. By varying the degree of doping and compensation it is possible, as shown in [4], to produce a controllable model of amorphous semiconductors, for in this case changes take place both in the amplitude and dimensions of the regions of the potential fluctuations and in the position of the Fermi level.

We note, however, that if fluctuations are produced as a result of a Poisson distribution of point defects and impurities, a rigid connection is