Stationary stimulated Mandel'shtam–Brillouin scattering in paramagnetic crystals under acoustic paramagnetic resonance conditions

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Stationary stimulated Mandel'shtam–Brillouin scattering is investigated in paramagnetic crystals under acoustic paramagnetic resonance conditions in an optical cavity, the crystal being an acoustic resonator. Generation of a single Stokes component is considered. The threshold condition for stationary stimulated Mandel'shtam–Brillouin scattering under acoustic paramagnetic resonance conditions is obtained. It is shown that the threshold for stimulated Mandel'shtam–Brillouin scattering is lowered in the case of an inverted population of the energy levels. A method for investigating acoustic paramagnetic resonance on the basis of stimulated Mandel'shtam–Brillouin scattering is proposed.

Theoretical investigation of stimulated Mandel'shtam-Brillouin scattering in piezosemiconductors [1,2] has shown that under conditions of acoustic instability (acoustic instability results when the drift velocity of the electrons exceeds the sound velocity in the piezosemiconductor, a situation which is achieved by the application of a constant electric field of sufficient magnitude), a significant decrease takes place in the threshold for stimulated Mandel'shtam-Brillouin scattering. In this connection, it is of interest to consider another source of acoustic instability, namely spin-phonon interaction. For this purpose, we consider stationary stimulated Mandel'shtam-Brillouin scattering in paramagnetic crystals under conditions of acoustic paramagnetic resonance (APR) in an optical cavity, assuming that the crystal represents an acoustic resonator. We limit ourselves to consideration of the generation of a single Stokes component (stationary stimulated Mandel'shtam-Brillouin scattering in an optical cavity filled to the borders with homogeneous matter with generation of an arbitrary number of Stokes components has been considered by Lugovoĭ and Strel'tsov. [3]).

1. Initial conditions: a) an open optical cavity is excited by an external laser beam of specified intensity, incident along the axis of the cavity; b) the axis of symmetry of a uniaxial paramagnetic crystal (the c axis) is parallel to the axis of the optical cavity; c) it is assumed that selection of axial modes is accomplished in the cavity, so that only characteristic oscillations with the smallest transverse numbers, differing only in the values of the axial index, possess sufficient Q; d) the reflection coefficients of the mirrors for the first Stokes component are close to unity, and those for the second are sufficiently small (use of interference mirrors), or an absorber is introduced in the cavity at the frequency of the second Stokes component. Then the second Stokes component will not be excited in the cavity; e) the mirrors of the resonators are assumed to be plane; f) magnetically dilute crystals are considered, so that the spin-spin interactions can be neglected.

With our initial conditions the electric field $\mathbf{E}(\mathbf{r}, t)$ in the cavity has the form [4]:[1)]

$$\mathbf{E}(\mathbf{r},t)=\mathscr{E}_0(t)\mathbf{E}_0(\mathbf{r}_\perp)\exp[i(k_0z-\Omega_0t)] + \sum_{s=1}^{n}\mathscr{E}_{-1s}(t)\mathbf{E}_{-1s}(\mathbf{r}_\perp)\exp[-i(k_{-1s}z+\omega_{-1s}t)]+\text{c.c.}, \quad (1)$$

where $\mathbf{E}_0(\mathbf{r}_\perp)\exp[ik_0z]$ is the axial mode, the eigenfrequency of which is close to the frequency of the external exciting beam; $\mathbf{E}_{-1s}(\mathbf{r}_\perp)\exp[-ik_{-1s}z]$ are the axial modes with eigenfrequencies close to the first Stokes frequency ω_{-1}. Here the spectrum of the function $\mathscr{E}_0(t)\exp(-i\Omega_0t)$ is concentrated near the frequency Ω_0, which we assume to be equal to the frequency of the external exciting beam; the spectra of the functions $\mathscr{E}_{-1s}(t)\exp(-i\omega_{-1s}t)$ are concentrated in the vicinity of ω_{-1}. The z axis is directed along the axis of the cavity.

The acoustic field in the crystal will be sought in the form

$$\Phi(\mathbf{r},t)=\sum_{l}\mathfrak{F}_l(t)\mathbf{E}_0(\mathbf{r}_\perp)\mathbf{E}_{-1l}(\mathbf{r}_\perp)\exp[i(k_lz-\omega_lt)]+\text{c.c.} \quad (2)$$

This notation for $\Phi(\mathbf{r}, t)$ will be made clear by the following presentation.

We obtain the equations which describe stimulated Mandel'shtam-Brillouin scattering in paramagnets under APR conditions from the equation of motion of an elastic medium

$$\rho_0\frac{\partial^2\Phi_i}{\partial t^2}=\frac{\partial T_{ji}}{\partial x_j}, \quad (3)$$

$$T_{ij}=c_{ijkl}U_{kl}+\gamma_{ijkl}E_kE_l+R_{ij}, \quad (3a)$$

and the Maxwell equation

$$\text{rot}\,\text{rot}\,\mathbf{E}=-\mu_0\frac{\partial^2\mathbf{D}}{\partial t^2}-\mu_0\frac{\partial\mathbf{J}_{ex}}{\partial t}, \quad (4)$$

$$D_i=(\varepsilon_{ij}-\gamma_{ijkl}U_{kl})E_j, \quad (4a)$$

where T_{ij} are the elements of the stress tensor, R_{ij} are the components of the magnetoelastic tensor, γ_{ijkl} and ϵ_{ij} are the electrostrictive and dielectric constants, c_{ijkl} are the elements of the elasticity tensor, $U_{kl} = \partial\Phi_k/\partial x$ are the elements of the deformation tensor, Φ_k are the components of the elastic displacement vector ($\mathbf{\Phi}$ is directed along the z axis, $\mathbf{\Phi}(0, 0, \Phi)$), E_k and D_i are the components of the electric field and the electric displacement, $\mathbf{J}_{ex}$ is the external current due to the external beam, ρ_0 is the density of the crystal, and μ_0 the magnetic permeability of the vacuum.

If we introduce the notation $R_{zz} = R$ and $U_{zz} = U$, then we can determine the necessary value of R from the relation

$$R=N\,\text{Sp}\left[\frac{\partial\mathscr{H}_c}{\partial U}\sigma\right],$$

where N is the concentration of paramagnetic ions, $\mathscr{H}_c$ is the Hamiltonian of spin-phonon interaction, and σ the density matrix. Using the stationary solution for the populations of the levels and the elements of the density matrix,[5] we can represent the expression for R in the form

$$R_l=-\hbar\varkappa^2T_2n_0\int_{-\infty}\frac{[(\Delta\omega)'T_2+i]\overline{U}_lg[(\Delta\omega)'']}{1+[(\Delta\omega)'T_2]^2+4\varkappa^2T_1T_2|\overline{U}_l|^2}d(\Delta\omega)''\exp[i(k_lz-\omega_lt)]+\text{c.c.} \tag{5}$$

$$\hbar\varkappa=\langle a|\mathscr{H}_c|b\rangle=\langle b|\mathscr{H}_c|a\rangle,$$

where a and b enumerate the energy levels of interest to us; n_0 is the initial difference in the populations of levels a and b (the lower and upper levels); $(\Delta\omega)' = \omega' - \omega_l$; $(\Delta\omega)'' = \omega_{ba} - \omega' = \omega_0 - \omega'$; T_1 and T_2 are the spin-lattice and spin-spin relaxation times for the pair of levels a and b; $U_l = \overline{U}_l \exp i(k_l z - \omega_l t)$ + complex conjugate; $g(\Delta\omega)$ are functions of the shape of the APR line.

Substituting the expressions (1), (2), (3a), (4a), (5) in Eqs. (3) and (4), we obtain in the approximation of slowly varying amplitudes

$$\dot{\mathscr{E}}_{-1s}=-\chi_sd_s\mathscr{E}_0\mathfrak{F}_s^*-\lambda_{-1s}\mathscr{E}_{-1s}, \tag{6a}$$

$$\dot{\mathscr{E}}_0=\chi_0\sum_s d_{s'}\mathscr{E}_{-1s'}\mathfrak{F}_{s'}-\lambda_0\mathscr{E}_0+F, \tag{6b}$$

$$\dot{\mathfrak{F}}_l=-\eta\mathscr{E}_0\mathscr{E}_{-1l}^*-(h_l+i\delta_l)\mathfrak{F}_l;$$

$$\eta=\gamma_1/2\rho_0v_0;\quad h_l=\tfrac{1}{2}v_0[\alpha_0^l+\alpha_{\omega_l}]; \tag{6c}$$

$$\delta_l=\omega_l\beta_l;\quad \chi_0=\gamma_2\omega_0/2N_0;\quad \chi_s=\gamma_2\omega_{-1s}/2N_{-1s};\quad \gamma_1=\gamma_{3311},$$

where $\gamma_2 = \gamma_{1133}$; v_0 is the sound velocity in the crystal without account of spin-phonon interaction; α_0^l is the phenomenological sound-absorption coefficient;

$$\alpha_{\omega_l}=\frac{\pi\hbar\varkappa^2\omega_ln_0g(\omega_0-\omega_l)}{\rho_0v_0^3[1+4\varkappa^2k_l^2T_1T_2|\mathfrak{F}_l|^2]^{1/2}} \tag{7}$$

is the sound absorption coefficient due to spin-phonon interaction;

$$\beta_l=-\frac{\pi\hbar\varkappa^2n_0T_2(\omega_0-\omega_l)g(\omega_0-\omega_l)}{2\rho_0v_0^2[1+4\varkappa^2k_l^2T_1T_2|\mathfrak{F}_l|^2]^{1/2}}$$

is a dimensionless quantity which characterizes the change in the sound velocity due to the spin-phonon interaction (the expressions for α_{ω_l} and β_l were obtained for large inhomogeneous broadening). Further,

$$N_0=\varepsilon_\parallel\int|E_0|^2dV,\quad N_{-1s}=\varepsilon_\parallel\int|E_{-1s}|^2dV$$

where the integration is carried out over the optical cavity and

$$d_s=k_s\int|(\mathbf{E}_0\mathbf{E}_{-1s}^*)|^2dV,$$

where the integral is only over the crystal. Finally,

$$\lambda_0(\lambda_{-1s})=\tfrac{1}{2}c\alpha_0^c(\alpha_{-1s}),$$

where c is the velocity of light; α_0^c, α_{-1l} are the phenomenological absorption coefficients of the optical modes; F is a term due to the external beam; $\epsilon_\parallel = \epsilon_{11} = \epsilon_{22}$.

It is obvious that

$$N_0/d_s\approx N_{-1s}/d_s\sim\varepsilon_\parallel V_1/k_sV_2, \tag{8}$$

where V_1, V_2 are the volumes of the optical and acoustic resonators, respectively.

3. Since the difference in populations of the energy levels is inversely proportional to the temperature, the effect of APR on the stimulated Mandel'shtam-Brillouin scattering will be perceptible only at low (liquid-helium) temperatures. We can then assume that $\lambda_{-s}, \lambda_0 \gg h_l$. Consequently, in accord with Lugovoĭ,[4] we can consider the stationary solutions for Eqs. (6a) and (6b) of the set (6). We obtain the following from Eq. (6a), assuming the left side to be equal to zero:

$$-\mathscr{E}_{-1s}=\frac{\chi_sd_s}{\lambda_{-1s}}\mathscr{E}_0\mathfrak{F}_s^*. \tag{9}$$

We obtain an equation for $W_l \equiv |\mathscr{F}_l|^2$, using Eq. (6c) and the relation (9):

$$\dot{W}_l=-2h_lW_l+2\eta\frac{\chi_ld_l}{\lambda_{-1l}}|\mathscr{E}_0|^2W_l. \tag{10}$$

Now, setting the left sides of (6b) and (10) equal to zero, applying (9), and solving the set of algebraic equations, we get the desired stationary solutions. We consider the small- and large-signal approximations.

In the small-signal approximation, we can write the relation (7) as

$$\alpha_{\omega_l}\approx\alpha_{\omega_l}'=\frac{\pi\hbar\varkappa^2\omega_ln_0}{\rho_0v_0^3}g(\omega_0-\omega_l). \tag{11}$$

Then the first of the states of equilibrium is determined by the equations (we denote the solutions by a tilde over the corresponding quantities): $\tilde{\mathscr{E}}_0 = F/\lambda_0 \equiv Z_0$, $\tilde{W}_l = 0$. The stimulated Mandel'shtam-Brillouin scattering is absent in the corresponding oscillation regime. The remaining equilibrium positions have the form (for all $l \neq \xi$)

$$|\tilde{\mathscr{E}}_0|_\xi^2=\frac{h_\xi'\lambda_{-1\xi}}{\eta\chi_\xi d_\xi},\quad \tilde{W}_l=0,$$
$$\tilde{W}_\xi=\frac{\lambda_0\lambda_{-1\xi}}{\chi_0\chi_\xi d_\xi^2}\left\{\frac{|Z_0|}{|\tilde{\mathscr{E}}_0|_\xi}-1\right\}. \tag{12}$$

The threshold conditions for generation of stimulated Mandel'shtam-Brillouin scattering can be determined by following Lugovoĭ:[4]

$$|Z_0|^2\geqslant|Z_0|_{\text{thr}}^2=\frac{h_{\xi_0}'\lambda_{-1\xi_0}}{\eta\chi_{\xi_0}d_{\xi_0}},$$

where the index ξ_0 is attached to the modes for which the quantity $h'_\xi\lambda_{-1\xi}/\eta\chi_\xi d\xi$ is minimal. ($h'_\xi = \tfrac{1}{2}v_0(\alpha_0^\xi + \alpha\omega\xi')$). We write down this threshold condition for the Poynting flux S_0, taking the relation (8) into account:

$$S_0=\frac{|Z_0|^2}{c\mu_0}\geqslant C_{33}c\frac{\varepsilon_\parallel^2}{\gamma_1\gamma_2}\frac{\alpha_{\xi_0}'\alpha_{-1\xi_0}}{k_{\xi_0}k_{-1\xi_0}}\frac{V_1}{V_2}\left[\text{W/m}^2\right], \tag{13}$$

where $\epsilon = \epsilon_{11} = \epsilon_{22}$; $C_{33} = c_{3333}$; $\alpha'_{\xi 0} = \alpha_0^{\xi_0} + \alpha'_\beta$, $\beta = \omega_{\xi 0}$. Consequently, for the case of inversion of population of the energy levels considered (i.e., when n_0 and hence also α'_β are negative), we can obtain a decrease in the threshold of stimulated Mandel'shtam-Brillouin scattering. As is seen from Eq. (12), the small-signal approximation is valid for $\alpha_0^{\xi_0} + \alpha'_\beta > 0$ and $|Z_0|^2 \sim |Z_0|^2_{\text{thr}}$.

In the large-signal approximation (this means larger values of $|Z_0|^2$ in comparison with $|Z_0|^2_{\text{thr}}$) we have

$$\alpha_{\xi_0}=\alpha_0^{\xi_0}+\alpha_\beta\approx\alpha_0^{\xi_0}$$

and the position of equilibrium is determined by the relations

$$|\tilde{\mathscr{E}}_0|_{\xi_0}^2\approx\frac{v_0\alpha_0^{\xi_0}\lambda_{-1\xi_0}}{2\eta\chi_{\xi_0}d_{\xi_0}},\quad \tilde{W}_{\xi_0}\approx\frac{\lambda_0\lambda_{-1\xi_0}}{\chi_0\chi_{\xi_0}d_{\xi_0}^2}\frac{|Z_0|}{|\tilde{\mathscr{E}}_0|_{\xi_0}}. \tag{14}$$

Under the large-signal conditions, saturation of the transition between the energy levels of interest to us takes place and the spin-phonon interaction does not have an effect on the stimulated Mandel'shtam-Brillouin scattering (cf. (12) and (14)).

4. We estimate the stimulated Mandel'shtam-Brill-

ouin scattering threshold without account of the spin-phonon interaction at the temperature of liquid helium (4.2°K). For this purpose, we use the expression (13), where we must replace α'_{ξ_0} by $\alpha_0^{\xi_0}$. Using the following typical values of the quantities for solids: $C_{33} = 5 \times 10^{10}$ N/m², $\alpha_0^{\xi_0} = 10$ m⁻¹, $\alpha_{-1\xi_0} = 10$ m⁻¹, $k_{\xi_0} = 4\pi 10^6$ m⁻¹, $k_{-1\xi_0} = 2\pi 10^6$ m⁻¹, $\epsilon_{\parallel}^2/\gamma_1\gamma_2 \approx 1$, $c = 3 \times 10^8$ m/sec, we obtain $S_0 \geq \sim 1.9 \times 10^7 V_1V_2$ W/m². The intensity of the laser beam necessary for the creation of such a field in the cavity, according to Lugovoĭ and Strel'tsov,[3] for mirror reflection coefficients ~0.8 and an optical-resonator length ~2 cm is $S_0^{(L)} \geq \sim 1.9 \times 10^6 V_1/V_2$ W/m². Considering such a relation between the volumes of the resonators, when the threshold is minimal, i.e., assuming that $V_1 \approx V_2$, we have $S_0^{(L)} \leq \sim 1.9 \times 10^6$ W/m². Using a He-Ne laser operating in the continuous mode (power ≈ 80 mW), and focusing its beam (so that the diameter of the beam ≈0.02 cm), we can achieve an intensity of the beam $\sim 2.5 \times 10^6$ W/m². Consequently, using ordinary lasers operating in a continuous mode, the threshold condition for stimulated Mandel'shtam-Brillouin scattering can be entirely satisfied.

We now estimate the value of α'_β at resonance ($\omega_{\xi_0} = \omega_0$) for ruby ($Cr^{3+}$ in Al_2O_3). The Hamiltonian of the spin-phonon interaction has the form

$$\mathcal{H}_c = \frac{3}{2} G_{33} U [S_z^2 - \frac{1}{3} S(S+1)],$$

where G_{33} is the constant of spin-phonon interaction in the Voigt notation.

Let the constant magnetic field $\mathbf{H}_0$ be directed at an angle to the c axis of the crystal equal to twice that of the pump ($\theta = 54°44'$). Then the energy levels (and there are four of them, S = 3/2) are distributed symmetrically with respect to the line of zero energy. In this case, the pair of levels of interest to us will be levels 2 and 3 (the enumeration is from 1 to 4 in the direction of increasing energy). To create an inverted population, pumping is carried out between levels 2—4 and 1—3 with frequency $\omega_p = \omega_{42} = \omega_{31}$; $\omega_{32} = \omega_0$. For a constant magnetic field $H_0 = 4000$ G, using the tables of matrix elements and wave functions for ruby,[6] we determine the transition frequencies (ν_p, ν_0) and the quantity which characterizes the spin-phonon coupling κ with account of the fact that for ruby $G_{33} = 6.04$ cm⁻¹; $\kappa = 8.36 \times 10^{11}$ sec⁻¹, $\nu_p = 24.0$ GHz, $\nu_0 = 9.0$ GHz. From (11) we obtain

$$\alpha_{\omega_0}' \approx \frac{\hbar \kappa^2 \omega_0 T_2^*}{C_{33} v_0} n_0, \quad g(0) \approx \frac{T_2^*}{\pi}.$$

In the strong-pump approximation $n_0 \approx h\nu_0 NI/4 k_B T$, where k_B is the Boltzmann constant, T the temperature in °K, I the inversion coefficient, which, in the case of equality of all the spin-lattice relaxation times, is equal to $I = 1 - \nu_p/\nu_0$; N is the concentration of paramagnetic ions. For the following values of the various quantities (T = 4.2°K): $v_0 \approx 3 \times 10^3$ m/sec; $C_{33} \approx 5 \times 10^{10}$ N/m², $T_2^* \approx 1.4 \times 10^{-8}$ sec, we get $\alpha'_{\omega_0} \approx -0.187 \times 10^{-18}$ N cm⁻¹ (N in cm⁻³). For the same frequencies and the same κ, but in the three-level scheme, $\alpha'_{\omega_0} \sim -0.5 \times 10^{-19}$ N cm⁻¹. Thus, for strong dilution (0.01—0.1%), the damping coefficient due to the spin-phonon interaction can be $\sim \alpha_0^{\xi_0}$ in absolute value, which, for liquid-helium temperatures (4.2°K), ≈ 0.1 cm⁻¹.

5. The following method of investigation of APR on the basis of the stimulated Mandel'shtam-Brillouin scattering can be proposed. By varying the intensity of the laser radiation and each time initiating stimulated Mandel'shtam-Brillouin scattering by a change in the constant magnetic field H_0, we can study the phenomenon of APR over the entire range of frequencies corresponding to the width of the APR line.

This method has excellent prospects for the following reasons: a) the hypersound necessary for the experiment is generated right in the crystal under study. Consequently, there is no problem in obtaining satisfactory coupling between the piezoelectric transducer and the paramagnetic crystal; b) from estimates, it is seen that the effect is perceptible (i.e., $|\alpha'_{\omega\xi}| \sim \alpha_0^{\xi}$) for crystals with very strong magnetic dilution, ~0.01%, and with small values of the constant of spin-phonon interaction; c) these experiments can be carried out with the help of ordinary low-power gas lasers (of the He-Ne type), working in a continuous regime.

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[1]) We assume that E_0, E_{-1s} lie in the xy plane.

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