Mandel'shtam-Brillouin Light Scattering Investigation of a Phonon System Under Paramagnetic Resonance Saturation Conditions

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The nonequilibrium phonon system is investigated by the Mandel'shtam-Brillouin light scattering technique at saturation of paramagnetic resonance of the Ce^{3+} ions in the $Ce_2Mg_3(NO_3)_{12}$ $24H_2O$ crystal under phonon "bottleneck" conditions. Pulsed saturation and a frequency deviation from the center of the EPR line resulted in the detection of an avalanche-type growth of the number of phonons produced behind the leading edge of the pulse. Transition and stationary processes were studied as functions of the magnitude of the detuning. Information is obtained on the phonon spectral distributions for saturation in the center of the EPR line and detuning of ± 100 MHz when the intensity of the avalanche reaches a maximum. The phonon spectrum in the avalanche is narrower by at least an order of magnitude than the EPR line width, and is shifted with respect to the line center beyond the saturation point. Kinetic equations are set up which describe the coupled spin-phonon-thermostat system with account of spin-spin interactions. In particular cases, the equations are identical with those of Provotorov's theory and the bottleneck phonon theory. The main features of the phenomenon under study can be explained qualitatively by the equations.

1. INTRODUCTION

UNTIL recently, the investigations of paramagnetic resonance have been carried out exclusively by observation of the change in the spin system under the action of a variable magnetic field of resonance frequency. Meanwhile, the interaction of the spin system with a system of phonons, coupled in turn with the thermostat surrounding the specimen studied, makes possible the investigation of various processes under conditions of paramagnetic resonance from the change in the state of the phonon system. Investigations of this type have been especially interesting at low temperatures, where the spin-lattice relaxation takes place mainly as the result of single phonon processes, because in this case the changes in the phonon system pertain only to the phonon resonance frequency and are therefore more noticeable. It was proposed in[1] to use the Mandel'-shtam-Brillouin light scattering (MBS) for the direct detection of changes in the phonon system of paramagnets. Theoretical consideration has shown that the frequency shift and the growth of intensity of the Mandel'shtam-Brillouin (MB) components can easily be measured under certain conditions. As is well known,[2] the relative change in frequency Ω of the light scattered at angle ϑ is equal to

$$\frac{\Delta\Omega}{\Omega} = \pm 2n \frac{v}{c} \sin \frac{\vartheta}{2},$$

where v is the speed of sound and c/n the speed of light in the crystal. The intensity of the scattered light is proportional to the number of phonons of frequency $\Delta\Omega$. In contrast with certain other methods which only permit us to find the mean values of the quantities characterizing the system of phonons as a whole, the great advantage of the MBS method is the possibility of tracking phonons that have a definite momentum and type of polarization.

The first experiments on observation of MBS under conditions of paramagnetic resonance aimed at the

study of the phenomenon of the phonon "bottleneck," which is encountered when the transition from the system of resonance phonons to the thermostat is the most difficult in the chain of interacting systems consisting of the radiofrequency field, spin system, phonon system, thermostat. The consequent significant increase in the number of resonance phonons under conditions of stationary saturation of the paramagnetic resonance was discovered in the research of[3] on MgO crystals with Ni^{2+} impurity ions and somewhat later, independently, by the authors of the present work on cerium magnesium nitrate.[4] The application of the method of pulse saturation together with the stationary saturation of the paramagnetic resonance allowed us to obtain valuable additional information, of which the most interesting was the discovery of an avalanche growth in the number of resonance phonons.[5] The detailed presentation of the results of the experimental and theoretical investigation of the phonon bottleneck in cerium magnesium nitrate (CeMN) is the subject of this paper.

2. METHOD OF EXPERIMENT

The phonon bottleneck by light scattering was observed with the apparatus whose block diagram is shown in Fig. 1.

The investigated sample S, which has the shape of a parallelepiped of volume 0.15 cm^3, is placed on the bottom of a rectangular UHF resonator R with H_{10} mode, placed in an optical cryostat. The light ray of a helium–neon laser with wavelength 6328 Å and power 80 mW is focused by the lens L_1 in the center of the sample. The scattered light is separated by the diaphragm D_1 at an angle of $90°$ to the incident light, is collimated by the lens L_2 and directed by the rotable mirror M into the scanning Fabry-Perot interferometer (the reflection coefficient of the plate is 97%). The objective O focuses the rays in the plane of the diaphragm D_2, which separates the central part of the
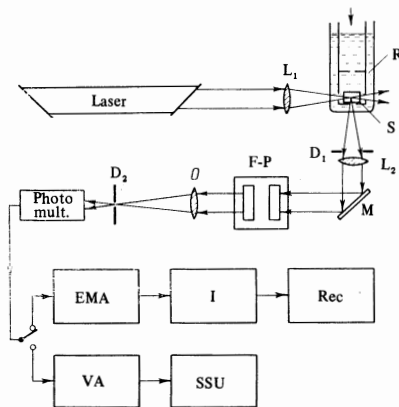
FIG. 1. Block diagram of experimental apparatus.

interference picture. Next, the light signals are transformed by an FEU-38 photomultiplier and fed to an electronic circuit consisting of two channels. One of these contains an electrometric amplifier EMA, an integrator I and an automatic recorder Rec and serves to record the MBS spectra. The other is connected to the video amplifier VA, which amplifies in the frequency range from zero to 1 MHz, and an 800-channel signal storage unit SSU with minimum time of analysis in a single channel equal to 10 microsec. This channel is intended for the investigation of transient processes of "heating" of the system of phonons by change in the intensity of the MBS components.

The optic axis of the crystal was perpendicular to the external constant magnetic field H and was identical with the direction of observation of the scattering. Experimentally,[6] such optical orientations give only a single pair of MBS components which are induced by the quasi-transverse elastic waves (for brevity, these are called transverse components in what follows). With the cooling the crystal to the working temperature of $1.5°K$, the intensity of these lines falls below the detection threshold. As our measurements in the range $10--15°K$ have shown, the shifts of the MBS components in CeMn remain practically unchanged below the boiling point of liquid nitrogen. Therefore, in the choice of conditions for observation of the phonon bottleneck, we used the results of the spectral measurements obtained for $77°K$.[6]

The frequency ω_1 of a saturated klystron with a power of about 200 mW was chosen to be equal to the value of the shift of the transverse components in the realized orientation of the crystal. Saturation was obtained both in the center of the EPR lines of the Ce^{3+} ions and on their wing, i.e., with detuning away from the exact resonance frequency ω_0, which is determined by the usual relation $\hbar\omega_0 = g\beta H$. The frequency difference $\Delta\omega = \omega_0 - \omega_1$ was produced by varying the magnetic field intensity H and consequently the value of ω_0. The advantage of this method is the preservation of the constant level of the rf power in the resonator. The "heating" of the controlled phonons was detected by the increase in the intensity of the corresponding MBS components. The effective temperature of the "hot" phonons was determined by comparison of the intensity (at the maximum of the spectral lines) of the

scattering by the nonequilibrium and equilibrium phonons for the temperatures 77 and $293°K$.

Initially, in the study of the stationary "heating" of the system of phonons, we used continuous saturation of the paramagnetic resonance. Later, the measurements were carried out with excitation of the spin system by periodic rectangular pulses (this regime was produced by means of a waveguide diode key) and the stationary process was studied together with the transient process which arises at the instant of initiating the saturating field. The duration of the pulses was 15 millisec and was sufficient for the transient to be practically finished within the time of action of each of them.

In the investigation of the dependence of the effective temperature of the "hot" phonons on the detuning, the optical receiver separated one of the excited MBS components and its integrated intensity was measured for each fixed value of $\Delta\omega$. The spectral distribution of the "hot" phonons was studied by measuring the mentioned quantity as a function of the angle of observation ϑ, which was changed in discrete amounts over a small range near $90°$. Decrease in the contribution to the resulting spectral form due to the finiteness of the aperture angles of the incident and scattered light was achieved by additionally restricting the corresponding light beams with diaphragms. We note that the contribution of the Doppler width $\delta\Omega$ of the laser beam to the phonon spectrum is of the order of the quantity $(\Delta\Omega/\Omega)\delta\Omega$ and is negligibly small.

3. EXPERIMENTAL RESULTS

In the first of the experiments mentioned, the orientation of the single CeMN crystal was such that the scattering took place from phonons which propagate along the $(\bar{1}01)$ direction in the set of coordinates used in[6]. The continuous saturation of the EPR line at the strict resonance frequency of 6.8 MHz led to a strong increase in the intensity of the corresponding MBS doublet. The value of the effective temperature T_{eff} of the resonance phonons amounted to $150°K$ and their spectral band was of the same order as the width of the unsaturated EPR line of Ce^{3+}, which is 270 MHz. The phonon "heating" is characterized by these parameters and under the conditions of pulse excitation of the spin systems at the line center.

Essentially new features of the effect of the phonon bottleneck were discovered in saturation on the wing of the EPR line. It turned out that saturation, not at the strict resonance frequency ω_0 but at frequencies which deviate from resonance by ± 100 MHz, corresponds to the maximum stationary "heating" of the phonon system. Furthermore, in pulsed saturation with detuning, an avalanche-like growth of the number of phonons was observed just after the leading edge of the pulse; thereafter, a comparatively slow decay occurred and the process assumed its steady state. Establishment of the equilibrium state occurred after the end of the saturating pulse (Fig. 2). The intensity of the phonon peak depends on the exact value of the detuning and for $\Delta\omega/2\pi \approx \pm 100$ MHz amounted to the maximum value, equal to $8000°K$. The larger part of the pulse, shown in Fig. 2, corresponds to the stationary "heating" with an effec-
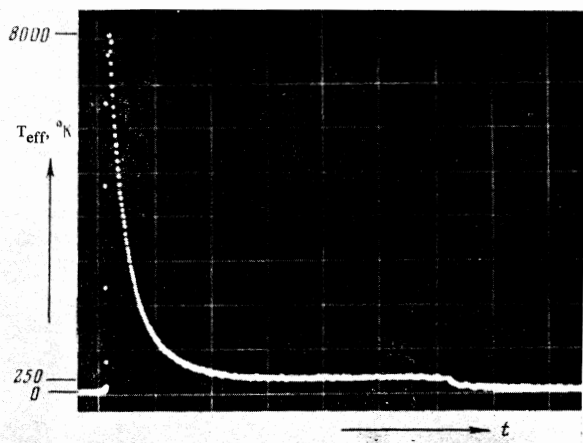
FIG. 2. Dependence of the scattered light intensity on time for pulse saturation on the wing of the EPR line (1 division on horizontal scale corresponds to 2.5 microsec.
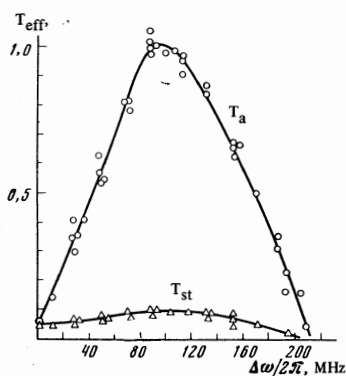

FIG. 3. Dependence of the effective temperature of the hot phonons at the peak of the avalanche and in the steady state on the value of the detuning.

FIG. 4. Spectrum of hot phonons for pulse saturation: a—on the wing of the EPR line, b—center of the line.

tive temperature of 250°K. The time of decay to the stationary level (to the half-maximum of the peak) amounts to 0.5 millisec.

The results of the measurement of the dependence of the characteristic parameters of the observed phenomenon on the value of the detuning ($\Delta\omega > 0$), obtained in another series of measurements, are shown in Fig. 3. Here T_a and T_s are the effective temperatures of the "hot" phonons at the peak of the avalanche and in the steady state, expressed in relative units. It is seen that the avalanche reaches maximum intensity for intermediate values of detuning, and decreases on both sides of the optimal value ~100 MHz, vanishing completely at the point $\Delta\omega = 0$. The effective temperature of the phonons in the steady state changes in stepwise fashion. It is noted that the avalanche is generated with maximum speed also in the region of optimal detuning. The time of decay of the avalanche, which is shorter for smaller $\Delta\omega$, increases rapidly with increase in $\Delta\omega$ above the optimal value. Saturation on the opposite wing of the EPR line leads to symmetric repetition of the graphs relative to the zero point, with only this difference that the maxima of the curves lie 5—19% lower than in Fig. 3.

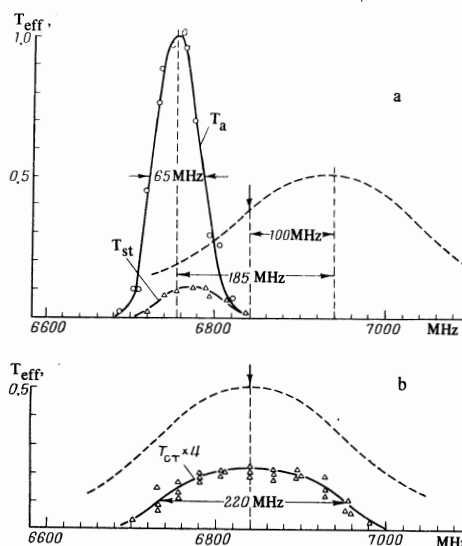Phonon avalanche was observed earlier (by indirect methods) in inversion of the spin level population under

"bottleneck" conditions.[7] The appearance of the avalanche as a result of simple saturation of the paramagnetic resonance appears rather strange at first glance. Important evidence for the explanation of this fact could be given by a spectral investigation of the observed phenomenon.

The spectral distributions of the effective temperatures T_a and T_s, found for $\Delta\omega/2\pi = 100$ MHz are shown in Fig. 4. For completeness, the spectrum of the bottleneck phonons, obtained at strict resonance saturation, is also shown here. The dashed curves indicate the shape and position of the unsaturated EPR line. An important feature of Fig. 4a is the fact that the maxima of the spectral distributions of the phonons do not coincide either with the frequency ω_0 or with the frequency ω_1, and are shifted still further from the saturation point along the wing of the EPR line. Moreover, the band of "hot" phonons is seen to be much narrower than the width of the EPR line. We note that the contours of these distributions are not true ones, since their widths were determined, within the limits of error of measurement, by the aperture angles of the incident and scattered light. This aperture width amounted to 60 MHz. An attempt at further decrease in the aperture width to the value equivalent to 30 MHz also did not lead to the establishment of the true band of "hot" phonons (this time only the distribution for T_a was obtained). Thus phonons in the band of frequencies below 30 MHz take part in the avalanche.

Measurements similar to those described were carried out for other orientations, which differ from the given rotation of the sample about the optic axis by angles of 90 and 270°, for conditions corresponding to new values of the shifts of the transverse MBS components. It was made clear that the change in the angle of propagation of the phonons in the crystal does not lead to a significant variation of the characteristics of the observed phenomenon. Only a somewhat different degree of "heating" of the phonon system was observed.

The results that have been stated refer to an un-

diluted sample. It would be interesting to test the behavior of the phonon avalanche for a different concentration of paramagnetic ions. With this purpose, we carried out experiments on a magnetically diluted sample, in which $3/4$ of the cerium ions were replaced with diamagnetic lanthanum.

As before, in the saturation an avalanche appears on the wing of the EPR line. Investigation of the dependence of its parameters on the value of the detuning and the observation angle led to a qualitative agreement with the results obtained previously. The difference was the following. The effect of "heating" of the phonons naturally decreases. To the maximum values of T_a and T_s correspond the detunings $\Delta\omega/2\pi = \pm(55-60)$ MHz, while the highest temperature of the phonons for the optimal detuning ± 60 MHz was achieved at a frequency that was about 45 MHz removed from the saturation point. The specified quantities indicate that a contraction in the frequency scale of the entire picture by a factor of almost two takes place with dilution. It is noteworthy that the dipole widths of the EPR lines in the investigated samples are found in the same relation.

Finally, we note that the value of $8000°K$, found in one of the experiments, does not correspond to the actual effective temperature of the phonons at the peak of the avalanche. Actually, this value is only the lower bound. According to what was said earlier, the phonon "heating" was estimated by a comparison of the light signals, which are proportional to the intensity of the MBS components, in scattering by nonequilibrium and equilibrium phonons. The intensity of the signal in the case of equilibrium phonons was determined by the whole aperture angle of observation, since the true band of phonons in the avalanche corresponds to smaller angles. As a result, the effective temperature of the phonons in the avalanche is seen to be lowered. This same consideration is valid for steady "heating" in saturation on the wing of the EPR line.

4. THEORY

The chief features of the observed picture of the avalanche-like growth of the number of phonons suggest that the essential role in this phenomenon is played by the reservoir of spin-spin interactions (SSI). This is especially shown by the fact that, for not strictly resonance saturation of EPR, the avalanche takes place at a frequency that is shifted still further on the wing of the EPR line relative to the frequency of saturating variable field, i.e., approximately in the range of frequencies of the induced radiation, which is determined by the heating or cooling of the SSI.[8,9] Thus, the problem arises of the description of the kinetics of three connected nonequilibrium systems—the Zeeman, the SSI and the phonons. Special cases of this problem have been solved: in the researches of Redfield[10] and Provotorov,[11] a theory of saturation of paramagnetic resonance with account of SSI has been constructed for the equilibrium phonon subsystem; Faughnan and Strandberg[12] investigated the kinetics of the effect of the phonon bottleneck without account of SSI. In this section, we shall construct a set of kinetic equations describing EPR saturation in the general case, when

all three subsystems—the Zeeman, SSI and phonon—are in nonequilibrium states, and consider the consequences of these equations. We note that an attempt at a semi-qualitative theoretical analysis of SPR saturation under conditions of phonon bottleneck with account of SSI has been undertaken in[9]. However, the heating of the phonons in[9] was considered in the steady state, while we are chiefly interested in transient processes. Moreover, the lack of clarity of interpretation of the results in[5] have raised criticism in the literature.[13]

A. Fundamental Equations

The Hamiltonian of the considered system can be represented in the form

$$\mathcal{H} = \mathcal{H}_z + \mathcal{H}_d + \mathcal{H}_{ph} + \mathcal{H}_{sph} + \mathcal{H}_r + \mathcal{H}_{sr},$$

$$\mathcal{H}_z = \omega_0 \sum_j S_{zj}, \qquad \mathcal{H}_d = \frac{1}{2} \sum_{ij} (A_{ij} S_{zi} S_{zj} + B_{ij} S_{+i} S_{-j}),$$

$$\mathcal{H}_{ph} = \sum_{k\sigma} \omega_{k\sigma} n_{k\sigma}, \qquad \mathcal{H}_{sph} = i \sum_{jmk\sigma} g_{k\sigma}{}^m S_{mj} e^{ikr_j}(b_{k\sigma} - b_{-k\sigma}^+), \qquad (1)$$

$$\mathcal{H}_r = \omega_1 B^+ B, \qquad \mathcal{H}_{sr} = V \sum_j (S_{+j} + S_{-j})(B^+ + B).$$

Here $\mathcal{H}_z$, $\mathcal{H}_d$, $\mathcal{H}_{ph}$, $\mathcal{H}_r$ are the respective Hamiltonians of the Zeeman, SSI, phonon and photon systems; $\mathcal{H}_{sph}$ and $\mathcal{H}_{sr}$ are the spin-phonon and spin-photon interactions; ω_1, $\omega_{k\sigma}$, ω_0 are the frequencies of the photons, phonons and Larmor precession; S_{mj} is the projection of the spin operator at the j-th node ($S_\pm = S_x \pm iS_y$); $n_{k\sigma} = b_{k\sigma}^+ b_{k\sigma}$; $b_{k\sigma}^+$, $b_{k\sigma}$ are the creation and annihilation operators of a phonon with wave vector k and polarization σ; B^+ and B^- are the corresponding photon operators; $g_{k\sigma}m$ and V characterize the value of the spin-phonon and spin-photon interactions; in this section, $\hbar = 1$ is Planck's constant.

For the derivation of the system of kinetic equations, we use the method of nonequilibrium statistical operator of Zubarev.[14] In correspondence with the ideas of this method, we shall assume that the state of the system can be completely determined by the average values of the dynamic variables S_z, $\mathcal{H}_d$, $n_{k\sigma}$ and their conjugate thermodynamic coordinates α, β, γ_k, which are proportional to the reciprocal temperature of the Zeeman system, SSI and phonons, respectively. This means that the indicated parameters should change but little during the course of the spin-spin relaxation T_2. It is convenient to introduce uniformity in the designation of the dynamic variables S_z, $\mathcal{H}_d$, $n_{k\sigma} \to P_m$ and of their corresponding thermodynamic coordinates α, β, $\gamma_{k\sigma} \to F_m$, where m enumerates these variables. Then the nonequilibrium statistical operator can be written in the form:[14]

$$\rho(t) = Q^{-1} \exp\left\{ -\sum_m P_m F_m(t) + \int_{-\infty}^{0} dt_1 e^{\varepsilon t_1} \sum_m \left[\frac{\partial P_m(t_1)}{\partial t_1} F_m(t + t_1) \right. \right.$$
$$\left. \left. + P_m(t_1) \frac{\partial F_m(t + t_1)}{\partial t} \right] \right\}, \qquad (2)$$

where $Q = \mathrm{Sp}\,\rho$, $A(t)$ is the operator A in the Heisenberg representation, $\varepsilon \to +0$. The meaning of the parameters $F_m(t)$ is determined from the condition that the average values $\langle P_m \rangle = \mathrm{Sp}\,\{P_m \rho\}$ must be

equal to the average $\langle P_m \rangle_l = \mathrm{Sp}\{P_m \rho_l\}$, calculated with the local-equilibrium statistical operator

$$P_l = Q_l^{-1} \exp\left\{-\sum_m P_m F_m(t)\right\}. \tag{3}$$

We write out explicitly the equations of motion for the operators S_z, $\mathcal{H}_d$ and $n_{k\sigma}$:

$$i\dot{S}_z = i\sum_{jk\sigma}(g_{k\sigma}^+ S_{+j} - g_{k\sigma}^- S_{-j})e^{ikr_j}(b_{k\sigma} - b_{k\sigma}^+)$$

$$\qquad + V\sum_j (S_{+j} - S_{-j})(B^+ + B),$$

$$i\dot{\mathcal{H}}_d = i\sum_{jmk\sigma}\{g_{k\sigma}^m[\mathcal{H}_d, S_{mj}]\}e^{ikr_j}(b_{k\sigma} - b_{-k\sigma}^+) \tag{4}$$

$$\qquad + V\sum_j \{[\mathcal{H}_d, S_{+j} + S_{-j}][B^+ + B]\},$$

$$\dot{n}_{k\sigma} = -\sum_{jm} g_{k\sigma}^m S_{mj} e^{ikr_j}(b_{k\sigma} + b_{-k\sigma}^+),$$

where $[A, B] = AB - BA$. We now average these equations with the help of the statistical operator (2), limiting ourselves to the components up to second order, inclusively, in the interactions $\mathcal{H}_{\text{sph}}$ and $\mathcal{H}_{\text{sr}}$. Inasmuch as we have $\langle P_m \rangle_l = 0$ for all P_m, the averaged equations take on an identical structure:

$$\frac{d}{dt}\langle P_m \rangle = \sum_n \int_{-\infty}^0 dt' e^{zt'}(\dot{P}_m, \dot{P}_n(t'))_l{}^t F_n(t+t'), \tag{5a}$$

$$(\dot{P}_m, \dot{P}_n(t'))_l{}^t = \int_0^1 d\lambda \left\langle \dot{P}_m \exp\left[\lambda \sum_r P_r F_r(t)\right]\dot{P}_n(t')\right.$$

$$\qquad \left. \times \exp\left[-\lambda \sum_r P_r F_r(t)\right]\right\rangle_l{}^t. \tag{5b}$$

In correspondence with the adopted approximation, we shall calculate the correlation functions (5b) in the zeroth approximation in $\mathcal{H}_{\text{sph}}$ and $\mathcal{H}_{\text{sr}}$. In the high-temperature approximation, we have

$$\langle S_+ S_-(t')\rangle_l{}^t = {}^2/_3 S(S+1)e^{-i\omega_0 t'}f(t'),$$
$$\langle b_{k\sigma}^+ b_{k'\sigma'}(t')\rangle_l{}^t = \delta_{kk'}\delta_{\sigma\sigma'}e^{-i\omega_{k\sigma}t'}n_{k\sigma}(t),$$

$$\langle S_z\rangle_l{}^t = -{}^1/_3 S(S+1)N\alpha(t); \quad \langle \mathcal{H}_d\rangle_l{}^t = -{}^1/_3 S(S+1)N\omega_l^2\beta(t),$$

$$\omega_l^2 = {}^1/_3 S(S+1)\sum_j ({}^1/_2 A_{ij}^2 + B_{ij}^2), \tag{6}$$

where $f(t)$ is the Fourier transform of the normalized function of the shape of the unsaturated EPR line, N the number of paramagnetic centers. We shall assume that $f(t)$ is determined completely by the SSI; therefore,

$$\langle S_+[S_-(t'), \mathcal{H}_d(t')]\rangle_l{}^t = \frac{2}{3}S(S+1)e^{-i\omega_0 t'}i\frac{\partial f(t')}{\partial t'} \tag{7}$$

and so forth. If the SSI is much less than the Zeeman energy, then we can everywhere neglect components containing the form function at a frequency that is always far from resonance, $f(\omega_0 + \omega_{k\sigma})$ and $f(\omega_0 + \omega_1)$. In addition, we shall neglect the anisotropies of the spin-phonon interaction and take it into account that the principal contribution to the sum over σ is made by the transverse vibrations. In this connection, it is convenient to introduce the following notation:

$$\sum_\sigma g_{k\sigma}^+ g_{k\sigma}^-(2n_{k\sigma}^0 + 1)\rho_0(\omega_0) = \frac{1}{\tau}; \quad \frac{{}^1/_3 a_0^2 S(S+1)N}{\sum_\sigma \rho_\sigma(\omega_0)\Delta} = b, \tag{8}$$

$$\frac{\alpha}{\alpha_0} = x, \quad \frac{\beta}{\beta_0} = y, \quad \frac{n_\omega - n_\omega^0}{n_\omega^0} = z_\omega. \tag{9}$$

Here τ is the time of spin-lattice relaxation in the absence of saturation; $\rho_0(\omega_0)$ is the spectral density of the lattice oscillators; Δ^2 the second moment of the absorption curve $f_{\omega \to \omega_0}$; α_0, β_0 and $n_{k\sigma}^0$ are the equilibrium values of the corresponding parameters. Transforming from a sum over k to integrals over the frequency (removing the slowly changing (in comparison with $f(\omega)$) function from under the integral over the frequency ω_0), we finally obtain:

$$-\frac{dx(t)}{dt} = \frac{1}{\tau}\int f_{\omega-\omega_0}z_\omega(t)\left[x(t) + \frac{\omega - \omega_0}{\omega_0}y(t)\right]d\omega + \frac{x-1}{\tau}$$

$$\qquad + Wf_{\omega_1-\omega_0}\left[x(t) + \frac{\omega_1 - \omega_0}{\omega_0}y(t)\right],$$

$$-\frac{dy(t)}{dt} = \frac{1}{\tau}\int \frac{\omega_0(\omega - \omega_0)}{\omega_l^2}f_{\omega-\omega_0}z_\omega(t)\left[x(t) + \frac{\omega - \omega_0}{\omega_0}y(t)\right]d\omega \tag{10}$$

$$\qquad + \frac{\Delta^2 y}{\omega_l^2 \tau} + Wf_{\omega_1-\omega_0}\frac{\omega_0(\omega_1 - \omega_0)}{\omega_l^2}\left[x(t) + \frac{\omega_1 - \omega_0}{\omega_0}y(t)\right],$$

$$\frac{dz_\omega(t)}{dt} = -\frac{b\Delta}{\tau}f_{\omega-\omega_0}\left\{(z_\omega(t)+1)\left[x(t) + \frac{\omega - \omega_0}{\omega_0}y(t)\right] - 1\right\} - \frac{z_\omega(t)}{\tau_{ph}},$$

where $W = V^2\langle B^+ B + 1\rangle$; the quantity $Wf_{\omega_0 \to \omega_1}$ is the probability of transition in the spin system under the action of the saturating variable field. In the last equation, we have introduced an additional term, which determines the lifetime of the phonons in the absence of the spin-phonon interaction. It is not difficult to see that in the special case $z \equiv 0$, Eqs. (10) are identical with the equations of Provotorov.[11] The equations of the theory of the phonon bottleneck[12] are obtained from (10) in the case $y = 0$ and for the condition that the distribution function of the number of phonons over the frequency z changes little in the region of the EPR line.[1]

B. The Steady State

We consider first the steady state of the EPR line, i.e., $\dot{x} = \dot{y} = \dot{z} = 0$. Inasmuch as it is impossible to solve the set of integral equations (10) in general form, we shall consider the case of a small departure from the phonon equilibrium distribution, when $b\tau_{ph}/\tau = 0 < 1$. Then, we shall have for strong saturation

$$x_0 + \frac{\omega_1 - \omega_0}{\omega_0}y_0 = 0, \quad x_0 = \frac{(\omega_1 - \omega_0)^2}{\Delta^2 + (\omega_1 - \omega_0)^2}, \tag{11}$$

$$z_\omega(\infty) = \sigma\Delta\frac{\Delta^2 + (\omega_1 - \omega_0)(\omega - \omega_0)}{\Delta^2 + (\omega_1 - \omega_0)^2}f_{\omega-\omega_0}. \tag{12}$$

It is then seen that in saturation of the EPR line at the resonance frequency $\omega_1 = \omega_0$, the distribution of the number of phonons over the frequencies for $\sigma \ll 1$ is identical with the shape of the EPR line. If now $\omega_1 \neq \omega_0$, then the maximal heating of the system of pho-

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[1]We note that the combination of temperatures of the Zeeman system and SSI, $x + y(\omega - \omega_0)/\omega_0$, defined in[9] as the inverse spin temperature at the frequency ω occur in natural fashion in Eq. (10). We must of course agree with the criticism of this description of the spin system in this relation that this parameter has no additional thermodynamic meaning.[13] Strictly speaking, it is also impossible to speak of the "temperature of the system of phonons at the frequency ω," and the third equation in (10) is actually the kinetic equation for the distribution function of the number of phonons over the frequencies.

nons takes place at a frequency that is shifted still further relative to ω_1 on the wing of the EPR line. On the opposite wing, the shape phonons are cooled. The case when z changes little in frequency in the range of the EPR line is much simpler to analyze. This occurs, for example, in the rapid spectral diffusion of phonons. However, this case is not realized experimentally, and we shall not consider it here.

C. Pulsed EPR Saturation

We now consider transient processes that arise in the phonon subsystem after switching on of the saturating variable field. In what follows, we shall see that the character of the change in state of the system with time depends essentially on the level of the saturating power. Therefore, it is expedient to separate the case which is distinguished by the relation between the transition probability in the spin system under the action of the variable field and the reciprocal of the time of spin-lattice relaxation.

Let us first study the more interesting case $Wf_{\omega_1 - \omega_0} \gg 1/\tau$. Here, naturally, we shall assume satisfaction of the inequality $Wf_{\omega_1 - \omega_0} \gg 1/T_2$, which is a consequence of the conditions of applicability of the developed theory. In this situation, the spin system develops, initially being cut off from the lattice and, during the time $\Delta t < \tau$ goes over into a state characterized by the parameters x_0 and y_0. These will be determined by Eq. (11) if we replace Δ in it by ω_l, which follows from the first pair of equations (10). Turning to the third equation of (10), we note that for $z_\omega = 0$, the number of phonons at the frequency ω increases linearly with time, while the rate of change is positive if $(\omega_1 - \omega_0)(\omega - \omega_0) + \omega_l^2 > 0$. After the number of phonons in this range of frequencies increases so much that $z_\omega \gg 1$, the rate of increase becomes proportional to the number of phonons, and its sign will be determined by the effective difference in the population of the energy levels of the spin system

$$x + \frac{\omega - \omega_0}{\omega_0} y = x(\omega).$$

For the initial conditions (11), $x(\omega) < 0$ if $(\omega - \omega_1)(\omega_1 - \omega_0) > 0$, and the $dz_\omega/dt > 0$. Thus, a nonlinear, avalanche-like growth in the number of phonons takes place, and the maximum in its frequency distribution is shifted still further along the wing of the EPR line in comparison with the stationary distribution (12). Moreover, by virtue of the nonlinearity of the process, a significant narrowing of the distribution function $z(\omega)$ takes place in comparison with the stationary case. It is important to emphasize that no avalanche of phonons develops when $\omega_1 = \omega_0$. The ratio of the temperatures of the Zeeman system and the SSI changes in proportion with the radiation of the phonons of the spin system, and saturation of the EPR line becomes more homogeneous. As a result, the sign of dz_ω/dt changes and the reverse process begins—the cooling of the phonon subsystem. If the saturating variable field remains switched on in this case, then the rate of cooling is determined by the relation between the saturating power and the rate of transfer of the energy from the hot phonons to the thermostat,

and the function z_ω tends to take the form that is characteristic for stationary EPR saturation.

An avalanche of phonons should arise also when the EPR line is saturated by a pulse whose duration is shorter than the spin-lattice relaxation time. It is only important that the spin system be capable of entering a state characterized by the parameters (11) or be close to it. The process of phonon cooling in this case should also be essentially nonlinear, and its duration will evidently be of the same order as the time of generation of the avalanche. As $t \to \infty$, the function z_ω tends in this case toward $z_\omega \equiv 0$.

5. DISCUSSION OF RESULTS

We consider the obtained experimental results from the point of view of the theory previously developed. Inasmuch as the intensity of the scattered light at the frequencies $\Omega \pm \Delta\Omega$ is proportional to the number of phonons $n(\omega)$, we shall equate the intensity of the scattered light directly with the distribution functions relative to the number of phonons over the frequency $z(\omega)$.

In EPR saturation at the frequency $\omega_1 = \omega_0$ in the steady state, the experimental function $T_{eff} \sim z(\omega)$ in Fig. 4b in general terms repeats the shape of the EPR line, in accord with (12). Some deviation toward a rectangular curve agrees with the qualitative analysis of Eq. (10) for $\sigma \gg 1$. The theoretical curve (12) also gives a good representation of the behavior of the experimental function T_{eff} in Fig. 4a for not strictly resonance stationary EPR saturation.

The picture of the generation and growth of the phonon avalanche experimentally studied by us is in complete agreement with the qualitative analysis of Eqs. (10) in part B of the preceding section. In particular, the avalanche arises only for the not strictly resonance EPR saturation, and it is seen from Fig. 4a that the maximum in the distribution of the number of phonons over the frequency is shifted still further on the wing of the EPR line relative to the maximum z_ω for stationary saturation. A marked narrowing of the frequency distribution of the phonons in the avalanche is shown in Fig. 4a. It has unfortunately not been possible to determine the actual degree of narrowing of the line, inasmuch as the width of the distribution lies within the limits of the aperture width of the optical detector, as has already been noted. For this same reason, the intensity of the scattered light (and consequently, T_{eff}) is a measure in this case of the total number of phonons in the avalanche, irradiated in the range of frequencies of the aperture band. We note that the dependence on the frequency detuning $\omega_1 - \omega_0$ of the function

$$f_{\omega - \omega_0} x(\omega) = f_{\omega - \omega_0} \frac{(\omega - \omega_1)(\omega_1 - \omega_0)}{\omega_l^2 + (\omega_1 - \omega_0)^2}, \qquad (13)$$

is proportional to the initial rate of the avalanche and has qualitatively the same shape as the corresponding curve for the effective temperature of the phonons at the peak of the avalanche in Fig. 3.

It should be noted that the irradiation of the phonons of the spin system takes place incoherently. This is indicated first of all by the fact that the fastest part of the process, the generation of the avalanche, lasts for

tens of microseconds, while the time of spin-spin relaxation during which the coherence is disrupted in the spin system, amounts to $T_2 \sim 10^{-9}$ sec in our case. In principle, for sufficiently large T_2 and strong spin-phonon interaction, coherent radiation of phonons is possible for inversion of the populations of the energy levels of the spin system;[15] however, the conditions here are very stiff and are rarely realized experimentally.

In conclusion, we show that if the investigated crystal is a resonator for some sound modes in the avalanche, then the growth of the process is possible in the regime of coherent sound generation. The study of MBS on phonons under these conditions is of undoubted interest.

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