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INCREASE OF PARALLEL PARAMAGNETIC SUSCEPTIBILITY BY RESONANT PUMPING

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We report here a multiple increase of the parallel magnetic susceptibility (χ) of a solid paramagnet under conditions when the temperature T_{ss} of the spin-spin interactions is strongly shifted by saturation of the paramagnetic resonance.

The study of the $\chi(\omega)$ dependence at frequencies ω much lower than the paramagnetic-resonance frequency ν_0 is the oldest method of investigating relaxation phenomena in spin system (the "parallel-field method" [1]). This dependence is usually of the form

$$\chi(\omega) = \sum_i \chi_i(\omega) = \sum_i \frac{\chi_i(0)}{1 + i\omega\tau_i}, \tag{1}$$

where τ_i can denote the spin-lattice relaxation time, the time for the establishment of equilibrium in the entire spin system, or else the cross-relaxation time (τ_{cr}) in a system with several resonance lines with close frequencies [2]. Since none of the $\chi_i(0)$ exceed the static susceptibility χ_0, the sensitivity of the "parallel field" method is low (we recall for comparison that when paramagnetic resonance is observed the susceptibility at the frequency ν_0 is $\chi_0(\nu_0/\delta\nu) \sim (10^2 - 10^3)\chi_0$, where $\delta\nu$ is the half-width of the resonance line.

The possibility of greatly increasing the absolute value of the parallel susceptibility, realized in the experiment described below, is based on a simultaneous application of two alternating magnetic fields to the paramagnetic specimen, a field $H_1 e^{i\omega t}$ parallel to H_0 and used to measure $\chi(\omega)$, and a field $h_1 e^{2\pi i \nu t}$ perpendicular to H_0 to saturate the paramagnetic resonance on the wing of the resonance line (with a detuning $|\nu - \nu_0| \sim \delta\nu$). As shown by one of us [3], such a saturation, which is known to decrease $|T_{ss}|$ strongly (cf. [4, 5]), should increase all the χ_i values on the order of $(T_0/T_{ss}) \sim (10^1 - 10^3)\chi_0$, where T_0 is the lattice temperature. We have verified this prediction experimentally by measuring $\chi_{cr}(\omega)$. As is well known, the cross-relaxation susceptibility is due to modulation of the z-component of the macroscopic magnetic moment of the sample M_z in the field $H_1 e^{i\omega t}$ owing to the redistribution of the Zeeman energy among two spin subsystems having close resonant frequencies $\nu_1 \simeq \nu_2$ (it is assumed that the detuning $\Delta_{12} \equiv \nu_1 - \nu_2$ depends on the external magnetic field and is therefore also modulated at the frequency ω). It can be shown (cf., e.g., [2]) that χ_{cr} is proportional to the difference $(\nu_1/T_{z1}) - (\nu_2/T_{z2})$, where T_{z1} and T_{z2} are the Zeeman spin temperatures of the corresponding subsystems. On the other hand, it is known [7, 6] that under the conditions

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of effective cross relaxation this difference tends to Δ_{12}/T_{ss}, so that the expected enhancement of χ_{cr} in comparison with the equilibrium value should amount to T_0/T_{ss} (the same conclusion is also arrived at by an exact calculation [3]).

The experiments were performed on a ruby crystal with chromium concentration 0.03% at $T_0 = 1.7°K$. The conditions for cross relaxation between the different transitions of the EPR spectrum of Cr^{3+} were set by choosing the angle θ between H_0 and the C axis of the crystal (cf., e.g., [8]). The imaginary part of the susceptibility χ''_{cr} was determined at frequencies $\omega/2\pi$ in the range $3 \times 10^5 - 6 \times 10^6$ Hz by measuring the absorption signal produced in an induction coil wound around the ruby sample and connected in the circuit of an ordinary autodyne NMR spectrometer.

Using a customary procedure of magnetic spectroscopy, we measured not $\chi''(\omega)$ but $\chi(H_0)$ at ω = const. The absorption signal was registered using deep modulation of the magnetic field (at 50 Hz) about the value H_0^{cr} corresponding to exact equality of the frequencies of two transitions of the EPR spectrum, $\nu_1 = \nu_2$, and had a maximum at this point (Figs. 1 and 2a). Such a shape of the $\chi''_{cr}(H_0)$ curve agrees with (1) if $\omega \gg \tau_{cr}^{-1}$ and describes the dependence of the cross-relaxation probability W_{cr} on Δ_{12} (cf., e.g., [9]).

$|T_{ss}|$ was lowered by saturating the wing of some EPR line of ruby at a wavelength 3.2 cm (in the case shown in Figs. 1 and 2, we saturated the low-frequency line wing, corresponding to a 2 - 3 transition if the energy levels are numbered upwards). As seen from Fig. 2b, the susceptibility signal was increased by many times in this case (the gain in the figure is $T_0/T_{ss} = +15$). When the opposite wing of the EPR line was saturated, we observed inversion of the $\chi_{cr}(H_0)$ curve (negative-absorption signal), but the gain had the same absolute value.
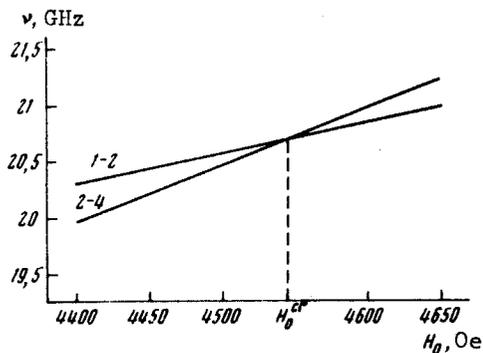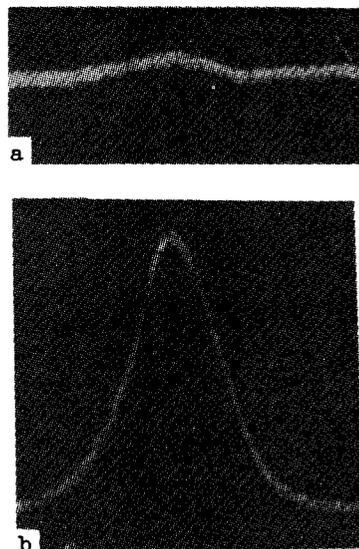

a

b

Fig. 1

Fig. 2

Fig. 1. Frequencies of the (1 - 2) and (2 - 4) transitions of the EPR spectrum of Cr^{3+} in ruby (the energy levels are numbered upwards) vs. the magnetic field H_0 at $\theta = 34°$.

Fig. 2. Oscillograms of the χ''_{cr} signal at $\theta = 34°$. The sweep corresponds to variation of H_0 from 4475 to 4625 Oe: a - under ordinary conditions, b - upon saturation of the EPR (2 - 3) line wing.

By suitable choice of the frequency we could, simultaneously with the observation of χ''_{cr}, register the NMR spectrum of the ^{27}Al nuclei in the ruby lattice (to this end, the coil axis was inclined somewhat to the H_0 direction). It turned out that the gain of the χ''_{cr} signal was exactly equal to the gain in the polarization of the ^{27}Al nuclei; furthermore, the NMR and χ''_{cr} signals returned to their equilibrium values, after the microwave was turned off, at the same time constant (8 sec in Figs. 1 and 2) characterizing the nuclear spin-lattice relaxation. We note that this result seems to be the most direct experimental proof of the existence of a thermal contact between the reservoir of the electronic spin-spin interactions of the paramagnetic impurity and the Zeeman system of the lattice nuclei (cf., e.g., [6]), and it is clear that this contact remains also in the absence of a saturating field.

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NONSTATIONARY STIMULATED SCATTERING BY POLARITONS IN LITHIUM IODATE

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The increased interest in stimulated Raman scattering by polaritons is due both to the possible high-efficiency conversion of laser frequencies and to the possibility of obtaining information on the dispersion characteristics of the medium. In addition, the extraction of infrared oscillations from a crystal makes it possible to produce a tunable laser operating in the far infrared [1].

The present communication is devoted to the first experiments on SRS by polaritons under nonstationary conditions (with picosecond pumping). The scattering was investigated, in particular, in an ionic lithium-iodate crystal belonging to the point group $P6_3(C_6^6)$ with two molecules per unit cell, and has a rich spectrum of infrared-active and inactive transitions [2, 3]. However, inasmuch as the stimulated scattering develops collinearly with the pump wave the number of oscillations that can be excited without supplementary conditions (such as transverse resonators) is limited. At the same time, excitation of oscillations with the aid of a broad spectrum can create possibilities for the observation of stimulated scattering by lower modes that lie in this case in the pump-spectrum line wing.