sound. The sources of sound waves are observed in He II also on the flat heater 2, which has a different construction. The visible bubbles of vapor in He II reach maximum dimensions (1 - 4 mm) within 2 - 10 msec, and their vanishing by the time the instant t_+ = 20 msec is reached is not accompanied by noticeable emission of sound.

[1] A.I. Gulyaev, Zh. Eksp. Teor. Fiz. 57, 59 (1969) [Sov. Phys.-JETP 30, 34 (1970)].

"QUENCHING" OF CROSS CORRELATION IN INHOMOGENEOUSLY BROADENED EPR LINES

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Spin-spin correlation has a significant influence on the processes of saturation and relaxation and multilevel systems [1], particularly in inhomogeneously broadened EPR lines. In measurements of the spin-lattice relaxation (SLR) times by the pulsed-saturation method, the cross relaxation can distort in a complicated manner the relaxation curves and lead to considerable errors in the determinations of the SLR times T_1. We have set up an experiment wherein the influence of the cross relaxation within the line can be completely eliminated (we call this the "quenching" of the cross relaxation), and have shown that the SLR time obtained in this case differs strongly from that obtained with the aid of the ordinary pulsed-saturation method. The "quenching" effect was obtained by rapidly sweeping the line during the saturation time. The entire line turned out to be then homogeneously saturated, and the restoration of any section of the line took place exponentially only as a result of the spin-lattice relaxation, without participation by the cross relaxation.

The gist of the effect can be easily understood by using two spin subsystems ("spin packets") as an example. If only one subsystem is saturated (the usual pulsed-saturation procedure), then the restoration of its population occurs in accordance with the doubly-exponential law [1]

$$A\exp[-(T_1^{-1} + T_{12}^{-1})t] + B\exp(-T_1^{-1}t).$$

Here T_1 are the SLR times of both subsystems and T_{12} is the characteristic cross relaxation time. The first term describes the cross relaxation to the second subsystem. It decreases with increasing duration τ of the saturation, but does not vanish even when $\tau \gg T_{12}$.

However, if both subsystems are saturated, then, using the equations of [1] we can show that the restoration of the population of the levels of both subsystems will proceed exponentially, with a time T_1, and the cross relaxation is completely eliminated (A = 0).

In a many-level system, such as an inhomogeneously broadened EPR line, there should obviously exist an entire spectrum of cross-relaxation times T_{12}^i and, since one may encounter in such a system values of T_{12}^i close to T_1, it is impossible to separate in the relaxation curves the spin-lattice exponential, i.e., it is impossible to determine T_1.

The results of the analysis for two spin subsystems can obviously be extended to the case of an inhomogeneously broadened EPR line. If the entire EPR is homogeneously saturated, say by rapidly scanning it during the time of action of the saturating pulse, then, provided all the spectral parts of the line have identical SLR times T_1, each part of the line (each "spin packet") will relax independently at a rate T_1^{-1}, and the transfer of energy inside the line will be excluded, since there is no spin-temperature gradient along the line.

It should be noted that in the usual procedure of pulsed saturation it is possible in the general case only to "burn a hole" in the inhomogeneously broadened line, but not to saturate it completely [2].

We note also that at very low concentrations of the paramagnetic ions, the spectral diffusion in the inhomogeneously broadened lines can proceed, owing to the electron-nuclear interactions, not as a result of spin-spin cross relaxation, but as a result of "forbidden" electron-nuclear transitions – the mechanism considered in [3]. An analysis of the equations in that reference shows that upon saturation of the entire inhomogeneously broadened line this mechanism is also completely "quenched."

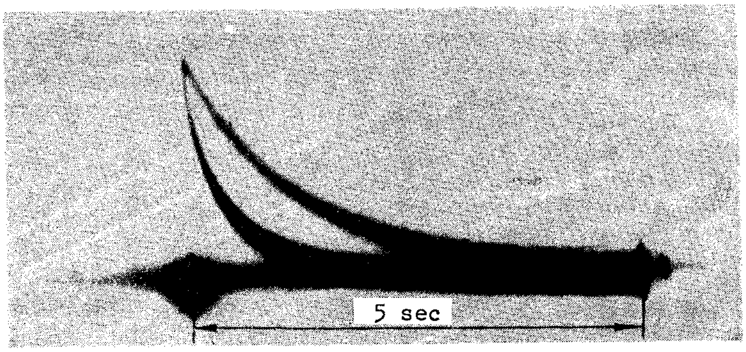

Fig. 1. Oscillograms of relaxation curves of Nd^{3+} in $Ca_5(PO_4)_3F$, obtained by the usual method of pulsed saturation (lower curve) and by "quenching" the cross relaxation (upper curve).

The validity of the considerations advanced above was verified by us for the EPR lines of Nd^{3+} in fluorapatite $Ca_5(PO_4)_3F$, whose EPR spectra and relaxation were investigated by us earlier in [4]. The EPR lines of Nd^{3+} in this crystal have an appreciable inhomogeneous broadening, owing to the strong anisotropy of the g-factor (line width ∿300 G at an Nd^{3+} concentration 0.68 at.% in a field ∿8 kG).

The experiment was performed in the following manner. The line was scanned by angular magnetic modulation, using a solenoid whose axis was perpendicular to the direction of the constant magnetic field, fed with a sinusoidal current pulse from the 50 Hz line through a special circuit, which made it possible to synchronize the saturating pulse with the zero phase of the line current. The duration of the saturation at the frequency ν = 9430 MHz was equal to the duration of the current pulse in the solenoid (20 msec). Owing to the strong anisotropy of the g-force, the oscillation of the direction of the resultant magnetic field through 2 - 3° could readily cause deep modulation of the EPR line, something impossible to achieve by ordinary amplitude magnetic modulation.

The figure shows oscillograms of the relaxation curves for a sample with 0.15 at.% Nd^{+} at 4.2°K, obtained after saturating the line without modulation (lower curve) and with simultaneous modulation of the line (upper curve)[1]. The lower curve corresponds to the usual procedure of pulsed saturation and can be described by a sum of two exponentials, $0.3\exp(-t/\tau_1) + 0.7\exp(-t/\tau_2)$, where τ_1 = 83 msec and τ_2 = 420 msec. The upper curve corresponds to the procedure of "quenching" the cross relaxation and is described by a single exponential with a time T_1 = 710 msec.

We note that the contribution of the cross relaxation of the tails of the

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[1] These curves were obtained in succession on a long-persistence oscilloscope screen, and were photographed on a single frame for clarity.

relaxation curves, which depends on the relations between T^{i}_{12} and T_1, may greatly distort the temperature and concentration dependences of the SLR. Our measurements in $Ca_5(PO_4)_3F:Nd^{3+}$ show that these dependences actually change when the cross relaxation is "quenched." Detailed results will be published later.

[1] A.A. Manenkov and A.M. Prokhorov, Zh. Eksp. Teor. Fiz. 42, 75 (1962) [Sov. Phys.-JETP 15, 54 (1962)]; N.V. Karlov and A.A. Manenkov, Kvantovye usiliteli (Quantum Amplifiers), M., 1966.
[2] L.L. Luishvili, M.D. Zviadadze, and G.R. Khutsishvili, Zh. Eksp. Teor. Fiz. 56, 290 (1969) [Sov. Phys.-JETP 29, 159 (1969)].
[3] N.A. Efremov and M.A. Kozhushner, ibid. 57, 534 (1969) [30, 293 (1970)].
[4] D.M. Daraseliya, G.V. Maksimova, and A.A. Manenkov, ZhETF Pis. Red. 10, 361 (1969) [JETP Lett. 10, 229 (1969)].
[5] P. L. Scott and C.D. Jeffries, Phys. Rev. 127, 32 (1962).

FOUR-PHOTON SCATTERING IN A RESONANT MEDIUM

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Several recent papers are devoted to spontaneous multiphoton processes, namely parametric luminescence and four-photon scattering [see [1], where other papers are cited]. Parametric luminescence is relatively easy to observe, whereas four-photon scattering under nonresonant conditions turns out to be a very weak effect. We report here observation of a very strong change of the angular and spectral distributions of powerful nonmonochromatic radiation passing through a resonant medium (potassium vapor); this radiation is interpreted as a consequence of four-photon scattering.

In our setup, a giant ruby-laser pulse (~50 MW) excited SRS in nitrobenzene (or α-chloronaphthalene). The SRS radiation, filtered out of the laser light, passed through a cell with potassium vapor and was registered with a DFS-8 spectrograph (grating with 1200 lines/mm), the slit of which was in the focal plane of the condenser. The photographic plate therefore recorded the angular distribution (along the height of the slit) and the frequency distribution of the light emerging from the potassium-filled cell).

When SRS in nitrobenzene was used (the width of the SRS spectrum is about 4 cm^{-1} and is shifted by 12 cm^{-1} toward the short-wave side away from the potassium resonance line λ = 7665 Å, ω_0 = 13042.9 cm^{-1}), the following phenomena were observed: At low vapor pressures ($p \lesssim 5 \times 10^{-4}$ Torr), slight diffusion of the radiation in frequency and in angle is observed (Figs. 1Aa, b). Characteristic "whiskers" (Fig. 1Ac) appear at frequencies lower than ω_0 in the interval 5×10^{-4} Torr $\lesssim p \lesssim 5 \times 10^{-2}$ Torr. The angle distance between the "whiskers" increases with pressure (the square of the angle depends linearly on p). At the same time, the diffusion (in terms of θ and ω) increases simultaneously in the region of the initial SRS spectrum. At $p \gtrsim 0.1$ Torr, the "whiskers" leave the field of view (which is determined by the height of the spectrograph slit), and only additional broadening of the frequency-angle diagram a is observed in the region of the SRS spectrum (Fig. 1Ae). In the latter case, radiation appears also in the region of both resonance lines (7665/99 Å), shifted by 1 - 2 cm^{-1} toward longer wavelengths from the atomic frequencies, with predominant concentration at $\theta \neq 0$ (Fig. 1Ae).