CONFIRMATION OF THE EXISTENCE OF SPIN-SPIN INTERACTION TEMPERATURE IN EPR

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Submitted 12 May 1967
ZhETF Pis'ma 6, No. 4, 578-581 (15 August 1967)

A theory of magnetic resonance in a solid, in which the dipole-dipole interactions of paramagnetic particles (nuclei or electrons) are regarded as a separate energy reservoir possessing a temperature T_{int} that differs in general from both the spin temperature in a magnetic field ("Zeeman system") and the lattice temperature, has been recently developed [1-5]. In particular, it was shown that in saturation of a homogeneously-broadened magnetic-resonance line at a frequency $\nu_0 \pm \Delta$ which is somewhat larger or smaller than the exact resonance frequency ν_0, the energy difference $h\Delta$ between the resonant and saturating quanta should be absorbed by the spin-spin reservoir, or respectively drawn from it, and this can lead to a strong increase or decrease (including a transition to negative values) of the temperature T_{int} and consequently to an appreciable change in the magnetic resonance line shape. In such a not-strictly-resonant saturation one can expect, in particular, the occurrence of a region of negative absorption (inversion of the magnetic resonance signal) [5] in one wing of the line and an increase in the absorption signal, compared with the equilibrium value, in the other (Fig. 1). So far as we know, there is no direct proof as yet of the suitability of the aforementioned theory for EPR; it was our aim to verify experimentally the applicability of the theory in this case.

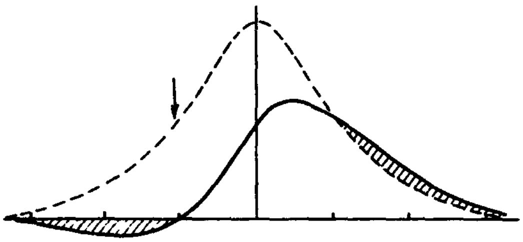

Fig.1. Theoretical line shape in not-strictly resonant saturation. Dashed line - no saturation. The arrow indicates the detuning of the saturating signal relative to the center of the line. The regions of inversion and of increased absorption are cross-hatched.

The experiments were performed in the 3-cm band at a temperature 1.8°K, on single crystals of $K_3Co(CN)_6:Fe^{3+}$ (the iron concentration was ~ 0.6%). This material was chosen because its EPR spectrum consists of only one line (effective spin S' = 1/2) that has no hf structure [6]. The line width $\Delta\nu$ was about 60 MHz; the line is apparently homogeneous, so that the reconstruction of the EPR signal after saturation (at the line center) is described by one exponential (time constant $\tau_1 \approx 40$ msec).

The not-strictly-resonant saturation of the EPR line was effected in our experiments by short (50 - 80 μsec) saturating pulses, and the indication was by means of a continuous weak signal with low-frequency modulation of the magnetic field. The time of passage through the EPR line was several milliseconds; thus, during the time necessary for the indication, the Zeeman system was practically isolated from the lattice. The same cannot be said, however, beforehand concerning the spin-spin reservoir, the temperature of which, which is shifted as a result of the not-strictly-resonant saturation, relaxes to the equilibrium value within some characteristic time τ_1'' which in general is shorter than τ_1. Consequently, the effects due to the strong shift of T_{int} should be most clearly manifest during the first instants after the saturating pulse is turned off. Since these effects (inversion and increase of absorption) are maximal in regions whose distance from the point at which saturation is reached is of the order of the line width (Fig. 1) we have carried out the indication at a frequency ν_{ind} differing from the saturation frequency ν_{sat} by approximately the same amount.

Some of the experimental results are shown in Figs.2a and b, * which show oscillograms of the absorption line at not-strictly-resonant saturation in two cases. Fig. 2a corresponds to a situation wherein the signals at the indication and saturation frequency are on the same side of the line peak at the instant when the saturating pulse terminates, while Fig. 2b corresponds to the case when these signals are on opposite wings of the line ($\nu_{sat} - \nu_{ind} = -1.0\Delta\nu$ and $-1.3\Delta\nu$, respectively). We see that in the former case an inversion region is indeed observed immediately after the end of the pulse, while in the second case a region of increased absorption (compared with the equilibrium value) is observed. Comparison of these results with the theoretical formulas of [5] leads to good agreement of it is assumed that the value of the local magnetic field is $\gamma H_{loc} \approx \Delta\nu/2$. It turns out here that Figs. 2a and b correspond to a temperature $T_{int} \approx 0.012°K$. The processing of the results, carried out with allowance for the change in the temperature T_{int} during time of passage of the indication signal through the line, has made it possible to estimate the time τ_1 of establishment of the equilibrium between the spin-spin reservoir and the lattice, namely $\tau_1' \sim 1 - 2$ msec. Thus, $\tau_1'/\tau_1 << 1$, from which it follows that in the case of continuous not-strictly-resonant saturation the effects connected with the shift of T_{int} will in this case be quite weak. The comparatively small value of τ_1' may be connected with the interaction between the spin-spin reservoir and the cobalt and potassium nuclei, which have appreciable magnetic moments (such a mechanism is considered in [7]).

Experiments were also performed in which isentropic passage through the EPR line was effected from the far wing to the peak under saturation conditions. After the saturation was removed, negative absorption was observed on the same line wing (inversion), thus demonstrating the possibility of a strong shift of the temperature T_{int}. It is important that inversion is attained in this case without passing through exact resonance (the line peak); we are dealing here essentially with the so called demagnetization in a rotating coordinate frame (ADRF), a phenomenon known in NMR [2].

The experiments show that an energy reservoir of spin-spin interaction can actually exist in paramagnetic crystals, and that its temperature can be shifted by using, in particular, the not-strictly-resonant saturation of the EPR line. We note also that in our investigation we obtained negative absorption (Fig. 2a) under conditions that differ from all other hitherto-employed method of obtaining inversion in systems with two Zeeman sublevels ($S = 1/2$).

The authors are deeply grateful to M. I. Rodak who helped with setting up and performing this work, and to M. E. Zhabotinskii and A. V. Frantsesson for help and useful discussions.

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*The photographs referred to here (Figs. 2a and b) were left out of the original Russian issue. They may be included as errata in a future issue. - Transl.

ERRATA

Vol. 6, No. 4. The following photographs were left out of the earlier edition of the Russian original and were not included in the translated version:

Article by T. L. Asatiani et al, (p. 83):

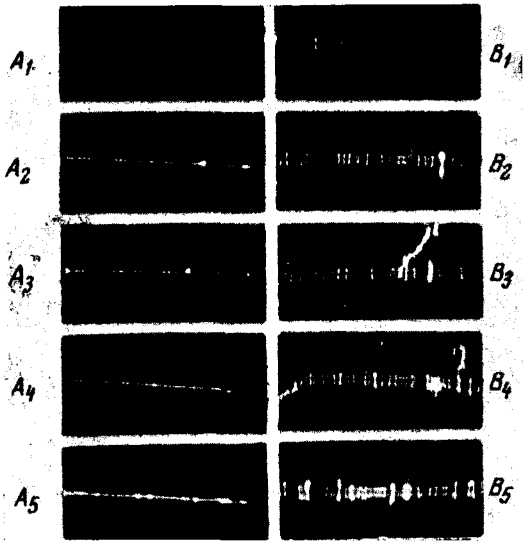

Fig. 1. Photographs of proton tracks in two projections at different energies in the case of a high-voltage pulse duration τ_{III}:
A_1, B_1 - $I/I_{min} = 1.2$; A_2, B_2 - $I/I_{min} = 3.4$;
A_3, B_3 - $I/I_{min} = 5.4$; A_4, B_4 - $I/I_{min} = 6.3$;
A_5, B_5 - $I/I_{min} = 8.5$.

Article by S. A. Akhmanov et al. (p. 85):

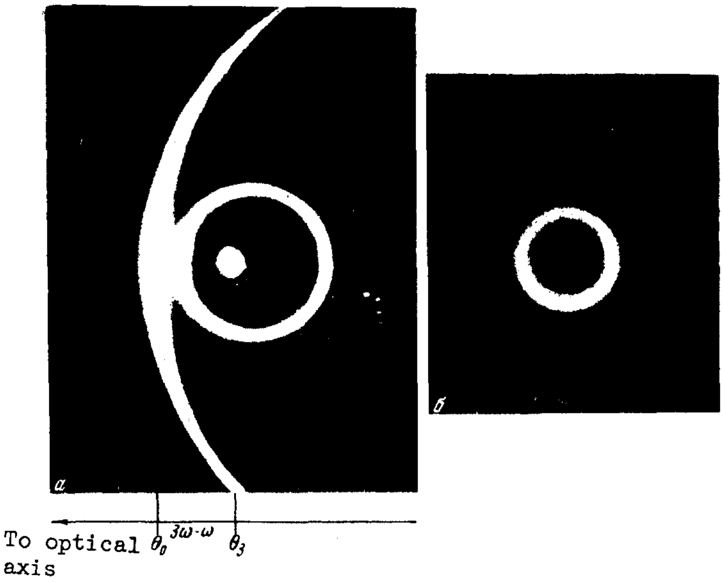

Fig. 2. Space structure in far field: a - emission at difference wavelength $\lambda_2 = 0.53$ μ obtained by mixing the pump and signal at $\lambda_1 = 1.06$ μ (bright spot, ring, and arc) and spontaneous noise (background); b - spontaneous noise passed through interference filter at $\lambda = 5570 \pm 25$ Å; photograph obtained by successive superposition on a single frame of photographs taken at $\theta_3 = \theta_0^{3\omega-\omega} + 5°20'$ (outer ring), $\theta_3 = \theta_0^{3\omega-\omega} + 4°20'$, and $\theta_3 = \theta_0^{3\omega-\omega} + 4°00'$ (two rings coalescing into a single internal one).

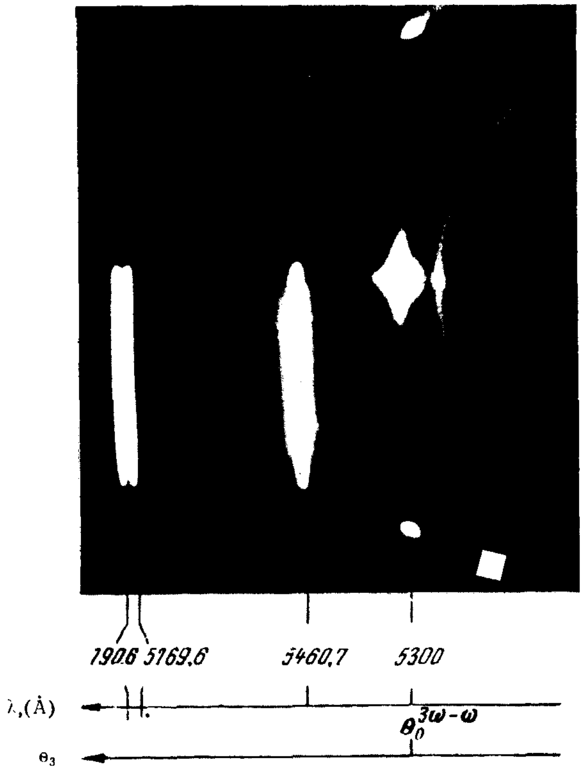

Fig. 3. Spectrograms of spontaneous noise, obtained by superimposing on one frame three spectrograms obtained at angles $\theta_3 - \theta_0^{3\omega-\omega}$ equal to 6°30' (a), 2°55' (b), and 0°35' (c) (arcs from left to right). The reference lines were obtained with a mercury lamp.

Article by V. A. Atsarkin et al. (p. 88):

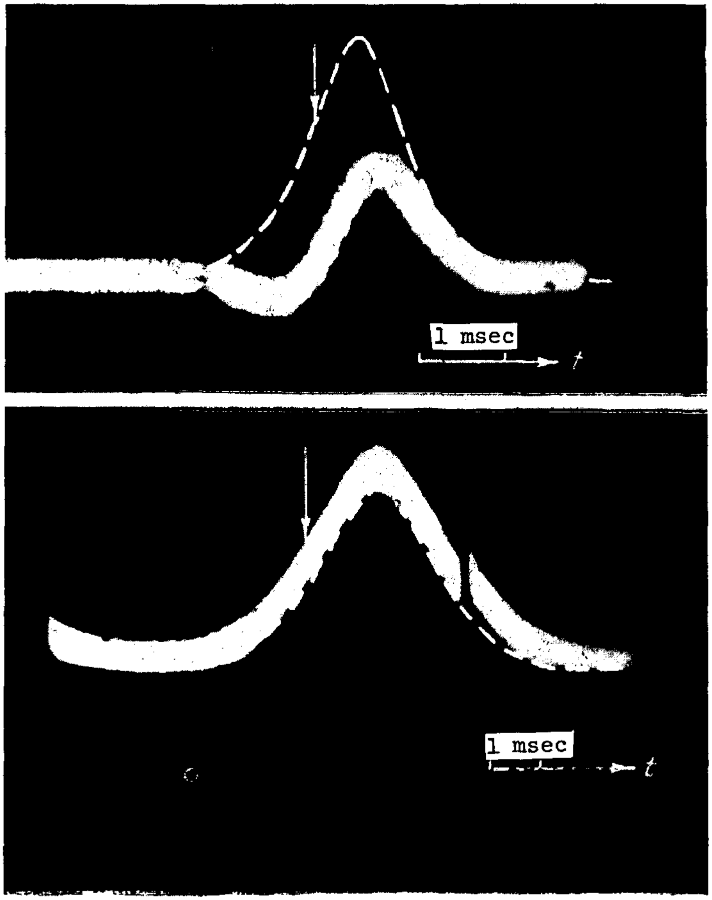

Fig. 2. Oscillograms of EPR line at not-strictly resonant saturation. The notation is the same as in Fig. 1. The magnetic field increases with time in the upper figure and decreases in the lower figure.