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EXPERIMENTAL INVESTIGATION OF THE MANIFESTATIONS OF THE SPIN-SPIN INTERACTION RESERVOIR IN A SYSTEM OF EPR LINES CONNECTED WITH CROSS RELAXATION

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The influence of saturation on the behavior of the EPR absorption spectra was investigated in BaF_2 crystals with admixture of 0.05% of Er^{3+} ions at 1.8°K. The relationship between the absorption in the spectral lines belonging to different magnetic complexes of Er^{3+} and the polarization of the nuclear spins of F^{19} was observed and investigated, including the stimulated spectral diffusion over the EPR spectrum due to saturation of the NMR of F^{19}. It is shown that the entire EPR spectrum of Er^{3+}, including the hyperfine structure due to the isotope Er^{167}, can be described in a manner similar to that of a single homogeneous line, by two temperatures: the Zeeman spin temperature of the center of gravity of the spectrum, and the temperature of the electron spin-spin reservoir; the latter is equal to the Zeeman spin temperature of the F^{19} nuclei. Such an approach explains, in particular, the stationary induced radiation at the outermost lines of the EPR spectrum, which was observed during the course of the experiments.

1. FORMULATION OF THE PROBLEM

IN the study of paramagnetic resonance in crystals, more frequent use is being made of the concept of reservoir of spin-spin interactions[1]; this concept employs two temperatures for the description of the spin system, namely the Zeeman temperature (T_Z) and the spin-spin temperature (T_{SS}).

Recent experimental investigations have shown that this concept is valid and fruitful in EPR, especially in problems connected with the shape of the resonance line under saturation conditions[2,3], cross relaxation[4,5], and dynamic polarization of nuclei[5,6]. Nonetheless, the problem of using the T_{SS} concept in the case of saturation of an inhomogeneous EPR line has not yet been solved. In a number of theoretical papers[7,8] this problem is used by solving the concept of "spin packets" constituting the inhomogeneous line; in others[9,10] the hypothesis of the "local field reservoir" having its own temperature is introduced instead.

As to the experimental study of this problem, the only experiment of this type is so far our own paper[3], in which we succeeded in confirming a number of important predictions of the theory, the object of the investigation did not make it possible to control the width, shape, and saturation factor of the individual "spin packets." We note that the introduction of "spin packets" within the limits of an unresolved line always presupposes a certain leeway in the choice of the parameters connected with the packets, and it is therefore difficult to compare the theory with experiment with sufficient conviction.[1)]

In this paper we undertook to approach the problem of saturation of the inhomogeneous line with the aid of an experiment with such paramagnetic crystals in which there exists an entire system of closely-lying but still resolved EPR lines, coupled by cross relaxation. In such a system the form and number of "spin packets" are completely controlled, just as the Zeeman spin temperature of each of them. It was also desirable to be able to vary the relative locations of the individual lines ("packets"), and in addition, to determine the temperature T_{SS} by using the direct thermal contact between the system of the electronic spin-spin interactions (SS) and the nuclear spins of the crystal lattice (the system Z_n)[12,13,5].

2. EXPERIMENTAL PROCEDURE

We chose as the object of the investigation BaF_2 with an admixture of Er^{3+} (concentration f = 0.05%)[2)]. In the crystal employed by us, the EPR spectrum consisted of four lines corresponding to four magnetic complexes Er^{3+}, the magnetic axes of which were directed along the trigonal axis of the unit cell. From the data of[14] it follows that $S = 1/2$, $g_{\|} = 5.94$, and $g_{\perp} = 7.13$ for each of the complexes, so that when working at 3.2 cm wavelength the spectrum falls in the range of magnetic fields ~1000 Oe. We also observed much weaker spectra belonging to the isotope Er^{167} (natural content 23%, nuclear spin $I = 7/2$), having a characteristic hyperfine structure (hfs). In addition to spectra of trigonal symmetry, we observed an isotropic EPR line belonging to Er^{3+} in a cubic surrounding; the number of such centers was about 10%.

The magnetic field H_0 was applied in a plane perpendicular to one of the trigonal axes, and could be rotated about this axis. This has made it possible to change the relative positions of the spectral lines. The spectra observed at certain values of H_0 are shown in Fig. 1.

[1)] By way of an example we can cite [11], where all of three free parameters were used in the packet model, and this of course makes it always possible to obtain the desired results from the theoretical formula.

[2)] The crystal was grown at the Kazan' State University.

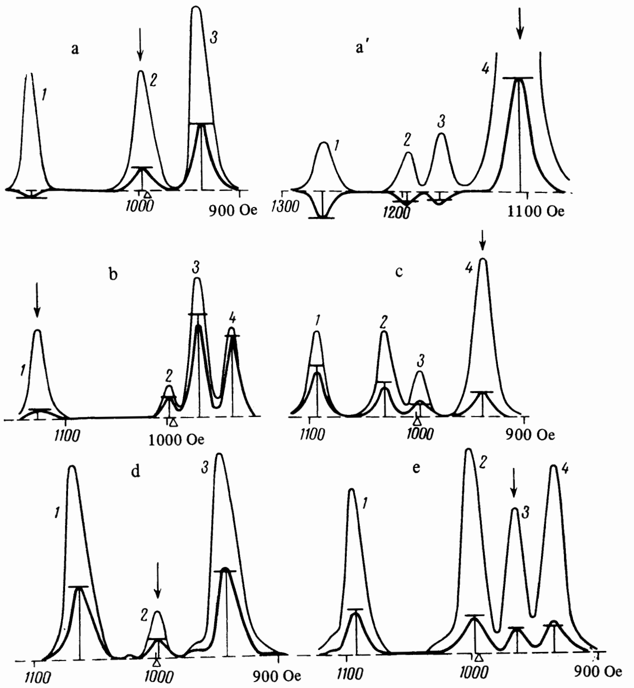

FIG. 1. Experimentally observed EPR absorption spectra of Er^{3+} in BaF_2 at different orientations of H_0 in the (111) plane. Thin lines—spectrum under equilibrium conditions, thick lines—under conditions of saturation of one of the lines. The saturation point is indicated by an arrow. The enhancement E of the polarization of the F^{19} nuclei has the following values: a—E = 2.6; b—E = 4.6; c—E = −3.0; d—E = 0.71; e—E = −0.74. The thick horizontal segments are the results of calculation of the line amplitudes by formula (3). The triangles denote the centers of gravity of the spectra. a′—part of the hyperfine structure due to the Er^{167} isotope at $\varphi = 14°$. The intense line to the right corresponds to the extreme left line of the spectrum a. E = 4.8.

The line near $H_0 = 1$ kOe belongs to the cubic spectrum; the weak spectrum of the isotope Er^{167} is hardly seen in Figs. 1a, b, c, d, e, and is partly shown in Fig. 1a′. Certain differences in the amplitudes of the fundamental four lines (when they do not overlap) are due to the difference in the orientation of the axis of the complexes relative to the microwave field. The experiments were performed at $T_0 = 1.8°K$. Under these conditions the spin-lattice relaxation time of the Zeeman spin system $\tau_{1e} \approx 0.2$ sec[15], and one could expect the cross relaxation within the limits of EPR spectrum to turn out to be much more effectively than the spin-lattice relaxation.

The indication of the EPR absorption spectrum was realized by means of a weak (unsaturating) microwave source under conditions of deep sinusoidal modulation of the magnetic field at 50 Hz. The saturation of the chosen line was carried out by periodic microwave pulses of duration ≈0.5 msec, synchronized with the field modulation. Inasmuch as the period of the modulation (20 msec) was much smaller than τ_{1e}, the saturation was practically stationary. The degree of saturation was monitored by means of the amplitude of the saturated line, indicated during the return passage of the magnetic field (the spectrum was scanned twice during one modulation period).

Simultaneously with indication of the EPR spectrum of Er^{3+}, we observed in the same sample also the NMR of the F^{19} nuclei (nuclear spin 1/2, spectroscopic splitting factor 4.007 Hz/Oe). The Zeeman temperature T_{Zn} of the nuclear spins was determined from the enhancement of the nuclear polarization $E = T_0/T_{Zn}$, which was proportional to the amplitude of NMR absorption line.

3. RESULTS

We investigated five different variants with different orientations of H_0 in the (111) plane, namely at $\varphi = 14$, 0, 24, 35, and 52°, where φ is the angle between H_0 and the projection of the $[1\bar{1}1]$ axis on the (111) plane. In each of the variants, all the EPR lines were saturated in sequence. Some of the experimental results are shown in Fig. 1, which shows also the enhancements E of the nuclear polarization.

A cursory examination of Figs. 1a, b, c, d, and e may give the impression that the differences in the degrees of saturation transfer from the directly saturated line to the remaining lines of the spectrum can be explained within the framework of the traditional concepts of "spectral diffusion" without allowance for the SS reservoir[16]. In fact, if we introduce the quantity $k_i = P_i/P_{i0}$, where P_i is the amplitude of the i-th line under conditions when the j-th line is saturated, and P_{i0} is the amplitude of the i-th line under equilibrium conditions, then it can be concluded from Figs. 1b and 1c that k_i increases monotonically with increasing distance from the saturation point. A similar growth of k_i is observed also for the lines 3, 2, and 1 of Fig. 1e. However, it can be noted that k_4 is somewhat smaller than k_3, and the effect observed in Fig. 1a cannot be explained at all from the point of view of the traditional concepts of "spectral diffusion." Here, when the line 2 is saturated, k_1 turns out to be not only smaller than k_2, but becomes negative, i.e., population inversion and stimulated emission takes place in line 1. A similar effect was observed also in the hfs lines of the isotope Er^{167} (see Fig. 1a′).

The inversion was even larger when the saturating pulses were shifted rapidly during the course of the experiment from the outermost lines of the spectrum to its middle. Thus, if the line 1 was first saturated (Fig. 1a), and then the saturation point was transferred after a time ~ 0.1 sec to line 2, then the value of k_1 at the first instant following was approximately −0.15, and only after a fraction of a second did it approach its stationary value (−0.06).

An analysis of the results has shown that the "anomalies" in the behavior of the values of k_i occurred for lines located farther from the center of gravity of the entire spectrum than the saturation point, but on the same side. The centers of gravity, calculated from the relation $\sum_i \delta_i n_i = 0$, where δ_i is the deviation of the center of the i-th line from the center of gravity of the spectrum and n_i is the number of ferromagnetic centers contributing to the i-th line, are shown in Figs. 1a, b, c, d, e by triangles.

We investigated also the dependence of the polarization of the F^{19} nuclei on the deviation δ_j of the saturating pulse from the center of gravity of the spectrum. The results are shown in Fig. 2, where each experimental point corresponds to saturation of the center of some

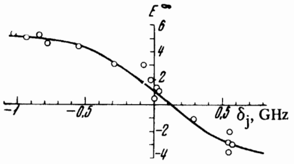

FIG. 2. Enhancement of the polarization of the nuclei F^{19} at the maximum saturation factor (E^{∞}) against the deviation (δ_j) of the saturation point relative to the center of gravity of the EPR spectrum. Points—experimental values, curve—result of calculation by means of formula (4) at $\delta_0 = 1.05$ GHz.

line. The results for all the investigated variants are shown on the common plot, since no correlation was observed between the orientation of H_0 and the dependence of E on δ_j. In order to facilitate the subsequent analysis, Fig. 2 shows the values of E^{∞} obtained by extrapolation of the experimental results to the case of limiting saturation of the line j (we use here the experimental dependence of E on the power of the saturating pulses). At the maximal experimentally reached saturating factors (5—10) the values of E were 10—20% lower than those of E^{∞}. A detailed analysis of Fig. 2 will be given in the next section; we note here that $E \approx 1$ in the case of saturation near the center of gravity.

4. COMPARISON OF EXPERIMENT WITH THEORY

The "anomalies" in the behavior of the values of k_i, noted in the preceding section, are quite similar to the course of the absorption coefficients on the wing of a homogeneous magnetic-resonance line if the line is not strictly resonantly saturated, a course predicted theoretically on the basis of the T_{SS} concept[17] and experimentally observed recently in EPR[2,5] and NMR[18]. The similarity is confirmed by comparing the effects occurring when the saturation point is shifted (see the preceding section) with isotropic passage of a homogeneous line under saturation conditions.

All this has induced us to attempt to describe the entire aggregate of cross-relaxation-coupled lines of Er^{3+} by means of only two temperatures, T'_Z and T'_{SS}, in a manner used for a single homogeneous line. Such a possibility is based also on the results of a theoretical calculation[8] of the cross relaxation of first order in a system of several resonant lines, performed under the assumption that a single SS reservoir exists for all the lines.

Let us assume, by analogy with the homogeneous line, that the absorption within the limits of the entire spectrum is described by a single expression

$$P(\delta) \infty g(\delta)\left(\frac{\nu_0}{T_z'}+\frac{\delta}{T_{ss}'}\right), \tag{1}$$

where $g(\delta)$ is the shape factor of the entire aggregate of lines regarded as a single inhomogeneous line, ν_0 is the frequency of the center of gravity of the spectrum, and δ is the running deviation from ν_0. Since in the absence of saturation we have $T'_Z = T'_{SS} = T_0$, we write k_i in the form

$$k_i=\frac{P(\delta_i)}{P_0(\delta_i)}=(\nu_0+\delta_i)^{-1}\left(\nu_0\frac{T}{T_z'}+\delta_i\frac{T_0}{T_{ss}'}\right). \tag{2}$$

Unlike a homogeneous line, where the temperature T_Z corresponds to a real Zeeman system, the quantity T'_Z now pertains to the center of gravity of the spectrum, where there can be no resonant lines at all, and therefore an experimental determination of T'_Z is difficult. The quantity T'_Z can be readily eliminated by expressing k_i in terms of k_j ($j \neq i$):

$$k_i=k_j\frac{\nu_0+\delta_j}{\nu_0+\delta_i}+\frac{T_0}{T_{ss}'}\frac{\delta_i-\delta_j}{\nu_0+\delta_i} \tag{3}$$

Using relation (3), we can determine T'_{SS} from any two experimental values of k_i and k_j, and then calculate k_i for the remaining lines and compare them with the experimental data. On the other hand, we can attempt to determine T'_{SS} from measurements of the nuclear polarization of F^{19}, assuming that T'_{SS} is equal to the true temperature of the spin-spin reservoir T_{SS}, and the latter, just as in ruby crystals[5], is equal to the spin temperature of the lattice nuclei T_{Zn}. Substituting then in (3) the measured value of E in place of T_0/T'_{SS}, we can calculate from a single value of k_j the amplitudes of all the remaining spectral lines.

We used the latter method, and always chose for the determination of k_j the line directly subjected to saturation. The results of the calculation are shown by the heavy horizontal segments in Fig. 1. We see that in all cases there is undisputedly good agreement between theory and experiment, when account is taken of the accuracy in the determination of E (approximately 10%). A similar agreement with (3) was obtained also when all the remaining lines were saturated in the variants of Fig. 1a, b, c, d, e.

Thus, it can be concluded that, first, the entire aggregate of lines can in our case actually be described in terms of two temperatures, T'_Z and T'_{SS} and, second, the temperature T'_{SS} is equal to the Zeeman temperature of the lattice nuclei T_{Zn} and obviously is the true temperature of the spin-spin reservoir T_{SS}.

The agreement between the calculation by means of formula (3) and experiment shows also that the monotonic growth of k_i with increasing distance from the saturation point (Fig. 1b, c) is not connected with a decrease in the rate of cross relaxation, as may appear at first glance, and is due to deviation of the SS reservoir from equilibrium, just as the "anomalous" course of k_i in Figs. 1a, 1a′, and 1e.

We note that usually $|\delta_i| \ll \nu_0$, thus ridding formulas (1)—(3) of an excessive parameter, namely the exact position of the center of gravity of the spectrum.

We proceed now to analyze the results shown in Fig. 2. Continuing the analogy with the homogeneous line, let us attempt to describe the dependence of $T_0/T_{SS} = E$ on δ_j by means of the formula[17]:

$$\frac{T_0}{T_{ss}}=-\frac{\nu_0}{\delta_j}\left(1+\frac{\delta_0^2}{\delta_j^2}\right)^{-1}+\left(1+\frac{\delta_j^2}{\delta_0^2}\right)^{-1}. \tag{4}$$

In the case of a homogeneous line, the parameter is $\delta_0 = (\gamma H_L)^2\tau_{1e}/\tau'_{1e}$, where $(\gamma H_L)^2$ is the mean square of the local fields due to the spin-spin interaction and τ'_{1e} is the time of the spin-lattice relaxation of the *SS* system. It is shown in[8] that for an inhomogeneous line made up of spin packets the formula (4) may remain in force if the cross-relaxation time is $\tau_{cr} \ll \tau_{1e}, \tau'_{1e}$, with

$$\delta_0^2=(\gamma H_L)^2\tau_{1e}/\tau_{1e}'+M_2^*, \tag{5}$$

where M_2^* is the second moment of the spin distribution over the packets relative to the center of gravity of the spectrum.

It is seen from Fig. 2 that when $\delta_0 = (1.05 \pm 0.1)$ GHz the experimental points fit satisfactorily the theoretical curve. To compare δ_0 with formula (5), we calculated M_2^* directly; it turned out that for the variants shown in Figs. 1a, b, c, d, e, the value of $(M_2^*)^{1/2}$ calculated without allowance for the hfs fluctuates between 0.52 and 0.6 GHz. Since in our case $\gamma H_L \sim 50$ MHz (this estimate was obtained by calculating the second moment of the homogeneous EPR line[19]), formula (5) is at first glance not confirmed.

The situation changes radically if we include in the calculation of M_2^* the hfs of the isotope Er^{167}, with constants[14] $A = 2.05 \times 10^{-2}$ cm^{-1} and $B = 2.46 \times 10^{-2}$ cm^{-1}. Allowance for these lines, which have low intensity but fall in the region of large $|\delta|$, yields $M_2^* = 1.0$ GHz, which coincides with δ_0 within the limits of measurement accuracy. Thus, formula (5) is confirmed if it is assumed that the entire hfs spectrum is also subject to effective (compared with the spin-lattice interaction) cross relaxation, and consequently is described by the same formulas (1)—(3). This is confirmed by Fig. 1a′, which shows the part of the hfs situated to the left of the main spectrum at $\varphi = 14°$ (line 4 of Fig. 1a′ corresponds to line 1 of Fig. 1a). It is interesting that in this case the inversion coefficient on the line 1 (Fig. 1a′) reaches -0.5, this being due to the large value of $|\delta_i - \delta_j|$ and the closeness of δ_j to δ_0.

5. COUPLING OF NUCLEAR SPINS WITH THE SS RESERVOIR

In the preceding sections we regarded the nuclear spin system of F^{19} only as a "thermometer" with which to determine the temperature T_{SS}. We now stop to discuss phenomena directly connected with the mechanism of the thermal contact of the SS and Z_n systems, namely stimulated spectral diffusion and spin-lattice relaxation of the nuclei.

Stimulated spectral diffusion is defined as the action of the resonant saturation of this system Z_n on the course and on the result of the cross relaxation in the EPR spectrum[5,20]. A decrease of the quantity $|T_0/T_{Zn}|$ (i.e., depolarization of nuclei), occurring under the saturation of the NMR, should lead to a corresponding decrease of T_0/T_{SS}, which in turn contributes to equalization of the Zeeman spin temperatures over the EPR spectrum (in our notation—to equalization of the values of k_i).

The effect of stimulated spectral diffusion is demonstrated in Fig. 3, which shows the result of the saturation of NMR of F^{19} under conditions of simultaneous saturation of line 2 of the EPR spectrum of Er^+ ($\varphi = 14°$, $k_2 = 0.22$). We see that in NMR saturation the inversion of the line 1 is replaced by positive absorption; the quantity k_3, to the contrary, decreases. Thus, as a result of saturation of the NMR k_1 and k_3 come close to k_2, T_{SS}^{-1} tends to T_{Zn}^{-1}, which in turn is close to zero. In our case, where initially $T_0/T_{SS} = 2.78$ (Fig. 3a), we have $T_0/T_{SS} = 1.84$ after NMR saturation (Fig. 3b; T_{SS} is calculated from formula (3)).

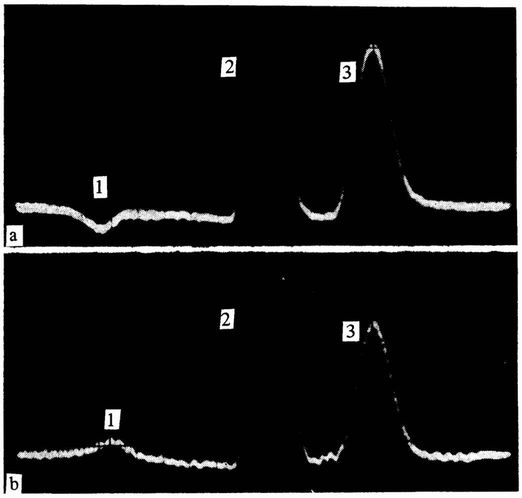

FIG. 3. Oscillograms of the EPR spectrum of Er^{3+}: a—saturation of one of the EPR lines (second on the left); b—simultaneous saturation of the NMR of F^{13}. The orientation of H_0 is the same as in Fig. 1a.

We also measured the time of spin-lattice relaxation of the F^{19} nuclei, namely $\tau_{1n} = 0.38$ sec. If it is assumed that the nuclear relaxation proceeds via thermal contact with the SS system, then in the absence of electronic cross relaxation $\tau_{1n} = \tau'_{1e}(c_n + c_{SS})/c_{SS}$, where c_n and c_{SS} are the specific heat of the Z_n and SS systems[13]. The electronic cross relaxation produces an additional way of relaxing the SS and Z_n system to the lattice, since a connection arises between the SS reservoir and the "difference" electronic Zeeman system, which relaxes to the lattice with a time τ_{1e}. The calculation for the case of cross relaxation between two EPR lines (i and j) shows[5] that if the rate w_{cr} is not bounded, and the "specific heat" of the Zeeman system is $c_\delta \gg c_{SS}, c_n$, then $\tau_{1n} \approx \tau_{1e}$, (according to[5], the "difference" Zeeman system is characterized by the quantity $\nu_i/T_{Zi} - \nu_j/T_{Zj}$).

It can be assumed that this calculation can be generalized to the case of cross relaxation in a system of many EPR lines, and in analogy with the results of the calculation of the saturation of such a system[8], the value of δ_0 determined from (5) is the characteristic frequency for the "difference" Zeeman. In this case $c_\delta \backsim f\delta_0^2$, and for our crystal $c_\delta/c_n = f\delta_0^2/2\nu_n^2 \gg 1$. The measured value $\tau_{1n} = 0.38$ sec indeed turns out to be quite close to $\tau_{1e} = 0.2$ sec; a certain deviation of τ_{1n} from τ_{1e} is apparently due to the limited cross-relaxation rate which introduces a delay $\sim \tau_{cr}c_n/c_{SS}$ in the nuclear relaxation process.

6. CONCLUSIONS

We have verified that the behavior of the group of EPR lines, connected by effective cross relaxation, is described completely, under conditions of saturation of some section of the spectrum, by two temperatures, T'_Z and T_{SS}, the latter being the true temperature of the spin-spin reservoir. This means that any analysis of cross relaxation without allowance for the shift of the temperature T_{SS} should lead to large errors both in the understanding of the nature of the process and in the concrete estimates of the values of τ_{cr}, the "length of spectral diffusion," etc. Moreover, the shift of saturation over the EPR spectrum (and obviously also over the "spin packets" within the inhomogeneous line) does not correspond in general to the simple diffusion equation, inasmuch as the process does not tend at all to equalize the Zeeman spin temperatures of the individual "packets" (the values of k_i). One can consequently speak of spectral diffusion only if the SS reservoir is "closed" on the lattice, i.e., $\tau'_{1e} \ll \tau_{1e}$, or else under conditions of stimulated spectral diffusion (see Sec. 5 of this article).

Notice should be taken of the deep analogy between the arbitrary system of EPR lines, connected by effective cross relaxation, and the single homogeneous line that follows from the applicability in both cases of Eq. (1). In both cases, in particular, it is possible to obtain stimulated emission on the wing sections of the spectrum. However, unlike the case of a homogeneous line, negative absorption in a system of lines (for example, line 1 in Fig. 1a) is connected with the real inversion of the populations on the Zeeman levels. Thus, we have here a new method of obtaining stationary population inversion, different from the usual three-level scheme.

Our results agree fully with the theoretical calculation for the case $\tau_{cr} \ll \tau_{1e}, \tau'_{1e}$[8]. By the same token, the fundamental premises of this calculation, namely the existence of a common SS reservoir for all types of electron spins and neglect of cross relaxations of higher order, have been experimentally confirmed. It is interesting that the "difference" Zeeman system, mentioned by us in Sec. 5, has apparently many properties of the "local-field reservoir"[9] with a characteristic frequency δ_0 and a temperature T_{SS}. The formation of such a reservoir does not contradict, therefore, the existence of "spin packets," and, to the contrary, is a result of cross relaxation between them.

In this paper we have used a recently proposed[5] method of measuring T_{SS} and monitoring cross relaxation by observing the nuclear spin of the lattice, and demonstrated the great possibilities of this method. Obviously, equality of the temperatures T_{Zn} and T_{SS} is not an exclusive property of ruby, and has a more general character.

We assume that the results obtained in this paper, pertaining to the extremely generalized case of an arbitrary set of spin packets, together with the previously published data[2-6,20,21] will make it possible to regard the question of the applicability of the concept of the spin-spin temperature in EPR as finally solved.

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[1] B. N. Provotorov, Zh. Eksp. Teor. Fiz. 41, 1582 (1961) [Soviet Phys.-JETP 14, 1126 (1962)]; Fiz. Tverd. Tela 4, 2940 (1962) [Sov. Phys.-Solid State 4, 2155 (1963)].

[2] V. A. Atsarkin and S. K. Morshiev, ZhETF Pis. Red. 6, 578 (1967) [JETP Lett. 6, 88 (1967)].

[3] V. A. Atsarkin, Zh. Eksp. Teor. Fiz. 58, 1884 (1970) [Soviet Phys.-JETP 31, 1012 (1970)].

[4] R. L. Kyhl and B. D. Nageswara-Rao, Phys. Rev. 158, 284 (1967).

[5] V. A. Atsarkin, A. E. Mefed, and M. I. Rodak, ZhETF Pis. Red. 6, 942 (1967) [JETP Lett. 6, 359 (1967)]; Phys. Lett. 27A, 57 (1968); Zh. Eksp. Teor. Fiz. 55, 1671 (1968) [Soviet Phys.-JETP 28, 877 (1969)].

[6] W. Th. Wenckebach, T. J. B. Swanenburg, H. Hoogstraate, and N. J. Poulis, Phys. Lett. 26A, 203 (1968). W. Th. Wehckebach, G. M. van den Heuvel, H. Hoogstraate, T. J. B. Swanenburg, and N. J. Poulis, Phys. Rev. Lett. 22, 581 (1969).

[7] L. L. Buishvili, M. D. Zviadadze, and G. R. Khutsishvili, Zh. Eksp. Teor. Fiz. 56, 290 (1969) [Soviet Phys.-JETP 29, 159 (1969)].

[8] M. I. Rodak, Fiz. tverd. Tela 12, 478 (1970) [Soviet Phys.-Solid State 12, 371 (1970)].

[9] S. Clough and C. A. Scott, J.of Phys. C.,1,919(1968).

[10] N. S. Bendiashvili, L. L. Buishvili, and M. D. Zviadadze, Zh. Eksp. Teor. Fiz. 58, 597 (1970) [Soviet Phys.-JETP 31, 321 (1970)].

[11] C. F. Hwang and D. A. Hill, Phys. Rev. Lett. 19, 1011 (1967).

[12] M. I. Kozhushner and B. N. Provotorov, in: Radiospektroskopiya tverdogo tela (Solid State Radiospectroscopy), Atomizdat, 1967, p. 5.

[13] L. L. Buishvili, Zh. Eksp. Teor. Fiz. 49, 1868 (1965) [Soviet Phys.-JETP 22, 1277 (1966)].

[14] A. A. Antipin, I. N. Kurkin, L. D. Livanova, and L. Z. Povtorova, and L. Ya. Shekun, Fiz. Tverd. Tela 8, 2664 (1966) [Sov. Phys.-Solid State 6, 2130 (1967)].

[15] A. A. Antipin, A. N. Katyshev, I. N. Kurkin, and L. Ya. Shekun, ibid. 9, 1370 (1967) [9, 1070 (1967)].

[16] A. M. Portis, Phys. Rev. 91, 1070 (1953).

[17] M. I. Rodak, Fiz. Tverd. Tela 6, 521 (1964) [Sov. Phys.-Solid State 6, 409 (1964)].

[18] A. E. Mefed and M. I. Rodak, Zh. Eksp. Teor. Fiz. 59, 404 (1970) [Soviet Phys.-JETP 32, 220 (1971)].

[19] C. Kittel and E. Abrahams, Phys. Rev. 90, 238 (1953).

[20] V. A. Atsarkin, Fiz. Tverd. Tela 12, 1775 (1970) [Soviet Phys.-Solid State 12, 1405 (1971)].

[21] V. A. Atsarkin and M. I. Rodak, ibid. 11, 613 (1969) [11, 493 (1969)].

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