Physics, Siberian Branch, Academy of Sciences of the USSR, Novosibirsk, 1969.
[15]A. V. Gurevich, R. A. Salimov, and N. S. Buchel'nikova, Teplofiz. Vys. Temp. 7, 852 (1969).
[16]O. A. Zolotovskiĭ, V. I. Koroteev, R. Kh. Kurtmullaev, and V. N. Semenov, Dokl. Akad. Nauk SSSR 197, 564 (1970) [Sov. Phys. Dokl. 16, 223 (1971)].
[17]G. E. Vekshteĭn and R. Z. Sagdeev, Pis'ma Zh. Eksp. Teor. Fiz. 11, 297 (1970) [JETP Lett. 11, 194 (1970)].
Translated by S. Chomet

Effective fields at diamagnetic impurities in rare-earth metals

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The hyperfine fields at the nuclei of impurity ^{119}Sn atoms in metallic Tb were measured in the regions of ferromagnetic and antiferromagnetic ordering in the temperature range 4.2–235°K. A strong deviation (up to 90%) of the plot of the temperature dependence of the hyperfine field from the plot of the spontaneous magnetization of the matrix is observed. A comparison with analogous measurements for the Dy matrix shows that the exchange constants can depend on the helicoid angle α. To explain the temperature anomaly, we consider, besides the previously discussed mechanisms, also a new mechanism connection with the rotation of the moments of the nearest environment of the impurity atom.

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The great variety of magnetic structures and the many magnetic properties of heavy rare-earth metals are attributed to the indirect exchange interaction of localized magnetic moments of the unfilled $4f$ shell via the conduction electrons and to interaction with the crystal field. According to the Ruderman-Kittel-Kasuya-Yosida (RKKY) model, the exchange interaction has an oscillating long-range character and leads to polarization of the conduction electrons. Information on the polarization of the conduction s electrons can be obtained from the magnetic field induced by them at the nuclei of a diamagnetic impurity. The investigation of the magnetic field at the nuclei of impurity tin in heavy rare-earth metals, carried out by the γ-resonance method, shows that the temperature dependence of these fields differs substantially from the magnetization curve for the matrix in the case of the Dy and Ho matrices.[1] An explanation of the observed anomalies within the framework of the s-f interaction model cannot be obtained without additional assumptions. A similar anomalous temperature dependence of the magnetic field at the impurity atom was confirmed in an investigation of the hyperfine interaction for another diamagnetic impurity (Cd) in the same matrices, carried out by the method of disturbed γ-γ correlations.[2,3]

In the present study we have used the γ-resonance method to perform more detailed measurements of the temperature dependence of the magnetic fields at Sn impurity nuclei in metallic Tb and Dy. The obtained data point to a complicated connection between the hyperfine field and the rotations of the moments on going from the collinear to the helicoidal type of order. We consider the possible mechanisms of the observed strong temperature anomalies of the hyperfine fields, and, in particular, the mechanism connected with the fact that the matrix ions have an orbital angular momentum.

EXPERIMENTAL PROCEDURE

The preparation and investigation of polycrystalline Dy samples with 0.3–0.5 at.% Sn impurity, enriched to 86.9% Sn^{119}, is described in[4]. An analogous procedure was used also for Tb samples with 0.5 at.% Sn impurity, with additional measures taken to increase the accuracy of the temperature measurements, especially within the limits of the narrow antiferromagnetic region[5] (221–229 °K). To this end, a semiconducting resistance thermometer of the "KG" type was clamped with the aid of a beryllium spring to the center of the sample, where a chromel-gold thermocouple (0.4 at.% Fe) was also located and connected in an automatic-control circuit. In addition, to exclude the influence of parasitic thermal emf's, the readings of the pickup at opposite directions of the current were averaged. The temperature was maintained constant within ≤ 0.1 °K, and the temperature gradient over the sample did not exceed 0.5 °K.

For investigations in a longitudinal magnetic field we used an installation with a superconducting solenoid that produced a field of intensity up to 40 kOe at a sample temperature of 4.2 °K.

RESULTS OF EXPERIMENTS

The characteristic form of the Mössbauer spectra in the case of Dy is shown in Fig. 1. The spectra have a complicated structure whose character changes qualitatively near the ferromagnetism–antiferromagnetism phase transition. The spectrum at 84 °K was obtained approximately a year after the others, and shows enhancement of the central part of the line spectrum, due

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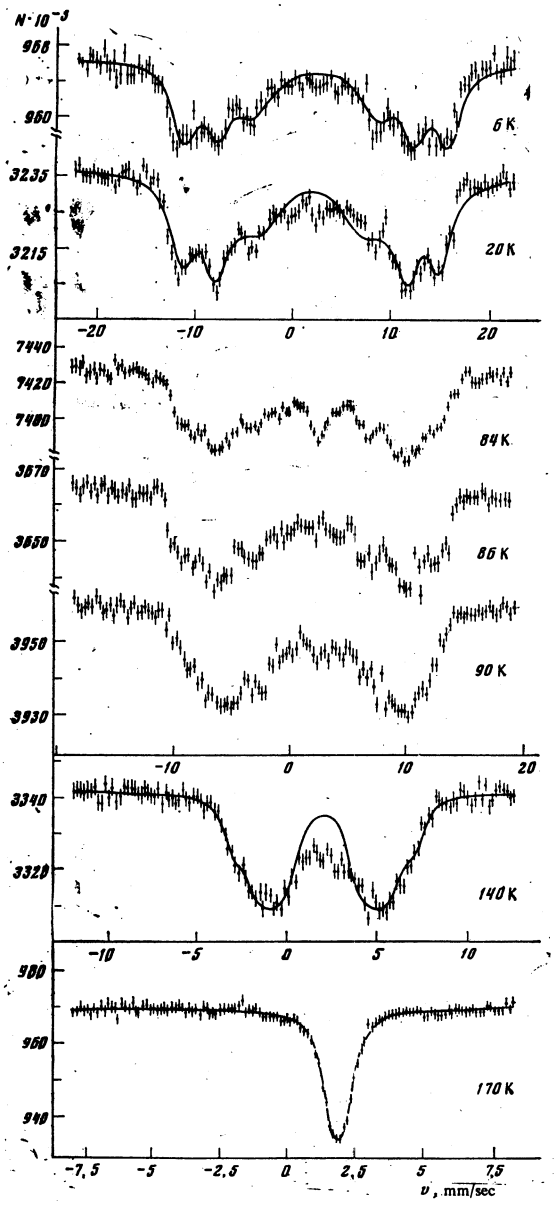

FIG. 1. Mössbauer spectra of Dy+0.5 at.% Sn at Sn^{119} nuclei. The solid lines are the result of the computer reduction. The spectrum at 84 °K was obtained a year later.

to the appearance of other phases in the sample. The models used for computer reduction of these complicated spectra are described in detail in[4]. The results of the reduction reduce mainly to the following.

1) In the ferromagnetic region there are two values of the hyperfine field, H_1 and H_2.

2) In the phase transition from the collinear to the helical structure near T_c, a large set or a continuous distribution of hyperfine fields in a certain interval from H_{min} to H_{max} is produced rather than two discrete values of the field H_1 and H_2.

The reason why it is correct to change the set of parameters that describe the spectra on going through T_c is the abrupt change in the form of the spectrum with changing type of ordering of the moments. The temperature dependence of the more intensive field H_1 (the relative intensity of the field H_2 changes from 0.4 at 6 °K to 0.8 at 86 °K and $H_2 < H_1$) and of the average field $H_{av} = (H_{max} + H_{min})/2$ is shown in Fig. 2. It is seen that in the ferromagnetic region the curve for H_1 deviates somewhat from the spontaneous magnetization curve of the pure matrix, and in the antiferromagnetic region the temperature dependence of H_{av} differs strongly from the magnetization curve and from the Brillouin function for a spin $\frac{5}{2}$. Near the phase-transition temperature T_c the abrupt change in the course of the experimental points characterizes the average change of the field H_1, which is equal to $\overline{\Delta H_1} = H_1 - H_{av}$.

The Mössbauer spectra for the case of Tb are shown for several temperatures in Fig. 3. The spectra given for 4.2 °K were obtained in an external magnetic field of 35 kOe and without a field in the same installation. In the paramagnetic temperature region (> 229 °K) a single line of width 1.04 ± 0.02 mm/sec is observed, just as in the case of the Dy matrix. In the narrow antiferromagnetic region, the single line broadens gradually with decreasing temperature, and in the ferromagnetic region (< 221 °K), at sufficiently low temperatures, the spectrum is split completely into six components.

The computer reduction of the spectra was carried out within the framework of the model of one sextet of Lorentz lines. The parameters in the ferromagnetic region were the field H, the line width Γ, and the isomeric shift δ. The value of H at 4.2 °K is 235 ± 1 kOe. It was found that the values of Γ increase from 1.5 mm/sec at 4.2 °K to 2.0 mm/sec at ~150 °K. On approaching the antiferromagnetic region, Γ increases somewhat to above 2.0 mm/sec. On the other hand, the width of the Mössbauer spectrum itself in a region somewhat higher than T_c is 1.66 mm/sec. It is obvious that it is impossible to describe the spectra in this region by using a value $\Gamma \geq 2$ mm/sec, and it is necessary to recog-

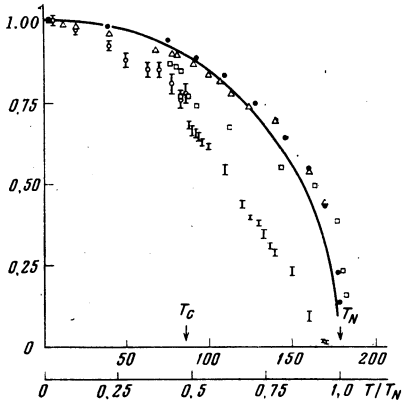

FIG. 2. Temperature dependences in Dy: ○—of the hyperfine field at the Sn^{119} nuclei in the ferromagnetic region H_1; the vertical sections are the average values of the hyperfine field at Sn^{119} nuclei in the antiferromagnetic region, H_{av}; ●—of the hyperfine field at the Dy^{161} nuclei,[6] △—of the spontaneous magnetization,[7] □—of the hyperfine field at Cd^{111} nuclei[2]; solid curve—Brillouin function for $(g-1)J = \frac{5}{2}$.

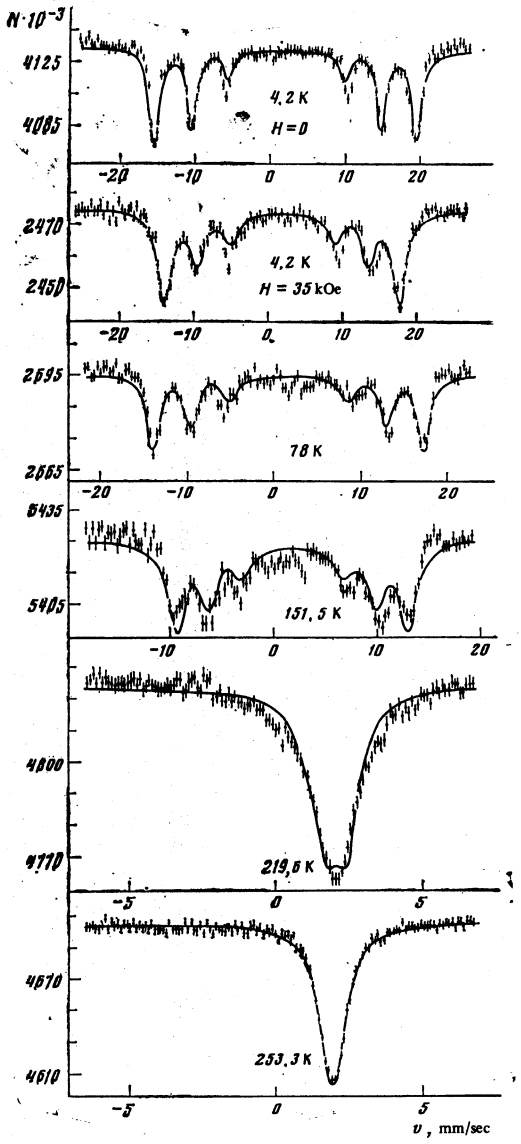

FIG. 3. Mössbauer spectra of Tb ± 0.5 at.% Sn at the Sn^{119} nuclei. Solid lines—results of computer reduction. The spectrum in an external field $H = 35$ kOe is shown additionally for 4.2 °K.

nize that Γ decreases sharply going through T_c. To describe the spectra in the antiferromagnetic region in the single-sextet model we have therefore fixed the parameter Γ at the 1.04 mm/sec level (just as in the paramagnetic region). This makes it possible to estimate the maximum value of the field in the antiferromagnetic region, which, as was found, decreases with increasing temperature from 6 kOe to zero.

Thus, in the case of Tb, just as in the case of Dy, it was necessary, on goint through the Curie point, to change the set of parameters needed to describe the spectra, thus attesting to the discontinuous character of the variation of the hyperfine interactions in the investigated phase transitions.

The results of the measurements of the hyperfine fields on impurity Sn in Tb at different temperatures

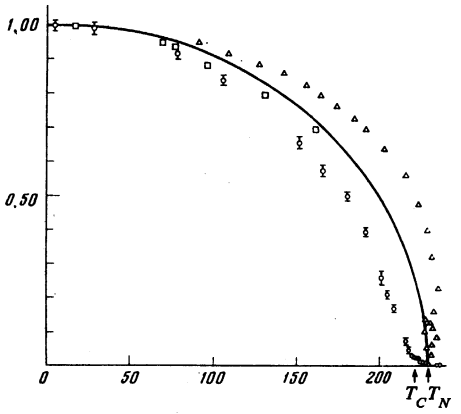

FIG. 4. Temperature dependences in Tb: ○ —of the hyperfine field at the Sn^{119} nuclei, △ —of the spontaneous magnetization[8], □— of the hyperfine field at Cd^{111} nuclei[9]; solid curve—Brillouin function for $(g-1)J=2$.

are shown in Fig. 4. The same figure shows the Brillouin function for spin 3, referred to $T_N = 229$ °K, and the spontaneous magnetization of Tb, which vanishes according to[8] in the temperature interval 230 ± 4 °K. It is seen that the observed temperature dependence differs strongly from the magnetization of the pure matrix, and also from the Brillouin function, and a rather abrupt inflection of the curve is observed near T_c. The error in the hyperfine field is the sum of the error due to the inaccuracy of the spectrometer velocity calibration and the error corresponding to the maximum permissible changes of the parameter H within the limits of the employed model.

The sign of the hyperfine field at an Sn nucleus in the Tb matrix, as expected, was found to be negative (Fig. 3). The isomeric shift relative to the compound $BaSnO_3$ amounts to 1.91 and 1.83 mm/sec for Dy and Tb, respectively.

For convenience in the analysis of our results, we represent them in a generalized form, as shown in Fig.

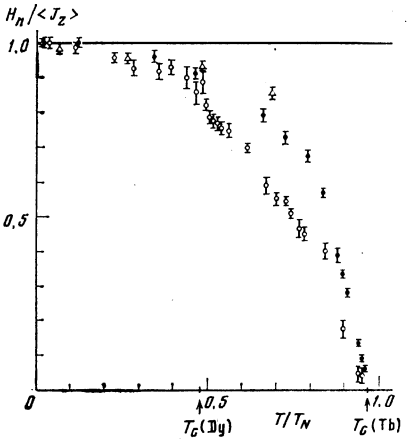

FIG. 5. Dependence of the relative field at Sn^{119} nuclei on the relative temperature: ○ —Dy; ● —Tb, present paper; △—Gd, according to the data of [10,11] (the abscissas represent T/T_c). Straight line—form of this dependence in the absence of temperature anomalies of the hyperfine field.

5. The abscissas represent here the relative temperature T/T_N in the same scale for Tb and Dy, while the ordinates represent the relative field $H_n/\langle J_z\rangle$, determined from Figs. 2 and 4. The curve for Tb terminates near T_c/T_N, for the lack of exact data for the spontaneous magnetization of Tb near T_N.

DISCUSSION OF RESULTS

We start with the premise that the magnetic field H_n at the atomic nucleus of a diamagnetic impurity in a rare-earth metal, induced as a result of s–f exchange, be proportional to the polarization of the conduction electrons at the site $\mathbf{r}_0$ occupied by the impurity:

$$H_n(T)=2A(Z)(g-1)\sum_{\mathbf{R}}A_{s-f}(\mathbf{r}_0-\mathbf{R})\langle J_z\rangle_{\mathbf{R}}. \quad (1)$$

The proportionality coefficient $A(Z)$ is determined here by the mechanism whereby the hyperfine field is produced for an impurity with an atomic number Z, g is the g factor, $A_{s-f}(\mathbf{r}_0-\mathbf{R}_0$ is an integral that depends on the exchange interaction between the f electrons localized at the lattice sights R and the conduction electrons from the nonlocal electron susceptibility $\chi(\mathbf{r}-\mathbf{r}_0)$; the quantity $\langle J_z\rangle_{\mathbf{R}}$ is the temperature-averaged projection of the total angular momentum and determines the spontaneous magnetization at the temperature T:

$$\langle J_z\rangle_{\mathbf{R}}=\sum_{J_z=-J}^{J}J_z\exp(-\mathcal{H}/kT)\left(\sum_{J_z=-J}^{J}\exp(-\mathcal{H}/kT)\right)^{-1}; \quad (2)$$

$\mathcal{H}$ in (2) is the interaction Hamiltonian for a rare-earth ion containing an exchange interaction, which can be written in the molecular-field approximation in the form

$$\mathcal{H}_{ex}=-2(g-1)^2\mathbf{J}_{\mathbf{R}'}\sum_{\mathbf{R}}A_{f-f}(\mathbf{R}-\mathbf{R}')\mathbf{J}_{\mathbf{R}}, \quad (3)$$

and the interaction $\mathcal{H}_{cr}$ with the crystal field.

The exchange integrals A_{f-f} are connected with the corresponding s-f exchange integrals by the relation

$$A_{f-f}\sim A_{s-f}^2, \quad (4)$$

which determines the connection between the curve for the hyperfine field (1) and the magnetization curve (2).

In the case of a matrix with collinear ordering of the moments, if $A(Z)$ is independent of the temperature, the two curves should either coincide or be different, depending on whether the integrals $A_{s-f}(r_0-R)$, which determine the relative field $A_n/\langle J_z\rangle$, is constant or temperature-dependent.

It must be emphasized that there is no connection between the corresponding relations (1) and (2) for matrices of $3d$ metals, in which there is also a direct d-d exchange on top of the indirect exchange via the electrons of the mixed conduction band.

It is seen from Fig. 5, that the relative field in the Tb matrix is not constant in an extended region of the collinear ordering of the moments, but decreases rapidly with increasing temperature, and is one order of magnitude lower at T_c than at 4.2 °K. A similar curve for the Sn^{119} impurity in the ferromagnetic Gd matrix, plotted from the data of[11] in Fig. 5, shows that in this matrix the relative field decreases much more slowly. A similar slower decrease of the relative field takes place for diamagnetic impurities of $4sp$, $5sp$, and $6sp$ as in ferromagnetic matrices of $3d$ metals, with exception of the basis of Sb in Fe,[12] where the anomaly is of opposite sign, and Sn in Co,[13] where a reversal of the sign of the hyperfine field is observed at high temperatures.

The anomalies of the curve of Fig. 5 cannot be attributed to matrix perturbation due to the diamagnetic impurity regarded as a magnetic vacancy. The calculations performed in[14–16,3] show that allowance for the magnetic vacancy weakens $\langle J_z\rangle$ of the matrix angular momenta that are adjacent to the impurity, has little influence on the temperature dependence of the local magnetization, and produces practically no distortion in this dependence in a region close to the temperature where the magnetic order vanishes. It was also shown that allowance for the thermal expansion of the matrix lattice, by reducing the data to a constant volume, does not influence the qualitative behavior of the results in the case of Sn in Fe.[17] We note that the curves in Fig. 5 have not been reduced to the constant-volume condition because of the absence of the necessary data. It can be assumed, however, that in our case, too, the existence of anomalies is not due to thermal expansion, all the more since the iron lattice expands with increasing temperature, while the Db and Dy lattices compress along the c axis, whereas the sign of the anomaly is the same in both cases.

The different mechanisms capable of explaining the observed behavior of the hyperfine fields at the nuclei of atoms of diamagnetic impurities in $3d$ and $4f$ metals has been discussed in a number of papers.[17,13,1–3] In the main, the following possible effects have been considered: 1) the change in the density of states of the electrons on the Fermi surface, 2) the dependence of the exchange integrals on the temperature, 3) the change of the electron structure due to the change in the magnetization of the matrix, 4) the influence of thermal oscillations of the impurity atoms, 5) the dependence of the electronic structure near the impurity on the temperature. Unfortunately, the role of the different effects still remains unexplained.

The large deviations of the hyperfine field at the Sn nuclei as function of the magnetization in the case of Dy were attributed in our earlier studies[4,18] to the appearance of helical ordering of the moments of the matrix. The observed deviations were interpreted as changes of the integrals H_{s-f} for the pure matrix. To reconcile these changes with the magnetization curves, it was suggested that besides the s-f exchange there exists also another exchange-interaction, for example via d electrons, which is responsible for the spontaneous magnetization but does not influence the spin density in the region of the impurity nucleus. No account was taken there of the possible variation of the coefficient $A(B)$

and of the electronic structure with changing temperature near the impurity.

It is seen from Fig. 5 that the curves of the relative field in the Dy and Tb matrices practically coincide in the region of the temperatures of collinear ordering of the moments ($0-0.5T/T_N$), while the curve for this matrix lies much lower in the region in the helical structure of Dy. This behavior gives grounds for assuming that the dependence of the hyperfine field on the temperature is practically the same for both matrices over the entire range of magnetic-ordering temperature. The additional decrease of the hyperfine field in the Dy matrix, which is due to rotations of the moments, can then be separated against the background of this dependence. Under these assumptions we can write within the framework of the 3-plane model

$$\frac{[H_n/\langle J_z\rangle]_{\mathrm{Dy,AFM}}}{[H_n/\langle J_z\rangle]_{\mathrm{Tb,FM}}}=\frac{A_0+2A_1\cos\alpha+2A_2\cos 2\alpha}{A_0+2A_1+2A_2} \tag{5}$$

where A_i are the temperature-independent constants of the s-f interaction with moments located in different basal planes. If we use the known value of the helicoid angle α for Dy, then it turns out that it is impossible to account for the experimental points corresponding to the left-hand side of (5) if the coefficients in the right-hand side remain unchanged. On this basis, we must admit either that our assumptions are incorrect or that the coefficients in (5) are themselves dependent on the angle α. In the latter case this serves as evidence in favor of the presence of two-ion exchange-interaction anisotropy in Dy, other possible manifestations of which were discussed in detail in[4].

Turning now to the temperature dependence of the hyperfine field in a rare-earth matrix, we indicate one other mechanism which is peculiar to these matrices and is capable of causing the temperature anomaly. This mechanism is connected with the fact that a diamagnetic impurity should be regarded not only as a magnetic vacancy, but also as a defect in a crystal field. If the effective charge at the site occupied by the impurity differs from the charge of the matrix ions by a value e' and produces a certain additional electric-field gradient $\mathbf{q}_i$, then the Hamiltonian of the interaction for the i-th ion from the nearest environment of the impurity should contain, besides the exchange interaction $\mathcal{H}_{i\,\mathrm{ex}}$ and the interaction $\mathcal{H}_{i\,\mathrm{cr}}$ with the crystal field of the pure matrix, also an additional term v_i' due to the interaction of the quadrupole moment of the electrons of the $4f$ shell with gradient $\mathbf{q}_i$:

$$\mathcal{H}_i=\mathcal{H}_{i\,\mathrm{ex}}+\mathcal{H}_{i\,\mathrm{cr}}+v_i'. \tag{6}$$

If the interaction v_i' is comparable with other interactions that determine the magnetic order, then the angles θ_i and φ_i, which characterize the directions of the moments $\mathbf{M}_i$ and minimize the Hamiltonian (6) will not be the same as in the pure matrix, and this should lead to a strong distortion of the magnetic order around the impurity. In this case one should expect primarily deviations of the magnetic moments of the nearest environment from the collinear ordering in that basal plane in which the impurity is located. This effect should lead to an additional decrease of the hyperfine field at the impurity, since the total polarization of the conduction electrons in the impurity cell, which is proportional to the vector sum of the moments of the nearest environment, will be smaller in this case.

The relative role of the interaction v_i' should increase with increasing temperature as a result of the decrease of the exchange interaction. This circumstance can explain why the anomaly for the rare-earth matrix increases much more than for the case of the $3d$ matrix in the temperature region close to the temperature where the magnetic order vanishes. An argument favoring this mechanism can be the fact that the temperature anomalies are much smaller for the hyperfine fields at the nuclei Sn^{119} and Cd^{111} in the Gd matrix (Fig. 5) and[19]), inasmuch as for Gd^{3+} ions with $L=0$ the interaction with the crystal field plays a much lesser role. If these arguments are valid, then we can expect the considered anomalies not to be very large in the case of the Eu matrix, just as in the case of Gd.

Figures 2 and 4 show data for the temperature dependence of the hyperfine field at Cd^{111} impurity nuclei in the same matrices, obtained in[2,9] by the method of perturbed angular correlations. It is interesting to note that the temperature anomalies for the hyperfine field at Cd are similar in character. In this case the anomalies are also large, but are smaller than for Sn. These differences should be attributed to differences in the charges Z of the impurity ions.

The radical change in the form of the spectrum, observed in the transition from the collinear to the helical ordering and interpreted as a manifestation of a set of hyperfine fields in the region of the helical structure of Dy, was attributed by us in[4,18] to an anisotropic interaction, as a result of which different basal planes become nonequivalent with respect to the hyperfine fields. From the point of view of the proposed mechanism of the impurity-induced rotation of the moments, the nonequivalence of the planes has another reason, connected with the fact that the directions of the moments located in different basal planes make different sets of angles with the gradients of the electric field of the impurity.

To verify the role of the proposed mechanism, numerical calculations and additional experiments are necessary. Until this is done, we prefer not to discuss other details that can be observed in the spectra.

[1]A. S. Kuchma and V. S. Shpinel', Zh. Eksp. Teor. Fiz. **62**, 1054 (1972) [Sov. Phys. JETP **35**, 556 (1972)].
[2]M. Forker and A. Hammesfahr, Z. Phys. **260**, 131 (1973).
[3]M. Forker and J. B. Fechner, Phys. Status Solidi **B65**, k9 (1974).
[4]S. K. Godovikov, M. G. Kozin, V. V. Turovtsev, and V. S. Shpinel, Phys. Status Solidi **B78**, 103 (1976).
[5]K. N. Taylor and M. I. Darby, Physics of Rare-Earth Solids, Chapman & Hall.
[6]D. M. Sweger and R. Segnan, Phys. Rev. **B9**, 3864 (1974).
[7]R. G. Jordan and E. W. Lee, Proc. Phys. Soc. **92**, 1074 (1967).
[8]B. K. Ponomarev and N. I. Moreva, Zh. Eksp. Teor. Fiz.

69, 1352 (1975) [Sov. Phys. JETP 42, 690 (1976)].
[9]B. Jonsson, L. Boström, J. Goldzewski, B. Lindgren, and E. Karlsson, Hyperfine interactions studied in nuclear reactions and decay, Contributed Papers, Upsala, Sweden, 1974, p. 178.
[10]V. Gotthard, H. S. Moller, and R. L. Mössbauer, Phys. Lett. 28A, 480 (1969).
[11]H. E. Nigh, S. Legvold, and F. H. Spedding, Phys. Rev. 132, 1092 (1963).
[12]Le Dang Khoi, P. Veillet, and I. A. Campbell, J. Phys. F 5, 2184 (1975).
[13]T. E. Cranshaw, J. Appl. Phys. 40, 1481 (1969).
[14]S. W. Lovesey and W. Marshall, Proc. Phys. Soc. 89, 613 (1966).
[15]H. P. van de Braak and W. J. Caspers, Z. Phys. 200, 270 (1967).
[16]H. P. van de Braak and W. J. Caspers, Phys. Status Solidi 24, 733 (1967).
[17]D. C. Price, J. Phys. F 4, 639 (1974).
[18]S. K. Godovikov. M. G. Kozin, V. V. Turovtsev, and V. S. Shpinel', Nineteenth All-Union Conference on Low Temperature Physics, Minsk, 1976, Abstracts of Papers, p. 568.
[19]L. Boström, G. Liljegren, B. Jonsson, and E. Karlsson, Physica Scripta 3, 175 (1971).

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Theoretical investigation of the effect of irreversible relaxation on signals such as photon echo following multipulse excitation

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With irreversible relaxation taken into account, a relatively complete and detailed classification is presented for the sets of responses, such as optical induction or photon echo, which appear in large samples of matter as a result of multipulse excitation. Allowance is made for the responses that appear during the intermediate stages of the excitation process and for the responses after the end of the excitation process. The excited samples are regarded as quantum systems consisting of a large number of identical noninteracting particles with a discrete nondegenerate finite equidistant energy-level spectrum describable with the aid of energy-spin concepts. The operator of the interaction of the field of the pulses is linear in the energy spin. The transverse and longitudinal irreversible relaxations are accounted for by phenomenological spin operators defined by time-dependent differential equations whose averaging yields equations of the Bloch type. A new matrix method of investigating the solutions of these equations is developed. Some examples of possible technical applications of the relations obtained in this paper are discussed. These relations, in particular, can help choose the most convenient sequences of exciting pulses for the measurement of the times of transverse and longitudinal irreversible relaxation, and can also be useful in the development of spin, acoustic, and optical memory elements.

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A natural continuation of Dicke's ideas[1] was the theoretical prediction of the photon-echo phenomenon.[2] Photon echo in ruby was observed experimentally in a number of studies.[3] A relatively detailed theory of photon echo was given in[4] on the basis of the formalism developed by Dicke.[1] A photon echo is a sharply directional coherent beam (spontaneous coherent emission) that contains information on the dynamics of optical quantum systems and on the external generators that illuminate the medium, and appears after two laser pulses are applied to a quantum system.[3,4] An analogous situation arises in the case when quantum systems are excited with hypersound and terasound ($\nu \sim 10^{12}$ sec^{-1}),[5] and also in the case of a combination of optical and acoustic exciting pulses.[6]

Multipulse spin-echo excitation has aroused considerable theoretical and experimental interest.[7-11] It appears that multipulse excitation should also be of interest in the region of photon and phonon[5] echo, all the more since experiments have already been performed on three-pulse excitation of signals of the photon-echo type.[12] In addition, the spiked structure of a giant laser pulse is a typical example of multipulse excitation.[13] This has made necessary a detailed theory of multipulse excitation, particularly with allowance for irreversible relaxation, or at least an initial treatment of the simplest and most graphic case. Such a theory is needed also for the analysis of problems connected with the development of spin memory elements.[14] Similar memory elements, using the phenomena of photon and phonon echo, are also possible.

Jaynes and Bloom[7] have considered different general matrix methods for the investigation of the solution of modified Bloch equations. They have also made the first attempt, to our knowledge, to analyze the general case of excitation of spin systems by an arbitrary number n pulses without allowance for irreversible relaxation. The methods considered in their papers, however,

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