Hyperfine interaction for ^{160}Tb in gadolinium

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We measured the temperature dependence of the anisotropy of the energy distribution of 299-keV γ photons accompanying the decay of polarized ^{160}Tb nuclei. The ^{160}Tb nuclei were polarized in a gadolinium matrix at infralow temperatures obtained by adiabatic demagnetization of potassium chrome alum. The temperature was measured with the aid of a nuclear thermometer by determining the angular anisotropy of the γ radiation of ^{54}Mn nuclei polarized in a nickel matrix. The hyperfine magnetic field and the quadrupole interaction for the impurity atoms ^{160}Tb in gadolinium are found to be $H = 3.06 \pm 0.31$ MG and $P = (1.38 \pm 0.21) \times 10^{-18}$ erg.

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The hyperfine interaction in rare-earth (RE) atoms has recently been the subject of intensive experimental research. Particular interest attaches to the investigation of the hyperfine interaction for the impurity atoms of RE in different metallic matrices. The most effective for such problems are methods connected with registration of nuclear radiations, since they make it possible to investigate systems with very small concentrations of the impurity atoms. Measurement of the parameters of the hyperfine interaction in such experiments makes it also possible to obtain the nuclear characteristics of the RE isotopes.

We have investigated the hyperfine interaction of ^{160}Tb impurity atoms in a gadolinium matrix by the method of nuclear orientation at infralow temperatures. These measurements were undertaken because the published data for Tb impurities in gadolinium, obtained by two different methods (NMR[1] and the nuclear orientation method[2]) do not agree. Exact values of the hyperfine interaction constants in rare-earth metal alloys are essential in order to check the existing theoretical ideas concerning their magnetic structure,[3] as well as to determine the magnetic and quadrupole moments of the atomic nuclei.

It is known that the magnetic moments of the ions in heavy rare-earth metals that are magnetically ordered at low temperatures become polarized also in the case of magnetic ordering $\langle J_z \rangle = J$, at low temperatures, owing to the presence of a large exchange field. The electric field gradient (EFG) produced by the proper $4f$ shell should be directed along the electronic magnetization. The crystal-field gradient is directed along the crystallographic c axis, which in the general case may not coincide with the direction of the electronic magnetization. The presence of the crystal field leads, generally speaking, to deviation of the electric field gradient from the direction of the axis. However, inasmuch as for all rare-earth metals (with the exception of gadolinium) the magnetic hyperfine interaction is much larger than the electric quadrupole interaction, we can retain, with good approximation, only the diagonal matrix elements of the EFG tensor. Then, obviously, the resultant quadrupole interaction, which is determined by the two contributions, will take the following form:

$$P = P_{\parallel} + 1/2 (3 \cos^2 \theta - 1) P_c,$$

where $P_{\parallel}$ is due to the intrinsic $4f$ electrons, and P_c is due to the crystal field and the conduction electrons. Here θ is the angle between the z axis (the direction of the electron magnetization) and the crystal c axis. Under these conditions, the Hamiltonian of the interaction takes the form[3]

$$\mathcal{H} = -a_0 I_z + P[I_z^2 - {}^1/_3 I(I+1)], \quad (1)$$

where $a_0 = g_y \beta_y H$ and $P = 3eQV_{zz}/4I(2I-1)$ are the constants of the magnetic-dipole and electric-quadrupole interaction constants, respectively (the remaining symbols are standard).

To obtain the constants a_0 and P we measured in the present study the temperature dependence of the anisotropy of the angular distribution of the γ photons with energy 299 keV, accompanying the decay of the polarized nuclei ^{160}Tb. The ^{160}Tb nuclei were polarized in the gadolinium matrix at infralow temperatures obtained by adiabatic demagnetization of a paramagnetic salt (potassium chrome alum).

The investigated sample was a dilute alloy Tb with gadolinium (the Tb content in the alloy was $\lesssim 0.1\%$), which was bombarded by neutrons in a reactor to obtain radioactive ^{160}Tb nuclei. The bombarded sample, in the form of a disk 4 mm in diameter and 0.2 mm thick, was soldered with indium to the end of a copper cold finger, the other end of which was pressed into a block of potassium chrome alum.[4] Besides the ^{160}Tb sample, a ^{54}Mn(Ni), sample was soldered to the end of the cold finger and served as a low-temperature thermometer. A magnetic field of 8 kOe was applied to the cooled samples and was sufficient, according to[5], to magnetize the gadolinium matrix practically to saturation. The measurement of the anisotropy of the angular distribution of γ quanta with energy 299 keV of ^{160}Tb and 835 keV of ^{54}Mn were carried out simultaneously, the γ rays were registered with a Ge(Li) detector having a volume 22 cm^3 (with an NTA-512 512-channel pulse-height analyzer). The detector was placed along the axis of the external orienting field.

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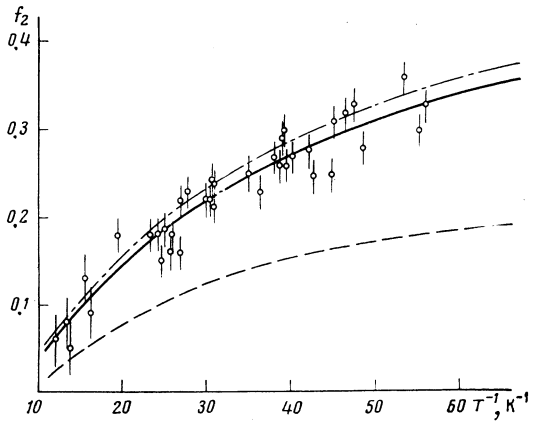

FIG. 1. Temperature dependence of the nuclear-orientation parameter f_2 for ^{160}Tb in gadolinium. The curves correspond to the hyperfine-interaction constants obtained in the present paper (solid line) and in [1] (dash-dot) and [2] (dashed).

The angular distribution of the γ photons emitted by the polarized nuclei can be represented in the form[6]

$$W(\vartheta)=1+\sum_{k=2,4,\ldots} A_k G_k f_k Q_k P_k(\cos\vartheta), \tag{2}$$

where A_k is a parameter that determines the angular dependence of the considered γ transition, G_k is the depolarization parameter due to the preceding unobservable transitions, f_k is a parameter describing the degree of orientation of the parent nucleus, Q_k is the correction for the finite solid angle of the detector, and $P_k(\cos\vartheta)$ are Legendre polynomials. The parameters A_k and G_k can be calculated, since the decay scheme of the ^{160}Tb nucleus is known, [7] and knowing the experimental geometry we can obtain the value of the coefficient Q_k. For the case of the considered γ transition with energy 299 keV, which is practically a pure dipole transition, we obtain

$$W(\vartheta)=1-0.9 f_2 P_2(\cos\vartheta). \tag{3}$$

We investigated in the experiment the counting rate of the γ photons at an angle $\vartheta=0$ in a time $\Delta t=2000$ sec and at different sample temperatures (the measurements were performed in the temperature interval 18 mK $\leq T \leq$ 88 mK). The 299-keV γ-radiation intensity of ^{160}Tb, and also the 835-keV radiation of ^{54}Mn, were determined by summing the counts in the channels corresponding to the γ line in the spectrum, and subtracting the background due to the harder γ radiation. The background was estimated in the following manner. Segments of the spectrum of the right and to the left of the peak were extrapolated until they crossed the verti- s that bounded the chosen energy window, and the area of the trapezoid under the line joining the intersection point was determined. The intensities determined in this manner were normalized to the corresponding values at high temperatures (~ 1 K), when there was no polarization.

From the normalized intensities $W(0)$ obtained in this manner we can determine with the aid of (3) the values of the nuclear-orientation parameter f_2. The temperature dependence of the experimental values $f_2(T)$ is shown in the Fig. 1 (the indicated errors are statistical). As already noted, the temperature in this experiment was measured with a nuclear thermometer by determining the anisotropy of the angular distribution of the γ photons from the polarized ^{54}Mn nuclei introduced into the lattice of metallic nickel. The temperature measurement method is based on the fact that the decay scheme and the nuclear magnetic moment of ^{54}Mn are known, as is also the value of the magnetic hyperfine field for Mn in Ni. [8] Inasmuch as in the case of 3d metals there are no effective quadrupole interactions, we can determine the temperature directly from the measurements of the angular anisotropy.

The obtained temperature dependence of the orientation parameter f_2 for ^{160}Tb is gadolinium was compared with the corresponding theoretical expression[6]

$$f_2=\frac{1}{I^2}\Big[\sum_m m^2 a_m-\frac{1}{3}I(I+1)\Big], \tag{4}$$

where $a_m=\exp(-E_m/kT)/\sum_m \exp(-E_m/kT)$ and $E_m=-a_0 m+P(m^2-4)$, with allowance for the fact that the spin of the ^{160}Tb nucleus is equal to 3. The parameters a_0 and P were selected with a BÉSM-6 computer by minimizing the functional

$$\chi^2=\sum\Big|\frac{f_{2i}^{\exp}-f_{2i}^{\text{theor}}}{\Delta f_{2i}^{\exp}}\Big|^2.$$

The corresponding theoretical curve is shown by the solid line in Fig. 1.

The calculation yields the following values of the hyperfine interaction parameters: $a_0=(8.60\pm 0.87)\times 10^{-18}$ erg and $P=(1.38\pm 0.21)\times 10^{-18}$ erg. This value of the magnetic hyperfine interaction parameter for ^{160}Tb corresponds to a hyperfine magnetic field $H=3.06\pm 0.31$ MG (with allowance for the fact that the magnetic moments of the ^{160}Tb nucleus is equal to 1.685 (8) n.m. [9]).

Figure 1 shows also the calculated curves corresponding to the parameters obtained in[1,2]. Kobayashi *et el.* [1] measured by the NMR method the hyperfine magnetic field and the quadrupole interaction at the ^{159}Tb nuclei in an alloy of terbium in gadolinium (at a terbium concentration ~ 10 at.%), and obtained $H=3.03(3)$ MG and $P=1.3(2)\times 10^{-18}$ erg (the last value was obtained by recalculation to the isotope ^{160}Tb, using the known values of the quadrupole moments of both isotopes). Fox and Hamilton, [2] using the procedure of oriented nuclei of ^{160}Tb in a dilute alloy of terbium and gadolinium, obtained values $H=3.4$ MG and $P=2.0\times 10^{-18}$ erg. Although the values of the compared parameters obtained in[4,2] do not differ greatly from one another, it is seen from Fig. 1 that the temperature dependences differ strongly at low temperatures. The experimental dependence obtained in the present study is close to the curve calculated with the constants of Kobayashi *et al.* [1]

According to the prevailing theoretical concepts and the available experimental data, the hyperfine magnetic field for heavy rare-earth metals and their alloys is

close to the corresponding value for the free ion. The hyperfine field for the Tb^{3+} ion is equal to 3.14 MG.[3] In a metal and an alloy, the presence of additional contributions from neighboring magnetic ions and from polarized conduction electrons, as a rule, decreases somewhat the value of the field in comparison with the field for the free ion. The data of[1] and the present results do not contradict this fact. The reason for the more substantial difference between the results of[2] remains unclear. It must be noted that the experimental conditions in[2] and in the present paper were practically identical.

As already noted, the quadrupole interaction constant determined in the experiment contains contributions from the intrinsic $4f$ shell and from the crystal field and the conduction electrons. From experiment with atomic beams (for the ^{159}Tb isotopes) we know the quadrupole-interaction constant for the Tb^{3+} ion, namely, $P_{II} = 2.55 \times 10^{-18}$ erg.[3] This yields for the ^{160}Tb isotope a constant equal to 1.14×10^{-18} erg. Comparing this value with the results of the present study (1.38×10^{-18} erg) we can conclude that the quadrupole interaction for terbium in gadolinium is determined mainly by the intrinsic $4f$ shell.

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Torsional vibrations and stimulated echo with a long memory in magnetic powders

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Two- and three-pulse echoes are studied in a number of magnetic powders at frequencies between 10 and 40 MHz in a stationary magnetic field. A long-lived (of at least several hours duration) stimulated echo is observed in powders consisting of particles with dimensions of the order of 1 μ. The results are interpreted on the basis of a model of torsional vibrations of the powder particles induced by an alternating magnetic field. The absence of damping of the stimulated echo is attributed to memory of the microcrystal orientation parameters.

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The phenomena of powder echo, first observed in magnetic materials,[1] and then in piezoelectrics[2,3] and metals,[4] are widely studied at the present time.[5–11] The experimental method is very similar to that which is used in the investigations of ordinary spin echo; upon irradiation of the powder at the instants of time $t = 0$, τ, and T by pulses of an alternating electromagnetic field, signals of two-pulse (A_2) and three-pulse (A_3) echoes are observed at the instants of time $t = 2\tau$ and $T + \tau$; the latter is frequently called the "stimulated" echo. The experiments are usually carried out with powders whose particles have a mean size of the order of the sound wavelength at the frequency of the alternating field, i.e., under conditions of acoustic resonance of the microcrystals. Just as in the case of magnetic resonance, the falloff of the echo signals upon change in the intervals between the sounding pulses is determined by the relations

$$A_2 \sim \exp(-2\tau/T_2), \quad A_3 \sim \exp(-2\tau/T_2 - T/T_1),$$

in which the characteristic times T_2 and T_1 appear. For most studied piezoelectrics and magnetic powders, the quantity T_2, depending on the dimensions of the crystals, the density of their packing, the viscosity of the medium in which the powders are located, and other factors, lies in the range 10^{-6}–10^{-3} sec. The time of "longitudinal relaxation" T_1 as a function of the charac-

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