Measurement of the spin amplitudes of neutron scattering by ^{159}Tb nuclei by the method of diffraction by antiferromagnets at infralow temperatures

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The difference $a_+ - a_-$ of the spin amplitudes for thermal-neutron scattering by terbium nuclei was measured in a diffraction experiment using a pulsed IBR-30 reactor and a cryostat with ^{3}He dissolved in ^{4}He. The result is $a_+ - a_- = -(0.35 \pm 0.14)$ F. The spin-incoherent scattering cross section obtained is $\sigma_{inc} = 5 \pm 4$ mb.

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INTRODUCTION

The study of the spin dependence of the interaction of thermal neutrons with nuclei yields information on the character of the nuclear forces in the case of light nuclei and on the properties of the so-called negative levels (with excitation energy lower than the binding energy of the neutron) for complex nuclei. The results can be used in other branches of physics, for example, to determine intra-atomic magnetic fields, to investigate hyperfine interactions, or to study nuclear magnetism. A known example of the spin dependence is the difference between the singlet and the triplet lengths of the neutron scattering by a proton. For the overwhelming majority of nuclei, however, there are no such data, since the usual measurements of the total and coherent neutron cross sections are insufficient for their determination.

A direct method of solving this problem is, for example, to use the diffraction of polarized neutrons by polarized nuclear targets [1]. Another method that improves the sensitivity of the experiments with polarized neutrons and polarized nuclei was proposed by Baryshevskiĭ and Podgoretskiĭ [2], who predicted the phenomenon of a neutron spin precession in a polarized nuclear target. Such measurements were initiated in Saclay. [3] A less universal but simpler method of determining the spin scattering amplitudes of neutrons by nuclei was given by Shapiro [4] and by Schermer and Blume [5]. This is the method of diffraction of unpolarized neutrons by an antiferromagnetic target with oriented nuclei. Herpin and Meriel [6], independently of the above-named authors, used this method to measure the spin amplitudes of holmium at liquid-helium temperature using a stationary reactor.

Much greater experimental possibilities arise on going to lower temperatures. We report the measurement of the spin-spin amplitude of ^{159}Tb by the neutron-diffraction time-of-flight method, using an IBR pulsed reactor and samples cooled to infralow temperatures in a cryostat with ^{3}He dissolved in ^{4}He.

MEASUREMENT METHOD

The s-scattering of neutrons by nuclei proceeds via states with spins $J = 1 \pm 1/2$. It is customary to separate in the corresponding spin scattering amplitude $a_\pm$ two terms, the first of which does not depend and the second does depend on the spin of the nucleus I and on the spin of neutron S:

$$a_\pm = A \pm B(\mathbf{IS}). \qquad (1)$$

The amplitude A determines the coherent cross section, and the so-called spin-spin amplitude B the spin-incoherent cross section for the scattering of unpolarized thermal neutrons.

$$\sigma_{coh} = 4\pi A^2, \qquad \sigma_{inc} = 4\pi I(I+1)B^2. \qquad (2)$$

The quantities A and B can be expressed in accordance with (1) in terms of the scattering amplitudes $a_\pm$ in the form

$$A = \frac{I+1}{2I+1}a_+ + \frac{I}{2I+1}a_-, \qquad B = \frac{1}{2I+1}(a_+ - a_-). \qquad (3)$$

Direct measurement of the magnitude and sign of the amplitude B is a difficult problem, which can be solved completely in principle in experiments with polarized neutrons and nuclei.

In 1966, Shapiro [4] pointed out an interesting possibility of measuring the amplitude B for a number of nuclei with the aid of diffraction of unpolarized neutrons at low temperatures by antiferromagnetic targets. We refer here to nuclei of elements with uncompensated electron shell, which produces strong intra-atomic magnetic fields. Cooling the target leads to polarization of the nuclei in each sublattice of the antiferromagnets as a result of the magnetic hyperfine interaction. The polarization produces a contribution of the coherent nuclear scattering to the "superstructure" diffraction maxima, where under ordinary conditions only magnetic scattering of the neutrons is observed. For a collinear antiferromagnet with one magnetic atom in the chemical unit cell, these maxima correspond to double the period of the cell.

A detailed analysis of the effects of nuclear polarization on the magnetic scattering of the neutrons was carried out by Schermer and Blume [5]. The formula obtained by them for the integral intensity of the superstructure maximum can be represented in the form

$$N = c\left|\sum_j \exp\{i\mathbf{k}\mathbf{r}_j\}(p_j\mathbf{q}_j + BI\mathbf{P}_{Nj})\right|^2, \qquad (4)$$

where the factor c takes into account the flux of the neutrons, their absorption, and the measurement geometry. The summation is carried out in the unit cell over all the atoms with coordinates $\mathbf{r}_j$. The magnetic-scattering amplitude p in units of 10^{-12} cm is $p = 0.27\,\mu_j f$, where μ_j is the magnetic moment of the ion (in Bohr magnetons) and f is the magnetic form factor of the atom. The magnetic-interaction vector **q** depends on the mutual orientation of the scattering unit vector **k** and the magnetization unit vector **h**, is equal to $\mathbf{q} = \mathbf{h} - \mathbf{k}(\mathbf{h}\cdot\mathbf{k})$. The equilibrium nuclear polarization P_N is given by the well known Brillouin formula

$$P_N = B_I\left(\frac{\mu_I H}{k_B T}\right) = \frac{2I+1}{2I}\,\mathrm{cth}\left(\frac{2I+1}{2I}\frac{\mu_I H}{k_B T}\right) - \frac{1}{2I}\,\mathrm{cth}\left(\frac{1}{2I}\frac{\mu_I H}{k_B T}\right), \quad (5)$$

where μ_I is the magnetic moment of the nucleus, H is the magnetic field at the nucleus, and T is the sample temperature. The polarization vector in the sublattice is collinear with its magnetization (parallel at $\mu_I H > 0$ and antiparallel at $\mu_I H < 0$).

Thus, measurement of the intensity of antiferromagnetic reflections in the presence and absence of nuclear polarization in the sublattices of the antiferromagnet makes it possible to determine the spin-spin scattering amplitude B. In accordance with formulas (4) and (5), it is necessary here to know the atomic and magnetic structures of the sample, its temperature, and also the magnitudes and signs of the hyperfine magnetic field and magnetic moment of the nucleus. It is obvious that the result of such an experiment can also be the solution of the inverse problem, i.e., the determination of the magnitude and sign of the hyperfine field H if the amplitude B is known.

EXPERIMENT AND RESULTS

1. The measurements were performed with the IBR-30 pulsed reactor by the neutron-diffraction time-of-flight method. The scattering angle was fixed, the neutrons incident on the sample had a continuous energy spectrum, and the wavelength satisfying the Bragg condition was determined from the time-of-flight of the neutrons over a specified distance. The reactor operated at an average power 15 kW (pulsed power 60 MW), the flight base was the distance 34.5 m from the reactor to the sample and 2 m from the sample to the detector. The detector was made up of small helium counters with gas pressure 10 atm.

The samples were cooled in a low-temperature container (dimensions 200 × 60 × 5 mm) of the cryostat with the ^{3}He dissolved in ^{4}He, in a manner similar to that used in [7], i.e., with the aid of liquid ^{4}He poured into the container and serving to transfer the heat from the sample to the dissolution chamber through a copper wall. The wall had a large surface area, strongly developed both on the container side and on the side of the dissolution chamber. The helium temperature in the sample container was measured as a calibrated carbon thermometer and was taken to be the sample temperature. It should be noted, however, that owing to uncontrollable thermal resistance between the sample and the liquid helium and to possible energy dissipation in the sample, the latter could have a higher temperature.

2. Preliminary measurements were performed with terbium dioxide and cobalt oxide, which have low intra-atomic fields, −315 kOe for terbium and +490 kOe for cobalt. The changes in the areas of the magnetic reflections (111) and (311) of terbium dioxide turned out to be less than 1%, indicating a very small value of the amplitude B for terbium. For cobalt, to the contrary, the value of B is known and is large [1], therefore measurements with the cobalt oxide served as a control. They yielded for the area of the magnetic reflection (111) a value of 20%, as against the 50% expected on the basis of latest data [8] on the field H.

The discrepancy can be attributed to the aforementioned possible difference between the sample temperature and the temperature of the thermometer (0.05°K), inasmuch as at low values of the magnetic fields the polarization of the nuclei is proportional to the sample temperature. In addition, the employed sample could have a noticeable fraction of another crystallographic modification [9] with a smaller value of H (lattice with vacancies).

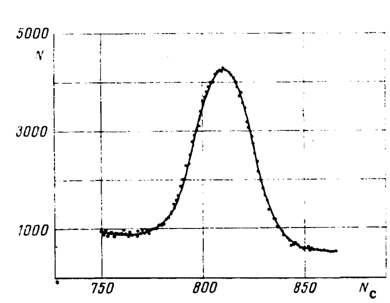
3. For the principal measurements, taking into account the experience with the preliminary measurements, we chose the intermetal TbAg, in which the magnetic field at the terbium nuclei is much larger. The sample weighing 180 g was prepared of terbium and silver of purity 99.99 by fusing stoichiometric amounts of the two components in an inert-gas atmosphere. The diffraction measurements were performed at a scattering angle of 42° in a "transmission" geometry.

The experimental spectrum for the reflection (110) obtained after 60 hours at 0.05°K, is shown in the figure. When the temperature was raised to 1.5°K, a small increase, but larger than the measurement error, of the peak area was observed. The results and the parameters used for the calculation are the following:

pq, F	H, Oe	T_1, K	T_2, K	P_{N_1}, %	P_{N_2}, %	$\Delta N/N$, %	B, F
23	$3.1\cdot10^6$	0.05	1.5	98	8	−(1.2±0.5)	−(0.088±0.034)

The magnetic-scattering amplitude pq was calculated here from magnetic-structure data [10] obtained with the aid of neutron diffraction. The magnetic structure was of the type (π, π, 0) with magnetic-cell parameters a = 7.24 Å and c = 3.62 Å, and with alternating (110) planes in which the magnetic moment of the ion was directed alternately parallel and antiparallel to the (110) direction. The value H of the hyperfine magnetic field in the free trivalent terbium ion was measured many times in other compounds. We can use this value, since the magnetic moment of the trivalent ion (TbAg ($\mu_J = (8.7-9.1)\mu_B$ [10]) is close to the value 9.0 μ_B of the free moment. We give the measured value of the relative change in the intensity [N(0.05°) − N(1.5°)]/N(1.5°) and the obtained value of the spin-spin amplitude B of terbium.

We note that in view of the saturation of the Brillouin curve (formula (5)) in large fields the uncertainty in the temperature, in the case of the TbAg sample, has practically no influence on the result. Nor does the presence of silver atoms in the unit cell of TbAg manifest itself, since the possible polarization of the silver nuclei is negligible owing to the small magnetic moment of the silver nuclei and the low value of the field H at nuclei of nonmagnetic ions in compounds of rare-earth elements.



Section of experimental neutron-diffraction curve of TbAg in the region of the reflection (110)M, obtained at temperature 0.05° K; N_c is the number of the analyzer channel of 40 μsec width and N is the number of the detector counts.

DISCUSSION OF RESULTS

The measurements of the spin-spin scattering amplitudes of thermal neutrons by terbium nuclei have demonstrated the high sensitivity of the method of diffraction by antiferromagnets at infralow temperatures. A value $-(0.35 \pm 0.14)$ F was obtained for the terbium-scattering amplitude difference $\Delta a - a_+ - a_-$. This is comparable in smallness with the value obtained [3] for ^{19}F by the method of diffraction of polarized neutrons by polarized nuclei. For the spin-incoherent cross section of neutron scattering by terbium nuclei we obtain, in accordance with formula (2), the value $(5 \pm 4) \times 10^{-3}$ b. The previous upper estimate [11] of the cross-section was 1 b.

Using the expression

$$a_\pm = R_\pm - \sum \lambda \Gamma_n{}^\pm / 2E_0{}^\pm,$$

which connects the scattering amplitude with the parameters of the resonances (neutron width Γ_n, resonance energy E_0), and assuming the effective radii of the potential scattering R_+ and R_- to be equal, we obtained the calculated value $\Delta a = -0.75$ (the spins of the resonances were taken from [12]). Predominance of the contribution of "positive" levels with J = 2 leads to a negative sign of Δa, and predominance of the contribution of the like "negative" levels to a positive sign of Δa. Since the calculated value takes into account only positive energies, the obtained experimental value of Δa indicates that levels with spin J = 2 predominate in the contribution made to the thermal region by the negative energies.

The small value of the spin-spin amplitude for the scattering of neutrons by terbium nuclei practically excludes the possibility of investigating the hyperfine interactions in various terbium compounds by diffraction of thermal neutrons at low temperatures. It is of interest to measure the spin-spin amplitudes of neutron scattering for different rare-earth elements.

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83