MAGNETIC HYPERFINE STRUCTURE OF THE Gd^{155} *LEVELS IN METALLIC GADOLINIUM AND IN THE INTERMETALLIC COMPOUND* $GdAl_2$

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The hyperfine structure of the ground and excited (86.5 keV) states of the Gd^{155} nucleus was investigated by means of the Mössbauer effect in metallic gadolinium and in the intermetallic compound $GdAl_2$ at a temperature of 80° K. The measurements were carried out in the lattice of samarium oxide using a Eu^{155} source, which, at a temperature close to that of liquid nitrogen, had an unsplit emission line of natural width. The absorption spectra obtained indicated that the spin of the 86.5 keV level was $^5/_2$, and the ratio of the g-factors of the excited and ground states was -2.1 ± 0.3. The values of the internal magnetic fields at the gadolinium nuclei in metallic gadolinium and in $GdAl_2$ were found to be 366 ± 55 kOe and 134 ± 20 kOe, respectively. An extrapolation to low temperatures gave a value of 167 ± 25 kOe for the field at the gadolinium nuclei in $GdAl_2$. The large difference between the values of the internal magnetic fields in gadolinium and in $GdAl_2$ was not accompanied by a large isomeric shift. No shift of the levels due to the quadrupole interaction was found in gadolinium or in $GdAl_2$. The measured value of the magnetic moment of the 86.5 keV level, $(+0.85 \pm 0.13)\mu_n$, did not agree with the value calculated from Nilsson's model.

1. INTRODUCTION

In recent years, the Mössbauer effect has been used effectively to investigate the magnetism of rare-earth elements and of their compounds and alloys. The magnetic hyperfine structure of the nuclear levels of isotopes of samarium, europium, dysprosium, erbium, thulium, and ytterbium has been investigated by means of the Mössbauer effect. Such investigations can be of special interest in the case of gadolinium, which is present in the form of a trivalent ion in the $^8S_{7/2}$ state both in the metallic phase and in all its compounds. In the first approximation, ions in the S-state should not exhibit a hyperfine interaction. However, it is known that the hyperfine interaction constants of transition elements in the iron group and of rare-earth elements (Gd^{3+}, Eu^{2+}) are not zero and not even very small. The existence of the hyperfine interaction indicates that the real wave function of an ion differs considerably from the wave function of the pure S-state. Recently, theoretical estimates have been made of the effects which would give rise to a nonzero magnetic field at nuclei of rare-earth ions in the S-state, but the nature of the hyperfine interaction in these cases has remained one of the less investigated problems.

The most convenient gadolinium isotope for investigations using the Mössbauer effect is Gd^{155}. The study of the hyperfine structure of the Gd^{155} levels is, however, complicated by the difficulties related to the preparation of a source of γ quanta with an unsplit emission line. In our earlier paper[1] (in which we dealt with the Mössbauer effect in gadolinium oxide involving γ transitions at energies of 60.0 and 86.5 keV), we showed that these difficulties could be overcome by using a Eu^{155} source in the samarium oxide lattice at temperatures close to the temperature of liquid helium. The emission line was found to be unsplit and its width was equal, or very close, to the natural width of the nuclear level. The probability of the Mössbauer effect under these conditions was low (little more than 1% for γ quanta of 86.5 keV energy) but still sufficient for measurements. In the present investigation, such a source was used to study the hyperfine structure of the Gd^{155} levels in metallic gadolinium and in the intermetallic compound $GdAl_2$.

2. EXPERIMENT

The measurement method was similar to that employed by us earlier[1] and therefore it will not

be discussed in detail in the present paper. Of the two γ transitions for which the Mössbauer effect was observed in Gd^{155}, the 86.5 keV transition corresponded to a considerably narrower line than the 60.0 keV transition. Therefore, the measurements of the hyperfine structure were carried out using the γ 86.5 keV transition. The lifetime of the 86.5 keV level was 7.2×10^{-9} sec according to Vergnes[2] and 9.6×10^{-9} sec according to Bozek et al.,[3] corresponding to the emission (absorption) line widths of 9.1×10^{-8} eV (0.31 mm/sec) and 6.8×10^{-8} eV (0.24 mm/sec), respectively. The separation between individual components of the hyperfine structure was expected to be greater than the line width in the absorption spectrum. The source of γ quanta, in the form of Eu^{155} in the lattice of samarium oxide, was at 90° K, and the absorbers were at 80° K. The measurements of the absorption spectra were carried out using an electrodynamic spectrometer; to isolate the 86.5 keV line in the γ radiation spectrum of Eu^{155}, we used a characteristic lead filter. The absorbers were made of powders; their thickness was 670 mg/cm^2 in the case of gadolinium and 590 mg/cm^2 in the case of $GdAl_2$ (natural mixture of gadolinium isotopes).

3. RESULTS OF MEASUREMENTS

The spectra of the resonance absorption of γ quanta of the 86.5 keV energy by Gd^{155} nuclei in metallic gadolinium and in the intermetallic compound $GdAl_2$ are shown in Fig. 1. Both the gadolinium and the compound $GdAl_2$ were in the ferromagnetic state at 80° K. In both cases, the absorption spectra had a hyperfine structure due to the interaction of the magnetic moments of the ground and excited states of Gd^{155} with the internal magnetic fields. The Gd^{155} isotope had a fairly large quadrupole moment (Q = 1.3 b) in the ground state. A quadrupole interaction was expected in metallic gadolinium, which has a hexagonal lattice. However, within the limits of the precision of the measurements, the spectra were found to be symmetrical with respect to zero source velocity, which indicated very small (or zero) values of the quadrupole interaction and the isomer shift. Therefore, in further analysis of the absorption spectra, the shift of the levels due to the quadrupole interaction and the isomeric shift were not taken into account. Since the hyperfine structure in the absorption spectrum of $GdAl_2$ was poorly resolved, the value of the ratio of g-factors of the excited and ground states of the nucleus was determined mainly on the basis of an analysis of the absorption spectrum of metallic gadolinium. It should be noted, however, that the form of the absorption spectrum of $GdAl_2$ was also found to be very sensitive to the value of the g-factor ratio.

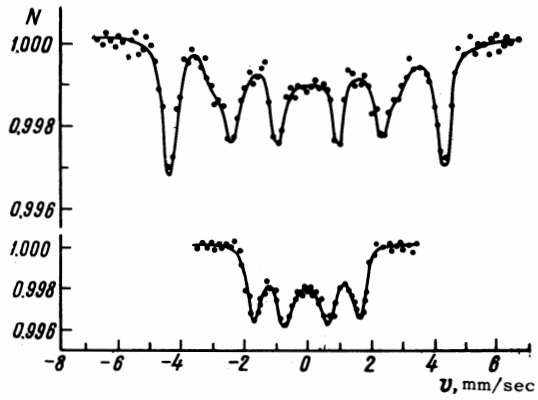

FIG. 1. Spectra of the resonance absorption of γ quanta of 86.5 keV energy by Gd^{155} nuclei in metallic gadolinium (upper curve) and in $GdAl_2$ (lower curve). The abscissa gives the velocity of the source of γ quanta relative to the absorber; the ordinate represents the intensity of the flux of quanta in relative units. The random error of the measurements was 0.04%.

The analysis of the absorption spectra was somewhat complicated by the fact that the spin of the 86.5 keV level of the Gd^{155} nucleus has not yet been measured. The known characteristics of the β and γ radiation of Eu^{155} and the value of the ground-state spin ($I_0 = {}^3/_2$) made it possible to establish only that the excited-state spin was either ${}^3/_2$ or ${}^5/_2$. For each of these values of the excited-state spin, we calculated the absorption spectra over a wide range of values of the ratio of the g-factors of the excited and ground states of the Gd^{155} nucleus. The energy of each of the components of the hyperfine structure was calculated from the usual formula

$$E = E_0 + g_g \mu_n H (m_g - \alpha m_e),$$

where E_0 is the energy of a γ transition in the absence of a hyperfine interaction, H is the magnetic field at a nucleus, μ_n is the nuclear magneton, m_g and m_e are the magnetic quantum numbers of the ground and excited states, $\alpha = g_e/g_g$, where g_g and g_e are the g-factors of the ground and excited states.

A comparison of the theoretical absorption spectra with the experimental results showed that for the excited-state spin $I_e = {}^3/_2$ agreement with experiment could not be obtained for any value of α. Thus, we established that the spin of the 86.5 keV level of the Gd^{155} nucleus was ${}^5/_2$.

Next, we determined, for $I_e = {}^5/_2$, the range of values of α, in which a satisfactory agreement was obtained between the theoretical and experi-

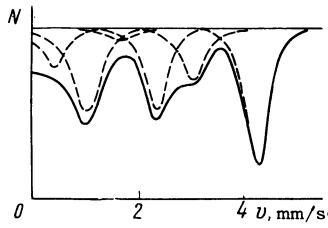

FIG. 2. Interpretation of the absorption spectrum of metallic gadolinium for $\alpha = -2.1$. The spectrum is symmetric with respect to the zero velocity and therefore only one half of the spectrum is given in the figure. The abscissa gives the velocity of the source, and the ordinate the intensity of the lines in relative units. The continuous curve is the sum of the individual components shown dashed.

mental absorption spectra. For the dipole transition $^5/_2 \rightarrow {}^3/_2$, there should be 12 components in the absorption spectrum, but two of them were of very low intensity and were not observed in the spectrum. The remaining 10 were not always well resolved because of the insufficiently strong magnetic field. The accuracy of the determination of α (and, consequently, of the value of the internal magnetic field) was limited by the poor resolution of those components of the hyperfine structure which were most sensitive to the value of α. Consequently, the following values were obtained for α and $g_g\mu_n H$:

$$\alpha = g_e/g_g = -2.1 \pm 0.3,$$

$$g_g\mu_n H = (0.65 \pm 0.10)\text{mm/sec} = (1.86 \pm 0.28)\cdot 10^{-7}\text{eV}.$$

Figure 2 shows the interpretation of the absorption spectrum of gadolinium for $\alpha = -2.1$.

Using the known value of the magnetic moment of the ground state of Gd^{155}, $\mu_g = -0.242\mu_n$, and the value of $g_g = \mu_g/I_g = -0.161$, we obtained: $g_e = +0.34 \pm 0.05$, $\mu_e = (+0.85 \pm 0.13)\mu_n$, H = (366 ± 55) kOe in the case of Gd, H = (134 ± 20) kOe in the case of $GdAl_2$.

4. DISCUSSION OF RESULTS

1. Internal magnetic fields. The value of the internal magnetic field at the gadolinium nuclei in metallic gadolinium was determined earlier by the investigation of the transmission of polarized neutrons through polarized targets[4, 5] and the magnetic field was found to be 324 ± 60 and 348 ± 34 kOe. These results are in good agreement with the magnetic field found in the present investigation. The measurements reported in [4, 5] were carried out at temperatures close to the absolute zero, but, bearing in mind the high value of the Curie point of gadolinium (293° K), we can ignore the difference between the temperatures at which the measurements were carried out in [4, 5] and in the present investigation. It has been shown also in [4-6] that the sign of the field in metallic gadolinium is negative.

The main source of the internal magnetic field in rare-earth elements is the orbital magnetism of the unfilled 4f-shell; for ions in the S-state this magnetism is zero. This is why the value of the field in gadolinium is much less than that in other rare-earth elements, in which it exceeds 10^3 kOe. Nevertheless, the value of the internal field is still high. To explain this, it is usually assumed that there is a mechanism which somehow perturbs the atomic core of an ion, disturbing its magnetic symmetry. Freeman and Watson[7] considered the exchange polarization of the electronic core of rare-earth ions in the S-state and obtained an approximate expression for the value of the internal magnetic field. For gadolinium, their estimate gave a value of −315 kOe, which was very close to the experimental value and had the correct sign. A certain contribution to the value of the field could have come from the exchange interaction with conduction electrons.[8] However, a comparison[5] of the hyperfine interaction constants of metallic gadolinium and Gd^{3+} ions, introduced as impurities into insulators (EPR data), showed that this mechanism was not very important for gadolinium because in all cases the hyperfine interaction constants had very similar values.

The intermetallic compound $GdAl_2$ is clearly the first compound in which the hyperfine interaction of the Gd^{3+} ion is considerably weaker. According to our data, at temperatures close to the temperature of liquid nitrogen, the internal field at gadolinium nuclei in this compound is 134 kOe. To compare it with the value of the field in gadolinium, it is necessary to make an extrapolation to low temperatures. Budnick et al.[9] have shown that the magnetization and the internal field increases by 20% when temperature is reduced to 4° K. Thus, the value of the internal field in $GdAl_2$ at low temperatures is found to be 167 ± 25 kOe, in good agreement with the nuclear magnetic resonance data.[9] Consequently, the hyperfine interaction for gadolinium nuclei in $GdAl_2$ is half that in metallic gadolinium. It may be mentioned that such a considerable difference between the values of the magnetic field is not accompanied by a change in the isomer shift. This may be regarded as indicating that the electron densities of gadolinium nuclei in the metal and in $GdAl_2$ are equal or differ only slightly. Unfortunately, at present, there are no data whatsoever on the isomeric shift for Gd^{155}. Therefore, we cannot ignore the possi-

bility of the accidental equality of the effective radii of the distribution of the nuclear charge in the ground and excited states, which would have led to the absence of the isomeric shift irrespective of the values of the electron densities in the region of a nucleus.

2. Spin and magnetic moment of the 86.5 keV excited state of Gd^{155}. As mentioned earlier, the spin of the 86.5 keV level of Gd^{155} has not yet been measured. Of the two possible values ($^3/_2$ or $^5/_2$), the value $^3/_2$ has been usually accepted. The justification for this has been the good agreement between the experimental value of the probability of the E1 γ transition from this level and the value of the probability calculated from Nilsson's model on the assumption that the 86.5 keV level was a state with the characteristics [651] $(^3/_2)^+$.[10] Vergnes[11] drew a general conclusion that the probabilities of the E1 γ transitions in deformed nuclei, in the range of mass numbers close to A = 155, were in good agreement with those calculated from the Nilsson model. Our spin value $^5/_2$ for the 86.5 keV excited state of Gd^{155} suggested that this conclusion should be treated with circumspection.

Moreover, the measured value of the magnetic moment of the 86.5 keV level indicated directly that the state is not pure in the sense of the Nilsson model. Attention was drawn to this earlier by Hrynkiewicz et al.,[3] who measured the absolute value of the g-factor of the 86.5 keV level by the perturbed angular correlation method; they obtained the value $|g_e| = 0.28^{+0.05}_{-0.04}$, which was in satisfactory agreement with the results of our measurements. The only state in Nilsson's scheme, whose characteristics could be identified with the 86.5 keV level of Gd^{155}, was the state [642] $(^5/_2)^+$. The magnetic moment calculated for such a state, both with [12] and without allowance for the renormalization of the spin g-factor, did not agree with the experimental value either in magnitude or in sign.

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