Magnetic hyperfine interaction and nuclear spin-lattice relaxation for $^{89}Y^m$ in $ZrFe_2$

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(Submitted 19 July 1982)
Zh. Eksp. Teor. Fiz. **84**, 1119–1124 (March 1983)

The anisotropy of the angular distribution of the 909 keV γ rays emitted in the decay of ^{89}Zr nuclei aligned at low temperatures in a ferromagnetic $ZrFe_2$ matrix has been measured. It is shown that annealing of the radiation defects produced by γ irradiation of the samples to induce the activity according to the reaction $^{90}Zr(\gamma,n)^{89}Zr$ plays a significant role. The magnetic hyperfine splitting is determined, as well as the nuclear spin-lattice relaxation time for the isomeric $^{89}Y^m$ ($T_{1/2} = 16.1$ sec) produced as a result of decay of the ^{89}Zr nuclei: for the annealed samples $\Delta = (5.76 \pm 0.14)$ mK and $T_1 = 37.3$ sec at a temperature of 15 mK. The hyperfine magnetic field for Y in the $ZrFe_2$ matrix is estimated.

PACS numbers: 75.30. – m, 71.50.Ej, 23.20.En, 23.20.Lv

INTRODUCTION

There have recently been many studies of the magnetic hyperfine interaction for nonmagnetic impurity atoms in ferromagnetic matrices. Results on hyperfine magnetic fields together with data from other magnetic measurements enable information to be obtained on the properties both of the system as a whole (impurity + matrix) and of the electronic structure of the matrix. The hyperfine magnetic fields at the impurity nuclei have been systematized for 3*d*-ferromagnetics as have a number of other characteristics of the impurity-matrix system, in particular the nuclear spin-lattice relaxation time. Besides working with pure 3*d* and 4*f*-ferromagnetic metals, it is of interest to study magnetic compounds in which atoms of these elements are the prime magnetic constituents. Investigation of nuclear spin-lattice relaxation in these systems can provide additional information on the nature of the hyperfine interaction and also, for example, on the conduction electron density at the Fermi level.

A knowledge of the value of the hyperfine fields is also of practical importance since it is essential for determining nuclear magnetic moments in experiments such as nuclear orientation, NMR on oriented nuclei, perturbed angular correlation, etc. It is also necessary in such experiments, carried out at low temperatures, to take account of the effects of nuclear spin-lattice relaxation since the relaxation time can be so long that it is difficult to achieve thermal equilibrium between the nuclear spins and the lattice.[1]

In the present work studies of nuclear orientation of ^{89}Zr and of the daughter isomer $^{89}Y^m$ have been undertaken in the ferromagnetic intermetallic compound $ZrFe_2$. The aim of this work was to determine the nuclear spin-lattice relaxation time and the hyperfine magnetic field for $^{89}Y^m$. As was shown elsewhere,[2] the compound $ZrFe_2$ with the Laves phase structure can be used successfully as a matrix for orienting the nuclei of a number of elements.

The determination of nuclear spin-lattice relaxation times in experiments with oriented nuclei is usually carried out by observing the rate of reduction of an initial nuclear orientation after its destruction by a thermal or radio-frequency pulse. However, the initial state of the system of nuclei before the relaxation is then indeterminate, which necessitates the introduction of an additional parameter in the analysis of the results. This requirement drops out if relaxation is observed in an isomeric (metastable) nuclear state, populated by decay of a parent radioactive nucleus, the state of which can be determined exactly.[3,4] This is just the situation for the $^{89}Zr \rightarrow {}^{89}Y$ decay investigated.

EXPERIMENTAL METHOD AND RESULTS

The specimens were prepared by melting the appropriate amounts of the basic components Zr and Fe in an arc furnace in an argon atmosphere, followed by homogenization *in vacuo* at 1000 °C for 95 h. The resulting bead was cut into ~0.4-mm thick plates by an electron beam. The plates were again heated *in vacuo* at 1000 °C for 40 h. The polished specimens in the form of 4-mm diameter, 0.3-mm thick disks were irradiated with 60- to 80-MeV γ rays in the linear electron accelerator of the Nuclear Research Institute of the Academy of Sciences (with tantalum radiator) for 5 h. The isotope to be studied resulted from the reaction $^{90}Zr(\gamma,n)^{89}Zr$ ($T_{1/2} = 78$ h). The ^{89}Zr decay scheme is shown in Fig. 1.

Measurements were made on four specimens. Two specimens were studied immediately after irradiation without additional annealing. Two other specimens, sealed into a pumped quartz ampoule, were heated at 900 °C for respec-

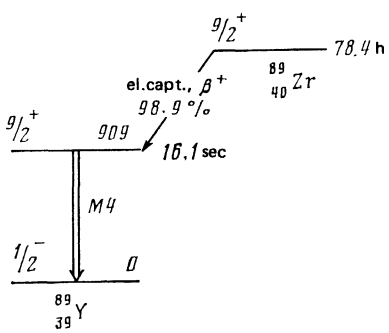

FIG. 1. Part of the decay scheme $^{89}Zr \rightarrow {}^{89}Y$.

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tively 90 and 170 h. The specimens were then soldered with pure indium to the end of the copper cold finger of an adiabatic demagnetization cryostat described earlier.[5] An external 6 kG orienting magnetic field was applied to the specimen. The γ rays emitted in the direction of the external magnetic field were recorded by a Ge(Li) detector. The specimen temperature was measured with a ^{54}Mn(Ni) nuclear thermometer soldered, together with the specimen, to the cold finger.

The angular distribution of the γ rays which accompany the decay of the oriented nuclei has the form

$$W(\theta)=\sum_{k=0,2,4\ldots} Q_k B_k U_k A_k P_k(\cos\theta), \qquad (1)$$

where Q_k is the coefficient which takes into account the geometrical dimensions of the detector, B_k is a function of the oriented parent nucleus and depends on the hyperfine splitting energy $\Delta=\mu H/Ik_B$ (μ is the nuclear magnetic moment, H the hyperfine magnetic field, I the nuclear spin and k_B is Boltzmann's constant) and the temperature T, U_k are disorientation coefficients resulting from preceding unobserved transitions, A_k are coefficients describing the properties of the observed γ transition and P_k are Legendre polynomials (θ is the angle between the nuclear orientation axis and the direction of emission of the γ rays).

The counting rate for 909-keV γ rays over 2000 seconds, at an angle $\theta=0°$, was measured in an experiment at a low temperature—$N_T(0)$—and at $T\sim 1$ K—$N_\infty(0)$, when there is no nuclear orientation (isotropic radiation). The anisotropy of the angular γ-ray distribution was determined as $1-W(0)=1-N_T(0)/N_\infty(0)$.

The decay ^{89}Zr$\rightarrow^{89}$Y (Fig. 1) takes place through an isomeric level with energy 909 keV, $^{89}Y^m$, which has half-life $T_{1/2}=16.1$ sec. Since the lifetime of the intermediate state $^{89}Y^m$ ($9/2^+$) is sufficiently long, reorientation of the nuclei is possible in this state as a result of the different hyperfine splittings in ^{89}Zr($ZrFe_2$) and $^{89}Y^m$($ZrFe_2$). Three different cases are then possible, determined by the relation between the nuclear spin-lattice relaxation time T_1 and the lifetime of the isomeric level τ.

1. $T_1\gg\tau$. There is then no reorientation in the intermediate state and the angular anisotropy of the 909-keV γ rays is completely determined by the orientation of the parent ^{89}Zr nucleus.

2. $T_1\ll\tau$. It is then to be expected that complete reorientation of the nuclei takes place in the $9/2^+$ ^{89}Y state. The measured angular anisotropy is determined by the orientation parameters of $^{89}Y^m$ in the $ZrFe_2$ matrix according to the corresponding hyperfine magnetic splitting of this state.

3. $T_1\sim\tau$. The anisotropy is a function of the orientation parameters of the nuclei ^{89}Zr and $^{89}Y^m$ and of T_1. The interpretation of results is then more difficult, but it is just in this case that the nuclear spin-lattice relaxation time can be estimated.

It is not known at the outset which of the three possibilities occurs in practice, so that the problem has to be solved experimentally.

The results of measuring the anisotropy of 909-keV γ rays (with multipolarity M 4) are shown in Fig. 2. Two sets of points are shown corresponding to the different conditions under which the specimens were prepared: annealed specimens (1) and unannealed specimens (2). It can be seen that the magnitude of the anisotropy for specimens which were heat-treated after γ-ray irradiation is higher than for the unannealed specimens. There was no noticeable difference between the specimens annealed for 90 and for 170 h.

The anisotropy expected was calculated on the assumption that it is produced completely by the orientation of the parent ^{89}Zr nucleus (case 1). The hyperfine field for Zr in $ZrFe_2$ is known from NMR results: $H=125$ kG.[6] Assuming that the magnetic moment of the ^{89}Zr nucleus is not appreciably different from the moments of the neighboring nuclei ^{85}Kr ($\mu=1.005$ n.m.[7]) and ^{87}Sr ($\mu=1.093$ n.m.[7]), the states of which are described by one and the same neutron configuration $g_{9/2}$, we can take $\mu(^{89}Zr)\approx 1$ n.m. Taking these values of μ and H, the calculated anisotropy $1-W(0)$ lies between 0.014 and 0.0035 in the temperature range of our measurements. The experimental values obtained exceed these calculated values appreciably (both for the annealed and unannealed specimens). This provides a basis for assuming that there is a reorientation of the nuclei (total or partial) during the intermediate isomeric state $9/2^+$ ^{89}Y. In this case the expression for the γ-ray angular distribution can be written in the form

$$\overline{W(\theta)}=\frac{T_1}{T_1+\tau}W_1(\theta)+\frac{\tau}{T_1+\tau}W_2(\theta), \qquad (2)$$

where $\overline{W(\theta)}$ is the observed angular distribution, while

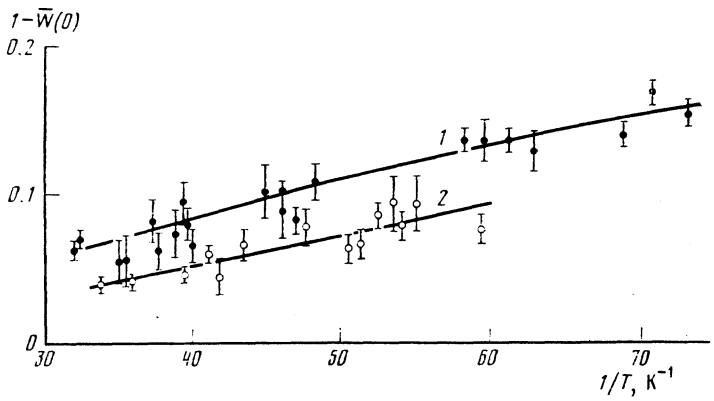

FIG. 2. Temperature dependence of the angular anisotropy of 909-keV γ rays. Full lines are the result of curve fitting using Eq. (2); 1—annealed specimens; 2—unannealed specimens.

$W_1(\theta)$ and $W_2(\theta)$ are the theoretical expressions of Eq. (1) for the angular distributions corresponding to orientation of only the parent (^{89}Zr) or of only the daughter ($^{89}Y^m$) nuclei: τ is the lifetime of the nucleus in the intermediate state. Equation (2) is an approximation and is valid on the assumption of a single-exponential relaxation process.

Two parameters in this equation are unknown: the magnetic hyperfine splitting Δ for $^{89}Y^m$ in $ZrFe_2$, which determines $W_2(\theta)$, and the spin-lattice relaxation time T_1. The values of these parameters were determined by fitting the theoretical expression of Eq. (2) to the experimental temperature dependence using the least squares method by minimizing the function

$$\chi^2 = \sum_i |(W_i^{exp} - W_i^{theor})/\Delta W_i|^2.$$

In doing this it was assumed that the temperature dependence of T_1 corresponded to either the "high temperature," $T_1T = C_K$, or to the "low temperature" $T_1 = (2/\Delta)C_K \tanh(\Delta/2T)$, Korringa approximations, where C_K is the Korringa constant. No difference was found between the results of calculations carried out for the two approximations. The following values were found for the parameters: $\Delta = (3.93 \pm 0.10)$ mK, $T_1T = (0.33 \pm 0.11)$ sec·K for unannealed specimens, $\Delta = (5.76 \pm 0.14)$ mK, $T_1T = (0.560 \pm 0.057)$ sec·K for annealed specimens.

DISCUSSION OF THE RESULTS

The difference in the values of the hyperfine magnetic splitting and spin-lattice relaxation constant for specimens annealed after irradiation in the accelerator and unannealed, indicates that radiation damage is of considerable importance in the present case. Radiation damage is produced by both the γ rays which lead to the reaction $^{90}Zr(\gamma, n)^{89}Zr$ and by the primary electron beam background, the intensity of which is considerably greater than the γ-ray beam intensity. An effect of radiation damage produced by neutrons on the hyperfine magnetic interaction for ^{181}Ta in $(Zr_{0.9}Hf_{0.1})Fe_2$ was found before.[8]

The hyperfine magnetic field for $^{89}Y^m$ in $ZrFe_2$ can be estimated from the experimentally determined hyperfine splitting Δ for $^{89}Y^m$. The magnetic moment of the isomer $^{89}Y^m$ has not been determined experimentally. It follows from the systematics of magnetic moments of nuclei in the region of mass numbers $A \sim 90$, that for the proton configuration $g_{9/2}$ the moment is practically independent of the number of neutrons.[9] It can be assumed from this that the nuclear magnetic moment of $^{89}Y^m(9/2^+)$ does not differ appreciably from the moment of $^{87}Y^m$: $\mu = 5.9(14)$ n.m.;[10] $6.10(^{+32}_{-15})$ n.m.[9] Taking the mean of these two values, we obtain the following estimate for the hyperfine magnetic field at Y in $ZrFe_2$ corresponding to unannealed and annealed specimens: $H = (81 \pm 8)$ kG; (118 ± 10) kG. The uncertainty in the value of the magnetic moment of the nucleus is included in the error shown.

We shall compare the magnitudes of the hyperfine fields for three neighboring elements Y, Zr and Nb in a $ZrFe_2$ matrix. It can be seen from Table I that the field on Y for an annealed specimen is close in value to the field on Zr determined in NMR experiments on the stable isotope ^{91}Zr. The field on Y for an unannealed specimen agrees, within the limits of uncertainty, with the hyperfine field for Nb, obtained in experiments on nuclear orientation, also with an unannealed specimen. It is reasonable to suppose that after annealing of radiation defects, the hyperfine field for Nb increases to the same extent as in our case for Y. If this is so, then it can be concluded that the hyperfine magnetic fields for the three neighboring elements Y, Zr and Nb on regular sites of the $ZrFe_2$ lattice are the same.

Analysis of the results on nuclear spin-lattice relaxation times show that the effect of annealing radiation damage is also very important in this case. The relaxation time T_1 for $^{89}Y^m$ in $ZrFe_2$ is, for example, 22 sec at $T = 15$ mK and 11 sec for $T = 30$ mK for unannealed specimens; for annealed specimens the relaxation times at the same temperatures are 37.3 and 18.7 sec. As can be seen, these values of T_1 are comparable with the mean lifetime of the isomeric level, $\tau = 23$ sec. In earlier work,[2] in which nuclear orientation of ^{177}Lu in a $ZrFe_2$ matrix was studied, a value $T_1 = 6.7$ sec was found for the nuclear spin-lattice relaxation time in the intermediate isomeric state $^{177}Hf^m$ ($T_{1/2} = 1.1$ sec) at $T = 30$ mK.

For the majority of impurity atoms in a Fe matrix, the values of $(gH)^2T_1T$ (g is the nuclear g-factor, H the hyperfine magnetic field) are the same within a factor of two.[4,11] Comparison of these two values for $^{177}Hf^m$ and $^{89}Y^m$ in a $ZrFe_2$ matrix, given in Table I, shows that our results for unannealed specimens are close to results for $^{177}Hf^m$, also obtained with an unannealed specimen. Unfortunately there are no other data on the nuclear spin-lattice relaxation time in a $ZrFe_2$ matrix with which our results could be compared.

The value of $(gH)^2T_1T$ obtained in the present work for a $ZrFe_2$ matrix is appreciably less than (by about an order of magnitude) the corresponding value for a Fe matrix. This

Table I. Results on hyperfine magnetic fields and nuclear spin-lattice relaxation times for nonmagnetic impurity atoms in a $ZrFe_2$ matrix.

Impurity	H_{hf}, kG	T_1T, sec · K	$(gH)^2T_1T \cdot 10^9$, G^2 sec · K	Method	Reference
$^{89}Y^m$ unannealed specimen	81±8	0.33±0.11	3.8	NO	Present work
$^{89}Y^m$ annealed specimen	118±10	0.560±0.057	13.8	NO	Present work
^{91}Zr	125	–	–	NMR	[6]
^{95}Nb	94±16	–	–	NO	[12]
$^{177}Hf^m$	200±20	0.201	4.2	NO	[2]

could be a reflection of the difference in electronic structures of these matrices.

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Translated by R. Berman