Measurements for spin inversion and noninversion in successive decays via nuclear magnetic resonance on oriented nuclei

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Nuclear magnetic resonance on oriented nuclei (NMR-ON) measurements were performed on the successive decays of ^{89}Zr-^{89}Y^{m} and ^{191}Os-^{191}Ir^{m} in Fe. The NMR-ON spectra of ^{89}Zr*Fe* and ^{191}Os*Fe* were obtained by detecting γ rays from the decay of the isomers, ^{89}Y^{m} and ^{191}Ir^{m}, respectively. For ^{89}Zr*Fe*, the anisotropy of the γ ray increased at the resonance. On the other hand, for ^{191}Os*Fe* the anisotropy of the γ ray decreased at the resonance. These phenomena were explained using the spin inversion and spin noninversion processes including the lifetimes of the isomers and spin lattice relaxation times. NMR-ON measurements for such spin inversion and noninversion processes were reported. The resonance spectra were also observed by detecting β rays from ^{89}Zr and ^{191}Os. In these experiments the magnetic moments of ^{89}Zr and ^{191}Os were determined to be -1.08 (2) μ_N and 0.962 (28) μ_N, respectively. The signs of the magnetic moments of ^{89}Y^{m} and ^{191}Ir^{m} were also determined to be positive. [S0556-2813(96)06109-2]

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I. INTRODUCTION

Nuclear magnetic resonance of oriented nuclei (NMR-ON) [1] is a powerful tool in the study of nuclear magnetic dipole moments. Recently, the technique has been applied in on-line experiments. In such experiments the radio isotopes are implanted as unoriented ''hot'' nuclei. Implantation is followed by a sequence of successive decay of short lifetime so that description of the degree of nuclear orientation, which involves competition between nuclear spin-lattice relaxation and nuclear decay becomes quite complex. To simplify the situation Stone [2] introduced the new method of time-resolved on-line nuclear orientation (TR-OLNO). The basic idea of this method is to measure the time dependence of the anisotropy of the angular distribution of the decay γ transitions following implantation of activity in short pulse. During the decay sequence the axis of magnetization of the sample remains unchanged, however the direction of polarization along that axis (the direction of the nuclear spin) depends upon the sign of the hyperfine interaction (the product of the nuclear moment and the hyperfine field) which may reverse at each decay. The effective relaxation time to thermal equilibrium at each stage in the decay chain depends upon whether or not inversion of the spin direction occurs in the preceding decay. Stone [2] calculated the time dependent anisotropy in both the spin inversion and noninversion cases and demonstrated the occurrence of the spin-inversion process for the sequence $^{183}\mathrm{Au} \rightarrow {}^{183}\mathrm{Pt}^{m}$ from analysis of the time dependent anisotropy of γ decay of ^{183}Pt^{m}.

In the present work we use NMR-ON to demonstrate both spin inversion and spin noninversion in successive decay in studies of ^{89}Zr-^{89}Y^{m} and ^{191}Os-^{191}Ir^{m} decays. When resonance is detected in β radiation, the results give information not only of the magnitude of the hyperfine interaction (and hence of the nuclear moment) but also its sign, which is usually not available from NMR-ON studies.

II. EXPERIMENTAL PROCEDURE

Sample of ^{89}Zr*Fe* was prepared by recoil implantation into iron of the precursor ^{89}Nb ($T_{1/2} = 2.0$ h) produced by the nuclear reaction $^{89}\mathrm{Y}(\alpha,4n)^{89}\mathrm{Nb}$. Targets of ^{89}Y were made by spattering onto thin Al foils. Stacks of alternating target foils and thin pure iron foils (thickness ~ 1 μm) were irradiated for 15 h by a 50-MeV α beam at the SF cyclotron of the Institute for Nuclear Study, University of Tokyo. Samples of ^{89}Zr*Fe* were handled in two ways after irradiation; one was given no heat treatment and the other was annealed at 800 °C for 1 h in vacuum. Sample of ^{191}Os*Fe* was prepared by irradiation of a small piece of a dilute alloy of Ir*Fe* (0.2 at. %), rolled to 2 μm thickness, for 12 days in the reactor at the Japan Energy Research Institute in a neutron flux of 2.9×10^{13} n/sec cm^2. After irradiation, the sample was annealed in vacuum for 1 h at 800 °C.

The samples were soft-soldered to the copper cold-finger of a ^{3}He/^{4}He dilution refrigerator and cooled to a temperature of 7 mK. The γ rays were detected by two pure Ge detectors placed at 0° and 180° with respect to B_0 which defined the orientation axis. The β rays were detected with two Si detectors of 50 mm^2 area and 0.5 mm thick, mounted on a heat shield at 0.7 K, inside the ^{3}He/^{4}He dilution refrigerator. Details of the detector system have been described in Ref. [3].

III. RESULTS AND DISCUSSION

A. ^{89}Zr-^{89}Y^{m} : an example of spin inversion

The partial decay scheme of ^{89}Zr is shown in Fig. 1. ^{89}Zr decays mainly (99.0%) to the 909-keV $\frac{9}{2}^+$ isomeric state of ^{89}Y^{m} ($T_{1/2} = 16.1$ s) by an allowed β^+/EC transition. The magnetic moment of ^{89}Zr was not previously measured. NMR-ON resonance spectra have been reported for ^{89}Y^{m}*Fe* [4] by observing resonant changes in the 909-keV anisotropy. However, depending upon the ratio of the

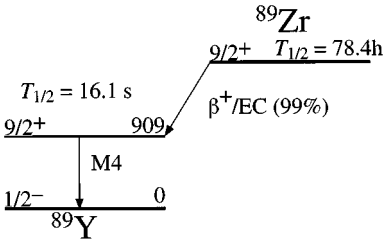

FIG. 1. Partial decay scheme of ^{89}Zr.

nuclear spin-lattice relaxation time of $^{89}Y^m$ in iron to the isomeric half-life it may also be possible to detect resonance in the parent isotope ^{89}Zr from observation of the same 909-keV transition. From the value of the Korringa constant measured by the integral low temperature nuclear orientation method for $^{89}Y^m$ in iron, $C_k = 0.44(4)$ sK [5], we estimate a relaxation time of 37 s at 10 mK. The tabulation of Shaw and Stone [6] may be used to estimate that the average B_2 parameter in the decaying relaxing $^{89}Y^m$ nuclei will be only 53% of its thermal equilibrium value. This means that at the time of decay the $^{89}Y^m$ sample retains considerable ''memory'' of the orientation of its long-lived ^{89}Zr parent. In this circumstance the possibility exists to detect NMR-ON of ^{89}Zr by observation of the decay of $^{89}Y^m$. The count rate of the 909-keV γ rays in both Ge detectors placed at 0° and 180° decreased at low temperature.

We first searched for the NMR-ON resonance of $^{89}Y^m$*Fe* at about 323 MHz as previously reported [4]. We could not detect the resonance with the annealed sample. However, with the unannealed sample we found the resonance, shown in Fig. 2, which exhibits a decrease in anisotropy (increase in count rate). Its observation shows that the parent ^{89}Zr nuclei would be in substitutional sites in the iron lattice. Next we searched for, and found, the resonance of the parent ^{89}Zr*Fe* at about 51 MHz using the same 909-keV γ-ray anisotropy. The resonance, shown in Fig. 3, exhibits an increase in anisotropy (decrease in count rate).

Understanding of the increase of anisotropy on resonance of the ^{89}Zr parent lies in the relative sign of the hyperfine interactions of ^{89}Zr*Fe* and $^{89}Y^m$*Fe*. The signs of the hyperfine fields acting on Zr and Y in iron are the same (negative), however, if the signs of the magnetic moments of ^{89}Zr and

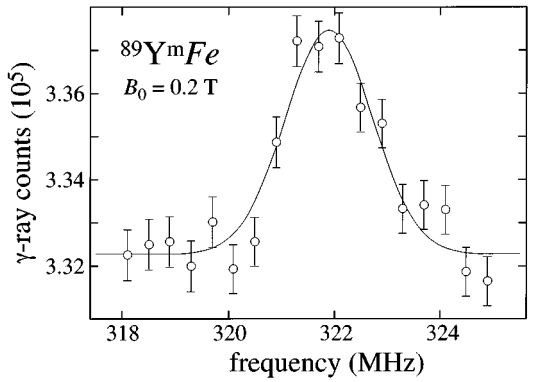

FIG. 2. NMR-ON spectrum of $^{89}Y^m$*Fe* taken with FM of ± 0.4 MHz by detecting 909-keV γ rays at 0° and 180°.

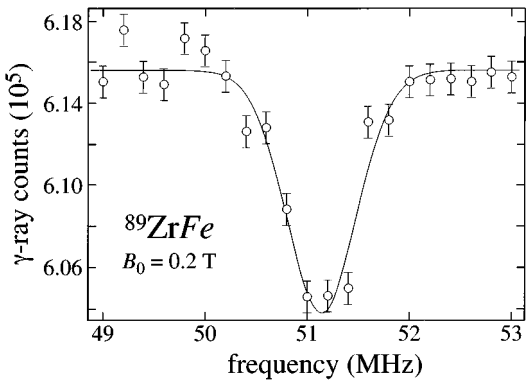

FIG. 3. NMR-ON spectrum of ^{89}Zr*Fe* taken with FM of ± 0.3 MHz by detecting 909-keV γ rays at 0° and 180°.

$^{89}Y^m$ differ, there is an inversion of the nuclear spin direction of $^{89}Y^m$ immediately following β decay, as compared with the thermal equilibrium direction, as shown in Fig. 4. Achievement of thermal equilibrium from this initial spin-inversion situation takes longer than from an initially unoriented $^{89}Y^m$ spin system. Resonance of ^{89}Zr reduced the initial spin inversion, speeding up the effective relaxation time and thus increasing the anisotropy of the 909-keV transition as observed. From the γ decay observation of the resonance in the polarizing field $B_0 = 0.2$ T we obtain

$$\nu(^{89}\mathrm{Zr}Fe{:}B_0 = 0.2\mathrm{T}) = 51.18(4)\ \ \mathrm{MHz}.$$

The absolute sign of the magnetic moment of ^{89}Zr can be determined independent of nuclear systematics by observation of β-ray anisotropy. The β-ray anisotropy was small due to the presence of other activities in the sample and the fact that the transition involves high spin states of the same j. A weak ^{89}Zr*Fe* NMR-ON resonance by detecting the β rays was observed simultaneously with the γ-ray resonance with center frequency 51.15(10) MHz as shown in Fig. 5, in agreement with the result given above. The sign of the β-ray asymmetry destruction on resonance unambiguously

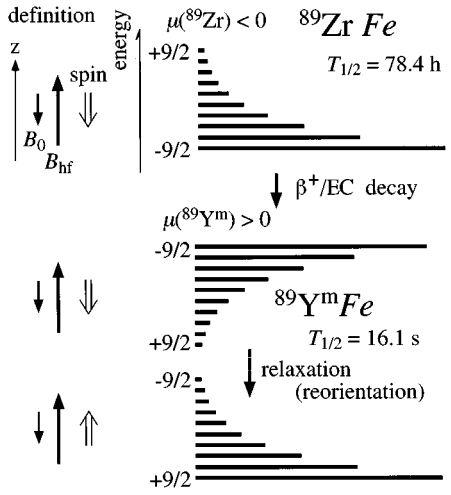

FIG. 4. Change of the spin polarized direction of ^{89}Zr-$^{89}Y^m$ in Fe.

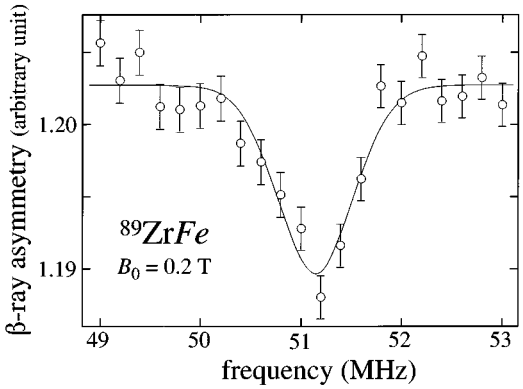

FIG. 5. β-NMR-ON spectrum of ^{89}Zr*Fe* taken with FM of $\pm$ 0.3 MHz by detecting the β-ray asymmetry of $N(0°)/N(180°)$ (arbitrary unit) with respect to the orientation axis (the direction of the external magnetic field).

gave the sign of the moment of ^{89}Zr as negative, hence the sign of the magnetic moment of ^{89}Y^m is positive. The hyperfine field B_{HF}(Zr*Fe*) was reported to be $-27.4(4)$ T by a room temperature PAC measurement [7]. If we assume that the impurity hyperfine field in iron follows the same temperature dependence as that of the hyperfine field of Fe itself [8], the field of Zr*Fe* at 4.2 K is expected to be $-28.1(4)$ T. Taking this field and the Knight shift K(Zr*Fe*) $= -0.16(12)$ [9] and neglecting a possibly hyperfine anomaly the magnetic moment of ^{89}Zr is deduced to be

$$\mu(^{89}\mathrm{Zr}) = -1.08(2)\,\mu_N .$$

It should be noted that the accuracy of the value is entirely limited by the uncertainty of the hyperfine field.

Since the configuration of ^{89}Zr is one neutron hole in the doubly magic $^{90}_{40}\mathrm{Zr}_{50}$, the result is important for nuclear shell model calculation. The Schmidt value of the $g_{9/2}$ neutron state is $-1.91\mu_N$. The deviation of the magnetic moment from the Schmidt value can be explained using the core-polarization formula [10]. With the neutron configuration of $(g_{9/2})^{-1}$ and the parameter set of the harmonic-oscillator potential $C = 30$ MeV, the calculated value is $-1.09\mu_N$ which is in good agreement with the experimental value.

B. ^{191}Os-^{191}Ir^m: an example of spin noninversion

As an example of spin noninversion in isomeric decay we report results on the sequence ^{191}Os-^{191}Ir^m. The partial decay scheme of ^{191}Os is shown in Fig. 6. The magnetic moment of ^{191}Os was not previously measured. The NMR-ON resonances of ^{191}Ir^m*Fe* and ^{191}Ir^m*Ni* [11] were measured and the magnitude of the magnetic moment of ^{191}Ir^m were determined as 6.026(36) μ_N. The half-life of the isomeric state is 4.9 s, relatively long compared to the spin-lattice relaxation time of 1.1 s at $B_0 = 0.2$ T deduced from observed relaxation following resonance [12]. Hence the average B_2 orientation parameter in the decay of ^{191}Ir^m nuclei is estimated from the tabulation of Shaw and Stone [6] to the 95% of its thermal equilibrium value. This suggests that the anisotropy of the 129-keV γ decay of ^{191}Ir^m will have a weak dependence upon the orientation of the parent ^{191}Os nuclei.

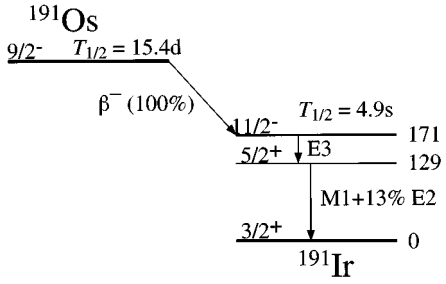

FIG. 6. Partial decay scheme of ^{191}Os.

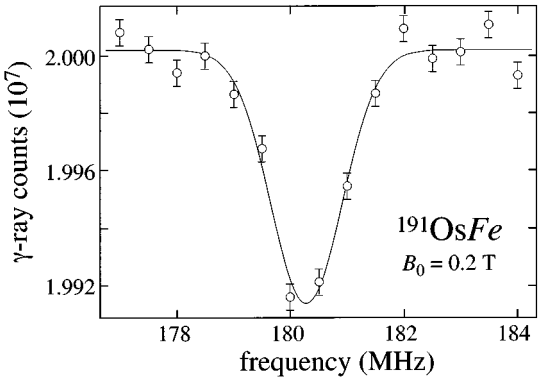

FIG. 7. NMR-ON spectrum of ^{191}Os*Fe* taken with FM of $\pm$ 0.5 MHz by detecting the 129-keV γ ray at 0° and 180°.

The sample was cooled to 7 mK in the polarizing field $B_0 = 0.2$ T with γ detectors at 0° and 180° which showed a strong increase in the 129-keV count rate with falling temperature. Searching for the ^{191}Os*Fe* resonance over a wide frequency region revealed a clear but small resonance at about 180 MHz. As shown in Fig. 7 the anisotropy is deduced (count rate decreased) in resonance as is also known to occur for the ^{191}Ir^m*Fe* resonance. The reduction in anisotropy on resonance of the parent is understood as arising from the fact that there is no spin inversion, i.e., no change of sign in the hyperfine interactions of parent and isomeric daughter, as shown in Fig. 8. The polarization of the ^{191}Ir^m spin immediately following β decay is in the same sense as their thermal equilibrium polarization and hence the isomeric spin system reaches equilibrium faster than if they had been initially unoriented. Detection of the ^{191}Os*Fe* polarization on resonance increases the effective relaxation time and hence decreases the anisotropy, as observed in Fig. 7. Thus we have an example of the spin noninversion case.

NMR-ON resonance was also observed in the β detectors at 0° and 180° as shown in Fig. 9 and the sign of the resonance signal determined the sign of the ^{191}Os hyperfine interaction as negative. The Os*Fe* hyperfine field is known to be negative, hence the ^{191}Os magnetic moment is determined to be positive. The sign of the magnetic moment of ^{191}Ir^m is also positive, as previously assumed on the basis of systematics. The observed ^{191}Os resonance yields

$$\nu(^{191}\mathrm{Os}Fe{:}B_0 = 0.2\mathrm{T}) = 180.3(1)\ \ \mathrm{MHz}.$$

The hyperfine field of ^{189}Os*Fe* has been determined as

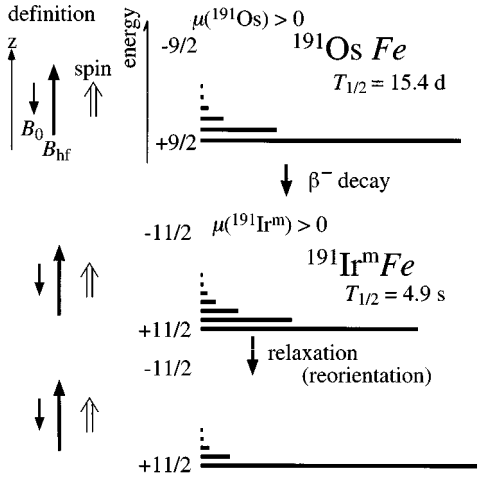

FIG. 8. Change of the spin polarized direction of ^{191}Os-^{191}Ir^m in Fe.

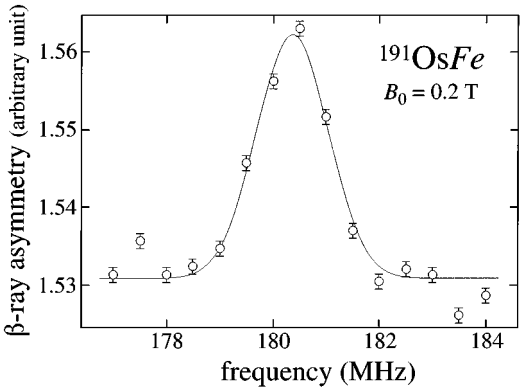

FIG. 9. β-NMR-ON spectrum of ^{191}Os*Fe* taken with FM of $\pm$ 0.5 MHz by detecting the β-ray asymmetry of $N(0°)/N(180°)$ (arbitrary unit) with respect to the orientation axis (the direction of the external magnetic field).

$-110.63(3)$ T from the spin echo NMR resonance at 371.0(1) MHz [13] and the known moment $\mu(^{189}\text{Os}) = 0.659933(4)\ \mu_N$ [14]. Large hyperfine anomalies are known in Os isotopes [13]. If we apply the empirical rule of Moskowitz and Lombardi [15] in the Hg region to ^{189,191}Os, a hyperfine anomaly $^{189}\Delta^{191}$ of 2.9% can be estimated. We therefore take an error of 2.9% on the hyperfine field and the resulting ^{191}Os magnetic moment is

$$\mu(^{191}\text{Os}) = +0.962(28)\mu_N .$$

This result is close to the calculation of 1.05 μ_N by Ekström and Rubinsztein [16] using Nilsson neutron orbit [505 $\frac{9}{2}$] and $g_s = 0.6\ g_s$(free).

IV. CONCLUSION

In on-line nuclear orientation there are long decay chains and in same cases nuclei have strong β decay with little γ activity. It is not easy to identify β decaying nuclei in complex decays and in such cases NMR-ON resonance signals have to be detected in, e.g., γ decay of daughter nuclei. When the nuclear spin-lattice relaxation time is comparable with the lifetime of the daughter nuclei reorientation in the successive decays depends on these times and upon the relative sign of successive hyperfine interactions.

We have demonstrated the practicality of measuring parent resonances *via* daughter γ decay in determining the magnetic moments of ^{89}Zr and ^{191}Os. We have shown that in the spin inversion case the anisotropy of the daughter γ rays is increased by the NMR-ON resonance whilst in the spin noninversion case it is decreased. There is thus good sensitivity to the relative sign of the parent/daughter hyperfine interactions. These results confirm the predictions of Stone [2]. He showed that the time evolution of the B_2 orientation parameter is extended in the sign inversion case and shortened in the spin noninversion case by a TR-OLNO study of the ^{183}Au-^{183}Pt^m decay sequence. Combinations of NMR-ON technique with the TR-OLNO method [2] in successive decays can be developed to study nuclear structure by on-line nuclear orientation.

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