Large hyperfine anomaly between ^{91}Y and ^{91m}Y in Fe

K. Nishimura,[1] S. Ohya,[2] T. Ohtsubo,[2] I. Sato,[2] J. Goto,[3] T. Izumikawa,[4] M. Tanigaki,[5]
A. Taniguchi,[5] Y. Ohkubo,[5] Y. Kawase,[5] and S. Muto[6]
[1]*Faculty of Engineering, Toyama University, Toyama 930-8555, Japan*
[2]*Faculty of Science, Niigata University, Niigata 950-2181, Japan*
[3]*Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan*
[4]*Radioisotope Center, Niigata University, Niigata 950-2181, Japan*
[5]*Research Reactor Institute, Kyoto University, Kumatori, Osaka 590-0494, Japan*
[6]*Neutron Science Laboratory, KEK, Tsukuba 305-0801, Japan*
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Nuclear magnetic resonance on oriented nuclei was carried out using a sample of dilute ^{91}Y and ^{91m}Y in iron prepared by ion implantation of radioactive precursor Rb isotopes. For ^{91}YFe, the beta-ray asymmetry was utilized to measure the magnetic hyperfine interaction frequency, $\nu_0 = 73.566(22)$ MHz, which led to the magnetic hyperfine field, $B_{\mathrm{HF}} = -29.4(2)$T, by using the known nuclear g-factor. The field shift of the resonant frequency, $d\nu_0/dB_0 = -2.501(17)$ MHz/T for ^{91}YFe, implies a Knight shift value, $K = 0.0(8)\%$. The interaction frequency and the field shift of ^{91m}YFe obtained via the gamma-ray detection confirmed the previously reported results. A ratio of the measured ν_0 and $d\nu_0/dB_0$ values for both ^{91}YFe and ^{91m}YFe gave rise to the hyperfine anomaly, $^{91}\Delta^{91m} = -4.2(8)\%$, which can explain discrepancies in the B_{HF} values of YFe as determined by various experiments.

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When a probe atom is located as a dilute impurity substitutionally in a ferromagnetic host matrix such as Fe, Ni and Co, the impurity nucleus will, in general, experience a magnetic hyperfine field B_{HF}.[1] The main contribution to B_{HF} originates from the Fermi contact interaction, and other noncontact interactions are usually small for 3d and 4d element impurities. The technique, nuclear magnetic resonance on oriented nuclei (NMRON), has been well established as a powerful method for the study of hyperfine interactions at dilute impurity nuclei in Fe, Ni and Co.[2] Since NMRON is the radioactive detection technique, the magnetic hyperfine interaction frequency, $\nu_0 = |g\mu_{\mathrm{N}}B_{\mathrm{HF}}/h|$, where g is the nuclear g-factor, in a broad range of elements and nuclear states has been investigated. In this method, an externally applied magnetic field B_0 is usually employed to polarize a ferromagnetic host, so the magnetic hyperfine-splitting frequency ν is given by,

$$\nu = \nu_0 + \mathrm{sgn}(B_{\mathrm{HF}})|g|\mu_{\mathrm{N}}(1+K)B_0/h,$$

where a parameter K implies the effects of Knight shift and diamagnetic shielding. The ν_0 values can be obtained from field shift measurements by extrapolating ν versus B_0 data to $B_0 = 0$. The field shift, $d\nu/dB_0 = \mathrm{sgn}(B_{\mathrm{HF}})|g|\mu_{\mathrm{N}}(1+K)/h$, depends on the g-factor, the K parameter and the sign of B_{HF}, but is independent of the absolute B_{HF} value.

The magnetic hyperfine field of dilute Y in Fe has been measured in several experiments.[3–8] The first B_{HF} value was derived from a NMR measurement of the stable isotope (^{89}Y, nuclear spin-parity $I^{\pi} = 1/2^{-}$); $B_{\mathrm{HF}} = 28.5(5)$T.[3] Early attempts to observe NMRON for Y in Fe failed due to metallurgical problems. Recently, NMRON spectra of ^{87m,89m,91m}YFe were observed using implantation of precursor nuclei.[6] The B_{HF} value was deduced to be −30.67(36)T from the magnetic hyperfine interaction frequency and field shift analysis assuming $K = 0$.[6] The magnitudes of these two values are inconsistent given the quoted errors. The authors[6] in the NMRON case claimed that the discrepancy was possibly due to sample preparation problems in the other experiments (high concentration is usually blamed), although they also suggested that an anomalously large Knight shift for Y in Fe could explain the difference between the B_{HF} values. More recently, NMRON of ^{87m}YFe was observed for a sample produced by direct implantation of Y at room temperature, which supported the previous ^{91m}YFe result.[8]

This work aims to investigate the magnetic hyperfine field and the Knight shift of Y in Fe with sufficient precision to resolve the discrepancy between the B_{HF} results of previous NMR and NMRON experiments. It is possible to derive both the magnetic hyperfine field and the Knight shift from the field shift data if the relevant nuclear g-factor is established independently. This is the case for ^{91}YFe; the nuclear g-factor of the ^{91}Y ground state has been measured precisely via an atomic beam experiment.[9] This probe nucleus ($I^{\pi} = 1/2^{-}$), unfortunately, decays without gamma emission, ruling out the more usual NMRON detection by gamma-ray anisotropy. Instead NMRON of ^{91}YFe is observed by detecting change in the beta-ray asymmetry. From the resonant frequency versus the external field shift thus observed, the Knight shift is found to be quite small. Gamma-ray detected NMRON of ^{91m}YFe was also carried out. Both samples were made in a similar manner. The resulting values of ν_0 and $d\nu_0/dB_0$ for ^{91}YFe and ^{91m}YFe indicate that the hyperfine anomaly could be large enough to explain the difference in the experimentally reported B_{HF} values.

Good sample preparation is essential for NMRON experiments. For this work the radioactive decay chain, ^{91}Rb $\rightarrow$ ^{91}Sr $\rightarrow$ ^{91m}Y $\rightarrow$ ^{91}Y $\rightarrow$ ^{91}Zr, was utilized in combination

with ion implantation. For detailed studies of sample preparation using ion implantation see Refs. 6 and 8. The ion implantation was carried out using the helium-jet loaded online isotope separator at the Research Reactor Institute, Kyoto University (KUR-ISOL). Radioactive ^{91}Rb nuclei were produced as fission products from a ^{235}U target in a neutron beam. After mass separation, the ^{91}Rb nuclei were implanted into an Fe foil (thickness 30 μm) with an accelerating voltage of 100 kV at room temperature. Details of the work at KUR-ISOL have been given elsewhere.[10]

The part of the Fe foil containing the activity was cut out, soft-soldered to the copper cold finger of a ^{3}He/^{4}He dilution refrigerator, and cooled to the base temperature of about 10 mK at Niigata University. The temperature of the sample was monitored with a ^{54}MnNi nuclear thermometer. A vertical external magnetic field B_0 up to 2.5 T, provided by a Nb_3Sn superconducting solenoid, was used in the ^{91}YFe case. A Si detector, mounted on a heat shield of 0.7 K inside a cryostat at 0° with respect the B_0 direction, provided the beta-ray signals. For the ^{91m}YFe measurements we used a pair of Helmholtz-type split coils to produce a horizontal B_0 (up to 1.0 T), allowing for the placement of four high-purity Ge detectors outside the cryostat both parallel and perpendicular to the B_0 direction to monitor the gamma rays. Consequently, the overall gamma-ray count rate was greatly improved. This configuration, at Niigata University, has been described previously in detail.[11] The gamma-ray anisotropy was obtained from the ratio of peak counts at the axial detectors to those of the equatorial detectors; no normalization of cold counts to warm counts was made in the NMRON spectra. NMRON measurements were performed applying a rf oscillating field with frequency modulation (FM) at 300 Hz perpendicular to the static field, B_0, and parallel to the foil plane.

The ground state of ^{91}Y decays via the first forbidden beta-decay to the ground state of ^{91}Zr with the Q_β value of 1544 keV and the branch ratio of 99.7%. NMRON spectra were acquired via beta-ray detection with $B_0=0.2$, 0.6, 1.0, 1.5, 2.0 and 2.5 T as shown in Fig. 1, where center frequencies obtained by Gaussian fits (solid line) of the data are 73.09(5), 72.06(2), 71.06(2), 69.84(3), 68.55(3), and 67.32(4) MHz, respectively. The large range of external fields B_0 was required to extract a precise value of the field shift given the small g-factor. Additionally use of high fields avoids potential problems when assigning Knight shifts to NMRON field shift data in Fe taken with $B_0<1$ T only.[12] The extracted values of the magnetic hyperfine interaction frequency and the field shift are, $\nu_0=73.566(22)$ MHz, and $d\nu/dB_0=-2.501(17)$ MHz/T, respectively. Taking the ^{91}Y nuclear g-factor as $-0.3282(16)$,[9] the magnetic hyperfine field of YFe and the Knight shift are deduced to be $B_{\mathrm{HF}}=-29.4(2)$T and $K=0.0(8)$%, respectively.

NMRON of ^{91m}YFe was recorded via the gamma-ray transition of 556 keV from the metastable state of the nuclear spin, $9/2^+$, to the ground state of spin, $1/2^-$ (M4 multipolarity). The spectra with $B_0=0.2$, 0.4, 0.6, 0.8 and 1.0 T are shown in Fig. 2, and the center frequencies were obtained as $\nu=307.89(3)$, 305.84(8), 303.99(7), 302.02(8), 299.71(16) MHz, respectively. A least-squares fit of the

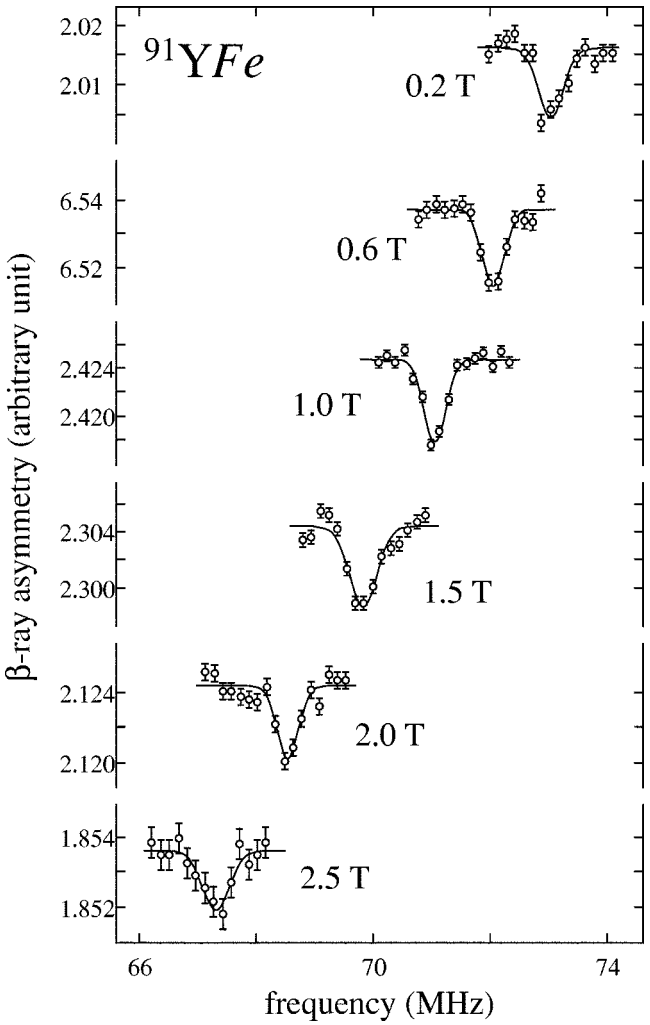

FIG. 1. Beta-NMRON spectra of ^{91}Y in Fe for external applied fields of 0.2, 0.6, 1.0, 1.5, 2.0, and 2.5 T. The solid lines are results of the fit the data point to a Gaussian function.

resonant frequency vs the external field yields the magnetic hyperfine interaction frequency and the field shift; $\nu_0=309.85(4)$ MHz, and $d\nu/dB_0=-9.80(22)$ MHz/T, respectively. These values are close to those reported previously for this system; $\nu_0=310.06(5)$ MHz, $d\nu/dB_0=-10.17(8)$ MHz/T (the highest $B_0=2.0$ T),[6] and $\nu_0=309.54(6)$ MHz.[7] Combining these values with the 556 keV gamma-ray anisotropy, the good-site fraction of ^{91m}YFe was found to be about 95%, which is larger than the earlier value of 86%.[7] The calculated destruction of the anisotropy (intensity of the NMR signal, corrected for the FM width) was about 23% for the spectrum at $B_0=0.2$ T.

The present B_{HF} value for ^{91}YFe, $-29.4(2)$T, is close to the value $(-)28.5(5)$T for ^{89}YFe from NMR; both the isotopes have the nuclear spin, 1/2. These B_{HF} values are somewhat lower than that of $-30.67(36)$T for ^{91m}Y (spin, 9/2) in Fe. The Knight shift value estimated in this work rules out the possibility for the anomalously large Knight shift for Y in Fe, suggested by Ref. 6. However, the present results of ν_0 and $d\nu/dB_0$ for ^{91m}YFe are found to be consistent with those of previous NMRON experiments. Indeed there is broad

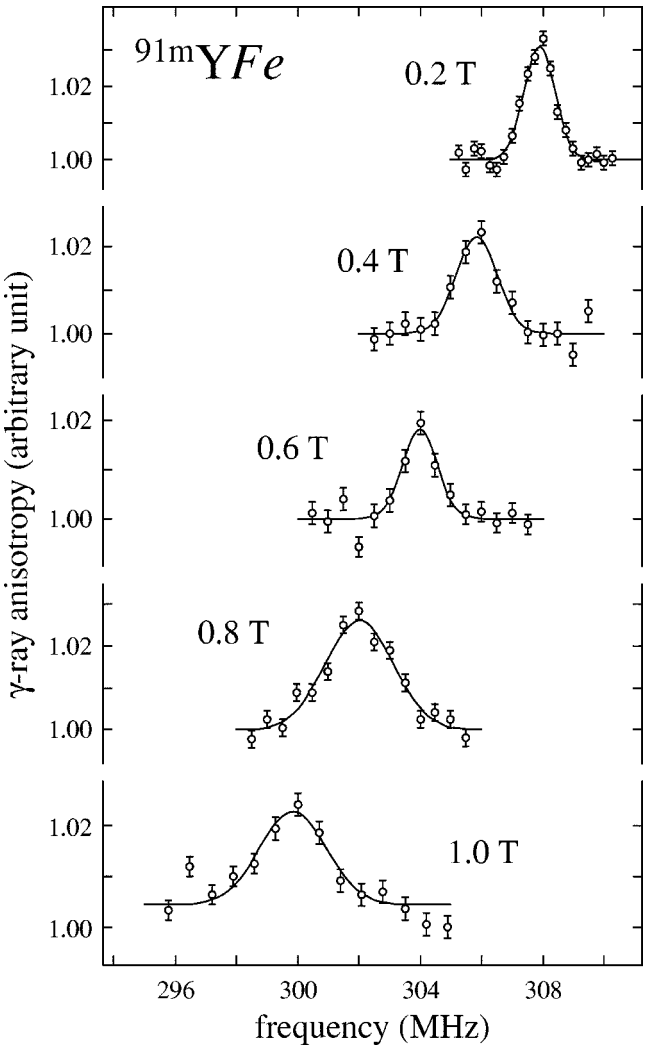

FIG. 2. NMRON spectra of ^{91m}Y in Fe for five different fields observed via 556 keV gamma transition.

agreement for the extracted hyperfine fields across all YFe studies involving the $I=9/2$ isotopes like ^{91m}Y and, separately for the $I=1/2$ isotopes ^{91}Y and ^{89}Y. It is these two data sets that are distinct, differing by some 4%. A possible origin of the difference between such B_{HF} values is the hyperfine anomaly Δ.

The influence of the nuclear magnetization distribution on the magnetic hyperfine interaction was first formulated by Bohr and Weisskopf.[13] In most cases actual effective B_{HF} values are smaller than those expected for the point dipole interaction, i.e., $B_{\mathrm{HF}}=B_{\mathrm{HF}}^{0}(1+\epsilon)$, where B_{HF}^{0} represents a point dipole interaction field at the origin and a parameter ϵ is mostly a negative value less than 1%. The magnitude of ϵ, however, can be rather large when the spin and the orbital contribution to the magnetic moment has opposite sign such as $p1/2$ and $d3/2$ nuclear states, i.e., $j=\ell-1/2$. Since the ground state of ^{91}Y probably has a $\pi(p1/2)$ nuclear configuration, and the metastable state of ^{91m}Y, a $\pi(g9/2)$, the Δ magnitude is considered to be large. Large hyperfine-anomaly values of about $(\pm)15\%$ have been reported for heavy elements, e.g., W isotopes in Fe.[14,15]

In NMRON experiments, neither B_{HF}^{0} nor ϵ are measurable, but the hyperfine anomaly $^{1}\Delta^{2}=(\epsilon_1-\epsilon_2)/(1+\epsilon_2)\approx\epsilon_1-\epsilon_2$ can be estimated from the relation:

$$^{1}\Delta^{2}=\frac{\nu_0^1}{\nu_0^2}\frac{d\nu^2/dB_0}{d\nu^1/dB_0}-1,$$

for two isotopes or two nuclear states in the same system. All the quantities appearing in the relation are directly determinable from NMRON measurements. Substituting the results of the current study into the above relation, the hyperfine anomaly is deduced to be

$$^{91}\Delta^{91m}=\frac{\nu_0^{91}}{\nu_0^{91m}}\frac{d\nu^{91m}/dB_0}{d\nu^{91}/dB_0}-1=-4.2(8)\%,$$

noting that the ν_0^{91m} and $d\nu^{91m}/dB_0$ values were recalculated using the present and reported[6] experimental data to be 310.005(23) MHz and −10.111(38) MHz/T for ^{91m}YFe, respectively. This recalculation reduced the errors of ν_0^{91m} and $d\nu^{91m}/dB_0$; consequently the error of $^{91}\Delta^{91m}$ was reduced by about 30%.

We note that our ^{91m}YFe field shift results have been collected for applied fields <1 T and therefore could be subject to scrutiny for possible small discrepancies in derived slope relative to the ^{91}YFe NMRON data set.[12] However as stated above, the parameters derived from these data are in quite good agreement with those of Ref. 6 for NMRON which was collected for applied fields up to 2 T. Variations in demagnetizing fields between individual (magnetic) hosts is a further issue in the assignment of hyperfine field values from NMRON data.[16] In this work we have been careful to prepare both types of specimen simultaneously and from the same Fe foil stock to mitigate any possible differences.

Comprehensive theoretical work with useful numerical tables for the Δ estimate has been reported by Stroke *et al.*[17] Fujita and Arima improved the expression for Δ to consider the core polarization and mesonic exchange currents.[18] If we take numerical values given by Stroke *et al.*[17] for the electron coefficients b and the radial integral values J relating with the electron density distribution inside a nucleus, a calculation with their formula for ϵ results in $\epsilon(^{91}\mathrm{Y})=-0.0169$ and $\epsilon(^{91m}\mathrm{Y})=-0.0040$, leading to $^{91}\Delta^{91m}=-1.3\%$. The single-particle model plus core polarization and mesonic exchange currents calculation using the expression by Fujita and Arima[18] gives a value, $^{91}\Delta^{91m}=-3.0\%$; $\epsilon(^{91}\mathrm{Y})=-0.0332$ and $\epsilon(^{91m}\mathrm{Y})=-0.0036$. Our result for the hyperfine anomaly supports this later estimation.

Since the hyperfine anomaly is rather large, the B_{HF}^{0} values, which are corrected for the effects of nuclear magnetism distribution, are preferable to the experimental B_{HF} for a comparison with theoretical ones. The ϵ values from the expression by Fujita and Arima[18] yield $B_{\mathrm{HF}}^{0}=-30.4(2)$T for ^{91}YFe and $B_{\mathrm{HF}}^{0}=-30.78(36)$T for ^{91m}YFe. These B_{HF}^{0} values strongly support the theoretical estimate of −29.4 T for Y in Fe based on *ab initio* band-structure calculations for $Fe_{15}X$ supercells, taking lattice relaxation effect into consideration.[19]

A combination of beta-/gamma-ray detections and ion implantation of the radioactive precursor isotope enabled NMRON of ^{91,91m}YFe, leading to precise values of the magnetic hyperfine field and the Knight shift. The origin of the difference between the B_{HF} values of $^{91}\mathrm{Y}(I^{\pi}=1/2^{-})$ and $^{91m}\mathrm{Y}(I^{\pi}=9/2^{+})$ in Fe has been explained by evoking a large hyperfine anomaly. Estimates for B_{HF} and Δ based on recent theories reproduce the present experimental values. It is hoped that these satisfactory results will stimulate further development in the theory to reproduce systematic variations of B_{HF} and Δ in Fe.

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