Measurement of the Hyperfine Field of Y*Fe* with Nuclear Magnetic Resonance on Oriented Nuclei after *In Situ* Production in Fe

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(Received 23 July 1990)

Nuclear magnetic resonance on oriented nuclei (NMR-ON) on Y as a dilute impurity in Fe was observed for the first time. As all attempts to observe NMR-ON resonances on Y*Fe* samples prepared by conventional techniques had failed, a new sample-preparation technique was applied. After the implantation of radioactive precursor isotopes and their *in situ* decay to Y, narrow NMR-ON resonances were observed. Our results indicate that all previous values for the hyperfine field of Y*Fe* in the literature do not represent the field at substitutional lattice sites.

PACS numbers: 75.50.Bb, 76.60.Jx, 76.80.+y

The magnetic hyperfine fields of most elements as dilute impurities in a ferromagnetic Fe host lattice are known with high precision. These hyperfine fields are interesting (i) *per se* for the understanding of magnetic properties of ferromagnets,[1] and (ii) for the determination of magnetic moments of short-lived nuclei from magnetic hyperfine-splitting frequencies using on-line nuclear-orientation techniques. Most known hyperfine fields originate from NMR measurements of the hyperfine splitting of stable isotopes as dilute impurities in Fe. For the detection of NMR a macroscopic amount of impurity nuclei is necessary; typical impurity concentrations are 0.1–2 at.%. An alternative technique is nuclear magnetic resonance on oriented nuclei detected via the anisotropy of radiation (NMR-ON).[2] Here, extremely small concentrations in the ppm region and below are sufficient for the resonance detection.

The measurements of dilute-impurity hyperfine fields require the preparation of stable or, at least, metastable alloys. With thermal methods, i.e., melting or diffusion, only metallurgically stable alloys can be prepared. Metastable alloys may be prepared with implantation techniques (recoil or mass-separator implantation), the stability of such alloys depending on the temperature. Such metastable alloys may have too short a lifetime even at room temperature. Low-temperature implantation might solve this problem. It is known, however, that with low-temperature implantation, lattice defects remain frozen, which, depending on the specific properties of the impurity-host system, may strongly influence the hyperfine interaction. Thus, a method would be desirable which allows the healing of the lattice defects, while simultaneously the impurity nuclei remain bound at the substitutional lattice sites with an undisturbed surrounding. These two requirements seem to be contradictory: On the one hand, thermal energy is necessary for the healing of the lattice defects. On the other hand, this thermal energy may be sufficient so that the impurity nuclei can leave the substitutional lattice sites or that lattice defects (or other impurities) are now trapped at the impurity nuclei.

Here we propose a new method to solve this problem: If the binding of the isotope of interest at substitutional lattice sites is too weak or if the probability for the trapping of lattice defects is too large so that a sample cannot be prepared by direct warm implantation, we implant a radioactive precursor for which the binding at substitutional lattice sites is strong enough and which does not trap lattice defects or other impurities. At lower temperatures, the precursor is allowed to decay to the isotope of interest, which remains bound with some probability at the substitutional lattice site with an undisturbed surrounding. The correct dilute-impurity limit for the hyperfine interaction can then be determined with NMR-ON.

The hyperfine field of dilute Y in Fe has been the subject of several spin-echo and nuclear-orientation measurements,[3–8] the results being inconsistent. Attempts to confirm the spin-echo result failed as it was found to be impossible to prepare dilute Y*Fe* alloys.[9] (Intermetallic compounds such as YFe_2 and YFe_3 exist, however, for which the magnetic properties are well known.) Systematic nuclear-orientation studies by Eder, Hagn, and Zech[10] showed that depending on the sample preparation and heat treatment different results are obtained, indicating that it is impossible to determine the substitutional hyperfine field of Y*Fe* with an integral measurement technique.[10]

The series of 4*d* elements (Y, Zr, Nb, Mo, Tc, Ru, Rh) as dilute impurities in Fe shows the following trend: Rh, Ru, Tc, and Mo are very soluble, and the alloys are thermally stable. Nb*Fe* alloys are less stable; NMR-ON measurements on ^{95}Nb*Fe* prepared thermally showed narrow resonance lines but weak resonance amplitudes, indicating that only a small fraction of Nb nuclei were substituted onto undisturbed lattice sites.[11] For implanted Nb*Fe* samples this fraction was found to be considerably larger.[12] Zr and Y are not soluble in Fe; it is not possible to prepare dilute alloys by thermal methods. Thus, samples of Y and Zr as dilute impurities in Fe can be prepared only by implantation techniques. As the binding of Y and Zr onto undisturbed substitutional sites in Fe seems to be rather weak, the new idea was to implant suitable radioactive Mo and Nb isotopes as precursors, for which the stability at substitutional sites is guaranteed, and let them decay to Y at lower temperatures. It could then be expected that a considerable fraction of the Y daughter nuclei remain at substitutional lattice sites with an undisturbed surrounding, allowing then the precise measurement of the hyperfine splitting with NMR-ON.

In this Letter we report the successful observation of NMR-ON on ^{87m}Y, ^{89m}Y, and ^{91m}Y in Fe. The samples were prepared by *in situ* decay after implantation of ^{87}Mo and ^{89}Mo, ^{87}Nb and ^{89}Nb, and ^{91}Rb. Our experiments show that all previous values for the hyperfine field of Y in Fe in the literature are erroneous.

In NMR-ON experiments, the anisotropy of the γ radiation is used as the detector for resonance; the resonance condition is given by

$$\nu = \nu_M + (|g|\mu_N/h)\,\mathrm{sgn}(B_{\mathrm{HF}})(1+K)B_0\,, \qquad \nu_M = |g\mu_N B_{\mathrm{HF}}/h|\,, \tag{1}$$

where ν_M is the magnetic hyperfine-splitting frequency, g is the nuclear g factor, B_{HF} is the hyperfine field, B_0 is the external magnetic field, and K is a parameter including the Knight shift and diamagnetic shielding. The measurement of the resonance frequency as a function of the external magnetic field B_0 yields ν_M from the extrapolation to $B_0=0$, and $g(1+K)$ from the slope. Thus the g factor can be determined from the resonance shift, independent of the knowledge of the hyperfine field, the uncertainty being given by the uncertainty of K. The parameter K has been measured for many different systems to be $|K| \lesssim 0.01$. Thus, with the assumption of $|K| \lesssim 0.01$ for Y*Fe*, the g factor *and* the hyperfine field can be determined from NMR-ON measurements with an accuracy of $\sim 1\%$.

Samples of ^{87m,89m}Y*Fe* were prepared with the recoil-implantation technique using a target stack consisting either of sixteen natural Y foils (thickness 5 μm) or sixteen ^{90}Zr foils (1.2 μm; enrichment 99%) each of which was followed by an Fe foil (1.5 μm; purity 99.999%). The target stacks were irradiated at the cyclotron in Karlsruhe with α particles: the Y-Fe stack (denoted in the following as sample I) for 2.5 h with an energy of 59 MeV and a current of 2.5 μA, and the ^{90}Zr-Fe stack (sample II) for 4 h with $E_\alpha = 100$ MeV and $I = 2.5$ μA. The following decay chains lead finally to Y nuclei:

$$^{89}\mathrm{Mo} \xrightarrow{2.2\ \mathrm{min}} {}^{89}\mathrm{Nb} \xrightarrow{2.0\ \mathrm{h}} {}^{89}\mathrm{Zr} \xrightarrow{78.4\ \mathrm{h}} {}^{89m}\mathrm{Y} \xrightarrow{16.1\ \mathrm{s}} {}^{89}\mathrm{Y}\,,$$

$$^{87}\mathrm{Mo} \xrightarrow{15\ \mathrm{s}} {}^{87}\mathrm{Nb} \xrightarrow{2.6\ \mathrm{min}} {}^{87}\mathrm{Zr} \xrightarrow{1.6\ \mathrm{h}} {}^{87m}\mathrm{Y} \xrightarrow{13\ \mathrm{h}} {}^{87}\mathrm{Sr}\,.$$

The compound nuclei with the kinetic energy of 3–4 MeV have a range of 0.2–0.3 μm; thus all nuclei which are produced in the rear surface area of the target foils are implanted homogeneously into the Fe foils.

The ^{91m}Y*Fe* sample (sample III) was prepared with the mass separator ISOLDE at CERN, implanting ^{91}Rb with an accelerating voltage of 60 kV into a 4-μm Fe foil. Here the following decay chain is relevant:

$$^{91}\mathrm{Rb} \xrightarrow{58\ \mathrm{s}} {}^{91}\mathrm{Sr} \xrightarrow{9.5\ \mathrm{h}} {}^{91m}\mathrm{Y} \xrightarrow{49.7\ \mathrm{min}} {}^{91}\mathrm{Y}\,.$$

After the implantations, the samples were soldered with GaIn to the Cu cold finger of a ^{3}He-^{4}He-dilution refrigerator with top-loading facility, and cooled to temperatures below 10 mK. The ferromagnetic domains of the Fe foils were polarized with an external magnetic field $B_0 = 0.1$–2.0 T. The γ rays were detected with four Ge detectors placed at 0°, 90°, 180°, and 270° with respect to the direction of B_0.

Sample I was cooled to temperatures below 10 mK within 4 h after the end of the irradiation. With $B_0 = 0.1$ T, at $T = 9.0(5)$ mK the following γ anisotropies $\varepsilon = W(0°)/W(90°) - 1$ were observed: ^{87m}Y, 381 keV, $-0.62(4)$; ^{89m}Y, 909 keV, $-0.59(2)$. Taking into account the NMR-ON resonance frequencies as given below, this is about 45% of the anisotropy which would be expected if all Y nuclei were on substitutional lattice sites. The NMR-ON resonances were searched for in the frequency region 210–330 MHz (frequency steps 3 MHz; modulation bandwidth ± 3 MHz) and detected at 313.8(5) MHz (^{87m}Y) and 322.6(2) MHz (^{89m}Y). In comparison to ^{89m}Y the resonance amplitude for ^{97m}Y was smaller by a factor of 4.7(7), which is probably due to the shorter half-lives of the Nb and Zr precursors in the $A = 87$ decay chain. Despite the small resonance effect, further measurements of the ^{87m}Y resonance were performed with a higher frequency resolution and a smaller frequency modulation bandwidth, for $B_0 = 0.1$ and 0.2 T. The resonance was found to be shifted from 313.56(16) MHz at 0.1 T to 312.7(2) MHz at 0.2 T, proving that a correct NMR-ON resonance had been observed. For ^{89m}Y the resonance centers were measured for $B_0 = 0.1$, 0.6, 1.1, and 1.6 T to be 322.69(4), 317.49(9), 312.13(36), and 306.87(21) MHz, respectively. Two NMR-ON spectra are shown in Fig. 1 (middle).

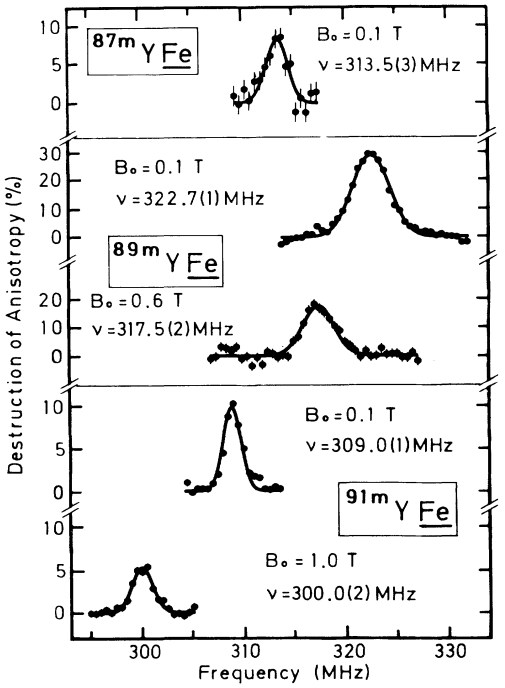

FIG. 1. Top: NMR-ON resonance of ^{87m}YFe (sample II, prepared via recoil implantation with a ^{90}Zr target at the cyclotron in Karlsruhe); linewidth $\Gamma = 2.2(3)$ MHz. Middle: NMR-ON resonances of ^{89m}YFe (sample I, prepared via recoil implantation with a Y target); $\Gamma = 3.7(1)$ MHz. Bottom: NMR-ON resonance of ^{91m}YFe [sample III, prepared with mass-separator implantation at ISOLDE (CERN) after implantation of ^{91}Rb]; $\Gamma = 2.2(2)$ MHz.

With sample II (prepared with the ^{90}Zr target) the γ anisotropies of ^{87m}Y and ^{89m}Y were only $\sim 50\%$ of the γ anisotropies observed with sample I (prepared with the Y target). An NMR-ON spectrum for ^{87m}YFe is shown in Fig. 1 (top). In comparison to the results obtained with sample I, the resonant anisotropy destructions were nearly doubled, the linewidths were smaller by $\sim 30\%$, but the center frequencies coincided well within the statistical errors (see Table I). This demonstrates that, in contrast to the temperature dependence of the γ anisotropy, which, as an integral technique, yields an average value for the hyperfine interaction, NMR-ON as a differential technique is highly sensitive only to nuclei on undisturbed substitutional lattice sites.

Two NMR-ON spectra for ^{91m}YFe (sample III) are shown in Fig. 1 (bottom). Measurements were performed for $B_0 = 0.1$, 0.5, 1.0, 1.5, and 2.0 T, with results of 309.02(3), 305.03(3), 300.00(7), 294.68(6), and 289.95(16) MHz, respectively.

The zero-field splittings $\nu(B_0 = 0)$ and the slope parameters $d\nu/dB_0$ were determined with least-squares fits; the results are compiled in Table I. The hyperfine fields as given in column 4 of Table I are derived with $K = 0$. The good agreement for ^{89m}Y and ^{91m}Y with their different precursor chains indicates that the hyperfine interaction of Y at undisturbed substitutional lattice sites in Fe has been observed. Assuming now that the uncertainty of K is 1% and adding the errors quadratically we get

$$B_{HF}(YFe) = -30.67(36)\ \mathrm{T},$$

which is different from all values in the literature. The only literature value which has to be considered seriously is the spin-echo result of 28.5(5) T.[3,4] The relative difference between these two values is 7.6(2.3)%, which must be explained. The following scenarios can be offered: (i) The spin-echo field is correct. Then the difference of 7.6(2.3)% has to be attributed to physical effects, such as hyperfine anomaly or Knight shift. An estimate of the hyperfine anomaly between the $\frac{1}{2}^-$ ^{89g}Y and the $\frac{9}{2}^+$ states yields an upper limit of 2%. Thus, at least 5.6% of the difference would remain. In principle, this could originate from an anomalously large Knight shift of YFe, which cannot be excluded *a priori* but which, in our opinion, is highly improbable. (ii) The spin-echo field does not represent the dilute-impurity

TABLE I. NMR-ON results for the hyperfine-splitting frequencies and resonance shifts. The hyperfine fields given in column 4 have been derived assuming $K = 0$ (see text).

System	ν_M (MHz)	$d\nu/dB_0$ (MHz/T)	B_{HF} (T)	$\mu/\mu(^{89m}Y)$
^{87m}YFe	314.65(13) [a]			
	314.58(12) [b]			
	314.61(9) [c]			0.9718(3)
^{89m}YFe	323.74(4) [a]	−0.1051(11)	−30.80(32)	
	323.73(5) [b]	−0.1034(19)	−31.31(57)	
	323.74(3) [c]			
^{91m}YFe	310.06(5)	−0.1017(8)	−30.49(24)	0.9577(2)
Average			−30.67(18)	

[a]Sample I: Y target.
[b]Sample II: ^{90}Zr target.
[c]Average value.

hyperfine field of Y in Fe. In the following we show that nuclear-physics arguments strongly support the second possibility.

The g factors derived with the hyperfine field of $B_{HF}=-30.67(36)$ T for ^{87m}Y, ^{89m}Y, and ^{91m}Y are 1.346(16), 1.385(16), and 1.326(16), respectively, while for $B_{HF}=-28.5(5)$ T, 1.448(25), 1.490(26), and 1.427(25) would be obtained. From a brute-force nuclear-orientation experiment, Marest, Haroutunian, and Berkes[13] report for ^{87m}Y $g=1.36^{+7}_{-3}$, which does not allow us to rule out one of the above possibilities. Häusser *et al.*,[14] however, reported measurements of magnetic moments in the $A=90$ region, which they could explain well with shell-model calculations. For example, for 8^+ ^{90m}Zr they report $g_{expt}=1.356(7)$, which is in excellent agreement with their theoretical value $g_{theor}=1.361$. Using their matrix elements, the subsequently measured g factor of 5^- ^{90m}Zr could also be explained well.[15] Assuming now ^{88}Sr to be the core, $\frac{9}{2}^+$ ^{89m}Y is a one-proton and 8^+ ^{90m}Zr a two-proton state. Häusser *et al.* quote $\delta g_{av}=-0.016$ for the additivity-violating contribution to the g factor of $|(\pi g_{9/2})^2\rangle_{8^+}$ ^{90m}Zr. According to their calculations, $g=1.372(7)$ is expected for ^{89m}Y, in excellent agreement with our value of 1.385(16), which is strong evidence that our hyperfine field $B_{HF}=-30.67(36)$ T is correct. These calculations exclude a g factor of 1.49 for ^{89m}Y, as would follow from the spin-echo field.

Thus we finally conclude that all hyperfine fields reported for *YFe* (Refs. 3 and 5–8) up to now do not represent the dilute-impurity hyperfine field, most probably due to metallurgical problems with the sample preparation. As demonstrated by our experiments, such problems can be overcome by *in situ* sample preparation via a suitable radioactive precursor combined with the highly efficient NMR-ON measurement of the hyperfine splitting. This new technique can probably also be applied to other elements for which no reliable data on the hyperfine fields exist up to now.

This work was supported by the German Federal Minister for Research and Technology (BMFT) under Contract No. 06 TM 108, and, partly, by the Kernforschungszentrum, Karlsruhe. We wish to thank Dr. P. Maier-Komor, K. Nacke, and W. Heimkes for the preparation of the targets, and E. Smolic, J. Hesol, H. Ratai, C. König, and R. Schmid for experimental help.

[1]M. Akai, H. Akai, and J. Kanamori, J. Phys. Soc. Jpn. **54**, 4246 (1985); **54**, 4257 (1985).

[2]E. Matthias and R. J. Holliday, Phys. Rev. Lett. **17**, 897 (1966).

[3]M. Kontani, K. Asayama, and J. Itoh, J. Phys. Soc. Jpn. **20**, 1737 (1965); M. Kontani and J. Itoh, J. Phys. Soc. Jpn. **22**, 345 (1967).

[4]Recalculated with $\mu(^{89}\text{Y})=-0.137\,418\mu_N$.

[5]G. Marest, R. Haroutunian, and I. Berkes, Hyperfine Interact. **4**, 425 (1978).

[6]A. L. Allsop, D. I. Bradley, V. R. Green, and N. J. Stone, Hyperfine Interact. **15/16**, 313 (1983).

[7]I. Berkes, M. Fahad, R. Hassani, M. Massaq, and N. J. Stone, Hyperfine Interact. **43**, 477 (1988).

[8]I. Berkes, O. El Hajjaji, M. Fahad, R. Hassani, J. Giroux, G. Marest, G. Marguier, N. J. Stone, J. Rikovska, and V. R. Green, in *Proceedings of the Fifth International Conference on Nuclei Far from Stability, Rosseau Lake, Ontario, Canada, 1987,* edited by I. Towner, AIP Conference Proceedings No. 164 (American Institute of Physics, New York, 1988), p. 193ff.

[9]P. C. Riedi (private communication).

[10]R. Eder, E. Hagn, and E. Zech (unpublished).

[11]M. Kopp and W. D. Brewer, Hyperfine Interact. **3**, 321 (1977).

[12]R. Eder, E. Hagn, and E. Zech, Nucl. Phys. **A451**, 46 (1986).

[13]G. Marest, R. Haroutunian, and I. Berkes, Phys. Rev. C **17**, 287 (1978).

[14]O. Häusser, I. S. Towner, T. Fästermann, H. R. Andrews, J. R. Beene, D. Horn, and D. Ward, Nucl. Phys. **A293**, 248 (1977).

[15]R. Eder, E. Hagn, and E. Zech, Nucl. Phys. **A468**, 348 (1987).