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Internal field distribution on Au in the Au-Fe system in the limit of very small concentration

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Nonsaturation of hyperfine fields on Au and Fe has been studied primarily with regard to the existence of the Aharoni angle. With the use of both nuclear orientation and magnetic resonance on ^{189m}Au in Fe, the hypothesis of a conical field distribution is shown to be unfounded—even at very moderate external magnetic field values—for well-prepared samples with small Au concentration. [^{181}Ta(^{12}C,xn)^{189m}Au, mass separation, ^{3}He-^{4}He dilution refrigeration, γ anisotropies, Ge detection].

The Au-Fe system, at almost any possible concentration ratio, has been studied intensively for several decades with a great variety of techniques. This system displays, indeed, a rich variety of effects from a Kondo system to a spin glass of various phases to a ferromagnet. We concentrate here on the behavior of Au at very low concentration in an Fe matrix, studied by integral low-temperature nuclear orientation (NO) measurements and nuclear magnetic resonance on oriented nuclei (NMR-ON).

An unresolved problem for heavy impurities in iron—necessary to explain observed data—is the need to assign nonunique hyperfine field distributions for the impurities in the host lattice. In practice one often resorts to a two-site model where some impurities feel the full hyperfine field while the others are not subject to any field at all. However, a not less possible solution has often been invoked for heavy impurities in iron: a cone angle between the internally distributed hyperfine field and the externally applied field. The cone angle was first introduced by Ben-Zvi *et al.*[1] in 1967 and supported by further work.[2-4] Theoretical studies were performed by Aharoni, Andriessen, and Postma.[5-7] We will show that through careful sample preparation the problem of a nonunique hyperfine field can be greatly reduced in significance, if not made to vanish altogether.

The results reported here were obtained using continuous ion implantation (in the ppm range) of Au isotopes into an iron matrix at an implantation temperature below 20 mK (for technical details of the setup, see Ref. 8). The $\frac{11}{2}^-$ isomeric states of Au are potentially useful probes since the magnetic moments are large and remarkably constant for varying mass number.[9-11] For example, the ratio of the magnetic moments $\mu(^{193m}\mathrm{Au})/\mu(^{195m}\mathrm{Au})$ is 1 to within about 0.1%, and it is accepted that, in the case of isolated proton states, the addition of neutrons has negligible influence on the magnetic moment value. For ^{197m}Au a slightly higher value was measured by Ligthart and Postma[4] using integral NO data, but a later NMR-ON measurement disproved[10] even this small upward trend. Lighthart[4] claimed a strong downward trend in ^{191m}Au, which was disproved again by an elaborate relaxation analysis of our NO data.[12] The very small lifetime ($t_{1/2}$=0.9 s) of the $\frac{11}{2}^-$ state makes ^{191m}Au a less than ideal hyperfine probe. All the other heavier odd Au isotopes have also short lifetimes (respectively, 3.9, 30.5, and 7.8 s for ^{193m}Au, ^{195m}Au, and ^{197m}Au) which necessitates additional corrections for relaxation effects to integral NO measurements. In this respect ^{189m}Au seems to be an ideal candidate due to its relatively long half-life of 4.6 min. On the other hand, the even Au isotopes have long lifetimes but small magnetic moments. The same is true for the ground states of the longer-lived odd Au isotopes. Hence, despite the large variety of potential probes only the ^{189m}Au $\frac{11}{2}^-$ isomeric state has long enough lifetime and a well-enough fixed decay scheme to allow a meaningful integral NO study—free of discussions on relaxation mechanisms and with large orientation effects in order to decorrelate the magnetic hyperfine interaction and the implantation behavior. For preparing samples we use on-line mass separation and implantation into a cooled high-purity Fe foil. This has the beneficial effect that the implanted dose is contaminant free and normally even below the 1 ppm level.

It should be kept in mind that NMR-ON studies are amenable to a very precise magnetic moment and/or substitutional hyperfine field determination: They lack, however, the possibility to study possible field distributions since they only deal with the substitutionally implanted atoms. Although defect structures have been reported occasionally in NMR-ON studies by us and others,[13,14] experimental tests on, e.g., ^{129m}Xe and ^{131}I in Fe single crystals show that these studies are restricted to a few favorable cases of well-defined monovacancy complexes. We therefore concentrated mainly on integral NO measurements of ^{189m}Au*Fe* in the initial phase of this study.

Figure 1 shows the recorded effect—defined as $W(0°)/W(90°)$—of the 166-keV transition in the ^{189m}Au decay as function of T^{-1}. As indicated before, the large effect and the strong saturation behavior together with the high statistical accuracy allow an independent determination of the nuclear moment as well as of the substitutional fraction. The latter refers to a simple hypothesis, in which we suppose that substitutionally landed Au isotopes experience the full hyperfine field, while the other ones (due to neighboring defect structures) do not contribute to the orientation effect. The fit to the data resulted in a substitutional fraction α=0.78(1) and a magnetic moment value of 6.22(20)μ_N.

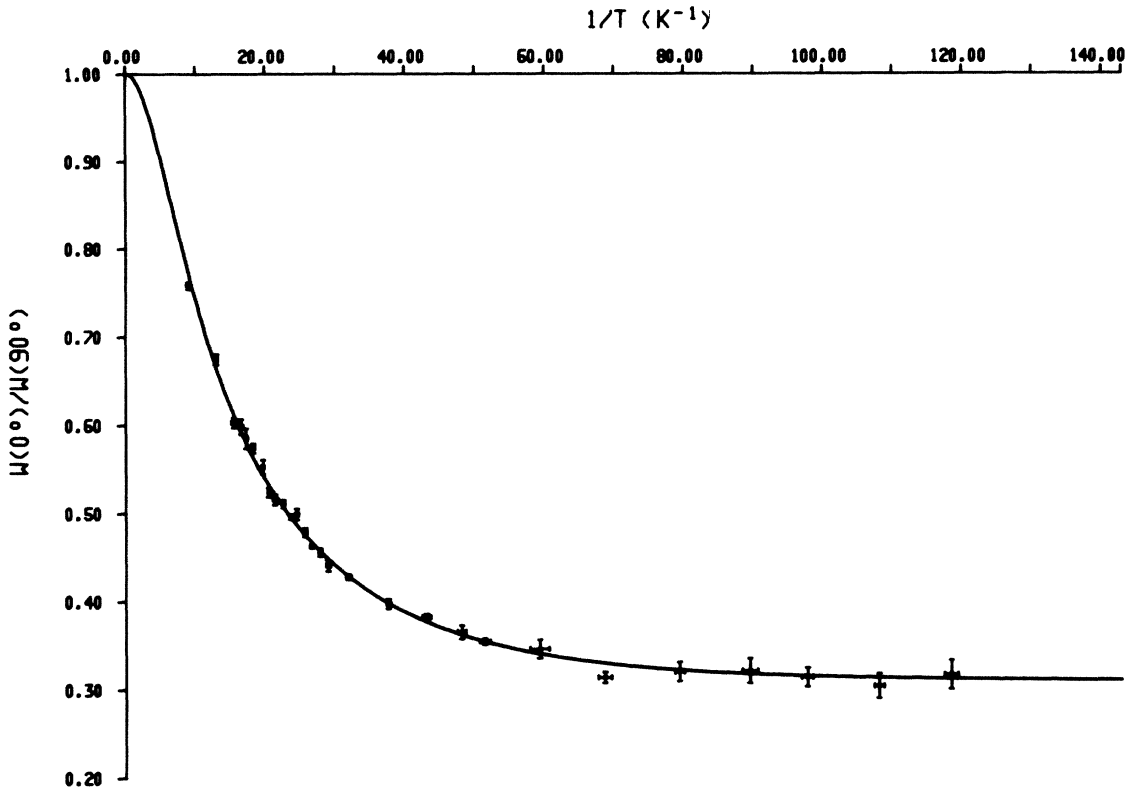

FIG. 1. The anisotropy on the 166-keV line.

The latter moment value is in good agreement with the values of the corresponding $\frac{11}{2}^-$ states in heavier isotopes ^{193m}Au, ^{195m}Au, ^{197m}Au. On the other hand, the substitutional fraction is low and points to a more complex internal field distribution. It is seemingly supported also by the fact that the substitutional fraction of ^{189m}Au*Fe* corresponds exactly to the one obtained in our study[12] of ^{191}Au, derived from a ^{191m}Hg implantation. However, both experiments contradict our previous experience with other soluble elements after low-temperature implantation at low dose.[15] Moreover, room-temperature implantations of both Au and Hg have been reported[16] leading to α values of about 90% measured in the same field we operate in (0.5 T).

Therefore, a possible explanation was looked for in a partial misalignment of the hyperfine field with respect to the external field. The noncollinearity of internal and external magnetic field—even in the case of full saturation—has been explained theoretically by Aharoni[6] by introducing effects of magnetostriction. The possibility of unsufficient external field for saturation was stressed by Krane, Murdoch, and Steyert.[2,3] In a more recent theoretical study it was pointed out that the Aharoni continuum model approach neglects exchange interactions. Introducing the latter an (unsuccessful) attempt was made to explain the difference in misalignment angle between Au and Hg.[7]

Unsufficient saturation and a corresponding distribution of direction in the hyperfine field are normally accounted for by introducing an average of the Legendre polynomials $P_k(\cos\theta_0)$, depending on the cone angle θ_0. From the saturation value of the 166-keV anisotropy one can, independently of the magnetic interaction parameter, deduce an experimental cone angle θ_0 of 18.9(1)°, which corresponds very well with the angle found by Ligthart for ^{193m}Au*Fe*.[4] In general, it agrees well with most cases where this effect was postulated and in which angles from 10° to 30° have been derived. Moreover, a new fit for the whole temperature region within the extended NO formalism results in $\alpha=1.00(1)$ and $\mu=5.84(15)\mu_N$. The moment value is still in rough agreement with the behavior of other $\frac{11}{2}^-$ isomers, while the α value corresponds to our expectations concerning cold implantation.[15] Unfortunately, a good fit using a cone angle does not prove that the hypothesis of field distribution is right, since a smaller α and a still acceptable μ lead to an equally good fit.

In order to verify the hypothesis of field distribution, we tried to fix the value of the moment of ^{189m}Au independently using NMR-ON. For this we need a high count rate on ^{189m}Au decay. The small production rate of ^{189m}Au and the limited beam time near a heavy ion accelerator make this a rather difficult endeavor. Moreover, because we implant in Fe, the search for resonance had to be performed around 1 GHz, where the loss of power in the transmission lines becomes high. In order to maximize the effect, any contamination or oxidation of the surface had to be removed carefully because of the very small skin depth. Therefore the surface was treated not only by the normal chemical etching and cleaning procedures, but also by thorough polishing to very small grain size.

The resonance was searched for in the region of 950–1010 MHz in steps of 3 MHz with a modulation width of 2.5 MHz. During continuous implantation and rf irradiation the temperature could be kept continuously below 20 mK. Hence the effect of the 166-keV transition remained in the saturation region. As an additional check, the temperature was monitored continuously using a ^{60}Co*Co* single crystal thermometer while the frequency region was repeatedly scanned up and down, alternating modulated and unmodulated rf-irradiation cycles with a time lapse between the cycles larger than the relaxation time of ^{189m}Au*Fe*. A broad resonance, shown in Fig. 2, was located with a center frequency of 978(18) MHz. Correcting for the external field and using the hyperfine field of Ref. 9 deduced for a $\frac{11}{2}^-$ Au state, we find $\mu(^{189m}\text{Au}) = 6.17(15)\mu_N$. Hyperfine anomalies for the $\frac{11}{2}^-$ states were shown[17] to be very small and can safely be disregarded. The moment value is in better agreement with the "two-site" approach but is not sufficiently accurate to discard the "cone-angle" hypothesis. Therefore, we planned a short integral run with an iron sample that was prepared the same way as during the NMR-ON experiment. Surprisingly, the saturation value in Fig. 3 corresponds to $\alpha = 0.96(1)$, a much higher substitutional fraction than during the first integral run. This, together with the moment value indicates that the lower implantation fraction in the initial experiments was only related to insufficient surface treatment.

The need for an explanation in terms of "misaligned" internal fields hence disappears with improved sample preparation. The question of sufficient external saturating field also cannot be separated from problems involving sample treatment. Especially when using ion implantation of heavy nuclei, surface magnetism is involved which can be correlated to the behavior of the bulk only for carefully prepared surfaces.

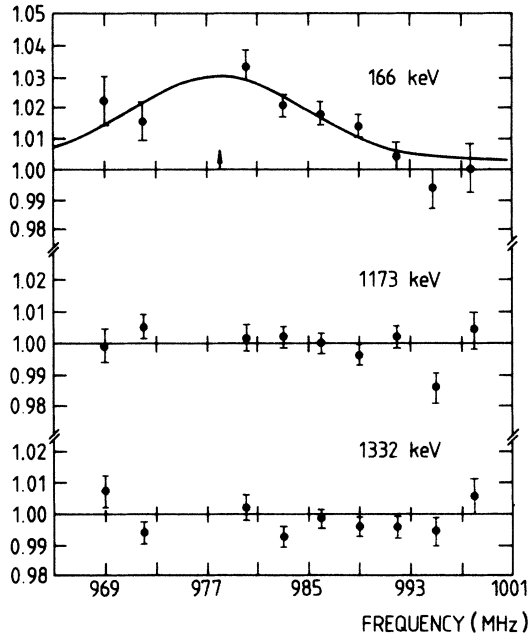

FIG. 2. The resonance on the 166-keV line. The nonresonant behavior on the 1173- and 1332-keV lines of ^{60}Co is shown on the lower curves.

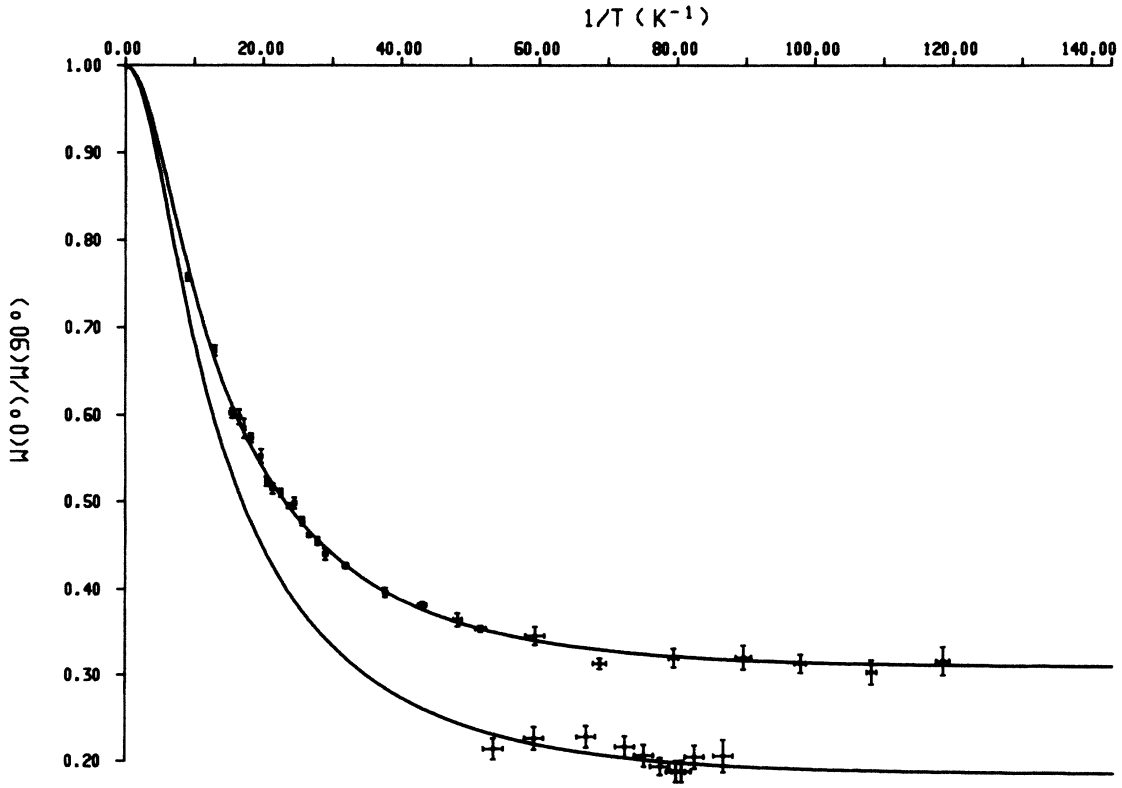

FIG. 3. Comparison of the anisotropies on the 166-keV line in two integral experiments with different sample preparation. The upper curve leads to $\alpha = 0.78(1)$, while the lower indicates $\alpha = 0.96(1)$.

Our conclusion from this work and many implantations at the KOOL and FOLBIS facilities[18] is that 100% substitutional implantations at low temperature can be achieved if the implanted surface is carefully prepared and if the implantation dose is kept low enough. Especially for the Au-Fe system there is no need to invoke a conical or other internal field distribution even at moderate (<0.5 T) fields. An increase of dose to 1 at.% may already lead to deviations from the really dilute case. An indication for this was pointed out for the ^{197}Au*Fe* system, which was studied with conventional NMR. Indeed, conventional NMR normally necessitates a Au content substantially above the implantation doses of ^{189m}Au reported here, which in turn leads to anomalies in the hyperfine splitting.[10]

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