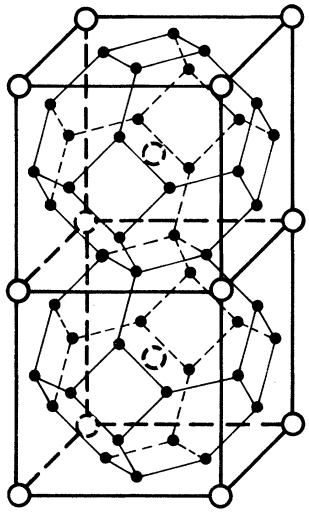

FIG. 3. Tetrahedral interstitial positions in a bcc lattice.

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Approach to Magnetic Saturation of Impurities in Iron: Effects on Nuclear Alignment, Perturbed Angular Correlation, Mössbauer, and γ-Ray Thermometry Measurements*

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Dilute alloys of Mn, Co, Ta, and Ir radioisotopes with iron were cooled to 25 mK; the impurity nuclei were oriented through applied magnetic fields up to $H=15.4$ kOe. At $H\approx 1$ kOe, sufficient to saturate the iron samples, nonalignments of nuclear hyperfine field with $\vec{H}$ of magnitude $\Theta\approx 20°$ were observed with $\sin\Theta\propto 1/H$. Errors in perturbed-angular-correlation measured g factors (10–40%), nuclear alignment parameters (10–30%), and ultralow-temperature Mössbauer and γ-ray thermometry (2 mK) due to insufficient H are discussed.

The large hyperfine fields associated with dilute impurities in ferromagnetic hosts are used in a number of research areas. However, unexplained discrepancies have been noted in nuclear g factors deduced from perturbed-angular-correlation (PAC) measurements,[1] nuclear alignment measurements,[2] in nuclear γ-ray anisotropy thermometry[3] (and its use to measure properties of

^{3}He down to 4 mK[4]) and in comparisons of Mössbauer and γ-ray anisotropy thermometry.[5] These peculiarities are primarily due to the failure to achieve magnetic saturation in the vicinity of the *impurity* atom.

The nucleus of an impurity either dissolved or implanted in a ferromagnetic material will be subjected to magnetic hyperfine fields of up to 10 MOe. If the ferromagnetic host is magnetically saturated by an externally applied magnetic field $\vec{H}$, it might be assumed that the hyperfine fields are aligned in the $\vec{H}$ direction. However, as discussed by Aharoni,[6,7] there is a tendency for impurities to orient along certain crystalline axes due to magnetostrictive, magnetocrystalline, exchange, and magnetostatic forces. This effect has been observed by Ben-Zvi *et al.*,[8] with Coulomb-excited ^{186}W and ^{148}Nd impurities recoil implanted into polycrystalline iron and nickel foils.

The inset in Fig. 1 shows the samples S which

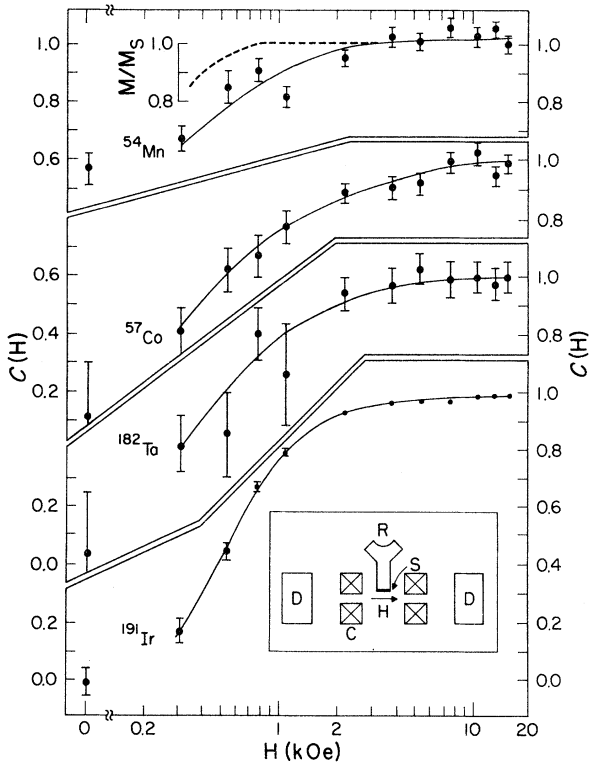

FIG. 1. Normalized orientation parameter $\mathfrak{C}(H)$, as defined in Eq. (1), versus applied field H. For ^{54}Mn, $E_\gamma = 835$ keV; for ^{57}Co, $E_\gamma = 122$ and 136 keV; for ^{182}Ta, $E_\gamma = 179$ and 264 keV; for ^{191}Ir, $E_\gamma = 129$ keV. The curves indicate approximate fits to the data. Also shown are the coinciding measured and theoretical relative magnetic saturation curve for the iron samples, and a schematic of the experimental arrangement.

are cooled to $T = 25$ mK by a ^{3}He-^{4}He dilution refrigerator R in a field $\vec{H}$, homogenous to 5% across the sample, produced by a pair of superconducting coils C. The γ-ray counting rates along the axis of the applied field were measured with two 40-cm^3 coaxial Ge(Li) detectors D as H was varied from 0 to 15.4 kOe. The sample S consisted of three iron foils, about 0.1 mm thick, about 2 mm wide, and 6 mm long, side by side to form a mosaic 0.1 mm thick. The long axes of the three foils were aligned in the $\vec{H}$ direction. Of the three 99.99%-pure Fe foils, one had ^{57}Co and ^{54}Mn diffused into it at 1200°C in a hydrogen atmosphere. Another iron foil contained 0.25 at.% natural Os, the third 0.07 at.% natural Ta. The Fe(Os) and Fe(Ta) foils were irradiated with thermal neutrons to form radioactive ^{191}Os and ^{182}Ta, respectively, and annealed at 900°C in H_2. The ^{191}Os activity is the parent of ^{191m}Ir ($\tau_{1/2} = 4.9$ sec) and, since the spin-lattice relaxation time of ^{191m}Ir in Fe is less than 0.1 sec (i.e., much less than $\tau_{1/2}$), the measured γ-ray anisotropies for this isotope will be characteristic of the ^{191m}Ir polarization. Metallographic examination of the three foils after the experiments showed no precipitation of impurities. Experiments at $T = 1$ K showed source motion due to $\vec{H}$ to be effectively eliminated by mechanical stabilization.

The angular distribution of γ radiation, normalized to the isotropic high-temperature limit is given by $W(\theta) = 1 + B_2 A_2 P_2(\cos\theta)$. (The contribution of the P_4 term can be neglected to an accuracy of better than 0.5%.) Here θ is the angle between the direction of $\vec{H}$ and the detector axis. (In the present case, the detector axes are parallel to the applied field.) For simplicity all parameters not depending on the nuclear polarization are included in the parameter A_2. Figure 1 shows the normalized coefficient

$$\mathfrak{C}(H) = B_2(H)/B_2(15.4\ \text{kOe}), \quad T = 25\ \text{mK}, \qquad (1)$$

where H is the externally applied field. Since the normalized $\mathfrak{C}(H)$ should be the same for two γ rays of a given isotope, in some cases we have averaged $\mathfrak{C}(H)$ for two γ rays of the isotope. The values of E_γ are listed in the caption to Fig. 1. A small correction was made to $\mathfrak{C}(H)$ to account for the fact that the nucleus feels the sum of the hyperfine field and the applied field, the two fields being in approximately opposite directions for the nuclei; e.g., if the applied field is 10 kOe and the internal field is -200 kOe the resultant field is -190 kOe at an impurity nucleus.

An ac hysteresis measurement on the samples

yielded the magnetization curve shown in Fig. 1. Magnetic saturation of the iron is observed when $H \geq 1$ kOe. Theoretical predictions for nearly pure polycrystalline iron yield similar results.[9-11]

For interpretation of the results, the nuclei are taken to be subject to a constant hyperfine field lying not along $\vec{H}$, but instead along the generating vector of a cone, making a polar angle of Θ with H. The angle Θ is a spatial rms average of the spectrum of angles defined by Aharoni,[6,7] and, with the exception of the Mössbauer experiments, is the precise quantity used in the equations under discussion (for small Θ). With the observation from Fig. 1 that the 15.4-kOe applied field essentially saturates $\mathfrak{C}(H)$, we can write

$$B_2(H)A_2P_2(\theta=0) = B_2(15.4\ \text{kOe})A_2P_2(\theta=\Theta) \quad (2)$$

or

$$\mathfrak{C}(H) = P_2(\cos\Theta). \quad (3)$$

We expect that $\mathfrak{C}(H)$ approaches unity for large H, and that the sign of H must not be significant. Hence, to lowest order, $\mathfrak{C}(H) \approx 1 - b/H^2$, where b is a constant. Also $P_2(\cos\Theta) = 1 - \frac{3}{2}\sin^2\Theta$, and thus $\sin\Theta \propto 1/H$.

When $\sin\Theta$ is plotted against H^{-1}, an approximately linear behavior (to within $\pm 5°$ in H) in the region $1 \geq H \geq 15$ kOe is observed. When $H = 1$ kOe, $\Theta \approx 25°$ for ^{191}Ir, ^{57}Co, and ^{182}Ta; and $\Theta \approx 20°$ for ^{54}Mn.

Aharoni,[6] in considering only magnetostrictive effects, also predicts $\sin\Theta \propto 1/H$. In a later publication, Aharoni[7] considers also magnetocrystalline, exchange, and magnetostatic forces for W and Nd, and finds a much weaker predicted dependence of $\sin\Theta$ upon $1/H$. The present results indicate a somewhat stronger relationship between Θ and H than is predicted by the latter.

It is noted that the angle Θ is smaller in Mn than in the other samples. (Qualitative measurements of ^{125}Sb, ^{60}Co, and ^{193}Os in Fe show results similar to those for Co, Ta, and Ir in the $H = 1$ kOe region.) Both Co and Mn are similar in size and electronic configuration to Fe. However, transition elements lying to the right of Fe in the periodic table are found to cause a widespread perturbation when introduced into the Fe lattice (out to 10 Å, involving ≈ 200 atoms). Moreover, Co is very magnetostrictive in Fe,[12] so that the observed cone angle for Co is larger than that for Mn. Recent experiments[13] show that Mn and Ta dilutely alloyed into iron have none other than a dilution effect on the over-all magnetic saturation of the iron. However, the large nuclear radius of Ta relative to iron results in a local strain and thus the large observed Θ.

Alignment of nuclei at a nonzero cone angle Θ with respect to $\vec{H}$ is important for interpretations of a number of measurements. For small Θ (large H) the sizes of errors which result from ignoring Θ can be estimated.

Perturbed angular correlations.—As shown by Ben-Zvi *et al.*,[8] in an inhomogeneous field varying from 0.4 to 1.5 kOe,[6] the resulting error in previous determinations of hyperfine couplings is about 20–30% and the cone angle is calculated to be about 30° for ^{186}W in Ni and Fe. Examination of the calculations of Ben-Zvi *et al.* indicates that this underestimation should decrease as $\Theta^2 \propto 1/H^2$.[14] A 20% underestimate of the hyperfine interaction was observed for ^{106}Pd in Fe for $H = 1$ kOe.[15] As another example integral PAC measurements of the magnetic moments of the $I = \frac{5}{2}$ levels in ^{191}Ir and ^{193}Ir with Ir diffused into iron as a source with an applied field $H = 1.2$ kOe have yielded $\mu = (0.42 \pm 0.05)\mu_N$ and $(0.48 \pm 0.08)\mu_N$,[16] respectively; whereas another measurement utilizing Coulomb excitation and recoil into gas, not involving a ferromagnetic host, yielded the higher values $\mu = (0.58 \pm 0.10)\mu_N$ and $(0.73 \pm 0.13)\mu_N$,[17] respectively. For further examples, see Ref. 1.

Nuclear orientation.—For $H = 1$ kOe, measurements ignoring Θ would result in underestimates of B_2 [Eq. (1)] by about 15%. Again, the magnitude of this error decreases like $1/H^2$, since $1 - P_{2,4}(\cos\Theta) \propto \Theta^2$ for small Θ, and $\Theta^2 \propto 1/H^2$. Corresponding errors would be observed in determinations of nuclear moments and in those particular nuclear decay parameters (i.e., U_k and A_k of Ref. 2) which depend directly on the value of B_k.

γ-ray anisotropy thermometry.—The anisotropy of γ rays emitted by ^{54}Mn and ^{60}Co impurities in Fe foils can be used to measure temperature below 50 mK[3,18] because all the parameters of the nuclear decays and hyperfine splittings involved are known. The temperature T has been deduced from the angular distribution $W(\theta)$ of radiation. In this way Sites, Smith, and Steyert[3] observed that a comparison of deduced ^{54}Mn and ^{60}Co temperatures showed a difference of 10% when $H = 1.5$ kOe; e.g., $T = 5.5$ mK for ^{54}Mn versus 6 mK for ^{60}Co. At higher temperatures the error was a smaller percentage. Johnson, Rapp, and Wheatley[4] measured the effect of a magnetic field on the melting curve of ^{3}He in the region $T = 4$–25 mK using Fe(^{54}Mn) thermometry. In

their work (see Fig. 2 of Ref. 4) an apparent discrepancy of 0.9 mK occurred at about 5 mK when the applied field was in the region from 2.1 to 6.4 kOe.

All the errors mentioned above can be understood by utilizing the cone angle Θ to determine overestimates of T. By comparison of the theoretical curves[3] of $W(0)$ versus T and $W(\Theta)$ versus T, taking the Θ characteristic of 2 kOe, the error is found to be about as follows: For ^{54}Mn at 3 and 20 mK, the error is about + 0.6 and + 0.3 mK, respectively; for ^{60}Co, + 1.1 and + 0.8 mK. (The plus sign indicates that the temperature from γ-ray anisotropies is larger than the thermodynamic temperature.) This large percentage error at the lower temperature is associated with the flatness of the curves[3] of $W(0)$ versus $\ln T$ at $T \to 0$. Thus, for thermometry below 10 mK, materials with smaller hyperfine splittings are required in order to avoid working in the flat region of the calibration curve. For example, thermometers such as ^{59}Fe and ^{65}Zn in Fe should have saturation-associated errors less than 2% at applied fields of 2 kOe in the $T = 3$ mK region.

Mössbauer thermometry.—For ^{57}Co in Fe the relative populations of the ^{57}Co magnetic substates are deduced from measurements of the relative intensities of the six 14.4-keV ^{57}Fe Mössbauer resonance lines.[5] Since the cone angle is probably very near to 0° for iron in iron and greater than zero for Co in iron, an error[19] of + 0.5 mK in the $T = 25$ mK region for $H = 1$ kOe can result if Θ is ignored. However, in this case the magnitude of the error should decrease as $1/H^4$.

Taylor[5] found the ^{54}Mn, ^{57}Co, and ^{60}Co anisotropy thermometers reading 5 ± 1.5, 4 ± 1.5, and 3 ± 1.5 mK, respectively, above a Mössbauer thermometer when $H = 1.1$ kOe, with the differences decreasing for larger applied fields, while the present results predict errors of $+\frac{1}{2}$, $+5\frac{1}{2}$, and $+3\frac{1}{2}$ mK, respectively, compared to the Mössbauer thermometer. Except for the large ^{54}Mn difference, Taylor's results are consistent with the present work.

In conclusion, for the materials studied, $\Theta = 20°/H(\text{kOe})$ to within 20%. For Mn, Θ appears to be near the lower extreme of the 20% spread, while values of Θ for other nuclei (Co, Sb, Ta, Ir, Os, W,[8] and Pd[15]) are near the upper extreme of this estimate. Estimates and corrections of previous PAC, γ-anisotropy, and thermometry errors are feasible; future experimenters should utilize externally applied polarizing fields greater than about 5 kOe, in order to reduce the cone angle Θ to a negligible value.

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[19]Taking the z axis as the $\vec{H}$ direction, a sudden change of quantization axis out of the cone into the z direction following the electron capture results in initial states which are mixtures of I_z values in the $I=\frac{5}{2}$ 136-keV level. A perturbation approximation was used to arrive at these numerical results.

Recoil Effects in Single-Nucleon-Transfer Heavy-Ion Reactions

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Using a distorted-wave Born-approximation treatment which exactly includes recoil for the data from the reaction $^{12}C(^{14}N,^{13}C)^{13}N$ at $E(^{14}N)=78$ MeV, we demonstrate the necessity for including recoil effects in calculations for single-nucleon–transfer reactions with heavy ions.

At sufficiently low energies ($Z_1Z_2e^2/\hbar v \gtrsim 10$), heavy-ion reactions appear to behave semiclassically.[1] Angular distributions for single-nucleon–transfer reactions seem to be well described[2,3] using formalisms[4] which neglect "recoil effects." In Fig. 1 the vector diagram is shown which is relevant to the distorted-wave Born-approximation (DWBA) amplitude used in theoretical calculations for such reactions. The vectors $\vec{r}_a$ and $\vec{r}_b$, which must be integrated over, can be expressed as[4]

$$\vec{r}_a=\vec{r}-(x/a)\vec{r}_{bx}, \quad \vec{r}_b=(A/B)\vec{r}+(x/B)\vec{r}_{bx}.$$

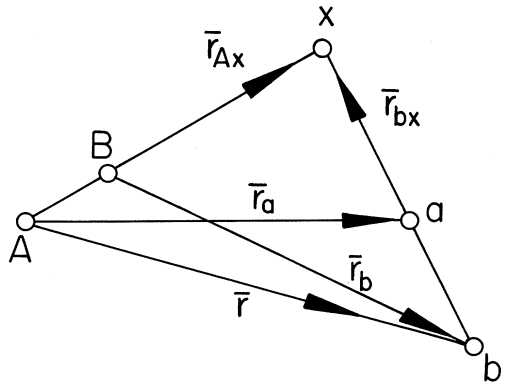

FIG. 1. Vector diagram for the reaction $A(a,b)B$, where $B=A+x$ and $a=b+x$.

Since this involves a six-dimensional integral, the "no-recoil" approximation is introduced which neglects the $\vec{r}_{bx}$ terms, with obvious simplifications. The dangers of this approximation were pointed out long ago.[5]

Recently measurements of heavy-ion single-nucleon–transfer reactions at higher energies[6-8] produced structureless angular distributions in disagreement with recoilless DWBA calculations and semiclassical predictions. The purpose of this paper is to show how the exact inclusion of recoil is necessary to obtain quantitative and qualitative agreement with the data of Ref. 6 for the reaction $^{12}C(^{14}N, ^{13}C)^{13}N$ at 78 MeV.

For a reaction $A(a, b)B$, where B (a) consists of a core A (b) with a particle x bound with angular momentum l_1 (l_2), the selection rules are[8,9]

$$\Delta\vec{j}=\vec{J}_A-\vec{J}_B, \quad \Delta\vec{l}=\vec{l}_1-\vec{l}_2,$$
$$\Delta\vec{s}=\vec{S}_a-\vec{S}_b, \quad \Delta\vec{j}=\Delta\vec{l}+\Delta\vec{s}.$$

If we assume that the directions of J_A and S_b do not change (inert core) in the reaction, then

$$\Delta j=j_1,$$

where $\vec{j}_1=\vec{l}_1+\vec{S}_x$, and

$$\Delta s=j_2,$$

where $\vec{j}_2=\vec{l}_2+\vec{S}_x$. Note that if $l_2=0$, the rules become the familiar values of (d,p) etc. reactions.