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Anomalous temperature dependence of the hyperfine field on osmium in nickel

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Using the ion-implantation perturbed-angular-correlation technique, the temperature dependence of the hyperfine field effective on osmium in nickel has been studied. First-excited states of ^{188}Os and ^{192}Os with known lifetimes and magnetic moments were used as probes of the magnetic interaction. The reduced hyperfine field $H(T)/H(0)$ differs markedly from the nickel reduced bulk magnetization $M(T)/M(0)$. The result confirms a prediction of Campbell correlating anomalous temperature dependence with excess residual resistivity of the alloy. Quantitative agreement with a local-moment model as well as with the Campbell model, however, is poor. Data for tungsten in nickel show no anomalous temperature dependence.

I. INTRODUCTION

The systematic study of the mechanisms which produce hyperfine magnetic fields at dilute transition-metal impurities in ferromagnets and the relative importance of these mechanisms for various impurities are important in understanding the electronic structure of these magnetic systems. The ability to predict and quantitatively describe the variation with temperature of hyperfine fields is a crucial test of any theoretical model.

The usual situation for dilute impurities in ferromagnets is that the temperature dependence of the reduced hyperfine field $h(T) = H_{hf}(T)/H_{hf}(0)$ follows that of the reduced bulk magnetization $\sigma(T) = M(T)/M(0)$, but for a few cases (*Ni*Ru,[1] *Fe*Mn,[2] *Ni*Mn,[3] *Fe*Os[4,5]) $h(T)$ shows a temperature dependence markedly different from $\sigma(T)$. An initial explanation for this anomalous behavior, the model of Jaccarino, Walker, and Wertheim,[6] assumes that the impurity in question has a localized moment μ_{loc} of spin J, which is more weakly coupled by a molecular field (MF) to the host moments than the host moments are to each other. This model does not specifically consider the charge screening of the impurity by the host's conduction electrons, which determines, via the Friedel sum rule, the depth of the impurity's scattering potential. Resulting differences in s-wave phase shifts for the spin-up and spin-down conduction bands leave a net negative spin polarization [conduction-electron polarization (CEP)[7]] at the impurity nucleus and thus, through the contact interaction, a hyperfine field which may be substantial. Later, Low[8] attempted a refinement of the local-moment concept by adding a term proportional to $\sigma(T)$ to take CEP into account, so that the MF-CEP model yields

$$h(T) = fB_J[\xi\sigma(T)/kT] + (1-f)\,\sigma(T)\,, \qquad (1)$$

where B_J is the Brillouin function for spin J, ξ is the impurity-host-exchange coupling parameter, and f is the fraction of $h(T)$ due to local moment.

Several theoretical objections to local-moment models can be raised, however: (i) Several values of J and f can give equally good fits to available $h(T)$ data for each anomalous case; (ii) neutron-diffraction data, where available, do not agree with MF-CEP local-moment values; (iii) no systematics based on the μ_{loc} hypothesis can explain or predict occurrences of anomalous $h(T)$, or why ξ turns out to be small for these cases.

Campbell has proposed[9] that $h(T)$ systematics can be understood using a model based on Friedel's description of transition impurities in ferromagnets, in which one studies changes in the impurity moments as a function of impurity-host charge difference Z. Around a certain critical difference Z_c, the moment (and $d\mu/dZ$) abruptly reverses sign as a bound state of spin-up d electrons crosses the Fermi level and empties. For impurities near Z_c, this bound state can be emptied through thermal smearing of the Fermi level, and $h(T)$ will decrease sharply. The transition at Z_c should be correlated with a peak in the residual resistivity[10] (this is true for the four anomalous cases cited above), and this systematics enables Campbell to predict the occurrence of an anomaly in *Ni*Os and other systems. If the μ_{high} state has $(h/\sigma)_{T=0} = \alpha$, μ_{low} has $(h/\sigma)_{T=0} = \beta$, and the energy separation between states is linear ($E_{high} - E_{low} = E_0 - \lambda T$; this is an *ad hoc* assumption), then

$$h(T) = \sigma(T)\,\frac{1+\gamma\eta\, e^{-E_0/kT}}{1+\eta e^{-E_0/kT}}\,, \quad \gamma = \beta/\alpha\,, \quad \eta = e^{\gamma/k}\,. \qquad (2)$$

E_0 and γ are to be determined from experimental data. The parameter η must be fixed in some way by theoretical estimates because of the large correlations that would result should both γ and η be used as free parameters in a least-squares-fitting routine to available data. Equation (2) yields good fits to the $h(T)$ data for *Ni*Mn, *Fe*Mn, and *Ni*Ru; and qualitative fits to *Fe*Os. The key aspect of the model, however, is the two-state transition concept and the correlation with residual-resistivity results. We have studied the *Ni*Os system, where the spin-up residual resistivity peaks as a function of Z, and we have also taken data for *Ni*W, which appears to be normal.

II. EXPERIMENTAL

We have studied $h(T)$ in the *Ni*Os and *Ni*W systems using the implantation-perturbed-angular-correlation (IMPAC)[11] method. IMPAC is an efficient way to produce low-dilution alloys without unwanted impurities, and is free of sample preparation difficulties present in many related techniques. Assumptions inherent in the interpretation of integral, i.e., time-independent IMPAC results are that (a) the static magnetic interaction is dominant, (b) the effective magnetic field at implant sites becomes constant in magnitude and direction in a time $t \ll \tau_N$ (the lifetime of the nuclear state involved), and (c) this field is the same for all implants and is the same effective field as in a conventionally prepared alloy. Sioshansi *et al.*[12] have recently shown that these assumptions are well satisfied for the *Ni*Os case, at least at room temperature. Their results, especially for ^{192}Os, show excellent agreement with perturbed-angular-correlation (PAC), NMR, and Mössbauer studies. In addition, transient-field[13] precession of the angular correlations during the slowing-down time (~1 psec) of the implant is small and calculable, and no anomalous attenuations at room temperature have been observed, indicating that Os implants probably come to rest at unique sites.

The measurements were performed in the standard geometry for this type of experiment (Fig. 1). Double-layer targets were prepared by plating 300–400-μg/cm^2 coatings of enriched ^{188}Os (87.7%) or ^{192}Os (98.7%) onto 11-mg/cm^2 nickel foil (this Os thickness yields resonable count rate while assuring deep implants). Similar targets of ^{184}W or ^{186}W on nickel were used. The targets were mounted in an external magnetic field of 2.0–2.5 kG, which polarized the nickel perpendicular to a detection plane containing four NaI (Tl) γ-ray detectors. A beam of 36- or 42-MeV $^{16}O^{5+}$ ions from the Rutgers-Bell tandem Van de Graaff accelerator was used to Coulomb excite the Os or W nuclei and implant them into the nickel host. In order to ensure that detected de-excitation γ rays corresponded to proper implanation and alignment of the Os nuclei in their $J^{\pi} = 2^+$, $m = 0$ first-excited states,

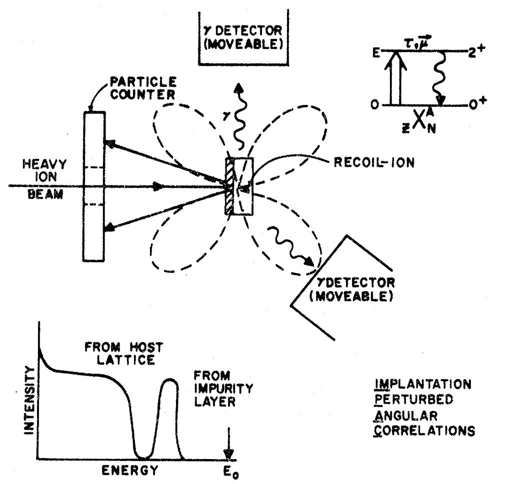

FIG. 1. Schematic implantation-perturbed-angular-correlation setup. The dashed rosette represents the γ-ray angular correlation. An external magnetic field is applied into or out of the plane. The lower inset is a schematic backscattered-particle spectrum.

coincidence was required with oxygen particles backscattered into a 400-mm^2 annular surface barrier detector.

For low-temperature measurements (4.5, 77, 200 K), targets were mounted in a hollow pole piece on an Air Products Corp. Helitrans Cryotip apparatus; a resistive sample heater allowed variation of the target temperature for different runs. Temperature was measured with a chromel-vs-gold thermocouple in good thermal contact with a thick copper target holder. For the beam currents used (a few nA) possible thermal gradients between beam spot and thermocouple are not important due to the high thermal conductivity of nickel.

For $T > 300$ K, a specially designed hot-target pole piece (Fig. 2) which developed a field of > 2.5 kG was employed. The targets were heated in vacuum by radiation from a tantalum-loop filament and temperature was measured by a chromel-alumel thermocouple mounted on a copper target holder whose geometry minimized thermal gradients between beam spot and thermocouple.

The geometry also makes these gradients easily calculable because the temperature is approximately constant throughout the copper (due to its high thermal conductivity) and because of the cylindrical boundary conditions imposed.[14] For each run the beam current was calculated from the target thickness (as determined from energy loss of backscattered particles) and the Rutherford backscatter rate from osmium. The nickel backing is thick enough to stop the beam, so that one knows the average power dissipated within the radius of the beam spot. This power input is confined by beam collimation to a small volume in the center of the target, and, since the thermal conductivity of nickel is low at these temperatures, a dynamic equilibrium is reached in which the temperature in the copper rises (~ 5 K) when the beam is turned on, but not quite as much as the temperature in the beam spot. In every case the gradients involved were calculated to be of the order of 2 K, and this correction was applied to the thermocouple reading. The fact that this reading was usually constant to within ± 1 K despite its sensitivity to variations in beam current is a good indication that such variations were small and that the temperatures assigned were accurate at least to ± 3 K.

To help keep the temperature constant, apart from variations in beam current, the entire target holder was insulated from the pole piece with ceramic standoffs and was surrounded by a thin (~ 500 μm) copper radiation shield with a swath cut out in the detection plane to allow for the escape of backscattered oxygen ions (a similar radiation shield is required at 4.5 K to keep cryotip-mounted samples from heating up). At the same time, damage to the surface barrier detector was avoided by mounting it in thermal contact with the beam line and by shielding its front surface with a 0.06 mm aluminized Mylar sheet.

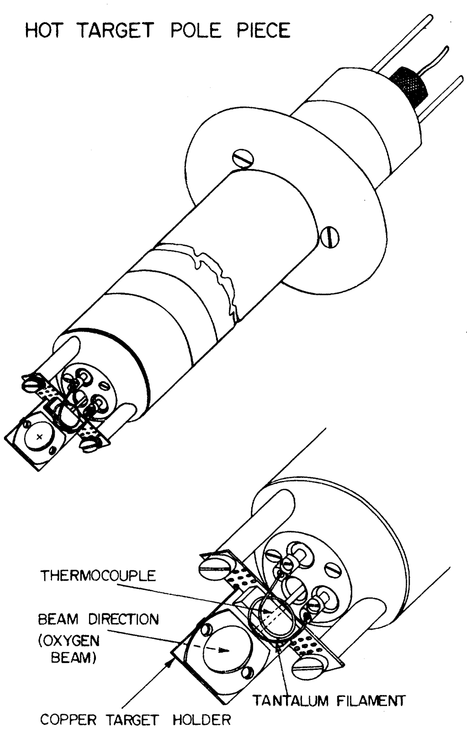

FIG. 2. Sketch of target heating apparatus.

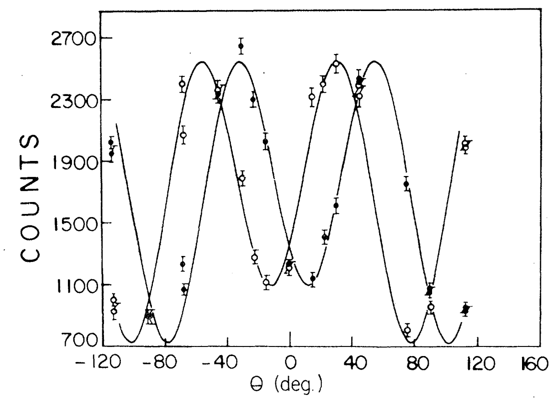

FIG. 3. Angular correlation for ^{192}Os at 300 K. Open circles: external field "up"; solid points: field "down." The solid lines are the best fit using $\omega\tau = 220$ mrad.

III. ANALYSIS AND RESULTS

Angular correlations were measured at 12 or more angles between $-112.5°$ and $+112.5°$, with field "up" and field "down" for all temperatures $T \leq 300$ K, and at selected higher temperatures in a search for possible attenuations due to hyperfine interactions with other than a unique magnetic field. Data at these temperatures were least-square fitted to obtain $\omega_L \tau$, to the usual expression for a time-integral correlation

$$W(\theta) = \sum_{k=0,2,4} b_k \frac{\cos k(\theta - \Delta\theta)}{1 + (k\omega_L \tau)^2} , \tag{3}$$

where $k\Delta\theta = \tan^{-1}(k\omega_L \tau_N)$ and $\omega_L = -g\mu_N H\tau_N / h$. τ_N is the nuclear excited-state mean life (0.414 nsec for ^{192}Os, 1.007 nsec for ^{188}Os, 1.53 nsec for ^{186}W, and 1.92 nsec for ^{184}W).

In all cases the experimental b_k's for ^{192}Os from Eq. (3) were found to be in agreement with those calculated for a $0 \xrightarrow{^{16}O} 2 \xrightarrow{\gamma} 0$ correlation; the only attenuations were due to particle- and γ-counter solid-angle effects. This indicates that, indeed, *Ni*Os is a good system for IMPAC at all temperatures studied.

At other temperatures data were taken only at slope angles of the correlation, and angular shifts were extracted by an iterative procedure which solves Eq. (3) for $\langle \omega_L \tau_N \rangle$ when given b_k and data at a series of angles. This procedure is most useful for the smaller (< 150 mrad) angular shifts which occur at higher temperatures. Data for ^{188}Os were only taken at shift angles, and the analysis was further complicated by 6.7% ^{189}Os and 3.2% ^{190}Os impurities present. A γ coincidence spectrum using a Ge(Li) detector revealed lines from ^{189}Os and ^{190}Os which could not be resolved from the dominant ^{188}Os line using NaI(*Tl*) crystals. The

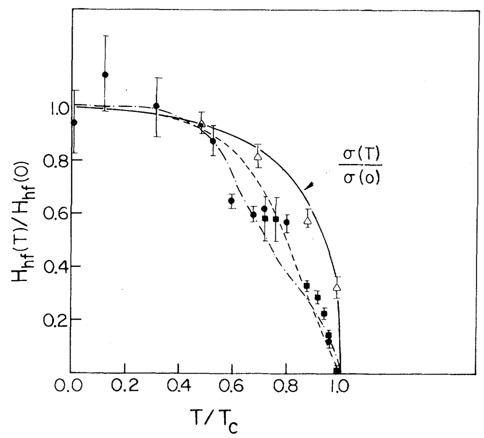

FIG. 4. Temperature dependence of the hyperfine field on osmium in nickel. The circles represent ^{192}Os results, the squares ^{188}Os. Four points (triangles) for tungstein in nickel are also shown. Solid line: nickel magnetization curve; dashed line: best fit to Eq. (2); dot-dash line: best fit to Eq. (1).

^{189}Os γ's were relatively isotropic and were treated as a constant background. The ^{190}Os line had to be corrected for its angular correlation and shift before subtraction. The agreement of the ^{188}Os result with ^{192}Os at 450 and 600 K (see Fig. 4) is independent evidence that the procedure adopted is a correct one.

Each $\omega_L\tau$ was also corrected for transient-field precession [which varies as $\sigma(T)$ and is about 7 mrad at low temperatures] and for beam bending, which was about 1 mrad. After these corrections one is left with $\omega\tau_{\text{static}}$ and $\omega\tau_{\text{static}}(T)/\omega\tau_{\text{static}}(0) = h(T)$. Almost all of the data taken below 500 K used ^{192}Os, while most of the higher-temperature data used ^{188}Os, since its longer lifetime yields larger angular shifts and makes it a more sensitive probe as $\omega\tau(T)$ diminishes near the Curie point of nickel (~ 631 K).

Our room-temperature result for ^{192}Os of $\omega\tau = 220 \pm 15$ mrad (Fig. 3) is in excellent agreement with the results of the Purdue group,[12] and our observation that $h(T)$ for *Ni*Os (Fig. 4) follows the Curie curve $\sigma(T)$ from 4.5 to 301 K agrees with the PAC experiments of Johansson *et al.*[4] Above room temperature, however, $h(T)$ drops sharply below $\sigma(T)$, flattens out between 400 and 550 K, then finally drops sharply again to zero as $T \to T_{\text{Curie}}$. Data were also taken at relevant temperatures for the *Ni*W system, using ^{186}W and ^{184}W. Campbell predicted that $h(T)$ would follow the Curie curve for this system since $Z = Z_c + 2$, and our experimental data do indeed confirm this.

IV. DISCUSSION

The results described indicate an anomalous temperature-dependent field for *Ni*Os and not for *Ni*W.

Quantitatively, however, Eq. (2) does *not* yield a good fit to our data (see Fig. 4). Letting γ and η vary independently does not significantly change the fit achieved by setting $\eta \sim 10$. In addition, the *Ni*Os data cannot be fit to a local-moment model regardless of spin. Both Eqs. (1) and (2) achieve a best $\chi^2 \cong 8$ because they cannot reproduce the two sharp changes in slope exhibited by the data. Campbell's function (2) contains a very explicit and *arbitrary* assumption about the linear dependence of the supposed separation between high- and low-moment states. This assumption was made to simplify the mathematics, but perhaps a much sharper transition or a completely different model is needed for the *Ni*Os case. In any event, Campbell's prediction of "anomalous" $h(t)$ for Os and "normal" behavior for W in Ni has been verified. Other systems for which Campbell predicts anomalies (*Ni*Cr, *Co*Mn, *Co*Ru, *Co*Os, and either *Fe*Ru or *Fe*Tc) remain to be studied.

In addition to the hyperfine-field temperature-dependence results, the unperturbed-angular-correlation data corroborate the implantation assumptions enumerated above. However, as there are no other data to compare ours with above room temperature, we cannot unambiguously rule out a reduced hyperfine field due to the ion-implantation process itself. As any damage should anneal more rapidly at the higher temperatures, we regard this possibility as remote.

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