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Neutron reflection as a probe of surface magnetism

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Polarized neutrons reflected from the surface of a ferromagnet are sensitive to the magnetization close to the surface as well as to the bulk magnetization. In a calculation using parameters appropriate to nickel it is shown how the two contributions can be separated. Neutron measurements can determine the first moment of the magnetic disturbances at the surface, with a sensitivity corresponding to one magnetic dead layer.

The problem of the magnetization at the surface of a ferromagnet has received a considerable amount of attention in recent years.¹ The consensus is now that at zero temperature only the magnetization of the topmost layers is affected by the presence of the surface, even in a metal; the detailed nature of the perturbation depending on the specific system. The effect of the temperature is to increase the thickness of the region in which the surface affects the magnetization. If at the surface layer the magnetization differs from the bulk by an amount $\Delta\mu_s$, the penetration of the disturbance at a distance z from the surface is^{2,3}

$$\Delta\mu(z) = \Delta\mu_s \exp(-z/\xi) \quad (1)$$

where ξ is a magnetic coherence length, and both $\Delta\mu_s$ and ξ are temperature dependent:

$$\xi(T) = \xi(0) \left(\frac{T}{T_C - T} \right)^\nu \quad (2)$$

$$\mu_s(T) = \mu_s(0) \left(1 - \frac{T}{T_C} \right) \quad (3)$$

Here $\xi(0)$ is of the order of one lattice spacing, and the exponent ν is less or equal to one. The linear temperature dependence of the surface magnetization μ_s up to the Curie temperature T_C is quite different from that of the magnetization of the bulk μ_∞ .

The form of the expressions (1)–(3) is still controversial. Their experimental verification is difficult, since only a few probes are at the same time surface and spin sensitive. Pioneering experimental work has been done using Hall effect,⁴ photoemission,³ polarized electron diffraction,⁵ and electron-capture spec-

troscopy.⁶ While these techniques are extremely sensitive to the magnetic state of the surface, the analysis of the data in terms of the layer-by-layer magnetization is rather difficult and subject to fairly drastic approximations. The purpose of the present Communication is to show how neutron scattering can be used, with all the advantages deriving from the availability of an accurate scattering theory.

It might seem paradoxical to propose neutron scattering as a probe to analyze phenomena at the surface, since the neutron scattering amplitude per atom is of the order of 10^{-12} cm, or of the order of a nuclear radius; thus neutrons can penetrate deeply into matter. However, their sensitivity to surface effects is enhanced by employing a beam of long-wavelength neutrons, sent at grazing incidence to the surface. Under these conditions the material interacts with the neutron wave as a continuous solid of refractive index n , which for a ferromagnet is dependent on the neutron spin⁷:

$$n^\pm = n_N \pm n_M = 1 - \frac{\lambda_0^2}{2\pi} \frac{1}{V} [b \pm C\mu(z)] \quad (4)$$

where λ_0 is the neutron wavelength, V the atomic volume, b the scattering amplitude per nucleus, and μ the magnetic moment per atom, converted to a length by a constant $C = 0.2695 \times 10^{-12}$ cm/ μ_B . The moment μ is assumed to be the only quantity dependent on the distance z from the surface. The signs are relative to the initial neutron polarization, parallel (+) or antiparallel (–) to the magnetization.

The optics of a plane wave in a medium with refractive index $n(z)$ has been thoroughly discussed in classical treatises.⁸ The reflectivity, or the normalized

intensity of the specular reflection, cannot be given in closed and exact form for arbitrary $n(z)$ but only for a restricted number of functional dependences. Of these the most well known is that of a semi-infinite slab of material, with constant refractive index n :

$$R = \left| \frac{\sin\theta_0 - (n^2 - \cos^2\theta_0)^{1/2}}{\sin\theta_0 + (n^2 - \cos^2\theta_0)^{1/2}} \right|^2, \quad (5)$$

where θ_0 is the glancing angle of the incident beam with the surface. For θ_0 lower than a critical angle θ_c the reflectivity is unitary (if the material is nonabsorbing). Above the critical angle, the reflectivity drops sharply, but it is still finite and practically detectable over a limited range ($R = 5 \times 10^{-3}$ at $\theta_0 = 2\theta_c$). The range $\theta_c < \theta_0 < 2\theta_c$ is of considerable interest in determining the magnetization of the material with polarized neutrons.

The intensity of the specular reflection is R^+ for neutron polarized parallel, R^- for neutrons polarized antiparallel to the magnetization. The ratio R^+/R^- , known as the flipping ratio R_F , is a very useful quantity, since it is largely independent from many experimental variables, and its precision approaches that of the statistics of the neutron counts. The behavior of the flipping ratio of the reflected beam for constant magnetization (CM) is illustrated in Fig. 1, with parameters close to those appropriate to nickel—perhaps the most studied among the elemental ferromagnets.⁹ The parameters used were a neutron wavelength of 5 Å (cold neutrons), a nuclear scatter-

ing amplitude $b = 1.03 \times 10^{-12}$ cm, and a magnetic moment of $0.6\mu_B$ per atom, contained in an atomic volume $V = 10.9 \text{ \AA}^3$. In the material there are two critical angles, $(\theta_c^\pm)^2 \approx (\lambda_0^2/\pi V)(b \pm C\mu)$, for the two polarization states of the neutrons. Below θ_c^- the reflectivity is unitary for both neutron states; hence the flipping ratio is unitary. Between θ_c^- and θ_c^+ the flipping ratio increases dramatically, since only the positively polarized neutrons are totally reflected. Then, for $\theta_0 > \theta_c^+$ the R_F decreases toward an asymptotic value, as may be better visualized by expanding the exact form of the flipping ratio in a power series of $C\mu/b$ for $\theta_0 > \theta_c$ and retaining the linear term:

$$(\sqrt{R_F})_{\text{CM}} \approx 1 + 2 \frac{C\mu}{b} \frac{\theta_0}{(\theta_0^2 - \theta_c^2)^{1/2}}, \quad (6)$$

where θ_c is the nonmagnetic critical angle

$$[\theta_c^2 = (\lambda_0^2/\pi) b/V].$$

A magnetic perturbation at the surface is most simply introduced in a step-function model by taking a constant decrement of the magnetic moment per atom $-\Delta\mu$ over a thickness z_0 of material. In this case the reflectivity is still given by an exact but lengthy closed expression.⁸ A simpler form is obtained by expanding the flipping ratio in powers of $\Delta\mu/b$, and retaining the linear term:

$$\sqrt{R_F} = (\sqrt{R_F})_{\text{CM}} \left[1 - \sin^2 \left(\frac{2\pi}{\lambda_0} z_0 p \right) \frac{4\sin\theta_0}{p} \frac{C\Delta\mu}{b} \right], \quad (7)$$

where $p \approx (\theta_0^2 - \theta_c^2)^{1/2}$ and the subscript CM refers to the magnetization in the bulk. The oscillatory term in Eq. (7) describes the interference pattern of a slab of scattering material of thickness z_0 . In all practical cases the argument is small; hence the expression for the flipping ratio can be further approximated by

$$\sqrt{R_F} = (\sqrt{R_F})_{\text{CM}} \left[1 - \left(\frac{4\pi}{\lambda_0} \right)^2 [\theta_0(\theta_0^2 - \theta_c^2)^{1/2}] \frac{2C}{b} M_1 \right]. \quad (8)$$

The magnetic disturbance at the surface has the effect of lowering the flipping ratio progressively with increasing θ_0 , so that R_F no longer tends to a constant value. The effect is proportional to the first moment of the magnetic disturbance at the surface $M_1 = \int z \Delta\mu(z) dz$, which in this simple model takes the form $M_1 = \frac{1}{2} z_0^2 \Delta\mu$. Thus, in principle, neutron reflection measurements can determine the first moment of the magnetic disturbance at the surface. However, several practical questions arise. First, what kind of precision is required in the experiment? Second, how safe is it to analyze an experimental R_F in terms of an ideal functional dependence? Third, to what extent is the first-moment approximation

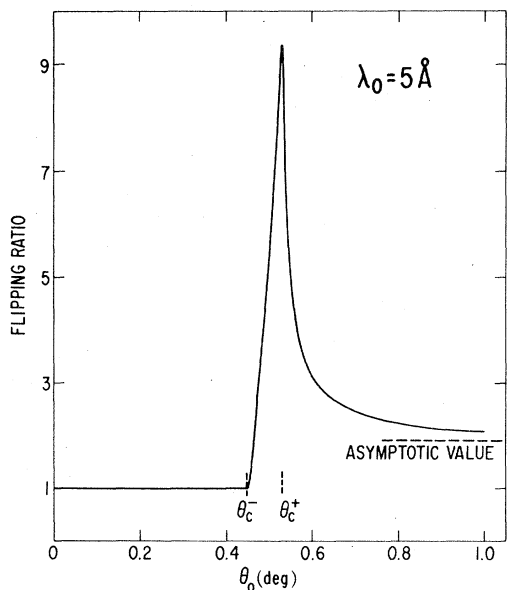


FIG. 1. Flipping ratio of the specular reflection of ferromagnetic nickel with a uniform magnetic moment of $0.6\mu_B$ /atom.

valid for a realistic spatial distribution of the magnetic disturbance? A promising answer to these questions is obtained with numerical calculations appropriate to nickel.

A reasonable description of the magnetic disturbance at the surface of nickel is provided by Eqs. (1)–(3) with the following parameters: in Eq. (2) $\nu = 1$ and $\xi(0) = 2.03 \text{ \AA}$ (the interlayer spacing of close-packed planes); in Eq. (3) $\mu_s(0) = \mu_\infty(0)$, while the temperature variation of the moments in the bulk was taken as a Brillouin function for $S = \frac{1}{2}$. Actually the temperature can be left as a hidden variable, and the moment disturbance defined as a function of a new parameter $z_1 = \sqrt{2M_1/\mu_\infty}$, which represents an effective dead-layer thickness. The refractive index $n(z)$ can be now readily defined for each atomic layer, and the calculation of the reflectivity⁸ carried out numerically.

An example of the calculated flipping ratio is presented in Fig. 2 for a temperature $T/T_C = 0.87$ or for an effective dead-layer thickness $z_1 = 5.2$ atomic layers; a value $\mu_\infty = 0.37\mu_B$ (where μ_B is the Bohr magneton) was assigned to the bulk magnetization. The decrease of the flipping ratio at increasing θ_0 is sharper than that obtained when the magnetization is constant up to the surface, and cuts across a similar curve obtained for $\mu_\infty = 0.33\mu_B$ at $\theta_0 \sim 0.9^\circ$, as is also seen in Fig. 2.

The experimental flipping ratio, and its functional dependence on θ_0 , can be measured rather accurately, since it is not too difficult to achieve a statistical accuracy per data point of the order of 10^{-3} . Experi-

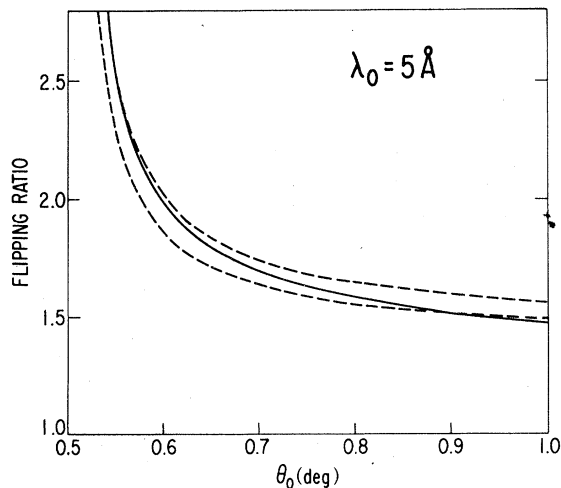


FIG. 2. Full line: flipping ratio of the specular reflection of nickel with a bulk moment of $0.37\mu_B/\text{atom}$ and a surface disturbance (in Bohr magnetons) $\Delta\mu = 0.29 \exp(-\frac{1}{6}z)$. Dashed lines: R_F for constant magnetization of $0.37\mu_B$ (upper curve) and $0.33\mu_B$ (lower curve).

mental error bars thus introduced indicate that the limit of detection of a magnetic disturbance occurs when only the first surface layer is magnetically dead. This observation brings with it some important consequences. It is quite unlikely to observe with neutrons disturbances in the surface magnetization at zero temperature. Conversely, the angular variation of the experimental flipping ratio at low temperature can be tested against the ideal functional form. Only then it is possible to analyze reliably the shape of the flipping ratio at finite temperature, in order to extract from it the first moment of the magnetic disturbance at the surface, and the bulk magnetization. The latter quantity can of course be compared with the result of conventional magnetic measurements.

Finally, it is worthwhile to test the validity of the first-moment approximation. For this purpose, the calculated flipping ratio was analyzed with the aid of Eq. (8), in order to extract an "observed" effective layer thickness $z_{1,\text{out}}$. The results, averaged for each temperature over a region $\theta_c < \theta_0 < 2\theta_c$, are presented in Fig. 3. The approximation is rather satisfactory except when the penetration length becomes very large, or close to the Curie point. There the flipping ratio may be capable of describing the magnetic disturbance in more detail; and indeed the description of the angular dependence of the flipping ratio in terms of Eq. (8) becomes poorer close to T_C . The importance of the approximations made is visualized by reporting (in Fig. 3) $z_{1,\text{out}}$ as obtained for a "square"

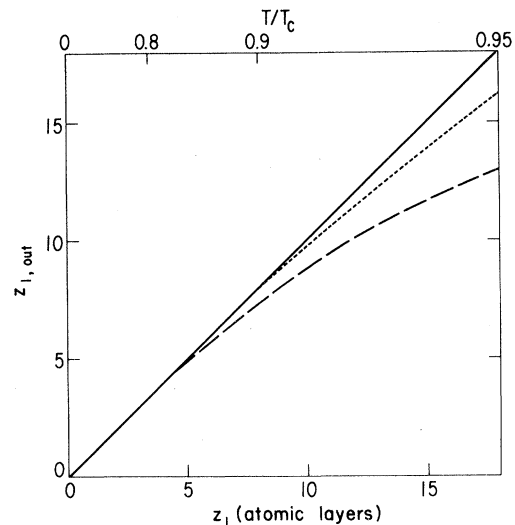


FIG. 3. Effective magnetic dead-layer thickness $z_{1,\text{out}}$, calculated from the neutron reflection in the linear approximation. Upper abscissa: normalized temperature; lower abscissa: effective magnetic dead-layer thickness at that temperature for nickel. Full line: an exact response. Dashed line: an exponentially decaying magnetic disturbance. Dotted line: a square magnetic disturbance.

magnetic disturbance where the first z_1 layers are indeed magnetically dead.

In conclusion, a simple formulation of the neutron reflectivity, and numerical calculations that can be easily extended to systems other than nickel, show that careful neutron measurements could determine the temperature dependence of the magnetization at the surface of ferromagnets. It is to be hoped that this old but viable technique will flank the newly developed surface probes in solving the important problems of the surface magnetism, such as the form

and the value of the critical exponents¹⁰ at the surface of Heisenberg and Ising ferromagnets.

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