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Magnetic Scattering of Electrons from Crystals and Polarization of the Scattered Beam

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The role played by the exchange interaction in the scattering of electrons from magnetic crystals has not been fully appreciated until recently. In this paper, we show that the interference between the Coulomb and the exchange scattering potentials can lead to large polarizations of a beam of elastically scattered low-energy electrons. A double-scattering experiment that can measure this effect is described.

I. INTRODUCTION

IN a recent letter[1] we examined the *inelastic* scattering of electrons from ferromagnets. We showed that for small scattering angles the dominant scattering mechanism is the electron-magnon interaction via the exchange term of the Hamiltonian. Because of the excellent energy resolution now available for electron-scattering experiments,[2] this exchange scattering was shown to provide a convenient means for probing the magnon spectra and for producing highly polarized electron beams. In the present paper, we discuss the *elastic* scattering of electrons and we show that the interference of the Coulomb and the exchange interactions is the dominant polarization mechanism for the elastic scattering of low-energy electrons at the smaller Bragg angles. This result is of particular interest since Mott[3] and others[4] have discussed in considerable detail the polarization due to the spin-orbit and dipole interactions, which are much weaker for the low-energy electron scattering at the angles we are considering, but few people have considered the possible importance of the exchange interaction in scattering phenomena.[5]

The model we use will be one where the incoming electrons are represented by plane waves and the scattering is described by a first-order Born approximation. Unlike the spin-orbit interaction, the exchange interaction contributes to the polarization of the electron beam in the first-order solution of the scattered wave. We show that at low energies very high polarizations can be achieved via this mechanism and an experiment is proposed for measuring these polarization effects.

[1] R. E. De Wames and L. A. Vredevoe, Phys. Rev. Letters **18**, 853 (1967).

[2] H. Boersch, J. Geiger, and W. Stickel, Phys. Rev. Letters **17**, 379 (1966).

[3] N. F. Mott, Proc. Roy. Soc. (London) **A124**, 425 (1929); **A135**, 429 (1932); N. F. Mott and H. W. W. Massey, *Theory of Atomic Collisions* (Oxford University Press, New York, 1949), 2nd ed.

[4] See, for example, P. S. Farago, in *Advances in Electronics and Electron Physics*, edited by L. Marton (Academic Press Inc., New York, 1965), Vol. 21, p. 1, where many other references may be found.

[5] D. L. Mills, J. Phys. Chem. Solids **28**, 2245 (1967); see also Ref. 4, p. 35.

II. INTERACTION HAMILTONIAN AND SCATTERING AMPLITUDE

The interaction Hamiltonian for an electron scattering from a magnetic crystal is

$$H_{\text{int}}=\sum_i V(\mathbf{r}-\mathbf{R}_i)+\sum_i G(\mathbf{r}-\mathbf{r}_i)\mathbf{S}_e\cdot\mathbf{S}_i$$
$$+\mu_e\cdot\{\nabla_r\times\sum_i[\boldsymbol{\mu}_i\times(\mathbf{r}-\mathbf{r}_i)/|\mathbf{r}-\mathbf{r}_i|^3]\}$$
$$+\sum_i[\boldsymbol{\mu}_i\times\nabla_{r_i}\phi(\mathbf{r}-\mathbf{r}_i)/2emc]\cdot\mathbf{P}_e$$
$$+\sum_i[\boldsymbol{\mu}_e\times\nabla_{r_i}V(\mathbf{r}-\mathbf{R}_i)/2emc]\cdot\mathbf{P}_e.\quad(1)$$

The first term is the Coulomb interaction between the atom at $\mathbf{R}_i$ and the incident electron, whose position we represent by $\mathbf{r}$. The second term is the exchange interaction between the magnetic electron at $\mathbf{r}_i$, having spin $\mathbf{S}_i$, and the incident electron whose spin we denote by $\mathbf{S}_e$. $G(\mathbf{r}-\mathbf{r}_i)$ is the exchange potential. The third term is the dipole interaction that leads to the contact and dipole-dipole terms. $\boldsymbol{\mu}_e$ and $\boldsymbol{\mu}_i$ are the magnetic moments of the scattering and magnetic electrons. The last two terms are the "spin-orbit" interactions of the host and scattered electron, respectively, where $\phi(\mathbf{r}-\mathbf{r}_i)$ is the Coulomb interaction between the magnetic electrons and the incident electron, m is the electronic mass, and $\mathbf{P}_e$ is the scattering electron momentum. The exchange, dipole, and host spin-orbit interactions are all magnetic terms, requiring the host atoms to have a net spin moment for their existence.

We note that, for the case of inelastic scattering, all five terms of this Hamiltonian can couple with phonons through their dependence on the atomic and host electron coordinates. The exchange, dipole, and host spin-orbit terms can also couple with magnons through their dependence on the magnetic electron spin coordinates. For moderate incoming electron energies $E_e\ll mc^2$ the dipole and spin-orbit interactions are negligible compared to the Coulomb and exchange interactions, and for this case one finds that the inelastic scattering from phonons is primarily due to the Coulomb interaction,[6]

[6] R. E. De Wames and W. F. Hall, Phys. Letters **23**, 649 (1966).

while the inelastic scattering from magnons is primarily due to the exchange interaction.[1]

All five terms of the Hamiltonian (1) contribute to the elastic (Bragg) scattering of the incident electrons. The elastic scattering amplitude for this interaction Hamiltonian in the Born approximation is

$$f(\mathbf{K},\psi)=(m/2\pi\hbar^2)\exp[-W(\mathbf{K})]\sum_i \exp(-i\mathbf{K}\cdot\mathbf{R}_i)$$
$$\times\{[4\pi Z_N e^2/(\Gamma^2+K^2)]+G(K)S_m^zS_e^z$$
$$+4\pi\beta^2F(K)S_m^{\perp}S_e^{\perp}+2\pi i\beta^2F(K)\sin\psi S_m^n(k_0/K)^2$$
$$+2\pi i\beta^2 Z_N D(K)\sin\psi S_e^n(k_0/K)^2\}. \quad (2)$$

k_0 is the wave number of the incident electron that has spin S_e, mass m, and charge e, and $\mathbf{K}$ is the transferred wave vector in the scattering, where the scattering angle is ψ. $\mathrm{Exp}[-W(\mathbf{K})]$ is the Debye-Waller factor, $\mathbf{R}_i$ is the lattice vector of the ith atom of the host, Γ^{-1} is the screening length of the host atoms that have a net spin moment S_m and nuclear charge Z_N, and β is the Bohr magneton $e\hbar/mc$. The superscripts z, $\perp$, and n on the spins denote components parallel to the magnetic axis (taken to be the Z axis), perpendicular to $\mathbf{K}$, and pependicular to the plane of scattering, respectively. $G(\mathbf{K})$ is the exchange potential, which has the integral form

$$G(\mathbf{K})=\frac{1024}{5\pi}\alpha^9e^2$$
$$\times\int\frac{\partial\mathbf{k}}{k^2}\left[\frac{2\alpha^2}{(|\mathbf{k}_0-\mathbf{k}|^2+\alpha^2)^4}-\frac{1}{(|\mathbf{k}_0-\mathbf{k}|^2+\alpha^2)^3}\right]$$
$$\times\left[\frac{2\alpha^2}{(|\mathbf{k}_0-\mathbf{k}+\mathbf{K}|^2+\alpha^2)^4}-\frac{1}{(|\mathbf{k}_0-\mathbf{k}+\mathbf{K}|^2+\alpha^2)^3}\right], \quad (3)$$

where α^{-1} is the mean orbital radius of the electrons contributing to the net spin moment S_m of the host atoms. A closed form solution for $G(\mathbf{K})$ can be obtained for three limiting cases:

$$G(\mathbf{K})=(\pi e^2/k_0^2)[4-(11.3-5.9\cos^2\theta)(K^2/\alpha^2)],\quad \text{for}\quad k_0^2\gg\alpha^2\gg K^2, \quad (4a)$$

where $\cos\theta=\mathbf{k}_0\cdot\mathbf{K}/k_0K$;

$$G(\mathbf{K})=(256\pi e^2/5k_0^2)(\alpha/K)^6,\quad \text{for}\quad k_0^2, K^2\gg\alpha^2 \quad (4b)$$

$$G(\mathbf{K})=(372\pi e^2/5\alpha^2),\quad \text{for}\quad k_0^2, K^2\ll\alpha^2. \quad (4c)$$

To describe the larger Bragg angle scattering processes, where the change in electron wave vector corresponds to several reciprocal lattice vectors, the solution (4b) is required. But, for small Bragg angles, where the change in wave vector corresponds to a reciprocal lattice vector $K^2\ll\alpha^2$, the solution (4a) (for high-electron energies) or (4b) (for low-electron energies) will be required. $F(K)$ is the magnetic form factor, which arises because the magnetic electrons are displaced from the nucleus. For the common case where the magnetic electrons are in d states,

$$F_d(K)=\frac{256}{3}\left(\frac{\alpha}{K}\right)^7\frac{48(\alpha/K)^5-40(\alpha/K)^3+3(\alpha/K)}{[4(\alpha/K)^2+1]^6}$$
$$=256(\alpha/K)^8,\quad\text{for}\quad K^2\gg\alpha^2$$
$$=1,\quad\text{for}\quad K^2\ll\alpha^2. \quad (5)$$

$D(K)$ is a factor containing the effects of the screening of the atomic potential in the spin-orbit interaction of the incident electron and has the form

$$D(K)=1+(\Gamma^2/\Gamma^2+K^2)-2(\Gamma/K)\arctan(K/\Gamma)$$
$$=1,\quad\text{for}\quad K^2\gg\Gamma^2$$
$$=-K^2/\Gamma^2,\quad\text{for}\quad K^2\ll\Gamma^2. \quad (6)$$

Comparing the magnitudes of the five scattering amplitudes in (2), for $K^2\gg\Gamma^2$, α^2 we find that

$$\frac{f_{\text{exch}}}{f_{\text{Coul}}}=\frac{64}{5}\left(\frac{S_m^z}{Z_N}\right)\left(\frac{K}{k_0}\right)^2\left(\frac{\alpha}{K}\right)^6S_e^z, \quad (7)$$

$$\frac{f_{\text{dip}}}{f_{\text{Coul}}}=512\left(\frac{S_m^{\perp}}{Z_N}\right)\left(\frac{\alpha}{K}\right)^8\frac{(\hbar^2K^2/2m)}{mc^2}S_e^{\perp}, \quad (8)$$

$$\frac{f_{\text{so (host)}}}{f_{\text{Coul}}}=i256\left(\frac{S_m^n}{Z_N}\right)\left(\frac{\alpha}{K}\right)^8\left(\frac{E_e}{mc^2}\right)\sin\psi, \quad (9)$$

$$\frac{f_{\text{so (elec)}}}{f_{\text{Coul}}}=i\left(\frac{E_e}{mc^2}\right)\sin\psi S_e^n. \quad (10)$$

We see from these ratios that for $E_e\ll mc^2$ the Coulomb scattering amplitude is much larger than the other four, and the amplitudes for the exchange and electron spin-orbit interactions are much larger than those for the dipole and host spin-orbit interactions. Therefore, in this paper we need concern ourselves only with the Coulomb, exchange, and electron spin-orbit interactions. Since $K=2k_0\sin\frac{1}{2}\psi$, all the scattering amplitudes in (2) decrease with increasing Bragg scattering angle. However, the ratio (7) *increases* as the scattering angle ψ becomes smaller. This has a very important consequence when considering interference effects between the Coulomb and the exchange interactions—that is, their interference is peaked in the region of maximum scattering amplitude. The importance of this point will be discussed in Sec. III.

III. POLARIZATION EFFECTS

It is well known[7] that any terms in the absolute square of the scattering amplitude that display a linear

[7] T. Y. Wu and T. Ohmura, *Quantum Theory of Scattering* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1962), Sec. J.

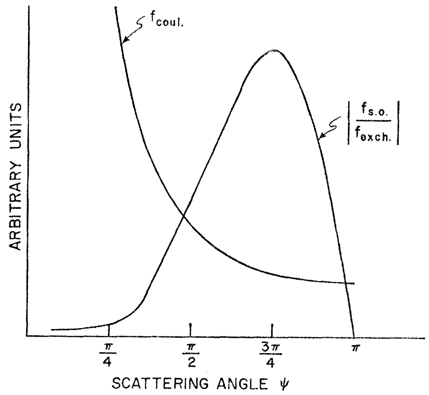

FIG. 1. Ratio of the electronic spin-orbit and the exchange scattering amplitudes as a function of the scattering angle. The angular dependence of the Coulomb scattering amplitude is also plotted.

dependence on the scattering electron spin will contribute to the polarization of the scattered beam. For example, if we consider a scattering amplitude of the form

$$f=a+\sum_\alpha b_\alpha\sigma_\alpha,$$

where σ_α is the Pauli matrix for the α spin component, then the polarization is given by

$$\langle\sigma_\alpha\rangle=2[R_e(a^*b_\alpha)/(|a|^2+\sum_\alpha|b_\alpha|^2)]. \qquad (11)$$

There is a linearly spin-dependent interference between the Coulomb and the exchange interactions in the absolute square of the scattering amplitude (2). But there is no interference between the Coulomb and the spin-orbit interactions since, in the first-order Born approximation, the spin-orbit amplitude is imaginary while the Coulomb amplitude is real. Therefore, the spin-orbit interaction will not contribute to the polarization (11) in first order.

In order to obtain contributions to the polarization of the scattered beam from the spin-orbit interaction, it is necessary to include the distortion of the incident wave in the solution of the scattering amplitudes, as Mott[3] did using a partial-wave analysis. For small scattering angles this correction will be small in magnitude compared to that of the first-order amplitude. Therefore, when considering small scattering angles, if the exchange amplitude dominates the magnitude of the first-order spin-orbit amplitude we can neglect the Mott contribution to the polarization. To show that this is the case we return to the total scattering amplitude (2) and find the ratio of the electronic spin-orbit and the exchange scattering amplitudes in the region k_0^2, $K^2\gg\alpha^2$.

$$\left|\frac{f_{\mathrm{so}}}{f_{\mathrm{exch}}}\right|=\frac{5}{2}\left(\frac{F_e}{mc^2}\right)\left(\frac{Z_N}{S_m^z}\right)\left(\frac{k_0}{\alpha}\right)^6\left(\frac{S_e^n}{S_e^z}\right)\sin^5\tfrac{1}{2}\psi\,\cos\tfrac{1}{2}\psi. \qquad (12)$$

The angular dependence of this ratio is displayed in Fig. 1 along with that of the Coulomb amplitude. We see from the figure that this ratio is sharply peaked at large scattering angles (having a maximum at $\psi\sim 3\pi/4$) where the Coulomb amplitude is small, but is very small for small scattering angles where the Coulomb amplitude is very large. For the case of an unpolarized electron beam scattering from iron, we find that the exchange amplitude still dominates the spin-orbit amplitude for scattering angles $\psi<\frac{1}{4}\pi$ when $E_e\lesssim 1$ keV. Only when the electron energy becomes very large will the spin-orbit scattering amplitude become competitive with the exchange amplitude at the small scattering angles where both the scattered currents *and* the effects of the Coulomb-exchange interference on the scattering are largest.[8]

Combining Eqs. (2) and (11) we calculate the polarization arising from the Coulomb-exchange interference to be

$$\begin{aligned}\langle\sigma_e^Z\rangle&=\frac{16\pi Z_N e^2 G(\mathbf{K})\,S_m^z(K^2+\Gamma^2)}{64\pi^2 Z_N^2 e^4+[G(\mathbf{K})\,S_m^z]^2(K^2+\Gamma^2)^2}\\&=\frac{4}{5}\frac{S_m^z}{Z_N}\left(\frac{\alpha}{k_0}\right)^6\sin^{-4}\tfrac{1}{2}\psi,\quad\text{for}\quad k_0^2,\,K^2\gg\alpha^2,\,\Gamma^2\\&=\frac{1860 Z_N S_m^z\alpha^2\Gamma^2}{100 Z_N^2\alpha^4+(93 S_m^z)^2\Gamma^4},\quad\text{for}\quad k_0^2,\,K^2\ll\alpha^2,\,\Gamma^2.\end{aligned} \qquad (13)$$

We see that the polarization increases as the energy and/or scattering angle decreases, and for the optimum case, where k_0, $K\ll\alpha$, Γ attains the maximum value given by the lower expression on the right-hand side of (13). For the case of iron we find this maximum value $\langle S_e^z\rangle_{\max}=0.86$ or a polarization of 86%. This maximum result requires electron energies $E_e\lesssim 50$ eV for the case of iron. As the energy and scattering angle increases, the polarization rapidly decreases. 350 eV electrons scattering from iron with a scattering angle of 60° ($k_0=K=10^9$ cm^{-1}), according to the upper expression on the right-hand side of (13), will be polarized by only 1.2%.

Because of the selective coupling of the exchange interaction to only those incoming electron spin components that are parallel to the magnetic axis, a beam of polarized electrons incident on a ferromagnet (or ferrimagnet) will undergo elastic Bragg reflection with

[8] One must be careful not to extend these arguments to larger scattering angles, where the distortion of the incident wave is too large to be treated by a Born-type expansion and a partial-wave solution is required. Recent experiments [W. Hilgner and J. Kessler, Phys. Rev. Letters **18**, 983 (1967)] suggest that the approximation of these effects as small second-order terms in a Born expansion begins to break down for 300 eV electrons scattering from the large Z molecules I_2 and C_2H_5I when the scattering angle is greater than ~70°.

an intensity that is dependent on the angle between the polarization axes of the beam and the ferromagnet. The maximum variation of the scattered intensity due to this effect will be observed by reversing the orientation of the crystal magnetic axis from a parallel to an antiparallel alignment with respect to the polarization of the incoming electron beam. The reason is clear. For the case of parallel alignment of the spins of the ferromagnet and the incoming beam, the interference between the Coulomb and exchange scattering amplitudes in (2) will make a maximum positive contribution to the elastic scattering. For the case of antiparallel alignment the interference will make a maximum negative contribution. Since the scattered electron current (intensity) is proportional to the square of the scattering amplitude (2), we find that the difference in scattered current arising from parallel versus antiparallel alignment of the spin axes, divided by the incident current I_0, is given by

$$\Delta I/I_0 = \langle \sigma_e^z \rangle^2. \quad (14)$$

IV. PROPOSED EXPERIMENT

Consider an experiment where one starts with an initially unpolarized beam of electrons and polarizes it by elastic reflection from a ferromagnetic (or ferrimagnetic) crystal. Then, if one measures the intensity of an elastic reflection of this polarized beam from a second ferromagnet that has its magnetic axis first parallel and then antiparallel to that of the first crystal, the difference in intensities, ΔI, from these two secondary reflections will allow a direct measurement of the interference between the Coulomb and exchange interactions. From (14) we see that this measurement will allow one to experimentally determine the exchange induced polarization.

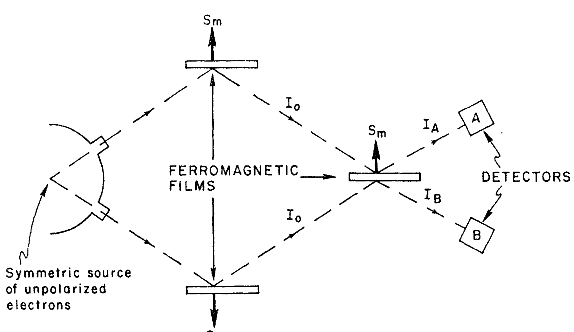

FIG. 2. Proposed arrangement for measurement of $\Delta I = I_A - I_B$.

An arrangement that will allow measurement of this intensity variation is illustrated in Fig. 2. The technique requires three crystals with their magnetic axes aligned as shown in the figure. Random fluctuations of the source intensity will be cancelled out by using a single source that is symmetric in its emission relative to the two beam axes.

Using our previous examples of low-energy electrons ($E_e < 50$ eV) and 350 eV electrons scattering from iron, we estimate from (14) that $\Delta I/I_0 = 0.74$ or 74% for the case of electrons with energy $E_e < 50$ eV, and $\Delta I/I_0 = 0.00014$ or 0.014% for the case of 350 eV electrons scattering by 60°. It thus becomes clear that this interference effect will be easily detectable only at low energies.

The measurement of $\Delta I/I_0$ will also provide an independent means of determining any quantity in the Coulomb-exchange interference in terms of the other parameters. For example, this experiment would allow one to measure the exchange potential as a function of the Coulomb potential. This technique could provide a valuable probe of the magnetic properties of ferromagnets and ferrimagnets.

It has been assumed that all of the magnetic scattering occurs from a single domain of the crystal. Although in some materials the domains become very small, it should be possible to confine a well-focused electron beam to a single domain.

Note added in proof. After this manuscript was submitted for publication, the first direct observation of coherent exchange scattering was made by Palmberg, De Wames, and Vredevoe[9] for low-energy electron diffraction from antiferromagnetic NiO. This direct observation of the magnetic scattering provides a more direct means of measuring the exchange scattering potential in magnetic crystals where the spin structure differs from that of the Coulomb potential. In crystals where the spin and potential structures are the same, such as in ferromagnetic iron, the magnetic scattering cannot be separated from that of the Coulomb potential, and it will be necessary to use other techniques, such as that proposed in this paper.

[9] P. W. Palmberg, R. E. De Wames, and L. A. Vredevoe, Phys. Rev. Letters **21**, 682 (1968).