$(4\pi)^{1/2}$ for normalization to 4π; also $Y_l^{0,c}$ and $Y_l^{0,s}$ should be multiplied by an additional factor of $(2)^{-1/2}$.

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Magnetic Scattering of X Rays from Electrons in Molecules and Solids

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The scattering of moderately high-energy x rays from electrons in magnetic solids is analyzed. We show that (a) the incoherent Compton scattering of polarized x rays can be used to determine the spin-dependent momentum distribution function of electrons in ferromagnetic materials, and (b) the coherent Bragg scattering of unpolarized x rays can be used to determine the magnetic structure of antiferromagnetic solids below their transition temperature.

Recently[1-3] there has been renewed interest in utilizing x rays to probe the electronic properties of molecules and solids.

In the extreme nonrelativistic limit, the x rays couple exclusively to the charge of the electrons. This implies that for electrons in solids the scattering cross section is independent of the magnetic properties of the medium. However, it is well known that the complete relativistic Compton amplitude does depend on the spin of the electron.[4]

The dominant charge scattering mechanism can be thought of as arising from the acceleration of the electron by the *electric* field of the wave and the *electric dipole* reradiation of the scattered field. Although it is not quantitatively correct it is qualitatively correct to think of the spin dependence of the scattering amplitude as, at least in part, arising from the same acceleration followed by a *magnetic* dipole reradiation of the scattered field. If one thinks of the electron as a little spinning ball with a radius of the order of the Compton wavelength $\lambda=\hbar/mc$ then the ratio of magnetic dipole to electric dipole radiation is roughly $k_1\lambda_c$, i.e., $\hbar\omega_1/mc^2$.

In this paper we will show that mildly relativistic x rays can be used to: (a) measure independently the momentum distributions of spin "up" and spin "down" electrons in magnetic solids, and (b) determine the magnetic crystal structure of antiferromagnetic solids below their transition temperatures. This magnetic Bragg scattering is analogous to conventional magnetic neutron scattering.

Since the binding energy of the outer electrons in atomic systems is small relative to typical x-ray energies, all of the physics we will discuss is contained in the formula for the scattering of light from free particles.[4] Binding effects will be included in the analysis only insofar as they modify the initial and final states of the electronic system. In essence we neglect the effect of binding on the intermediate states. This type of correction can be shown to modify *all* of the leading terms to be discussed here by quantities of order $E_B/\hbar\omega_1$.

Consider the scattering of light from a single free electron of momentum $\vec{p}_1$ and spin σ. This scattering is diagrammed in Fig. 1. The initial (final) photons are characterized by the four-vector k_1 (k_2) and polarization vector ϵ_1 (ϵ_2). The quantity

$$k^\mu = k_1^\mu - k_2^\mu \equiv (\vec{k}, \omega) \tag{1}$$

is the four-dimensional momentum transfer.

The complete second-order (e^2) relativistic Compton amplitude for the scattering of light, i.e., the sum of the two Feynman diagrams shown in Fig. 2, is proportional to M, where

$$M = \frac{\not{\epsilon}_2 \not{k}_1 \not{\epsilon}_1}{\omega_1} + \frac{\not{\epsilon}_1 \not{k}_2 \not{\epsilon}_2}{\omega_2}. \tag{2}$$

The slashed quantities in Eq. (2) are the usual four-dimensional dot products of vector quantities with the conventional Dirac matrices, i.e.,

$$\not{k}_1 = \omega_1 \gamma_4 - \vec{k}_1 \cdot \vec{\gamma}\ . \tag{3}$$

The scattering amplitude is obtained by taking matrix element of M between plane-wave Dirac spinors.

We remind the reader that the four-dimensional matrix $\vec{\gamma}$ explicitly contains the spin of the electron, i.e.,

$$\vec{\gamma} = \begin{pmatrix} 0 & \vec{\sigma} \\ -\vec{\sigma} & 0 \end{pmatrix}.$$

We will be interested in terms including the first nonrelativistic correction to the limiting low-energy form of Eq. (2). The appropriate reduction is simply achieved by first going to a two-dimensional representation for the four-dimensional Dirac matrices and then expanding all quantities in powers of $\hbar\omega_1/mc^2$. The result, to order $\hbar\omega_1/mc^2$, for the scattering amplitude is given by[5]

$$M_{\alpha\beta} = A\delta_{\alpha\beta} + i\vec{B} \cdot \vec{\sigma}_{\alpha\beta}\ , \tag{4}$$

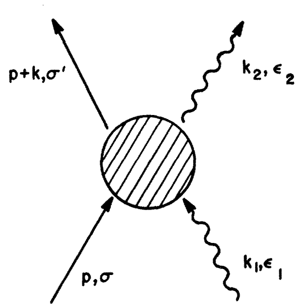

FIG. 1. Diagrammatic representation of the Compton scattering from an electron in the solid. The solid (wiggly) lines represent the incoming and outgoing electrons (photon), respectively.

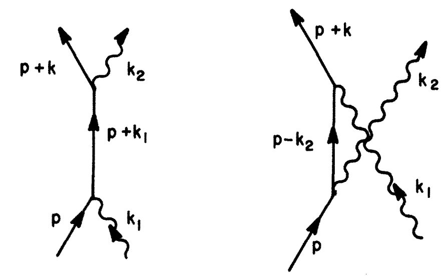

FIG. 2. Lowest-order Feynman diagrams for the Compton scattering amplitude.

where

$$A = 2\vec{\epsilon}_1 \cdot \vec{\epsilon}_2 \tag{5}$$

and

$$\vec{B} = -\frac{\omega_1}{m}\left[(\vec{\epsilon}_1 \cdot \vec{\epsilon}_2)(\hat{k}_1 \times \hat{k}_2) - \tfrac{1}{2}(\vec{p}' \cdot \vec{p}')(\vec{\epsilon}_1 \times \vec{\epsilon}_2) - (\vec{p}' \times (\vec{p}' \times (\vec{\epsilon}_1 \times \vec{\epsilon}_2)))\right]\ . \tag{6}$$

The vector $\vec{p}'$ is defined by

$$\vec{p}' = \hat{k}_1 - \hat{k}_2\ . \tag{7}$$

The leading term in Eq. (4), i.e., the quantity A, gives the usual Thompson scattering cross section. The term proportional to B leads to the spin-dependent part of the scattering amplitude. For arbitrary angles of scattering, the terms proportional to the quantity B, the spin-dependent terms, are reduced from the leading spin-independent terms by $\hbar\omega_1/mc^2$. For 50-keV x rays this is a 10% effect. In the limit where the initial electrons momentum is small compared to the photons momentum it is the recoiling electron whose moment couples to the radiation field.

To obtain the scattering amplitude from the approximate M [Eq. (4)], we must take its expectation value between two-dimensional Pauli spinors. The cross section (aside from trivial density-of-states factors) is proportional to the square of M. The interference term between the large Thompson term A and the small spin-dependent term $\vec{B}$ will give the dominant relativistic modifications in the scattering cross sections.

Consider the Compton scattering from a single free electron with a definite initial spin. To obtain the cross section we must square the matrix element M and sum over all final states (spin included) consistent with energy conservation. We find that

$$\sum_f |\langle\, i\ |\ M_{\alpha\beta}\ |\ f\,\rangle|^2 = \langle\, i\, |\, (A^* \delta_{\alpha\beta} - i\vec{B}^* \cdot \vec{\sigma}_{\alpha\beta})\, (A + i\vec{B} \cdot \vec{\sigma}_{\alpha\beta})\, |\, i\,\rangle\ . \tag{8}$$

The leading term in $|B|$, i.e., the interference term, is proportional to $\mathrm{Im}(A^*B)$. For *real* polarization vectors $\vec{\epsilon}_1$ and $\vec{\epsilon}_2$ this term vanishes. In order to observe the leading correction terms in the scattering cross section Eq. (8) implies that one must have complex, i.e., circularly polarized radiation. The cross section for light circularly polarized parallel (antiparallel) to the spin of a single electron at rest is easily computed and is given by[5]

$$\left(\frac{d\sigma}{d\Omega}\right)^{\pm} = \frac{1}{2}\left(\frac{e^2}{mc^2}\right)^2 \left(1+\cos\theta \pm \frac{2\omega_1}{m}(\cos\theta)(\cos\theta-1)\right) . \quad (9)$$

The asymmetry of the cross section about $\theta = \pi$ is due to the presence of the spin of the initial electron and the circular polarization of the incident photon. This particular effect has been used by several workers to measure the degree of circular polarization of γ rays in β-decay experiments.

If we are interested in learning something new about electrons in magnetic solids or in complex magnetic molecules, we must consider the Compton scattering from many electrons. In the so-called impulse approximation (IA) it is a simple matter utilizing Eq. (4) to compute the scattering cross section.[1,4] We must sum, with the appropriate phase factors, the amplitude M over all the electrons. In this case M becomes

$$M_{\alpha\beta} = \sum_i [A\delta^i_{\alpha\beta} + i\vec{B}\cdot\vec{\sigma}^i_{\alpha\beta}]e^{i\vec{k}\cdot\vec{r}_i} . \quad (10)$$

The cross section is easily computed (see Ref. 1). We take the absolute square of M, Eq. (10), multiply by the energy conservation δ function, sum over final states assuming they are plane waves, and obtain the cross section per particle per unit volume of material. To leading order in B_z,

$$\frac{d\sigma}{d\omega\, d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 \int \{ |A|^2[n_{\vec{p}\uparrow} + n_{\vec{p}\downarrow}] + 2[\mathrm{Im}(A^* B_z)][n_{\vec{p}\uparrow} - n_{\vec{p}\downarrow}]\} \times \delta\left(\omega - \frac{k^2}{2m} - \frac{\vec{k}\cdot\vec{p}}{m}\right)\frac{d^3p}{(2\pi)^3} . \quad (11)$$

The z axis is the direction of magnetization in the specimen, and

$$n_{\vec{p},\sigma} = \langle a^\dagger_{\vec{p},\sigma} a_{\vec{p},\sigma}\rangle \quad (12)$$

is the momentum distribution for a definite spin σ.

The interference term is proportional to the difference $(n_{\vec{p}\uparrow} - n_{\vec{p}\downarrow})$ which in turn is a measure of the spin polarization in a magnetic solid. The coefficient of proportionality $[2\,\mathrm{Im}(A^* B_z)]$ is a simple product of the vectors $\hat{k}_1\,\hat{k}_2$, u_z, $\vec{\epsilon}_1$ $(\vec{\epsilon}_1^{\,*})$, and $\vec{\epsilon}_2$ $(\vec{\epsilon}_2^{\,*})$. In order to simplify the results somewhat we consider the case where we average over the final polarization ϵ_2, and take the vector $\hat{k}_1$ along the z axis. If we now circularly polarize the incident photon, i.e.,

$$\epsilon_1 = (\hat{u}_x \pm i\hat{u}_y)/\sqrt{2}, \quad (13)$$

and take the difference of the cross section for the two circular polarizations, we find that

$$\left(\frac{d\sigma}{d\omega\, d\Omega}\right)^{\Delta} = \left(\frac{d\sigma}{d\Omega}\right)^{\Delta}_0 \int (n_{\vec{p}\uparrow} - n_{\vec{p}\downarrow}) \times \delta\left(\omega - \frac{k^2}{2m} - \frac{\vec{k}\cdot\vec{p}}{m}\right)\frac{d^3p}{(2\pi)^3} , \quad (14)$$

where

$$\left(\frac{d\sigma}{d\Omega}\right)^{\Delta}_0 = \frac{4\hbar\omega_1}{mc^2}\left(\frac{e^2}{mc^2}\right)^2 \cos\theta(\cos\theta - 1) \quad (15)$$

and θ is the scattering angle.

We could just as well have used unpolarized light and measured the difference of the cross sections for the two degrees of circular polarization of the scattered photon. For arbitrary scattering angle only $(d\sigma/d\Omega)^{\Delta}_0$ would change. In either case the spin-dependent Compton experiment is difficult to perform since neither polarized x-ray sources nor polarization detectors are readily available.

To this point we have discussed the *incoherent* Compton scattering from electrons in matter. We would now like to investigate the *coherent* Bragg scattering from electrons in solids.[6]

Since the scattering amplitude depends on the spin of the electrons, Bragg scattering utilizing x rays *will* reflect the magnetic structure of the lattice. This is directly analogous to the situation that occurs in neutron scattering.[7] As in the case of neutrons such scattering does not require a polarized x-ray beam. In the Compton effect we do not measure the spin of the recoiling electron. In Bragg scattering the electron has the same initial and final polarization. In any spin-dependent scattering at least two spins or polarizations must be specified in order that there be an observable spin-dependent modification of the cross section.

In order to compute the Bragg scattering cross section, the matrix element, Eq. (9), is taken between the *same* initial and final electronic states. The scattering is elastic and the momentum is transferred, at the appropriate Bragg scattering angles, to the lattice. For an insulating antiferromagnet the matrix element which depends on the spin of the electrons is simply

$$M' = iB_z \langle 0 | \textstyle\sum_i \sigma^z_i e^{i\vec{k}\cdot\vec{r}_i} | 0\rangle \sum_n e^{i\vec{k}\cdot\vec{R}_n}(\pm) . \quad (16)$$

The states $\langle 0 |$ are the atomic orbitals and the sum over i in Eq. (16) is over those electrons localized on a single magnetic site. The sum on the index n, over different crystalline sites, is multiplied by a $\pm$ sign depending on the orientation of the sublattice magnetization. It yields the Bragg peaks when the momentum transfer coincides with the magnetic reciprocal-lattice vector K_n'.

When spin-orbit coupling is negligible and all of the magnetic electrons are in the same orbital (a not uncommon situation) then Eq. (17) becomes

$$M' = iB_z F^M(\vec{k})\, 2S_z\, \delta(\vec{k} - \vec{K}_n') \quad . \tag{17}$$

Here S_z is the total spin on a single magnetic site and $F^M(\vec{k})$ is the Fourier transform of the single atomic orbital of the outer magnetic electrons, i.e.,

$$F^M(\vec{k}) = \int e^{i\vec{k}\cdot\vec{r}}\, |\varphi^M|^2 d^3r \quad . \tag{18}$$

In order to estimate the importance of the magnetic Bragg scattering we choose to compare it to the ordinary Bragg scattering. Since the Bragg scattering is zero at the *new* points of the magnetic lattice the interference term in the cross section vanishes. It is the square $R \equiv (B/A)^2$ which is relevant. To within polarization factors this ratio is

$$R = \left[B_z \frac{F^M(K_n')}{F(K_n')} \frac{N_M}{2N} \right]^2 \quad . \tag{19}$$

The quantity N_m/N is the relative number of magnetic electrons and $F(K)$ is the usual atomic form factor.

It is well known that the form factor $F^M(\vec{k})$ decreases rapidly with increasing k since it is only the outer electrons which contribute to it. The wave vector k must be of order a^{-1} (a being a typical atomic distance) in order that $F^M(k) \approx 1$. This in turn implies (as for ordinary Bragg scattering) that the scattering of hard 10–50-keV x rays must be in the forward direction. In the forward direction the dominant term in $\vec{B}$ is

$$\vec{B} \cong -(\omega_1/m)(\vec{\epsilon}_1 \cdot \vec{\epsilon}_2)\hat{\vec{k}}_1 \times \hat{\vec{k}}_2 \quad . \tag{20}$$

If Eq. (20) is substituted into Eq. (19), it gives us a relative scattering intensity at the magnetic Bragg peak of

$$R = [(\hbar/mc)\vec{K}_n' F^M(\vec{K}_n')\, N_m/2N]^2 \quad . \tag{21}$$

In writing Eq. (21) we have assumed that the form factor for Bragg scattering is unity. The ratio R in Eq. (21) is independent of the incident energy. It only depends on the momentum transfer K_n', i.e., the lattice geometry. In a specific experimental situation the function $K_n' F^M(K_n')$ is slowly varying so that there will always be several reciprocal-lattice vectors where R is near its maximum value. The optimum magnetic Bragg scattering angle, in any given experiment, will depend almost exclusively on the geometry of the reciprocal lattice. Under typical conditions, however, R is approximately 10^{-6}.

Although R is a small quantity, it is a number which is definitely within the range of observation. The use of x rays to determine magnetic structures would be interesting in itself, but useful because of the fact that x-ray sources are easily available, whereas more conventional neutron piles are scarce.

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