Theories of Compton Scattering by Magnetic Materials *

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Z. Naturforsch. **48a**, 261–265 (1993); received December 14, 1991

Theoretical work on the cross-section for Compton scattering by magnetic materials is surveyed. Exact results for scattering by a free polarized electron are contrasted with corresponding results obtained perturbatively for a model of bound electrons with a finite width to the momentum distribution.

Key words: Compton scattering; Scattering cross-section; Magnetic materials; Spin density in momentum space.

1. Introduction

Radiation scattering experiments on magnetic materials continue to provide new and exciting science. Recent examples include polarization and resonance studies of x-ray magnetic scattering in rare-earth metals [1] and white-beam non-resonant diffraction of synchrotron radiation from ferromagnetic iron [2]. Material properties probed in such experiments are weak structural modulations that accompany the magnetic ordering of some rare-earth metals and that are described by a so-called spin-slip model, and the spin and orbital contributions to the magnetic-moment distribution. For the most part, neutron scattering is the preferred method for the study of magnetic configurations, structures and excitations. Notable among recent achievements is the confirmation of an interplay between spin and orbital moments in actinide compounds [3, 4]. There are fresh challenges, to both experimental and theoretical methods, presented by the development of new magnetic materials with attractive properties for applications in various devices [5].

In the present paper we gather results for Compton and total scattering of photons by magnetic materials. Discussions are at a basic level in as much that no specific experimental data are analysed. Rather, key elements of standard theoretical models are summarized and compared. Hence, we begin by recording the cross-section for photon scattering by a free polarized electron initially at rest, a result that is an outcome of a straightforward exercise with QED [6]. Moving to a more realistic model, in which the momentum distribution for the electrons has a finite width, we consider the impulse approximation to the cross-section for pure charge scattering, and its expansion as a function of the initial momentum of the electron. This work provides a link between studies by Ribberfors [7] and Grotch et al. [8], which are at first sight quite different. A derivation in § 4 of the Compton profile for (non-relativistic) charge scattering hopefully sheds fresh light on the real nature of the approximations involved. The method employed is similar to that used for the derivation of Compton profiles for spin and orbital magnetic moments [9]. These topics are addressed in §§ 5 and 6, where following on from the work by Grotch et al. we provide an expression for the Compton profile for scattering by a material with pure spin moments, and the behaviour of the total scattering from spin and orbital moments. For the special case of pure spin scattering it is stressed that the polarization-induced contribution to the cross-sections for models of free and bound electrons are different, at least at the level of current theoretical work. Before embarking on discussions of these various topics, perhaps it is worth remarking that the theory of photon scattering by electrons is based on QED which is thoroughly tested and proven [10].

2. Scattering from a Free Polarized Electron

The cross-section for scattering by a free polarized electron can be calculated without approximation. Here we record the result for the special case in which the free electron is *initially at rest*, and the primary photon beam possesses circular polarization. The result for unpolarized photons and electrons is usually referred to as the Klein–Nishina formula.

* Presented at the Sagamore X Conference on Charge, Spin, and Momentum Densities, Konstanz, Fed. Rep. of Germany, September 1–7, 1991.

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Let the ratio of the secondary and primary photon energies be denoted by $b = (q'/q)$, where $\boldsymbol{q}$ and $\boldsymbol{q}'$ are the corresponding wave vectors. If the primary photon is scattered through an angle θ, the energy–momentum relation is

$$g(1 - \cos\theta) = \left(\frac{1-b}{b}\right), \tag{2.1}$$

where $g = (\hbar q/mc)$ is the ratio of the primary photon energy to the rest mass of the electron. The degree of primary circular polarization is described by a Stokes parameter P_2, and the spin moment of the electron is $\langle \boldsymbol{s} \rangle$. With this notation, the differential cross-section for scattering into a solid-angle element $\mathrm{d}\Omega$ is [6]

$$\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega} = \frac{1}{2} r_e^2 b \{(1 + \cos^2\theta) - (1-b)(b + \cos^2\theta) - 2P_2(1-b)\langle \boldsymbol{s} \rangle \cdot (\hat{\boldsymbol{q}} \cos\theta + b\hat{\boldsymbol{q}}')\}. \tag{2.2}$$

In the limit $b \to 1$ we recover Thomson's formula. This limit is achieved for $\hbar \to 0$ (which means $g \to 0$) or $\theta \to 0$ for any value of the primary energy.

It is possible to choose a scattering geometry such that the contribution induced by the circular polarization vanishes. One obvious scheme is to arrange the spin polarization perpendicular to the plane defined by the primary and secondary wave vectors $\boldsymbol{q}$ and $\boldsymbol{q}'$. If, on the other hand, $\langle \boldsymbol{s} \rangle$ is in the scattering plane, and the unit vectors in (2.2) are expressed in the form

$$\hat{q}_z = \cos\alpha \quad \text{and} \quad \hat{q}'_z = \cos(\alpha - \theta),$$

where α is the angle between $\langle \boldsymbol{s} \rangle$ and $\boldsymbol{q}$, the condition at which the spin contribution vanishes,

$$\hat{q}_z \cos\theta + b\hat{q}'_z = 0,$$

can be expressed in the form

$$\tan\alpha \tan\theta = -(1 + 1/b). \tag{2.3}$$

This relation is exact for the case of a free electron at rest before the scattering event.

3. Impulse Approximation for Pure Charge Scattering

A realistic model for the interpretation of data for electrons in molecules and solids must incorporate a non-trivial distribution for the momenta of the electrons. Let the distribution be denoted by $\varrho(\boldsymbol{k})$ and normalized such that

$$\int \mathrm{d}\boldsymbol{k}\, \varrho(\boldsymbol{k}) = 1.$$

For an electron at rest $\varrho(\boldsymbol{k}) = \delta(\boldsymbol{k})$, where $\delta(\boldsymbol{k})$ is the delta-function.

For this model it is not possible to calculate the cross-section without making approximations, except for some very special cases which we shall not discuss. The most successful approximation to describe scattering of energetic radiation (deep inelastic processes) appears to be one based on the impulse approximation, where the target particle is assumed to behave as a free particle for the brief duration of the scattering event. Applied to electrons in condensed matter there is need for the further assumption that scattering is dominated by independent electron events, i.e. spatial correlations between target particles (electrons) can be safely neglected. Physical intuition leads one to expect the independent-particle (or incoherent) approximation to be valid when the photon wavelength is small compared to the mean separation between target particles.

In words, the impulse approximation to the cross-section is the cross-section for scattering by a free electron with momentum $\boldsymbol{p}_i$ with an average over $\boldsymbol{p}_i$ performed according to the distribution $\varrho(\boldsymbol{p}_i)$. Since the cross-section for scattering by a free electron with an arbitrary initial momentum can be reduced to a simple analytic expression, the impulse approximation to the cross-section for a bound electron can be expressed in a closed form that can be evaluated (numerically) for a given momentum distribution. The analytic expression for the impulse approximation to the cross-section is provided by Ribberfors [7], Eqn. (8). For our part we have evaluated this cross-section as an expansion in the initial momentum of the target electron motivated by the realization that, in most cases of interest, the mean value of the momentum in the initial bound state is relatively very small, being of order $(mc/137)$. The result for the partial differential cross-section is

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'} = \frac{1}{2} r_e^2 (q'/q) \int \mathrm{d}\boldsymbol{p}\, \varrho(\boldsymbol{p})\, \delta\left\{\hbar\omega - \frac{\hbar^2}{2m}(k^2 + 2\boldsymbol{k}\cdot\boldsymbol{p})\right\} \cdot \left([1 + \cos^2\theta] - \frac{2\hbar}{mc}\boldsymbol{p}\cdot(\hat{\boldsymbol{q}} + \hat{\boldsymbol{q}}')\cos\theta(1 - \cos\theta)\right). \tag{3.1}$$

If $\varrho(\boldsymbol{p}) = \varrho(-\boldsymbol{p})$, the leading-order correction to the standard expression for the Compton profile vanishes. In this instance the first finite correction is of order $\boldsymbol{p}^2$, and the appropriate expression is obtained from the general expression by straightforward tedious algebra.

The standard Compton profile, often written in terms of the van Hove response function,

$$S(\boldsymbol{k}, \omega) = \int \mathrm{d}\boldsymbol{p}\, \varrho(\boldsymbol{p})\, \delta\left\{\hbar\omega - \frac{\hbar^2}{2m}(k^2 + 2\boldsymbol{k}\cdot\boldsymbol{p})\right\}, \tag{3.2}$$

has been shown to accurately describe deep inelastic scattering events. Note that, in this limit, the total intensity is independent of $\boldsymbol{k}$,

$$\hbar \int_{-\infty}^{\infty} \mathrm{d}\omega\, S(\boldsymbol{k},\omega) = 1\,, \tag{3.3}$$

while the result

$$\int_{-\infty}^{\infty} \mathrm{d}\omega\, \omega\, S(\boldsymbol{k},\omega) = (k^2/2\,m) \tag{3.4}$$

agrees with the exact f-sum rule (relativistic forces between the electrons are here assumed to be negligible). Using (3.2),

$$\int_{-\infty}^{\infty} \mathrm{d}\omega\, \omega^2\, S(\boldsymbol{k},\omega) = \left(\frac{k^2}{2\,m\,\hbar}\right)\left\{\frac{(\hbar\, k)^2}{2\,m} + \frac{4}{3}\langle \mathrm{KE}\rangle\right\}, \tag{3.5}$$

where $\langle \mathrm{KE}\rangle$ is the average value of the kinetic energy of an electron in the initial state. The result (3.3) can be used to gauge the value of the impulse approximation to describe data, whereas (3.4) tests the quality of the data. The result (3.5) then provides a reliable estimate of $\langle \mathrm{KE}\rangle$. Higher-order sum-rules are more complicated and possibly of little value in data analysis.

4. A Derivation of the Impulse Approximation for the Compton Profile

The following derivation of the impulse approximation, given for pure charge scattering, should shed fresh light on the real nature of the various approximations made, although at first sight it could appear meretricious. The full van Hove response function for (non-relativistic) charge scattering is the time Fourier transform of the density autocorrelation function,

$$\langle n^+(\boldsymbol{k})\, n(\boldsymbol{k},t)\rangle = \sum_{jj'} \langle \exp(-\,i\,\boldsymbol{k}\cdot\boldsymbol{R}_{j'}) \exp(i\,\boldsymbol{k}\cdot\boldsymbol{R}_j(t))\rangle\,, \tag{4.1}$$

where $\boldsymbol{k} = \boldsymbol{q} - \boldsymbol{q}'$, $\boldsymbol{R}_j$ is the position of the jth electron and $\boldsymbol{R}_j(t)$ is a Heisenberg operator. On writing the latter out in full, the correlation function on the right-hand side of (4.1) is

$$\langle \exp(-\,i\,\boldsymbol{k}\cdot\boldsymbol{R}_{j'}) \exp(i\,t\,H/\hbar) \exp(i\,\boldsymbol{k}\cdot\boldsymbol{R}_j) \cdot \exp(-\,i\,t\,H/\hbar)\rangle\,, \tag{4.2}$$

in which H is the Hamiltonian for the target electrons. Next we perform some rearrangements that bring (4.2) to a convenient form.

First we bring the operator $\exp(-\,i\,H\,t/\hbar)$ from the right to the left. This does not change the correlation function because the average value (denoted by angular brackets) is taken with respect to the states of H, namely

$$\langle \ldots \rangle \propto \mathrm{Tr}\{\exp(-\,H/k_{\mathrm{B}}\,T)\ldots\}\,,$$

and the trace operation is invariant with respect to a cyclic permutation of operators. The second step is to insert unity before $\exp(i\,\boldsymbol{k}\cdot\boldsymbol{R}_j)$ in the guise of

$$\exp\{i\,\boldsymbol{k}\cdot(\boldsymbol{R}_{j'} - \boldsymbol{R}_{j'})\} = 1\,,$$

and so create the combination

$$\exp(-\,i\,\boldsymbol{k}\cdot\boldsymbol{R}_{j'}) \exp(i\,t\,H/\hbar) \exp(i\,\boldsymbol{k}\cdot\boldsymbol{R}_{j'})\,.$$

Here, the exponential operators involving $\boldsymbol{k}\cdot\boldsymbol{R}_{j'}$ constitute a unitary transform on H in which the momentum operator $\boldsymbol{p}_{j'}$ becomes $(\boldsymbol{p}_{j'} + \hbar\,\boldsymbol{k})$; the transformed Hamiltonian is denoted by $H_{j'}$. Following these two steps, the correlation function of interest is

$$\langle \exp(-\,i\,t\,H/\hbar) \exp(i\,t\,H_{j'}/\hbar) \exp\{i\,\boldsymbol{k}\cdot(\boldsymbol{R}_j - \boldsymbol{R}_{j'})\}\rangle\,. \tag{4.3}$$

The product of the exponential operators that involve H and $H_{j'}$ can be combined using the Campbell–Backer–Hausdorff formula,

$$\exp(-\,i\,t\,H/\hbar) \exp(i\,t\,H_{j'}/\hbar) = \exp\left\{\frac{i\,t}{\hbar}(H_{j'} - H) + \frac{t^2}{2\,\hbar^2}[H, H_{j'}] + \ldots\right\}, \tag{4.4}$$

in which additional terms on the right-hand side are labelled by ascending powers of t.

To proceed to the impulse approximation, based on the leading term in (4.4), let H be the sum of kinetic terms for identical particles of mass m, and a potential energy V that is not a function of the momenta (relativistic interactions are therefore excluded). In this instance,

$$H_j - H = \frac{\hbar}{2\,m}(\hbar\,k^2 + 2\,\boldsymbol{k}\cdot\boldsymbol{p}_j) \tag{4.5}$$

and

$$[H, H_j] = i\,\hbar^2\,(\boldsymbol{k}\cdot\boldsymbol{\nabla}_j V)/m\,. \tag{4.6}$$

Hence, neglect of the term in t^2 in (4.4), and all higher-order terms, implies that for the duration of the scattering event $t \sim (\hbar/E)$ the mean value of $(\hbar\,k^2 + 2\,\boldsymbol{k}\cdot\boldsymbol{p})$ exceeds the mean value of $t\,(\boldsymbol{k}\cdot\boldsymbol{\nabla} V)$. This can be transformed to an inequality for the primary energy E, namely

$$E \gg \left(-\frac{1}{k}\right)\left(\frac{\mathrm{d}V}{\mathrm{d}r}\right)_a, \tag{4.7}$$

where a is the radius of the atomic orbital. Note that (4.7) is well satisfied for a sufficiently large value of the scattering vector $\boldsymbol{k}$. On taking $(a\,k) \sim 1$ and $V \sim (Z\,e^2/r)$, the inequality (4.7) provides the familiar estimate

$$E \gg (Z\,e^2/a) \sim Z\,R_\infty,$$

where R_∞ is the Rydberg energy unit.

The second aproximation to be made involves spatial correlations. Given that k is large, the exponential of $\{(\boldsymbol{R}_j - \boldsymbol{R}_{j'}) \cdot \boldsymbol{k}\}$ in (4.3) will oscillate rapidly, taking positive and negative values, as the sums in (4.1) range over all the electrons. In consequence, all but the terms $j = j'$ will largely cancel, i.e. the incoherent approximation can be safely invoked. Combining this observation together with the approximation for the dynamics reduces (4.3) to

$$\left\langle \exp\left\{\frac{i\,t}{2\,m}(\hbar\,k^2 + 2\,\boldsymbol{k}\cdot\boldsymbol{p}_j)\right\}\right\rangle;\; j = j', \quad \text{and zero otherwise.}$$

Using

$$\int \mathrm{d}\boldsymbol{p}\,\delta(\boldsymbol{p} - \boldsymbol{p}_j) = 1$$

and the standard definition for the momentum distribution,

$$\langle \delta(\boldsymbol{p} - \boldsymbol{p}_j)\rangle = \varrho(\boldsymbol{p}), \tag{4.8}$$

we arrive finally at the standard Compton profile,

$$\begin{aligned} S(\boldsymbol{k},\omega) &= \left(\frac{1}{2\,\pi\,\hbar\,N}\right)\int_{-\infty}^{\infty} \mathrm{d}t \exp(-\,i\,\omega\,t)\langle n^+(\boldsymbol{k})\,n(\boldsymbol{k},t)\rangle \\ &= \int \mathrm{d}\boldsymbol{p}\,\varrho(\boldsymbol{p})\,\delta\{\hbar\,\omega - E_k - \hbar^2\,\boldsymbol{k}\cdot\boldsymbol{p}/m\}, \end{aligned} \tag{4.9}$$

where $E_k = (\hbar\,k)^2/2\,m$ is the recoil energy. Note that the two approximations involved in reaching the standard Compton profile, one for dynamic and the other for static correlations, have a common denominator in as much as they are valid for sufficiently large k.

Another scheme by which to arrive at (4.9) is to use the short-time expansion,

$$\boldsymbol{R}_j(t) = \boldsymbol{R}_j(0) + \frac{t}{m}\boldsymbol{p}_j, \tag{4.10}$$

where $\boldsymbol{R}_j(0) = \boldsymbol{R}_j$ and $\boldsymbol{p}_j$ are, of course, conjugate variables. Taken together with the incoherent approximation, which means keeping just the self-terms $j = j'$ in (4.1), it leads directly to (4.9). The next term in (4.10) would involve the force acting on the jth particle, as in (4.6). This is negligible compared to the retained momentum term over a time $t \sim (\hbar/E)$ provided the foregoing inequalities are well satisfied.

5. Compton Profile for Bound Magnetic Electrons

A realistic model for the interpretation of photon scattering by a magnetic material is based on *unpaired* electrons in atomic (bound) orbitals, in which the momentum distribution has a finite width. The scattering probability for such a model has been calculated perturbatively by Grotch et al. [8] for circularly polarized primary photons. Contributions of order e^2 (single Compton scattering) arise from first-order perturbation theory, in which interactions occur at a single space–time point, as well as from second-order perturbation theory. For the latter case the emission and absorption of photons occur at different space–time points. The authors checked their complicated expression for the cross-section by performing an independent calculation of the free relativistic cross-section, including polarization effects. In the appropriate limit this result confirms their cross-section.

Grotch et al. perform a systematic expansion of their cross-section in powers of $(1/m)$. In this way corrections of order $g = (E/m\,c^2)$ and $(\hbar\,p_i/m\,c)$ in the amplitude are obtained. As far as the cross-section is concerned, the leading-order momentum terms are precisely those given in (3.1). The polarization-induced spin-dependent scattering is quite complicated, because contributions come from both the first-order and second-order perturbation terms, whereas orbital magnetism comes solely from the latter. In the following expression, adapted from Grotch et al., orbital scattering is not included, and just leading-order momentum and spin corrections to Thomson (non-relativistic) scattering are retained:

$$\begin{aligned} \frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E'} &= \frac{1}{2}r_e^2\,b\int \mathrm{d}\boldsymbol{p}\,\delta\{\hbar\,\omega - E_k - \hbar^2\,\boldsymbol{k}\cdot\boldsymbol{p}/m\} \\ &\cdot\Big\{(1+\cos^2\theta)\,\varrho(\boldsymbol{p}) - \frac{2\hbar}{m\,c}\boldsymbol{p}\cdot(\hat{\boldsymbol{q}}+\hat{\boldsymbol{q}}')\cos\theta\,(1-\cos\theta)\,\varrho(\boldsymbol{p}) \\ &\quad - 2\,g\,P_2\,\langle \boldsymbol{s}_j\,\delta(\boldsymbol{p}-\boldsymbol{p}_j)\rangle\cdot[(1-\cos\theta)(\hat{\boldsymbol{q}}\cos\theta + \hat{\boldsymbol{q}}') \\ &\quad + \tfrac{1}{2}(b-1)\,\{\hat{\boldsymbol{q}}(1+\cos\theta) + \hat{\boldsymbol{q}}'(1-3\cos\theta)\}]\Big\}, \end{aligned} \tag{5.1}$$

in which $b = (q'/q)$ as in (2.2). The spin-dependent scattering in (5.1) is different from that in (2.2) because the cross-sections are for different models (bound and free electrons), and they are computed to different levels of accuracy ((2.2) is exact, whereas (5.1) is obtained by expansion in $1/m$). The additional orbital contribution not included in (5.1) is discussed by Lovesey [9].

The spin-dependent scattering in (5.1) vanishes when $\tan\alpha$ (cf. (2.3)) satisfies

$$\tan\alpha\sin\theta\{1-\cos\theta+\tfrac{1}{2}(b-1)(1-3\cos\theta)\} \quad (5.2)$$
$$+(1-\cos\theta)\{2\cos\theta+\tfrac{1}{2}(b-1)(1+3\cos\theta)\}=0.$$

The difference between this result and (2.3) is attributed to the points made following (5.1).

6. Total Scattering

Total scattering is related to instantaneous correlations, as is evident from the integral of $S(\boldsymbol{k},\omega)$ taken over all ω ($\boldsymbol{k}$ fixed). In one sense it is the opposite extreme to Bragg scattering, which is proportional to the time average ($t\to\infty$) of the correlations and strictly elastic. More precisely, the total and Bragg scattering cross-sections for pure charge scattering are proportional to $\langle|n_{\boldsymbol{k}}|^2\rangle$ and $|\langle n_{\boldsymbol{k}}\rangle|^2$, respectively [9].

The following expressions for the polarization-induced charge–magnetic interference contribution to the total cross-section are derived for the model of bound electrons used by Grotch et al. [8] and described in the previous section. The spin contribution can be derived from (5.1). If the primary photon energy is sufficiently large, scattering is only mildly inelastic ($q\sim q'$ or $b\sim 1$), and on integrating over ω the spin contribution to (5.1) is just

$$-r_e^2\, g\, P_2(1-\cos\theta)\langle\boldsymbol{S}\rangle\cdot(\hat{\boldsymbol{q}}\cos\theta+\hat{\boldsymbol{q}}'). \quad (6.1)$$

The corresponding result for the orbital contribution, which is to be added to (6.1), is

$$-\tfrac{1}{2}r_e^2\, g\, P_2\sin\theta(1+\cos\theta)(k/q)\{\hat{k}_x\langle L_z\rangle-\hat{k}_z\langle L_x\rangle\}. \quad (6.2)$$

Here, the z-axis defines the quantization axis (in a fully saturated magnet the non-zero components of $\langle\boldsymbol{S}\rangle$ and $\langle\boldsymbol{L}\rangle$ are parallel to the z-axis). Note that the orbital contribution vanishes for $\theta\to 0$ and $\theta\to\pi$. Optimum geometries are discussed by Collins et al. [11]. The latter work demonstrates that the special position at which the sum of (6.1) and (6.2) vanishes does provide a unique value for the ratio of the spin and orbital magnetizations. Such information is not directly available by other experimental methods [4].

7. Conclusions

The brief survey of some key features of the theory of Compton scattering has aimed to link work for pure charge scattering with technically much more complicated calculations for a realistic model of a magnetic material. For a polarized free electron the cross-section can be calculated without approximation. The result provides insight and confidence for corresponding results for charge and spin scattering by bound electrons tackled perturbatively. However, similar comfort is not available for the theory of orbital scattering since there is no such scattering from free electrons (orbital scattering is induced for free electrons by application of a magnetic field)*.

Acknowledgement

Discussions and correspondence with S. P. Collins, G. H. Lander, R. E. Peierls, S. Manninen, P. Platzman, and N. Sakai are gratefully acknowledged.

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* *Note added in proof:* Relativistic corrections for pure charge scattering one analysed from a different viewpoint by F. Sacchetti, Phys. Rev. B **36,** 3147 (1987).