Magnetic x-ray scattering at relativistic energies

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The photon magnetic scattering cross section is examined in the high-energy limit in order to define the validity of the nonrelativistic approach usually employed to interpret the experimental results. A proper definition of the magnetic contribution is introduced and a high-energy limit is deduced. It is shown that the elastic magnetic cross section decreases or stays constant on increasing the photon energy when the momentum transfer is held constant in agreement with recent experimental results. [S0163-1829(98)01433-7]

For a long time x-ray scattering has been one of the most important tools for condensed-matter studies. The advent of the very high brilliance synchrotron radiation sources has even extended the range of possible applications of the x ray in the analysis of condensed-matter properties. In particular, it became possible to use the weak photon-electron magnetic-moment interaction in order to determine the magnetic properties of a many-electron system. This interaction has been demonstrated to be rather useful as a companion technique to magnetic neutron scattering in the study of the magnetic properties of matter.

In order to perform accurate studies employing x-ray magnetic scattering it is mandatory to have a fully accurate formulation of the corresponding cross section. Unfortunately it is *impossible* to derive an *exact* expression of the photon-scattering cross section off a many-bound-electron system even at the lowest nonvanishing order.[1–3] When the magnetic interaction is considered the situation is even more complex. A standard description is that given by Blume[4] and Blume and Gibbs[5] derived within the standard nonrelativistic approach of the photon-electron interaction. This approach is considered to be adequate when the photon energy is small with respect to the electron rest energy ($mc^2 = 511$ keV). In the case of magnetic Compton scattering a more accurate development has been proposed in Ref. 6, in relation to an accurate nonrelativistic Hamiltonian of a combined system of many electrons and photons. More recently Arola, Strange, and Gyorffy[7] presented a formal extension of the standard perturbation relativistic theory to a many-electron system in a form suitable for numerical calculations. In this paper, we derive a formulation useful for the interpretation of experimental results.

One has to remember that in calculating the electron-photon cross section the main parameter is the fine-structure constant $\alpha = e^2 \cong \frac{1}{137}$ (in the following we shall use natural units $c=1$ and $h=2\pi$). This (small) constant is the development parameter that is used to describe the cross section. However, if one has to derive a nonrelativistic formulation of the electron-photon interaction a new (small) parameter has to be introduced, formally $1/c$. Therefore there are two (small) parameters governing the cross section, but it is not obvious which is the correct procedure for the development of the exact (unknown) cross section. In fact, one can use the exact electron-photon interaction to derive the cross section at the lowest (nonvanishing) order in α in order to develop the resulting expression in power of $1/c$. However, this is the most difficult approach. In Refs. 4 and 5 the opposite procedure is used, namely, the electron-photon interaction is developed in power of $1/c$ and the cross section is derived at the lowest (nonvanishing) order in α. It is not clear how to establish whether the two procedures are, at least in principle, equivalent.

In the case of the scattering from a single free electron it has been known for a long time[8,9] that the exact scattering amplitude developed with respect to the incoming photon energy contains a first-order (in $1/c$) magnetic scattering. This contribution is formally related to the relativistic invariance,[8,9] that is, the magnetic photon scattering is a pure relativistic effect.

Considering the general situation we examined the magnetic photon scattering looking at the high-energy behavior of the cross section. One can note that in a full relativistic formulation the magnetic contribution to the cross section cannot be defined in a straightforward way because the electron magnetic moment is intrinsic to the theory and does not explicitly appear in the photon-electron interaction as has been already observed in the first study[10] of the magnetic Compton cross section in a many-body system. In the following, we shall consider as quasielastic a process where the incoming and outgoing photon energies are close to each other. On increasing the photon energy these processes are generally confined in the forward direction so that the simplified assumption $\mathbf{k}_0 \cong \mathbf{k}$ is valid.

If we assume that the exact relativistic many-body states of the system one is examining are known, the quasielastic cross section developed at the lowest nonvanishing order in the coupling constant α is given by

$$d\sigma/d\Omega = \rho_F |K_{F0}|^2, \tag{1}$$

where ρ_F is the density of the final states of the whole system and K_{F0} is the matrix element of the electron-photon interaction. K_{F0} can be written as

$$K_{F0} = e^2 \sum_I \frac{\left\langle F \left| \sum_i \alpha_i \cdot \mathbf{A}(\mathbf{x}_i) \right| I \right\rangle \left\langle I \left| \sum_j \alpha_j \cdot \mathbf{A}(\mathbf{x}_j) \right| 0 \right\rangle}{E_0 - E_I}, \tag{2}$$

where α_i is the vector of the Dirac matrices of the ith electron, whose position is $\mathbf{x}_i$, and $\mathbf{A}(\mathbf{x}_i)$ is the vector potential operator describing the photon field; $|0\rangle$, $|I\rangle$, and $|F\rangle$ are the initial, intermediate, and final states of the whole system

composed by the photon and the target; and E_0 and E_I are the total energies of the corresponding states. It is clear that Eqs. (1) and (2) are not explicitly covariant, but we prefer to use a noncovariant form because in a condensed-matter experiment the laboratory frame is a special reference where one is interested in studying various observables.

As observed in Ref. 10 the electron spin is formally contained in the Dirac matrices that are expressed in terms of the Pauli matrices. However, the complex structure of Eq. (2) does not allow for a straightforward definition of the magnetic terms. In the available fully relativistic formulations at the lowest nonvanishing order in the coupling constant α (Refs. 11 and 12), the magnetic contributions are deduced at the end of a long calculation as those terms that depend explicitly on the electron polarization. This approach is of little use for condensed-matter applications where one needs a well-defined cross section containing the *unknown* observables of the target system possibly completely disentangled from the properties of the probe.

Therefore all meaningful approaches useful for condensed-matter applications must pass through some approximation that should be carefully examined in comparison to the experimental situation.

In order to identify the structure of the magnetic terms we start examining the usual Compton cross section for the scattering off a single free electron. Instead of the standard formulation, based on the Feynman approach, we follow that described by Heitler,[12] who shows that the transition probability from the initial electron state $|0\rangle$ to the final state $|F\rangle$ is proportional to the square modulus of the matrix element:

$$K_{F0}=\{u^{\dagger}[2(\mathbf{e}_0\cdot\mathbf{e})+(\mathbf{e}\cdot\alpha)(\mathbf{k}_0\cdot\alpha)(\mathbf{e}_0\cdot\alpha)/k_0 + (\mathbf{e}_0\cdot\alpha)(\mathbf{k}\cdot\alpha)(\mathbf{e}\cdot\alpha)/k]u_0\}/(2\mu), \tag{3}$$

where $\mathbf{e}_0$ and $\mathbf{e}$ are the polarization vectors of the incoming and outgoing photons, u_0 and u are the initial and final electron spinors, and μ is the electron rest energy. Starting from Eq. (3) and summing the square modulus of K_{F0} on the spin of u_0 and u, one gets the usual Klein and Nishina formula. However, if one considers Eq. (3) as it is, it is easily seen that the first term in the right-hand side of Eq. (3) cannot give rise to spin-dependent contributions, while magnetic contributions are given by those terms containing three times the Dirac matrix. Therefore, one can define as magnetic those terms that involve three times the Dirac matrix. Having this in mind it is evident that the magnetic contribution to the photon scattering is a relativistic effect, as the matrix element of the product of three Dirac matrices is proportional to the product of *large* and *small* components of the Dirac spinors, which is zero in a nonrelativistic theory. Incidentally one can also observe that the small component of the Dirac spinor is proportional to the large one through the spin operator.

As a consequence of this discussion, to arrive at a cross section useful for our purpose we adapt the approach described by Heitler[12] to derive the Klein and Nishina Compton cross section. The main difficulty one finds in studying Eq. (2) is the fact that the energy denominator explicitly depends on the unknown energies of the intermediate states, which in turn depend on the target Hamiltonian that is unknown and can be considered as the subject of the experimental investigations. In the case of a single free electron these energies are known and the momentum conservation holds at each vertex so that the Feynman formulation can be applied. When the bound many-particle states have to be considered, one has to introduce a proper approximation because no rules are available at the interaction vertex and the target states have to be considered unknown.

In order to disentangle photon and target in Eq. (2) we first observe that, as usual, two intermediate states are possible with no photons or two photons, corresponding to the usual direct and crossed terms. The matrix element for the direct term is given by

$$K_{F0}^{(d)}=e^2\sum_I \frac{\left\langle F\left|\sum_i (\alpha_i\cdot\mathbf{e})\exp[\mathbf{i}\mathbf{k}\cdot\mathbf{x}_i]\right|I\right\rangle\left\langle I\left|\sum_j (\alpha_j\cdot\mathbf{e}_0)\exp[-\mathbf{i}\mathbf{k}_0\cdot\mathbf{x}_j]\right|0\right\rangle}{E_0-E_I}, \tag{4}$$

where one can write $E_0=k_0+N(\mu+\varepsilon_0)$ and $E_I=N(\mu+\varepsilon_I')$. In Eq. (3), N is the number of electron in the system, assumed to be arbitrarly large, ε_0 is the ground-state binding energy per electron, and ε_I' is the *unknown* intermediate-state binding energy per electron. The simplest approximate intermediate state we can use when the photon energy is high enough is that where a single electron is removed from the ground state leaving unchanged the state of the other $N-1$ electrons and the excited electron is described using the Wentzel-Kramers-Brillouin approximation by a plane wave of wave vector equal to that of the photon involved in the matrix element. In this condition the matrix element of Eq. (4) can be treated in way similar to that appropriate to independent free electrons because the momentum conservation rule is again obtained at each interaction vertex. In other words, one has to reduce the complex *unknown* many-body propagator to that of the single excited electron. It is worthwhile to remember that, in any case, the incoming photon energy cannot be increased without any limit and the simplified theory one is developing has a validity only below the pair-creation threshold so that the radiative corrections can be safely neglected. Within the present approximation one has $E_I=(N-1)(\mu+\varepsilon_0)+\mu+\varepsilon_I$ and $E_0-E_I=(k_0+\mu+\varepsilon_0)-(\mu+\varepsilon_I)$, where ε_I is the binding energy of the excited electron.

Therefore the intermediate-state propagator becomes

$$G_I(E_0)=\sum_I |I\rangle\frac{1}{E_0-E_I}\langle I| = \sum_I |I\rangle\frac{(k_0+\mu+\varepsilon_0)+(\mu+\varepsilon_I)}{(k_0+\mu+\varepsilon_0)^2-(\mu+\varepsilon_I)^2}\langle I|.$$

Moreover, assuming that the photon wavelength is very short as compared to the atomic size, that is, the range of the ground-state wave function, one has that the matrix element of $\exp[-i\mathbf{k}_0\mathbf{r}]$ in the ground state is vanishingly small. Therefore on taking the various matrix elements of Eq. (4) a single electron is peaked up and the contribution to $K_{F0}^{(d)}$ will come mainly from those intermediate states with one electron with momentum $\mathbf{p}\cong\mathbf{k}_0$. In this condition the intermediate-state wave function will be an eigenfunction of the single-particle Dirac equation, which, because of the high momentum can be treated in the WKB approximation

$$[\alpha_i\cdot\mathbf{p}_i+V(\mathbf{r}_i)+\mu\beta_i]\Psi_I\cong(\mu+\varepsilon_I)\Psi_I \cong[V(\mathbf{r}_i)+(k_0^2+\mu^2)^{1/2}]\Psi_I,$$

where the $\mathbf{r}_i$ dependence of Ψ_I is described by a plane wave of momentum $\mathbf{p}_i\cong\mathbf{k}_0$. Because of this dependence the matrix elements of Eq. (4) are performed by integration over the coordinate of the excited electron only (apart from a trivial repetition because of the antisymmetry of the wave function). Moreover, in the present approximation, all the possible intermediate states have the excited electron with the *same* momentum and hence $(\mu+\varepsilon_I)^2$ does not depend on the intermediate state itself. Therefore one has

$$G_I(E_0)=\frac{\mu+k_0+\varepsilon_0+\alpha\mathbf{k}_0+\mu\beta}{(k_0+\mu+\varepsilon_0)^2-(\mu+\varepsilon_I)^2}\sum_I|I\rangle\langle I|. \quad (5)$$

Thanks to the form of this approximation, the sum over the intermediate states can be performed by closure. Inserting Eq. (5) into Eq. (4) and taking care of the fact that $\mathbf{x}_i=\mathbf{x}_j=\mathbf{r}$ one gets

$$K_{F0}^{(d)}=e^2\sum_i\int d\mathbf{r}_i\left\{\Psi_F^\dagger(\alpha_i\cdot\mathbf{e})\exp[i\mathbf{k}\cdot\mathbf{r}_i][k_0+\mu+(\alpha_i\cdot\mathbf{k}_0)+\mu\beta_i](\alpha_i\cdot\mathbf{e}_0)\exp[-i\mathbf{k}_0\cdot\mathbf{r}_i]\Psi_0\,\frac{1}{2\mu k_0[1-V(\mathbf{r}_i)/\mu+\varepsilon_0/\mu]}\right\}, \quad (6)$$

where in the numerator the terms of the order of ε_0 and $V(\mathbf{r}_i)$ have been neglected as compared to μ and k_0, while in the denominator the $k_0\to\infty$ limit has been taken. Further simplification can be obtained by considering that β anticommutes with α and μ $(1-\beta_i)\Psi_0$ is negligible as compared to $\mu\Psi_0$ because it is of the order of $\varepsilon_0\Psi_0$. In this way the final desired relationship is obtained:

$$K_{F0}^{(d)}=\sum_i e^2\int d\mathbf{r}_i\,\frac{\Psi_F^\dagger[(\alpha_i\cdot\mathbf{e})(\alpha_i\cdot\mathbf{e}_0)+(\alpha_i\cdot\mathbf{e})(\alpha_i\cdot\mathbf{k}_0)(\alpha_i\cdot\mathbf{e}_0)]\exp[-i\mathbf{Q}\cdot\mathbf{r}_i]\Psi_0}{2\mu[1-V(\mathbf{r}_i)/\mu+\varepsilon_0/\mu]}, \quad (7)$$

where $\mathbf{Q}=\mathbf{k}-\mathbf{k}_0$ is the momentum transfer. The contribution to the matrix element of the crossed term is readily obtained by making the substitution $\mathbf{k}_0\leftrightarrow\mathbf{k}$ and similar substitutions for the polarization vectors.

As one can see there is a one-to-one correspondence between Eq. (7) and Eq. (3) that is valid in the case of a single free electron. The most important difference is that there is no longer a momentum conservation rule between the initial and final states so that the elastic scattering occurs. Using Eq. (7) and the corresponding one for the crossed term, K_{F0} can be derived and hence the cross section of Eq. (1) can be obtained. The charge contribution arising from those terms that are proportional to $\mathbf{e}_0\cdot\mathbf{e}$ have been already discussed in Ref. 1. Considering that the magnetic cross section is quite small we shall approximate the energy denominator of Eq. (7) by 2μ. Then one gets for the magnetic contribution

$$\frac{d\sigma}{d\Omega}\cong\frac{{r_0}^2}{4k_0^2}\,|\{[i\mathbf{Q}\cdot(\mathbf{e}_0\times\mathbf{e})]\langle F|\Xi_{\mathbf{Q}}|0\rangle+[(\mathbf{e}_0\cdot\mathbf{e})(\mathbf{k}+\mathbf{k}_0)+(\mathbf{k}_0\cdot\mathbf{e})\mathbf{e}_0+(\mathbf{k}\cdot\mathbf{e}_0)\mathbf{e}]\langle F|\mathbf{j}_{\mathbf{Q}}|0\rangle\}|^2, \quad (8)$$

where r_0 is the classical electron radius, $\Xi_{\mathbf{Q}}$ is the Fourier transform of a modified electron density operator, and $\mathbf{j}_{\mathbf{Q}}$ is the Fourier transform of the current density operator. They are given by

$$\Xi_{\mathbf{Q}}=\tfrac{1}{3}\sum_i\alpha_i\sigma_i\exp[i\mathbf{Q}\cdot\mathbf{x}_i],\quad \mathbf{j}_{\mathbf{Q}}=\sum_i\alpha_i\exp[i\mathbf{Q}\cdot\mathbf{x}_i], \quad (9)$$

where σ_i is the 4×4 matrix obtained inserting the Pauli matrix on the principal diagonal. It is worthwhile to remember that $\mathbf{j}_{\mathbf{Q}}$ contains both the orbital and spin current, while $\Xi_{\mathbf{Q}}$ is proportional to the spin density.

The first important point one can observe in Eq. (8) is that the prefactor of the magnetic cross section *decreases* as a function of the incoming photon energy. This result is in apparent contrast with the common idea that the magnetic photon scattering cross section *increases* as the photon energy is increased. Actually this is also not the case in the nonrelativistic calculation of Refs. 5 and 6. In fact, as already mentioned, if one calculates the cross section for pure spin scattering at *constant* momentum transfer one obtains a nonincreasing result. In particular, depending on the photon polarization, one obtains a result having the same energy dependence as Eq. (8) or a constant trend.

It is also interesting to observe that a magnetic-scattering contribution can be thought to be present also in the usual spin-summed Klein and Nishina Compton cross section, which can be interpreted as a *paramagnetic scattering*. In fact all terms different from the Thomson term can be considered as *magnetic* in the sense that they are originated as a relativistic effect, like the electron spin, and are the result of the presence of three Dirac matrices in the matrix element. Therefore the *paramagnetic* term in the Klein and Nishina formula is

$$\frac{d\sigma}{d\Omega}=\frac{r_0^2}{4}\,(k/k_0+k_0/k-2). \quad (10)$$

We see that Eq. (10) has the same trend as Eq. (8) when the momentum transfer is taken as constant and the Compton formula is employed. This is particularly simple in the low-energy limit $(k_0\ll\mu)$ because one has

$$k/k_0 \cong 1 - 2k_0/\mu \sin^2(\theta/2) + [2k_0/\mu \sin^2(\theta/2)]^2,$$

$$k_0/k = 1 + 2k_0/\mu \sin^2(\theta/2),$$

where θ is the scattering angle. Then Eq. (10) becomes

$$\frac{d\sigma}{d\Omega} \cong \frac{r_0^2}{16} \frac{Q^4}{k_0^2 \mu^2}. \tag{11}$$

This equation is virtually exact in the appropriate limit and shows the same trend as Eq. (8). As already observed, the usual low-energy approximation of Refs. 4 and 5 does not provide a result exactly equal to that of Eq. (11). This is an indication that this approximation is accurate only to the order $1/c$, so that pure magnetic scattering, which is at the order $1/c^2$, is not correctly described by this approximation. This conclusion suggests that a correct low-energy approximation can be obtained using a more accurate interaction term along the line of Ref. 6, while the interference term between magnetic and charge scattering is correctly described by the $1/c$ approximation of Refs. 4 and 5.

It is useful to observe that in the case of a single bound electron in the nuclear Coulomb field there exists the calculation performed by Goldberger and Low,[13] which results in a general formula that is equal to Eq. (7) in the appropriate limit. In Ref. 13 a more accurate approximation for the Coulomb propagator is given so that the small imaginary part of the scattering amplitude is also obtained.

The results given in Eq. (8) could be treated numerically along the lines of Ref. 7; nonetheless it is interesting to derive an approximate analytical result from such an equation. First of all, one can readily see that there is only one term that is not decreasing as a function of the photon energy at constant momentum transfer, namely,

$$\frac{d\sigma}{d\Omega} = \frac{r_0^2}{4k_0^2} |(\mathbf{e}_0 \cdot \mathbf{e})(\mathbf{k} + \mathbf{k}_0) \langle 0 | \mathbf{j_Q} | 0 \rangle\}|^2, \tag{12}$$

where we focused on elastic scattering as such a case appears to be accessible to an experimental investigation.[14] Since $\mathbf{j_Q}$ is proportional to the α Dirac matrix, the matrix elements correspond to the superposition of the large and small components of the Dirac spinor. In an independent-particle approximation, if χ is the large component and φ is the small one, the following approximation at the order $1/c$ can be used:

$$\varphi \cong (\sigma \cdot \mathbf{p}) \chi / (2\mu),$$

χ and φ being two-row spinors. Using this approximation, after some manipulation one gets

$$\mathbf{j_Q} \cong (ihc/2\pi\mu)\ \mathbf{Q} f(\mathbf{Q}) S_\perp, \tag{13}$$

where $f(\mathbf{Q})$ is the atomic form factor and $S_\perp$ is the spin component perpendicular to the scattering plane. Inserting Eq. (13) into Eq. (12) one obtains a relationship identical to that deduced in Ref. 14 from the nonrelativistic approach of Refs. 4 and 5. Equation (13) is valid to the same $1/c$ order used in Refs. 4 and 5; however, in such a case the nonrelativistic approximation is formally used for the intermediate states, which are, on the contrary, in the extreme relativistic condition when the photon energy is increased. Moreover, the present description is adequate to treat the small binding effects that seem in any case quite small and of the same order as the difference between the ordinary and modified form factors.[1] Another consequence of Eq. (8) is the behavior of the orbital current contribution. The orbital current is rather high but tends to be parallel to $\mathbf{Q}$ and hence perpendicular to $\mathbf{k}_0$, therefore the corresponding cross section behaves like $Q^4/(k_0\mu)^2$, so that it decreases on increasing the incoming photon energy, at constant momentum transfer. Finally, one can observe that the present result is not confined to relativistic energies, but its validity condition is that the photon energy is much larger than the electron binding energy. The experimental findings of Ref. 14 are in good agreement with Eq. (13) up to fully relativistic energies, namely, $k_0 \cong \mu$.

As a final remark one can recall that, in the case of an independent-electron approximation and for central field, a fully numerical approach is possible,[7,15] at least in the case of purely elastic scattering. However, it does not appear obvious if it is possible to apply the numerical method to the case of extended systems and for quasielastic scattering, even in the case of an independent-electron approximation. To our best knowledge there is no attempt to make such a calculation. A direct numerical calculation allows for the determination of both real and imaginary parts of the scattering amplitude in a wide energy range but, of course, it does not allow for an understanding of the physical mechanism involved in the process. Another relativistic approach to the magnetic cross section is that of Ref. 16, where all the electrons are described by free independent-electron states, thus employing the older results of Tolhoek.[17] Also this procedure results in a similar contribution for the elastic spin-only scattering, but it is intrinsically related to an independent-particle approximation, while Eq. (8) contains many-body quantities.

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