Magnetic x-ray Compton scattering

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A theory of magnetic Compton scattering is formulated. The cross section is found to be insensitive to orbital effects, in agreement with recent experiments on transition-metal and rare-earth ferromagnets. When the transferred energy is sufficiently small to generate substantial corrections to the impulse approximation, the sensitivity to orbital moments of ordinary nonresonant magnetic scattering is recovered.

In x-ray Compton scattering from bound electrons,^{1,2} the size of the momentum transfer, $\mathbf{q} = \mathbf{k}_1 - \mathbf{k}_2$, is large with respect to the reciprocal of the (average) interparticle separation: $q|\mathbf{r}_1 - \mathbf{r}_2| \ge 1$; coherence effects are therefore negligible, and one-particle properties are probed. Also, the energy transfer, $\hbar \omega = \hbar(\omega_1 - \omega_2)$, greatly exceeds the outer-electron binding energy; the impulse approximation (IA) applies, yielding the double-differential cross section³

$$\frac{d^2\sigma}{d\Omega \ d\omega_2} = r_0^2 \frac{\omega_2}{\omega_1} |\boldsymbol{\epsilon}_2^* \cdot \boldsymbol{\epsilon}_1|^2 \int \frac{d\mathbf{p}}{(2\pi)^3} |\varphi(\mathbf{p})|^2 \\ \times \delta \left(\hbar \omega - \frac{(\hbar q)^2}{2m} - \frac{\hbar \mathbf{q} \cdot \mathbf{p}}{m} \right), \tag{1}$$

directly related to the Compton profile, that is, the projection of the electron momentum density along the scattering wave vector. Here, $\varphi(\mathbf{p})$ stands for the electron ground-state wave function in momentum space; $\boldsymbol{\epsilon}_1$ and $\boldsymbol{\epsilon}_2$ denote photon polarizations. For simplicity, the cross section has been written out for a single electron.

Equation (2) is obtained by considering the \mathbf{A}^2 term (the coupling to the electron diamagnetic current) in the interaction Hamiltonian, which is taken in the weakly relativistic limit⁴ and treated in the lowest order Born approximation. [The coupling is weak: $r_0 = e^2/(mc^2) = \alpha \chi_e$.] Corrections to the \mathbf{A}^2 scattering arise from the couplings of radiation to the electron paramagnetic current, the $\mathbf{p} \cdot \mathbf{A}$ term, and spin. The latter is known to result in a cross section proportional to the spin-polarized momentum distribution in a ferromagnetic solid, as predicted by Platzman and Tzoar⁵ and observed by Sakai and Ono.⁶

The paramagnetic term enters the scattering amplitude to second order in perturbation theory and, far from an absorption threshold, can be written as a transverse current operator^{7,8}

$$-i\gamma e^{i\mathbf{q}\cdot\mathbf{r}}\frac{i}{\hbar k_1^2}\left(\mathbf{k}_1-\mathbf{k}_2\frac{\omega_1}{\omega_2}\right)\times\mathbf{p}\cdot\boldsymbol{\epsilon}_2^*\times\boldsymbol{\epsilon}_1,$$

with $\mathbf{k}_1 - \mathbf{k}_2 \omega_1 / \omega_2 \simeq \mathbf{q}$, and $\gamma = (\hbar \omega_1) / (mc^2)$. As shown by Trammell,⁹

$$e^{i\mathbf{q}\cdot\mathbf{r}}\mathbf{q}\times\mathbf{p} = g(\mathbf{q}\cdot\mathbf{r})\mathbf{q}\times\mathbf{p} + g'(\mathbf{q}\cdot\mathbf{r})\mathbf{q}\times[\mathbf{r}(\mathbf{q}\cdot\mathbf{p}) + (\mathbf{L}\times\mathbf{q})],$$
(2)

with $g(x) = (e^{ix} - 1)/(ix)$, displaying the angular momentum contribution to the current.

Such a feature of expression (2) has led to the theoretical prediction^{10,11} that magnetic Compton scattering should provide information on both spin and orbital magnetization densities; including the possibility of a separation of the two contributions. The technique could thus complement magnetic x-ray diffraction^{12,13} and dichroism.¹⁴

Experiments aimed at verifying these theoretical ideas were reported by Collins et al.¹⁰ Data collected on metallic iron and cobalt were shown to agree well with the predictions. (Notice that in these systems the orbital contribution to the magnetic moment is small with respect to its spin counterpart.) However, significant departures from the expected values were observed in HoFe2, a ferrimagnet with dominant orbital magnetization. This ambiguous outcome was clarified by further experimental work showing that magnetic Compton scattering arises solely from the spin magnetization in the sample.^{15,16} These experiments detected the chargemagnetic interference (see below), by employing 45–50 keV ingoing circularly polarized photons, $\hbar \omega \simeq 5$ keV, and reversing the direction of the external (aligning) magnetic field (asymmetric ratio). To date no satisfactory theoretical explanation for the lack of orbital information in magnetic Compton scattering has been put forward.

For weakly bound electrons, those that determine the magnetic properties of the material, the IA yields accurate results.³ As will be shown, this approximation severely re-

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stricts the form of the electron current operator. As a result, the scattering response function is seen to contain no information on the orbital magnetization density. Corrections to the IA, which are very small for magnetic Compton scattering, can be calculated by time-dependent perturbation theory.¹⁷ The potential the electrons are moving in, absent in the IA, comes into play; its action results in a "bending" of electron motion, thus displaying the onset of an evolution towards the nonresonant x-ray scattering regime, where the orbital momentum is indeed observable.¹³

X-ray scattering. Neglecting the coupling of radiation to electron spin, the cross section for nonresonant x-ray scattering is given by^{7,8}

$$\frac{d^{2}\sigma}{d\Omega \ d\omega_{2}} = r_{0}^{2} \frac{\omega_{2}}{\omega_{1}} \sum_{f} \left| \langle f | \sum_{j} e^{i\mathbf{q}\cdot\mathbf{r}_{j}} \left[\boldsymbol{\epsilon}_{2}^{*} \cdot \boldsymbol{\epsilon}_{1} - i\gamma \left(\frac{i\mathbf{q} \times \mathbf{p}_{j}}{\hbar k_{1}^{2}} \right) \cdot \boldsymbol{\epsilon}_{2}^{*} \times \boldsymbol{\epsilon}_{1} \right] |g\rangle \right|^{2} \\ \times \delta(E_{g} - E_{f} + \hbar \omega), \qquad (3)$$

where *j* runs over all electrons in the target.

In momentum space, the intensity of interference scattering takes the form

$$\sum_{\mathbf{p}_{1}\mathbf{p}_{2},f} M_{g \to f}(\mathbf{p}_{1},\mathbf{p}_{2},\mathbf{q})[\mathbf{q} \times (\mathbf{p}_{1}+\mathbf{p}_{2}) \cdot (\boldsymbol{\eta}+\boldsymbol{\eta}^{*}) + \mathbf{q} \times (\mathbf{p}_{1}-\mathbf{p}_{2}) \cdot (\boldsymbol{\eta}-\boldsymbol{\eta}^{*})], \quad (4)$$

where

$$M_{g \to f}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{q}) = \langle g | \mathbf{p}_1 - \hbar \mathbf{q} \rangle \langle \mathbf{p}_1 | f \rangle \langle f | \mathbf{p}_2 \rangle \langle \mathbf{p}_2 - \hbar \mathbf{q} | g \rangle$$
$$\times \delta(E_g - E_f + \hbar \omega), \tag{5}$$

with $\eta = (\epsilon_2 \times \epsilon_1^*)(\epsilon_2^* \cdot \epsilon_1)$. (To simplify the notation, the expression has been written for a single electron, omitting unessential factors.) General features of the scattering process upon reversal of an external magnetic field can be determined by analyzing the behavior of Eq. (4).

When an external magnetic field is reversed, the eigenstates of the system are turned into their complex conjugates with the same eigenvalues. (As we are dealing with spindiagonal matrix elements, spin labels are omitted throughout this work.) Then the property: $\langle \psi^* | \mathbf{p} \rangle = \langle -\mathbf{p} | \psi \rangle$ results in $M_{g^* \to f^*}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{q}) = M_{g \to f}(-\mathbf{p}_2, -\mathbf{p}_1, -\mathbf{q})$. In a system with *inversion symmetry*, the ground state has and the final state can be taken to have, *definite parity*. Using the relation $\langle -\mathbf{p} | \psi \rangle = \pm \langle \mathbf{p} | \psi \rangle$, we have $M_{g \to f}(-\mathbf{p}_2, -\mathbf{p}_1, -\mathbf{q}) = M_{g \to f}(-\mathbf{p}_2, -\mathbf{p}_1, -\mathbf{q})$

$$M_{g^* \to f^*}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{q}) = M_{g \to f}(\mathbf{p}_2, \mathbf{p}_1, \mathbf{q}).$$
(6)

Notice that in Eq. (4) the terms proportional to $\eta + \eta^*$ and $\eta - \eta^*$ contain factors which are, respectively, symmetric and antisymmetric under exchange of \mathbf{p}_1 and \mathbf{p}_2 . From Eq. (6), we immediately conclude that, if parity is a good quantum number, the $(\eta + \eta^*)$ term remains unchanged, whereas the $(\eta - \eta^*)$ term changes sign upon reversal of an external magnetic field.

The symmetric part (present for arbitrary polarization) of Eq. (4) gives a nonvanishing asymmetric ratio only when

 $|g\rangle$ does not have a definite parity; this is the case of noncentrosymmetric structures. The antisymmetric part (circular polarization) yields a nonvanishing asymmetric ratio, irrespectively of the properties of the ground state under parity transformations; if $|g\rangle$ has definite parity, the matrix element changes sign upon field reversal and we have a genuine magnetic effect (orbital magnetism). The foregoing derivation formalizes and adds to Blume's remarks⁷ on the symmetry of Eq. (3).

Compton scattering. Magnetic Compton scattering is best analyzed by rewriting the interference cross section, Eq. (3), after discarding the terms proportional to $\exp[i\mathbf{q} \cdot (\mathbf{r}_j - \mathbf{r}_{j'})], j \neq j'$, as their contribution can be neglected at large momentum transfers.¹⁸ We have

$$\frac{d^2\sigma}{d\Omega \ d\omega_2}\Big|_{\text{int}} = r_0^2 \frac{\omega_2}{\omega_1} \frac{\gamma}{\hbar k_1^2} \frac{1}{2\pi\hbar} \sum_j \int dt e^{i(\omega - \hbar q^2/2m)t} F_j(\mathbf{q}, t),$$
(7)

with the response function given by

$$F_{j}(\mathbf{q},t) = \langle g | \boldsymbol{\eta} \cdot \mathbf{q} \times \mathbf{p}_{j} e^{-i(\hat{H}/\hbar + \mathbf{q} \cdot \mathbf{p}_{j}/m)t} + e^{-i(\hat{H}/\hbar + \mathbf{q} \cdot \mathbf{p}_{j}/m)t} \mathbf{q}$$
$$\times \mathbf{p}_{j} \cdot \boldsymbol{\eta}^{*} | g \rangle$$
(8)

 $(\mathbf{q} \times \mathbf{p} \text{ commutes with } e^{i\mathbf{q}\cdot\mathbf{r}})$. Here, $\tilde{H} = H - E_g$, with H the electron Hamiltonian: $H = \sum_j p_j^2 / 2m + V$, where V includes one- and two-body potentials, neglecting the spin-orbit interaction.

In the Compton regime, the energy transfer is much larger than any energy associated with the ground-state magnetic electrons; the "collision time" ($\sim \omega^{-1}$) is, therefore, very short. A suitable time-dependent perturbation expansion of the cross section is obtained from expression (8) when combined with the identity

$$e^{-i(\tilde{H}/\hbar + \mathbf{q} \cdot \mathbf{p}_j/m)t} = \left[1 - \frac{i}{\hbar} \int_0^t e^{-i(\tilde{H}/\hbar + \mathbf{q} \cdot \mathbf{p}_j/m)t'} \tilde{H} e^{i\mathbf{q} \cdot \mathbf{p}_j t'/m} dt'\right] \times e^{-i\mathbf{q} \cdot \mathbf{p}_j t/m}.$$
(9)

To first order, the response function is then given by

$$F_{j}(\mathbf{q},t) = F_{j}^{(0)}(\mathbf{q},t) + F_{j}^{(1)}(\mathbf{q},t), \qquad (10)$$

with

$$F_{j}^{(0)}(\mathbf{q},t) = \langle g | e^{-i\mathbf{q} \cdot \mathbf{p}_{j}t/m} \mathbf{q} \times \mathbf{p}_{j} | g \rangle \cdot (\boldsymbol{\eta} + \boldsymbol{\eta}^{*}), \qquad (11)$$

and

$$F_{j}^{(1)}(\mathbf{q},t) = -\frac{i}{\hbar} \int_{0}^{t} dt' [\langle g | \boldsymbol{\eta} \cdot \mathbf{q} \times \mathbf{p}_{j} e^{-i\mathbf{q} \cdot \mathbf{p}_{j}t'/m} \tilde{H} e^{i\mathbf{q} \cdot \mathbf{p}_{j}(t'-t)/m} + e^{-i\mathbf{q} \cdot \mathbf{p}_{j}t'/m} \tilde{H} e^{i\mathbf{q} \cdot \mathbf{p}_{j}(t'-t)/m} \mathbf{q} \times \mathbf{p}_{j} \cdot \boldsymbol{\eta}^{*} | g \rangle].$$
(12)

Centrosymmetric structures. The zeroth-order term, $F_j^{(0)}(\mathbf{q},t)$, readily yields the scattering intensity in the IA; we find (again, unessential factors are omitted)

$$\int d\mathbf{p}\langle g|\mathbf{p}\rangle\langle \mathbf{p}|g\rangle \mathbf{q}\times\mathbf{p}\cdot(\boldsymbol{\eta}+\boldsymbol{\eta}^*)\delta\left(\hbar\omega-\frac{(\hbar q)^2}{2m}-\frac{\hbar\mathbf{q}\cdot\mathbf{p}}{m}\right).$$
(13)

When $|g\rangle$ is an eigenstate of the parity operator (we are seeking a genuine magnetic effect), then the reversal of an external magnetic field results in

$$\langle g^* | \mathbf{p} \rangle \langle \mathbf{p} | g^* \rangle = \langle g | \mathbf{p} \rangle \langle \mathbf{p} | g \rangle.$$
 (14)

The asymmetric ratio vanishes in this case; *no orbital magnetism is detectable in the IA, irrespectively of the experimental conditions*, i.e., choice of photon polarization and scattering geometry. The physical content of this result will now be illustrated by analyzing the structure of the orbital current in the IA regime.

Consider the general response function for interference scattering at large momentum transfers

$$e^{-i\hbar q^2 t/2m} F_j(\mathbf{q},t) = \boldsymbol{\eta} \cdot \langle g | \mathbf{q} \times \mathbf{J}_j(\mathbf{q},t) \rho_j^{\dagger}(\mathbf{q}) | g \rangle + \boldsymbol{\eta}^* \cdot \langle g | \rho_j(\mathbf{q},t) \mathbf{q} \times \mathbf{J}_j^{\dagger}(\mathbf{q}) | g \rangle, \quad (15)$$

with $\mathbf{J}_{j}(\mathbf{q},t) = e^{iHt/\hbar} \mathbf{J}_{j}(\mathbf{q}) e^{-iHt/\hbar}$, and similarly for ρ_{j} . Transverse-current and charge densities are given by

$$\mathbf{q} \times \mathbf{J}_{i}(\mathbf{q}) = e^{-i\mathbf{q} \cdot \mathbf{r}_{j}} \mathbf{q} \times \mathbf{p}_{i}$$

and

$$\rho_i(\mathbf{q}) = e^{-i\mathbf{q}\cdot\mathbf{r}_j},$$

respectively.

The nature of the current operator is determined by its time evolution. As observed, the interaction time is very short in the Compton regime; the potential terms, *V*, do not play any role and the time dependence of operators is controlled by the kinetic part of the Hamiltonian: $H \rightarrow H_0 = \sum_i p_i^2/2m$. We have

$$e^{iH_0t/\hbar}\mathbf{q}\times\mathbf{J}_i(\mathbf{q})e^{-iH_0t/\hbar}=e^{-i\mathbf{q}\cdot\mathbf{r}_j(t)}\mathbf{q}\times\mathbf{p}_i\,,\qquad(16)$$

with

$$\mathbf{r}_j(t) = \mathbf{r}_j(0) + \frac{\mathbf{p}_j t}{m};$$

that is, the current generated by freely moving electrons. It contains no orbital magnetism. The angular momentum part of Eq. (2) is lost; this can be checked by substituting Eq. (16) into (15); \mathbf{r}_j drops out of the expressions and we recover the matrix element of Eq. (11)

$$e^{-i\hbar q^2 t/2m} \langle g | e^{-i\mathbf{p}_j \cdot \mathbf{q} t/m} \mathbf{q} \times \mathbf{p}_j | g \rangle \cdot (\mathbf{\eta} + \mathbf{\eta}^*),$$

which is invariant upon field reversal when $|g\rangle$ has definite parity.¹⁹

The first order term, as from Eq. (12), is conveniently split into symmetric and antisymmetric components

$$F_{j}^{(1)}(\mathbf{q},t) = \sum_{\mathbf{p}^{(1)}\mathbf{p}^{(2)}} \langle g | \mathbf{p}^{(1)} \rangle \langle \mathbf{p}^{(1)} | V | \mathbf{p}^{(2)} \rangle \langle \mathbf{p}^{(2)} | g \rangle$$
$$\times [(\boldsymbol{\eta} + \boldsymbol{\eta}^{*}) \cdot \mathbf{f}_{s}(\mathbf{p}_{j}^{(1)}, \mathbf{p}_{j}^{(2)}, \mathbf{q}, t)$$
$$+ (\boldsymbol{\eta} - \boldsymbol{\eta}^{*}) \cdot \mathbf{f}_{a}(\mathbf{p}_{j}^{(1)}, \mathbf{p}_{j}^{(2)}, \mathbf{q}, t)], \qquad (17)$$

$$\mathbf{f}_{a}(\mathbf{p}_{1},\mathbf{p}_{2},\mathbf{q},t) = \mathbf{q} \times (\mathbf{p}_{1}-\mathbf{p}_{2}) \frac{e^{-i\mathbf{q}\cdot\mathbf{p}_{1}t/m} - e^{-i\mathbf{q}\cdot\mathbf{p}_{2}t/m}}{2\hbar\mathbf{q}\cdot(\mathbf{p}_{1}-\mathbf{p}_{2})/m},$$
(18)

with $\mathbf{f}_a(\mathbf{p}_1, \mathbf{p}_2, \mathbf{q}, t) = -\mathbf{f}_a(\mathbf{p}_2, \mathbf{p}_1, \mathbf{q}, t)$. (The explicit form of the symmetric part is not required in the following.) $|\mathbf{p}^{(1)}\rangle = |\mathbf{p}_1^{(1)} \cdots \mathbf{p}_N^{(1)}\rangle$ denotes an *N*-particle plane-wave state.

Consider the case of a ground state with definite parity. By applying the rules previously given, it is readily shown that the antisymmetric part of Eq. (17) changes sign upon field reversal $(g \rightarrow g^*)$, thus describing a genuine magnetic effect. The result is interpreted as follows. Adding $F_i^{(1)}(\mathbf{q},t)$ to the cross section amounts to considering the probability that the electron is scattered by the potential Vwithin a lapse of time ω^{-1} . As Compton scattering is very fast, the probability is very small (corrections are of the order: $\varepsilon_{el}/\varepsilon_a \sim 10^{-3}$, that is, the ratio between a magneticelectron characteristic energy and the recoil energy, as shown below) and the effect not observable in practice. The antisymmetric part (circular polarization) of Eq. (17) has, however, an important meaning: the function \mathbf{f}_a is nonzero only for $\mathbf{p}_1 \neq \mathbf{p}_2$, thus displaying the "bending" of electron motion. Upon reducing the photon energy transfer, higher order terms become significant in Eq. (9); when $\hbar \omega$ is of the order of the magnetic electron ground-state characteristic energies, the complete time-evolution operator is required to describe its propagation; bound electron motion and full orbital magnetization are recovered. This is the case of x-ray magnetic scattering.

Noncentrosymmetric structures. Compton scattering in noncentrosymmetric structures, as described by $F_j^{(0)}(\mathbf{q},t)$ for a parity-broken ground state, remains to be discussed. Equation (14) is no longer valid in this case; the asymmetric ratio (for arbitrary polarization) reads

$$\frac{d^{2}\sigma}{d\Omega \ d\omega_{2}} \Big|_{\mathrm{IA}}^{\Delta} = r_{0}^{2} \frac{\omega_{2}}{\omega_{1}} \frac{\gamma}{\hbar k_{1}^{2}} \int d\mathbf{p}^{(1)} [|\langle \mathbf{p}^{(1)}|g \rangle|^{2} -|\langle -\mathbf{p}^{(1)}|g \rangle|^{2}] \sum_{j} \mathbf{q} \times \mathbf{p}_{j}^{(1)} \cdot (\boldsymbol{\eta} + \boldsymbol{\eta}^{*}) \times \delta \Big(\hbar \omega - \frac{(\hbar q)^{2}}{2m} - \frac{\hbar \mathbf{q} \cdot \mathbf{p}_{j}^{(1)}}{m} \Big).$$
(19)

The quantity $|\langle \mathbf{p}^{(1)}|g \rangle|^2$ is not invariant upon reversal of orbital motion when parity is broken, and the cross section provides information on the asymmetry of the electron wave function (projected along the scattering wave vector) in momentum space.

Moment analysis. The accuracy of the IA in centrosymmetric structures can be assessed by evaluating the exact moments M_n of the dynamic structure factor for interference scattering; they are given by

$$M_n(\mathbf{q}) = i^n \frac{d^n}{dt^n} e^{-i\hbar q^2 t/2m} \sum_j F_j(\mathbf{q},t) \bigg|_{t=0}$$

Assuming inversion symmetry to hold, we find that $M_0=0$; furthermore, we have

where

$$M_1 = m^{-1}(\boldsymbol{\eta} + \boldsymbol{\eta}^*) \cdot \langle g | \sum_j \mathbf{q} \times \mathbf{p}_j(\mathbf{q} \cdot \mathbf{p}_j) | g \rangle, \qquad (20)$$

and

$$M_{2} = (2/\hbar)\varepsilon_{q}M_{1} + \hbar m^{-1} \boldsymbol{\eta} \cdot \langle g | \sum_{j} \mathbf{q} \times \nabla_{j}(\mathbf{q} \cdot \nabla_{j}V) | g \rangle$$
$$+ im^{-1}(\boldsymbol{\eta} - \boldsymbol{\eta}^{*}) \cdot \langle g | \sum_{j} (\mathbf{q} \cdot \nabla_{j}V) \mathbf{q} \times \mathbf{p}_{j} | g \rangle, \quad (21)$$

with $\varepsilon_q = (\hbar q)^2/2m$, the recoil energy.²⁰ For interference scattering, M_2 is the lowest moment containing the potential *V*. (In the case of pure charge scattering, the \mathbf{A}^2 term alone, *V* makes its first appearance in M_3 .³) M_1 and the *V*-independent terms of M_n , with $n \ge 2$, are correctly reproduced by the IA, that is, by $F_j^{(0)}$. It is straightforward to verify that first-order corrections to the IA, as given by $F_i^{(1)}$, correctly reproduce the *V*-dependent part of M_2 .

An order of magnitude estimate of the size of the corrections to the IA, responsible for a nonvanishing asymmetric

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ratio in centrosymmetric systems, can be inferred from M_2 , taken in the large-q limit. We find

$$\frac{M_2 - M_2^{\rm IA}}{M_2} \sim \frac{1}{\varepsilon_q} \frac{\hbar^2 \langle \hat{\mathbf{q}} \times \nabla(\hat{\mathbf{q}} \cdot \nabla V) \rangle}{\langle \hat{\mathbf{q}} \times \mathbf{p}(\hat{\mathbf{q}} \cdot \mathbf{p}) \rangle} \sim \frac{\varepsilon_{\rm el}}{\varepsilon_q}, \qquad (22)$$

with $\varepsilon_{\rm el}$ a magnetic-electron characteristic energy. The same result can be obtained by evaluating the corresponding quantity for higher momenta. [Notice that the correction to the IA for pure charge scattering are found to be of the order: $(\varepsilon_{\rm el}/\varepsilon_q)^{2.3}$]

To summarize, this work has provided an explanation for the absence of an orbital contribution to magnetic Compton scattering. Within the IA, very accurate in describing the process, orbital magnetic effects are seen to vanish. Corrections to the IA cross section bear a reminiscence of bound electron orbital motion, however, as shown by the moment analysis, these contributions are very small and not observable in practice.

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- ²⁰These averages are nonzero for a magnetic or a low-symmetry crystal.