X-Ray Resonance Exchange Scattering

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Large resonant magnetization-sensitive x-ray scattering is predicted to occur in the vicinity of $L_{\rm II}$, $L_{\rm III}$, and $M_{\rm II}$–$M_{\rm V}$ absorption edges in the rare-earth and actinide elements, and at the K and L edges in the transition elements. These "magnetic" resonances result from electric multipole transitions, with the sensitivity to the magnetization arising from exchange. For some transitions, the magnetic scattering will be comparable to the charge scattering. The general features of the observed $L_{\rm III}$ resonance in Ho are discussed.

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In the course of investigating the magnetic spiral structure of a holmium crystal using the x-ray magnetic scattering of synchrotron radiation, Gibbs *et al.*[1] observed a large resonant enhancement (by a factor of 50) in the magnetic satellite intensities when the energy of the incident x rays was tuned through the $L_{\rm III}$ absorption edge. Second-, third-, and fourth-harmonic satellites were also observed at resonance. A complex polarization dependence was found, with the resonance peaks for the σ-σ and σ-π components of the magnetic scattering being separated by about 6 eV for the first two harmonics, and occurring at the same resonance energy for the third and fourth harmonics.

This behavior (resonant increase, additional harmonics, polarization dependence) can be understood on the basis of electric quadrupole ($E2$) transitions to $4f$ levels and electric dipole ($E1$) transitions to $5d$ levels. It is noteworthy that this "magnetic" scattering results from *electric* multipole transitions. This is due to the exclusion principle allowing only transitions to unoccupied orbitals, resulting in an "exchange interaction" which is sensitive to the magnetization of the f and d bands.

To get a strong resonant enhancement, the scattering must involve a low-order electric multipole transition ($E1$ or $E2$) between a core level and either an unfilled atomic shell, or a narrow band.[2] In the latter case, the atomiclike nature of the transition is increased because the core hole gives an additional binding of the excited level.

For the rare earths, enhanced magnetic resonance scattering will occur at the $L_{\rm II}$, $L_{\rm III}$, $M_{\rm II}$, and $M_{\rm III}$ absorption edges, involving the $E2$ transitions to the tightly bound $4f$ shell, and the $E1$ transitions to the $5d$ band. Although the latter transition is $E1$, the strength of the magnetic scattering depends on the induced polarization and exchange splitting of the band, resulting in a contribution of comparable magnitude to the $E2$ transition to the $4f$ shell. At the $M_{\rm IV}$ and $M_{\rm V}$ edges, the very strong $E1$ transition to the $4f$ shell will give a resonant magnetic scattering amplitude on the order of $100r_0$! The resonances at the K and $L_{\rm I}$ edges will be relatively weak, involving $E3/M2$ transitions to the $4f$ shell, and $E2$ transitions to the $5d$ band. The $L_{\rm II}$ and $L_{\rm III}$ resonances lie in the 1–2-Å region, well suited for diffraction studies of magnetism in crystals. The strong $E1$ resonances at the $M_{\rm IV}$ and $M_{\rm V}$ edges lie in the 5–10-Å region, but can still be used for diffraction studies of the long-range antiferromagnetic spirals, and for grazing incidence studies of surface magnetism, and they will give rise to very strong magneto-optical effects in reflection and transmission in magnetic samples.

Similar magnetization-sensitive electric multipole resonances should be useful for the study of the magnetic properties of the transition elements and the actinide series. These large resonant enhancements should also be important for the study of two-dimensional magnetic ordering, with possible applications to high-T_c superconductors.

The coherent elastic-scattering amplitude for nonresonant magnetic x-ray scattering from a magnetic ion is given by[3–7]

$$f^{(\rm mag)} = ir_0(\hbar\omega/mc^2)f_{\rm D}[\tfrac{1}{2}\mathbf{L}(K)\cdot\mathbf{A}+\mathbf{S}(K)\cdot\mathbf{B}],$$

where $\mathbf{L}(K)$ and $\mathbf{S}(K)$ are the atomic orbital and spin magnetization densities, $\mathbf{A}$ and $\mathbf{B}$ are polarization vectors determined by $\mathbf{k}_0,\mathbf{e}_0,\mathbf{k}_f,\mathbf{e}_f$, and $f_{\rm D}$ is the Debye-Waller factor. The x-ray magnetic scattering is considerably weaker than the charge scattering, with magnitudes typically $\approx 0.01r_0$. The total coherent elastic-scattering amplitude is $f\approx f_0+f'+if''+f^{(\rm mag)}$, where $f_0\propto -Zr_0$ is the usual Thomson contribution, and $f'+if''$ is the contribution from dispersive and absorptive processes. The resonant scattering processes we consider below contribute to $f'+if''$.

For an electric 2^L-pole resonance (EL) in a magnetic ion, the contribution to the coherent scattering amplitude is given by[2,8,9]

$$f_{EL}^{(\mathrm{xres})}(\mathbf{k}_f\mathbf{e}_f;\mathbf{k}_0\mathbf{e}_0)=4\pi\lambda\!\!\!^{-} f_{\mathrm{D}}\sum_{M=-L}^{L}[\mathbf{e}_f^*\cdot\mathbf{Y}_{LM}^{(e)}(\hat{\mathbf{k}}_f)\mathbf{Y}_{LM}^{(e)*}(\hat{\mathbf{k}}_0)\cdot\mathbf{e}_0]F_{LM}^{(e)}(\omega), \tag{1}$$

where

$$F_{LM}^{(e)}(\omega)=\sum_{\alpha,\eta}\left[\frac{p_\alpha p_\alpha(\eta)\Gamma_x(\alpha M\eta;EL)/\Gamma(\eta)}{x(\alpha,\eta)-i}\right].$$

$|\alpha)$ is the initial ground state of the ion, and $|\eta)=|\alpha(h\mu_h)^{-1}(n\mu_n)^{+1})$ is the excited state with an electron excited to the level $(n\mu_n)$ leaving a hole in the core level $(h\mu_h)$, where μ_n and μ_h are the appropriate spin and angular momentum indices. The excited states $(n\mu_n)$ are more tightly bound than the corresponding states in the unexcited ion due to the potential of the core hole. p_α gives the statistical probabilities for the various possible initial state $|\alpha)$, and $p_\alpha(\eta)$ gives the probability that the $(n\mu_n)$ state is unoccupied in $|\alpha)$. $p_\alpha(\eta)$ is determined by the overlap integrals of the "old" orbitals which are occupied in the initial state $|\alpha)$ with the orbital $(n\mu_n)$ which is a "new" level in the presence of the $(h\mu_h)$ core hole, a familiar procedure in shakeoff calculations. Γ_x is given to lowest order in kr by[9]

$$\Gamma_x(\alpha M\eta;EL)=8\pi\left(\frac{e^2}{\lambda\!\!\!^{-}}\right)\left(\frac{L+1}{L}\right)\left|(\alpha|\sum_i j_L(kr_i)Y_{LM}(\hat{\mathbf{r}}_i)|\eta)\right|^2, \tag{2}$$

where j_L is the spherical bessel function of order L. Summed over M, Γ_x gives the *partial* width for *EL radiative decay* from $|\eta)\rightarrow|\alpha)$. $\Gamma(\eta)$ is the *total width* for the excited state $|\eta)$, which is determined by *all* radiative (from any shell) and nonradiative (Auger, Coster-Kronig) deexcitations of $|\eta)$. $\Gamma(\eta)$ is typically ≈ 1–10 eV, so that the scattering will be fast $\approx 10^{-16}$ s, accounting for the presence of the Debye-Waller factor in Eq. (1).[8] In the resonance denominator, $x(\alpha,\eta)=[\epsilon(\eta)-\epsilon(\alpha)-\hbar\omega]/[\Gamma(\eta)/2]$ gives the deviation from resonance in units of $\Gamma(\eta)/2$. The polarization dependence is determined by the vector spherical harmonics $\mathbf{Y}_{LM}^{(e)}$ for an EL transition,[10] and the relevant factors which appear in Eq. (2) can be expressed as products of the components of $\mathbf{k}$ and $\mathbf{e}$,

$$\mathbf{e}\cdot\mathbf{Y}_{LM}^{(e)}(\hat{\mathbf{k}})=\left[\frac{4\pi(2L+1)}{3(L+1)}\right]^{1/2}\sum_{\mu=-1}^{1}C(1\,L-1\,L;\mu\,M-\mu)Y_{L-1,M-\mu}(\hat{\mathbf{k}})Y_{1\mu}(\hat{\mathbf{e}}).$$

For the *electric dipole transitions* $(E1)$, we have

$$[\mathbf{e}_f^*\cdot\mathbf{Y}_{1\pm1}^{(e)}(\hat{\mathbf{k}}_f)\mathbf{Y}_{1\pm1}^{(e)*}(\hat{\mathbf{k}}_0\cdot\mathbf{e}_0]=\left[\frac{3}{16\pi}\right][\mathbf{e}_f^*\cdot\mathbf{e}_0\mp i(\mathbf{e}_f^*\times\mathbf{e}_0)\cdot\hat{\mathbf{z}}_J-(\mathbf{e}_f^*\cdot\hat{\mathbf{z}}_J)(\mathbf{e}_0\cdot\hat{\mathbf{z}}_J)],$$

$$[\mathbf{e}_f^*\cdot\mathbf{Y}_{10}^{(e)}(\hat{\mathbf{k}}_f)\mathbf{Y}_{10}^{(e)*}(\hat{\mathbf{k}}_0)\cdot\mathbf{e}_0]=\left[\frac{3}{8\pi}\right][(\mathbf{e}_f^*\cdot\hat{\mathbf{z}}_J)(\mathbf{e}_0\cdot\hat{\mathbf{z}}_J)],$$

giving the scattering amplitude

$$f_{E1}^{(\mathrm{xres})}=\tfrac{3}{4}\lambda\!\!\!^{-}\{\mathbf{e}_f^*\cdot\mathbf{e}_0[F_{11}^{(e)}+F_{1-1}^{(e)}]-i(\mathbf{e}_f^*\times\mathbf{e}_0)\cdot\hat{\mathbf{z}}_J[F_{11}^{(e)}-F_{1-1}^{(e)}]+(\mathbf{e}_f^*\cdot\hat{\mathbf{z}}_J)(\mathbf{e}_0\cdot\hat{\mathbf{z}}_J)[2F_{10}^{(e)}-F_{11}^{(e)}-F_{1-1}^{(e)}]\}, \tag{3}$$

where $\mathbf{z}_J$ is the direction of the quantization axis defined by the local moment of the ion. Thus there are only three distinct polarization responses for $E1$ scattering[9]: The first term, $\mathbf{e}_f^*\cdot\mathbf{e}_0$, is independent of the direction of the magnetic moment. The second term, $-i(\mathbf{e}_f^*\times\mathbf{e}_0)\cdot\mathbf{z}_J$, depends linearly on the direction of the magnetic moment, and will give first-harmonic satellites in an antiferromagnet. If we use the linear σ,π polarizations as basis (which are perpendicular and parallel to the $\mathbf{k}_0$-$\mathbf{k}_f$ scattering plane, respectively), incident σ_0 scatters only to π_f, while incident π_0 scatters to both σ_f and π_f. The third contribution, $(\mathbf{e}_f^*\cdot\mathbf{z}_J)(\mathbf{e}_0\cdot\mathbf{z}_J)$, depends quadratically on the moment direction, and will give second-harmonic satellites in a spiral antiferromagnet (and also a contribution to the zeroth harmonic). This term scatters either σ_0 or π_0 to both σ_f and π_f. The appearance of first- and second-harmonic magnetic satellites is a characteristic signature of a dipole transition ($E1$ or $M1$).

Two simplified examples illustrate the main ideas: First, if the ground state of the ion has a single hole in the $4f$ shell, then by Hund's-rule, in the fully aligned case, the empty orbital is $m_l=-3$, $m_s=-\frac{1}{2}$, which is also a state of good $j=\frac{7}{2}$, $m_j=-\frac{7}{2}$. Then in Eq. (1), $p_\alpha=1$ for the Hund's-rule ground state $|\alpha)$, and $p_\alpha(m_l,m_s)=1$ for $(m_l,m_s)=(-3,\downarrow)$, and zero otherwise. (Here we ignore the question of overlap integrals discussed below, but this should be less important for the highly localized $4f$ orbitals.) Then at the M_{V} edge, only $M=-1$ is allowed, the transition being $|3d_{5/2},m_j=-\frac{5}{2})\leftrightarrow|4f_{7/2},m_j=-\frac{7}{2})$. This corresponds to a circularly polarized electric dipole oscillator, with left-hand circulation about $+\mathbf{z}_J$. As a consequence, $F_{11}=F_{10}=0$ in Eq. (3), and $F_{1-1}=[\Gamma_x(\alpha,-1,\eta;E1)/\Gamma(\eta)]/[x-i]$ as given by Eqs. (1) and (2). All three polarization terms then contribute with equal amplitude to $f_{E1}^{(\mathrm{xres})}$ in Eq. (3), and so the magnetic scattering will be as large as the charge scattering. Fluorescence yield calculations[11] indicate that $(\Gamma_x/\Gamma)\approx 10^{-2}$ for the rare earths, so that magnetic resonance scattering amplitudes $\approx 100r_0$ should be possible for these soft x-ray resonances, giving very strong magneto-optical effects in both reflection and transmission.

As a second example, consider the L_{III}-edge resonance $2p_{3/2}\leftrightarrow 5d$, treating the "impurity" $5d$ states in the presence of the core hole as atomic levels $|5d,m_l,m_s)$, with an exchange splitting Δ between the (lower energy) $|5d,m_l,\uparrow)$ orbitals and the $|5d,m_l,\downarrow)$ orbitals induced by the $4f$ moment. For such a system, the probability $p_\alpha(5d,m_l,m_s)$ that the impurity orbital $|5d,m_l,m_s)$ is empty would be determined by the overlap integrals of the occupied orbitals in the ground state $|\alpha)$ with the impurity orbital. For simplicity, we will assume that $p_\alpha(5d,m_l,m_s)\equiv p(m_s)$ independent of m_l. The partial radiative width is then

$$\Gamma_x((2p_{3/2}m_j)M(5d,m_l,m_s))=C^2(1,\tfrac{1}{2},\tfrac{3}{2};m_l-M,m_s,m_j)C^2(1,1,2;m_l-M,m_s,m_l)\,|\chi|^2,$$

where $|\chi|^2=\frac{24}{5}ke^2|(2p_{3/2}||kr||5d)|^2$, and the scattering amplitude becomes

$$f_{EL}^{(\mathrm{xres})}=F[\mathbf{e}_f^*\cdot\mathbf{e}_0 n_h+i(\mathbf{e}_f^*\times\mathbf{e}_0)\cdot\mathbf{z}_J P/4], \qquad (4)$$

where $P=[n_e(\uparrow)-(\Delta/\Gamma)n_h$, $n_e(\uparrow)=5[p(\downarrow)-p(\uparrow)]$ is the net number of spin-up *electrons* in the $5d$ band, $n_h=5[p(\downarrow)+p(\uparrow)]$ is the number of *holes* in the $5d$ band, and $F=\lambda|\chi|^2]/3\Gamma(x-i)$. We have assumed that $\Delta\ll\Gamma$, giving an unresolved resonance doublet, and x is the deviation from the central frequency. There is no quadratic magnetic contribution $(\mathbf{e}_f^*\cdot\mathbf{z}_J)(\mathbf{e}_0\cdot\mathbf{z}_J)$ because of the assumed m_l independence of $p_\alpha(5d,m_l,m_s)$. The linear magnetic contribution is seen to arise from both the spin polarization of a partially occupied band, and from the exchange splitting of the empty states, with the sign and magnitude of the "polarization factor" P depending on the relative magnitude of the two contributions. As an order-of-magnitude estimate for Ho, taking $\Delta\approx 0.3$ eV, $n_e(\uparrow)\approx 0.3$, $Fn_h\approx 30r_0/(x-i)$, $\Gamma\approx 10$ eV, and $n_h\approx 8$, gives $P\approx +0.07$, and a linear magnetic scattering contribution of $\approx +0.06r_0 i(\mathbf{e}_f^*\times\mathbf{e}_0\cdot\mathbf{z}_J)/(x-i)$.

For electric quadrupole transitions ($E2$), $f_{E2}^{(\mathrm{xres})}$ will contain thirteen distinct terms—order (0):

$$(\mathbf{k}_f\cdot\mathbf{k}_0)(\mathbf{e}_f^*\cdot\mathbf{e}_0),$$

order (1):

$$i(\mathbf{k}_f\cdot\mathbf{k}_0)(\mathbf{e}_f^*\times\mathbf{e}_0)\cdot\mathbf{z}_J+[\mathbf{k}_f\leftrightarrow\mathbf{e}_f,\mathbf{k}_0\leftrightarrow\mathbf{e}_0],$$

order (2):

$$(\mathbf{k}_f\cdot\mathbf{k}_0)(\mathbf{e}_f^*\cdot\mathbf{z}_J)(\mathbf{e}_0\cdot\mathbf{z}_J)+[\mathbf{k}_f\leftrightarrow\mathbf{e}_f]+[\mathbf{k}_0\leftrightarrow\mathbf{e}_0]+[\mathbf{k}_f\leftrightarrow\mathbf{e}_f,\mathbf{k}_0\leftrightarrow\mathbf{e}_0]+i(\mathbf{k}_f\times\mathbf{k}_0)\cdot\mathbf{z}_J(\mathbf{e}_f^*\times\mathbf{e}_0)\cdot\mathbf{z}_J,$$

order (3):

$$i(\mathbf{k}_f\cdot\mathbf{z}_J)(\mathbf{k}_0\cdot\mathbf{z}_J)(\mathbf{e}_f^*\times\mathbf{e}_0)\cdot\mathbf{z}_J+[\mathbf{k}_f\leftrightarrow\mathbf{e}_f]+[\mathbf{k}_0\leftrightarrow\mathbf{e}_0]+[\mathbf{k}_f\leftrightarrow\mathbf{e}_f,\mathbf{k}_0\leftrightarrow\mathbf{e}_0],$$

order (4):

$$(\mathbf{k}_f\cdot\mathbf{z}_J)(\mathbf{k}_0\cdot\mathbf{z}_J)(\mathbf{e}_f^*\cdot\mathbf{z}_J)(\mathbf{e}_0\cdot\mathbf{z}_J).$$

The relevant coefficients with which these terms contribute for each M will be given elsewhere. In a spiral antiferromagnet, each order will give rise to a separate magnetic satellite. The appearance of four harmonic satellites is a characteristic signature of a quadrupole resonance ($E2$ or $M2$). The polarization dependences of the four $E2$ magnetic contributions will generally allow all the possible combinations of $\sigma\leftrightarrow\sigma$, $\sigma\leftrightarrow\pi$, and $\pi\leftrightarrow\pi$ scattering. (In contrast, the first-order $E1$ harmonic only allows $\sigma\leftrightarrow\pi$ and $\pi\leftrightarrow\pi$ scattering.)

These ideas give a simple explanation for the complex resonance spectra obtained at the L_{III} edge in Ho (Ref. 1): The double-peak structure is the superposition of resonance arising from different transitions. The lower resonance several eV below the edge is the $E2$ transition $2p_{3/2}\leftrightarrow 4f$, giving rise to four harmonics, which were predicted and observed. The appearance of only two harmonics at the high-energy resonance, and the absence of $\sigma\leftrightarrow\sigma$ scattering in the first harmonic, identify this as an $E1$ resonance, presumably the $2p_{3/2}\leftrightarrow 5d$ transition. In Fig. 1, we give theoretical curves for the four harmonics. These curves are a combination of calculation and parametrization, carried out under the simplest approximations. The purposes here is qualitative illustration, and not a detailed fit to the data. We have calculated $f_{E2}^{(\mathrm{xres})}$ using nonrelativistic Hartree-Fock wave functions assuming a Hund's-rule ground state for the Ho ion, with four empty atomic orbitals $|4f,m_l,\downarrow)$, $m_l=-3$ to 0. For $\sigma\leftrightarrow\sigma$ scattering, the calculated peak resonance-nonresonance ratio $f_{E2}^{(\mathrm{xres})}/f^{(\mathrm{mag})}=4.3i$ for the first harmonic, giving a 19/1 intensity ratio, in reasonable agreement with the observations. $f^{(\mathrm{mag})}$ only contributes to the first harmonic, interfering with $f_{E2}^{(\mathrm{xres})}$ for $\sigma\leftrightarrow\sigma$ scattering, giving a pronounced symmetry of the first-harmonic $\sigma\leftrightarrow\sigma$ resonance curve, with constructive interference predicted on the high-frequency side of resonance, as observed. The calculation of $f_{E1}^{(\mathrm{xres})}$ is more complicated. The $5d$ states are more extensive than the highly localized $4f$ orbitals, and it is necessary to include banding (or "hopping"), crystal-field mixing of the m_l

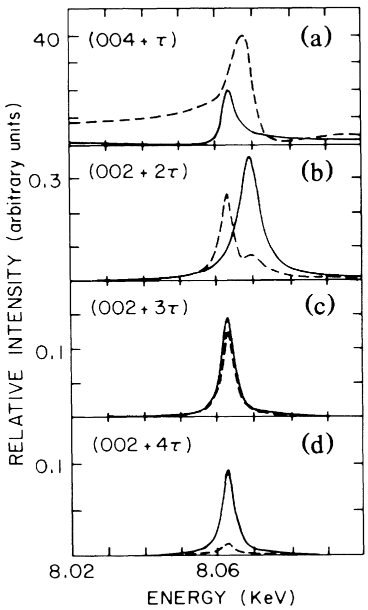

FIG. 1. Relative scattered intensities (theoretical) vs x-ray energy for the L_{III} edge in Ho: (a) $(004+\tau)$, (b) $(002+2\tau)$, (c) $(002+3\tau)$, and (d) $(002+4\tau)$. The solid lines give the $\sigma\leftrightarrow\sigma$ scattering, and the dashed lines give $\sigma\leftrightarrow\pi$.

orbitals, and exchange splitting. In addition, band occupation and induced polarization must be determined, as discussed above. These calculations are underway, but regardless of the detailed nature of the states, the parametric form for the polarization dependence is given by Eq. (3). In Fig. 1(a), the magnitude of $F_{11}-F_{1-1}$ was chosen to give a 2/1 ratio for the peak $E1$ and $E2$ intensities, and the sign was chosen to give constructive interference with $f^{(\mathrm{mag})}$ on the low-frequency side of resonance, to agree with experiment. As shown in Eq. (3), either sign is possible. The present choice of sign and magnitude corresponds to $P(\mathrm{Ho})\approx+0.11$, in reasonable agreement with the previous simple estimate of $+0.07$. In Fig. 1(b), the magnitude of $2F_{10}-F_{11}-F_{1-1}$ was chosen to give a 4/3 ratio for the peak $E1$ and $E2$ intensities. The dominant $E1$ contribution is predicted to be $\sigma\leftrightarrow\sigma$, but with a small $\sigma\leftrightarrow\pi$ contribution which interferes with the $E2$ resonance which is predicted to be almost entirely $\sigma\leftrightarrow\pi$ for the (002) reflection, in good agreement with the observations. The $E1$ contribution to the second harmonic indicates that there is a nonsphericity in the $5d$ polarization. The third and fourth harmonics arise entirely from the $E2$ resonance, and both $\sigma\leftrightarrow\sigma$ and $\sigma\leftrightarrow\pi$ contributions are predicted, in agreement with experiment.

The theory gives excellent qualitative and reasonable quantitative agreement with the observations even with these simple approximations. More sophisticated calculations will be required to give accurate fits to the data. The sensitivity of the measurements to the details of the magnetic properties, and the large enhancement of the magnetic scattering, promise to make this new spectroscopy an important new probe for magnetic studies.

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