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LETTER TO THE EDITOR

**Magnetic absorption dichroism and sum rules in itinerant magnets**

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**Abstract.** In this letter we discuss x-ray magnetic dichroism in magnetic materials where an itinerant model of the magnetic behaviour is appropriate. Inspired by progress made in interpreting dichroism spectra in a localized approach, we show that dichroism spectra are an excellent measure of the orbital and spin magnetic moments in itinerant magnets. By performing an energy decomposition of the sum rules we show that the structure found in dichroism spectra reflects the energy dependence of the magnetic moment.

The availability of intense x-rays from synchrotron sources has generated great interest in using x-rays as a probe of magnetism in condensed matter [1]. Among the most important effects are resonant magnetic scattering [2–4] and circular magnetic dichroism (CMXD) [5–7]. Recently there has been considerable progress in our understanding of circular dichroism and what it tells us about the properties of materials. In particular Thole *et al* [8] and Carra *et al* [9] have performed model calculations leading to an approximate magneto-optical sum rule which relates the integrated CMXD signal to the orbital and spin magnetic moments of the electrons in the dipole allowed states. This has proved to be a very useful and enlightening step forward in our understanding. However, there are still several questions to be answered. Firstly, the orbital and spin magnetic moments are energy integrated quantities, but the dichroism curves are often highly structured [5–7] and so the sum rules do not extract all the information from a dichroism experiment. Secondly, the models used in references [8, 9, 10] were based on a localized description of magnetism. There are a large number of materials where the magnetism is well described by an itinerant theory, and it is not at all clear that the sum rules will translate to these materials. Wu *et al* [11] have addressed this question for the orbital moment sum rule in a very detailed study and found that the sum rule holds for real materials to the 5–10% level. In this paper we describe further progress on this topic as we consider both the spin and orbital sum rules. Furthermore, we examine the energy decomposition of the sum rules and show explicitly how these are related to the magnetic moment.

To tackle such problems requires a description of electrons in condensed matter which includes both relativity and magnetism. Such a description has been provided by Strange *et al* [12, 13], who developed a relativistic spin-polarized version of the KKR method of band theory. Their expression for the  $4 \times 4$  bispinor Dirac Green's function is

$$G(\mathbf{r}, \mathbf{r}', E) = \sum_{\kappa\kappa'm_jm'_j} Z_{\kappa m_j}(\mathbf{r}, E) \tau_{\kappa m_j \kappa' m'_j}(E) Z_{\kappa' m'_j}^\dagger(\mathbf{r}', E) - \sum_{\kappa m_j} Z_{\kappa m_j}(\mathbf{r}, E) J_{\kappa m_j}^\dagger(\mathbf{r}', E) \quad (1)$$

where  $Z_{\kappa m_j}(\mathbf{r}, E)$  and  $J_{\kappa m_j}(\mathbf{r}, E)$  are the regular and irregular solutions of the Kohn–Sham–Dirac equation at energy  $E$ ,  $\kappa$  and  $m_j$  are the usual relativistic quantum numbers [14], and  $\tau_{\kappa m_j \kappa' m_j'}$  are matrix elements of the scattering path operator which may be obtained from band theory [13]. From this Green's function many observables can be evaluated. The particular observables we are interested in are the spin and orbital contributions to the magnetic moment given respectively by

$$\mu_{\text{spin}}(E) = -\frac{\mu_B}{\pi} \text{Im Tr} \tilde{\beta} \tilde{\sigma} G(\mathbf{r}, \mathbf{r}, E) \quad (2a)$$

where the trace includes integration over  $\mathbf{r}$ , and [15]

$$\mu_{\text{orbit}}(E) \approx -\frac{\mu_B}{\pi} \text{Im Tr} \tilde{\beta} \tilde{L}_z G(\mathbf{r}, \mathbf{r}, E). \quad (2b)$$

Here  $\tilde{\beta}$  and  $\tilde{\sigma}$  are the usual Dirac matrices in the standard representation. Both equations (2a) and (2b) have to be integrated up to the Fermi energy  $E_F$  to give the total spin and orbital magnetic moments.

The scattering Green's function can also be used in the interpretation of spectroscopies and Durham [16] has developed a particularly powerful and systematic formalism for studying the interaction of electromagnetic radiation with electrons in a solid from first principles. His work uses multiple scattering theory to implement the diagrammatic perturbation theory of Keldysh. The framework he sets up can be translated without change into relativistic quantum theory. In this theory the absorption rate for a photon of wavevector  $\mathbf{q}$  and polarization  $\lambda$  by excitation of an electron from a core state described by the Dirac spinor  $\psi_c(\mathbf{r})$  is given by

$$W^\lambda = -\frac{1}{\Gamma} \int d^3r \int d^3r' \psi_c^\dagger(\mathbf{r}) X_{q\lambda}(\mathbf{r}) \text{Im} G(\mathbf{r}, \mathbf{r}'; E_c + \hbar\omega) X_{q\lambda}^\dagger(\mathbf{r}') \psi(\mathbf{r}'). \quad (3)$$

Here  $E_c$  is the energy of the core state,  $\omega$  is the frequency of the incident photon, and  $X_{q\lambda}$  is the usual interaction vertex

$$X_{q\lambda} = -e\alpha \cdot \mathbf{A}(\mathbf{r}) = -e\alpha \cdot \epsilon_\lambda e^{i\mathbf{q} \cdot \mathbf{r}} \quad (4)$$

where  $\alpha$  represents the Dirac matrices again in the standard representation, and  $\mathbf{A}(\mathbf{r})$  is the vector potential associated with the photon of wavevector  $\mathbf{q}$ .  $\epsilon_\lambda$  is a vector describing the polarization of the incident photon.  $\Gamma$  represents the lifetime of the initial state. The above theory has been implemented and has yielded dichroism spectra in agreement with experiment for a wide variety of materials [17].

In a single localized ion model Thole *et al* [8] wrote down a sum rule showing that the integrated dichroism signal is directly proportional to the orbital contribution to the magnetic moment. Their expression is

$$\rho = \frac{\int_{j\pm} d\omega (W^+ - W^-)}{\int_{j\pm} d\omega (W^+ + W^- + W^0)} = \frac{1}{2} \frac{l(l+1) - c(c+1) + 2}{l(l+1)(4l+2-n)} \langle L_z \rangle \quad (5)$$

where  $c$  is the  $l$  quantum number of the core state being excited and  $n$  is the number of occupied valence states with the dipole allowed quantum number  $l$ . Hence  $(4l+2-n) = n_h$  is the number of holes in the valence shell.  $W^+$ ,  $W^-$ , and  $W^0$  are the absorption rates for left circularly, right circularly and linearly polarized light respectively.  $\langle L_z \rangle$  is the expectation value of the orbital magnetic moment. The integration is over the absorption edge for both the  $j = l + 1/2$  and  $j = l - 1/2$  states. A similar expression relating the dichroism to the

spin-magnetic moment has been derived by Carra *et al* [9]

$$\eta = \frac{\int_{j+} d\omega(W^+ - W^-) - \frac{c+1}{c} \int_{j-} d\omega(W^+ - W^-)}{\int_{j\pm} d\omega(W^+ + W^- + W^0)} = \frac{l(l+1) - 2 - c(c+1)}{6c(4l+2-n)} \langle 2S_z \rangle + \frac{l(l+1)[l(l+1) + 2c(c+1) + 4] - 3(c-1)^2(c+2)^2}{6lc(l+1)(4l+2-n)} \langle T_z \rangle. \quad (6)$$

Here  $\langle 2S_z \rangle$  is the expectation value of the spin magnetic moment.  $\langle T_z \rangle$  is a dipolar operator expected to be negligible in cubic systems. Henceforth we ignore it. These sum rules have proved remarkably successful in determining the magnetic moment of a number of materials, although one should note that the orbital moment sum rule has a different sign in references [8] and [9]. In deriving the sum rules these authors neglected differences between radial integrals if the levels are spin-orbit split. We have analysed this approximation in iron and found that it holds remarkably well. It is valid to within about 5% in all cases, and for many transitions it is a lot better than this.

Now we are in a position to test the validity of these sum rules for itinerant magnets. The magnetic moment can be calculated directly from equations (2) or we can calculate the absorption rate for right, left and linearly polarized x-rays and determine the magnetic moment from equations (5) and (6). We have selected the  $L_2$  and  $L_3$  edges of iron as our example. In this case equation (5) can be written

$$\langle L_z \rangle = 2n_h \rho \quad (7)$$

and equation (6)

$$\langle 2S_z \rangle = 3n_h \eta \quad (8)$$

where  $n_h$  is the number of holes in the d band, and in our calculation we found  $n_h = 3.44$ . Firstly we will consider the integrated quantities. From (2a) we find the spin contribution to the magnetic moment in iron from the d electrons is  $2.025\mu_B$  whereas the sum rule gives  $1.922\mu_B$ . For the orbital moment we find equation (2b) gives  $0.056\mu_B$  and the sum rule gives  $0.048\mu_B$ . This is a remarkable degree of agreement between the two approaches. There is a small uncertainty in the value calculated from the sum rule, because we have to select a cut off for the energy integrations. Our integration was done over a range of 10.5 eV from the absorption edge. There is also some uncertainty in the value of the orbital moment in iron; Carra *et al* [9] (and references therein) find that the ratio of orbital to spin moments is around a factor of two greater than our calculation. Experimental determination of the orbital component of the moment must await experiments of the type suggested by Blume [1]. One can ask how this agreement comes about. The magnetic moment comes from the electrons below  $E_F$  whereas the absorption experiments probe the empty states above  $E_F$ . Consider first the spin magnetic moment. If all the iron d bands were filled they would make no contribution to the spin magnetic moment. However they are not filled, and the filled part of the d bands determines the magnetic moment. The spin magnetic moment the empty d bands above  $E_F$  would have if they were filled must be the negative of this. Hence the dichroism experiment probes the spin magnetic moment of the empty states above  $E_F$  and this is related by a change of sign to the magnetic moment of the occupied states. This is illustrated in figure (1) for iron, where the full line is the spin magnetic moment associated with the empty states above  $E_F$  as a function of energy calculated from equation (2a) and the dashed line is the right hand side of equation (7) where the energy dependence arises because we have not done the energy integration in the numerator of  $\rho$ . An exactly analogous argument holds for the orbital moment and we illustrate this in figure (2). Here the full line is the orbital magnetic moment associated with the empty states above  $E_F$  as a

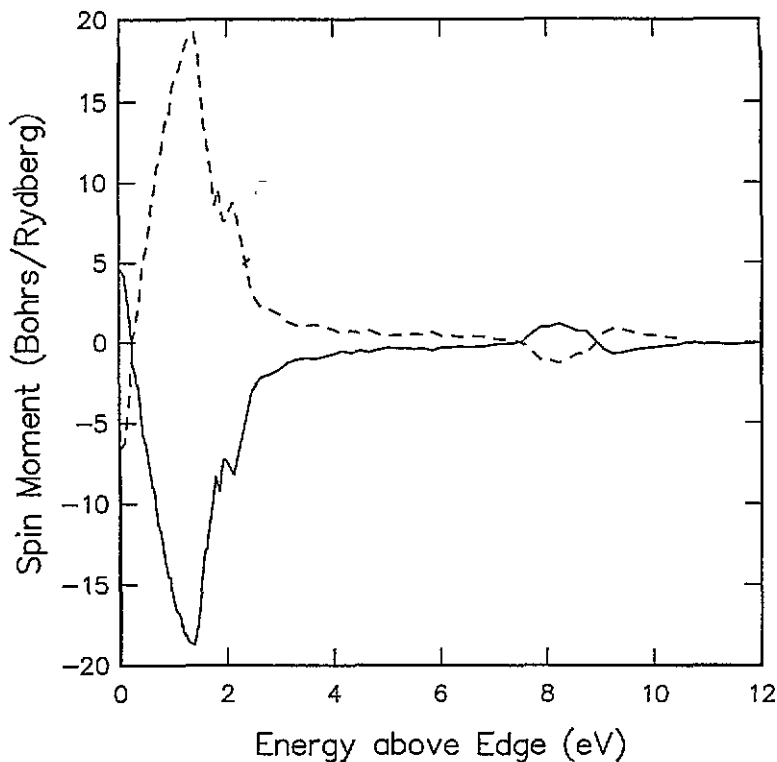


Figure 1. The full line shows the spin magnetic moment as calculated from equation (2a), the dashed line is the spin magnetic moment calculated from equation (8).

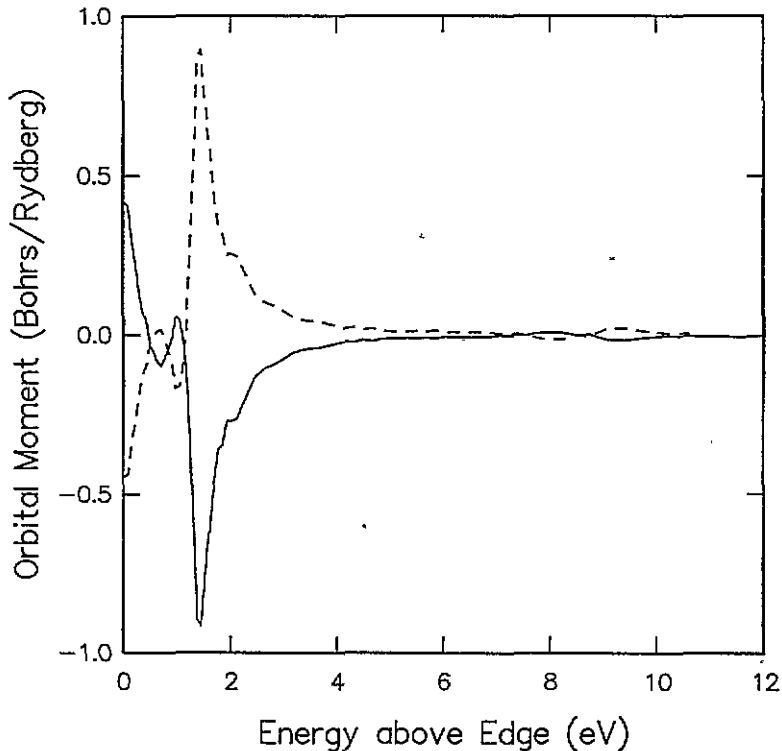


Figure 2. The full line shows the orbital magnetic moment as calculated from equation (2b), the dashed line is the orbital magnetic moment calculated from equation (7).

function of energy calculated from equation (2b) and the dashed line is the right hand side of equation (8) again with the energy integration in the numerator not performed. Clearly in both cases there is excellent agreement in the magnitude of the curves and they are of opposite sign. This indicates that the sum rules hold not just in their energy integrated form, but also at all individual energies. Wu *et al* [11] have also come to this conclusion for the orbital sum rule.

There are some difficulties with this approach which should not be ignored. Firstly, to get the correct magnetic moment from the sum rule, one should integrate up to the top of the dipole allowed bands and no further. Although an approximate value for this is easy to obtain from the density of states, a precise value is not well defined, hence some uncertainty is introduced into the calculation. Secondly, we work in the dipole approximation, and from the  $L_2$  and  $L_3$  edges this allows transitions to s states as well as d states. The s state contribution to the magnetic moment is completely ignored in the analysis above, although it does arise through hybridization with the d bands. Finally one must be wary of using the above methods to describe dichroism in compounds and alloys, where the fraction of the unit cell associated with each ion, and hence the number of electrons on each ion, is an arbitrary quantity, and so there would be further uncertainty in the parameters used in the sum rules.

From this analysis we can conclude that the techniques developed to determine the spin and orbital contribution to the magnetic moment in localized magnets from the results of absorption dichroism experiments are remarkably robust and can be transferred to the case of itinerant magnets. Furthermore, figures (1) and (2) suggest that it is not just the energy integrated quantities for which this holds true, but that they hold at all individual energies as well.

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