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Orbital-magnetization sum rule for x-ray circular dichroism: A simple proof

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A simple and physically intuitive proof for the sum rule relating the integral of the x-ray circular dichroism from a core shell to the value of the orbital magnetic moment in ferromagnets is described. It provides a formalism in which further results of interest can be derived by elementary methods.

I. INTRODUCTION

A useful approximate sum rule, by which the integral of the circular dichroism signal from the two spin-orbit partners of a core edge can be related to the orbital magnetic moment in a ferromagnet, was recently derived by Thole *et al.*[1] Their proof is based on elegant methods of angular momentum theory but is rather complex and lacks the appeal of intuitive physical concepts, such as those introduced in the interpretation of x-ray dichroism spectra by Imada and Jo.[2]

The purpose of the present paper is to show that the sum rule can be derived by rather elementary and physically transparent methods, without any loss of generality. The basic idea is to express the electronic states of the system in a basis of one-electron wave functions (Slater determinants) which are eigenstates of L_z. The essence of the argument is presented in the next section, while the final section is devoted to concluding remarks.

II. DERIVATION OF THE SUM RULE

Consider an ion with an incomplete outer shell with angular momentum l, and let l^n be the ground-state configuration. The ground state $|0\rangle$ can be expanded in a set of Slater determinants $\{n_{m\sigma}\}$ labeled by the quantum numbers of the $h=2(2l+1)-n$ missing electrons in the l shell:

$$|0\rangle=\sum_{\{n_{m\sigma}\}} c^{(0)}(\{n_{m\sigma}\})|\{n_{m\sigma}\}\rangle ,$$
$$\{n_{m\sigma}\}=\{n_{-l\uparrow},n_{-l\downarrow},\ldots,n_{m\sigma},\ldots,n_{l\uparrow},n_{l\downarrow}\} , \tag{1}$$

where each of the $n_{m\sigma}$ is either 0 or 1, and they add up to h. Obviously, we can express properties of the ground state in terms of the expansion coefficients $c^{(0)}(\{n_{m\sigma}\})$ and, in particular, the z component of the angular momentum is

$$\langle 0|L_z|0\rangle=-\sum_{\{n_{m\sigma}\}}|c^{(0)}(\{n_{m,\sigma}\})|^2\sum_{m,\sigma} m n_{m,\sigma} . \tag{2}$$

Similarly, each final state $|f\rangle$ in the configuration $c^{2(2c+1)-1}l^{n+1}$, where c is a core shell with orbital angular momentum c, can be expanded in a basis of Slater determinants $\{c_z,\sigma_c;n'_{m\sigma}\}$ specifying the core-hole quantum numbers, as well as those of the $h-1$ holes of the l shell, $n'_{m\sigma}$. The prime acknowledges the difference in radial one-electron wave functions between ground and excited states. Consider the integrated strength of dipole transitions D_q ($q=-1,0,1$) between the ground and excited configurations,

$$I_q=\sum_{|f\rangle}\omega_{0f}|\langle 0|D_q|f\rangle|^2 , \tag{3}$$

under the following assumptions: (i) for all transitions, the radial matrix element is the same, (ii) ω_{0f} is replaced by an average $\bar{\omega}$. These are equivalent to the assumptions of Thole *et al.*[1] and imply small errors if the spin-orbit splitting of the c shell and the multiplet splitting are small compared to $\hbar\bar{\omega}$, which is the case for x-ray transitions. Then,

$$I_q=\bar{\omega}\sum_f|\langle 0|D_q|f\rangle|^2=\bar{\omega}\langle 0|D_q\sum_f|f\rangle\langle f|D_{-q}|0\rangle . \tag{4}$$

Here $\sum_f|f\rangle\langle f|$ is a projection operator over the $c^{2(2c+1)-1}l^{n+1}$ configuration, and it can be expressed as $\sum_{f'}|f'\rangle\langle f'|$ in terms of any complete basis f' for the configuration. We choose the Slater determinants $\{c_z,\sigma_c;n'_{m\sigma}\}$ and obtain

$$I_q=\bar{\omega}\sum_{\{n_{m\sigma}\}}\sum_{\{c_z,\sigma_c;n'_{m\sigma}\}}|c^{(0)}(\{n_{m,\sigma}\})|^2|\langle\{n_{m,\sigma}\}|D_q|\{c_z\sigma_c;n'_{m,\sigma}\}\rangle|^2 . \tag{5}$$

Notice that selection rules on m prevent the appearance of terms mixing different components of $|0\rangle$.

It is easily shown that the above D_q matrix element is proportional to

$$n_{c_z+q,\sigma_c}\begin{pmatrix} c & 1 & l \\ -c_z & -q & c_z+q \end{pmatrix}^2 \tag{6}$$

via the radial matrix element and some overlap integrals of one-electron wave functions which are the same for all terms in the sum. These factors disappear when intensity ratios are considered, e.g.,

$$\frac{I_1-I_{-1}}{I_1+I_0+I_{-1}}=I_{\rm dic}\ , \tag{7}$$

the quantity appearing in Eq. (5) of Ref. 1. Substituting well-known expressions for three j symbols, namely,

$$\begin{pmatrix} j+1 & 1 & j \\ -m & -1 & m+1 \end{pmatrix}^2=\frac{(j-m)(j-m+1)}{(2j+1)(2j+2)(2j+3)}\ ,$$

$$\begin{pmatrix} j+1 & 1 & j \\ -m & 0 & m \end{pmatrix}^2=\frac{2(j+m+1)(j-m+1)}{(2j+1)(2j+2)(2j+3)}\ ,$$

into Eqs. (6) and (5), we find, for example, for $l=c+1$, after some algebra,

$$I_{\rm dic}=\frac{\sum_{\{n_{m\sigma}\}}|c^{(0)}\{n_{m\sigma}\}|^2\sum_{m=-l}^{l}\sum_{\sigma}n_{m,\sigma}m/(2l+1)l}{\sum_{m=-l}^{l}\sum_{\sigma}n_{m,\sigma}/(2l+1)}\ ,$$

or, comparing with Eq. (2),

$$I_{\rm dic}=-\langle 0|L_z|0\rangle/\{l[2(2l+1)-n]\}$$

in agreement with Eq. (5) of Thole *et al.*

III. CONCLUDING REMARKS

The sum rule of Thole *et al.*[1] is derived with elementary methods and without loss of generality. The use of Slater determinants of one-electron l_z,σ_z eigenfunctions simplifies the algebra and provides a rigorous formulation for the physically intuitive argument of Imada and Jo.[2]

Although deriving again a known result may appear as an irrelevant achievement, it is perhaps worth noticing that the proposed formalism may turn out to be helpful in providing further results. It is, in fact, our conviction that considerable information on magnetic systems is contained in x-ray dichroism spectra but that more progress in the theory is necessary to extract it. As an example one can mention the information about the mixing of states with different l-shell occupations in the ground state,[3] for which the sum rule discussed here is not applicable in a straightforward fashion.

Furthermore, the description in terms of Slater determinants of one-electron wave functions with specified angular momentum character is similar to that adopted in band theory, and may therefore help to bridge the conceptual gap between atomic and band description of systems with local correlations.

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