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Theoretical analysis of magnetic dichroism in the x-ray-resonance scattering of cubic systems at the *K* and *L* edges

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Magnetic dichroism is investigated in second-order processes, especially for x-ray-resonance scattering, in a semirelativistic treatment. The matrix element of the cross section is expressed by the Green function of the system for the unoccupied states and by the Fourier-transformed density for the occupied states. The lattice harmonic expansion of the Green function is used, the coefficients of which can be calculated by a band-structure investigation. For cubic systems and resonances at the *K* and *L* edge, respectively, the matrix elements are evaluated for dipole and quadrupole contributions. It is analyzed how to measure different contributions independently by observing Bragg peaks in and outside the scattering plane and by changing the polarization of the incoming and of the scattered photon.

I. INTRODUCTION

Magnetic properties of materials can be studied via x-ray experiments by taking advantage of the beam polarization of the synchrontron radiation. The success of preliminary investigations stimulated experimental and theoretical developments. Synchrotron beamlines are available and improved, e.g., Vettier[1] reported that at the European Synchrotron Radiation facility a new magnetic scattering beamline is to be opened to users in 1995. Using new sources the problem associated with the small scattering cross section can be overcome. Fields of interest are the second-order processes magnetic x-ray Bragg scattering, magnetic x-ray resonance scattering, magnetic Compton scattering and the first-order processes magnetic dichroism in the x-ray absorption spectra and magnetic dichroism in the photoelectron emission.

Magnetic x-ray dichroism may be caused by the spin polarization of the valence electrons and by the spin-orbit splitting of the valence states in ferromagnetic materials and in the rare earth, by the spin-orbit splitting of the core states, and by the relativistic corrections in the interaction of the light with the electrons.

Linear and circular magnetic x-ray dichroism at the absorption edge was observed, e.g., by van der Laan *et al.*,[2] Schütz *et al.*,[3] and Chen *et al.*[4] Ebert *et al.*[5] succeeded to explain such spectra by spin-polarized relativistic band-structure calculations where spin-polarization and spin-orbit coupling are treated on a common level. Multiple scattering contributions could be included using the relativistic scattering path operator. The spin and angular momentum contribution to the spin-polarization of the unoccupied states can be investigated.

Magnetic circular dichroism in the core-level photoemission was investigated by Baumgarten *et al.*[6] Thole and van der Laan[7,8] analyzed that different combinations of the three polarizations in the experiment allow to distinguish eight fundamental spectra. Angular-resolved measurements[9] of photoelectrons from valence bands can be compared with the electron energy bands of magnetic materials including the spin-polarization and the spin-orbit interaction.

Our considerations are concentrated to the second-order processes. The cross section for the scattering of photons by electrons was calculated by Lipps and Tolhoek[10] including the polarization. The amplitude was given by Platzman and Tzoar[11] and by de Bergevin and Brunel.[12] Platzman and Tzoar have shown that it is possible to determine the spin-dependent momentum distribution of the electrons by the incoherent Compton scattering of polarized x rays and to determine the magnetic structure of antiferromagnetic solids by the coherent Bragg scattering. de Bergevin and Brunel reported Bragg scattering results for zinc-substituted magnetite. Magnetic Compton profiles have been measured for iron[13–16] and nickel.[16] Special features in the low-momentum region could be explained[17,18] by the negatively polarized *s*-*p* band electrons.

A second-order resonance process can be observed in the emission spectra, too. Strange, Durham, and Gyorffy[19] describe that information is available about the spin polarization of the occupied and the unoccupied part of the energy bands from dichroic x-ray fluorescence experiments.

Blume[20] analyzed the magnetic scattering of x rays and discussed the possibility of resonance effects in the Bragg scattering. At the *K* edge this resonance effect was first observed by de Bergevin and Brunel[21] for iron and by Namikawa *et al.*[22] for nickel. The resonance effect is enhanced, if there is a spin-orbit splitting of the core level. Thus, large effects have been predicted in the vicinity of the $L_{\mathrm{II,III}}$ and the $M_{\mathrm{IV,V}}$ edge.[23,24] Magnetic x-ray resonant scattering recently was investigated for Ho,[23,25] UAs,[24] and TmSe.[26] It was pointed out that quadrupole transitions give a remarkable contribution,[27,28] if localized $4f$ states are involved.

We analyze the resonant scattering for cubic systems as iron and nickel. The transition metals are investigated intensively, nevertheless there is a considerable interest at

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the electronic properties of the cubic ferromagnetic $3d$ metals. Recently Hammond, Fahsold, and Kirschner[29] have investigated the elastic and inelastic reflection of spin-polarized low-energy electrons from Fe(110), van der Laan *et al.*[30] have observed the magnetic circular dichroism in Ni $3p$ core-level photoemission, Waddill, Tobin, and Pappas[31] have measured the $2p$ core-level photoemission from thin Fe layers on a Cu(001) substrate. Schneider, Venus, and Kirschner[32] have shown that more detailed features can be investigated by magnetic circular dichroism in angle-resolved photoemission spectra. Ebert *et al.*[33] have presented a completely relativistic treatment of the polarization dependence of the $2p$-core-level photoemission spectra of Fe. Stähler, Schütz, and Ebert reported on magnetic K-edge absorption measurements and relativistic calculations in $3d$ elements. Smith *et al.*[35] have calculated magnetic circular dichroism at the L_{II} and L_{III} edges of Ni and Fe using a relativistic tight-binding band-structure approach.

We do not include orbital contributions. Orbital contributions are more important for rare earths. Carra *et al.*,[36] Thole *et al.*,[37] and Altarelli[38] have derived sum rules to split off orbital and spin contribution to the magnetic moment. Jo, Yoshida, and Sawatzky[39] have shown that including an orbital contribution $0.07\mu_B$ it is possible to explain the observed magnetic circular dichroism in the $2p$ and $3p$ x-ray absorption spectra of Ni.

We briefly discuss basic formulas and perturbation theory in Secs. II and III. In Sec. IV the Green function is discussed and the cross section is expressed by the Green function in Sec. V. The spin summation is evaluated in Sec. VI. The matrix elements at the K and L edge are calculated in Sec. VII and VIII, respectively, and the results are discussed in Sec. IX.

II. HAMILTONIAN

We consider the interaction of electrons in a solid with the photon field including relativistic corrections. In the Hamiltonian

$$H = m_e c^2 \beta + c\tau \underline{\boldsymbol{\sigma}} \cdot [\mathbf{p} - e\,\mathbf{A}(t)] + V_{\mathrm{eff}} - \frac{e\hbar}{2m_e} \beta \underline{\boldsymbol{\sigma}} \cdot \mathbf{B}_{\mathrm{eff}} \,, \qquad (1)$$

$$\beta = \begin{bmatrix} I & 0 \\ 0 & -I \end{bmatrix}, \quad \tau = \begin{bmatrix} 0 & I \\ I & 0 \end{bmatrix}, \quad \underline{\boldsymbol{\sigma}} = \begin{bmatrix} \boldsymbol{\sigma} & 0 \\ 0 & \boldsymbol{\sigma} \end{bmatrix}$$

the Coulomb and exchange interaction is described by V_{eff} and $\mathbf{B}_{\mathrm{eff}}$, which usually are calculated in the local density approximation. A static external field $\mathbf{B}_0$ can be added to $\mathbf{B}_{\mathrm{eff}}$. I is the unit matrix and $\boldsymbol{\sigma}$ the Pauli spin vector.

In our semirelativistic treatment we consider the Pauli equation. We include the exchange splitting of the electron energy bands

$$H^0 = \frac{\mathbf{p}^2}{2m_e} + V_{\mathrm{eff}} - \frac{e\hbar}{2m_e} \boldsymbol{\sigma} \cdot \mathbf{B}_{\mathrm{eff}} \,. \qquad (2)$$

In the core states the spin orbit splitting $[\mathbf{s} = (\hbar/2)\boldsymbol{\sigma}]$

$$H^0_{LS} = \frac{1}{4} \left[\frac{\hbar}{m_e c} \right]^2 \frac{1}{r} \frac{dV_{\mathrm{eff}}}{dr} \frac{2\mathbf{L}\cdot\mathbf{s}}{\hbar^2} \qquad (3)$$

can be included. Thus the wave functions, called $|i\rangle, |f\rangle$ in the following, are two-component spinors. The interaction of the photons with the electron contains linear and quadratic terms in the photon field[17]

$$H^{(1)} = -\frac{e}{m_e} \mathbf{A}\cdot\mathbf{p} - \frac{e\hbar}{2m_e} \boldsymbol{\sigma}\cdot\mathbf{B} \,, \qquad (4)$$

$$H^{(2)} = \frac{e^2}{2m_e} \mathbf{A}^2 + \frac{e^2\hbar}{4m_e^2 c^2} \boldsymbol{\sigma}\cdot(\mathbf{A}\times\dot{\mathbf{A}}) \,. \qquad (5)$$

We restrict the electromagnetic field

$$\mathbf{E} = -\dot{\mathbf{A}}, \quad \mathbf{B} = \nabla\times\mathbf{A} \qquad (6)$$

(scalar potential $\phi=0$) to an expression where the vector potential contains two photons:

$$\mathbf{A} = \sqrt{\hbar/2\varepsilon_0\Omega\omega}\,[\boldsymbol{\epsilon} e^{i(\mathbf{q}\cdot\mathbf{r}-\omega t)} + \boldsymbol{\epsilon}^* e^{-i(\mathbf{q}\cdot\mathbf{r}-\omega t)}] + \sqrt{\hbar/2\varepsilon_0\Omega\omega'}\,[\boldsymbol{\epsilon}' e^{i(\mathbf{q}'\cdot\mathbf{r}-\omega' t)} + \boldsymbol{\epsilon}'^* e^{-i(\mathbf{q}'\cdot\mathbf{r}-\omega' t)}] \,. \qquad (7)$$

The factor volume Ω is canceled in the final result, therefore we put $\Omega=1$ in the following formulas. $\mathbf{q}=\hat{\mathbf{q}}\omega/c$ is the wave vector of the plane wave. A real $\boldsymbol{\epsilon}$ describes linearly polarized photons. Circularly polarized photons can be included by complex $\boldsymbol{\epsilon}$. $\boldsymbol{\epsilon}$ is always perpendicular to the wave vector $\boldsymbol{\epsilon}\cdot\hat{\mathbf{q}}=0$, $\boldsymbol{\epsilon}'\cdot\hat{\mathbf{q}}'=0$.

The resonance scattering is a second-order process. $H^{(2)}$ contributes in the first-order perturbation theory and $H^{(1)}$ in the second order. We only include those terms

$$\begin{aligned} H^{(1)} &= -\frac{e}{m_e} \left[\boldsymbol{\epsilon}\cdot\mathbf{p} + i\frac{\hbar}{2}\boldsymbol{\sigma}\cdot(\mathbf{q}\times\boldsymbol{\epsilon}) \right] \sqrt{\hbar/2\varepsilon_0\omega}\, e^{i\mathbf{q}\cdot\mathbf{r}} e^{-i\omega t} - \frac{e}{m_e} \left[\boldsymbol{\epsilon}'^*\cdot\mathbf{p} - i\frac{\hbar}{2}\boldsymbol{\sigma}\cdot(\mathbf{q}'\times\boldsymbol{\epsilon}'^*) \right] \sqrt{\hbar/2\varepsilon_0\omega'}\, e^{-i\mathbf{q}'\cdot\mathbf{r}} e^{i\omega' t} \\ &\equiv W_1 e^{-i\omega t} + W_1'^{\dagger} e^{i\omega' t} \,, \end{aligned} \qquad (8)$$

$$\begin{aligned} H^{(2)} &= \frac{e^2}{m_e} \left[\boldsymbol{\epsilon}'^*\cdot\boldsymbol{\epsilon} - i\frac{\hbar(\omega'+\omega)}{4m_e c^2} \boldsymbol{\sigma}\cdot(\boldsymbol{\epsilon}'^*\times\boldsymbol{\epsilon}) \right] \frac{\hbar}{2\varepsilon_0} \frac{1}{\sqrt{\omega'\omega}} e^{-i(\mathbf{q}'-\mathbf{q})\cdot\mathbf{r}} e^{i(\omega'-\omega)t} \\ &\equiv W_2 e^{i(\omega'-\omega)t} \end{aligned} \qquad (9)$$

which describe the absorption of the photon $\mathbf{q},\boldsymbol{\epsilon},\omega$ and the emission of the photon $\mathbf{q}',\boldsymbol{\epsilon}',\omega'$.

III. PERTURBATION THEORY

Time-dependent perturbation theory results in the matrix element

$$M(f,\mathbf{q}'\boldsymbol{\epsilon}';i,\mathbf{q}\boldsymbol{\epsilon})=\langle f|W_2|i\rangle+\sum_n^{\text{unocc}}\left[\frac{\langle f|W_1'^{\dagger}|n\rangle\langle n|W_1|i\rangle}{E_i+\hbar\omega'-E_n}+\frac{\langle f|W_1|n\rangle\langle n|W_1'^{\dagger}|i\rangle}{E_i-\hbar\omega-E_n}\right]\delta(E_f+\hbar\omega'-E_i-\hbar\omega)dN' \tag{10}$$

for the process where an electron in an initial state $|i\rangle$ is excited into the state $|f\rangle$ and a photon $\mathbf{q}\boldsymbol{\epsilon}$ is scattered into $\mathbf{q}'\boldsymbol{\epsilon}'$.

At this point we have to remember that we have a many-electron system, say N_e electrons. We have to include the Pauli principle. The total Hamiltonian is a sum over expressions (1) with $\mathbf{r}_i$ instead of $\mathbf{r}$ and the wave function is a Slater determinant with orthogonal one-particle states, called $|f\rangle, |n\rangle, |i\rangle$ in (10). Thus, the sum n runs over unoccupied states only. Furthermore we get different expressions for elastic and inelastic photon scattering.

The evaluation of the matrix element

$$\langle b|\sum_j^{N_e} O(\mathbf{r}_j)|c\rangle\langle c|\sum_k^{N_e} O(\mathbf{r}_k)|a\rangle$$

of a one-particle operator $O(\mathbf{r})$ gives different sums for elastic and inelastic scattering. Here we follow the notation of Blume[20] and Namikawa *et al.*[22] where $|b\rangle$, $|c\rangle$ and $|a\rangle$ are N_e-electron wave functions. For the inelastic scattering (Fig. 1) we have a final state with a definite hole at $|i\rangle$ and an electron in the definite state $|f\rangle$. Thus, the hole $|h\rangle$ in the intermediate state has to be placed at $|i\rangle$ and there remains only a sum n over unoccupied states.

The Compton scattering is an inelastic process. We do not observe the scattered electron and we get contributions from all electrons $|i\rangle$. The number of final states dN' for the scattered photon contains their density of states $\rho(\omega')$

$$dN'=\rho(\omega')d\Omega'dE'=\frac{1}{(2\pi)^3}\frac{\omega'^2}{\hbar c^3}d\Omega'dE' \ . \tag{11}$$

Thus the cross section is given by

$$\frac{d^2\sigma}{d\Omega'dE'}=\frac{1}{I_0}\sum_i^{\text{occ}}\sum_f^{\text{unocc}}\frac{2\pi}{\hbar}|M(f,\mathbf{q}'\boldsymbol{\epsilon}';i,\mathbf{q}\boldsymbol{\epsilon})|^2 \times\delta(E_f+\hbar\omega'-E_i-\hbar\omega)\rho(\omega') \ . \tag{12}$$

$I_0=c/\Omega$ is the current density of the incoming photon.

For the elastic scattering (Fig. 2) the final state $|b\rangle$ is equal to the initial state $|a\rangle$ and two sums remain in the matrix element. Thus the cross section for the x-ray-resonance scattering is determined by

$$\frac{d\sigma}{d\Omega'}=\frac{1}{I_0}\frac{2\pi}{\hbar}\left|\sum_i^{\text{occ}}M(i,\mathbf{q}'\boldsymbol{\epsilon}';i,\mathbf{q}\boldsymbol{\epsilon})\right|^2\rho(\omega) =\frac{1}{I_0}\frac{2\pi}{\hbar}|M(\mathbf{q}'\boldsymbol{\epsilon}';\mathbf{q}\boldsymbol{\epsilon})|^2\rho(\omega) \ . \tag{13}$$

The integration over dE' results in $\omega'=\omega$ and $|\mathbf{q}'|=|\mathbf{q}|$.

Due to the Bloch character of the wave functions $|i\rangle$ and $|n\rangle$ the integration in the matrix element can be restricted to one elementary cell and the structure factor $|S(\mathbf{K})|^2$ appears in the cross section where

$$S(\mathbf{K})=\frac{1}{N}\sum_{\mathbf{R}}e^{-i\mathbf{K}\cdot\mathbf{R}} \tag{14}$$

gives contributions, if the scattering vector $\mathbf{K}=\mathbf{q}'-\mathbf{q}$ is equal to a reciprocal lattice vector. N is the number of unit cells. The constant factors in (8), (9), and (11) can be summarized into the square of the classical electron radius $r_0=e^2/4\pi\varepsilon_0 m_e c^2$ which gives the order of magnitude of the cross section.

IV. GREEN FUNCTION

The sum over the intermediate states can be expressed by the Green function

$$\underline{G}(E)=\sum_n\frac{|n\rangle\langle n|}{E-E_n}=\begin{bmatrix}G_\uparrow & 0\\ 0 & G_\downarrow\end{bmatrix} \ . \tag{15}$$

The one-particle Green function of the many-electron system has poles in the upper half of the complex energy plane for states below the Fermi energy E_F and poles in the lower half plane for $E>E_F$. Equation (15) is just the second part. Because we assume that the states $|n\rangle$ are eigenfunctions of σ_z, $\underline{G}$ is diagonal in the spin space.

The imaginary part of the Green function (15) describes the density of states (DOS) of the unoccupied states

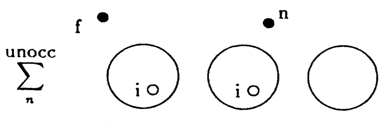

FIG. 1. From right to left: initial state (Fermi sphere), intermediate, and final state of the total system for an inelastic process.

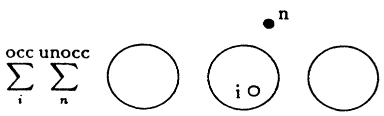

FIG. 2. Initial state (Fermi sphere), intermediate, and final state of the total system for an elastic process.

$$D(E)=-\frac{1}{N\pi}\mathrm{Im}(G_\uparrow(E)+G_\downarrow(E))\equiv-\frac{1}{N\pi}\mathrm{Im}G(E)\ ,\tag{16}$$

$$D_s(E)=-\frac{1}{N\pi}\mathrm{Im}(G_\uparrow(E)-G_\downarrow(E))\equiv-\frac{1}{N\pi}\mathrm{Im}G_s(E)\ ,$$

where $G=G_\uparrow+G_\downarrow$ gives the total DOS $D(E)$ and $G_s=G_\uparrow+G_\downarrow$ describes the spin DOS $D_s(E)$. A factor $1/N$ usually is added, where N is the number of elementary cells. The real part of the Green function can be calculated from the imaginary part using a Kramers-Kronig relation. D and D_s and their local (at a special site) and partial (for a special angular momentum) components result from band-structure calculations. We investigate what components can be measured by the magnetic x-ray resonance scattering.

The Green function with $\mathbf{r}$ and $\mathbf{r}'$ within one atomic sphere can be expanded into lattice harmonics:[40]

$$G(\mathbf{r}',\mathbf{r},E)=\sum_{\Gamma\gamma}\sum_{ll'}G_{ll'}^{\Gamma}(r',r,E)K_l^{\Gamma\gamma}(\mathbf{e}')K_{l'}^{\Gamma\gamma}(\mathbf{e})\ .\tag{17}$$

This and the following relations hold for both G and G_s. $K_l^{\Gamma\gamma}$ is a lattice harmonic corresponding to the representation Γ of the point group of the atomic site. It is a linear combination of spherical harmonics $Y_{lm}(\mathbf{e})$. γ describes different functions of the representation Γ according to its dimension n_Γ. It is an advantage of the expansion (17) that the coefficients $G_{ll'}^{\Gamma}$ are independent of γ.

If we restrict our considerations to cubic systems with s, p, and d states in the occupied and unoccupied part of the conduction band, then we do not have mixed terms l,l' and a unique relation between Γ and l, see Table I. Including f states there is a mixing between $l=1$ and $l=3$.

TABLE I. Cubic irreducible representations built from spherical harmonics of angular momentum l.

1	Γ	n_Γ
0	Γ_1	1
1	Γ_{15}	3
2	$\Gamma_{12},\Gamma_{25'}$	2,3
3	$\Gamma_{2'},\Gamma_{25},\Gamma_{15}$	1,3,3

If we insert (17) into (16)

$$D(\mathbf{r}',\mathbf{r},E)=-\frac{1}{N\pi}\mathrm{Im}G(\mathbf{r}',\mathbf{r},E)$$

$$=\sum_{\Gamma\gamma}\sum_{ll'}A_{ll'}^{\Gamma}(r',r,E)K_l^{\Gamma\gamma}(\mathbf{e}')K_{l'}^{\Gamma\gamma}(\mathbf{e})\ ,\tag{18}$$

we get the expression for the partial DOS

$$D_l^\Gamma(E)=\int_0^{R_{\mathrm{at}}}dr\,r^2A_{ll}^{\Gamma}(r,r,E)\ ,$$

$$D(E)=\sum_\Gamma\sum_l n_\Gamma D_l^\Gamma(E)\tag{19}$$

from the imaginary part $A_{ll'}^{\Gamma}=(-1/N\pi)\mathrm{Im}G_{ll'}^{\Gamma}$ of the coefficients G_{ll}^{Γ} in (17) by integration over an atomic cell.

V. CROSS SECTION

In the matrix element (10) of the cross section

$$\frac{d\sigma}{d\Omega'}=r_0^2|S(\mathbf{K})|^2|M(\mathbf{q}'\boldsymbol{\epsilon}';\mathbf{q}\boldsymbol{\epsilon})|^2\tag{20}$$

we get a zeroth-order contribution and relativistic corrections of the order $\hbar\omega/mc^2$. The resonance scattering (13) is determined by those terms of the matrix element (10) with vanishing denominator. Thus, in the first term of (10) all occupied states are included—core states and valence states. In the second term the summation is restricted to the core levels and the third term can be neglected.[22] For the matrix element remains

$$\begin{aligned}M(\mathbf{q}'\boldsymbol{\epsilon}';\mathbf{q}\boldsymbol{\epsilon})=\ &\boldsymbol{\epsilon}'^*\cdot\boldsymbol{\epsilon}n(\mathbf{K})-i\frac{\hbar\omega}{2m_ec^2}\mathbf{e}_z\cdot(\boldsymbol{\epsilon}'^*\times\boldsymbol{\epsilon})n_s(\mathbf{K})\\&+\sum_i^{\mathrm{edge}}\left[\frac{1}{m_e}\boldsymbol{\epsilon}'^*\cdot\langle i|\mathbf{p}'e^{-i\mathbf{q}'\cdot\mathbf{r}'}\underline{G}\mathbf{p}e^{i\mathbf{q}\cdot\mathbf{r}}|i\rangle\cdot\boldsymbol{\epsilon}+i\frac{\hbar\omega}{2m_ec}\boldsymbol{\epsilon}'^*\cdot\langle i|\mathbf{p}'e^{-i\mathbf{q}'\cdot\mathbf{r}'}\underline{G}\boldsymbol{\sigma}e^{i\mathbf{q}\cdot\mathbf{r}}|i\rangle\cdot(\hat{\mathbf{q}}\times\boldsymbol{\epsilon})\right.\\&\left.-i\frac{\hbar\omega}{2m_ec}(\hat{\mathbf{q}}'\times\boldsymbol{\epsilon}'^*)\cdot\langle i|e^{-i\mathbf{q}'\cdot\mathbf{r}'}\boldsymbol{\sigma}\underline{G}\mathbf{p}e^{i\mathbf{q}\cdot\mathbf{r}}|i\rangle\cdot\boldsymbol{\epsilon}+\frac{(\hbar\omega)^2}{4m_ec^2}(\hat{\mathbf{q}}'\times\boldsymbol{\epsilon}'^*)\cdot\langle i|e^{-i\mathbf{q}'\cdot\mathbf{r}'}\boldsymbol{\sigma}\underline{G}\boldsymbol{\sigma}e^{i\mathbf{q}\cdot\mathbf{r}}|i\rangle\cdot(\hat{\mathbf{q}}\times\boldsymbol{\epsilon})\right]\ .\end{aligned}\tag{21}$$

The summation over the occupied states (with spin $\uparrow$) in the first term results to the Fourier-transformed density

$$n_\uparrow(\mathbf{K})=\sum_i^{\mathrm{occ}}\langle i|e^{-i\mathbf{K}\cdot\mathbf{r}}|i\rangle=\int d^3r n_\uparrow(\mathbf{r})e^{-i\mathbf{K}\cdot\mathbf{r}}\ .\tag{22}$$

We have introduced $n=n_\uparrow+n_\downarrow$ and $n_s=n_\uparrow-n_\downarrow$. Due to $\boldsymbol{\epsilon}\cdot\mathbf{q}=0$ the operator $\mathbf{p}$ commutates with the plane-wave factor $\exp(i\mathbf{q}\cdot\mathbf{r})$ in (21). We have introduced unit vectors by $\mathbf{q}=(\hbar\omega/c)\hat{\mathbf{q}}$.

VI. SPIN OPERATORS

In the cross section we have to evaluate matrix elements of spin operators. Each operator can be expressed by the four operators

$$I=\begin{bmatrix}1 & 0\\ 0 & 1\end{bmatrix},\quad \sigma_z=\begin{bmatrix}1 & 0\\ 0 & -1\end{bmatrix},$$
$$\sigma_x=\begin{bmatrix}0 & 1\\ 1 & 0\end{bmatrix},\quad \sigma_y=\begin{bmatrix}0 & -i\\ i & 0\end{bmatrix}, \tag{23}$$

unit matrix, and Pauli spin matrices. Instead of σ_x,σ_y,

$$\sigma_+=\sigma_x+i\sigma_y=\begin{bmatrix}0 & 2\\ 0 & 0\end{bmatrix},\quad \sigma_-=\sigma_x-i\sigma_y=\begin{bmatrix}0 & 0\\ 2 & 0\end{bmatrix} \tag{24}$$

can be used. An arbitrary operator

$$\underline{M}=\begin{bmatrix}M_\uparrow & M_+\\ M_- & M_\downarrow\end{bmatrix}=\frac{1}{2}(MI+\mathbf{M}\cdot\sigma) \tag{25}$$

can be characterized by a scalar $M=M_\uparrow+M_\downarrow$ and a vector $\mathbf{M}$

$$M_x=M_++M_-\ ,$$
$$M_y=i(M_+-M_-),\ M_z=M_\uparrow-M_\downarrow\equiv M_s \tag{26}$$

or using (24) by

$$\mathbf{M}\cdot\sigma=M_+\sigma_++M_-\sigma_-+M_s\sigma_z\ . \tag{27}$$

In the cross section the Green function has to be multiplied with spin operators. Using the relation $\mathbf{a}\cdot\sigma\sigma\cdot\mathbf{b}=\mathbf{a}\cdot\mathbf{b}+i\sigma\cdot(\mathbf{a}\times\mathbf{b})$ we get

$$2\mathbf{a}\cdot\sigma\underline{M}=\mathbf{a}\cdot\mathbf{M}I+(\mathbf{a}M+i\mathbf{a}\times\mathbf{M})\cdot\sigma\ ,$$
$$2\underline{M}\sigma\cdot\mathbf{b}=\mathbf{M}\cdot\mathbf{b}I+\sigma(M\mathbf{b}+i\mathbf{M}\times\mathbf{b})\ ,$$
$$\begin{aligned}2\mathbf{a}\cdot\sigma\underline{M}\sigma\cdot\mathbf{b}=&[\,\mathbf{a}\cdot M\mathbf{b}+i\mathbf{a}\cdot(\mathbf{M}\times\mathbf{b})]I\\ &+\mathbf{a}\cdot\sigma\mathbf{M}\cdot\mathbf{b}+\mathbf{a}\cdot\mathbf{M}\sigma\cdot\mathbf{b}-\mathbf{a}(\sigma\cdot\mathbf{M})\mathbf{b}\\ &+i(\mathbf{a}\times M\mathbf{b})\cdot\sigma\ .\end{aligned} \tag{28}$$

a,**b** can contain the momentum operator. Therefore we do not commute them with $M,\mathbf{M}$. The Green function (15) is an operator with vanishing off-diagonal elements $M_\pm$. In this case (25) simplifies to

$$\underline{G}=\tfrac{1}{2}(GI+G_s\sigma_z) \tag{29}$$

and in (28) only G and G_s appear.

VII. RESONANCE SCATTERING AT THE *K* EDGE

To get the resonance scattering at the K edge we have to sum over the electrons in the K shell. Then $|i\rangle$ is an eigenfunction of σ_z with quantum numbers $m_s=\pm\frac{1}{2}$ and the summation over m_s is equal to the trace of the spin operator in the matrix element. We have $\mathrm{tr}I=2$ and $\mathrm{tr}\sigma_i=0$, thus only terms containing I in (28) contribute. Furthermore we restrict the expressions to the dipole and quadrupole contribution:

$$\begin{aligned}M(\mathbf{q}'\epsilon';\mathbf{q}\epsilon)=&\{\epsilon'^*\cdot\epsilon n(\mathbf{K})\}_{(\mathrm{I})}-\left\{i\frac{\hbar\omega}{2m_ec^2}\mathbf{e}_z\cdot(\epsilon'^*\times\epsilon)n_s(\mathbf{K})\right\}_{(\mathrm{II})}+\left\{\frac{1}{m_e}\epsilon'^*\cdot\langle 1s|\mathbf{p}'G^{(p)}\mathbf{p}|1s\rangle\cdot\epsilon\right\}_{(\mathrm{III})}\\ &+\left\{\frac{1}{m_e}\epsilon'^*\cdot\langle 1s|\mathbf{p}'(\mathbf{q}'\cdot\mathbf{r}')G^{(d)}(\mathbf{q}\cdot\mathbf{r})\mathbf{p}|1s\rangle\cdot\epsilon\right\}_{(\mathrm{IV})}\\ &-\frac{\hbar\omega}{2m_ec}\{\epsilon'^*\cdot\langle 1s|\mathbf{p}'G_s^{(p)}(\mathbf{q}\cdot\mathbf{r})|1s\rangle\mathbf{e}_z\cdot(\hat{\mathbf{q}}\times\epsilon)+(\hat{\mathbf{q}}'\times\epsilon'^*)\cdot\mathbf{e}_z\langle 1s|(\mathbf{q}'\cdot\mathbf{r}')G_s^{(p)}\mathbf{p}|1s\rangle\cdot\epsilon\}_{(\mathrm{V})}\\ &+\left\{\frac{(\hbar\omega)^2}{4m_ec^2}(\hat{\mathbf{q}}'\times\epsilon'^*)\cdot(\hat{\mathbf{q}}\times\epsilon)\langle 1s|\mathbf{q}'\cdot\mathbf{r}'G^{(p)}\mathbf{q}\cdot\mathbf{r}|1s\rangle\right\}_{(\mathrm{VI})}\\ &-i\left\{\frac{(\hbar\omega)^2}{4m_ec^2}\mathbf{e}_z\cdot\left[(\hat{\mathbf{q}}'\times\epsilon'^*)\times(\hat{\mathbf{q}}\times\epsilon)\right]\langle 1s|\mathbf{q}'\cdot\mathbf{r}'G_s^{(p)}\mathbf{q}\cdot\mathbf{r}|1s\rangle\right\}_{(\mathrm{VII})}\ .\end{aligned} \tag{30}$$

We number the different terms with (I) to (VII). This numbering differs from that one of Namikawa *et al.*[22] because we do not split off the expressions into real and imaginary parts. Furthermore we have some additional terms, which do not contribute under the experimental conditions discussed in that paper. The first term (I) gives the Thomson scattering. It contains the density of all electrons. The second term (II) is the normal magnetic scattering coming from the spin density of the electrons. Both terms result from the first and second term in (21), respectively. The following terms contain the unoccupied states described by the Green function G for all states and G_s for the spin polarization. Due to the selection rules special angular momentum components (p,d) contributions are listed in Table I. (III) is the dipole term and (IV) the quadrupole contribution of the third term in (21). p and d states, respectively, appear in these expressions. There is no contribution from the s states due to $\epsilon\cdot\mathbf{q}=0$. The next two lines of (21) are collected in (V). In this term the p-like part of the spin density of the unoccupied states contributes. The lowest line of (21) gives (VI) and (VII) using the last relation of (28).

TABLE II. Order of magnitude of the terms (I) to (VII) in (30) coming from prefactor, factors p/m_ec and qr. In the last column a factor $1/\text{a.u.}=1/\alpha^2 m_e c^2$ is added from G in (III) to (VII).

	Prefactor	p/m_ec	qr	Total
I	1			1
II	$\hbar\omega/2m_ec^2$			$(Z\alpha)^2/4$
III	m_ec^2	$(Z\alpha)^2$		Z^2
IV	m_ec^2	$(Z\alpha)^2$	$(Z\alpha)^2/4$	$Z^2(Z\alpha)^2/4$
V	$\hbar\omega/2$	$Z\alpha$	$Z\alpha/2$	$Z^3(Z\alpha)^2/8$
VI	$(\hbar\omega)^2/4m_ec^2$		$(Z\alpha)^2/4$	$Z^2(Z\alpha)^4/64$
VII	$(\hbar\omega)^2/4m_ec^2$		$(Z\alpha)^2/4$	$Z^2(Z\alpha)^4/64$

(VII) again includes the spin density.

We get first-order magnetic resonance terms in the square of the matrix element from the product of (I) with (V) and (VII) and from the product of (II) with (III), (IV), and (VI), respectively. In the special case of linear polarized light we do not have a linear term from the magnetic scattering term (II), because $n(\mathbf{K})$ is real if there is a center of inversion. The real part of the Green function cancels in the mentioned products, thus the contributions to the magnetic resonance scattering are proportional to the partial density of states.

To estimate the order of magnitude we use the following approximations. (i) the ratio of the spin density and the total density is less than $\frac{1}{10}$; (ii) if we approximate the $|1s\rangle$ state by a hydrogenlike wave function an expression $\mathbf{p}/mc$ gives (besides a unit vector) a factor $Z\alpha$ where Z is the nuclear charge and α the fine-structure constant; (iii) $\hbar\omega$ can be approximated by Z^2 Ry, or $Z-1$ instead of Z. Thus $\hbar\omega/m_ec^2 \approx (Z\alpha)^2/2$ holds; (iv) an expression qr in (30) can be replaced by $(\omega/c)(a_0/Z)\times Zr/a_0=(\frac{1}{2})Z\alpha \times Zr/a_0$, where a_0 is the Bohr radius; (v) the density of states for d electrons is of the order of magnitude 100/a.u./atom and a few percent of this value for p electrons; (vi) at least we have to estimate the radial integrals with the radial parts of the s, p, and d wave functions, in some cases containing a factor Zr/a_0.

The estimation was done for nickel and the results are listed in Tables II and III. We distinguish between the terms containing the occupied states (n, n_s) and the terms containing the unoccupied states (G, G_s). In n and n_s we have contributions from all angular momenta in G and G_s only from a special angular momentum (p or d) as indicated in the matrix element.

TABLE III. Spin (index s or not), angular momentum (p,d), and radial matrix element (including Zr/a_0 or not) in the contributions of (30). Estimated relative numerical values for Ni including the factors of Table II.

	s	Integration	Value
I	n		1
II	n_s		3×10^{-4}
III	G	$\langle s\|p\rangle\langle p\|s\rangle$	2×10^{-1}
IV	G	$\langle s\|Zr/a_0\|d\rangle\langle d\|Zr/a_0\|s\rangle$	5×10^{-1}
V	G_s	$\langle s\|p\rangle\langle p\|Zr/a_0\|s\rangle$	3×10^{-5}
VI	G	$\langle s\|Zr/a_0\|p\rangle\langle p\|Zr/a_0\|s\rangle$	5×10^{-6}
VII	G_s	$\langle s\|Zr/a_0\|p\rangle\langle p\|Zr/a_0\|s\rangle$	2×10^{-7}

As mentioned we get magnetic resonance contributions from the products (I) with (V) and (VII) and from the products (II) with (III), (IV), and (VI). The order of magnitude is roughly 10^{-5} or less. Which products contribute depends on the polarization. If it is possible to measure the resonance effect for different directions of $\boldsymbol{\epsilon}$ and $\boldsymbol{\epsilon}'$ with respect to each other and with respect to the direction of magnetization $(\mathbf{e}_z)$, then the different terms can be separated.

VIII. RESONANCE SCATTERING AT THE *L* EDGE

We calculate the contributions for the L_{II} and L_{III} edge separately. Thus $|i\rangle$ in (21) is a $2p_{1/2}$ state or a $2p_{3/2}$ state, respectively, and the sum over μ contains the two terms given in (A1) or the four terms of (A2). In contrast to the considerations for the K shell in Sec. VII these states cannot be eigenstates of σ_z and all terms of (28) appear. The evaluation of the terms containing I and σ_z is straightforward, the result is given in (A3) and (A4). To evaluate terms containing σ_x, σ_y or σ_+, σ_- it seems to be necessary to fix the direction of the x axis. But we like to avoid this. We still have five directions $\mathbf{e}_z, \mathbf{q}, \boldsymbol{\epsilon}, \mathbf{q}', \boldsymbol{\epsilon}'$ and the suitable choice of the x axis depends on their relative positions and on the directions of the lattice vectors. But, it is possible to derive an invariant expression of these matrix elements which only contains $\mathbf{e}_z$ and which is given in (A5).

The matrix elements are listed in the Appendix. We have divided them into three groups according to expressions resulting from I, σ_z and σ_x, σ_y, respectively. We like to discuss the difference between the results for the $2p_{1/2}$ and $2p_{3/2}$ state. In the first group (A6) we have to add a factor 2 and in the second (A7) and third groups (A8) we have to change the sign. Thus, adding the $2p_{1/2}$ and $2p_{3/2}$ contributions (A6) remains with a factor 3, which cancels a factor $\frac{1}{3}$ appearing in the formulas. We get the result for a p state without spin-orbit splitting. Thus the terms caused by the spin-orbit splitting are (A7) and (A8). Separating the contributions for the two levels we can get information on the spin polarization, because the largest term, the first line in (A7), contains G_s whereas the corresponding term in (A6) contains G.

IX. CONCLUSIONS

We have calculated the matrix element of the cross section (20) for the elastic scattering of a photon (13) at a spin-polarized system. The matrix element contains three types of terms as can be seen from the K-edge resonance scattering result (30). (i) The classical term (I) describes the Thomson scattering and contains the Fourier-transformed electron density of all occupied states. (ii) A relativistic correction of the order $\hbar\omega/m_ec^2$ (II) is determined by the Fourier-transformed spin density of the occupied states. (iii) Resonance corrections

TABLE IV. The products containing linear resonant terms of the spin-polarized contributions.

Product	Part of G
I×V	$n\times\mathrm{Im}G_s$
I×VII	$n\times\mathrm{Im}G_s$
II×III	$n_s\times\mathrm{Im}G$
II×IV	$n_s\times\mathrm{Im}G$
II×VI	$n_s\times\mathrm{Im}G$

(III–VII) contain the Green function of the unoccupied states with respect to all electrons G and the spin-polarized part G_s, respectively. We avoid replacing the operator $\mathbf{p}$ by $\mathbf{r}$, otherwise we cannot introduce the Green function. $\mathbf{p}$ can be expressed by $\mathbf{p}=\mathbf{e}\,\mathbf{e}\cdot\mathbf{p}-\mathbf{e}\times(\mathbf{e}\times\mathbf{p})=\mathbf{e}(\hbar/i)\partial/\partial r-r\mathbf{e}\times\mathbf{L}$ and this expression acts on the core-state wave function changing the radial or angular momentum part.

In the cross section (20) we have to square the matrix element and usually we neglect the square of the correction. Thus, which terms appear also depends on the polarization, whether we have linearly polarized light with real ϵ or circular polarized light with complex ϵ. In the following we shall restrict the considerations to linearly polarized light.

As mentioned we get magnetic resonance contributions from the product of (I) with (V) and (VII) and from the products of (II) with (III), (IV), and (VI), respectively. We can split off the Green function into real and imaginary parts. For linearly polarized light (I) is a real term and (II) is an imaginary one. Thus in the products always the imaginary part of the Green function, the density of state matrix (18), contributes as listed in Table IV.

In these products the spin polarization of the unoccupied states G_s is involved, on the one hand, and the spin polarization of the occupied states n_s on the other hand.

Furthermore it is possible to select special contributions by a suitable choice of the characteristic directions, magnetization $\mathbf{e}_z$, wave vector $\mathbf{q}$, and polarization ϵ of the incoming photon. The wave vector $\mathbf{q}'$ of the scattered photon is fixed by the Bragg condition to special directions. If it is possible to measure the polarization of the scattered photon then ϵ' can be varied. There appears the same combination of these vectors in terms of the K edge (30) as in the terms for the L edge (A3)–(A5).

$\mathbf{e}_z$ is involved only in those terms that contain spin-polarized components n_s, G_s. Due to the symmetry (17) of the Green function for a cubic-system term (III) in (30) is proportional to $\epsilon'^*\cdot\epsilon$. From the quadrupole term (IV) we get three invariants $I_1=\epsilon'^*\cdot\mathbf{q}'\mathbf{q}\cdot\epsilon$, $I_2=\epsilon'^*\cdot\epsilon\mathbf{q}'\cdot\mathbf{q}+\epsilon'^*\cdot\mathbf{q}\epsilon\cdot\mathbf{q}'$ and $I_3=\sum\epsilon_i'^*q_i'\epsilon_i q_i$, where i means the Cartesian components with respect to the lattice. The first term I_1 vanishes due to the transversality of the light. On the other hand this term results from the Γ_1 part of the Green function. Therefore in (IV) we do not have a contribution $G^{(s)}$. The Γ_{12} part is the linear combination $I_3-I_1=I_3$, thus, only I_3 remains. The $\Gamma_{25'}$ is the linear combination I_2-2I_3.

In Table V we consider which terms of (30) or of the analogous terms in (A3)–(A5) contribute for special conditions. $\mathbf{e}_z$ lies in the surface and $\mathbf{e}_x$ may be the normal direction.

In cases 1–8 the wave vector $\mathbf{q}$ lies in the xy plane and in cases 9–16 in the xz plane. We consider a Bragg peak in the scattering plane and a Bragg peak perpendicular to the scattering plane, respectively. For these cases four combinations of the polarization vectors are involved. We restrict the consideration to linearly polarized light.

We find quite different situations. We can get contributions from all terms. The contribution from the magnetic terms II, V, VII can vanish, but there are special conditions where only the magnetic terms contribute. Thus, it is a question of intensity of the source whether by a suitable combination of different measurements the terms can be analyzed separately.

Summarizing we have shown that magnetic resonance scattering is a suitable tool to investigate the spin polarization of occupied and unoccupied states. We have eval-

TABLE V. Contributing terms of (30) for different directions of the wave vectors and the polarizations. *xy* means that the vector lies in the *xy* plane and *z* means it has the *z* direction. 0 indicates that the term does not contribute.

		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
	$\mathbf{q}$	*xy*	*xy*	*xy*	*xy*	*xy*	*xy*	*xy*	*xy*	*xz*	*xz*	*xz*	*xz*	*xz*	*xz*	*xz*	*xz*
	ϵ	*xy*	*xy*	*xy*	*xy*	*z*	*z*	*z*	*z*	*xz*	*xz*	*xz*	*xz*	*y*	*y*	*y*	*y*
	$\mathbf{q}'$	*xy*	*xy*	*xz*	*xz*	*xy*	*xy*	*xz*	*xz*	*xz*	*xz*	*xy*	*xy*	*xz*	*xz*	*xy*	*xy*
	ϵ'	*xy*	*z*	*xz*	*y*	*z*	*xy*	*xz*	*y*	*xz*	*y*	*z*	*xy*	*xz*	*y*	*z*	*xy*
I	$\epsilon'^*\cdot\epsilon$	+	0	+	+	+	0	+	0	+	0	+	+	0	+	0	+
II	$(\epsilon'^*\times\epsilon)_z$	+	0	+	+	0	0	0	0	0	+	0	+	+	0	0	+
III	$\epsilon'^*\cdot\epsilon$	+	0	+	+	+	0	+	0	+	0	+	+	0	+	0	+
IV	$\epsilon'^*\cdot\mathbf{q}\mathbf{q}'\cdot\epsilon$	+	0	+	+	0	0	+	+	+	0	+	+	0	0	+	+
	$\epsilon'^*\cdot\epsilon\mathbf{q}'\cdot\mathbf{q}$	+	0	+	+	+	0	+	0	+	0	+	+	0	+	0	+
	$\sum\epsilon_i q_i' q_i\epsilon_i$	+	0	+	0	0	0	0	0	+	0	0	+	0	0	0	0
V	$\epsilon'^*\cdot\mathbf{q}b_z$	+	0	+	+	0	0	0	0	0	0	0	0	+	0	+	+
	$b_z'\mathbf{q}'\cdot\epsilon$	+	0	0	+	0	0	0	+	0	+	0	+	0	0	0	+
VI	$\mathbf{b}'^*\cdot\mathbf{b}\mathbf{q}'^*\cdot\mathbf{q}$	+	0	0	+	+	0	+	+	+	0	+	0	0	+	+	+
VII	$(\mathbf{b}'^*\times\mathbf{b})_z\mathbf{q}'^*\cdot\mathbf{q}$	0	0	0	0	+	0	+	+	0	+	+	0	+	0	+	0

uated the cross section for the K edge (30) and the L edge (A3)–(A5). To evaluate the different contributions we have to calculate the Green function (17) or the radial expressions $A_{ll'}^{\Gamma}$ (18). They can be written as products of radial wave functions and weights which are integrals over the Brillouin zone containing the coefficients of the Bloch functions. The Brillouin zone integration can be performed using the tetrahedron method.[41,42]

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APPENDIX: MATRIX ELEMENTS AT THE L EDGE

In Sec. VII the matrix element (21) is given for the case that the core state is a $1s$ state. Now we extend these considerations for a $2p_{1/2}$ and a $2p_{3/2}$ state, respectively. The states characterized by quantum numbers j,l,μ are

$$\begin{aligned} |\tfrac{1}{2}1+\tfrac{1}{2}\rangle &= -\sqrt{\tfrac{1}{3}}Y_{10}\chi_+ + \sqrt{\tfrac{2}{3}}Y_{11}\chi_- \,, \\ |\tfrac{1}{2}1-\tfrac{1}{2}\rangle &= +\sqrt{\tfrac{1}{3}}Y_{10}\chi_- - \sqrt{\tfrac{2}{3}}Y_{1-1}\chi_+ \end{aligned} \tag{A1}$$

for the $2p_{1/2}$ state and

$$\begin{aligned} |\tfrac{3}{2}1+\tfrac{3}{2}\rangle &= Y_{11}\chi_+ \,, \\ |\tfrac{3}{2}1+\tfrac{1}{2}\rangle &= \sqrt{\tfrac{2}{3}}Y_{10}\chi_+ + \sqrt{\tfrac{1}{3}}Y_{11}\chi_- \,, \\ |\tfrac{3}{2}1-\tfrac{1}{2}\rangle &= \sqrt{\tfrac{1}{3}}Y_{1-1}\chi_+ + \sqrt{\tfrac{2}{3}}Y_{10}\chi_- \,, \\ |\tfrac{3}{2}1-\tfrac{3}{2}\rangle &= Y_{1-1}\chi_- \end{aligned} \tag{A2}$$

for the $2p_{3/2}$ state. The Y_{lm} are spherical harmonics. We have to sum over the electrons in one level, thus over μ, and we evaluate the spin part. We can use the relations

$$\sum_{\mu} \langle \tfrac{1}{2}1\mu|\cdot I\cdot|\tfrac{1}{2}1\mu\rangle = \tfrac{2}{3}\sum_{m} \langle Y_{1m}|\cdots|Y_{1m}\rangle \,, \tag{A3}$$

$$\sum_{\mu} \langle \tfrac{1}{2}1\mu|\cdot\sigma_z\cdot|\tfrac{1}{2}1\mu\rangle = -\frac{2}{3}\sum_{m} m\langle Y_{1m}|\cdots|Y_{1m}\rangle \,, \tag{A4}$$

$$\begin{aligned} \sum_{\mu} \langle \tfrac{1}{2}1\mu|\cdot\sigma_x b_x + \sigma_y b_y\cdot|\tfrac{1}{2}1\mu\rangle &= \tfrac{2}{3}\sqrt{3}[\,\langle Y_{10}|\cdots|\mathbf{e}_r Y_{00}\rangle\cdot i(\mathbf{e}_z\times\mathbf{b}) \\ &\quad - i(\mathbf{e}_z\times\mathbf{b})\cdot\langle Y_{00}\mathbf{e}_r|\cdots|Y_{10}\rangle] \,. \end{aligned} \tag{A5}$$

z is the axis of quantization, but we did not like to fix the x and y axis. This was done in (A5) by expressing the spherical harmonics Y_{1m} by the unit vector $\mathbf{e}_r=\mathbf{r}/r$. Thus (A5) does not contain special components of the vector $\mathbf{b}$ and only spherical harmonics with $m=0$ remain which are independent on the position of the x axis. Notice that expressions like (A3) and (A4) which contain two spherical harmonics with the same magnetic quantum number m also do not depend on the choice of the x axis.

We have similar results for the $p_{3/2}$ state. We have to add a factor 2 in (A3) and to change the sign in (A4) and (A5).

Evaluating the matrix element for the $2p_{1/2}$ state we can distinguish between three types of terms according to matrix elements of (A3), (A4), and (A5), respectively. At first we get terms which correspond to (III)–(VII) of (30) for the K edge:

$$\begin{aligned} &+\frac{1}{3m_e}\sum_{m}\langle Y_{1m}|\epsilon'^{*}\cdot\mathbf{p}'G^{(s,d)}\mathbf{p}\cdot\epsilon|Y_{1m}\rangle \,, \\ &+\frac{1}{3m_e}\sum_{m}\langle Y_{1m}|\epsilon'^{*}\cdot\mathbf{p}'(\mathbf{q}'\cdot\mathbf{r}')G^{(f)}(\mathbf{q}\cdot\mathbf{r})\mathbf{p}\cdot\epsilon|Y_{1m}\rangle \,, \\ &-\frac{\hbar\omega}{6m_e c}\sum_{m}\{\,\langle Y_{1m}|\epsilon'^{*}\cdot\mathbf{p}'G_s^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|Y_{1m}\rangle b_z \\ &\qquad\qquad - b_z'^{*}\langle Y_{1m}|(\mathbf{q}'\cdot\mathbf{r}')G_s^{(s,d)}\mathbf{p}\cdot\epsilon|Y_{1m}\rangle\} \,, \\ &+\frac{(\hbar\omega)^2}{12m_e c^2}\sum_{m}\mathbf{b}'^{*}\cdot\mathbf{b}\langle Y_{1m}|\mathbf{q}'\cdot\mathbf{r}'G^{(s,d)}\mathbf{q}\cdot\mathbf{r}|Y_{1m}\rangle \,, \\ &-\frac{(\hbar\omega)^2}{12m_e c^2}\sum_{m} i\mathbf{e}_z\cdot(\mathbf{b}'^{*}\times\mathbf{b})\langle Y_{1m}|\mathbf{q}'\cdot\mathbf{r}'G_s^{(s,d)}\mathbf{q}\cdot\mathbf{r}|Y_{1m}\rangle \,. \end{aligned} \tag{A6}$$

We wrote $(\hat{\mathbf{q}}\times\epsilon)=\mathbf{b}$ and $(\hat{\mathbf{q}}'\times\epsilon'^{*})=\mathbf{b}'^{*}$. In comparison with the result for the $1s$ level (30) we have the sum over the magnetic quantum number of a p state and a factor $\frac{1}{3}$ appears. The radial part of the wave functions is not noticed explicitly. Clearly we get contributions from the unoccupied s and d states instead of from p states.

The second group of terms result from matrix elements of σ_z (A4). They differ from (A6) in the sign and by a factor m according to the difference between (A3) and (A4) and by interchanging G and G_s according to the two different contributions in (29):

$$
\begin{aligned}
&-\frac{1}{3m_e}\sum_m m\langle Y_{1m}|\boldsymbol{\epsilon}'^{*}\cdot\mathbf{p}'G_s^{(s,d)}\mathbf{p}\cdot\boldsymbol{\epsilon}|Y_{1m}\rangle\ ,\\
&-\frac{1}{3m_e}\sum_m m\langle Y_{1m}|\boldsymbol{\epsilon}'^{*}\cdot\mathbf{p}'(\mathbf{q}'\cdot\mathbf{r}')G_s^{(f)}(\mathbf{q}\cdot\mathbf{r})\mathbf{p}\cdot\boldsymbol{\epsilon}|Y_{1m}\rangle\ ,\\
&+\frac{\hbar\omega}{6m_e c}\sum_m m\{\langle Y_{1m}|\boldsymbol{\epsilon}'^{*}\cdot\mathbf{p}'G^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|Y_{1m}\rangle b_z-b_z'^{*}\langle Y_{1m}|(\mathbf{q}'\cdot\mathbf{r}')G^{(s,d)}\mathbf{p}\cdot\boldsymbol{\epsilon}|Y_{1m}\rangle\}\ ,\\
&-\frac{(\hbar\omega)^2}{12m_e c^2}\sum_m m\,\mathbf{b}'^{*}\cdot\mathbf{b}\langle Y_{1m}|\mathbf{q}'\cdot\mathbf{r}'G_s^{(s,d)}\mathbf{q}\cdot\mathbf{r}|Y_{1m}\rangle\ ,\\
&-\frac{(\hbar\omega)^2}{12m_e c^2}\sum_m mi\mathbf{e}_z\cdot(\mathbf{b}'^{*}\times\mathbf{b})\langle Y_{1m}|\mathbf{q}'\cdot\mathbf{r}'G^{(s,d)}\mathbf{q}\cdot\mathbf{r}|Y_{1m}\rangle\ .
\end{aligned}
\tag{A7}
$$

The third group of terms contains spherical harmonics with different m values. They are recalculated according to (A5) using the unit vector $\mathbf{e}_r$ to avoid defining an $\mathbf{x}$ axis:

$$
\begin{aligned}
&\frac{\hbar\omega}{6m_e c}\sqrt{3}\{-\langle Y_{10}|\boldsymbol{\epsilon}'^{*}\cdot\mathbf{p}'G^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|\mathbf{e}_r Y_{00}\rangle\cdot i(\mathbf{e}_z\times\mathbf{b})+i(\mathbf{e}_z\times\mathbf{b})\cdot\langle Y_{00}\mathbf{e}_r'|\boldsymbol{\epsilon}'^{*}\cdot\mathbf{p}'G^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|Y_{10}\rangle\\
&\qquad-\langle Y_{10}|\boldsymbol{\epsilon}'^{*}\cdot\mathbf{p}'G_s^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|\mathbf{e}_r Y_{00}\rangle\cdot\mathbf{b}+\mathbf{b}\cdot\langle Y_{00}\mathbf{e}_r'|\boldsymbol{\epsilon}'^{*}\cdot\mathbf{p}'G_s^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|Y_{10}\rangle\\
&\qquad-\langle Y_{10}|(\mathbf{q}'\cdot\mathbf{r}')G^{(s,d)}\mathbf{p}\cdot\boldsymbol{\epsilon}|\mathbf{e}_r Y_{00}\rangle\cdot i(\mathbf{e}_z\times\mathbf{b}'^{*})+i(\mathbf{e}_z\times\mathbf{b}'^{*})\cdot\langle Y_{00}\mathbf{e}_r'|(\mathbf{q}'\cdot\mathbf{r}')G^{(s,d)}\mathbf{p}\cdot\boldsymbol{\epsilon}|Y_{10}\rangle\\
&\qquad-\langle Y_{10}|(\mathbf{q}'\cdot\mathbf{r}')G_s^{(s,d)}\mathbf{p}\cdot\boldsymbol{\epsilon}|\mathbf{e}_r Y_{00}\rangle\cdot\mathbf{b}'^{*}+\mathbf{b}'^{*}\cdot\langle Y_{00}\mathbf{e}_r'|(\mathbf{q}'\cdot\mathbf{r}')G_s^{(s,d)}\mathbf{p}\cdot\boldsymbol{\epsilon}|Y_{10}\rangle\}\ ,\\
&\frac{(\hbar\omega)^2}{12m_e c^2}\sqrt{3}\{-b_z'^{*}i(\mathbf{e}_z\times\mathbf{b})\cdot\langle Y_{00}\mathbf{e}_r'|(\mathbf{q}'\cdot\mathbf{r}')G_s^{(s,d)(}\mathbf{q}\cdot\mathbf{r})|Y_{10}\rangle-b_z i(\mathbf{e}_z\times\mathbf{b}'^{*})\cdot\langle Y_{00}\mathbf{e}_r'|(\mathbf{q}'\cdot\mathbf{r}')G_s^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|Y_{10}\rangle\\
&\qquad+\langle Y_{10}|(\mathbf{q}'\cdot\mathbf{r}')G_s^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|\mathbf{e}_r Y_{00}\rangle\cdot i(\mathbf{e}_z\times\mathbf{b})b_z'^{*}+\langle Y_{10}|(\mathbf{q}'\cdot\mathbf{r}')G_s^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|\mathbf{e}_r Y_{00}\rangle\cdot i(\mathbf{e}_z\times\mathbf{b}'^{*})b_z\\
&\qquad+\langle Y_{10}|(\mathbf{q}'\cdot\mathbf{r}')G^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|\mathbf{e}_r Y_{00}\rangle\cdot[(\mathbf{b}'^{*}\times\mathbf{b})\times\mathbf{e}_z]-[(\mathbf{b}'^{*}\times\mathbf{b})\times\mathbf{e}_z]\cdot\langle Y_{00}\mathbf{e}_r'|(\mathbf{q}'\cdot\mathbf{r}')G^{(s,d)}(\mathbf{q}\cdot\mathbf{r})|Y_{10}\rangle\}\ .
\end{aligned}
\tag{A8}
$$

The results for the $2p_{3/2}$ core state are similar and we have to add a factor 2 in (A6), because all these terms result from a matrix element of type (A3). We have to change the sign in (A7) and in (A8), because these terms result from matrix elements of type (A4) and (A5), respectively.

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