Origin of magnetic dichroism in angular-resolved photoemission from ferromagnets

N. A. Cherepkov
State Academy of Aerospace Instrumentation, 190 000 St. Petersburg, Russia
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It is shown that recently discovered effects of magnetic x-ray dichroism in angular-resolved photoemission from core levels of ferromagnets appear due to the spin-orbit and exchange splitting of core levels. The effects are proportional to the state multipoles characterizing the alignment and orientation of the hole levels, and can be observed with circularly polarized, linearly polarized, and unpolarized light. Possible applications of these effects in investigations of magnetic structures and adsorbates are pointed out.

Recently, it was discovered experimentally that the shape of photoelectron spectra from core levels of magnetized Fe is changed when the direction of magnetization is reversed. The effects were observed for both circularly[1] and linearly[2–4] polarized light. These effects are evidently connected with the local magnetic field acting upon the substrate atoms, and with the spin-orbit and exchange splitting of core levels. In this respect they are similar to the well-known Kerr and Faraday effects observed in optical absorption or reflectance. New effects were discovered due to appearance of synchrotron-radiation sources which produce intense and highly polarized radiation in the soft-x-ray region.

In the first experiment Baumgarten *et al.*[1] used the grazing incidence of circularly polarized radiation with respect to the surface, so that photon spin and direction of magnetization were nearly parallel. Electrons were collected at the angle 55°. They observed that the shape and intensity of photoelectron spectra from $2p_{1/2}$ and $2p_{3/2}$ levels of Fe depend on the relative orientation of photon spin and sample magnetization. The difference of two intensities for two opposite directions of magnetization we will call circular magnetic dichroism in the angular distribution (CMDAD). Roth *et al.*[2,3] and Sirotti and Rossi[4] observed a similar effect but with linearly polarized light. Again, they used grazing incidence of radiation, and collected photoelectrons emitted in the direction of the surface normal. The direction of photon polarization was either perpendicular or parallel to the surface, and the direction of magnetization was either perpendicular or parallel (approximately) to the photon beam. It was shown that depending on the relative orientation of the light polarization and the direction of sample magnetization, the shape of the $3p$ line of Fe is changed. The difference between photoelectron intensities for two opposite directions of magnetization, which we call, following Ref. 2, linear magnetic dichroism in the angular distribution (LMDAD), was observed when the direction of magnetization **M** was perpendicular to both direction of the photon beam **q** and the photon polarization **e**, that is for p-polarized light. For s-polarized light LMDAD is zero, but the line shapes are different for $\mathbf{M}\perp\mathbf{e}$ and $\mathbf{M}\parallel\mathbf{e}$. In Ref. 2 also the spin polarization of photoelectrons has been measured, but this problem is more complicated and will not be discussed here.

The theoretical explanation of CMDAD, similar in essence with the present one but formulated less transparently, was given in Ref. 5, while LMDAD remains to our knowledge unexplained. The analogous effects in angular integrated spectra have been considered in Ref. 6. For qualitative understanding of the observed effects in all publications[1–4] the analogy with the well understood spin polarization effects in atomic photoionization[7] has been used. But though this analogy can give some hints, it is not appropriate for the problem under consideration, and in some cases leads to wrong conclusions.

The aim of this paper is to show that all effects observed in Refs. 1–4 can be successfully explained if we accept the pure atomic description of the photoemission process, provided the splitting of the hole state due to the exchange interaction with the $3d$ subshell is taken into account. The applicability of this model is justified because in all measurements the photoelectron energy was high enough, ≥ 40 eV. In ferromagnets the hole state with a given total angular momentum j due to the exchange interaction is split into components with a given projection m_j. In other words, each component of the final ionic state is polarized. It is easy to show (as it was shown in Ref. 8 that the spin polarization effects are identical for one-electron and closed subshells), that the photoionization of a closed subshell in the situation when the final hole state is polarized, is exactly equivalent to the photoionization of a one-electron subshell which is initially polarized. The latter problem has been considered in Refs. 9–11, and we will directly use below the equations obtained in Ref. 10. The polarization of the hole state will be described, following Ref. 10, by state mul-

TABLE I. State multipoles ρ^{n}_{N0} for magnetic sublevels of $np_{1/2}$ and $np_{3/2}$ states.

	$j=1/2$		$j=3/2$			
m_j	$\frac{1}{2}$	$-\frac{1}{2}$	$\frac{3}{2}$	$\frac{1}{2}$	$-\frac{1}{2}$	$-\frac{3}{2}$
ρ^{n}_{00}	$\frac{1}{\sqrt{2}}$	$\frac{1}{\sqrt{2}}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$	$\frac{1}{2}$
ρ^{n}_{10}	$\frac{1}{\sqrt{2}}$	$-\frac{1}{\sqrt{2}}$	$\frac{3}{2\sqrt{5}}$	$\frac{1}{2\sqrt{5}}$	$-\frac{1}{2\sqrt{5}}$	$-\frac{3}{2\sqrt{5}}$
ρ^{n}_{20}	0	0	$\frac{1}{2}$	$-\frac{1}{2}$	$-\frac{1}{2}$	$\frac{1}{2}$
ρ^{n}_{30}	0	0	$\frac{1}{2\sqrt{5}}$	$-\frac{3}{2\sqrt{5}}$	$\frac{3}{2\sqrt{5}}$	$-\frac{1}{2\sqrt{5}}$

tipoles, as they have been defined in Ref. 12. For the particular case of a p subshell the values of the state multipoles $\rho^{\mathbf{n}}_{N0}$ are given in Table I for each magnetic sublevel.

We shall use in the following the unit vectors $\mathbf{n}$, $\boldsymbol{\kappa}$, and $\mathbf{q}$ to define the directions of the sample magnetization, the electron ejection and the photon beam, respectively. The coordinate system and the light polarizations are defined in Fig. 1. From Eq. (10) of Ref. 10 it follows that in the case when the spin-orbit interaction in continuous spectrum is neglected, the parameters C^j_{kLN} with $N>2$ for p shells are zero, therefore we will restrict the consideration here by terms with $N\leq 2$.

Using the definition of the angular distribution of photoelectrons given by Eq. (7) of Ref. 10, we obtain the following general expression for LMDAD:

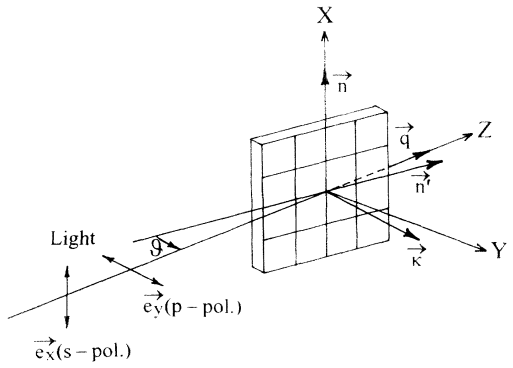

FIG. 1. Definition of the geometry of experiments and of the coordinate system.

$$I_j^{\mathrm{LMDAD}}(\boldsymbol{\kappa},\mathbf{n})=I_j(\boldsymbol{\kappa},\mathbf{n})-I_j(\boldsymbol{\kappa},-\mathbf{n})$$
$$=\frac{\sigma_{nlj}(\omega)}{2\pi}\cdot\frac{3i}{2}C^j_{221}(2j+1)^{1/2}\rho^{\mathbf{n}}_{10}$$
$$\times\{(\boldsymbol{\kappa}\cdot\mathbf{q})(\mathbf{q}\cdot[\boldsymbol{\kappa}\times\mathbf{n}])\pm[(\boldsymbol{\kappa}\cdot\mathbf{q})(\kappa_x n_y+\kappa_y n_x)-2(\mathbf{n}\cdot\mathbf{q})\kappa_x\kappa_y]\}\ . \tag{1}$$

Here the upper and lower signs refer to the s- and p-polarized light, respectively, $\sigma_{nlj}(\omega)$ is the partial photoionization cross section, and C^j_{221} is the dimensionless parameter like the angular asymmetry parameter β. For s-polarized light Eq. (1) gives a zero result for both directions of magnetization $\mathbf{n}$ and $\mathbf{n}'$ in Fig. 1, in accord with measurements.[3] For p-polarized light and $\mathbf{n}\perp\mathbf{q}$ we have

$$I_j^{\mathrm{LMDAD}}=\frac{\sigma_{nlj}}{2\pi}3iC^j_{221}(2j+1)^{1/2}\rho^{\mathbf{n}}_{10}\sin\vartheta\cos\vartheta\ . \tag{2}$$

ϑ is the angle of the grazing incidence of light. It is proportional to the state multipole $\rho^{\mathbf{n}}_{10}$ which defines the orientation of the hole state.

In general the parameters C^j_{kLN} (Ref. 10) have the same sign for the states with $j=l+\frac{1}{2}$ and $j=l-\frac{1}{2}$. In the particular case of a p subshell and $N=1$, the parameters $C^{3/2}_{kL1}$ and $C^{1/2}_{kL1}$ are nearly equal:

$$C^{3/2}_{kL1}=\frac{\sqrt{5}}{2}C^{1/2}_{kL1}\ . \tag{3}$$

Therefore, the relative sign and the magnitude of LMDAD (2) for different magnetic sublevels is defined by the sign and the magnitude of the state multipoles $\rho^{\mathbf{n}}_{10}$. As it follows from Table I, LMDAD has a different sign for sublevels with the different sign of m_j. So, measurements of LMDAD enables one to distinguish between components of the hole state with different signs of projection m_j. The curves for LMDAD obtained in Refs. 2–4 changes sign at some point, which gives the middle of the $3p_{3/2}$ multiplet. The $3p_{1/2}$ doublet is evidently masked by a larger contribution of the $3p_{3/2}$ levels.

The next value which has been measured in Ref. 3 and remained unexplained, is the difference between photoelectron spectra obtained with s-polarized light when the direction of magnetization is changed from $\mathbf{n}$ to $\mathbf{n}'$ (see Fig. 1). Again from Eq. (7) of Ref. 10 one can derive the following expression for this difference:

$$I_j^{\perp}\equiv[I_j(\boldsymbol{\kappa},\mathbf{n})-I_j(\boldsymbol{\kappa},\mathbf{n}')]_{\mathbf{k}\perp\mathbf{n}}$$
$$=-\frac{\sigma_{nlj}(\omega)}{4\pi}(2j+1)^{1/2}\rho^{\mathbf{n}}_{20}$$
$$\times\left[\frac{3}{\sqrt{2}}C^j_{202}+3\sqrt{5/7}C^j_{222}+\frac{3}{2\sqrt{7}}C^j_{242}\right]\ . \tag{4}$$

It is worthwhile to note that this value does not depend on the angle of the light incidence ϑ, which is a simple consequence of the fact that the dipole photoeffect is defined by the polarization vector ($\mathbf{e}_x$ in this case), and not by the photon momentum $\mathbf{q}$. The variation of this value within a given multiplet is completely defined by the alignment tensor $\rho^{\mathbf{n}}_{20}$ because all other values in (4) are constants. The $p_{1/2}$ state could not be aligned, therefore $\rho^{\mathbf{n}}_{20}$ for this state is zero. So, only the $3p_{3/2}$ multiplet contributes to the curve measured in Ref. 3, and as it follows from Table I, the components with $|m_j|=\frac{1}{2}$ and $|m_j|=\frac{3}{2}$ give the contributions of the opposite sign and of the same magnitude. Figure 2 shows the result of the simplest possible simulation of the points observed in Ref. 3 for the $3p_{3/2}$ multiplet of Fe by four equidistant Lorentzians of equal magnitudes and widths, and of the signs defined by the state multipoles $\rho^{\mathbf{n}}_{20}$. The agreement with experiment is rather good, and from the figure the following positions for four magnetic sublevels can be deduced: 51.77, 52.13, 52.50, and 52.86 eV. The change of sign of the LMDAD curve [Fig. 3(c) of Ref. 3] occurs at 52.3 eV,

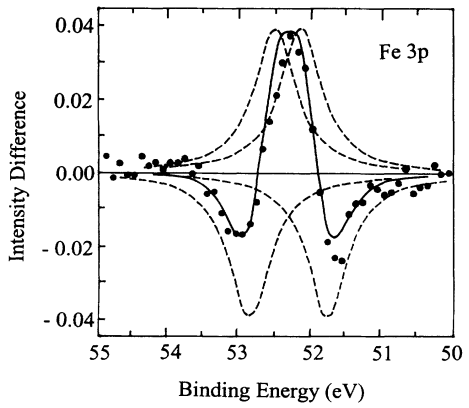

FIG. 2. The difference between two photoelectron spectra corresponding to two directions of magnetization $\mathbf{n}$ and $\mathbf{n}'$ in Fig. 1, for s-polarized light. Full line: the simulation of this difference by four Lorentzians shown by dashed lines; points: experiment (Ref. 3).

i.e., exactly in the middle of this multiplet, in accord with the conclusion made above. A similar fit of the $3p$ photoemission spectra made in Ref. 4 gives somewhat different positions of four equidistant magnetic sublevels, and also unequal amplitudes. The introduction of unequal amplitudes into our fit can improve the agreement with the experiment. The structure observed in LMDAD in Refs. 3 and 4 extends to higher-binding energies, which is connected with the contribution of the $3p_{1/2}$ states.

Finally, let us consider CMDAD. From Eq. (7) of Ref. 10 the following expression is obtained:

$$I_j^{\mathrm{CMDAD}}(\boldsymbol{\kappa},\mathbf{n})=\frac{\sigma}{2\pi}(2j+1)^{1/2}\rho_{10}^{\mathbf{n}} \times\{\mp\sqrt{3/2}C_{101}^{j}(\mathbf{n}\cdot\mathbf{q}) \pm\sqrt{3}C_{121}^{j}[\tfrac{3}{2}(\boldsymbol{\kappa}\cdot\mathbf{q})(\boldsymbol{\kappa}\cdot\mathbf{n})-\tfrac{1}{2}(\mathbf{n}\cdot\mathbf{q})] +\tfrac{3}{2}iC_{221}^{j}(\boldsymbol{\kappa}\cdot\mathbf{q})(\mathbf{q}\cdot[\boldsymbol{\kappa}\times\mathbf{n}])\}\ , \tag{5}$$

where the upper and lower signs refer to the left and right circularly polarized light. For the particular geometry used in Ref. 1(a) (the magnetization direction $\mathbf{n}'$ in Fig. 1), we have from (5)

$$I_j^{\mathrm{CMDAD}}=\mp\frac{\sigma}{2\pi}(2j+1)^{1/2}\rho_{10}^{\mathbf{n}} \times\left[\sqrt{3/2}C_{101}^{j}+\frac{\sqrt{3}}{2}C_{121}^{j}\right]\cos\vartheta\ . \tag{6}$$

In general, CMDAD (5) is not equal to CDAD, which is the difference between photoelectron currents for opposite light polarizations and the fixed direction of magnetization (due to the last term in (5), which in the case of CDAD is substituted by another term with the coefficient C_{122}^{j}). But for the geometry used in Ref. 1(a), CMDAD and CDAD are equal, as it was also proved experimentally.

From (6) it follows that for the geometry under consideration CMDAD, like LMDAD, is defined by the state multipole $\rho_{10}^{\mathbf{n}}$, therefore the qualitative information which can be obtained from the measurements of these two values, is exactly the same. There is one interesting point which should be stressed. CMDAD has been measured in Ref. 1 for the $2p$ level of Fe where the $2p_{1/2}$ and $2p_{3/2}$ sublevels are well separated in energy. It was observed that, coming from low-binding energies, CMDAD changes sign from positive to negative at the $2p_{3/2}$ line, and in the opposite direction at the $2p_{1/2}$ line. Since the parameters C_{101}^{j} and C_{121}^{j} are nearly equal for the $j=\frac{3}{2}$ and $j=\frac{1}{2}$ levels, and the state multipoles $\rho_{10}^{\mathbf{n}}$ for both $j=\frac{3}{2}$ and $j=\frac{1}{2}$ levels have the same sign as the projection m_j, this observation can be considered as experimental proof that the magnetic sublevels for the $2p_{1/2}$ and $2p_{3/2}$ levels have reverse ordering. This result has been obtained also theoreticaly in Ref. 5. From the values of the $\rho_{10}^{\mathbf{n}}$ state multipoles and relation (3) for the C_{101}^{j} and C_{121}^{j} parameters it also follows that the square under the CMDAD curve at the $p_{3/2}$ level should be four times larger then at the $p_{1/2}$ level, which is approximately fulfilled in the experimental observation [see Fig. 1(b) of Ref. 1(a)]. From (5) it also follows that CMDAD exists for a "forbidden geometry" considered in Ref. 1(b) when $\mathbf{n}\perp\mathbf{q}$.

So, we have shown that all effects of magnetic dichroism in the angular-resolved photoemission from magnetized Fe observed in Refs. 1–4, have mainly an atomic origin and are connected with the existence of the energy splitting of core levels on projections of the total angular momentum m_j. This splitting appears due to the exchange interaction with the open $3d$ subshell, and it can be smaller than the spin-orbit splitting, as is the case in the Fe $2p$ subshell, or of the same order of magnitude, as it is evidently the case in the Fe $3p$ subshell. In both cases the widths of magnetic sublevels are comparable to or larger than their energy splitting, therefore they, in principle, could not be resolved by the methods of ordinary photoelectron spectroscopy. Effects of magnetic dichroism in the angular-resolved photoemission give the unique opportunity to resolve them quite unambiguously, and it can be done from a rather simple analysis of the experimental data. Comparison of numerical calculations performed with atomic wave functions (which are now in progress[13]) with experimental data, will give more exact information on the applicability of the simple atomic model and on the contribution of the solid state effects.

From the theory presented above it follows that measurements with linearly polarized light are quite sufficient to obtain all the information on the levels structure, so that measurements with circularly polarized light, which are usually more complicated, can be avoided. Moreover, the same information can be obtained even from measurements with unpolarized light. MDAD for unpolarized light is defined by the first term in the square brackets in (1) [or by the last term in (5)], which has the same sign for two orthogonal light polarizations. The value $I_j^{\perp}$ for unpolarized light is also different from zero and is defined by more complicated equations as compared to (4). For another geometry of experiment $I_j^{\perp}$ has been considered in Ref. 14. Thus, there are many possibilities to obtain the same information which has been obtained

from the experiments performed in Refs. 1–4.

From the above analysis it follows that the state multipoles ρ_{30}^{n} do not contribute to the effects of magnetic dichroism as long as the spin-orbit interaction in the continuous spectrum is small. From the preliminary analysis it follows that ρ_{30}^{n} will give the contribution of the order of unity to the spin polarization of photoelectrons in the experiment like that performed in Ref. 2. Higher state multipoles will also contribute in subshells with $l > 1$, for example, in the valence band of Fe.[15]

The theory presented here can be applied to any situation where the final ionic state is polarized. In particular, it is fully applied to the photoemission from atoms adsorbed at a surface. It is well known that the *np* hole states of rare-gas atoms adsorbed at a surface have an additional splitting into the $|m_j| = \frac{3}{2}$ and $|m_j| = \frac{1}{2}$ sublevels,[16] that is they are aligned. Therefore for these atoms LDAD and CDAD can be investigated, as is proposed in Ref. 11. In the case of magnetic surfaces adsorbed atoms will be not only aligned but also oriented, and the consideration of this paper will be fully applied. In general, methods discussed in this paper can be used in all cases where the local magnetic or electric field causes the additional splitting of atomic levels. Then MDAD, LDAD, and CDAD effects will be a sensitive probe of this splitting, and thereby of the local field.

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[1](a) L. Baumgarten, C. M. Schneider, H. Petersen, F. Schäfers, and J. Kirschner, Phys. Rev. Lett. **65**, 492 (1990); (b) C. M. Schneider, D. Venus, and J. Kirschner, Phys. Rev. B **45**, 5041 (1992).

[2]Ch. Roth, F. U. Hillebrecht, H. B. Rose, and E. Kisker, Phys. Rev. Lett. **70**, 3479 (1993).

[3]Ch. Roth, H. B. Rose, F. U. Hillebrecht, and E. Kisker, Solid State Commun. **86**, 647 (1993).

[4]F. Sirotti and G. Rossi, Phys. Rev. B **49**, 15 682 (1994); G. Rossi, F. Sirotti, N. A. Cherepkov, F. Combet Farnoux, and G. Panaccione, Solid State Commun. **90**, 557 (1994).

[5]H. Ebert, L. Baumgarten, C. M. Schneider, and J. Kirschner, Phys. Rev. B **44**, 4406 (1991).

[6]B. T. Thole and G. Van der Laan, Phys. Rev. B **44**, 12 424 (1991); G. Van der Laan and B. T. Thole, *ibid.* **48**, 210 (1993).

[7]N. A. Cherepkov, Adv. At. Mol. Phys. **19**, 395 (1983).

[8]N. A. Cherepkov, Sov. Phys. JETP **38**, 463 (1974).

[9]H. Klar and H. Kleinpoppen, J. Phys. B **15**, 933 (1982).

[10]N. A. Cherepkov and V. V. Kuznetsov, J. Phys. B **22**, L405 (1989).

[11]N. A. Cherepkov, V. V. Kuznetsov, and V. A. Verbitskii, J. Phys. B (to be published).

[12]K. Blum, *Density Matrix Theory and Application* (Plenum, New York, 1981).

[13]N. A. Cherepkov and F. Combet Farnoux (unpublished).

[14]N. A. Cherepkov and V. V. Kuznetsov, J. Phys. B **25**, L445 (1992).

[15]J. Bansmann, C. Westphal, M. Getzlaff, F. Fegel, and G. Schönhense, J. Magn. Magn. Mater. **104-107**, 1691 (1992).

[16]U. Heinzmann and G. Schönhense, *Polarized Electrons in Surface Physics*, edited by R. Feder (World Scientific, Singapore, 1985), p. 467.