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Magnetic linear dichroism and spin polarization in 3*d*-band photoemission

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Spin-orbit effects in normal photoemission from ferromagnetic cubic (001) surfaces with in-plane magnetization $\vec{M}$ have been studied by symmetry considerations and by relativistic one-step-model Green-function calculations for d-band emission from Ni(001). Spin-averaged spectra due to s-polarized light in two orthogonal states show an intensity asymmetry, i.e., magnetic linear dichroism (MLD). For off-normally-incident p-polarized light, there is another type of MLD, which is maximal for $\vec{M}$ normal, and zero for $\vec{M}$ parallel to the plane of incidence. In the latter case, the electron spin polarization $\vec{P}$ has components exclusively due either to spin-orbit coupling or exchange. Individual spectral features are explained by the relativistic bulk band structure and by analytical considerations.

The interplay between exchange interaction and spin-orbit coupling in ferromagnets in conjunction with electromagnetic radiation is well known to produce a variety of observable effects. These include firstly the magneto-optical effects in the visible and infrared light regime (cf., e.g., Refs. 1 and 2, and references therein). In circularly polarized x-ray absorption, an intensity asymmetry upon helicity reversal, i.e., a "magnetic circular dichroism" (MCD) was predicted by theory,[3] verified experimentally[4] and subsequently fruitfully pursued in numerous studies (see, e.g., Refs. 5–7, and references therein). In photoemission (see, e.g., Refs. 8 and 9, and references therein), related effects are the intensity asymmetries upon reversal of the magnetization known as MCD and MLD (where MLD denotes magnetic linear dichroism). Experimentally, both have been observed in emission from core levels (see, e.g., Ref. 10, and references therein), and MCD also from valence states.[11,12] Theoretical photoemission studies have been made in a many-body framework,[13] describing MCD for core and valence levels, and using an effective single-particle relativistic multiple-scattering formalism to calculate MCD and MLD from core levels[14,15] and MCD from Ni(001) valence states.[16]

In this paper, we present a theoretical study of MLD and further spin-orbit coupling effects in normal photoemission from ferromagnetic cubic (001) surfaces with in-plane magnetization. We first employ symmetry arguments to obtain qualitative insight into both core and valence emission. Subsequently, we present numerical d-band photoemission results for the prototype ferromagnetic surface Ni(001).

As has been demonstrated for nonmagnetic surfaces,[17] qualitative information on the intensity I and the spin polarization vector $\vec{P}$ of photoelectrons can be obtained by symmetry arguments. This holds for both valence-band and core-level photoemission. Under a spatial symmetry transformation S of the total system (crystal, magnetization $\vec{M}$, incident light, electron detection direction), $\vec{P}$ is invariant. For various S this dictates that some components P_k, where $k=x$, y, z, vanish. In the absence of a suitable S, P_k is generally nonzero.

We now apply this to photoemission normal to a ferromagnetic cubic (001) semi-infinite crystal or adsorbate system with $\vec{M}$ along the x axis, i.e., parallel to the surface plane (x,y). The x axis is either along the [100] or the [110] direction. $\vec{M}$ reduces the spatial symmetry from C_{4v} to only one mirror plane, (y,z). The photon field is, in the electric dipole approximation, sufficiently characterized by its magnetic vector potential $\vec{A}$.

First consider normally incident s-polarized light with $\vec{A}$ along x or y. As there is no spatial transformation between these two cases, which would leave $\vec{M}$ unaltered, the intensities I of the photocurrent are, in general, different from each other, i.e., there is MLD in the sense that an observable quantity has different values for the two orthogonal linear photon states. Since reflection at the (y,z) plane is a symmetry operation, P_y and P_z must vanish, leaving only P_x along $\vec{M}$.

For p-polarized light at off-normal incidence, there are two different cases. First, take $\vec{A}=(0,A_y,A_z)$, i. e., $\vec{M}$ normal to the plane of incidence. The (y,z) mirror reflection implies $P_y=P_z=0$, leaving only P_x nonzero, as in the absence of spin-orbit coupling. Since a finite P_x is also produced by spin-orbit coupling alone (i.e., in the limit $\vec{M}\rightarrow 0$),[17] the actual value of P_x for the ferromagnetic system must be due to the joint action of magnetic exchange and spin-orbit coupling. Reflection at the (x,z) plane, which is not a symmetry operation but changes A_y into $-A_y$ and reverses the direction of $\vec{M}$, implies $I(p_+,\vec{M})=I(p_-,-\vec{M})$, where p_+ and p_- label the two directions of $\vec{A}$, i.e., light incidence at the same polar angle but at azimuthal angles 0° and 180°. Since there is no spatial transformation from p_+ to p_-, which leaves $\vec{M}$ unchanged, we have further $I(p_+,\vec{M}) \neq I(p_-,\vec{M})=I(p_+,-\vec{M})$, i.e., there is a MLD, which is observable either by keeping $\vec{M}$ fixed and changing p_+ into p_- or by reversing $\vec{M}$ for fixed p_+. The magnetic origin of this effect is confirmed by considering the limit $\vec{M}\rightarrow\vec{0}$. Reflection at the (x,z) plane then implies $I(p_+)=I(p_-)$.

In the second case with p-polarized light, $\vec{A}=(A_x,0,A_z)$, i.e., $\vec{M}$ is parallel to the plane of incidence. From the (y,z) and the (x,z) reflections we now obtain $I(p_+,\vec{M})=I(p_-,\vec{M})$ and $I(p_\pm,\vec{M})=I(p_\pm,-\vec{M})$, i.e., there is no MLD. Since there is no symmetry operation for the total setup, all components of $\vec{P}$ are nonzero, with $P_x(p_+,\vec{M}) = P_x(p_-,\vec{M})$ and $P_{y,z}(p_+,\vec{M}) = -P_{y,z}(p_-,\vec{M})$. For $\vec{M}=\vec{0}$, only $P_y \neq 0$ (cf. Ref. 17), while in the absence of spin-orbit coupling only $P_x \neq 0$. P_z requires the simultaneous presence of ferromagnetism and spin-orbit coupling.

For a quantitative investigation of the above phenomena we have chosen the prototype ferromagnetic surface Ni(001). Spin-resolved photoemission spectra and the corresponding bulk band structure were calculated within fully relativistic Green-function theory for ferromagnetic crystalline surface systems.[18] The effective quasiparticle potential is taken in the muffin-tin shape approximation with a spin-dependent bulk potential. Since for Ni this leads to exchange splittings of about 0.6 eV as opposed to an average value of about 0.3 eV observed in photoemission experiments, we modified it by an *ad hoc* spin-dependent self-energy correction reducing the splitting between the majority- and minority-spin potentials by a factor 0.5. For the uniform inner potential, which is complex and energy dependent, we use—in the absence of realistic first-principles self-energy calculations—as real parts 15 eV for the initial and 13 eV for the final states in view of matching calculated photoemission peak positions with their experimental counterparts.[19] We note that a decrease of the real self-energy part with increasing energy is in qualitative accordance with electron gas results and has been employed successfully in a number of previous photoemission studies (see, e.g., Refs. 20 and 21, and references therein). For the imaginary part of the inner potential we adopt energy-dependent forms increasing (in absolute value) away from the Fermi energy E_F as $0.025(E-E_F)$ for the lower states and as $0.04(E-E_F)^{1.25}$ for the upper states. These forms are qualitatively plausible, since the inelastic electron (hole) scattering cross section is 0 at E_F and increases with energy above (below) E_F. Further below E_F, an approximately linear behavior has been found in many-body model calculations for Ni.[22] Strictly, our above forms should be modified to reproduce the quadratic behavior on both sides very close to E_F, which is required by Fermi liquid theory. Since the photoemission spectra calculated in the following exhibit sizable peaks only below about 0.4 eV and agree quite well with experimental data, such quadratic modification would, however, not affect our present results.

We now present normal-emission photoelectron spectra calculated at photon energy 21.22 eV for ferromagnetic Ni(001) with in-plane magnetization $\vec{M}$ (along [110]). Since the photoemission features can be qualitatively understood in terms of direct bulk interband transitions, let us first look at the relativistic band structure (bottom panel of Fig. 1). Spin-orbit effects on ferromagnetic band structures are well known: spin-up and spin-down bands hybridize with each other in a manner dependent upon the direction of the magnetization $\vec{M}$. Consequently, the spin polarization vector $\vec{P}=P\vec{P}_0$ (i.e., the expectation value of the spin operator) of the electron states is still characterized by the unit vector $\vec{P}_0 \| \vec{M}$, but the degree of spin polarization P may vary continuously between $+1$ and -1 along a given band.[23] While the actual values of P have been obtained in our calculations, we prefer for clarity's sake to distinguish the bands in Fig. 1 only by $P>0$ and $P<0$ as of predominant majority- and minority-spin type (solid and dashed lines). For the relevant

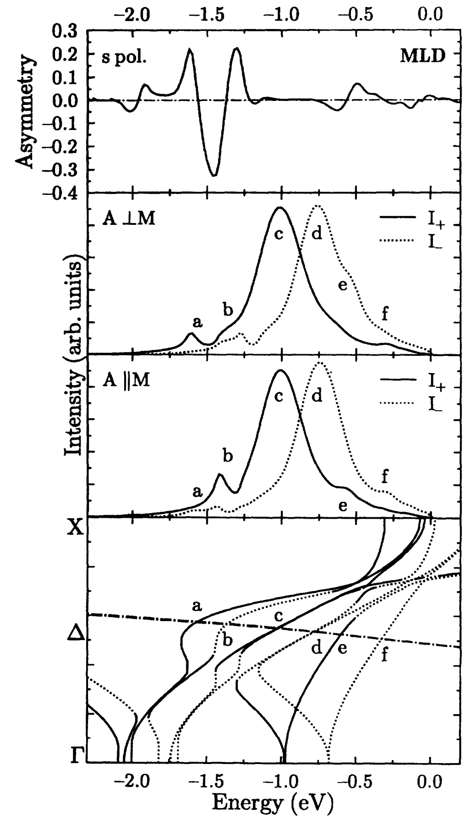

FIG. 1. Ferromagnetic Ni(001) with magnetization $\vec{M}\|[110]$ (parallel to the surface): Relativistic bulk band structure along [001] for occupied states with dominant majority- (solid lines) and minority-spin orientation (dotted lines) together with upper bands (of predominant spatial symmetry type Δ_6^1) shifted downward by the photon energy $\hbar\omega=21.22$ eV (bottom panel). Normal photoemission by *s-polarized light* with photon energy $\hbar\omega=21.22$ eV: spin-resolved spectra I_+ and I_- (with respect to the direction of $\vec{M}$) for vector potential $\vec{A}\perp\vec{M}$ and $\vec{A}\|\vec{M}$ (second and third panel) plus the asymmetry between the spin-averaged spectra of the latter two cases (i.e., magnetic linear dichroism) (top panel). The energy zero is the Fermi energy. Crossing points between lower and upper band structures and the corresponding peaks in the spectra are labeled $a-f$.

final state band (displaced downward by the photon energy 21.22 eV), the spin splitting is within the line thickness.

In view of understanding individual photoemission peaks and their spin polarization, we briefly elucidate the nature of the electronic half-space states (in the absence of an absorptive potential) in a simple manner (rather than in the formal terms of magnetic double-group theory[24]). Since a magnetization $\vec{M}$ exists parallel to the surface, the relevant point group is no longer C_{4v} (with a fourfold rotation axis normal to the surface), but the magnetic point group $\underline{2}\underline{m}m$. The underlining of 2 and m indicates that the twofold rotation about the surface normal and the mirror operation with respect to the plane, which is normal to the surface and parallel to $\vec{M}$, are combined with time reversal (which transforms $\vec{M}$ into $-\vec{M}$). In the presence of spin-orbit coupling, all electron states can be expressed as

$$|\Sigma^1\rangle|\sigma\rangle+|\Sigma^2\rangle|\bar{\sigma}\rangle+|\Sigma^3\rangle|\bar{\sigma}\rangle+|\Sigma^4\rangle|\sigma\rangle, \qquad (1)$$

where $\bar{\sigma}=-\sigma$ and $|\Sigma^i\rangle$ denotes a spatial part of the single-group symmetry type Σ^i associated with the nonmagnetic point group $2mm$ (i.e., C_{2v} in Schoenflies notation), and the Pauli spinors $|\sigma\rangle$ are aligned with respect to $\vec{M}$. The spin orientation of individual photoemission peaks is determined by electric dipole transition matrix elements $\langle i|\vec{A}\cdot\vec{x}|f_\sigma\rangle$, where the initial states $|i\rangle$ and the final states $|f_\sigma\rangle$ are expressed according to Eq. (1), with the latter having the form $|\Sigma^1\rangle|\sigma\rangle$ with $\sigma=+,-$ outside the crystal (at the detector). The nonvanishing partial matrix elements are easily identified, since $\vec{A}$ does not affect the Pauli spinors and couples the spatial parts $|\Sigma^k\rangle$, $k=1,\ldots,4$, subject to the usual nonrelativistic dipole selection rules.

For s-polarized light, our numerical calculations yield photoelectron spin polarization only along $\vec{M}$, in line with our above symmetry considerations. The resulting partial intensity spectra $I_\tau=I(1+\tau P_x)/2$, $\tau=\pm$ (shown in the two central panels of Fig. 1), are interpreted using the above states and matrix elements and recalling that A_x (A_y) couples spatial parts $|\Sigma^3\rangle$ ($|\Sigma^4\rangle$) and $|\Sigma^1\rangle$.

At point c, there are two degenerate initial states, one with a dominant part $|\Sigma^3\rangle|\sigma\rangle$ and the other with $|\Sigma^4\rangle|\sigma\rangle$. From the former (latter), a transition takes place into $|\Sigma^1\rangle|+\rangle$ for $\vec{A}\|\vec{M}$ ($\vec{A}\perp\vec{M}$), i.e., peak c is positively polarized in both cases. Analogously, peak d is negatively polarized. Thus, peaks c and d are essentially the same as in the absence of spin-orbit coupling.

Peaks a, b, e, and f, however, are brought about by spin-orbit coupling and depend strongly on the orientation of $\vec{A}$ relative to $\vec{M}$. At a, the initial state consists of $|\Sigma^1\rangle|+\rangle$ with an admixture $|\Sigma^4\rangle|+\rangle$ and a weaker one (due to the larger energetic separation) $|\Sigma^3\rangle|-\rangle$. The first admixture leads to a positive-spin peak a for $\vec{A}\perp\vec{M}$ and the second one to a weaker negative-spin peak a for $\vec{A}\|\vec{M}$. Similarly, a fairly strong positive-spin and a weak negative-spin peak b are expected for $\vec{A}\|\vec{M}$ and $\vec{A}\perp\vec{M}$. The two extra features at b, which occur in our calculated spectra (Fig. 1), are absent in the simple model of transitions between real bulk bands. At the band crossing point e, the majority-spin initial state contains admixtures $|\Sigma^3\rangle|+\rangle$ and $|\Sigma^4\rangle|-\rangle$. Consequently, a majority-spin emission peak occurs for $\vec{A}\|\vec{M}$ and a minority-spin one for $\vec{A}\perp\vec{M}$. At f, the situation is analogous. At E_F, where we have applied a Fermi function cutoff, the minority spectra are seen to be still finite (though very small), a consequence of the imaginary self-energy part of the photoelectron (upper state).

As anticipated in our symmetry considerations, the asymmetry, i.e., the normalized difference between the spin-averaged spectra for the two directions of $\vec{A}$ does not vanish (see top panel of Fig. 1). This MLD is rather small in the region of the main peaks c and d, which are only mildly affected by spin-orbit coupling, but reaches values up to 30% around features a and b, which owe their existence to spin-orbit coupling.

For p-polarized light incident at polar angle $\theta=45°$, calculated photoemission spectra are shown in Fig. 2. Consider first the case that the plane of incidence is normal to $\vec{M}$. As derived from symmetry, the spin orientation is along $\vec{M}$. The maximal information is contained in the spin-resolved spectra for incidence at azimuthal angles 0° and 180°, which we characterize by the labels p_+ and p_- (second panel of Fig. 2). We notice again the majority- (minority-) spin peaks c (d) already found for s-polarized light, but their heights are different for p_+ and p_-. This difference stems from the admixture of $|\Sigma^1\rangle|+\rangle$ in the initial state [cf. Eq. (1)] (due to spin-orbit coupling) and the resulting matrix element contribution involving A_z. Obviously, the spin-averaged spectra for p_+ then differ from those for p_- and there is a MLD (as shown in the top panel of Fig. 2). More insight is obtained by considering the limit of vanishing $\vec{M}$. As we confirmed by numerical calculations, peak $c(p_+)$ then merges with $d(p_-)$ and $c(p_-)$ with $d(p_+)$. There is no more dichroism, but a net spin polarization, which is positive (negative) for p_+ (p_-). This is the spin polarization effect found earlier with p-polarized light from nonmagnetic surfaces.[17] Our "magnetic" spectra can therefore be viewed as arising from the nonmagnetic case by an exchange splitting of both the p_+ and the p_- spin-resolved spectra. This interpretation also applies to peaks a and b. Compared to their counterparts for

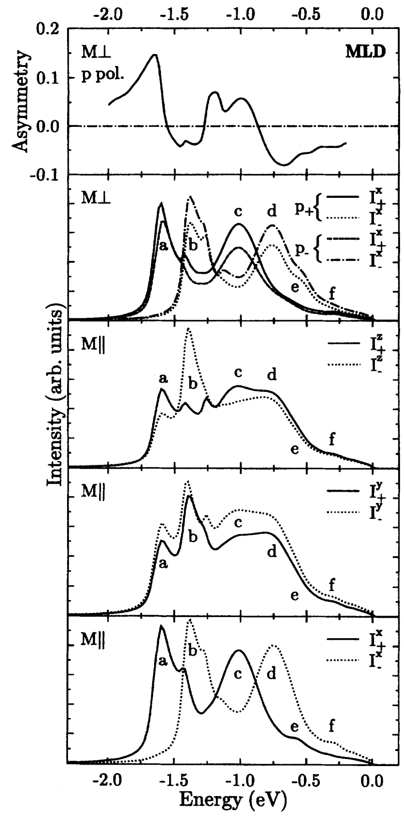

FIG. 2. Normal photoemission from ferromagnetic Ni(001) with magnetization $\vec{M}\|[110]$ by *p-polarized light* with $\hbar\omega=21.22$ eV incident at polar angle 45° relative to the surface normal. For azimuthal incidence angles 90° (labeled p_+) and 270° (p_-) relative to $\vec{M}$ (i.e., for $\vec{M}$ perpendicular to the incidence plane), the topmost panel shows the asymmetry between the spin-averaged intensities (i.e., MLD) and the second panel the corresponding partial intensities resolved with respect to the spin polarization component P^x (parallel to $\vec{M}$). For azimuthal incidence angle 0° (i.e., for $\vec{M}$ parallel to the incidence plane), the partial intensities I_+^k and I_-^k for $k=x$, y, and z corresponding to the spin polarization vector components P^k are shown in the lower three panels. The peak labels $a-f$ correspond to those in Fig. 1.

s-polarized light, they are much larger, since A_z provides matrix element contributions from the dominant initial state parts of spatial symmetry Σ^1.

In the case "plane of incidence parallel to $\vec{M}$" our numerical results (lower three panels of Fig. 2) confirm the symmetry-derived existence of three components of $\vec{P}$, with P_x and P_y resulting from exchange and spin-orbit interaction, respectively. It is interesting to note that P_z, which requires both interactions, is generally smaller (except for the feature near -1.4 eV).

While we are not aware of MLD measurements, which we could compare our results with, our spin-resolved intensity spectra for s-polarized light (third panel of Fig. 1) and our I_σ^x spectra for p-polarized light (bottom panel of Fig. 2) agree quite well with experimental ones.[19] In particular, the latter data even exhibit the small peak b (Fig. 1), near which we find the large values of the "s-polarization MLD."

In conclusion, our numerical calculations of normal photoemission from ferromagnetic Ni(001) confirm the existence of two spin-orbit-induced magnetic linear dichroism effects anticipated on symmetry grounds and actually predict values large enough to be accessible experimentally. Since these effects require neither circularly polarized light nor electron spin analysis, they promise a simpler way of studying magnetic properties of surfaces including ultrathin ferromagnetic films. The strong dependence of the p-polarized-light MLD on the orientation of $\vec{M}$ relative to the plane of incidence suggests its use for investigating magnetic domain structures. As a prerequisite for such applications, we feel that experiments on Ni(001) or some other ferromagnetic clean surface and a detailed comparison with theoretical results are highly desirable.

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