Spin-Dependent Photoemission Intensities from Solids

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We observed that the photoemission *intensity* from solids may depend explicitly on the *spin state* of the photoexcited electrons. This new phenomenon is explained by spin-dependent transmission through the surface of solids with nonvanishing spin-orbit coupling. It is demonstrated for photoemission from Pt(111) with elliptically polarized synchrotron radiation.

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Electron-spin polarization analysis in momentum-resolved photoemission has been shown recently to yield valuable information on the electronic band structure of solids, which was not obtainable by conventional intensity measurements. In photoemission[1] and inverse photoemission[2] from ferromagnets the electron spin provides an unequivocal identification of occupied or empty minority and majority bands. In momentum-resolved photoemission from nonmagnetic solids with circularly polarized light, electron-spin analysis provided a direct experimental determination of the double group symmetries of relativistic energy bands.[3,4] In all photoemission experiments so far the measured *intensities* were found to be *independent* of the magnetization of the sample or the polarization of the incoming light. The reason is that in the uv range the interaction of the magnetic component of the photon field with the magnetic moment of the electron is far too small to yield measurable intensity effects.[5] Hence, in all previous experiments an explicit spin analysis of the electrons had to be made, which requires more or less elaborate equipment,[1,4] involving a very substantial loss of intensity, even with a modern detector.[4] In the following we discuss and demonstrate a mechanism by which the measured intensity depends explicitly on the spin state of the excited electrons. With suitably chosen geometrical conditions a spin-polarization analysis may be replaced by two relative-intensity measurements. In this sense the crystal to be investigated may serve as its own spin-polarization detector.

The physical phenomenon as such is visualized in Fig. 1. For simplicity we adopt the three-step model of photoemission for the moment, and comment on the one-step model later on. Assume that we have excited electrons from two adjacent bands somewhere inside the solid into a common final band, that they have equal intensities, and that they are completely but oppositely spin polarized. This situation could, for example, arise for emission from an exchange-split band in a ferromagnet. In a nonmagnetic solid the two oppositely polarized electron groups could stem from a spin-orbit–split band when circularly polarized light is used.[3,4] For simplicity we assume inelastic processes to be independent of the spin and the crystal to have a center of inversion. Then the two electron groups travel through the crystal without changing their relative intensities nor their spin polarization, until they arrive at the surface. When detected outside the solid at a nonzero angle θ, then—this is the experimental fact—the relative intensities are no longer equal. For example [see Fig. 1(a)], the first peak (up-spin) is diminished relative to the second peak (down-spin). If we observed at the angle $-\theta$, the first peak would be enhanced over the second peak. When we invert the initial spin orientation, e.g., by reversing the magnetization of the ferromagnet or by using circular-polarized light of opposite helicity, we obtain the result of Fig. 1(b). Here, the first peak (now down-spin) is enhanced relative to the second peak. If the two peaks were unpolarized initially, i.e., if they contained up- and down-spins in equal amounts, the relative intensities would remain equal. Thus, we see that an initial inequality of spin populations is transformed into a relative intensity difference or, in other words, that the spin polarization is translated into an intensity asym-

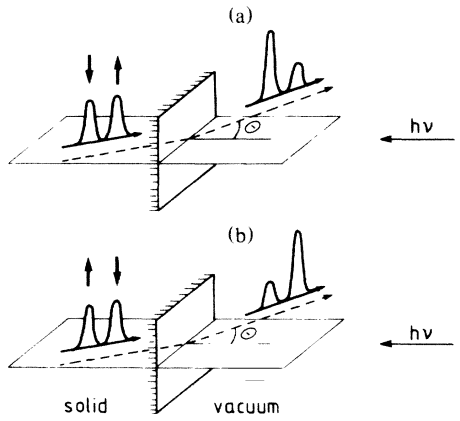

FIG. 1. Schematic description of the effects of spin-dependent transmission of photoelectrons through an interface. (a) Initially equal intensities of photoexcited electrons with opposite spin polarizations are changed by the transmission step. (b) Reversal of the initial spin polarizations reverses the relative intensity difference after transmission.

metry. For such a phenomenon to occur—and we prove below that it does exist—a mechanism is needed which, e.g., weakens the transmission of up-spin electrons through the surface and enhances the transmission of down-spin electrons.

This spin-selective filter mechanism is provided by the presence of spin-orbit interaction in the solid. Therefore, the matching of Bloch spinors on the bulk side of the surface to free-electron states on the vacuum side of the surface is spin dependent, if significant spin-orbit coupling is present in the solid. Since this is, in principle, always the case the phenomenon described here is of a general nature. It is not limited to the solid-vacuum interface, but is a general interface phenomenon, provided that the degree of spin-orbit coupling is significantly different on the two sides of an interface. A related phenomenon, the spin-dependent diffraction, is well known from spin-polarized LEED. For example, the intensity asymmetry of (normally degenerate) back-diffracted beams is used in the "LEED detector"[6] for spin-polarization analysis. Since the (spin dependent) "LEED state" is the appropriate final-state wave function in a one-step model of photoemission,[7,8] spin-dependent transmission effects are to be expected[9] in photoemission also.

In the following we shall confront our highly simplified picture of Fig. 1 with experimental results, and we will use them to discuss further characteristic features of the spin-dependent transmission through the surface. In our case the spin-polarized electrons are excited in Pt by elliptically polarized light. The experiments have been done at the 6.5-m normal-incidence monochromator of the electron storage ring BESSY in Berlin. The uv light has about 90% circular polarization, and the photon helicity can be reversed easily by use of only the synchrotron radiation from above or below the storage-ring plane. The electron spectrometer system has been described recently.[4] The light impinges along the surface normal onto Pt(111). The spectrometer is set at a polar angle θ with respect to the normal and at an azimuthal angle ϕ with respect to the ΓLUX mirror plane of the crystal. A typical experimental result is shown in Fig. 2(a) for $h\nu = 12$ eV. We see two intensity distributions I^+ and I^-, obtained with negative ($-$) and positive ($+$) light helicity, respectively. For the two prominent peaks near the Fermi energy we observe precisely the predicted behavior: For the positive helicity we find the second peak enhanced and the first peak weakened while for negative helicity the opposite happens. With linearly polarized light the average of both was measured. These intensity differences are due to the opposite polarization signs of the two peaks when excited by circularly polarized light, while both are unpolarized when excited by linearly polarized light. From the two intensities I^+ and I^- we may form a normalized intensity asymmetry A via

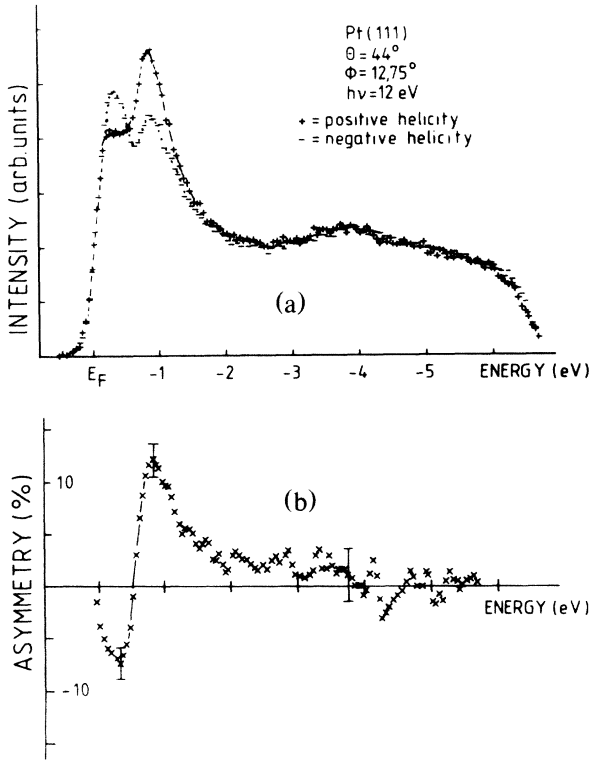

FIG. 2. (a) Experimental intensity spectra from Pt(111) obtained with circularly polarized light of positive and negative helicity. Note the change of relative intensities of the two peaks near E_F. (b) Intensity asymmetry indicating opposite spin polarizations of the two leading peaks. The asymmetry decays towards zero because of an increasing contribution of unpolarized secondary electrons.

$$A = (I^+ - I^-)/(I^+ + I^-), \tag{1}$$

which is shown in Fig. 2(b). It changes from negative to positive with increasing binding energy, and gradually decays to zero towards the region of the secondary electrons, which are known to be unpolarized.[4] This demonstrates the direct relationship between asymmetry and spin polarization. Further experimental observations are (i) the asymmetry vanishes at $\theta = 0°$, i.e., at normal take-off, and (ii) the asymmetry vanishes at $\phi = 0°$, i.e., in the mirror plane, for all polar angles θ. These observations are explained in the following by a detailed consideration of the transmission step.

It is known from spin-polarized LEED[5,10] that because of spin-orbit coupling a diffracted beam with wave vector $\mathbf{k}'$ may change in intensity when the polarization vector $\mathbf{P}_0$ of the primary beam (wave vector $\mathbf{k}$) is reversed. A generalized asymmetry vector $\mathbf{A}_{\mathbf{k}\mathbf{k}'}$ may be defined by

$$\mathbf{A}_{\mathbf{k}\mathbf{k}'} \cdot \mathbf{P}_0 = A(P_0), \tag{2}$$

where $A(P_0)$ is the asymmetry defined in Eq. (1). Since the symmetry vector owes its existence to the spin-dependent coupling of free-electron waves to Bloch spinors of the crystal, it determines not only the asymmetry of back-diffracted beams in LEED, but also the asymmetry of transmitted beams in photoemission. The asymmetry vector has to obey certain symmetry relations in space and time, determined by the crystal symmetry and the experimental conditions. For example, in elastic transmission along the surface normal, time-reversal symmetry dictates that the asymmetry vector vanishes. (For the same reason an unpolarized beam cannot become polarized in normal back diffraction.) Therefore, from Eq. (2) the asymmetry in normal photoemission must vanish, even if the photoelectrons are spin polarized. This is consistent with the experimental result (i) above, i.e., that one measures the same spectra for both helicities, as well as for linearly polarized light. For reasons of spatial symmetry the asymmetry vector stands normal to a mirror plane of the crystal.[5] In brief, this is seen from the following: The asymmetry vector **A** is an axial vector, i.e., it may be thought of as formed by the vector product of two polar vectors. Under the mirror operation each component of the vector product parallel to the mirror plane therefore changes sign. Invariance of **A** with respect to the mirror operation is thus achieved only if each of its parallel components vanishes identically. More details are given in Ref. 9. Our experimental finding (ii) (vanishing intensity asymmetry for observation in the mirror plane) can thus only be explained if the polarization vector $\mathbf{P}_0$ [Eq. (2)] lies in the mirror plane. This was very recently predicted by Borstel[11] for direct transitions within a mirror plane, and is proven here experimentally.

While there are special conditions where the intensity asymmetry vanishes, in the general case (e.g., in Fig. 2) an asymmetry is always to be expected: When the emission plane, defined by the surface normal and the direction of observation, does not coincide with a mirror plane the asymmetry vector is no longer normal to the emission plane. It does, however, have a component normal to the emission plane. The polarization vector of the excited electrons no longer lies in the emission plane and will in general have a normal component.[12] Thus, the scalar product in Eq. (2) will in general not vanish, since there are at least two vector components parallel to each other. In the case of photoemission from ferromagnets the magnetization may be chosen to be normal to a mirror plane. Strong intensity asymmetry will then result for observation in the mirror plane since then spin polarization $\mathbf{P}_0$ and asymmetry **A** are parallel or antiparallel to each other.

The asymmetry vector is known from LEED to depend on the kinetic energy of the electrons. Its variation typically occurs on the energy scale of LEED intensity variations, i.e., of the order of 5 eV. This is a slow variation relative to the scale of polarization changes in photoemission (say 0.5 eV). Thus, over limited portions of the photoemission spectrum the asymmetry may be assumed to be approximately constant. This was checked experimentally for the doublet near E_F in Fig. 2 by variation of the photon energy within several electronvolts. The magnitude of the asymmetry was found to change, but the characteristic minus-plus feature near E_F remained. It is thus clear that the intensity doublet contains electrons of opposite spin-polarization signs when excited by circular-polarized light. This information on the spin polarization is obtained by pure intensity measurements, without explicit spin-polarization analysis. Since the spin polarization is related to the symmetry properties of relativistic energy bands, this opens the way to an experimental characterization of electronic bands which goes beyond the conventional mapping of eigenvalues. In the present case the plus-minus asymmetry structure showed clearly that the doublet belongs to the topmost spin-orbit–split *d* band in Pt. The spin-orbit interaction imposes particular symmetry properties of the wave functions, giving rise to the opposite spin-polarization sign, as well as to the energetic splitting of their eigenvalues.[12]

We wish to mention that the intensity asymmetries due to spin-dependent transmission through an interface may also find an important application in band-structure studies of ferromagnets. In this case linearly polarized or even unpolarized light is sufficient, since the bands are split in energy. From the intensity asymmetry of two spectra measured with reversed sample magnetization the spin character of the bands may be determined.

Explicit polarization analysis will not be made obsolete by intensity asymmetry measurements, but these are expected to find wide application since intensity measurements are typically 3 orders of magnitude faster than polarization measurements, even with the most advanced polarization detectors.

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