Spin-dependent photoemission intensities from platinum (111)

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Photoemission spectra have been obtained from a Pt(111) surface using nearly circularly polarized light with positive and negative helicity from the Berlin storage ring BESSY. Strong intensity asymmetries, i.e., normalized intensity differences for both helicities, with absolute values of more than 45% have been found, indicating an almost complete spin selection during the transmission of the excited electrons into the vacuum. Clearly antisymmetric behavior of the asymmetry with respect to a crystal mirror plane has been observed in accordance with theoretical expectations.

I. INTRODUCTION

In recent years circularly polarized light has been used with great success in spin-resolved photoemission experiments.[1–9] The measurement of the spin polarization of the emitted electrons provided additional information about electronic states in solids as compared to conventional photoemission experiments without spin analysis. The present paper deals with intensity measurements of electrons excited by circularly polarized light and emitted in a direction away from the surface normal of the sample. Although the spin polarization was not analyzed, the electron spin is essential for the effect which was investigated, i.e., the dependence of the intensities of emitted electrons on their spin and, therefore, on the helicity (or handed-ness) of the exciting light.

Intensity variations of photoelectrons due to changes of the light polarization are routinely used in photoemission experiments to determine the nonrelativistic symmetries of electronic states. Intensities obtained with linearly polarized light with its electric field parallel to the sample surface, often incident normally onto the surface, are compared to intensities with linearly polarized light containing a component of the electric field normal to the surface (incidence away from the surface normal). Intensity differences are caused by different transition matrix elements due to different parities of the excited electron states.

In the present paper we discuss intensity variations with a change in the circular polarization of the incident light: Different intensities of emitted electrons may be measured if the sample is irradiated with circularly polarized light with positive and negative helicity, respectively, if the light intensity is kept constant. The difference from the case mentioned above is that the number of electrons that are excited is equal for both helicities (if the spin degeneracy of the electronic states is not removed by exchange interaction as in ferromagnetic materials). Only when the electrons leave the crystal, different intensities for a given emission direction may occur. This will be explained in more detail in what follows.

Circularly polarized light is irradiated normally onto a platinum (111) surfaces. Due to spin-orbit coupling electrons that are excited by absorption of a photon generally will be spin polarized. A change in the helicity of the light leaves the intensity of the *excited* electrons inside the crystal with a general wave vector $\mathbf{k}_0$ unaffected, but reverses their spin polarization $\mathbf{P}_0$ to $-\mathbf{P}_0$. This is due to time-reversal symmetry combined with spatial inversion symmetry:[10] Time reversal changes the helicity of the light and inverts spin and momentum of the excited electrons. If spatial inversion symmetry is present, then for each state with energy E, wave vector $\mathbf{k}$, and spin $\mathbf{s}$, another state with E, $\mathbf{k}$, and $-\mathbf{s}$ exists. Therefore, excitation with light with positive and negative helicity produces the same amount of electrons with energy E and wave vector $\mathbf{k}$, but with opposite spin $\mathbf{s}$ and $-\mathbf{s}$, respectively. Since linearly polarized light may be considered as a coherent superposition of circularly polarized light with positive and negative helicity and with equal amplitude, another consequence of inversion symmetry is that the polarization of electrons excited with linearly polarized light should be zero. Of course, all these statements hold only if the spin degeneracy of the electronic states is not removed by exchange interaction as in ferromagnetic materials.

It should be mentioned here that inversion symmetry is only present in the bulk of the crystal, but that it is disturbed at the surface. In special cases like band-gap emission of electrons located very close to the surface, i.e., transitions of electrons into free-electron states which are strongly attenuated toward the sample interior, the spin polarization may not be inverted exactly by a change of the helicity. Or, equivalently, electrons excited with linearly polarized light may exhibit a nonzero spin polarization[11,12] even if emitted along the surface normal. However, the peaks in the spectra that will be presented in Sec. III of this paper are most certainly due to direct transitions between bulk states so that inversion symmetry is expected to be valid in this case.

So far we have considered only the excitation of electrons with circularly polarized light, and, assuming constant light intensity, we have found a dependence of the spin polarization on the helicity, but not of the electron intensity. After the excitation a fraction of the excited electrons will reach the crystal surface and will be

transmitted into the vacuum. The electron transport to the surface should not depend on their spin due to inversion symmetry and is therefore not considered here. In a simple approximation the electrons are considered to be in an eigenstate of a Hamiltonian with the periodic bulk potential and are therefore not affected by it. At the surface, however, this translational periodicity is broken. A part of them will be reflected back into the sample. Others will ultimately leave the crystal. Since the periodicity of the crystal is broken normal to the surface, the normal component of the wave vector is changed during the transition, whereas the wave-vector component parallel to the surface is conserved modulo a reciprocal-lattice vector, which is most often the zero vector. Generally, the wave vectors $\mathbf{k}_0$ inside and $\mathbf{k}$ outside the sample will not be parallel, with the exception of normal emission.

The scattering process at the surface is complicated due to multiple-scattering effects similar to those in low-energy electron-diffraction (LEED) experiments. For our purpose the most important point is that the scattering process is spin dependent. This may be made plausible by contemplating the scattering of an electron at a single atom. The potential that the electron experiences contains a contribution of the spin-orbit interaction. This term is proportional to the dot product of the electron spin and its angular momentum. If the spin is not parallel to the scattering plane, the sign of this term will change if the spin is reversed, e.g., by changing the helicity of the exciting light. This may indicate the basic mechanism for the spin dependence of the scattering at the crystal surface, even though the real process is much more complex.

The consequence of this effect is that scattering of electrons with wave vector $\mathbf{k}_0$ into $\mathbf{k}$ may occur with different probabilities for different spin directions. This shall be illustrated with the help of Fig. 1. Circularly polarized light with positive helicity impinges normally onto the sample surface [Fig. 1(a)]. As an example, it is assumed that two energetically separated transitions take place inside the crystal with equal intensities as indicated by the schematic spectrum of the excited electrons on the left-hand side of Fig. 1(a). The two peaks are assumed to be both completely polarized, i.e., the electrons in each peak shall all be in the same spin state, but with opposite spin polarization as indicated by the arrows. The scattering of the excited electrons at the surface depends on their energy on a scale of several electron volts as in LEED experiments. We assume the energy separation of the two peaks to be small enough so that we can ignore the energy dependence for the moment. Still the scattering process is spin dependent. As an example, we assume that preferential scattering of "spin-down" electrons in the direction towards the detector occurs. Then the number of *detected* electrons will be higher for the peak with lower energy than for the peak with higher energy as is indicated by the schematic spectrum on the right-hand side of Fig. 1(a). Figure 1(b) shows the same experiment if the helicity of the light is changed to negative. The arrows indicate that the spin polarization is inverted by the change of the helicity. Since spin-down electrons are still favored by the scattering process, the higher intensity is now found in the peak with higher kinetic energy. Thus, the measured intensity of photoelectrons may depend on their spin, which, in turn, depends on the helicity of the circularly polarized light.

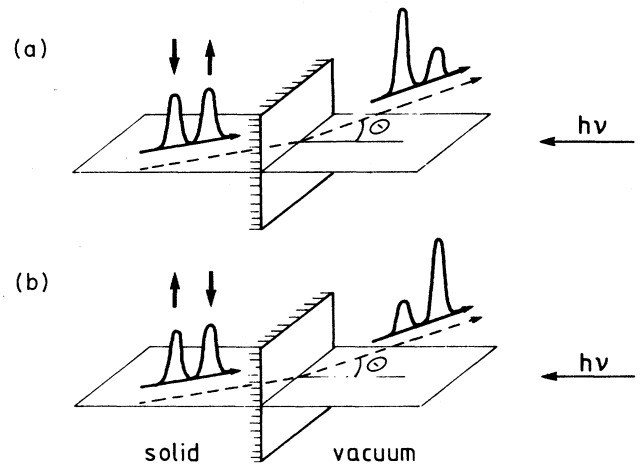

FIG. 1. Sketch to describe the principle of intensity asymmetries. Explanation is given in the text.

In order to have a measure of this effect that is independent of the light intensity, an intensity asymmetry A is defined by

$$A_{\mathbf{k}_0\mathbf{k}}(\mathbf{P}_0)=\frac{I^+-I^-}{I^++I^-} . \tag{1}$$

I^+ and I^- are the measured photoelectron intensities with positive and negative helicity, respectively, while the light intensity is kept constant. The asymmetry depends on the wave vectors $\mathbf{k}_0$ inside and $\mathbf{k}$ outside the crystal as well as the direction and absolute value of the polarization $\mathbf{P}_0$ of the electrons excited with light of positive helicity. From the definition (1) it follows that the asymmetry may have values between -1 and $+1$. It should be mentioned here that generally the spin polarization is also changed during the transmission into the vacuum.[3]

The existence of spin-dependent photoemission intensities has been demonstrated for the first time by Oepen *et al.*[13] at a Pt(111) surface. Since only one spectrum was published showing maximum asymmetry values below 15%, we were motivated to explore these asymmetry effects more intensely. Our aim was to find larger asymmetry values which would mean that the surface can act as an effective spin filter. We wanted to verify some theoretical expectations about the intensity asymmetries derived from the spatial symmetry of the sample. These expectations will be discussed in what follows.

To describe the asymmetry effect independently of the polarization $\mathbf{P}_0$, an asymmetry vector may be defined so that

$$\mathbf{A}_{\mathbf{k}_0\mathbf{k}}\cdot\mathbf{P}_0=A_{\mathbf{k}_0\mathbf{k}}(\mathbf{P}_0) . \tag{2}$$

The length of the asymmetry vector is again restricted to the interval $[-1,+1]$. Now the behavior of the asym-

metry with respect to a crystal mirror plane shall be considered. Figure 2 shows that the direction of emission from a single crystal—in this case from a platinum (111) sample—can be characterized by a polar angle θ to the surface normal, and by an azimuthal angle ϕ, which here is defined so that $\phi=0°$ if the electron-emission direction lies in a ΓLUX mirror plane.

A mirror operation at the ΓLUX mirror plane changes the azimuthal angle ϕ from $+\phi$, while θ is conserved. Both vectors $\mathbf{A}_{\mathbf{k}_0\mathbf{k}}$ and $\mathbf{P}_0$ are axial vectors, so that they behave like angular-momentum vectors under a mirror operation. This means that their components parallel to the mirror plane change signs, while the components normal to it do not. Since both vectors transform equally their scalar product is left unchanged so far.

The photon spin is also an axial vector. For positive helicity it is oriented parallel, and for negative helicity antiparallel to the direction of propagation of the light. Since the photon spin only has components in the mirror plane, it is inverted by the mirror operation. To maintain the same definition of the asymmetry for emission along $(\theta,-\phi)$ as for $(\theta,+\phi)$, the helicity must be changed back. As a consequence the polarization vector is inverted. This leads to a change of sign in the scalar product $\mathbf{A}_{\mathbf{k}_0\mathbf{k}}\cdot\mathbf{P}_0$ and therefore in the asymmetry. The result of these symmetry considerations is that if the asymmetry is measured in the directions $(\theta,+\phi)$ and $(\theta,-\phi)$, equal absolute values but opposite signs should be found:

$$A(\theta,-\phi)=-A(\theta,+\phi)\ . \tag{3}$$

For the special case $\theta=0°$, i.e., emission in the ΓLUX mirror plane, it follows from Eq. (3) that the asymmetry must vanish:

$$A(\theta,\phi=0°)=0\ . \tag{4}$$

The reason is that in this case the asymmetry vector must be normal to the mirror plane and the polarization vector must be parallel to it. Therefore the scalar product between both vectors is zero. For electrons emitted in the direction of the surface normal, the asymmetry vector itself must vanish: Since there are three equivalent mirror planes oriented at 120° to each other, the asymmetry vector must be orthogonal to all of them, and the only possibility of fulfilling this condition is that the asymmetry vector is the zero vector:

$$\mathbf{A}(\theta=0°)=\mathbf{0}\ . \tag{5}$$

A series of experiments was performed to verify especially the antisymmetric behavior of the asymmetry with respect to a crystal mirror plane. The results will be presented and discussed in Sec. III. In Sec. II, a few experimental remarks will be made.

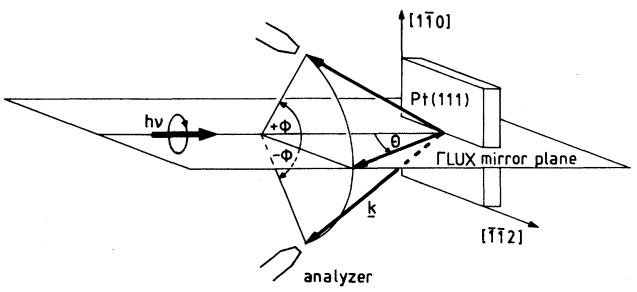

FIG. 2. Scattering geometry of the experiment. Elliptically polarized light is incident normally onto a Pt(111) surface. Emission direction is characterized by the polar angle θ to the surface normal and an azimuthal angle ϕ to a ΓLUX mirror plane of the crystal.

II. EXPERIMENTAL REMARKS

The photoemission experiments were done at the 6.5-m Normal Incidence Monochromator (NIM) at the Berlin storage ring BESSY (Berliner Elektronspeicherring-Gesellschaft für Synchrotronstrahlung). This monochromator[14] yields elliptically polarized light in the photon energy range between 6 and 30 eV. The intensity fraction of circularly polarized light is around 90%. The light was incident normally onto a platinum (111) single crystal. The polar emission angle θ could be chosen by rotating the spectrometer[15,16] about an axis within the sample surface. The azimuthal angle ϕ was varied by rotating the sample about its surface normal.

The results to be shown in this paper were obtained at a photon energy of 18 eV. The total resolution of the monochromator and the spectrometer is estimated to be 0.3 eV. Since the light intensities were not exactly equal for positive and for negative helicity, the photoelectron intensities obtained with light with negative helicity were multiplied by a constant factor that was chosen so that the intensities for both helicities were equal at binding energies below 6 eV where the asymmetry should be close to zero. Because mainly s-type electrons and electrons which have suffered considerable energy losses are detected, for both kinds of electrons the polarization should be close to zero. This assumption was confirmed by normal-emission spin-polarization spectra from both Pt(111) (Ref. 8) and Pt(110) (Ref. 4) surfaces at the same photon energy (18 eV). No other correction was applied to the data.

The sample was cleaned prior to the experiments by repeated cycles of sputtering with argon ions, heating in oxygen to remove carbon, and flashing in UHV. During the measurements it was kept clean by flashing the crystal to approximately 1300 K once every hour. The good quality of the sample surface was checked by Auger-electron spectroscopy[17] and LEED.

III. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 3 shows a series of spectra which were obtained with elliptically polarized light at a photon energy of 18 eV. The polar angle θ was fixed at $\theta=62°$ and the azimuthal angle ϕ to a ΓLUX mirror plane was varied over a wide range. Solid and dashed lines show spectra measured with positive and negative helicity, respectively. For most angles ϕ, major differences between both spec-

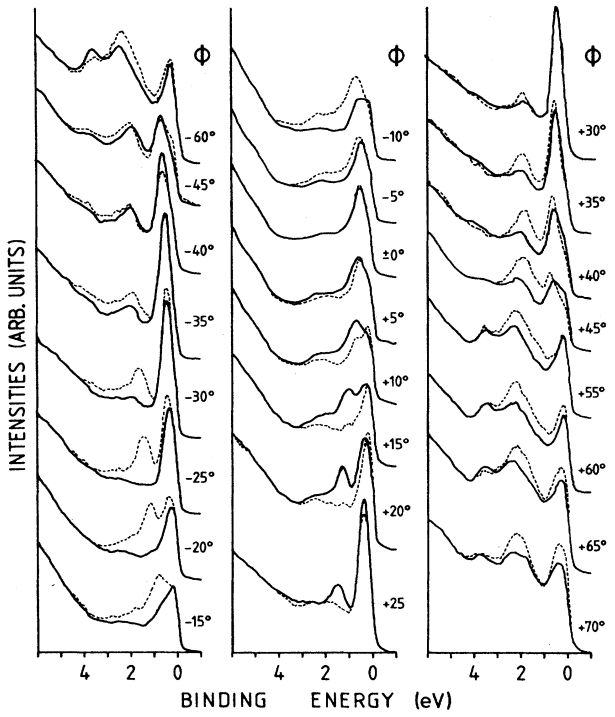

FIG. 3. Photoelectron spectra from Pt(111) at a polar emission angle $\theta=62°$ and various azimuthal angles ϕ to a ΓLUX mirror plane. Elliptically polarized light was incident normally onto the sample. Solid and dashed lines show spectra obtained with light of positive and negative helicity, respectively.

tra can be seen, but this is not so for $\phi=0°$, i.e., emission in the mirror plane. Actually, the position of the mirror plane was determined only roughly (for geometric reasons) by LEED, and then more exactly by the rather sensitive condition that the spectra for both helicities should be equal (except for a constant factor due to different light intensities) in accordance with Eq. (4). At $\phi=\pm 2.5°$ already clearly visible deviations from this condition were found.

At $\phi=+5°$ the intensity is higher for positive helicity than for negative helicity over a substantial energy range in the spectra, and vice versa in the spectra at $\phi=-5°$. This is the behavior that is expected after Eq. (3) together with the definition of the asymmetry Eq. (1). For higher absolute values of ϕ this effect becomes more pronounced. The highest absolute values of the asymmetry are found in the peak with a binding energy of approximately 1.5 eV around $\phi=\pm 25°$. For example, at $\phi=-25°$ this peak is only seen in the spectrum for negative helicity, while in the spectrum for positive helicity only a rather constant background is present.

The interpretation of the peaks in the spectra of Fig. 5 is not easy since a band structure of platinum for general wave vectors is not available. Therefore we did linear combinations of atomic orbitals–augmented-plane-wave (LCAO-APW) calculations using the relativistic interpolation scheme of Smith and Mattheis.[18] This scheme contains various parameters which were optimized by a least-squares fit to reproduce the valence bands of the relativistic, self-consistent band structure of Eckardt and Noffke[19,20] along high-symmetry lines, which is shown in Fig. 4. The interpolation scheme then gives the dispersion relations of the valence bands for general wave vectors $\mathbf{k}_0$.

To calculate possible transitions for given emission directions and photon energies we also needed the dispersion relation of the excited states. We did not use the calculated unoccupied bands above the Fermi level for two reasons: First, according to Hora and Scheffler[21] the dispersion of the excited states with $\mathbf{k}_0$ should follow rather nearly-free-electron behavior than the calculated ground-state bands above the Fermi energy at final-state energies around 14–18 eV, which is the energy range of interest here. In particular, the band gaps of the empty ground-state bands due to hybridization in this energy range should not be present for the excited states in photoemission experiments. In addition, the spectra gave no evidence for contributions of more than one final-state band, or for band gaps in the final states. The second reason is of computational nature: The unoccupied bands can only be calculated with an extended interpolation scheme which we could not use since it would have increased the computation time excessively.

For these reasons we chose a parabolic band as an approximation to the final states which followed the dispersion relation

$$E(\mathbf{k})=-1.5\ \text{eV}+\frac{\hbar^2\mathbf{k}^2}{2\times 1.15m_0}\ . \tag{6}$$

Several branches of this parabolic band can be seen in Fig. 4 as dotted lines. Some parts of them follow closely parabolic parts of the calculated ground-state bands. Along the Λ line a mapping of the initial-state bands could be successfully performed using the corresponding branch of the same parabola. The choice of the final-state band is somehow arbitrary but it may be hoped that

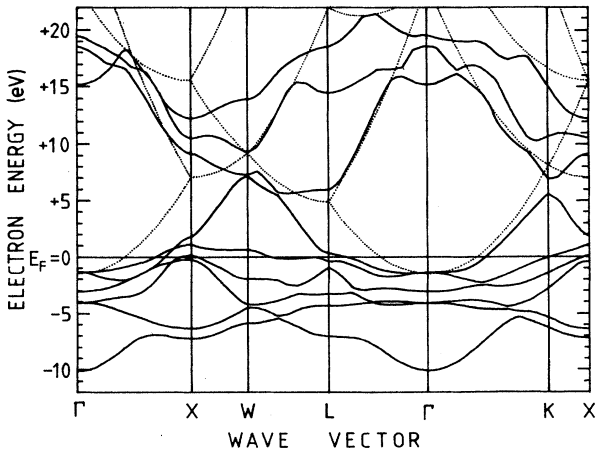

FIG. 4. Relativistic self-consistent band structure of platinum after Eckardt and Noffke (Refs. 19 and 20) along high-symmetry lines. Dotted lines show several branches of a parabolic band with $E_0=-1.5$ eV and $m^*=1.15m_0$.

it is good enough for a qualitative analysis of the photoemission peaks.

It is now possible to calculate initial-state energies for all possible transitions with fixed components of $\mathbf{k}_0$ parallel to the crystal surface.[22] The calculation is done iteratively by use of the values of the angles θ and ϕ, of the photon energy (18 eV), and of the work function which could be estimated from the length of the spectra to be 5.6 eV. The result is shown as solid lines in Fig. 5 versus the azimuthal angle ϕ, together with the measured binding energies of the peaks in the spectra of Fig. 3 (solid circles). Generally, the agreement is quite good. The strongest discrepancy occurs for band 3 around $\phi=0°$. The corresponding $\mathbf{k}_0$ vectors are near the point X where band 3 has a maximum. At $\phi=\pm 40°$ additional peaks are measured which are not reproduced by the calculation. But at these angles a minimum of band $E(\phi)$ for band 6 is found though it is located slightly above the Fermi level and was therefore not included in Fig. 5.

The agreement between experiment and theory is at least good enough to identify the initial-state bands for all transitions. A comparison with Fig. 3 shows that the peak at a binding energy of approximately 1.5 eV at $\phi\approx\pm 20°$ is produced by electrons excited from band 4. The asymmetry of this peak as calculated after Eq. (1) is plotted versus ϕ in Fig. 6. The antisymmetry with respect to the mirror plane at $\phi=0°$ as expected from Eq. (3) is found indeed. The deviation from perfect antisymmetry is most probably due to geometrical errors. The maximum absolute value of the asymmetry is in excess of 45%. If it is taken into account that the fraction of circularly polarized light is only 90% and that an unpolarized background is always present in the spectra, this result indicates that the scattering of the excited electrons at the crystal surface leads to an almost complete spin selection in this case. To explain this experimental result three conditions must be fulfilled. (1) The excited electrons must be highly polarized. Polarization measurements in the ΓLUX mirror plane,[3] along the Λ line[8] and the Σ line[4] of platinum, showed maximum polarization

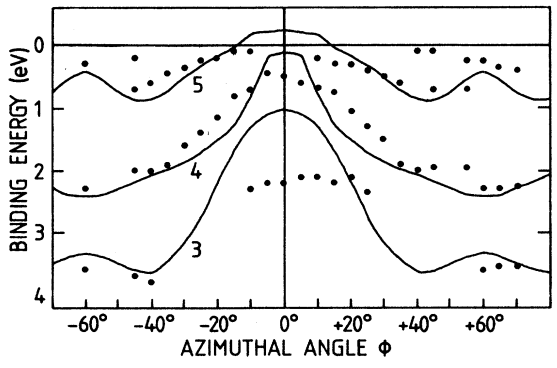

FIG. 5. Binding energies of the peaks in the spectra of Fig. 3 vs the azimuthal angle ϕ (solid circles). Lines show the calculated dispersion of the transitions from some initial-state bands to the parabolic band shown in Fig. 4.

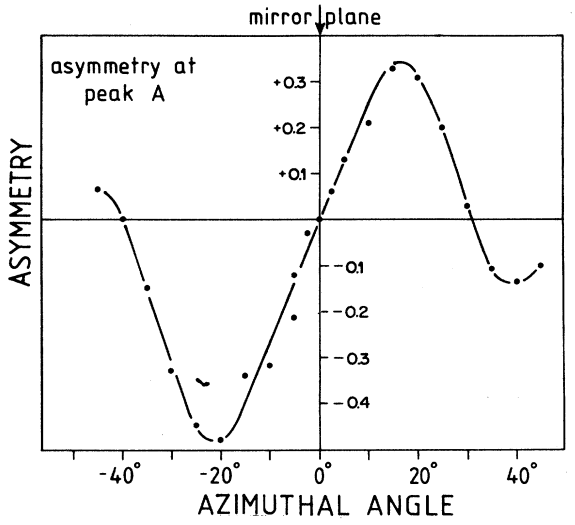

FIG. 6. Asymmetry of peak A in Fig. 3 vs the azimuth ϕ. Clearly, antisymmetric behavior with respect to the ΓLUX mirror plane ($\phi=0°$) can be seen.

values of 55% at room temperature. It cannot be expected that the polarization of the excited electrons should be considerably higher in the present case. Therefore, (2) the asymmetry vector must have a length close to 1. This surprising result means that almost total spin selection occurs during the scattering process, and that (3) both the polarization and asymmetry vectors must be oriented almost parallel or antiparallel to each other, respectively, to give a scalar product of more than 0.45.

For electrons excited from band 4 the linear combination of atomic orbitals–orthogonalized-plane-wave (LCAO-OPW) calculation gives an initial wave vector $\mathbf{k}_0$ of (0.03, 0.58, −0.68) after reduction to the first Brillouin zone for an emission direction of ($\theta=62°$, $\phi=-25°$) and a photon energy of 18 eV. The wave vector is given in units of $2\pi/a$, where a is the length of the cubic unit cell. This means that the electrons with the highest asymmetry originated from a point not too far away from point K. An inspection of Fig. 4 shows that at point K band 4 shows a binding energy of about 1.5 eV, in agreement with the measured and calculated binding energy. Polarization measurements have been done earlier at a Pt(110) surface.[4] Electrons excited from band 4 near point K and emitted normally showed a polarization of about 30%. This value cannot be compared directly to the asymmetry value from Fig. 3, though, because during the polarization measurement the light was incident along the Σ line so that the polarization vector must also be parallel to the Σ direction, whereas in this case the light incidence was aligned with the Λ line. Also, the photon energy was substantially lower for the polarization experiment so that the electrons were excited into the lowest branch of the final-state parabola seen in Fig. 4 in the Σ direction.

For electrons excited from band 5 not all of the conditions for high asymmetry which were mentioned above

are fulfilled since the peak near the Fermi edge shows very small asymmetry. The asymmetry vector probably does not change much within an energy difference of about 1 eV to the peak with the highest asymmetry. Neither is it expected that the spin polarization of the excited electrons is close to zero, because in the ΓLUX mirror plane as well as along the Λ line substantial spin polarization of electrons excited from band 5 was measured. The wave vector of the electrons excited from band 5 at $\phi = -25°$ is also close to point K. The polarization measurement at the Pt(110) surface[4] gave polarization values only slightly lower than for band 4, but with opposite sign. If the direction of light incidence is changed away from the Σ line, the direction of the polarization vector will also change in an unpredictable way. The only polarization measurement of electrons emitted away from the surface normal was done also at a Pt(111) surface[3] in the ΓLUX mirror plane ($\phi = 0°$). The polar angle θ as well as the photon energies were somewhat lower than in this case, but the result which is of interest here was that electrons stemming from band 5 showed a polarization vector almost normal to the polarization vector of electrons from band 4. The directions of the polarization vectors $\mathbf{P}_0$ inside and $\mathbf{P}$ outside the crystal are generally not the same, and the azimuthal angle was different. Therefore this result cannot be compared directly with the results of the asymmetry measurements. But the only probable explanation for the missing asymmetry of the electrons excited from band 5 is exactly the same, i.e., that the spin-polarization vector is oriented almost normally to the asymmetry vector and, therefore, also to the polarization vector of the electrons excited from band 4.

The direction of the asymmetry vector itself cannot be extracted from the present results. Indeed it seems impossible to measure it even if combined asymmetry and polarization measurements were done. The only alternative is to calculate the asymmetry vector by a method similar to relativistic LEED calculations.

IV. CONCLUSIONS

Photoemission intensity asymmetries have been measured at platinum (111) surface. Clearly antisymmetric behavior of the asymmetry with respect to a crystal mirror plane was observed in accordance with theory. Absolute values of the asymmetry of more than 45% were found, indicating an almost complete spin selection during the electron transmission process through the sample surface. At lower polar angles a measurement of the asymmetry may even provide some qualitative information about the spin polarization of the excited electrons without the complicated and time-consuming explicit spin analysis.

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