

On the energy resolution of a GaAs-based electron source for spin-resolved inverse photoemission

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Abstract. The spin resolution in inverse photoemission spectroscopy is achieved by injecting spin-polarized electrons, usually produced by GaAs-based cold cathodes that replace hot-filament electron guns of spin-integrated setups. The overall energy resolution of the system can be enhanced by adjusting either the optical bandpass of the photon detector or the energy distribution of the electron beam. Here we discuss the influence of the photocurrent and the photocathode temperature on the energy broadening of the electron beam through the inverse photoemission spectra of the spin-split Shockley surface state of Au(111). First, we find that cooling down the GaAs photocathode to 77 K increases the band gap and reduces the number of allowed vertical transitions, monochromatizing the electron beam with an enhancement of about 30 meV for the energy resolution. Second, we observe a correlation between the generated photocurrent at the electron source, and the space-charge effects at the sample as a reduction of lifetime and spin asymmetry of a polarized bulk state. These observations allow defining a threshold of current density for the optimum acquisition in the measurements of spin-resolved inverse photoemission in Au.

1 Introduction

The exploration of unoccupied states with total spin control is now possible in inverse photoemission (IPES) by fully decoupling the spin polarization vector of the electron beam from its wavevector [1]. The decoupling of the polarization orientation of the electron beam from the wavevector allows exploring the unoccupied bands with the spin orientation more suitable to explore every state. Another critical factor when studying the bands is the energy resolution in order to distinguish states close in energy. It has already been demonstrated that spin-resolved measurements of spin-polarized inverse photoemission (SPIPES) allow discriminating unoccupied spin-dependent states whose energy splitting is below the energy resolution [2]. Any enhancement of the total energy resolution ΔE_{tot} in IPES setups will therefore allow to better discriminate states close in energy, as required in particular for the study of spin-textures in systems with e.g., spin-orbit coupling (SOC), magnetic exchange or Rashba effect. The total energy resolution depends on the thermal distribution of the electron beam and the bandpass energy of the photon detector with usual detection in the vacuum ultraviolet (VUV) regime. In isochromat IPES, where only the kinetic energy of the electron beam is varied, the bandpass detection of the emitted photons consists of a combination of the photoionization threshold of the detection gas (high-pass) and the transmission cutoff of an optical window (low-pass) in

Geiger-Müller counters. A modification of the bandpass characteristic through the decrease of the low-pass energy transmission allows for enhancement of the optical resolution of the photon detector system. The surface quality and purity [3], and the temperature of the window [4,5], usually made of alkaline earth fluoride crystals, are the most explored parameters. The temperature dependency of the cutoff wavelength of VUV transmission windows, explained by a photoexcitation model of excitons at thermally distorted lattice regions [5,6], allows to reach up to 165 meV for the overall energy resolution [4]. In more elaborated devices the high-pass is modified by using the oxygen and krypton absorption lines by incorporating additional windows [7]. However, the increase of energy resolution by the aforementioned methods is usually accompanied by a decrease in the quantum yield [8] which is quite disadvantageous due to the intrinsic cross-section of IPES. Thus, an alternative to enhance the energy resolution is by adjusting the beam distribution of the GaAs-based electron source. Only a few studies deal with the photoemission broadening of the polarized electrons, see for instance [9,10]. Varying the temperature of the electron source rather than the temperature of the window of the photon detector is an alternative that does not reduce the counting rate in IPES since it does not filter the energy of the beam electrons. For these reasons, we report here the effects of the temperature of a GaAs-based electron source on the energy resolution in isochromat IPES by analyzing the spin-polarized Shockley surface state of Au(111). From

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the phase space considerations of the IPE process, it would be desirable to have a high-intensity source with a small beam size and high flux. Yet, even when samples could stand relatively high currents, the electron-electron repulsion becomes important in low-energy and high-brightness beams, affecting not only the energy but also the momentum resolution [11,12]. The selected transfer energy in the electron optics has been chosen to reduce them [1]. Therefore, the effects of the space-charge are observed as a decrease in the lifetime of the surface state while increasing the electron current at the target.

2 Methodology

The Au(111) surface was prepared by Ar ion sputtering (1 keV) and annealing at 800 K under the UHV environment. The cycles were repeated until a clear LEED pattern with sixfold symmetry was obtained (not shown). The SPIPES measurements were subsequently performed.

The spin-resolution is incorporated in the room temperature measurements on freshly-prepared samples by the near-infrared photoemission (~ 830 nm) from a negative electron affinity GaAs wafer, already detailed for isochromat IPES [1]. The net polarization of the photocathode is reported to be $P = 0.30 \pm 0.03$. The photocathode was cooled down to LN₂ boiling point (77 K) through an in-house cold trap at the 3-axis photocathode manipulator and stabilized in about 1 hr. The temperature is monitored by a contact thermocouple at the photocathode surface. Because thermal fluctuations of the GaAs lattice modify the sweet spot of the NIR optical excitation, impacting the transmission of the electron beam to the sample during the cooling process, the experiment was performed until the electron transmission ($\sim 55\%$), and the photocathode temperature stabilized.

3 Results

The degree of polarization of electronic states due to SOC is more significant in heavy-atom compounds and it can be enhanced by surface adsorbates as occurs in noble-metal surfaces with Rashba splitting [13]. However, if the Rashba parameter is very small, the spin effects may not be observed in IPES due to the constraints on the energy resolution, usually of hundreds of meV. A prototypical system for studying the Rashba splitting is the Shockley surface state of Au, studied below [14-16] and above the Fermi level [1,17], with a spin that is tangent to the concentric Fermi surface. We present in Fig. 1 the SPIPES raw spectra of Au(111) along the ΓM direction at room temperature. The spectra are normalized to the impinging current of $\sim 0.6 \mu A$. The spin-polarized Shockley surface state of the L-gap and surface-projected bulk bands are observed, and the

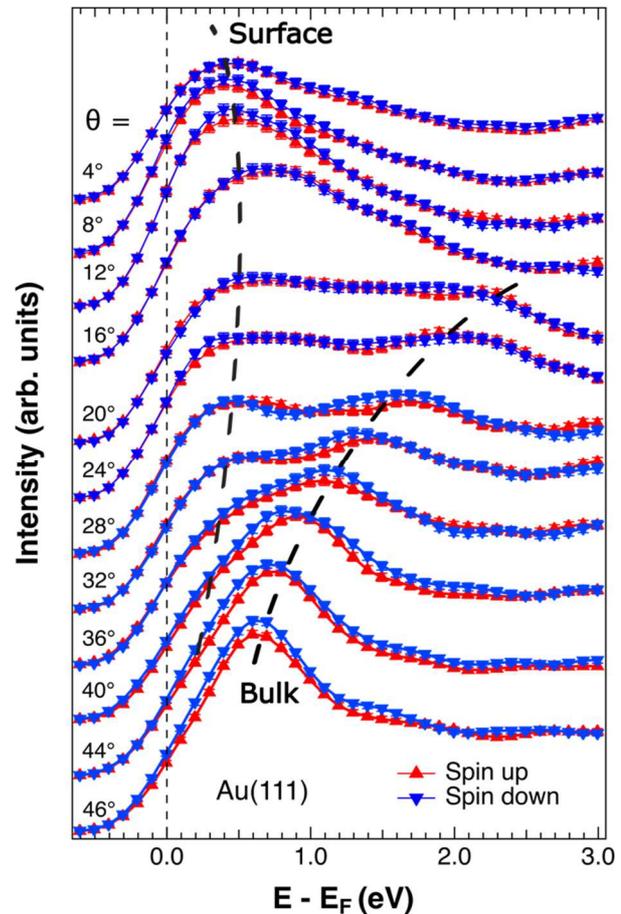


Fig. 2. SPIPES spectra of Au(111) along ΓM at room temperature. Surface and bulk states are present. Lines are added as a guide to the eye.

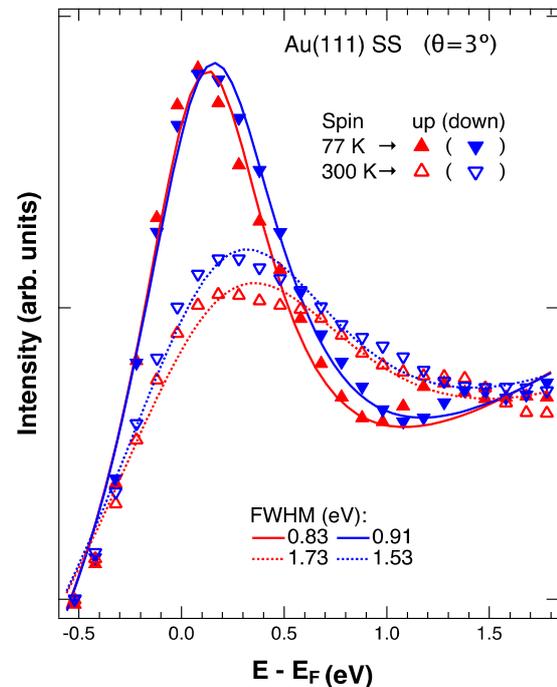


Fig. 1. SPIPES spectra of Shockley surface state of Au(111) at $\theta = 3^\circ$ as a function of the temperature of the GaAs photocathode. In the LT, the binding energy of the state is more defined as follows from the FWHM.

details on the surface resonance and spin-polarized bulk states have been discussed elsewhere [1,17]. The lifetime of the Shockley surface state is apparently smaller when approaching normal incidence due to the particular experimental constraints. It is thus desirable to increase the energy resolution to better estimate the spin-split final state binding energies. Therefore, we focus first on the thermal distribution of the electron beam while studying the surface state at an almost normal incidence ($\theta=3^\circ$), as shown in Fig. 2.

Two spectra are contrasted as a function of the photocathode temperature: the 77 K low temperature (LT) and the 300 K high temperature (HT) spectra. Both measurements are normalized to a target current kept at about $0.6 \mu\text{A}$ for both sets of measurements. The system presents the surface Rashba effect so that the binding energy of the polarized final states are shifted in energy with respect to each other for a given wavevector. It is evident that the IPES spectral linewidth of the Shockley surface state of Au(111) is decreased when the GaAs temperature goes from HT to LT, facilitating the determination of the binding energy of the state. The difference of the full width at half maximum (FWHM) is 0.9 (0.6) eV for the spin up (down) component. By increasing the bandgap energy of GaAs, the optical transitions are then more concentrated around the Γ -point as indicated by the rise of the spectral intensity in the LT surface-emission while maintaining the same inelastic background of the HT at ~ 1.5 eV. In this scenario, the effective polarization of the GaAs photocathode should also increase due to the monochromatized electron beam. A difference in the binding energy is possibly due to a variation of the photocathode work function at these two temperatures. Experimentally, this variation can be compensated by modifying the acceleration potential of the electron source [1]. Anyway, here we are concerned with the broadness of the SPIPES features. The SPIPES spectral broadening is mainly affected by two parameters: the thermal energy spread of the beam and the space-charge effects of the cathode. If we concentrate at first on thermal effects, the broadening when considering a Maxwellian momentum distribution projected along one transverse direction is given by [11, 18]:

$$\Delta k(T) = \frac{2}{\hbar} \sqrt{2mk_B T \ln(2)}, \quad (1)$$

where T is the photocathode temperature. Since electrons arriving at the sample are free electrons, their energy broadness is:

$$\Delta E = \frac{\hbar^2}{m} k \Delta k. \quad (2)$$

And combining Eq.1 and Eq.2 we get that

$$\Delta E(k, T) = \frac{2\hbar}{m} k \sqrt{2mk_B T \ln(2)}. \quad (3)$$

If only the photocathode temperature is varied, the change between energy distributions can be quantified at each wavevector k_j . Assuming that the binding energy of the spin-down state is $E^\downarrow \sim 0.2$ eV, we get that $k_{\parallel} \sim 0.06 \text{ \AA}^{-1}$ and therefore $\Delta E(300) - \Delta E(77) = 62 - 32 \text{ meV} = 30 \text{ meV}$. In other words, there is a difference of 30 meV in the thermal distribution of the electron beam when the photocathode is at 77 K.

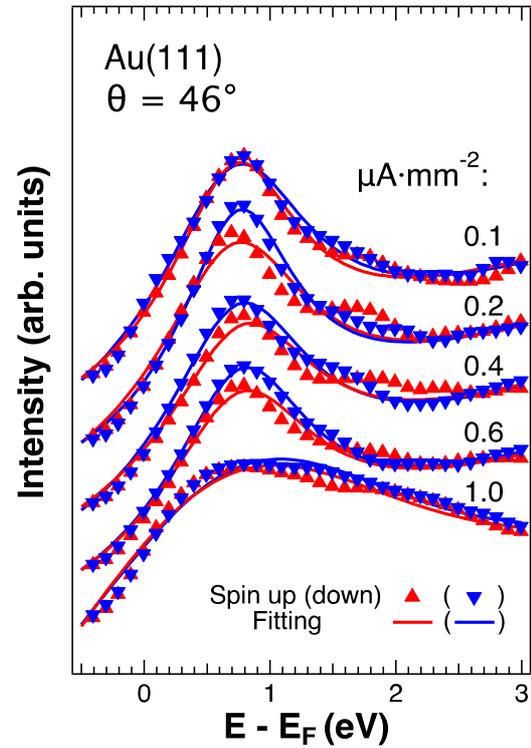


Fig. 3. SPIPES spectra of Au(111) *sp*-bulk state ($\theta = 46^\circ$) as a function of the current density over the sample. The data (triangles) are normalized to maximum intensity and fitting lines are shown. The effects of the current density are observed in: (i) the spin asymmetry and (ii) the spectral broadening of the state.

The above energy resolution of the inverse photoemission setup has been determined by avoiding the space-charge effect. The space-charge at the photocathode surface may broaden the thermal energy distribution of the electron source if the emitted current is too high. This eventuality is usually prevented by decreasing the voltage in the extractor lenses at the electron source. Experimentally, we studied the space-charge effects by varying the impinging current into the *sp*-bulk state ($\theta = 46^\circ$) of the Au(111) surface. This state presents spin asymmetry and pseudo-Rashba splitting as experimentally observed and ascribed to initial-state effects [15]. The SPIPES spectra of the state as a function of current density are presented in Fig. 3 and their corresponding FWHM are presented in Fig. 4. The space-charge affects the electron beam for relatively high current densities over the sample. It can be seen a gradual increment of the FWHM of the state as a function of the current density, up to the point that it is difficult to determine the final state binding energy when the beam current approaches $1.0 \mu\text{A}\cdot\text{mm}^{-2}$. Thus,

it is critical to characterize the current density over the specific sample since this may lead to misinterpretations of the lifetime of the state. This is why we have determined the most favorable current densities for exploring Au(111) with SPIPES and in the range between 0.2 and 0.6 $\mu\text{A}\cdot\text{mm}^{-2}$.

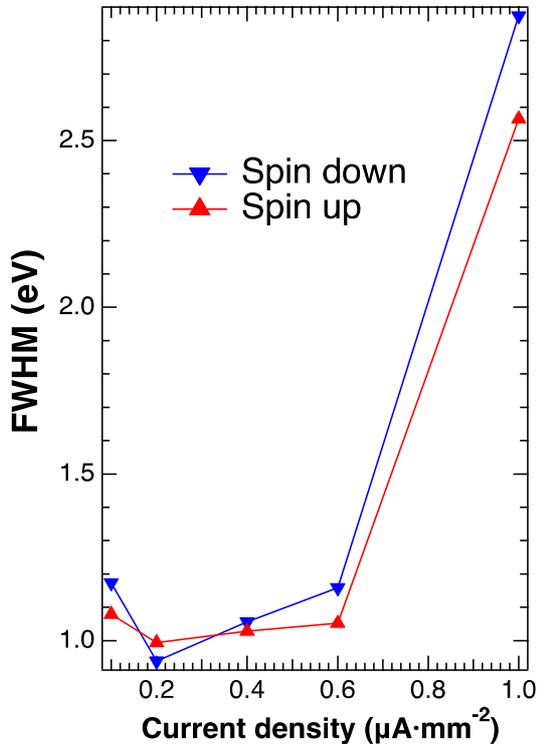


Fig. 4. FWHM of the SPIPES spectra of Au(111) *sp*-bulk state ($\theta = 46^\circ$) as a function of the current density over the sample.

In summary, cooling down the photocathode to 77 K increases the energy resolution attributed to the electron beam broadening by 30 meV while not reducing the electron beam intensity. This study has been performed by previously determining experimental conditions where the space-charge effects are negligible for the bulk state of Au(111).

The authors strongly acknowledge CNRS (Institut de Physique) for funding the spin- and angle-resolved inverse photoemission project and Labex Palm–Grant No. ANR-10-LABX-0039-PALM–for extra funds (SPOUSE and IP-OP projects). We also acknowledge funding by DIMAG project from 2019 FLAG-ERA ANR call.

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