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Spin-orbit splitting of the Shockley surface state on Cu(111)

A. Tamai,^{*} W. Meevasana,[†] P. D. C. King,[‡] C. W. Nicholson, A. de la Torre,^{*} E. Rozbicki, and F. Baumberger^{*,§}

SUPA, School of Physics and Astronomy, University of St. Andrews, St. Andrews, Fife KY16 9SS, United Kingdom

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We present angle-resolved photoemission data from Cu(111). Using a focused 6 eV continuous-wave laser for photoexcitation, we achieve a high effective momentum resolution, enabling detection of the Rashba spin splitting in the Shockley surface state on Cu(111). The magnitude of the spin splitting of $\Delta k \sim 0.006 \text{ \AA}^{-1}$ is surprisingly large and exceeds values predicted for the analogous surface state on Ag(111), but is reproduced by first-principles calculations. We further resolve a kink in the dispersion, which we attribute to electron-phonon coupling.

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Two-dimensional electron gases (2DEGs) are essential elements of electronic devices and have played a pivotal role in fundamental condensed-matter physics for decades. They are most commonly realized at the interface of conventional semiconductors and have been used extensively to study the complex phases that emerge in seemingly simple electronic systems in the presence of many-body interactions, magnetic fields, disorder, or lateral quantum confinement. More recently, 2DEGs created at the interfaces and surfaces of bulk-insulating transition-metal oxides^{1–5} and topological insulators^{6,7} have generated much interest because of their unconventional properties and potential for novel applications.

A different class of 2DEGs exists on the surface of many elemental metals. Whereas semiconductor 2DEGs derive from the confinement of bulk states into a thin layer, electronic surface states on metals result directly from the broken translational symmetry, which permits new, evanescent solutions of the Schrödinger equation in projected bulk band gaps. Prototypical examples are found at the Brillouin-zone center of the noble metal (111) surfaces. These Shockley surface states derive from the gap at L , and have a free-electron dispersion, densities of a few times 10^{13} cm^{-2} , and Fermi wave lengths of 30–80 Å. They played an important role in the study of lateral quantum confinement effects at metal surfaces^{8–11} and attracted attention as model systems for benchmarking the capability of scanning tunneling spectroscopy and angle-resolved photoemission spectroscopy (ARPES)^{12–15} in deriving intrinsic quasiparticle lifetimes in weakly correlated systems.

If confined by an asymmetric potential well breaking inversion symmetry, the spin degeneracy in a 2DEG can be lifted by the Rashba effect.¹⁶ Such nonmagnetic but fully spin-polarized electronic systems realized on metal surfaces^{17–20} or in semiconductor hosts^{7,21,22} have been investigated intensely for fundamental reasons and because of their importance for spintronic devices, most notably the spin field-effect transistor.^{23,24} In this context, the Shockley surface state on Au(111) served as an early model system permitting the first direct spectroscopic measurements of the characteristic spin momentum locking in a Rashba-split 2DEG.^{17–19} However, despite the large number of spectroscopic studies of the analogous surface states on the (111) surfaces of the lighter noble metals, a Rashba splitting could not be observed to date

for Cu and Ag and is thus generally assumed to be below the resolution of electron spectroscopic techniques.

Here we present ARPES data from Cu(111). Using a 6 eV laser focused to a spot size of $\sim 3 \text{ }\mu\text{m}$ as the excitation source, we successfully reduce the common broadening of photoemission lines due to the integration over residual inhomogeneity and roughness of the sample surface, and simultaneously increase the momentum resolution of the electron optics. Our experimental linewidths at the Fermi level are more than a factor of three narrower than in the best published data. This is sufficient to resolve for the first time a clear Rashba-type splitting in the dispersion of the Shockley surface state.

A clean Cu(111) surface was prepared by repeated cycles of Ar ion sputtering followed by annealing at 500 °C. Photoelectrons were excited using a continuous wave (cw) laser system consisting of a 820 nm diode laser with tapered amplifier (Sacher Lasertechnik) and two frequency doublers with actively stabilized bowtie cavities (LEOS solutions). This setup provides $> 1 \text{ mW}$ cw radiation at a wavelength of 205 nm ($\sim 1 \times 10^{15}$ photons/s with $h\nu = 6.05 \text{ eV}$) in a neV bandwidth and a stability of the photon energy of better than 10 μeV during a typical experimental run of a few hours. For the experiments presented here, the UV radiation was focused into a spot of $\sim 3 \text{ }\mu\text{m}$ diameter on the sample and the photon flux was reduced to around 10^{13} photons/s in order to protect the detector from excessive count rates. Control experiments were performed with a monochromatized He plasma light source. Photoelectrons were analyzed using a SPECS Phoibos 225 spectrometer. Its energy and angle resolution was set to $< 2.5 \text{ meV}$ / $\sim 0.3^\circ$ and all measurements were done at 6 K. Density functional theory (DFT) calculations including spin-orbit coupling were performed using the WIEN2K code²⁵ for bulk truncated slabs with thicknesses up to 31 layers of Cu.

The Shockley surface state on Cu(111) is an iconic electronic system and has been investigated by angle-resolved photoemission for more than 30 years.^{26–28} We therefore start the discussion of our observations by demonstrating that the laser-ARPES data is fully consistent with measurements at higher photon energy. This is nontrivial since the sudden approximation underlying the interpretation of conventional ARPES data in terms of the single-particle spectral function ultimately must fail as the photon energy approaches the ionization threshold. Eventually, the interaction of the

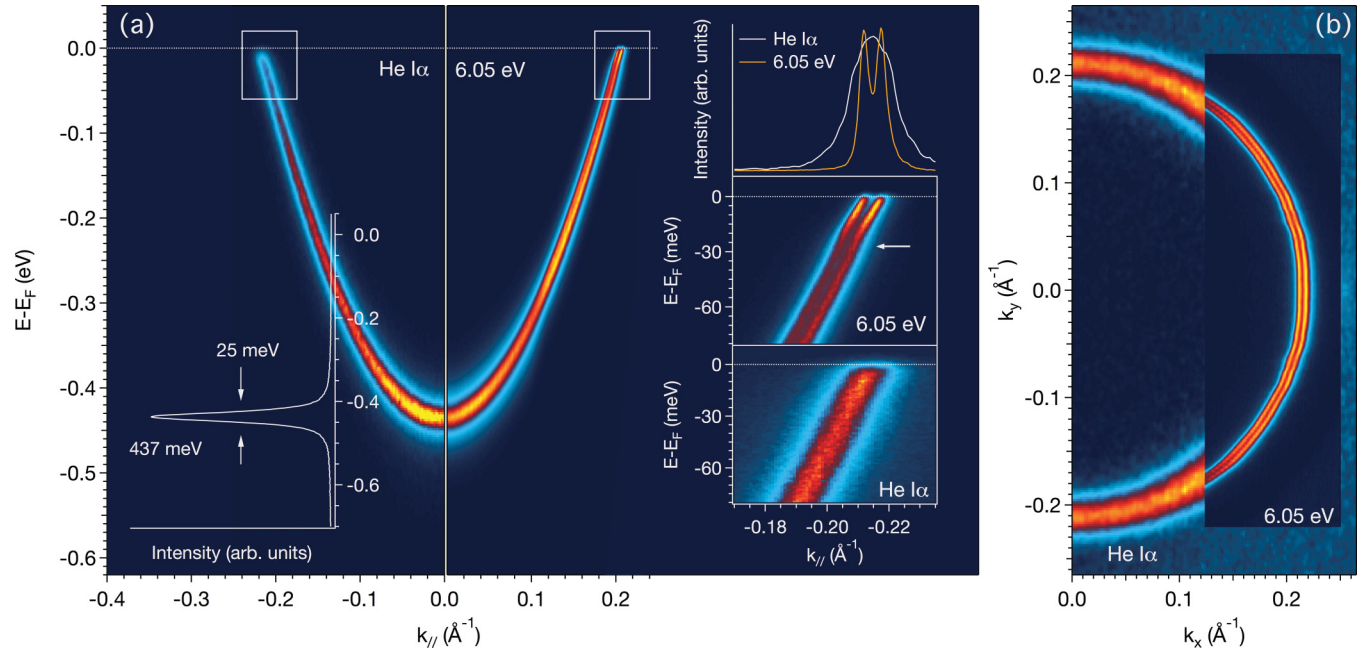


FIG. 1. (Color online) Conventional and laser-ARPES data from the Cu(111) surface state. (a) The full parabolic dispersion measured with He I α (left) and laser excitation (right). The insets show momentum distribution curves at the Fermi level (E_F) and expand the most crucial region of the dispersion near E_F , revealing the momentum-independent splitting of the dispersion, which is characteristic of Rashba systems with small wave vectors. (b) A section of the Fermi surface measured with He I α and laser excitation, respectively.

photoelectron with the $(N - 1)$ system left behind will become non-negligible and increasing relaxation of the latter might reduce the photoemission linewidth and induce a shift towards higher kinetic energy. In order to check for such signatures of a failure of the sudden approximation, we compare in Fig. 1 laser-ARPES data with measurements from the same sample performed with a monochromatized He discharge source using the He I α line with $h\nu = 21.2$ eV. The gross dispersion measured with laser excitation can be fitted with an effective mass of $\sim 0.41 m_e$ and a binding energy of ~ 437 meV, in good agreement with our He I α data and the literature.^{26–28} The quantitative agreement extends to the linewidth at the band bottom. From a fit to a Lorentzian convolved with the (nearly negligible) instrumental resolution, we find a full width at half maximum of 25 meV, in excellent agreement with the best published conventional ARPES data from Cu(111) (Ref. 28) and theoretical results.^{13,29} This strongly suggests that all many-body interactions contributing to the quasiparticle lifetime are contained in the laser-ARPES spectra.

The splitting of the quasiparticle band is barely visible on the scale of the full occupied bandwidth in Fig. 1, but revealed clearly in the expanded view of the near- E_F region in the laser-ARPES data. This panel further reveals a subtle kink in the dispersion of both branches around an energy of 30 meV, which we attribute to weak electron-phonon coupling. Although this kink is hardly discernible in the He I α data, its position and magnitude are consistent with previous measurements at higher photon energy that detected a very small reduction of the linewidth near E_F .²⁹ This confirms that low-energy loss features, including signatures of electron-phonon coupling in ARPES data taken with photon energies around 6 eV, can be

interpreted within the sudden approximation, consistent with earlier findings on cuprates.³⁰ Finally, we show in the top-right inset in Fig. 1(a) that the observed splitting is smaller than the linewidth of a resolution-limited momentum distribution curve taken with He I α radiation. Hence, our laser-ARPES data is fully consistent with conventional ARPES measurements of the Cu(111) surface. The qualitative differences from earlier work can thus be attributed to instrumental advances leading to a significantly improved resolution.

Fits to momentum distribution curves indicate that within our experimental resolution, the splitting Δk in the dispersion does not depend on binding energy and Fermi-surface angle. The dispersion is therefore described by two parabolic subbands of equal effective mass and energy at the band bottom, which is characteristic for a Rashba splitting of a free-electron-like band near the Brillouin-zone center. However, the magnitude of the splitting of $\Delta k \sim 0.0057 \text{ \AA}^{-1}$ is surprising. Contrary to naive expectations based on atomic spin-orbit splittings, we find that Δk is significantly larger than calculated for Ag(111) and only four times smaller than that found experimentally and theoretically for Au(111).^{17–19} It was, however, pointed out by several authors that such simplistic scaling relations fail to quantitatively explain existing data and density functional calculations of the band structure for a number of spin-orbit coupled systems.^{15,31} Rather, the magnitude and also the sign of the Rashba parameter appear to be dominated by the asymmetry of the wave function near the nuclei, which can be obtained from electronic structure calculations.^{31,32} Indeed, the experimentally observed splitting on Cu(111) is in fair agreement with our density functional calculations that include spin-orbit interaction, as shown in Fig. 2. This strongly supports the interpretation of our

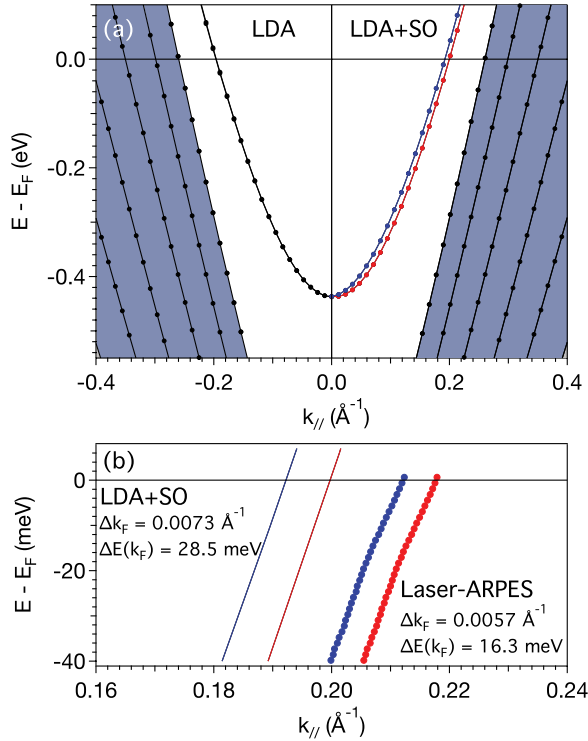


FIG. 2. (Color online) (a) Density functional calculations of the surface-state dispersion in a slab of 31 layers. The inclusion of spin-orbit interaction results in a Rashba-type splitting of the dispersion, consistent with the experiments. (b) Quantitative comparison between the local-density approximation plus spin-orbit (LDA + SO) calculation and the laser-ARPES data.

data in terms of a Rashba splitting of the Shockley surface state.

We now focus on the quasiparticle self-energy near the chemical potential extracted from our laser-ARPES data with improved resolution. The raw dispersion data displayed in Fig. 1 shows a clearly discernible kink at an energy of ~ 30 meV (white arrow), indicating a sharp structure in the self-energy Σ at this frequency. In order to extract quantitative values of Σ , we approximate the line shape of the spin-split momentum distribution curves by two Lorentzians with a width of $2 \text{ Im}\Sigma$ convoluted with a Gaussian accounting for the experimental resolution. The real part of the self-energy is obtained by subtracting a parabolic fit of the bare electron dispersion from the energy of the quasiparticle pole. The gross shape of the real and imaginary part of Σ follows a simple Debye model for the electron-phonon interaction with an Eliashberg function $\alpha^2 F(\omega) \propto \omega^2$ for frequencies up to $\omega_{\text{Debye}} = 27$ meV and a coupling constant of $\lambda = 0.16$ [dotted line in Figs. 3(a) and 3(b)], in good agreement with the value deduced in Ref. 33 from the temperature dependence of the linewidth. Assuming a momentum resolution of $\sim 0.003 \text{ \AA}^{-1}/0.3^\circ$ full width at half maximum, we find a residual quasiparticle line width $2 \text{ Im}\Sigma$ at the chemical potential of ~ 2 meV, consistent with a very high surface quality. The total phonon contribution to the linewidth, given by the increase of $2 \text{ Im}\Sigma$ between $\omega = 0$ and ω_{Debye} , is ~ 10 meV, significantly larger than found in the many-body calculations by Eiguen *et al.*,²⁹ but in excellent agreement

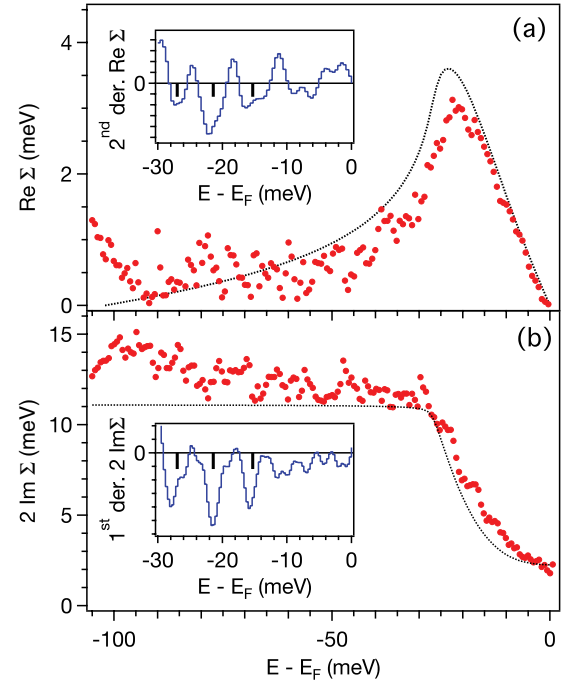


FIG. 3. (Color online) (a), (b) Real and imaginary part of the self-energy of the Shockley surface state obtained from fits to the laser-ARPES data. A Debye model of the electron-phonon interaction calculated for $T = 6$ K and a coupling constant $\lambda = 0.16$ is shown as a thin dotted line. The fine structure in the self-energy is visualized through the second derivative of $\text{Re}\Sigma$ and the first derivative of $\text{Im}\Sigma$, respectively, shown in the insets to (a) and (b). The minima of this function at 15, 21, and 27 meV indicate the frequency of maxima in the Eliashberg function $\alpha^2 F(\omega)$ and are shown as vertical bars.

with the experimental data of the same authors taken with He I radiation.^{14,29}

Intriguingly, our data show a subtle fine structure in the self-energy in the form of faint shoulders in the real part and small steps in the imaginary part at corresponding energies. This is indicative of a more complex Eliashberg function $\alpha^2 F(\omega)$ with multiple maxima.³⁴ In order to reveal their position, we show the second derivative of $\text{Re}\Sigma$ and the first derivative of $\text{Im}\Sigma$ in the insets of Figs. 3(a) and 3(b), respectively. The minima of these functions correspond to peaks in $\alpha^2 F(\omega)$ and are observed most clearly at frequencies of 15, 21, and 27 meV (marked by black bars), in excellent agreement with the dominant contributions from bulk phonon modes to $\alpha^2 F(\omega)$ calculated by Eiguen *et al.*²⁹ At very low energy, the results from our analysis are less conclusive. However, rather than the single sharp surface phonon mode at 13 meV predicted by Eiguen *et al.*, our data indicate several weak peaks of $\alpha^2 F(\omega)$ in the range of 7–11 meV. We note that resolving these features approaches the limits of the imaging precision of modern electron spectrometers. In order to minimize artifacts introduced by the electron optics, we performed these experiments without grid in front of the multichannel plate of the detector and selected a geometry that images the relevant energy range below the Debye frequency onto the most homogeneous part of the entrance slit to the hemisphere.

We finally comment on the surface sensitivity of laser ARPES. The universal curve of the electron inelastic mean free path³⁵ suggests a probing depth around 40 Å for $h\nu \sim 6$ eV, comparable to excitation with hard x rays. Yet, the intensity from the Cu surface state is large and exceeds that of bulk states in several layered materials that we investigated with the same setup. We attribute this to a particularly favorable matrix element. The Shockley surface state is located in the projected gap at L and its transition-matrix element peaks for the same perpendicular final-state momenta, which can be reached with a photon energy around 70 eV,³⁶ and are again approached at very low photon energies, as used in this study and by Gartland *et al.*²⁶ Moreover, for a surface state on a three-dimensional (3D) free-electron metal, bandlike final states are available for most excitation energies because they strongly disperse in k_z . This is generally not the case for layered

materials where bandlike final states disperse weakly in k_z and are thus available for selected photon energies only, which frequently suppresses matrix elements at low photon energies in quasi-2D materials.

In conclusion, we presented laser-ARPES measurements from Cu(111) that resolve for the first time a Rashba-type spin splitting in the Shockley surface state. This demonstrates that instrumental advances in ARPES continue to reveal unexpected effects, promising new insight even in the most widely investigated systems.

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*Present address: Département de Physique de la Matière Condensée, Université de Genève, 24 Quai Ernest-Ansermet, 1211 Genève 4, Switzerland.

†Present address: School of Physics, Suranaree University of Technology, Nakhon Ratchasima 30000, Thailand, and Thailand Center of Excellence in Physics, CHE, Bangkok 10400, Thailand.

‡Present address: Kavli Institute at Cornell for Nanoscale Science, Ithaca, New York 14853, USA.

§Present address: Swiss Light Source, Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland.

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