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Auger neutralization of He^+ on Cu surfaces: Simulation of azimuthal scans

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ABSTRACT

Charge exchange by Auger neutralization (AN) plays an important role in surface analysis techniques such as low energy ion scattering (LEIS). Recent advances in the theoretical description of AN have included a model based on a linear combination of atomic orbitals (LCAO) approach, which is able to calculate accurate neutralization probabilities of He^+ due to AN in LEIS. Previous investigations have shown that the neutralization probability is strongly influenced by the distance dependent shift of the He 1s level. In this study simulations of He^+ scattered from Cu(100) and Cu(110) surfaces at fixed azimuth angles are presented. Additionally, the azimuth dependence of ion- and neutral-yield for He^+ scattered from Cu(100) is simulated and compared to experimental data. Calculations were performed using the LCAO model in combination with molecular dynamics simulations. The excellent agreement between simulation and experiment provides evidence that the obtained values for the level shift are a characteristic property of the surface.

1. Introduction

Low energy ion scattering (LEIS) is a very popular method to investigate the composition and structure of metal and semiconductor surfaces [1,2]. In this technique, a beam of noble gas ions, typically He^+, with incident energies, E_0, in the range of several hundred eV to several keV is directed on the surface of interest. When the energy distribution of backscattered ions is analyzed, LEIS exhibits excellent surface sensitivity, which is owed to efficient neutralization of He^+ within the target material. Besides its application as a surface analysis technique [3–6], LEIS is in consequence also an excellent tool to study charge exchange mechanisms between He^+ ions and metal/semiconductor surfaces.

The state of the art description of charge exchange in the LEIS energy regime distinguishes between three different mechanisms, which lead to neutralization of ionized projectiles: Auger neutralization (AN), resonant neutralization in a close collision [1] and quasi-resonant neutralization [7,8]. Due to these mechanisms, only a certain fraction of backscattered projectiles, P^+, arrive at the detector in an ionized state. The presence and the efficiency of the different neutralization mechanisms depend on the projectile target combination as well as the primary energy of the projectile. For the sake of completeness, one should also mention that for certain conditions neutralized projectiles can undergo resonant ionization in a close collision [1] and Auger-ionization [9], which leads to an increased ion yield.

For He^+ ions scattered from Cu surfaces, AN is the only possible mechanism for charge exchange when the empty He-1s state is below the conduction band of the target. In this configuration, an electron from the target can tunnel to the ion while the gain in binding energy is dissipated via excitation of an electron from the conduction band or a plasmon, respectively.

The present study will focus on the theoretical description of AN in LEIS. Recent advances in this field have included the use of a linear combination of atomic orbitals (LCAO) approach to calculate AN-probabilities [10]. This LCAO theory is able to calculate P^+ for a given scattering geometry with good accuracy. However, investigations have shown that the distance dependent shift of the He 1s level has a large influence on the probability of AN [11]. Two different questions will be discussed in detail in the following sections:

(i) It is known that not all Cu surfaces are described equally well with the same model for the distance dependent level shift [11]. What are the differences in the level shift models which lead to best agreement between theory and experiment for the Cu(100), Cu(110) and polycrystalline Cu surfaces? Is it possible to correctly reproduce first and second layer contributions to the measured signal with a single model for the level shift?

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(ii) Is this model for the level shift additionally able to simulate more complex scenarios like neutral and ion yields as a function of azimuthal angle (azimuthal scans) of He^+ scattered from low index surfaces of Cu(100)?

Atomic units ($e = \hbar = m_e = 1$) will be used throughout this investigation unless otherwise stated.

1.1. Theory and simulation

The probability of AN is commonly described by an AN transition rate, Γ. Since the pioneering work of Hagstrum [12], many different studies were concerned with developing a accurate theory of AN at metal surfaces [13–17]. The present investigation makes use of a LCAO approach which was developed recently. For a thorough description of the theory we refer the reader to Refs. [18,19]. This theory has already been successfully applied to study experimental results in LEIS [20,11].

A key quantity for the calculation of the AN-rate is the energy level, E_a, of the He 1s state with respect to the Fermi level. E_a, is not constant, but may change as the projectile approaches the target: At comparatively large ion-surface distances ($\sim$5 a.u.), the image charge interaction leads to a noticeable decrease in the binding energy of the He 1s level [21]. At closer distances, the interaction between projectile and conduction band electrons of the target can lead to an increase in the binding energy. At even closer distances the interaction with core level electrons of the target will finally lead to a strong promotion of the level [22]. When the ion-atom distance is smaller than a critical distance, r_{min}, the He 1s level is promoted such that it is in resonance with the conduction band and resonant neutralization in a close collision starts to be effective. For a given scattering geometry an ion-atom distance smaller than r_{min} requires E_0 to exceed a certain threshold energy E_{th}. For the He – Cu system E_{th} amounts to $\sim$2 keV [1]. For the sake of completeness, one should also mention that eventually, the level will cross E_F and AN will cease to be active, while it is possible to observe resonant ionization (RI) of already neutralized projectiles. In the present study, incident energies are chosen in order to restrict charge exchange to AN ($E_0 \leqslant E_{th}$).

It has been shown in [11] that the distance dependent shift of the He level is an important parameter in the calculation of Γ. However, experimental data on this effect is very scarce, as only few investigations were concerned with studying this shift for specific systems [23,24]. Unfortunately, the exact distance dependence of the level shift for the He – Cu system is not known. As a consequence, the present investigation uses the following approximations:

(i) Constant level shift: here the He level is assumed constant at a certain energy below E_F.
(ii) Hard wall level shift: this model is similar to model (i) except for a steep rise of E_a at a position corresponding to r_{min}. The hard wall level shift model was already applied in [11]

To calculate P^+ due to AN, one can make use of the rate equation $dP^+ = -P^+\Gamma dt$, to deduce the following expression:

$$P^+ = e^{-\int \Gamma(\vec{r}(t))dt} \tag{1}$$

To evaluate this equation, one needs knowledge of the projectile trajectory, $\vec{r}(t)$, and the AN-rate along this trajectory. Since the calculation of Γ for each point of the trajectory would be far too time consuming, Γ is calculated for specific points at the surface under investigation and $\Gamma(r)$ is determined by linear interpolation between these points.

Projectile trajectories are calculated with the molecular dynamics (MD) simulation package Kalypso [25]. Collision processes between projectile and target atom were modeled by the Thomas–Fermi–Moliere potential with Firsov screening length and a correction factor of 0.75 [26]. Lattice vibrations of the target atoms and electronic stopping of the projectiles were not considered in the simulation.

For simulations at a specific azimuth angle, only the trajectories are considered where the projectile is backscattered in a single collision from a first or second layer atom. To save computation time, the acceptance angle in the simulation was chosen slightly wider (±1.6°) than in the experimental setup (±0.46°).

For the simulation of azimuth scans the following procedure was applied: it can be estimated, that for 2 keV He^+ and Cu(100) only a very small amount of primary projectiles ($\sim$1%), which are backscattered from atoms below the second layer contribute to the ion yield. Thus, MD-simulations were performed for thick targets (8 layers) and for thin targets (2 layers). Simulations contain an adequate number of trajectories with impact parameters located within a reduced unit cell utilizing the crystal symmetry. Projectiles with a scattering angle of 129° (±1.6°) and a final energy within 15 eV of the single scattering energy were filtered. It was tested that a change in the acceptance angle does not lead to significantly different results. The remaining backscattered projectiles were sorted with respect to their azimuthal exit angle, using angular bins of 6° size. The yield of projectiles which are backscattered into a certain azimuthal bin is denoted $N(\phi)$ and $N_2(\phi)$ for the thick and thin target, respectively. The difference $N_d(\phi) = N(\phi) - N_2(\phi)$ is automatically considered neutral. Besides this contribution to the neutral yield from deeper layers, there is a certain amount of primary projectiles which will be neutralized by AN in the course of being backscattered from a first or second layer atom. To calculate this amount, P^+ according to Eq. (3) was evaluated for all filtered projectiles in the simulation with the thin target. From these results, a mean P^+ for a certain azimuthal bin was deduced, $P^+(\phi)$. Based on $P^+(\phi)$ it is possible to calculate the yield of ions, $A^+(\phi) = N_2(\phi) \cdot P^+(\phi)$, and the yield of neutrals $A^0(\phi) = N_d(\phi) + N_2(\phi) \cdot (1 - P^+(\phi))$, respectively.

2. Experiment

Experimental results in the present investigation were recorded in the TOF-LEIS setup ACOLISSA [27]. Cu single crystal samples were cleaned by multiple sputtering-annealing cycles. Surface cleanness was checked by Auger-electron spectroscopy and LEIS. LEED images were recorded in order to check surface crystallinity, and exhibited clear spots demonstrating well-defined (100) and (110) surfaces. When a crystal is in a double alignment condition with respect to primary beam and detector axis, P^+ can be evaluated in a straightforward way using the yield of ions A^+ and the yield of neutrals A^0:

$$P^+ = \frac{A^+}{A^+ + A^0} \tag{2}$$

To separate ions from neutrals our setup makes use of a post-acceleration lens located between sample and detector. To obtain A^0, only the single scattering peak in the neutral spectrum is evaluated. However, this peak is located above a background signal which is due to backscattering from deeper layers [28]. Consequently, A_0 can be determined in two different ways: either by subtracting this background or by considering this background a part of A_0. Experimental results for P^+, which are presented in the next section, were evaluated with the background subtracted A_0.

When P^+ is plotted as a function of the inverse perpendicular velocity, $1/v_\perp = 1/v_{\perp,in} + 1/v_{\perp,out}$, a single-exponential fit to the data ($P^+ = exp(-v_c/v_\perp)$) yields the characteristic velocity v_c, which is