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Velocity Scaling of Ion Neutralization in Low Energy Ion Scattering

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The ion fraction P^+ is measured for He$^+$ ions scattered by 129° from a Cu surface. Both the primary energy and the angles of incidence and of exit are varied. From our results we conclude the following: along the incoming and outgoing trajectories, neutralization is due to Auger processes and depends on the normal velocity component $v_\perp$ only. At higher energies, additional charge exchange is due to collision induced neutralization and reionization, both depending on the total ion energy only. Also in this regime P^+ depends on $v_\perp$, but via a two-valued function of the scattering geometry at fixed energy.

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In a typical low energy ion scattering (LEIS) experiment, a (solid) target is bombarded with noble gas ions (He$^+$) with a primary energy E_0 of ~1–10 keV, at perpendicular incidence ($\alpha = 0$), while ions scattered by a large angle θ are detected. In this regime, projectiles are scattered from surface atoms almost exclusively by binary collisions. If only ions are detected, sensitivity to the outermost atomic layer is gained, since He projectiles that are backscattered from deeper layers leave the surface as neutral He atoms [1]. On this basis, LEIS has become a widely used surface analytical tool of quantitative composition and structure analysis [2–4].

In LEIS, information on the concentration of a certain species in the surface is obtained from the intensity of an ion peak at a certain energy [3], from the knowledge of the scattering cross section $d\sigma/d\Omega$, the ion fraction P^+, and instrumental parameters such as detector solid angle and detection efficiency. The Thomas-Fermi-Moliere potential, which is a screened Coulomb potential [5], is a reasonable basis to calculate $d\sigma/d\Omega$. The theoretical basis for P^+ is less safe. Since the pioneering work by Hagstrum [6], it is generally accepted that Auger neutralization (AN) plays an important role, at least at low energies. Hagstrum derived a relation for the probability P_A^+ to survive AN in the charged state when approaching or leaving the surface. Because of the nonlocal character of AN, P_A^+ depends on the velocity component $v_\perp$ perpendicular to the surface ($v_\perp = \vec{v} \cdot \vec{e}$ where $\vec{e}$ is the surface normal):

$$P_A^+ = \exp(-v_c/v_\perp). \tag{1}$$

Here, v_c characterizes the neutralization efficiency and has the dimension of a velocity, since $v_c = \int ds[1/\tau_A(s)]$ is obtained by integration of the Auger transition rate $1/\tau_A(s)$ from 0 to ∞ over the distance s to the surface. Note that in this context the ion velocity v—and its parallel component $v_\parallel$—is always small compared to the target Fermi velocity v_F. Consequently, the effective occupation of the target states in the rest frame of the projectile is very well described by the Fermi-Dirac distribution and therefore shifted Fermi sphere effects [7] are negligible here [8].

Van Ween and Haak [9] introduced a local model, where neutralization takes place in an interaction between the projectile and one target atom and an expression for P^+ is obtained, which contains a different constant V_c and depends on the (total) ion velocity v:

$$\mathrm{P}_l^+ = \exp(-V_c/v). \tag{2}$$

Apart from neutralization along the trajectory discussed so far, charge exchange may occur also by a local process (the close collision with the backscattering center), leading to collision induced neutralization (CIN) and reionization (CIR) [10]. Since these processes require a distance of closest approach in the collision smaller than a critical value r_{th}, the associated probabilities P_{CIN} and P_{CIR} have nonvanishing values for a given scattering angle only at energies E larger than a certain threshold energy E_{th}. At a given energy E, the values of P_{CIN} and P_{CIR} depend only on the impact parameter (or equivalently on the scattering angle), and are independent of the scattering geometry (angles α andβ).

The survival probability P^+ for the total trajectory is then obtained as (see, e.g., [11])

$$P^+ = [P_{\mathrm{in}}^+(1 - P_{\mathrm{CIN}}) + (1 - P_{\mathrm{in}}^+)P_{\mathrm{CIR}}]P_{\mathrm{out}}^+. \tag{3}$$

P_{in}^+ and P_{out}^+ denote the survival probability on the ingoing and outgoing trajectory, respectively, and may be calculated either from Eq. (1) or from Eq. (2). The two addands in Eq. (3) describe survivals and reionized projectiles, respectively. At $E < E_{\mathrm{th}}$, $P_{\mathrm{CIN}} = P_{\mathrm{CIR}} = 0$, and Eq. (3) simplifies to $P^+ = P_{\mathrm{in}}^+ P_{\mathrm{out}}^+ = e^{-v_{cx}(1/v_{0x}+1/v_{fx})}$, where v_{0x} and v_{fx} denote either v_0 and v_f (initial and final velocity) or their perpendicular components $v_{0\perp}$ and $v_{f\perp}$. Depending on the choice of v_{0x} and v_{fx}, v_{cx} denotes either V_c or v_c.

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Summarizing the experimental data available at that time, Boers [12] concluded that the most promising ansatz would be $P^+ = P^+(0)e^{-v_c/v_f}$ with $P^+(0)$ the probability that the projectile is in the positive charge state immediately after the close collision. There is an ongoing debate, whether for a given target P^+ depends intrinsically on v or on $v_\perp$ or just on the final velocity v_f. Note that P^+ measurements varying only the ion energy cannot distinguish between total and normal velocity scaling. In [13], both the ion energy and scattering geometry were varied, but an unresolved dependence of v_c on v was found for $P^+_{\rm out}$. More recently, mostly energy dependent measurements were performed and results were presented as a function of v [14,15]. Lately, for Ne neutralization in noble/transition metals, the introduction of a preexponential factor was suggested in order to properly describe $P^+(v)$, without proper foundation [16].

Ab initio calculations showed the importance of collision induced processes and the good qualitative validity of Eq. (3) [17,18], while the absolute values of P^+ for He$^+$ and Al were, due to the complexity of the model, only in fair agreement with experiment.

The aim of this study is to understand how P^+ is determined by the interplay of different neutralization processes in LEIS. To reach this goal, P^+ has to be measured for a prototypic system with a high threshold energy $E_{\rm th}$ [10], so that both neutralization regimes ($E < E_{\rm th}$ and $E > E_{\rm th}$, respectively) are accessed. In these experiments, both the primary energy E_0 and the scattering geometry (α, β) have to be varied. From this, it should become clear how P^+ evolves.

Therefore, we performed LEIS measurements for polycrystalline Cu targets (a Cu film evaporated onto a Si substrate and a massive Cu sheet) and He ions in the range 1–7 keV. The scattering angle $\theta = \pi - \alpha - \beta$ was 129°, and the scattered ions were analyzed by means of a time-of-flight (TOF) measurement. The angles α and β were varied in the range from 0° to 50°, thereby rendering contributions from multiple scattering negligible [19,20]. Along part of the outgoing trajectory, ions and neutrals were separated by postacceleration of the ions. A typical spectrum is shown in Fig. 1. The number of detected ions A_+ of the ion peak is given by

$$A_+ = N_0 n_s \frac{d\sigma}{d\Omega} \Omega P^+ \eta_+. \qquad (4)$$

Here, N_0 is the number of primary ions, n_s the number of surface atoms per area, Ω the solid angle, and η_+ the detection efficiency for the ions. In the single scattering approximation [21], the height of the neutral spectrum H_0 at the kinematic high energy limit kE_0 (with k the kinematic factor) is given by

$$H_0 = N_0 \frac{\epsilon}{[\varepsilon_e]} \frac{d\sigma}{d\Omega} \Omega (1 - P^+) \eta_0. \qquad (5)$$

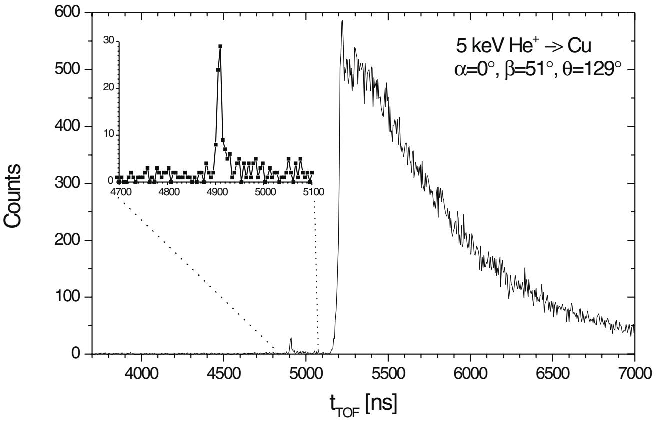

FIG. 1. TOF-LEIS spectrum of 5 keV He$^+$ projectiles scattered from a Cu target, measured with postacceleration to separate ions (inset) and neutrals.

Here, ϵ is the energy width of one channel, $[\varepsilon_e]$ the electronic stopping cross section factor [22,23], and η_0 the detection efficiency for the neutral He atoms.

The ion fraction P^+ was obtained in two independent ways: (i) from the ratio A_+/H_0 (see Fig. 1), and (ii) from Eq. (4). The former method [24] is based on the fact that in a Rutherford backscattering spectrum H_0 is independent of multiple scattering [25]. Within the single scattering approximation, this also holds true for LEIS spectra. The validity of this assumption in the present regime was shown in [26] for $0.8 < E_f/kE_0 < 1$. The latter method is well established. Both data sets yielded concordant results within 20% using the stopping power from [23] and the detector efficiency from [27,28], which gives confidence to the accuracy of our data, in view of the complementarity of the two evaluation procedures and their possible systematic errors.

In Fig. 2, the resulting P^+ data are presented in a semilogarithmic plot as a function of $1/v_\perp \equiv 1/v_{0\perp} + 1/v_{f\perp}$. Three different regimes (I–III) can be distinguished. For

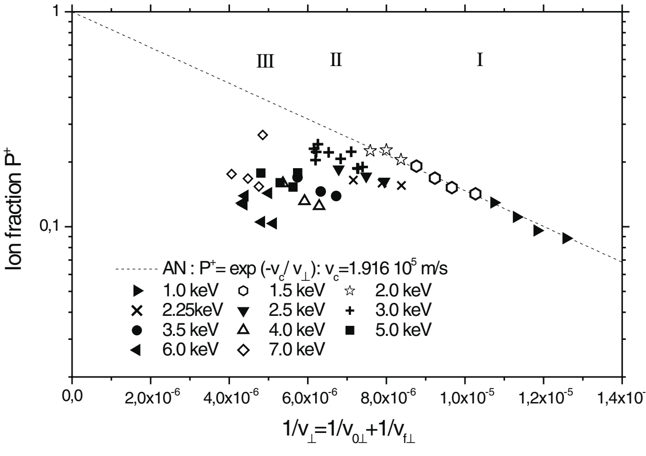

FIG. 2. Experimental results for the ion fraction P^+ for He$^+$ projectiles scattered from a Cu target, as a function of $1/v_\perp = 1/v_{0\perp} + 1/v_{f\perp}$ (see text).

large $1/v_\perp$ values (regime I, corresponding to primary energies 1–2 keV) all the data follow a single straight line (within the experimental uncertainty of 7%) independent of the actual scattering geometry (α, β). The data are perfectly described by Eq. (1) with $v_c = 1.916 \pm 0.012 \times 10^5$ m/s. In this regime of low energies, only the nonlocal neutralization mechanism (Auger neutralization) is active, and P^+ is a unique function of $1/v_\perp$. The so determined value for v_c is in excellent agreement with results from literature obtained for smaller scattering angles [29]. By conversion of the results reported in [14,15] from a $1/v$ to a $1/v_\perp$ scaling, a similar value ($v_c = 1.6 \times 10^5$ m/s) can be extracted.

At higher energies, a sudden decrease in P^+ is observed, at 2.25 keV and at ~3.5 keV. The intermediate regime (II) is established by data in the range of primary energies between 2.25 and 3 keV. Its main feature is a pronounced decrease of P^+ to a value of ~75% with respect to the extrapolated AN line, obtained at low energies. However, the slope of P^+ seems to be very similar to that in the low energy regime. The transition from low energy to intermediate energy regime is quite abrupt (between 2.0 and 2.25 keV), in perfect agreement with the reported threshold energy $E_{\rm th}$ for reionization [14]. The physical origin of the decrease in P^+ is obviously, that collision induced processes start to contribute, with $P_{\rm CIN} > P_{\rm CIR}$ (otherwise P^+ would increase rather than decrease), in qualitative agreement with theoretical findings [17,18]. Assuming $P_{\rm CIR} = 0$, the reduction factor of ~0.75 could be interpreted as (1-$P_{\rm CIN}$) according to Eq. (3), if $P_{\rm CIR} > 0$ follows $P_{\rm CIN} > 0.25$. An important consequence of the presence of collision induced processes is that the data seem to extrapolate to a value different from 1 for $1/v \rightarrow 0$. This is a very general finding, which is valid for any target at energies $E > E_{\rm th}$. Note that a larger scatter of the data is observed in regime II, although the precision of the data is the same as at low energies (regime I).

In Fig. 2, in regime III (3.5 to 7 keV) another decrease in P^+, and again a strikingly large scatter of the data, measured at fixed energy by variation of the geometry (α and β), is observed. In order to elucidate the seeming spread of the data, we show in Fig. 3 just the regimes II and III (from 2 to 7 keV), together with model calculations for P^+, with proper choice of $P_{\rm CIN}$ and $P_{\rm CIR}$. It is obvious that the variation of α and β yields P^+ values, which are not a unique function of $v_\perp$. This is because for a given system $P_{\rm CIN}$ and $P_{\rm CIR}$ are only a function of E and impact parameter b, while the probability for AN depends only on $v_\perp$. As a consequence, for a given value of $v_\perp$, grazing incidence will lead to $P^+_{\rm in} \ll 1$ and $P^+ \approx P_{\rm CIR} P^+_{\rm out}$, and grazing exit will strongly reduce P^+ in any case. Consequently, for a specific value of $1/v_\perp$ and of θ, two P^+ values exist, depending on geometry, with an apex at $\alpha = \beta$. The separation between these two values is mainly due to $P_{\rm CIR}$; since in the present regime $P_{\rm CIR}$ increases with increasing energy, the two values are further separated at higher energies. Thus, the probabilities for the collision induced processes can be deduced from the data (see Fig. 4). Note that these findings apply to the entire LEIS regime, since v_c and $E_{\rm th}$ are the only specific properties of Cu that enter. It is the present choice of energy range, scattering angle, and scattering geometry, and the variation of both, the geometry and the energy, which made it possible to reveal how the observed P^+ evolves out of local and nonlocal charge exchange processes, which depend on velocity and energy, respectively.

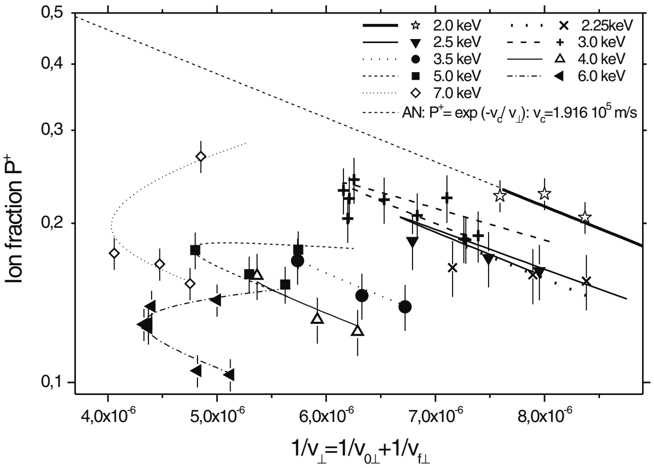

FIG. 3. Experimental results for incident energies ≥ 2 keV for the ion fraction P^+ for He$^+$ projectiles scattered from a Cu target, together with model calculations based on Eq. (3) (lines).

Finally, the limiting behavior of P^+ for $1/v_\perp \rightarrow 0$ should be considered. In the energy range of experiments presented here, the extrapolation of $P^+(1/v_\perp \rightarrow 0)$ yields a value <1. At higher velocities, $P_{\rm CIN}$ and $P_{\rm CIR}$ will

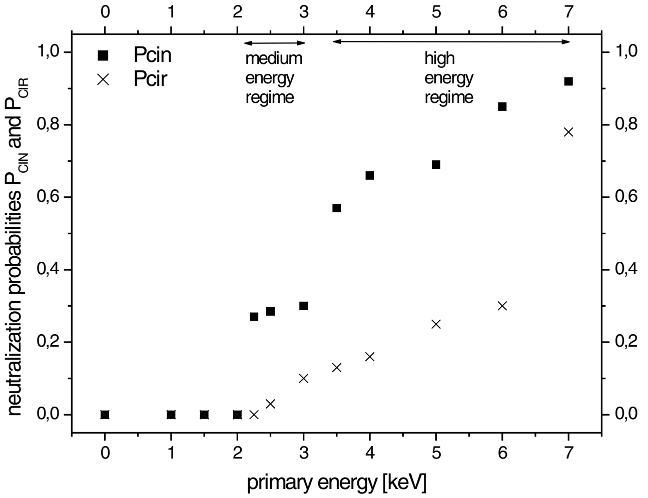

FIG. 4. Results for the probabilities $P_{\rm CIN}$ and $P_{\rm CIR}$ for collision induced neutralization and reionization, as a function of the primary energy E_0.

eventually decrease again, due to the reduction of interaction time, and lead to $P_{\mathrm{CIN}} \ll 1$ and $P_{\mathrm{CIR}} \ll 1$, so that $P^+ \approx P^+_{\mathrm{in}} P^+_{\mathrm{out}}$ will be valid again, unless new inner shell effects come into play.

To summarize, from the present investigation one can draw the following conclusions: below the reionization threshold ($E < E_{\mathrm{th}}$), charge exchange in LEIS is entirely due to a nonlocal process (Auger neutralization), which only depends on the perpendicular component of the velocity $v_\perp$, and P^+ is perfectly described by Eq. (1). At higher energies ($E > E_{\mathrm{th}}$), P^+ is governed by local processes (collision induced neutralization and collision induced reionization) and by a nonlocal process (Auger neutralization), and thus depends on the energy as well as on $v_\perp$. From experiments like the one presented here, P_{CIN}, P_{CIR}, and v_c can uniquely be determined for any system. These findings are generally valid and reveal the relevance of different charge exchange processes and the scaling properties of the ion fraction in LEIS.

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