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Neutralization of energetic He ions scattered from clean and Cs-covered Si(100)

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We report on a comprehensive study of the neutralization of (75—180)-keV He ions scattered from the UHV-cleaned and Cs-covered Si(100) surface, using surface-sensitive channeling techniques. It is shown that ions are neutralized exclusively at the solid surface on the ion's outward path. Angular depth and work-function dependence results are discussed. A model is proposed which includes resonant transitions to a broadened He $n=2$ quantum level, and is compared with experimental data.

I. INTRODUCTION

Ion neutralization in solids, a process basic to our understanding of fundamental ion-solid interactions, has been an active and challenging area of research for many years. Most theoretical and experimental efforts have focused on electron capture and loss in the low-ion-velocity or adiabatic regime.[1] In this regime the Born-Oppenheimer approximation for decoupling the nuclear and electronic motion is employed and theoretical analysis has shown that Auger transitions and tunnel-resonance processes (where possible) are the dominant means of ion neutralization. Recently, theoretical and experimental studies concerning the neutralization of low-velocity He (Ref. 2) and Cs (Ref. 3) ions at solid surfaces have shown that tunnel-resonance processes make a larger contribution than expected due to broadening of the resonant ion levels which allows overlap with electronic states in the solid. In the high-ion-velocity regime ($v > v_0$ where v_0 is the Bohr velocity) the Born approximation is employed.[4,5] Here, *ab initio* calculations employing simple solid wave functions have achieved only qualitative success and extrapolation to the intermediate-velocity regime ($v \sim v_0$) studied here has yielded poor agreement. Most of the ion-neutralization studies in this regime have been carried out in gases and scaling of these results to solid densities has provided a means of predicting ion fractions of ions scattered from solid surfaces.[6] This scaling is reasonable from the standpoint that electron capture at very high ion velocities occurs from the core levels of the target atoms. Comparisons of gas-phase results in the intermediate-velocity regime[7] to those presented in this paper yield poor agreement and hence suggest that further elucidation of the physical mechanisms involved in charge transfer at these velocities is needed.

Recently, increased interest from both fundamental and practical considerations has attracted attention to the intermediate-ion-velocity regime, i.e., He ion energies of $\sim$50—200 keV. Here the ion moves at velocities comparable to electron velocities in the solid and hence complicates theoretical attempts to study neutralization processes in this regime.

From a practical viewpoint, this energy regime has become increasingly important in ion-scattering studies of clean and adsorbate-covered crystal surfaces.[8] Quantitative studies of surface structures with the use of electrostatic analyzers necessarily require information on the neutralization processes which determine the scattered ion fraction. Previous neutralization studies have been carried out almost exclusively on so-called "practical" surfaces where the state of the solid surface is not well characterized.[9-11] This situation has led to considerable speculation on the contribution of carbon and oxide layers to the determination of the ion fraction and has prevented the formulation of a clear understanding of the neutralization processes which occur at these energies.

In this paper we report on a study of the neutralization of He ions scattered from the clean and cesium-covered Si(100)2×1 surface prepared and studied under UHV conditions. We have found that (1) neutralization occurs at the solid surface with no evidence of a dependence on the depth from which the ion scatters, and (2) there exists no observable dependence of the ion fraction on the scattered ion takeoff angle. These findings differ with results from low-velocity experiments (10 eV to 10 keV) which show both angular and scattering depth dependences.[12,13] We have also observed interesting changes in the ion fraction upon adsorption of cesium on the clean Si surface. We propose a model which explains not only our experimental observations but also trends expected in the low- and high-velocity regimes.

The sensitivity of the ion fraction to surface cleanliness was demonstrated in two studies in which a consistent difference was observed between practical and Ar-ion-bombarded or annealed surfaces, although in those cases detailed knowledge of the state of the surface was lacking.[14] These observations have motivated us to perform our measurements in an ultrahigh vacuum chamber with a base pressure of $\sim 2\times 10^{-10}$ Torr, coupled via differentially pumped sections to an Ortec 300-keV ion accelerator. Figure 1 shows a schematic of the experimental setup and idealized channeled scattering spectra. The Si surface was prepared by standard cleaning techniques.[15] The surface phase which was monitored with a "view from behind" low-energy electron diffraction system, exhibited a sharp "2×1" pattern, characteristic of the clean Si(100) surface. A He ion beam (75–200 keV) was directed onto the sample and the scattered ions were detected by a surface-barrier detector mounted behind a pair of electrostatic deflection plates. With the application of sufficient voltage to the deflection plates, scattered ions were swept from the path of the detector permitting only neutral He to be measured. Spectra of both ions and neutrals were collected with the deflection plates at ground potential. The ratio of ions to ions plus neutrals is called the ion fraction. Channeling combined with grazing exit-angle detection of the scattered particles provided enhanced surface selectivity and accurate comparison between the surface and bulk scattered ion fractions.[16]

The scattered yields associated with the Si surface peak and the deposited Cs were accurately converted to areal densities via comparison with both a Bi-implanted Si standard and the scattered intensity from the Si sample oriented in a nonchanneling direction. Each ion-fraction data point required two spectra whose accumulated dose was 7.5×10^{14} He ions/cm^2. The statistical error associated with the number of counts in the Si surface peak was approximately ±3% and ±1–5% for the Cs (depending upon the amount of adsorbed Cs present). Heavy metal impurities, monitored with ion scattering, were present on the surface in concentrations of $<1\times 10^{13}$ atoms/cm^2 (<0.01 ml). No carbon or oxygen was observed with ion scattering.

II. DISCUSSION OF THE DATA

A. Depth dependence

The sensitivity of the ion fraction to surface cleanliness was first pointed out by Buck *et al.*[14] In their measurements a consistent discrepancy between a practical and Ar-ion-bombarded "clean" surface was observed although even in that case detailed knowledge of the state of the surface was not known. A systematic study of neutralization at clean and cesium-covered Si surfaces is presented here. Figure 2 shows the He ion fraction as a function of the scattered projectile velocity for incident energies between 75 and 180 keV. The He ion beam has been directed along the ⟨001⟩ crystallographic direction which is normal to the (100) surface. The open circles represent the ion fractions of those particles which have scattered from the surface only, i.e., from the measured integrated surface peak intensities. The solid circles correspond to the measured ion fractions from particles which are directed along a nonchanneling or so-called random direction. The surface sensitivity from channeling is derived from the shadowing of subsequent atoms by the first in each crystal string and hence is independent of the resolution of the detection system. At 150 keV the ion beam in-

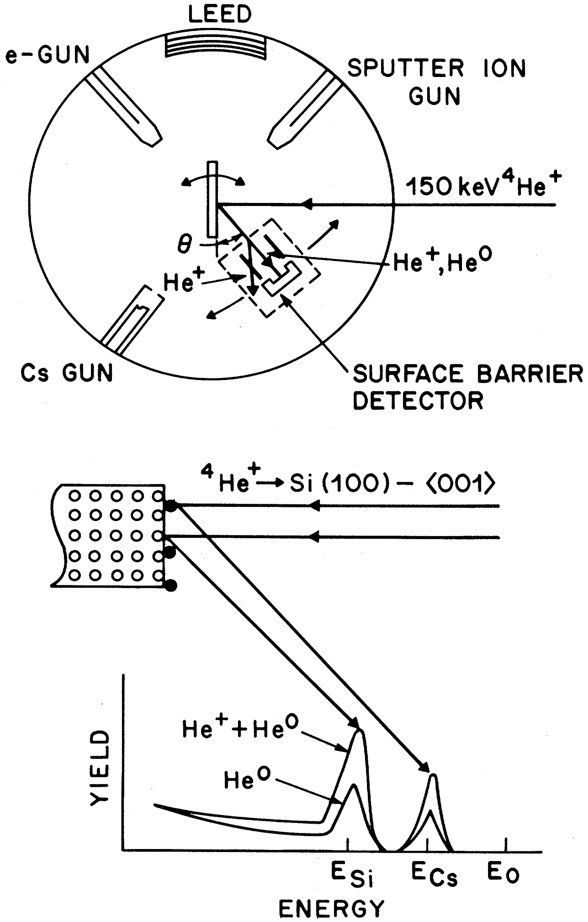

FIG. 1. Sketch of the experimental apparatus and idealized channeled scattering spectra of both neutrals and ions plus neutrals.

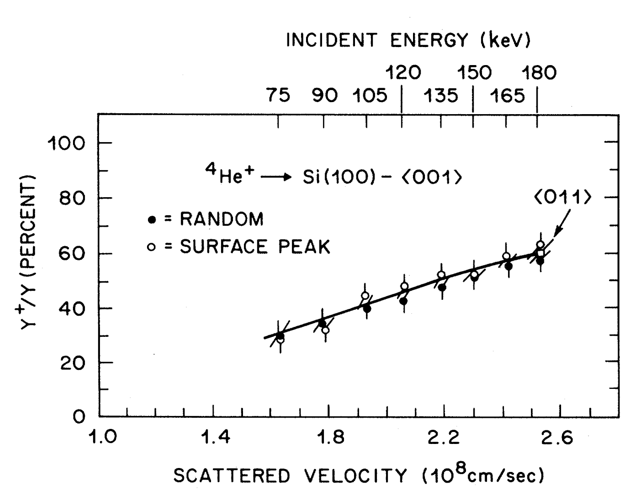

FIG. 2. Ion fractions from surface peak (open circles) and random (solid circles) measurements as a function of scattered He ion velocity. The solid curve is a guide to the eye.

teracts with only 1.8±0.15 atoms (~10 Å) in each ⟨001⟩ string and at 75 keV the He beam see only 1.3±0.17 atoms per row (~7 Å).[17] Conversely, when the beam is aligned in a random direction, the depth sensitivity is a function of the resolution of the detector which in our measurements corresponds to about 30 Å. The agreement between surface and random data shows conclusively the lack of any strong depth dependence in the neutralization process. In measurements to be described later we show that this lack of depth dependence continues to hold for any scattering depth. These experimental results are in reasonable agreement with those of Buck *et al.*[11] and represent not only a concurrence with their findings, but also an increase in the precision allowing an improved depth sensitivity of the scattered ion fraction. The use of channeling and grazing exit-angle detection has allowed us to set a limit on the depth below the surface required to determine the ion fraction observed, at 7 Å, which is the depth resolution of the experimental technique. Furthermore, the use of atomically clean surfaces, which differs from the practical or Ar-bombarded surfaces used in Buck's study, has allowed us to show conclusively that the ion fraction is determined at the solid surface. Buck's ion fraction versus scattered velocity data is also consistently ~4% lower than results from this study and may be due to impurities on their Si surface or depth-sensitivity effects which fold in lower-velocity scattered particles (energy loss associated with the subsurface scattered particles) with those which scatter from the surface. These findings are reinforced in the next subsection where the results of a controlled cesiation of the Si surface and the subsequent effects on the scattered ion fraction are described. To determine the dependence of the ion fraction on the incident-ion-beam direction, the He beam was directed along the ⟨011⟩ crystallographic axis which is 45° from the normal ⟨001⟩ axis. A measurement of the ion fraction for this configuration is represented by the open square in Fig. 2 and agrees with measurements made along the ⟨001⟩ direction, within experimental error. The conclusion here is that the ion fraction is insensitive to the incident-beam direction as well as scattering depth. The solid curve is drawn to guide the eye.

B. Angular dependence

In Fig. 3 the ion fraction for 150-keV He ions is measured as a function of takeoff angle with respect to the surface for both channeling and random orientations. Here, too, the data show good agreement between the two cases. Further, there is no evidence of any angular dependence, in good agreement with results at somewhat higher velocities obtained by other workers.[18] Adiabatic theory (dashed curve in Fig. 3), predicts the ion survival probability to vary as $e^{-v_c/v\sin\theta}$ where θ is the projectile takeoff angle and v_c is a characteristic velocity.[12] This model is in clear disagreement with the experimental results presented here. Low-velocity theory and experiment show an ion fraction approaching 0 at grazing exit angles and which increases exponentially with increasing exit angle. It is in the angular range studied here that we see the remarkable contrast between low-velocity results and those of the higher-velocity regime studied in this experiment. A small but noticeable slope is present in the data and is due to the decrease in scattered ion velocity as the scattering angle is increased, simply a kinematic effect. These new results clearly show that the ion fraction observed depends only on the speed of the outgoing ions and not their velocity. Henceforth, the use of velocity here implies its absolute magnitude only.

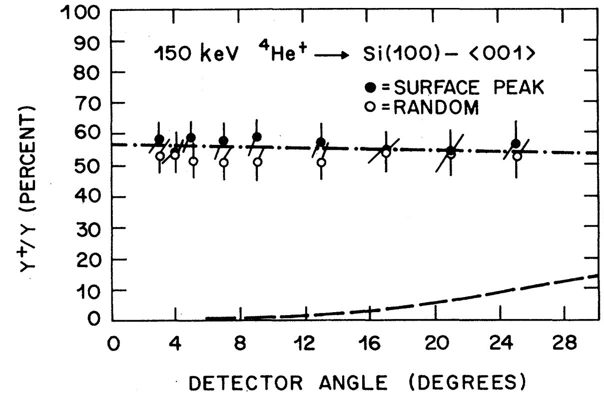

FIG. 3. Ion fractions from surface peak (solid circles) and random (open circles) measurements as a function of takeoff angle. The dot-dashed line is a guide to the eye, the dashed curve from adiabatic theory. (See Ref. 12.)

C. Cs coverage dependence—work-function dependence

We have also studied the change in the ion fraction of Si scattered He upon controlled deposition of cesium. In the measurements to be described, Cs was deposited on the Si surface and the ion fractions were measured for those He projectiles which scattered from the underlying Si. This is possible since the kinematics of the scattering process results in an energy separation of the Si and Cs scattered He ions which is easily resolved in our detection system. The effects of Cs adsorption on Si(100) is to monotonically decrease the work function from 5 to 1.5 eV at ~0.55 monolayer coverage. Higher Cs coverages result in an increase in the work function to the Cs metal value of 2.14 eV.[19–21] Cesiation of the Si surface provides a unique means of studying the effect of a large work-function change on the mechanisms and efficiency of charge exchange at a solid surface. Figure 4 shows four data sets which display the dependence of the ion fraction on Cs coverage. The curves display similar features although the details of the curve shapes are velocity dependent. The closed circles are results from measurements of surface peak intensities while the open boxes are results from random measurements. The agreement between the random (depth resolution of ~30 Å) and channeling (depth resolution of <10 Å) measurements yield further evidence that the ion fraction is determined at the solid surface. The dashed curves represent fits calculated from the model to be presented in the next section.

This system allows a further test of depth-dependent effects. Detailed comparisons have been made between the ion fractions of two important cases. In the first case ions scatter from the surface with some particular velocity.

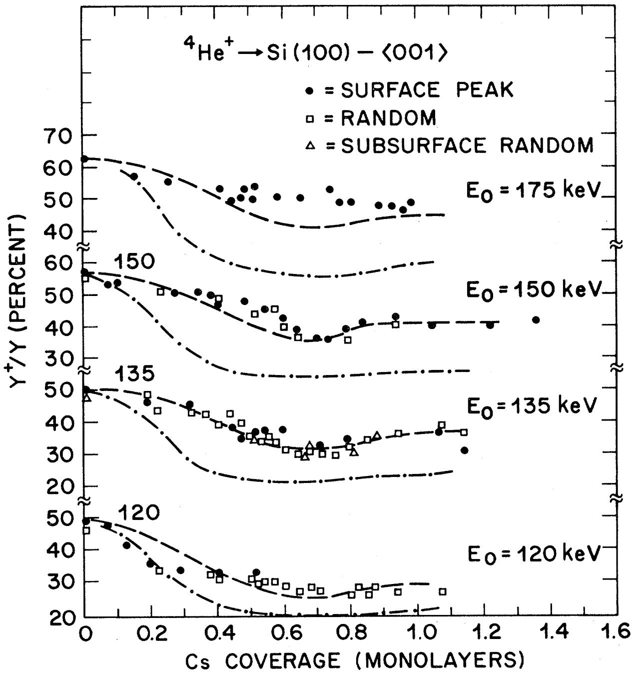

FIG. 4. Ion fractions from surface peak (solid circles) and random (open squares) measurements as a function of Cs coverage for incident-beam energies, E_0. Open triangles (135 keV curve) are ion fractions determined from subsurface scattered ions whose incident energy was 150 keV, as discussed in the text. The dashed curves are fits to the data from the model and include both broadening ($\sigma = 2.0$ eV) and shifting (to 1.5 eV below the vacuum level) of the $n = 2$ level as discussed in the text. The dotted-dashed curves are results from the model where shifting of the $n = 2$ level has not been included, i.e., the $n = 2$ level remains 4.77 eV below the vacuum level.

The ion fraction measured is compared with a second case where ions of a higher initial velocity penetrate the solid, scatter at some depth, and emerge from the surface with the same velocity as ions in the first case. In this manner we can directly compare ion fractions determined from scattering at any depth within the surface. These results are displayed in the 135-keV curve of Fig. 4 where the solid circles and open triangles represent the surface and subsurface scattered projectiles, respectively. This measurement was carried out on both clean and Cs-covered surfaces for incident He ion energies of 175 and 150 keV as well. Agreement within experimental error is noted in all cases. This information lends further support for capture at the surface since the only important parameter is the velocity with which the ion exits the surface.

We have also compared the ion fractions of He scattered from the deposited Cs (not shown) for low coverages, with that of Si. After differences in the scattered ion velocity were accounted for (Cs is more massive than Si), the ion fractions of the two cases showed good agreement. The lack of atomic specificity implies that capture at these ion velocities occurs not from the core levels of the target atoms but from the valence levels associated with the valence and conduction bands of the surface. We give further support for this argument in the next section.

These results are in sharp contrast with those from high-velocity experiments. Specifically, experimental results from Kido *et al.*, at high He ion velocities (1.0 MeV He), exhibit a strong dependence upon the Z of the target atoms associated with electronic shell effects on the capture cross section.[22] Further target atom Z-dependence effects at high-projectile velocity ($v \sim 2-3v_0$) have been observed with protons transmitted through thin foils.[10] As in the clean Si case, the Si scattered ion fraction at a Cs coverage of 0.79 monolayers displayed no dependence upon takeoff angle. These data yield conclusive evidence that the ion retains no memory of its path, or scattering depth within the solid, and that the ion fraction is determined only at the solid surface.

It is possible to deduce several conclusions concerning the mechanisms of electron transfer directly from the experimental data alone. It is well known that electron pickup at high velocities involves capture from the core of the target atoms.[6] We may argue that this is not the case in the intermediate-velocity regime studied here. Particles collected in our detector which have scattered from subsurface Si must pass though the surface with impact parameters (with respect to the surface atoms) which are large, on the order of angstroms (small impact-parameter collisions would scatter the ions from the path of the detector and hence we would not observe those doubly scattered ions with any significant probability). Upon cesiation of the surface we observe significant changes in the ion fraction for both surface and subsurface scattered ions. If capture occurred from the core of the target atoms we would expect to see large differences between surface scattered He (where electron transfer is strongly affected by the large work-function change) and subsurface scattered He (where the Cs coverage of the surface has a diminished influence on the capture process). As the subsurface scattered He passed through the surface at large impact parameters there would be little or no charge transfer with the cores of the surface atoms and hence we would then observe a difference in the ion fractions which reflected the difference in the environments from which the respective ions scattered. This is, of course, not the case as evidenced from our data which display the agreement between surface and subsurface scattered ion fractions. We may now ask why we observe no takeoff-angle dependence in the ion-fraction data. At low velocities these dependencies are indeed observed. An explanation for the lack of angular dependence in the data may be arrived at by noting trends in the low-energy theory and data. It is evident from the adiabatic theory put forward first by Hagstrum and later by other workers that the distance from the surface at which the electron capture occurs decreases with increasing projectile velocity.[1,23] At low velocities capture occurs at a distance from the surface on the order of several angstroms where the electron-density distribution is constant and independent of the detailed nature of the surface atomic arrangement. At the velocities under study here, which are approximately a factor of 10 higher than those for which adiabatic theory is strictly valid, it is qualitatively expected that electron capture will occur closer to the surface where now the electron-density distributions appear spherical in nature.[24] Electron capture which occurred in this spherical electron-density distribution would certainly yield ion fractions which displayed no dependence on takeoff angle since the exiting ion would observe the same electron density in all scattering directions.

It is also of interest to determine the processes involved in electron transfer to the ion. The ion-fraction dependence on Cs coverage (work function) of the Si surface provides evidence that electrons in the Si valence bands are resonantly transferred to the $n=2$ level (or the manifold of states which lie at slightly smaller binding energies than the $n=2$ state). We present arguments to support this proposition. Figure 5 displays the basic processes to be discussed and is based on the earlier ideas of Hagstrum.[11] At large distances from the Si surface the He ion has a $1s$ half-filled state at 24.8 eV below the vacuum level and a $2s$ state at 4.77 eV. The Fermi level and inner potential of Si are 5 and 17 eV below the vacuum level, respectively. As the ion approaches the solid, several important effects occur. The ion levels shift to smaller binding energies due to screening effects in the electron distribution at and near the surface. Because the ion is moving rapidly, these screening effects are smaller than in the static case and are dependent upon the ion velocity.[25]

Furthermore, the levels are broadened due to the finite lifetime of the $n=2$ state associated with the rate of resonant transfer of the electron between the ion and solid. There may also be a contribution to the broadening, σ, associated with the ion's velocity, i.e., due to the finite time that the ion spends near the surface (a 100-keV ion spends 2.5×10^{-16} sec within 5 Å of the Si atom from which it has scattered at the surface). When broadening of the ion levels is included, electrons can resonantly tunnel between the ion and the solid. Tunneling can occur to any of the manifold of states above and including the $n=2$ quantum level for which there is sufficient overlap with occupied electronic states in the solid, although for simplicity we have chosen to focus on the $2s$ level. In addition, electrons can transit to the $1s$ state via Auger processes although this has been shown to be a slower or less probable window for electron transfer. The reduction of the work function may be viewed as an upward shift of the Fermi level towards the vacuum level. As this level moves upward an increased rate of transitions to the $n=2$ level is expected. The data in Fig. 4 reflect this work-function variation.[26] In order to study the nature of the dependence of the ion fraction on the work-function change in a more quantitative manner, we have represented the broadened $n=2$ state with a Lorentzian, whose width σ is considered as a fitting parameter. We approximate the density of states in the solid as a step function $D(e)$ and fold this with the Lorentzian to obtain the effect of the work-function change on the neutralization of the He ions which scatter from the Si surface. We therefore write

$$\frac{\sigma}{\pi}\int_W^{e_f}\frac{D(e)}{(e-e_0)^2+\sigma^2}\,de = \frac{1}{\pi}\left[\tan^{-1}\left[\frac{e_f-e_0}{\sigma}\right]-\tan^{-1}\left[\frac{W-e_0}{\sigma}\right]\right],$$

where e_f, e_0, σ, and W are the Fermi level, He $n=2$ quantum level, the line broadening, and the Si inner potential, respectively.

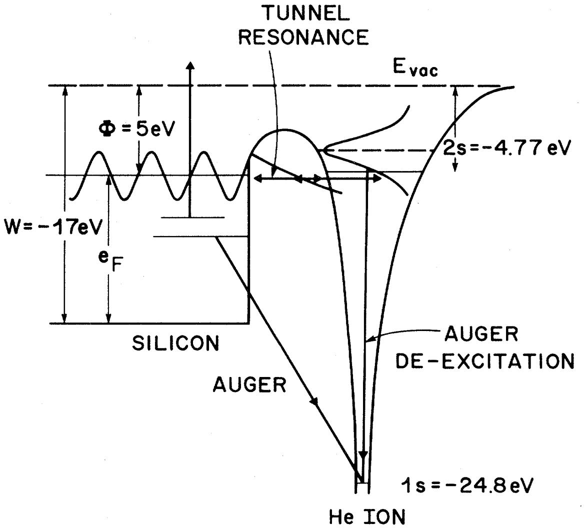

FIG. 5. Sketch of the ion-solid interaction [after Hagstrum (Ref. 1)]. The diagram shows the modes of electron transfer between the solid and the ion when tunneling to a broadened He $n=2$ level is included.

The results of this calculation are plotted in Fig. 4 for two cases where (1) the $n=2$ level remains unshifted (dot-dashed curves), i.e., screening effects have been neglected and (2) where screening has been included resulting in a shift of the level by 3 eV (dashed lines).[27] Note that both screening-induced shifts as well as broadening of ~2 eV are required to give a more reasonable agreement between theory and experiment. Note also that if the $n=2$ level were to have only its natural linewidth associated with the excited state of an isolated He atom, the fitted curve would be nearly a step function and hence show poor agreement with the data. Since our best fits with the data yield broadening of 2 eV we can use this information to estimate the resonant transition rate between the ion and the solid. The broadening, to first order, is given by $\sigma=\hbar w$ where w is the transition rate. This simple calculation gives a transition rate of 3.0×10^{15}/sec or 1 transition in 3.3×10^{-16} sec. We see that the residence time of the ion near the Si atom from which it has scattered is very nearly the same as the time needed for one transition to occur, a result which is reasonable from the point of view that our measured ion fractions in this range are on the order of 50%.

We may also understand, within the context of the ideas discussed here, trends observed in the lower-ion-velocity regime. As the ion velocity decreases, both resonant and Auger processes become more probable due to the increased residence time of the ion near the surface. We may expect that since resonant transfer occurs not only from the solid to the ion but also from the ion back to the solid with some probability, a dynamic equilibrium of resonant electron transfer between the ion and the solid is established. If resonant tunneling were the only process involved in the neutralization of the ion then we would expect the ion fraction to display only a weak dependence on velocity. At lower ion velocities, capture via the slower Auger processes including Auger deexcitation will now begin to populate the He 1*s* state. It is noted that there are no unoccupied states in Si which are in resonance with the He 1*s* level and hence an electron captured to this state will remain there with high probability. At the ion velocities studied in this work, the probability of populating the He 1*s* state via Auger processes is small due to the small ion residence time near the surface. We may also understand, within the framework of these arguments, the lack of depth dependence in the intermediate-velocity regime studied here. Capture in the bulk may proceed via resonant transfer between the ion and the solid (we assume as before that Auger processes are sufficiently slow to be neglected) but an electron captured to the He $n=2$ level will be extremely weakly bound (if at all) within the bulk of the solid.[25] We therefore expect that the ion fraction of the subsurface scattered ions to be determined exclusively at the surface where reduced screening permits stable bound states to exist on the ion. At lower velocities (He ion energies of several keV) capture via Auger processes becomes more probable. The Auger processes result in capture to the 1*s* state where the electron is more tightly bound and hence we expect to see scattering depth dependences in the ion-fraction data. Thus, with the inclusion of Auger processes which become more important at lower ion velocities, we can understand not only the decrease of the ion fraction with decreasing ion velocities but also the onset of scattering depth dependences as well. We therefore see that as the ion velocity is decreased there is a natural evolution from the processes which operate at high velocities to those which occur at low velocities, namely the well-established Auger neutralization observed by Hagstrum.

The previous approximations, while relatively simple, allow us to understand the important physics involved in electron transfer between an energetic ion and a solid surface. Several improvements would include a more realistic density of states and somewhat more quantitative estimates of the ion-level broadening which may include contributions associated with the time-dependent ion-solid interaction. In Fig. 4 the 135- and 150-keV calculated curves agree well with the data while the 175- and 120-keV curves deviate quantitatively. This deviation indicates the need for a more detailed treatment of the time-dependent ion-solid interaction which should include changes in the screening response of the solid due to the Cs coverage of the surface.

In conclusion, our data show that (1) the ion fractions observed are determined at the solid surface, on the ion's outward path; the ion retains no memory of its path within the solid, (2) there is no dependence of the ion fraction on takeoff angle, (3) the ion fraction displays a dependence on the solid work function, and (4) electron capture occurs not at the core of the Si atoms nor at the relatively large distances associated with electron capture at low-ion velocities, but instead at intermediate distances of $\lesssim 1$ Å where the electronic density distributions are spherical. We have explained these effects by invoking the contribution of tunnel-resonant transitions to the broadened He $n=2$ quantum level. Furthermore, we have been able to estimate, within this model, a transition rate for resonant transfer of electrons between the solid and the energetic ion.

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[26] A small coverage difference (~ 0.1 monolayer) exists between our measurements and those of Ref. 21.

[27] A detailed discussion of screening-induced level shifts is given in Ref. 25. Level shifts versus ion solid distance has been calculated for low-ion velocities in Ref. 1.