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Electron emission from He^+ interacting with a Cs overlayer on W(110)

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Low-energy ($10 < E_0 < 500$ eV) He^+ ions are employed to explore the mechanism of electron emission from the Cs-saturated W(110) surface. The intense peak at the Fermi level exhibits a marked Doppler shift while a lifetime broadening of the peak is less pronounced than that expected from Auger deexcitation. These findings reveal that the intense electronic emission comes from the incoming trajectories before Auger deexcitation of He^* ($^3S_1$) takes place.

The electronic interaction of atoms and molecules with solid surfaces has attracted much interest in connection with a number of dynamical processes. In particular, investigations of a decay channel of excited atoms[1-5] or ion neutralization[6-8] have recently shed light on the electronic structure of the alkali-metal layers. The previously accepted picture of almost complete ionization of the alkali-metal atoms at very low coverage has been called into question by a series of studies of metastable deexcitation spectroscopy[1-4] (MDS) because alkali-metal s-derived states have been directly observed at any coverage. The interpretation of the results was, however, complicated by the existence of a very effective transformation process at the alkali-metal-covered surface;[2-4] the singlet He^* ($^1S_0$) metastable is readily converted into the triplet ($^3S_1$) species and the conversion rate changes with alkali-metal coverage. So far, all of the structures appearing around the Fermi level have been related to the alkali-metal s state. Very recently, however, it has been claimed[5] that a narrow intense maximum located just below E_F has a resonancelike feature and should be discussed separately from the portion coming from the ordinary Penning transitions. The former is attributed to the decay of the core-excited He^- states and hence has nothing to do with the existence of the alkali-metal s state.

The pronounced feature of ejected electrons just below E_F has already been reported by Hagstrum and coworkers[6,7] in the ion-neutralization-spectroscopy (INS) study of He^+ ions on potassium-covered surfaces. In these works, they have concluded that the mechanism underlying the strong electron emission is resonance neutralization (RN) followed by Auger deexcitation (AD) and hence provides essentially the same result as MDS. One of the most significant differences in these two approaches is that the energetic He^+ ion beams can be used in INS while essentially the thermal-energy He^* beam is employed in MDS. With use of the ion beam, therefore, the electronic-transition process leading to the strong electron emission can be explored from other points of view. This paper is devoted to this subject. It is found that the decay of He^* ($^3S_1$), descended from He^+ interacting with a Cs-saturated W(110) surface, causes the significant Doppler shift in the spectral-peak position and that the lifetime broadening of the spectral peak is less pronounced compared to that expected from Auger deexcitation of He^*.

The experimental setup has been described elsewhere[9] and only the features related to this study are briefly summarized here. The sample chamber was evacuated down to an ultrahigh-vacuum condition (1×10^{-8} Pa) and equipped with facilities for low-energy ion scattering (ISS), low-energy electron diffraction (LEED), and ultraviolet photoelectron spectroscopy (UPS). The He^+ ions with kinetic energy E_0 ranging from 10 eV to 1 keV were generated in a discharge-type ion source and were mass analyzed by a Wien filter. The ion source was attached to the sample chamber through three differentially pumped vacuum chambers containing lens systems so that the pressure in the sample chamber is kept below 2×10^{-8} Pa during the measurements. The He^+ beam was incident upon a surface with a various glancing angles of α and electrons emitted into a certain angle β, defined between the He^+-incidence and e^--detection directions, were analyzed by a hemispherical electrostatic energy analyzer operating with a constant energy resolution of 200 meV. The W(110) surface was prepared with a standard oxygen treatment. The sample cleanliness was checked by means of LEED, UPS, and ISS with use of $E_0=1$ keV He^+ ions. Cesium was evaporated from a carefully outgassed dispenser source (SAES Getters, Inc.). Both clean and Cs-saturated surfaces showed a sharp 1×1 pattern in LEED.

Typically shown in Fig. 1(a) is a spectrum of ejected electrons from the Cs-saturated W(110) surface obtained with a $E_0=20$ eV He^+ beam, the experimental geometry being shown in the inset. The Fermi-level position relevant to the decay of He^* ($^3S_1$) is obtained in comparison with the normal-emission UPS spectra, shown in Fig. 1(b), by considering the difference in the excitation energy of HeI (21.2 eV) and He^* (19.8 eV). In addition to the pronounced structure just below E_F, a broad maximum is observed around the binding energy of -12 eV. This can easily be ascribed to the Cs $5p$ state from the comparison with the UPS spectra. The spectral features seem to be consistent with the results of MDS as well,[4] implying that incident He^+ is resonantly neutralized into He^* ($^3S_1$) with high probability before AD takes place. This is reasonable since the work function of the surface (1.6 eV) is much smaller than the binding energy of the He^* triplet state (4.7 eV). It should be noted that, in the INS spectra, the doublet in the Cs $5p$ peak is not clearly separated whereas the structure just below E_F is quite

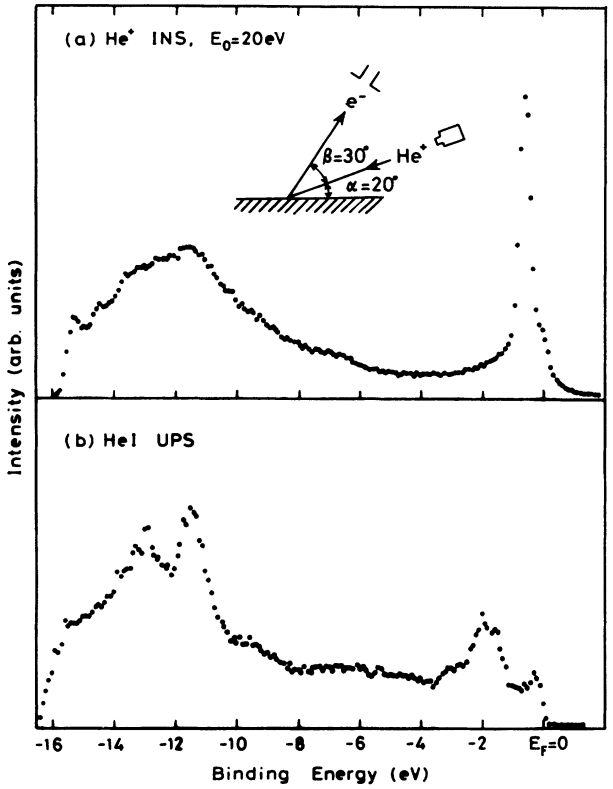

FIG. 1. (a) The INS energy spectrum using $E_0=20$ eV He^+ ions and (b) the normal-emission UPS (HeI) spectrum obtained at the Cs-saturated W(110) surface. The experimental geometry in INS is shown in the inset.

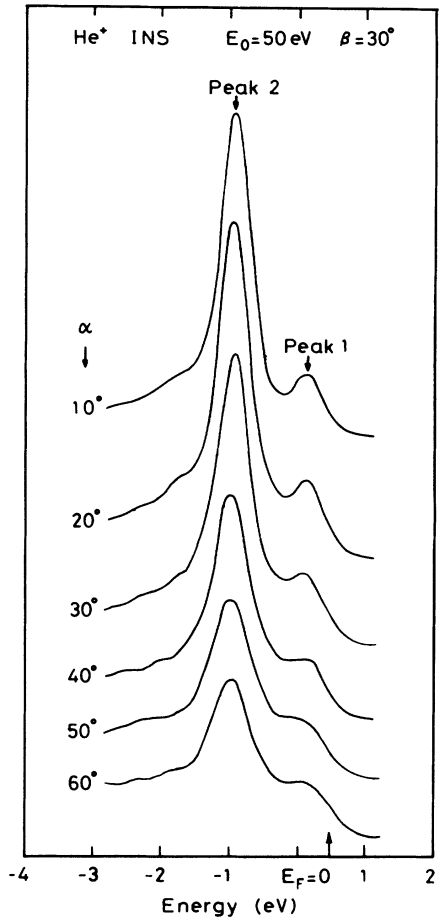

FIG. 2. The INS spectra of $E_0=50$ eV He^+ obtained at the Cs-saturated W(110) surface as a function of glancing angle α, β being fixed at 30°.

sharp with a full width at half maximum (FWHM) of 0.45 eV.

Figure 2 shows the INS spectra around E_F obtained by the incidence of the $E_0=50$ eV He^+ ions as a function of α; β is fixed at 30°. In contrast to Fig. 1(a), the two peaks are clearly separated energetically. Though the peak intensity decreases with α, the peak position is almost unchanged. The high-energy peak (referred to as peak 1) appears above E_F corresponding to AD of He^* ($^3S_1$) while the position of the low-energy peak (peak 2) is located at a lower-energy side of the main peak in Fig. 1(a). The variation of the peak intensity in Fig. 2 clearly shows that the probability for the electron emission is dependent on the time spent by He^+ in a certain region of the surface. This is also confirmed by the fact that the peak intensity decreases more rapidly with α by increasing the primary He^+ energy. Moreover, the peak intensity shows no azimuthal-angle dependence. It is well known in ISS (Refs. 9 and 10) that the ion flux hitting the surface atom is significantly dependent on the azimuthal angles due to the shadowing and focusing effect. The absence of this effect indicates that the role of the collisional regime can be ruled out in the strong electron emission.

The energy position of peak 2 and the high-energy cutoff of peak 1 (indicated by an arrow on the abscissa of Fig. 2) are measured as a function of E_0 under the same scattering geometry of $\alpha=20°$ and $\beta=30°$, and the results are plotted in Fig. 3 against the velocity of the incident He^+ ions. Peaks 1 and 2 shift linearly toward opposite directions with ion velocity. Also shown in Fig. 4 is the position of peak 2 relative to E_F as a function of β with α being fixed at 20°. The result clearly shows that the peak shift increases with increasing the velocity component of incident He^+ toward the analyzer. These behaviors remind us of the Doppler shift: It is well known that the motion of the electron source produces a shift in the kinetic energy of the ejected electrons. Since the velocity of He^+ (He^*) is much smaller than the velocity of the electron, we can write

$$E_L = E_K + (2mE_K)^{1/2} v_0 , \tag{1}$$

where E_L (E_K) is the electron energy in the laboratory (He^*) frame, m is the electron mass, and v_0 is the velocity of incident He^+ in the detection direction. The values calculated with use of $E_K=18.2$ eV (corresponding to the excitation energy of He^* minus work function) are shown in Figs. 3 and 4 by solid lines. As shown in Fig. 3, on the other hand, the shift of peak 1 is opposite in direction to that expected from the primary He^+ beam, so that it is probable that peak 1 comes from the reflected He^+ ions. However, backscattered He^+ is only a small portion of incident He^+ due to a relatively small cross section and a

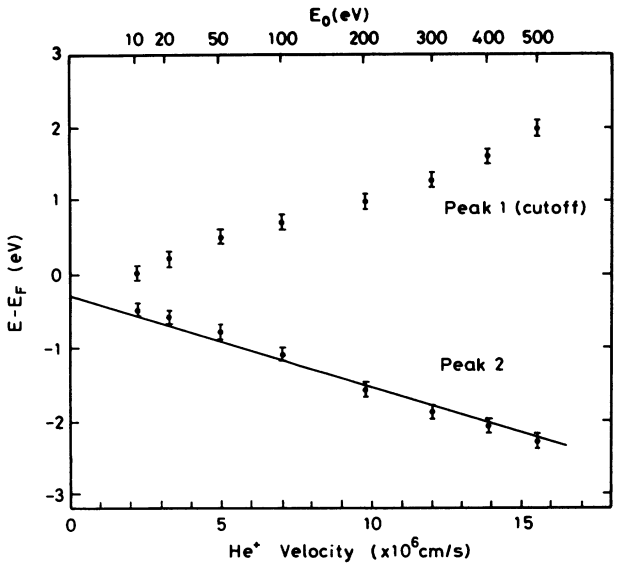

FIG. 3. The energy positions of peak 2 and the high-energy cutoff of peak 1 (typically shown by arrows on the abscissa of Fig. 2) are plotted against the He^+ velocity. The measurements were made under the same scattering geometry of $\alpha=20°$ and $\beta=30°$. The calculated values of the Doppler shift of the ejected electrons from He^* ($^3S_1$) in the incoming trajectory are shown by a solid line.

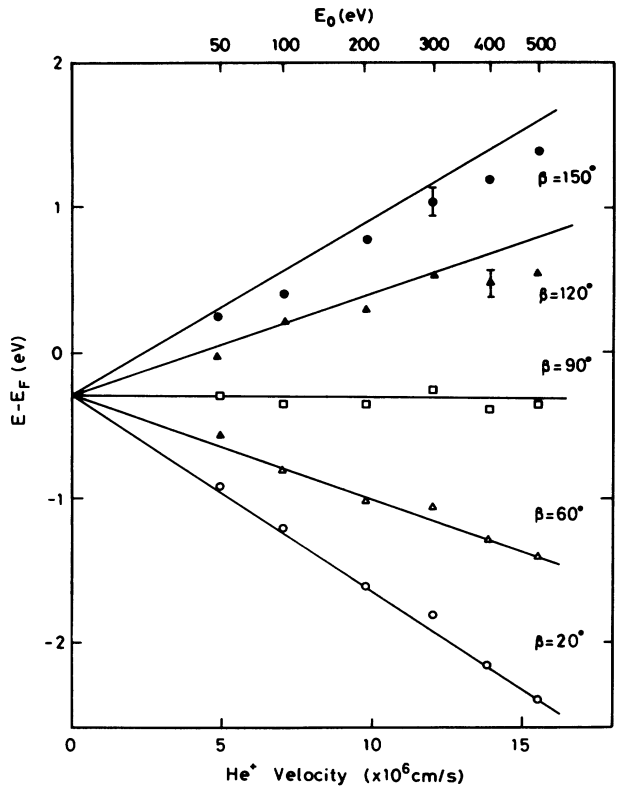

FIG. 4. The energy position of peak 2 as a function of the velocity of incident He^+. The measurements were made at various angles of β with α fixed at 20°. The calculated values of the Doppler shift of the ejected electrons from He^* ($^3S_1$) in the incoming trajectory are shown by solid lines.

large neutralization probability and its spatial distribution should be rather broad, leading to a broad energy distribution rather than the sharp peak. In fact, peak 1 is usually observed superposed on the broad contribution which can be ascribed to the emission from the backscattered He^+. A more detailed discussion of peak 1 will be presented in a separate paper. It should be noted that all spectral peaks can substantially be ascribed to the decay of He^* ($^3S_1$) and no contribution from the singlet states is recognized.

Despite the marked Doppler shift of the spectral-peak positions, the peak shape is less affected by the energy or the experimental geometries. Plotted in Fig. 5 is the FWHM of peak 2 versus the incident He^+ velocity perpendicular to the surface. Besides a linear dependence on $v_\perp$, a meaningful width remains at the zero-velocity limit. The spectral width is known to be determined by the shift and the lifetime of the electronic states in the vicinity of a surface.[11,12] The energy level shifts upwards due to the image charge effect. Then the level-shift broadening of the ejected-electron spectra comes about if the image shift of the relevant electronic levels ($1s$, $2s$) is different from each other as a function of the distance. On the other hand, a limited lifetime of the level also gives rise to the peak broadening. The lifetime broadening of the level is given by the Heisenberg uncertainty principle written as

$$\Delta E=\hbar/\tau=\hbar R(s)\,,\qquad R(s)=A\exp(-as)\,, \tag{2}$$

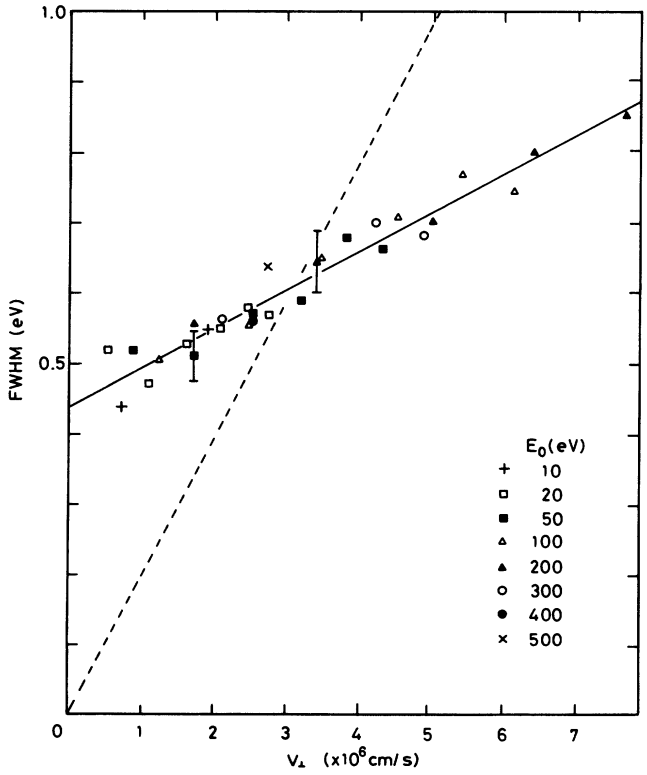

FIG. 5. FWHM of peak 2 as a function of the velocity perpendicular to the surface. The lifetime broadening expected from Auger deexcitation is also shown by a dashed line.

where τ is the lifetime and $R(s)$ is the total transition probability at the distance s from the surface at which the electronic transition occurs. According to Hagstrum, Takeishi, and Pretzer,[11] the lifetime broadening can be given as

$$\Delta E = \hbar a v, \tag{3}$$

and the corresponding transition rate

$$R = a v. \tag{4}$$

If Auger neutralization or AD is relevant, the most probable value of $a \sim 3$ Å^{-1} has been obtained.[11] The lifetime broadening based on this evaluation is shown by the dashed line in Fig. 5 and is rather steep compared with our results. Conventionally, the remaining peak width at the zero-velocity limit might provide the density of state of the occupied portion of the Cs 6s state. If that were the case, the inclination of the data points in Fig. 5 would be much steeper since the electron ejection should be caused by AD of He* ($^3S_1$). In this context, it should be mentioned that the Cs 5p peaks in Fig. 1(a) are broad relative to the corresponding peaks in the HeI UPS spectra shown in Fig. 1(b) as well as the MDS spectra by Maus-Friedrichs *et al.*,[4] the trend of which stems from the more pronounced broadening associated with AD mentioned above. Considering these findings, we conclude that peak 2 originates not from AD of He* ($^3S_1$) but from other decays occurring farther from the surface before entering the Auger-decay region (< 2 Å). Our result is consistent with the recent MDS study by Hemmen and Conrad,[5] in which the ejected electron close to E_F comes from the decay of core-excited He$^-$ state.

[1] B. Woratschek, W. Sesselmann, J. Kuppers, G. Ertl, and H. Haberland, Phys. Rev. Lett. **55**, 1231 (1985).

[2] J. Lee, C. Hanrahan, J. Arias, F. Bozso, R. M. Martin, and H. Metiu, Phys. Rev. Lett. **54**, 1440 (1985).

[3] B. Woratschek, W. Sesselmann, J. Kuppers, G. Ertl, and H. Huberland, Phys. Rev. Lett. **55**, 611 (1985).

[4] W. Maus-Friedrichs, M. Wehrhahn, S. Dieckhoff, and V. Kempter, Surf. Sci. **237**, 257 (1990).

[5] R. Hemmen and H. Conrad, Phys. Rev. Lett. **67**, 1314 (1991).

[6] H. D. Hagstrum Phys. Rev. Lett. **43**, 1050 (1979).

[7] H. D. Hagstrum, P. Petrie, and E. E. Chaban, Phys. Rev. B **38**, 10264 (1988).

[8] R. Souda, W. Hayami, T. Aizawa, S. Otani, and Y. Ishizawa, Phys. Rev. Lett. **69**, 192 (1992).

[9] R. Souda, T. Aizawa, C. Oshima, S. Otani, and Y. Ishizawa, Phys. Rev. B **40**, 4119 (1989).

[10] M. Aono and R. Souda, Jpn. J. Appl. Phys. **24**, 1249 (1985).

[11] H. D. Hagstrum, Y. Takeishi, and D. D. Pretzer, Phys. Rev. **139**, A526 (1965).

[12] P. Nordlander and J. C. Tully, Phys. Rev. B **42**, 5564 (1990).