Formation of Feshbach Resonances Associated with Doubly Excited He States in Slow Collisions of He^{++} Ions with Low Work Function Surfaces

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We report electron energy spectra from He^{++} (100 eV) ions colliding under grazing incidence with partially cesiated W(110). We propose that at low surface work functions (≤ 2 eV) the formation of Feshbach resonances associated with the doubly excited He states of the configurations $(2l\,2l')$ occurs. This is concluded from the variation of the relative population of the doubly excited He states with the surface work function. Furthermore, a peak can be identified in the spectra which appears to be due to the decay of the Feshbach resonances by one-electron emission.

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The electron emission induced in He^{++} collisions with clean and alkalated metal surfaces has found considerable interest in recent years because its study offers a convenient way to improve our knowledge of the dynamics of inelastic ion-surface collisions [1–5]. The following picture concerning the sequence of electronic transitions leading to neutralization of He^{++} has emerged (see Fig. 1).

For clean surfaces [1–3] the majority of He^{++} ions is neutralized in two Auger capture (AC) processes whereby the first one leads to the formation of He^{+} in its ground state and the second one to the formation of He^{0}. In some minor fraction of the collision events also the formation of doubly excited He states with the configuration $(2l\,2l')$ occurs as a consequence of the formation of He^{+*} ($n = 2$) via resonant capture [1–3] or Auger capture [5, 6] followed by the resonant capture of a second electron by the He^{+*} ($n = 2$) [1–3]. For simplicity we have neglected in Fig. 1 the various channels for the decay of the species He^{+*}, etc., via interatomic Auger processes,

as, in particular, Auger deexcitation (AD) (Penning ionization) involving electrons of the surface (see [1–5]).

For low work function surfaces resonant transfer (RT) of electrons from the surface by the projectile becomes much more likely with the consequence that in a large number of collision events the upper row of the reaction scheme is passed through from the left to the right before He^{+} is formed by autoionization of He** [(AU)$_1$ in Fig. 1] with high probability. The He^{+} ions are again neutralized via resonant capture into singly excited states of He. When capturing resonantly a second surface electron, the temporary negative ion He^{-*}($1s2s^2$; ^2S) is formed [4, 7]. These species finally decay into the neutral He ground state via interatomic and intra-atomic Auger processes [4, 7], Auger deexcitation, and autodetachment [(AU)$_2$ in Fig. 1]. These conclusions concerning the dynamics of the process could be confirmed [5] by the simulation of the electron spectra using the method described in [2, 8].

For electron collisions it has been demonstrated both experimentally and theoretically (see [9] for a review) that the He^{++} ion can attach three electrons in ($n = 2$) states, forming He^{-**} in the configurations $(2s^2 2p)$ and $(2s2p^2)$. These states can be considered as Feshbach resonances of the He** parent states $(2s2p)$ and $(2p^2)$. In this Letter we provide evidence that these Feshbach resonances are also formed in slow He^{++} collisions with partially cesiated W(110): The He** intermediates (formed by the resonant capture of two surface electrons) capture an additional surface electron provided the surface work function is sufficiently low (≤ 2 eV). This means that the reaction scheme for collisions of He** with low work function surfaces should be completed as indicated in Fig. 1 by the dashed part.

The apparatus was documented previously [5]. Briefly, He^{++} ions are generated in a low voltage gas discharge source and mass/charge selected by a Wien filter. They impinge upon a W(110) crystal (oriented with the [001] direction along the beam direction and held at room temperature) grazingly under 5° with respect to the surface. A hemispherical analyzer is positioned at a fixed detec-
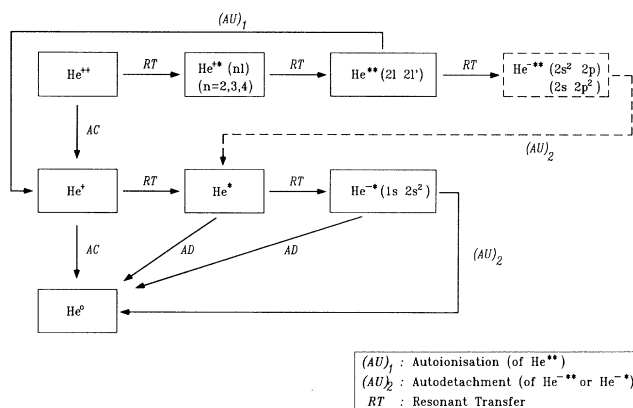

FIG. 1. Reaction scheme for slow He^{++} collisions with low work function metal surfaces. Arrows denote some of the possible electronic transitions between the involved intermediate charge and excitation states.

tion angle of 90° with respect to the beam axis. The spectrometer records the energy spectra at constant pass energy (ΔE = 0.4 eV FWHM). A small (< 5 V) bias voltage is applied between the crystal and the electro-static analyzer in order to compensate their work function difference. Electrons leaving the crystal with zero kinetic energy thus arrive at the analyzer with a kinetic energy given by the sum of the bias voltage and the dif-ference between the work functions of the crystal and the analyzer. These electrons define the low energy edge of the spectra. Lowering the crystals work function by ad-sorption of alkali atoms results in a shift of the low energy edge in the electron spectra by the change in the crystals work function. The bias voltage for the experiments pre-sented here is chosen in such a way that electrons leaving the crystal with zero energy for a surface work function of 1.4 eV arrive at the spectrometer with approximately 2 eV. A $1/E$ correction was applied to all spectra in order to compensate the energy dependence ($\propto 1/E_{kin}$) of the analyzer's transmission. Cs atoms are offered to the sur-face by means of dispenser sources (SAES Getters Inc). The variation of the work function of W(110) with alkali coverage has been reported by us in [10].

Figure 2 shows a set of electron spectra obtained for 100 eV He^{++} ions colliding with partially cesiated W(110) surfaces as a function of the Cs coverage be-tween zero and one monolayer (1 ML) coverage; 1 ML corresponds to the Cs saturation coverage at room tem-perature. Similar spectra, but only up to 0.6 ML, were reported by us in [5]. Figure 3 presents the change of the electron spectra when the fully cesiated W(110) surface is exposed to NaCl. By this procedure the work function of the surface increases and, in particular, the surface den-sity of states at the Fermi level decreases strongly [11]. Most of the features seen in the spectra were identified in [4, 5]. The symbols show the initial intermediate state (see Fig. 1) which is responsible for the electron emission and the Auger processes which are responsible for the electron emission (AD, Auger deexcitation; AC, Auger capture; AU, autoionization of He** or autodetachment of He^{-*}). The feature at 8 eV is now identified as Auger deexcitation of He^{+*} (n = 4) to the He^{+*} (n = 2).

We will now concentrate on the strong emission ob-served near the high energy end of the spectra which so far, on the basis of the existing simulations of the energy spectra with the method described in [2], is assumed to be entirely due to autoionization of He$^{**}(2l\,2l')$ [2, 3, 5]. For work functions > 2 eV we observe two peaks which were identified as due to autoionization of the He** states from the two groups of states, $2s^2$; ^1S and $2s2p$; ^3P, on the one hand (lower energy peak), and $2s2p$; ^1P and $2p^2$; ^3P and ^1D, on the other hand [1, 2] (higher energy peak) (emission marked in black). Around the work emission from the autoionization of $2p^2$; ^3P and ^1D and $2s2p$; ^1P
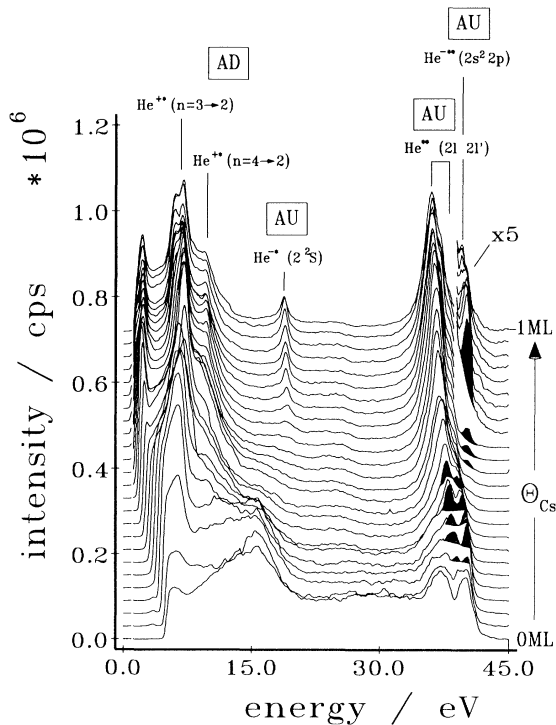

FIG. 2. Electron energy spectra at the impact of 100 eV He^{++} ions at partially cesiated W(110) as a function of the Cs coverage. The angle of incidence is Ψ = 5°.
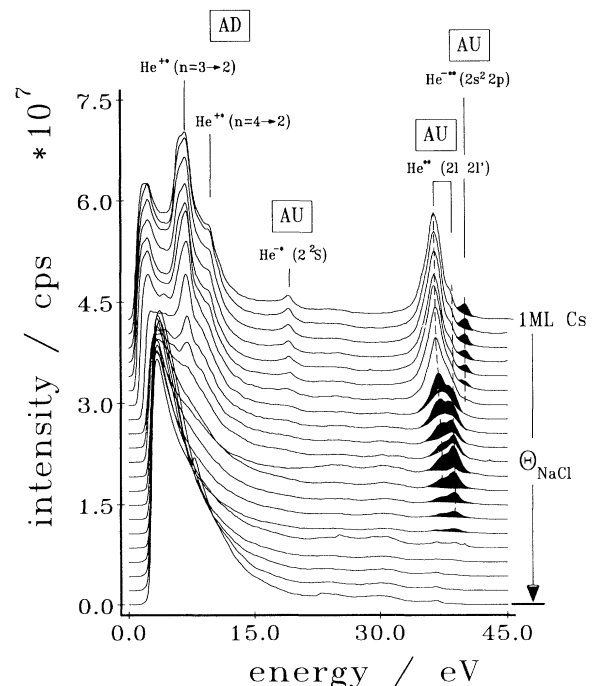

FIG. 3. Electron energy spectra at the impact of 100 eV He^{++} ions at cesiated (1 ML) W(110) as a function of ex-posure to NaCl (arbitrary units). The angle of incidence is Ψ = 5°.

disappears almost entirely, and instead a new feature appears on the high energy side of the spectra (marked in black). We have attributed this additional feature to the formation of higher excited He** states of the configuration $(2snl; n > 2)$ in [4,5]. No explanation for the disappearance of the high energy component of the emission due to the decay of He**$(2l\,2l')$ as soon as the surface work function is lowered by alkali adsorption could be given, though. Furthermore, the double-peak structure from the autoionization of He** reappears upon exposure of the fully cesiated surface to NaCl, i.e., if the work function is enlarged again (see Fig. 3).

Both these facts can be explained simultaneously by adapting a model proposed in [7] (and quantified in [12]) in order to explain the singlet to triplet conversion in He metastable collisions to the present situation. It assumes that the conversion of He($2\,^1S$) metastables into He($2\,^3S$) metastables proceeds via the He$^{-*}(1s2s^2; \,^2S)$ resonance. For a qualitative discussion of our situation we assume that the collision process populates only He** states of the configuration $2l\,2l'$; the states He^{+*} $(n = 3,4)$ which can form higher excited states [as He**$(3l\,nl)$, etc.] by attaching an additional electron are most likely efficiently deexcited to He^{+*} $(n = 2)$ via fast Auger deexcitation (see Fig. 2). We propose that the He** atoms in $2s\,2p$; ^1P and $2p^2$; ^3P and ^1D resonantly capture an additional surface electron when the surface work function becomes lower than about 2 eV. This leads to the formation of the Feshbach resonances He^{-**} $2p^2\,2s$; ^2D and $2s^2\,2p$; ^2P.

Figure 4 shows schematically the interaction of He** and He^{-**} states with the surface. The influence of the image forces is shown only for the states involving He^{-**}; at smaller internuclear distances the $(n = 2)$ electrons do not shield the He^{++} core well [2,8], and the image interaction should be considered also for the states involving He**. The relative position of the potentials is drawn for a low work function surface (1.8 eV). The excitation energies of the doubly excited states and temporary negative ion states of the free particles as given in [9] have been used. z_c denotes the position of the outermost crossing between the states involving He^{-**} and the lowest state correlating with He**; for $z < z_c$ He^{-**} becomes stable against the decay to He** via resonant ionization involving surface electrons. On the other hand, as long as $z > z_c$ the resonances He^{-**} will decay efficiently into He** $2s\,2p$; ^3P and $2s^2$; ^1S by the ejection of an electron into the surface (Fig. 4). This model explains the disappearance of the peak from autoionization of He** $2p^2$; ^1D and ^3P and $2s\,2p$; ^1P as a conversion into the low energy group of He** states via the Feshbach resonances.

As soon as $z < z_c$, at least the Feshbach resonance He$^{-**}(2s^2 2p)$ becomes stable against the decay to He**$(2s^2; \,^1S)$ and $(2s2p; \,^3P)$, and some fraction of He^{-**} will decay into excited states of the neutral helium, He* $(1s2l)$ [9] via a one-electron-emission process. The energy of these electrons should be approximately 3 to 4
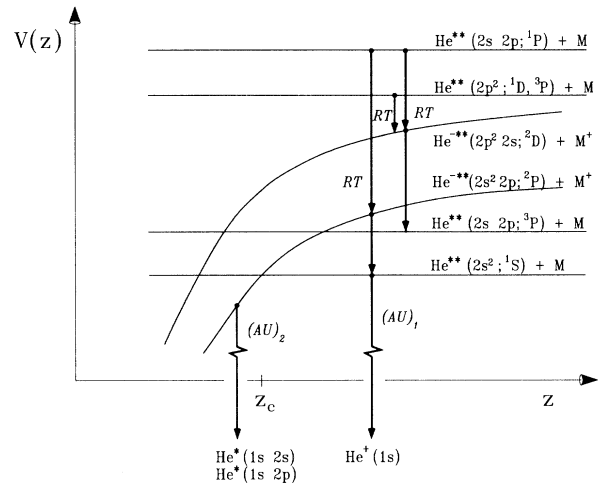

FIG. 4. Potentials (schematically) for the interaction of He** and He^{-**} with low work function surfaces. Arrows denote the channels for the decay of the various states.

eV higher than that from autoionization of He** $(2s^2; \,^2S)$ and $(2s2p; \,^3P)$ to He$^+$ considering the fact that the final states after $(AU)_2$ are He*$(1s2l)$ and taking into account the image shift of He$^+$. Thus, we explain the high energy peak appearing for work function values below 2 eV as due to the decay of the Feshbach resonance He$^{-**}(2s^2 2p)$ to excited states He*$(1s2l)$.

The findings of Fig. 3 are also consistent with the proposed explanation: The exposure to NaCl inhibits the resonant capture of a surface electron by the He** as soon as the work function increases beyond 2 eV and, in addition, the surface density of states has decreased considerably. Consequently, the conversion process between the two groups of He** states is inhibited, and the double-peak structure from the autoionization of both groups of He** states reappears (see Fig. 3). At sufficiently large exposures the NaCl band gap appears, and even the formation of He** from He^{++} becomes inhibited [11]. We would like to mention that the exposure of the cesiated surface to oxygen produces a similar behavior of the electron emission.

Summarizing, we have presented evidence that Feshbach-type resonances of helium with the configurations $(2s^2 2p)$ and $(2s2p^2)$ are formed in slow He^{++} collisions with low work function surfaces. Their formation leads to a redistribution in the relative populations of the various He** 2 states and to the appearance of a peak in the electron spectra due to the one-electron decay of He^{-**} to excited states of He.

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