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Electron emission in slow collisions of He projectiles (He*, He⁺, He⁺⁺) with Li- and LiF-surfaces

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Abstract

We report the spectra of the electrons emitted in slow collisions of He⁺ (50 eV) and He⁺⁺ (100 eV) ions with a Li covered W(110) surface and with LiF films grown on Li/W(110). The preparation of the surfaces was made under in situ control of the metastable impact electron spectroscopy (MIES). The interpretation of the spectra is made on the basis of simulated electron spectra: For collisions with Li/W(110) the ion induced electron spectra are mainly due to Auger processes of the projectiles after their resonant capture of electrons from the surface. Both Auger deexcitation and intra-atomic Auger processes, such as the autodetachment of He^{-*} (1s2s²) and the autoionization of the "hollow atoms" He^{**} (2l2l') (l, l' = 0, 1), are observed. For collisions with LiF films the neutralization of the projectiles takes place mostly by Auger capture processes involving two electrons from the surface. The resonant capture of an electron by the He⁺⁺ into He^{+*} (n=2) state still occurs, but is weak. The formation of "hollow atoms" does not take place in front of dense LiF films. There are some indications that the quasi-resonant electron transfer involving F 2s electrons plays also a role in the interaction of He⁺⁺ with the surface.

Keywords: Amorphous thin films; Auger ejection; Electron emission; Ion impact electron spectroscopy; Lithium; Lithium fluoride; Metastable impact electron spectroscopy

1. Introduction

Ion impact electron spectroscopy (IIES), i.e. the analysis of the electron energy spectra induced by the impact of slow ions with surfaces, has provided new insight into the collision dynamics. In particular, the main electronic transition processes taking place in contact with the surface could be identified [1,2]. This has become facilitated by the fact that such electron spectra can now be simulated in a semi-quantitative way [3,4]. IIES on insulator surfaces or on surfaces covered by insulating adlayers is less popular although several efforts

have been made [5–10]. Several reasons account for this fact: (1) difficulties do exist to prepare defect-free insulator surfaces [11], (2) insulator surfaces are subject to modifications in their electronic structure under heavy particle impact [12,13], and (3) such surfaces are subject to charging phenomena due to the removal of electrons from the insulator valence band during the collision process [5–8].

The study of the possible mechanisms for the transfer of electrons between heavy particles and insulator surfaces is important for (i) the understanding of the mechanisms for desorption of heavy particles induced by electronic transitions (DIET) [12,13], and (ii) the understanding of the mech-

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anisms leading to the neutralization of highly charged ions in collisions with insulating surfaces [14,15]. Such collisions were so far mostly studied on metallic surfaces; the interest often focused on the formation of so-called “hollow atoms” (the formation of doubly-excited states He^{**} ($2l2l'$) ($l, l'=0, 1$) in the case of helium collisions). Some doubts remained as far as the origin of the emitted electrons is concerned: it is not yet clear which part of the electron emission originates from the precollision phase, e.g. before the projectile hits the surface. Collisions with insulator surfaces may answer this question: as was demonstrated for He^{++} collisions with alkali halide surfaces [6,7], the collision dynamics of highly charged ion collisions with insulator and metal surfaces is different.

In this paper we report the energy spectra of the electrons emitted in collisions of He^* atoms of thermal energy, He^+ (50 eV), and He^{++} (100 eV) ions with Li adlayers and with LiF films. Detailed information on the collision dynamics, in particular on the electronic transition processes during the collision, is obtained from a comparison of the experimental spectra with those simulated in the way described in Refs. [3,4]. These data need to be supplemented by the total electron yields from the study of the electron emission statistics as was done recently for collisions of hydrogen, nitrogen, and argon ions with polycrystalline LiF [16].

2. Apparatus and results

The results were obtained with the apparatus described in Refs. [17–19] (see also the references given therein). A source for metastable impact electron spectroscopy (MIES) [20] was newly added. MIES is employed in order to monitor the growth of the Li layer on W(110) and the LiF film on W(110) covered by 1 monolayer (ML) of Li.

Monoenergetic and mass analyzed beams of He^+ and He^{++} ions (50 and 100 eV, respectively) impinge grazingly (5° with respect to the surface plane) along the [001] direction of a W(110) crystal held at room temperature during the measurements. The beam of He^* metastable atoms utilized for MIES hits the surface under 45° . The spectra of the emitted electrons are recorded under

normal emission in the plane formed by the beam direction and the surface normal.

The work function of the surface is varied between 5.3 and about 2.3 eV by the deposition of various amounts of Li on the surface. The energy spectra are taken at regular time intervals of about 1 min while offering alkali atoms to the W(110) surface at a slow rate (below 0.05 ML/min).

LiF molecules are supplied to the surface by thermal evaporation (1100 K) of single crystal chips (see also Refs. [5–8]). The exposure rate is constant for all measurements. Under the chosen conditions a closed LiF adlayer is deposited onto the Li/W(110) substrate in less than 20 min. We come to this conclusion because after this exposure time MIES does not detect any features from the substrate and the LiF valence band is fully developed including the bandgap.

The energy spectra are collected at constant pass energy ($\Delta E=0.3, 0.1, 0.45$ eV FWHM for He^* , He^+ , He^{++} , respectively). The spectra of the slow electrons were taken in the following way: the difference in the work functions of the crystal (typically partially alkaliated) and the analyzer was overcompensated by biasing the electrostatic analyzer. We apply a small bias voltage between the tungsten surface and the analyzer in order to have the low-energy cut-off of the spectrum for clean W(110) at 5.3 eV. The variation of the position of the low-energy cut-off of the spectra reflects then the change of the surface work function with coverage starting from the value of the clean W(110) surface (5.3 eV). This procedure affects the collection efficiency below about 10 eV electron energy; it was checked that this procedure did not produce artefacts in the low energy part of the spectra. Therefore the spectra reported in this paper, do not represent truly angle-resolved electron spectra, but are to some extent angle-integrated at energies below 10 eV.

Fig. 1 displays the MIES spectra obtained during the growth of the Li adlayer on W(110) as well as the change of the spectra when the Li adlayer (first complete layer at room temperature) is exposed to LiF molecules.

Figs. 2 and 3 display ion impact electron spectra (IIES) for grazing surface collisions (5° with respect to the surface plane) of He^+ (Fig. 2) and He^{++}

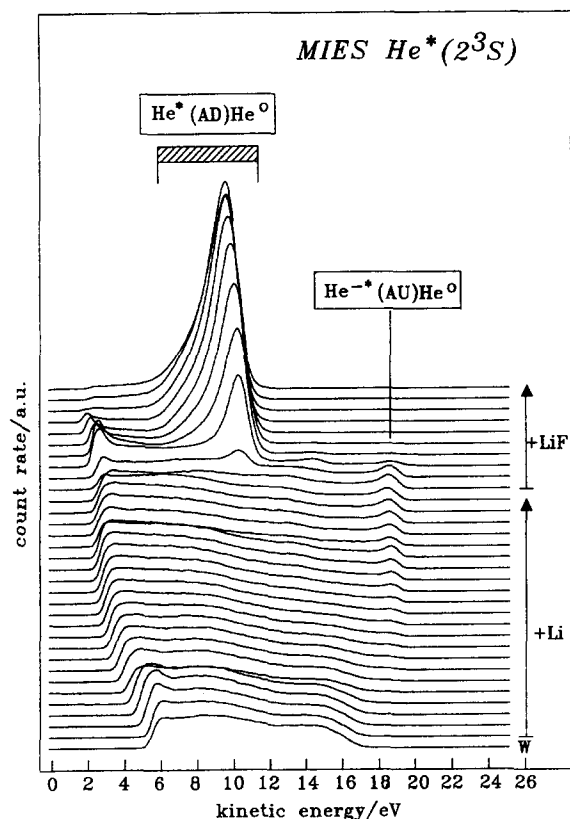


Fig. 1. Energy spectra of the electrons emitted in collisions of thermal energy He^* metastable atoms during the growth of a Li adlayer on W(110) and during the exposure of Li(1 ML)/W(110) to LiF molecules. The exposure rates remain constant during the film growth. During the exposure to Li the coverage changes by about 0.05 ML between adjacent spectra. The angle of ion incidence is 45° with respect to the surface.

(Fig. 3) projectiles obtained under the same conditions as discussed above. Spectra obtained during the exposure of W(110) to Li atoms were reported previously [21]. As in the photoelectron spectroscopy the total width of the MIES and IIES spectra is given by the maximum potential energy which can be converted into kinetic energy of the emitted electron, e.g. the ionization potential of He minus the work function of the surface. Thus, the total width of the spectra is work function dependent. We have indicated the processes that are responsible for electron ejection (see Section 3). Fig. 4 presents an enlarged version of the high-energy part of the IIES spectra with He^{++} projectiles. In

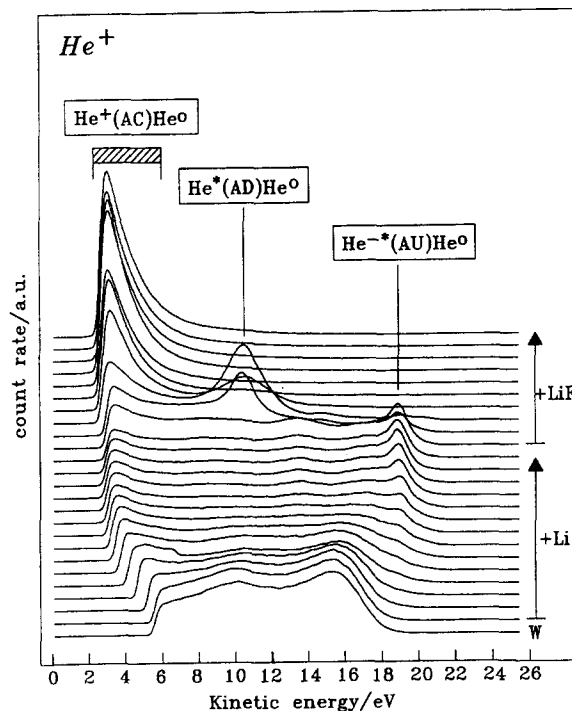


Fig. 2. Energy spectra of the electrons emitted in 50 eV collisions of He^+ ions during the growth of a Li adlayer on W(110) and during the exposure of Li(1 ML)/W(110) to LiF molecules; otherwise as in Fig. 1. The angle of ion incidence is 5° with respect to the surface.

order to demonstrate the strong decrease in the probability for “hollow atom” formation the exposure of W(110) starts with the top curve; the work function dependence of the structure due to autoionization (see Section 3) is also indicated.

Figs. 5 and 7 present simulations of selected experimental spectra (see Section 3) performed on the basis of the reaction schemes presented in Fig. 6 (see Section 3).

3. Interpretation of the results

The interpretation of the spectral features seen in collisions of ions or metastable inert gas atoms with metallic surfaces, such as clean W(110) and Li/W(110), has been given at several occasions [1,17,21–24]. In order to come to a sound interpretation of the structures seen for collisions with LiF layers, the experimental results will be com-

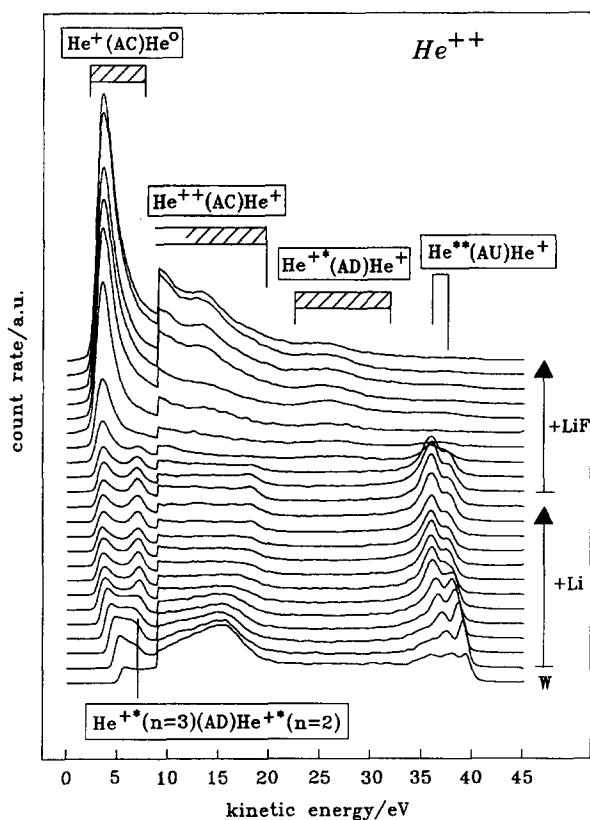


Fig. 3. Energy spectra of the electrons emitted in 100 eV collisions of He^{++} ions with Li(1 ML)/W(110) exposed to LiF molecules. Low-energy part of the spectra is reduced by a factor of 2. Otherwise as in Fig. 2.

pared with model spectra simulated as described in Refs. [3,4]:

(i) A reaction scheme (such as presented later in Figs. 6a and 6b) is chosen. It gives the sequence of events during the collision; in particular, it indicates which electronic processes can lead to electron emission.

(ii) For the interaction of He^* and the He^{q+} ions ($q=1, 2$) with the LiF film we have assumed that it can be approximated under the present conditions by a superposition of a repulsive Moliere-type potential and the image potential (in a.u.) [25,26]

$$V(z) = Z_{\text{eff}}^2(z)(\epsilon - 1)/(\epsilon + 1)1/4z.$$

$Z_{\text{eff}}(z)$ are the distance-dependent effective charges, z is the distance from the image plane; we have

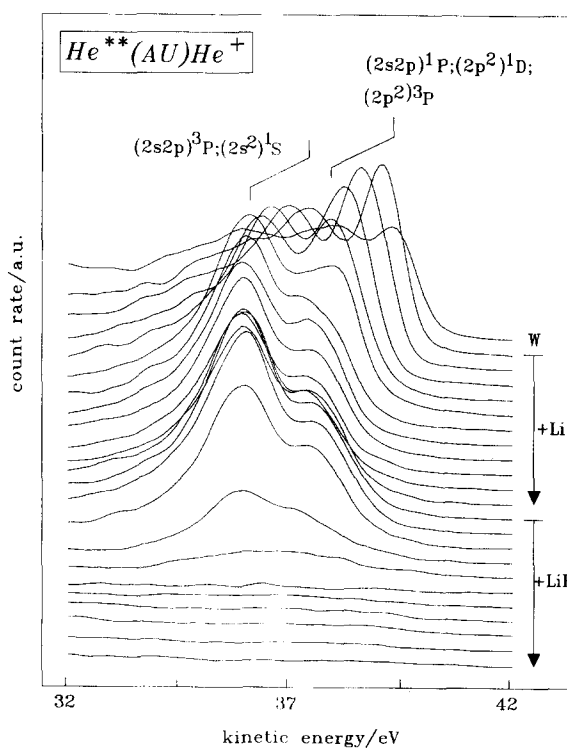


Fig. 4. High energy part of the spectra shown in Fig. 3.

assumed for the calculations that it is located 2.5 a.u. in front of the surface. The Z_{eff} are state-dependent, and are modeled as described in Refs. [3,4]. For ϵ we have chosen $\epsilon = 8.95$ [26,27], the static value. At present we have still to wait for more reliable potentials which may come from the determination of the image charge acceleration of helium ions in front of LiF surfaces [28]. The choice of the potentials is important for the spectral shape of the contributions to the electron spectra from the various electronic transition processes.

(iii) The transition rates required for the simulations on the basis of the reaction scheme of Fig. 6b are presented in Fig. 7c as a function of the projectile-surface distance z . The model that we use in order to obtain estimates for the z -dependence of the rates [3,4] is probably only justified for the interaction of the projectiles with conducting surfaces. From our simulations we find that the electron energy spectra are mostly sensitive to the absolute values of the employed rates around a

distance of about 3 a.u. from the surface (where most of the processes occur).

(iv) The surface density of states (SDOS) of the LiF adlayer is derived from the MIES spectra (see Fig. 5): these spectra are caused by one electronic transition process only (Auger deexcitation) and reflect the SDOS in a rather direct way (slightly modified by the projectile's motion with respect to the surface). We have assumed that this SDOS can also be employed for the simulation of the He^{q+} ion impact electron spectra.

3.1. MIES spectra

The adlayers were grown under the control of MIES. In particular, we studied the transition from a situation where hollow atoms, in this case He in the states $\text{He}^{**}(2l2l')$ ($l, l' = 0, 1$), are formed easily to a situation where hollow atoms are not expected to be formed in collisions of He^{++} with surfaces. We have shown that for He^{++} collisions with Li(1 ML)/W(110) hollow atoms are formed with large probability [2,21]. This holds for other low work function surfaces as well [24]. On the other hand, hollow atoms are not expected to be formed for He^{++} collisions with alkali halide surfaces such as LiF [5,9]. The W(110) substrate covered by one complete adlayer (1 ML) of Li was exposed to LiF molecules until

- the band gap of LiF has fully developed [29], and does not show any spectral feature due to surface defects, and
- the F 2p valence band has fully emerged, e.g. features a width of approximately 4 eV [29].

The MIES results obtained during the formation of the Li adlayer were analyzed in Ref. [30] and will not be discussed here any further. The results obtained after the formation of a dense LiF film (third curve from the top in Fig. 1) were simulated by using the surface density of states (SDOS(1)) shown in Fig. 5 (insert), a work function of 2.7 eV and a separation of 7.9 eV between the valence band maximum and the Fermi level. This corresponds to a separation of 10.6 eV between the valence band maximum and the vacuum level in good agreement with the findings of Ref. [29]. The SDOS derived from the MIES results is in good agreement with that of Ref. [31] (projected

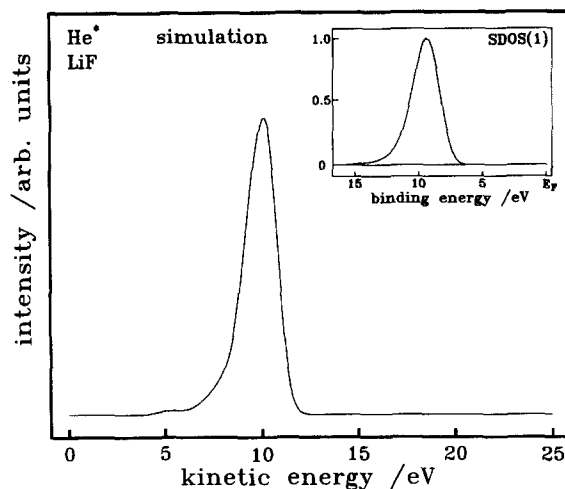


Fig. 5. Simulated electron energy spectrum for collisions of He^* metastable atoms with LiF films (for comparison with top spectrum in Fig. 1). The insert shows the surface density of states (SDOS(1)) employed for the calculations.

to the surface F atom) after taking into account a phonon broadening [32] of 1 eV of the valence band states.

It is assumed for the simulations that the electron emission is entirely due to Auger deexcitation involving F 2p electrons (see the structure denoted by $\text{He}^*(\text{AD})\text{He}$ in Fig. 1); resonance ionization of the metastables is inhibited by the LiF bandgap. Thus, electron emission due to the Auger capture process cannot occur. The simulated MIES spectra shown in Fig. 5 agree well with the experimental data. The SDOS derived in this manner will be used in Section 3.2.2 in the simulation of IIES spectra for He^+ and He^{++} collisions.

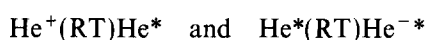
3.2. Ion impact electron spectroscopy

The electron emission spectra were studied during the growth of the Li adlayer on W(110) and during the growth of a LiF film on Li(1 ML)/W(110) for:

- He^+ (50 eV) collisions under 5° with respect to the surface plane,
- He^{++} (100 eV) collisions under the same conditions.

3.2.1. Electron emission from Li/W(110)

The interpretation of the electron energy spectra obtained during the adsorption of alkali atoms on W(110) (Figs. 2–4) has been given on several occasions [1,17,21–24] and is based on a qualitative agreement with simulated spectra. We do not present a detailed analysis of these spectra here. Instead, the reaction scheme of Fig. 6a summarizes the collision dynamics for the surface covered by Li qualitatively (some of the weaker processes, as f.i. the Auger capture processes, have been omitted for clarity): for He^+ collisions the two consecutive resonant electron transfer (RT) processes



lead to electron emission caused by (i) the interatomic Auger process, Auger deexcitation (AD) [33,34] via



[17], and (ii) the intra-atomic Auger process, autodetachment (AU) of He^{-*} [21,35]



The electron transfer between He^{++} and the surface is, as for He^+ collisions, mainly resonant (RT) (see reaction scheme of Fig. 6a). This leads to the formation of He^{+*} ($n=2, 3$) and He^{**} ($2l2l'$) ($l, l'=0, 1$) species [1,21,24]. These states manifest themselves by their decay via inter- and intra-atomic Auger processes, Auger deexcitation and autoionization (of He^{**}).

The strong feature between 35 and 40 eV in Figs. 3 and 4 is due to the autoionization of “hollow atoms” in the states He^{**} ($2s^2$), He^{**} ($2s2p$) and He^{**} ($2p^2$) [3,21,36]. The probability for their formation is rather high for collisions with low workfunction surfaces, such as for Li(1 ML)/W(110). The position of the structure due to autoionization of He^{**} depends on the electronic structure of the surface, in particular on the work function. In addition, the relative intensities from the decay of the two groups of states, $(2s2p)^3\text{P}$; $(2s^2)^1\text{S}$ and $(2s2p)^1\text{P}$; $(2p^2)^3\text{P}$, ^1D depends on the electronic structure of the surface.

The sharp structure, labeled He^{+*} ($n=3$) (AD) He^{+*} ($n=2$) in Fig. 3, is due to the Auger deexcitation of He^{+*} ($n=3$) ions formed by the

resonant capture of one surface electron by the He^{++} ions.

As pointed out previously [3,21,22,24], the interaction of the products He^+ and He^* with the surface gives additional channels for electron emission. The corresponding features are the same as already discussed above for He^+ collisions.

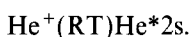
3.2.2. Electron emission from Li/W(110) exposed to LiF

Electron emission spectra induced by the interaction of He^{q+} ($q=1, 2$) ions and He^* projectiles with alkali halide films were reported by us previously [5–8]. We follow essentially the discussion of Ref. [6].

For He^+ collisions the probability for formation of He^{-*} quickly decreases upon exposure of Li/W(110) to LiF because of (i) the increase of the surface workfunction, and (ii) the decrease of the surface density of states near the Fermi level E_F upon F adsorption. Instead, a structure, labeled



appears in Fig. 2. It is caused by Auger deexcitation (AD) of He^* ($1s2s$) interacting with fluorine atoms. The Auger deexcitation process involves a F 2p electron (instead of a W 5d electron as in collisions with Li/W(110)). Apparently, the He^* atoms are still formed via the resonant capture of a surface electron to the projectile, e.g. by



We have supported this conclusion by simulating the spectra of electrons emitted when a He^* atom collides with fluorine (not shown). Good agreement in the position of the structure is obtained when assuming that the surface density of states is strongly enhanced at binding energies between 7 and 10 eV with respect to the Fermi level E_F by the surface adsorbed fluorine atoms [37] on the one hand, and is strongly decreased near E_F by the transfer of charge from the Li to the F atoms on the other hand.

With further increase of the exposure the LiF bandgap completely inhibits the resonant transfer of electrons to the projectile



Fig. 6 displays the reaction scheme with the dominant reaction channels active under these conditions. The He^* ions can be neutralized only by Auger capture



involving two electrons from the valence band of the LiF film. We neglect the possibility for resonant neutralization into the $\text{He} 1s^2$ ground state. This process should however be weak because there are no occupied states of the LiF film in resonance with the $\text{He} 1s$ level.

Fig. 7a (curve 1) shows the result of a simulation of the He^+ induced spectra under these conditions; we have used the SDOS(1) derived from the MIES data (displayed in Fig. 5). The Auger capture rate as a function of z (AC_2) is displayed in Fig. 7c. The simulation correctly predicts the strong rise below about 7 eV, but fails to reproduce the weak intensity above this energy (up to about 13 eV). This contribution can be understood qualitatively in at least two different ways:

(1) The film exhibits geometric imperfections at the surface, such as steps, kinks, corners etc. In violent collisions of the He^+ projectile molecular effects such as discussed in Refs. [38,39] may become important: the $\text{He} 1s$ binding energy of the He^+ which is in close contact with the surface is considerably larger (of the order of 5 to 10 eV) than in the undisturbed He^+ ion. This corresponds to the “demotion” of the $\sigma 1s$ orbital often observed in ion–atom collisions [40]. Thus, more potential energy is available for the Auger capture process, and consequently electrons with kinetic energy higher than simulated with the smooth surface can be ejected.

(2) Electrons with kinetic energies higher than 7 eV are obtained when occupied defect states above the valence band maximum are present in the LiF band gap; such states could be induced by the ion impact [12,13]. The simulation (2) in Fig. 7a was performed with SDOS(2) shown in Fig. 7a (insert); it differs from SDOS(1) solely by its defect-induced part.

Further studies are required to distinguish between these two (and eventually additional) possibilities to explain the observed high energy tail.

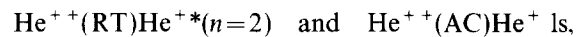
For He^{++} collisions the features due to the

resonant transfer of surface electrons rapidly decrease upon LiF exposure. This holds in particular for the narrow structures due to the decay of He^{++} ($n=3$) and of the “hollow atoms” He^{**} . Collisions with a closed LiF film show no indication for the formation of “hollow atoms” (see Fig. 4). A new structure appears at intermediate exposures, labeled

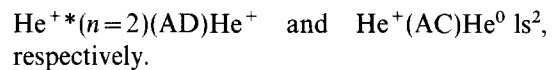


Simulations suggest that it is due to Auger deexcitation involving collisions of He^{++} with fluorine atoms involving F 2p electrons.

After the formation of a closed LiF film, e.g. when the bandgap characteristic for LiF has appeared, the only electronic transitions that apparently can lead to electron emission appear to be (see reaction scheme of Fig. 6b)

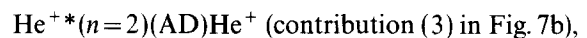


followed by

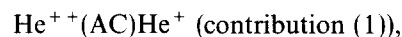


In particular “hollow atoms” He^{**} ($2l2l'$) are not formed anymore.

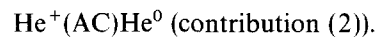
We have confirmed these statements concerning the collision dynamics by performing simulations based on the scenario discussed above (see also the reaction scheme shown in Fig. 6b). As for He^+ impact we have employed SDOS(2). The relevant transition rates are presented in Fig. 7c as a function of z . The simulation is shown in Fig. 7b. The comparison with curve 18 of Fig. 3 identifies (i) the weak contribution between about 24 and 30 eV as



(ii) the shoulder between above 13 eV extending to about 24 eV as



and (iii) the emission below about 7 eV as



Although no qualitative changes are produced, the actual shape and magnitude of the contributions (2) and (3) depends somewhat on the presence of defect-induced states in the bandgap. Moreover,

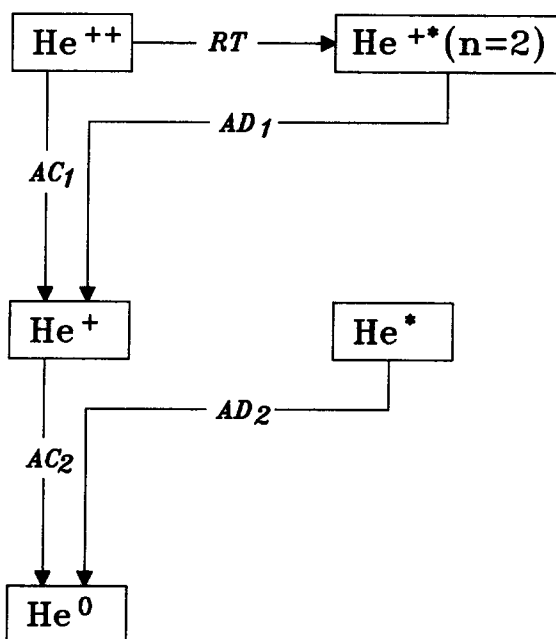
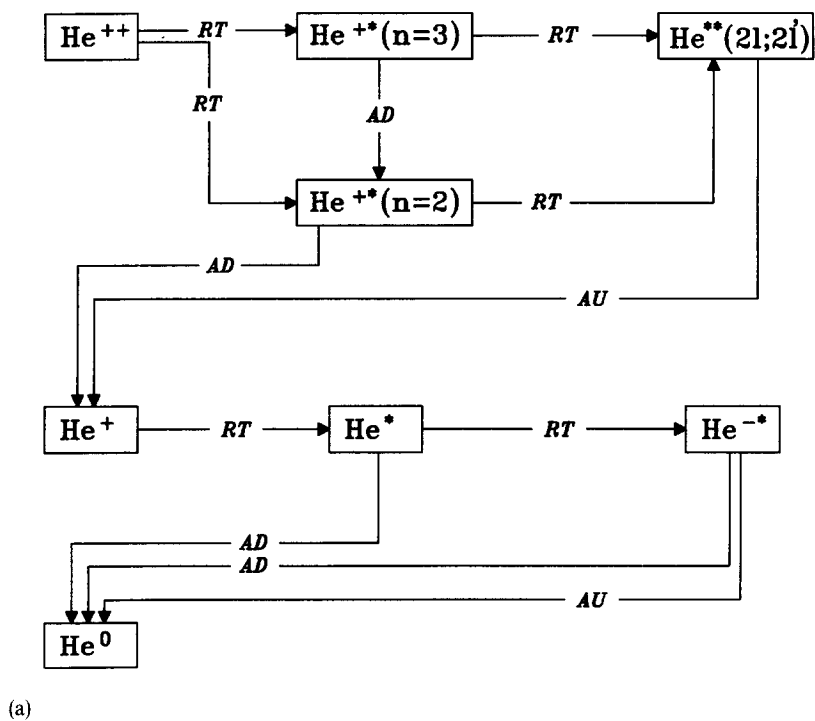


Fig. 6. (a) Reaction scheme for collisions of He projectiles with a Li covered metal surface. AD: Auger deexcitation; RT: resonant transfer; AU: autodetachment or autoionization. (b) Reaction scheme for collisions of He projectiles with LiF surfaces. RT: resonant transfer; AC: Auger capture; AD: Auger deexcitation.

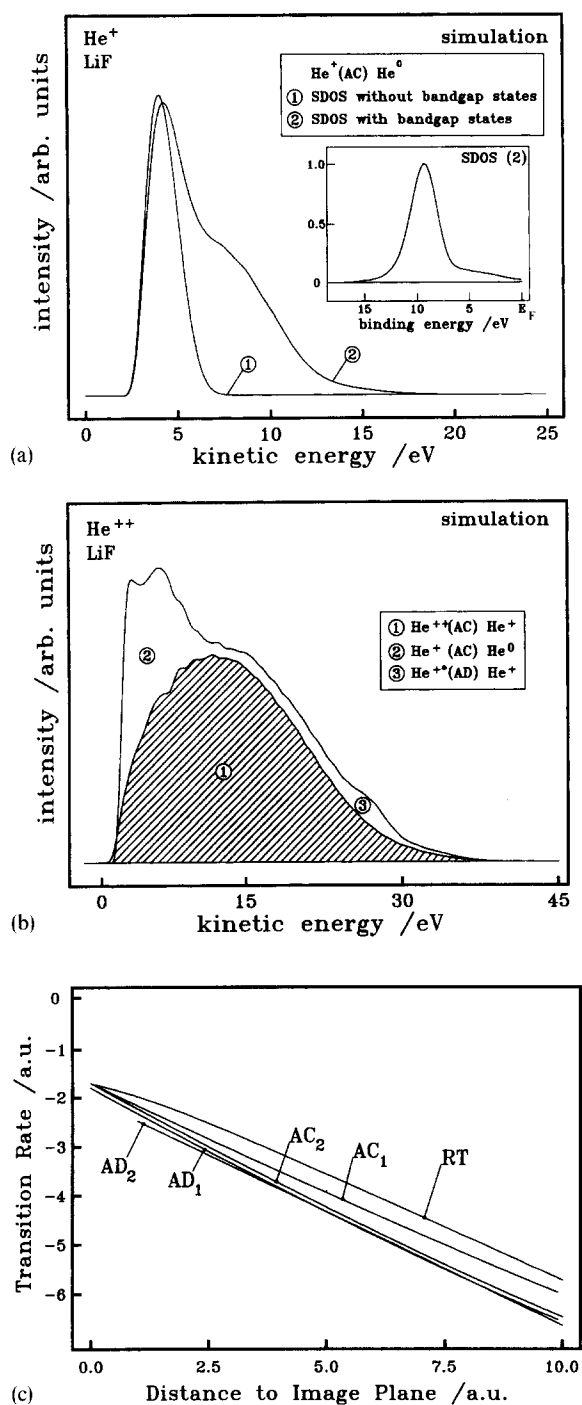


Fig. 7. (a) Simulated electron energy spectra for collisions of He^+ ions with LiF films. The insert shows the surface density of states (SDOS(2)) employed for the calculation of Figs. 7a and 7b. (b) Simulated electron energy spectra for collisions of He^{++} ions with LiF films. (c) Rates for the various electronic transitions shown in Fig. 6b.

we cannot exclude that the spectra below about 10 eV are to some extent influenced by secondary electron emission phenomena.

Any more quantitative analysis of the He^{++} induced electron emission spectra must take into account the formation of defects initiated by the injection of vacancies into the valence band of the LiF film by the transfer of electrons from the valence band to the projectile. As is shown in the reaction scheme of Fig. 6b up to 4 vacancies per He^{++} collision may be injected into the valence band.

We cannot exclude that a radiationless transition may be in competition with the electronic transition processes discussed so far. This could be the quasi-resonant capture [41–44] of F 2s electrons into the 1s He^{++} orbital leading to the formation of He^+ ions: the level shift of 1s He^{++} close to the surface may bring this level into resonance with F 2s.

4. Summary

A Li adlayer on W(110) and LiF films on a W(110) surface saturated with Li were grown under the control of metastable impact electron spectroscopy (MIES). Ion impact electron spectroscopy (IIES) was applied in order to gain information on the dynamics of electron transfer processes in front of these surface structures: we have studied the electron emission induced by slow He^+ (50 eV) and He^{++} (100 eV) ions in grazing collisions.

The large probability for resonant transfer of electrons from the Li covered W(110) surface to the projectile leads to the formation of the excited states, $\text{He}^* 1s2s$, $\text{He}^{-*} 1s2s^2$ and $\text{He}^{**} (2l2l')$ ($l, l' = 0, 1$) (“hollow atoms”), respectively. The energy spectra reflect the inter- and intraatomic Auger processes from the decay of these states.

For collisions with LiF films the probability for resonant transfer of electrons into excited states of the projectile is small because of the LiF bandgap. As a consequence the formation of the “hollow atoms” He^{**} does not occur anymore. The neutralization of the projectiles He^+ (He^{++}) takes now the place mainly by one (two consecutive) Auger capture processes. The quasi-resonant electron

transfer of a F 2s electron into the He 1s orbital may also contribute to the formation of He⁺ in He⁺⁺ collisions with the LiF film.

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