EXCITATION OF Li(2p) IN SLOW COLLISIONS OF Li $^+$ IONS WITH CESIATED W(110) SURFACES

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The excitation of the Li(2p) state in low-energy collisions of Li $^+$ ions with low work function surfaces is studied for impact energies below 1.0 keV. The yields of excited atoms and electrons are measured for the scattering from cesiated and oxidized cesiated W(110) surfaces characterized by AES, LEED, and $\Delta\phi$. It is concluded that Auger deexcitation of the 2p state populated by resonant charge transfer between the Li $^+$ projectile and the partially cesiated surface strongly influences the photon yield. For the oxidized surface it is shown that resonant electron exchange between the solid and the Li(2p) state is not the mechanism for projectile excitation. It is proposed that projectile excitation is caused by a direct transition between the 2s and 2p states of the Li $^+$ projectile which is neutralized on its way towards the surface.

1. Introduction

The mechanism for neutralization of alkali ions into excited states in collisions with clean metal surfaces at high collision energies (100 keV, typically) is well established [1]: resonant electron transfer from occupied metal states occurs into excited states of the projectile. According to the Doppler shift the highest energy which metal electrons can have in the rest frame of the projectile is $\hbar k_F v_{ion}$ above the Fermi level ($T = 0$ K) [2]. Here k_F is the Fermi wave vector, and v_{ion} the projectile velocity component parallel to the surface. Thus, excitation by resonant neutralization will occur when

$$(E^* - E_F) < \hbar k_F v_{ion}, \tag{1}$$

where E^* is the binding energy of the electron in the considered excited state. Consequently, there will be a threshold for excitation at several keV impact energy, and a monotonous rise of the excitation probability towards larger impact energies [1].

Resonant electron transfer between the ground state of alkali atoms and metal surfaces at low impact energies (400 eV, typically) was studied extensively by Los and coworkers (see ref. [3] for a review). Basically, efficient

resonant electron transfer between projectile and metal occurs when the distance between projectile and surface is smaller than some freezing distance which is of the order of 8 a.u. for alkali atoms, typically.

When applying this picture to the excitation of Li(2p) in low energy Li^+ collisions (such that the Doppler shift mechanism does not yet operate, below 2 keV, typically) we should expect the following behavior:

As a starting point we assume that the surface can be described by the Fermi energy and the macroscopic work function ϕ. Excitation by resonance neutralization (RN) into Li(2p) should take place as soon as $\phi < E^*(2p)$ holds at projectile–metal distances smaller than the freezing distance. $E^*(2p)$ is the binding energy of the 2p electron. Thus one would predict a threshold as a function of ϕ for 2p excitation at $\phi = 2.7$ eV which is the 2p binding energy at the freezing distance modified by the image charge potential. The excitation probability should be weakly dependent on the projectile energy. Surfaces with low work functions can be produced conveniently by partially cesiating the metal surface [3]. Depending upon the amount of adsorbed Cs the metal work function will decrease from its value for the clean metal, 5.3 eV for W(110), to 1.4 eV for a Cs coverage of about 0.64 ML.

Earlier studies [10] have shown that the local electrostatic potential significantly influences the electron exchange process. This has the consequence that the 2p excitation as a function of ϕ will not necessarily have a well defined threshold at $\phi = 2.7$ eV, but may already occur at larger ϕ.

In order to test these predictions we have studied the Li(2p) excitation for the scattering of Li^+ from cesiated W(110) surfaces for which the work function varies in the range 5.3–1.4 eV. We have carried out the following experiments:

(1) Comparison of the photon yield stimulated by Li^+ and $Li^0(2s)$ collisions: The dependence of the 2p yield on the initial charge state of the projectile furnishes information whether some "charge equilibrium" is reached prior to the excitation process. For sufficiently small work functions ($\phi < 3.0$ eV) excitation by RN into excited states should not operate for neutral projectiles because ionization of the projectile has to preceed RN into excited states.

(2) Comparison of the yields for Li(2p) excitation and electron emission as a function of the cesium coverage: Electron emission is expected to occur when the Li(2p) state is deexcited to Li(2s) by an Auger process in front of the surface, and therefore will furnish information on the influence of radiation-less deexcitation processes following RN into the 2p state.

(3) Oxidation of the cesium adlayer: This removes the metal-like states in the toplayer of the surface formed by the Cs atoms. Therefore the oxidized surface cannot any longer, not even approximately, be considered as a free electron metal as in the case of the cesiated surface. This should strongly affect the 2p excitation yield.

2. Apparatus

A detailed description of the surface scattering apparatus can be found elsewhere [4–6]. Its scheme is given in fig. 1 of ref. [6]. A number of important changes have been made, and will be described briefly.

The closed cycle He cryopump is replaced by a titanium sublimation pump in order to be able to bake the scattering chamber at 200°C. The base pressure in this chamber is now below 10^{-10} Torr. A CMA for AES is incorporated into the scattering chamber in order to verify the cleanliness of the prepared W(110) surface prior to the preparation of the Cs adlayer, and for the determination of the adatom concentration after preparing the adlayers. A quadrupole mass filter has been added for residual gas analysis.

Most of the results reported required a monoenergetic Li^+ ion beam: Li metal is heated to 400°C in order to produce a Li vapor atmosphere of about 10^{-4} Torr at the location of a Re ribbon held at about 1100°C. The Li^+ ions produced by surface ionization at the hot ribbon are extracted, focused, and accelerated to the desired energy. The beam flux as measured with a Faraday cup is 3×10^{10} ions s^{-1} mm^{-2} at 1 keV beam energy at the W(110) target. The energy width is estimated to 0.1% FWHM. A detailed description of the source can be found elsewhere [7].

The Li^+ beam impinges upon a W(110) surface which replaces the polycrystalline tungsten ribbon employed in our previous work. The procedure for cleaning the crystal is described in ref. [5]. During the measurements the crystal is at room temperature. Some results are reported which were obtained with Cs multilayers on W(110); for this purpose the crystal was cooled to about 150 K.

The cesiated and oxidized cesiated W(110) surfaces were characterized in some detail in an ion-pumped standard apparatus for surface diagnostics (Model TNB-X of Perkin-Elmer) bakeable at 250°C. The base pressure in this apparatus is 2×10^{-11} Torr. It is equipped with AES, LEED, $\Delta\phi$, and a quadrupole mass filter.

3. Results

The data are represented as yields for product formation, the total number of products of a given species divided by the projectile beam current. Such yields could be measured as a function of the beam energy, the incidence angle with respect to the surface normal, and, in particular, as a function of the coverage of W(110) by Cs, and of the exposure to oxygen of the partially cesiated surfaces. We have ourselves concentrated to the study of the 2p excitation and the production of electrons for selected incidence angles and energies as a function of the Cs coverage.

3.1. Results for clean and oxygen covered W(110)

The W(110) surface showed the expected LEED pattern without extra spots. C and O impurities before cesiation were < 0.05 of the W(350 eV) AES signal.

The ion fraction P_+ of the scattered projectiles is unity within the accuracy of the experiment both for the clean and the oxygenated W(110) (see also ref. [10]).

Table 1 compares absolute values for the photon yields for clean W(110) ($\phi = 5.3$ eV), partially cesiated W(110) ($\theta_{Cs} = 0.20$ giving the maximal photon yield at 500 eV impact energy), and an oxidized Cs monolayer on W(110). Results are for 1 keV impact energy and near-grazing incidence. The efficiency of the photon detection system was calibrated with a light standard. The absolute values of the yields are estimated to be accurate within a factor of two. Uncertainty comes mainly from the possible error in the calculation of the solid angle accepted by the photon detector.

If clean W(110) is exposed to oxygen, the photon yield rises by a factor of 10 although the work function rather increases. Because of the low yields no systematic studies have been made with high work function surfaces.

3.2. Results for partially cesiated W(110)

For Cs coverages $\theta_{Cs} > 0.6$ ML extra spots characteristic of a hexagonal structure of the Cs film appear in addition to the normal LEED spots. This

Table 1
Absolute photon yields for Li$^+$ collisions at 1 keV impact energy and 85° incidence angle for selected surface conditions and values for the higher excited states for W(110) covered by 1 ML Cs

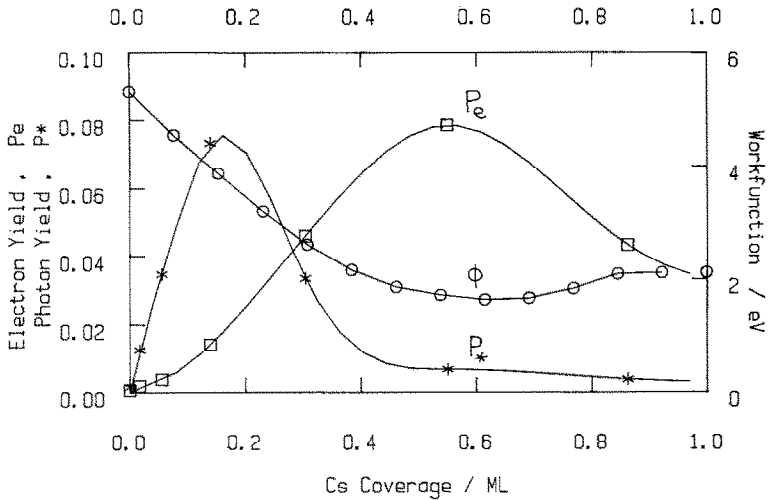| Condition	Yield
W(110), clean	
($\phi = 5.3$ eV)	$< 3 \times 10^{-4}$
W(110)/O	
(O exposure = 10 L)	3×10^{-3}
W(110)/Cs	
($\theta_{Cs} = 0.2$ ML)	8×10^{-2}
W(110)/O, Cs	
(O exposure = 6 L, $\theta_{Cs} = 1$ ML)	24×10^{-2}
W(110)/Cs	
(yields in units of Li(2p–2s) at $\theta_{cs} = 1$ ML)	
Li(2p)	1.0
Li(3d)	0.1
Li(3s)	0.02

Fig. 1. The Li(2p–2s) photon yield, P_*, the yield for electron emission, P_e, (arbitrary units) for 1 keV Li$^+$ ions colliding with partially cesiated W(110) versus the cesium coverage for 85° incidence angle of the Li$^+$ beam with respect to the surface normal. Also shown is the surface work function versus the cesium coverage.

pattern persists up to the saturation of the surface with Cs. For $\theta_{Cs} < 0.6$ ML the substrate spots are observed only.

Fig. 1 compares the yields for photon and electron emission, P_* and P_e, as a function of θ_{Cs}; also shown is the θ_{Cs} dependence of the work function. The beam parameters are given with the figures. The coverage by Cs, θ_{Cs}, has been determined from the magnitude of the Cs(564 eV) signal: it saturates at 1 ML Cs which is the highest coverage which can build up at room temperature. Results for 500 eV impact energy are qualitatively similar; the maximum of P_* seems to shift to slightly smaller coverages.

The work function measurements were made in the surface diagnostics machine, but compare well with some checks for $\phi(\theta_{Cs})$ carried out in the scattering machine.

No rise of P_* occurs when a second cesium layer starts to build up on a W(110) surface cooled to 150 K. The formation of the multilayers was monitored by AES.

An indication for the occurrence of a maximum in P_* as a function of θ_{Cs} was already seen in ref. [4] for the scattering of neutral Li from polycrystalline tungsten.

Table 1 gives some information about the excitation of other Li states. 2p excitation is stronger by about one order of magnitude than 3d excitation which is the next strongest populated Li state. Emission from the deexcitation of d-states ($n = 3$ to 6) and s-states ($n = 3, 4$) was observed.

Within the reproducibility of the measurements (0.05 L) the maximum electron yield coincides with the work function minimum which is not the case for the maximum photon yield. The electrons could not be energy analyzed, but were identified by their behaviour in an applied magnetic field. Again qualitatively similar results have been obtained for 500 eV impact energy.

When neutral Li atoms of the same energy are employed as projectiles, P_* and P_+ are very similar to the results in fig. 1 for Li$^+$ projectiles. Electron emission was not studied for neutral projectiles.

The neutrals were produced by colliding the Li$^+$ ions under grazing incidence with a polycrystalline tungsten ribbon of sufficiently low work function mounted in front of the single crystal target, and removing the remaining ions. An alternative way of producing Li neutral beams with higher intensity and better beam quality is described in ref. [5]. The method employed here has the advantage that the switching from ionic to neutral projectiles can be carried out without breaking the vacuum, and changing the surface conditions.

The ion fraction P_+ of the scattered projectiles decreases linearly with increasing θ_{Cs}, and is below 0.15 at $\theta_{Cs} = 0.3$; for $\phi < 3.0$ eV the surface scattered projectiles are therefore predominantly neutral. Detailed studies of P_+ as a function of θ_{Cs} and the projectile energy can be found in ref. [10].

3.3. Results for oxidized cesiated W(110)

The following discussion refers to a W(110) surface saturated by Cs which is exposed to oxygen.

The LEED pattern characteristic for the Cs film persists up to the oxygen exposure where the minimum work function occurs (fig. 3). With oxygen coadsorption the Cs spots become sharper and stronger. They disappear rather suddenly at the oxygen exposure where the work function minimum is located, and for larger exposures we find the W(110) pattern only.

In order to further characterize the oxidation process of a Cs monolayer on W(110) AES and $\Delta\phi$ measurements were made as a function of the oxygen exposure (fig. 2). Measurements were carried out in the diagnostics apparatus, but similar results were also obtained in the scattering apparatus. Qualitatively similar AES and $\Delta\phi$ results have been reported for other alkali–substrate combinations [13].

Together with what is known from UPS [8] and combined UPS, $\Delta\phi$, and MIES (metastable impact electron spectroscopy) studies [9] of the oxidation of Cs films and monolayers we characterize the surface structure as follows: up to an oxygen exposure of 2.2 L (minimum of the work function) the oxygen is incorporated below the toplayer which remains metallic and mainly consists of Cs atoms. The work function rise and the kink in the Auger signal indicate that the oxygen now starts to form the toplayer. Once the work function
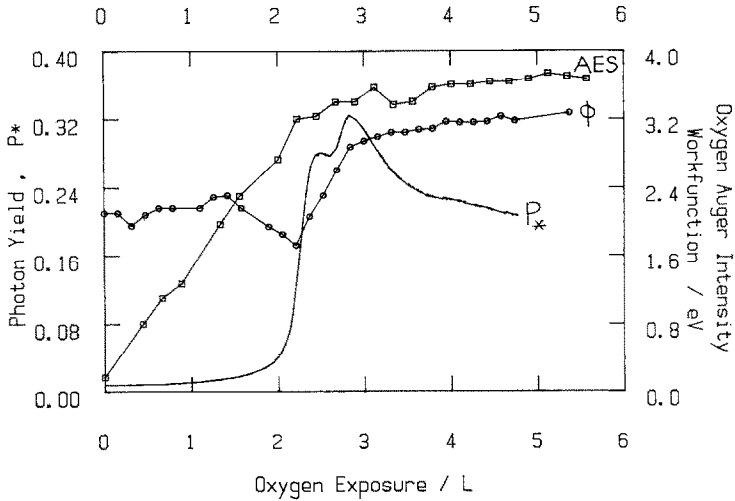
Fig. 2. The oxygen (512 eV) Auger signal, the work function of W(110) precovered by 1 ML cesium, and the photon yield versus the oxygen exposure.

reaches 2.8 eV, there are no free Cs valence electrons left in the toplayer because all of them are involved in the binding of the O_2 molecules [8,9].

Fig. 3 presents P_* and P_e (1 keV Li^+ collisions) for a W(110) surface precovered by 1 ML Cs which is exposed to oxygen. Within the reproducibility of the measurements the pronounced increase of both P_* and P_e takes place
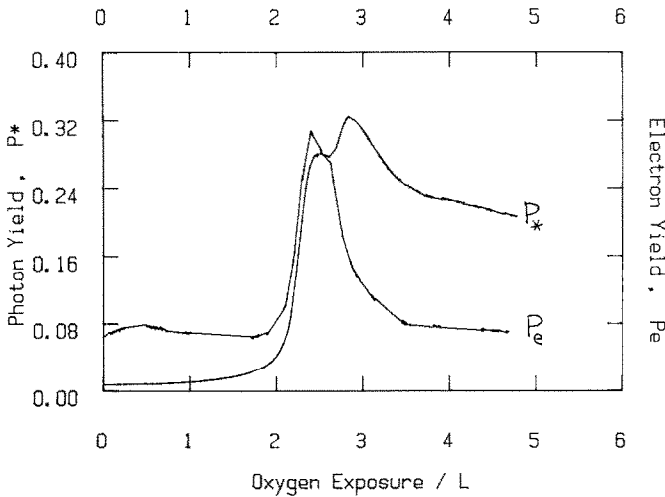

Fig. 3. P_* and P_e versus the oxygen exposure of W(110) precovered by 1 ML cesium. Beam parameters as in fig. 1.

in the exposure range where the work function minimum is found. The peak of P_e is confined to the exposure range where $\phi < 2.4$ eV, and the maximum of P_e is close to the minimum of ϕ; P_* has a shoulder at this value, but its main maximum occurs at a larger oxygen exposure.

Photon yields of a similar magnitude are observed when Cs multilayers are oxidized. No desorption of Cs or oxygen could be observed with AES during the progress of the oxidation process.

P_* behaves similarly as a function of the oxygen exposure when neutral projectiles are used. Electron emission was not studied.

4. Discussion

4.1. Clean and cesiated W(110)

Very little 2p excitation is observed on clean W(110) up to the highest studied energy of 1.0 keV. This is in agreement with ref. [1] where no Na(3p) excitation occurred for Na$^+$ ions colliding with Ni(111) below 2 keV. Their interpretation is as follows: at such low energies there is no resonance between occupied metal states and the Na(3p) level; the additional energy gained by the Doppler shift in the rest frame of the projectile is not sufficiently large to bring them into resonance with the projectile's 3p level (see eq. (1)). This small excitation yield could possibly be influenced by defects of the metal surface.

The cesiation brings occupied metal states in resonance with Li(2s) and even with Li(2p). The Li$^+$ ion yield P_+ behaves as was reported in ref. [10]: the charge state of the reflected projectile switches from predominantly ionic to neutral when the work function becomes lower than the 2s binding energy (modified by the image force) at the freezing distance.

In the following we will attempt to give an interpretation of the results for cesiated W(110) by treating the cesiated surface as a free electron metal and anticipating that resonant electron transfer between the alkali ion and occupied states of the metal leads to 2p excitation (see introduction). Adopting the simplest picture that the variation of the electrostatic potential along the surface can be neglected resonance between 2p and occupied metal states occurs when the work function lowers to 2.7 eV which is the binding energy of the 2p electron at the freezing distance. Thus one would expect to see the onset for 2p excitation at $\phi = 2.7$ eV. Fig. 1 shows however a nearly linear rise of P_* starting from $\phi = 5.3$ eV. A maximum of P_* is observed around $\theta_{Cs} = 0.2$ ML. As can be seen from fig. 1 this maximum appears well before the minimum of the work function where the maximum electron emission is found, and where also the maximum in the Li$^-$ ion yield occurs [11,12].

The linear rise of P_* can be understood when assuming that even single Cs adatoms may modifiy the electrostatic potential of the surface in such a way

that RN into the 2p state becomes feasible. This argumentation has already been used in ref. [10] in order to explain the linear decrease of P_+ starting from $\phi = 5.3$ eV.

The following considerations offer a convincing explanation for the coverage dependence of P_e in fig. 1: when RN into the 2p state takes place, it is followed by rapid Auger deexcitation (AD) of the 2p state to the 2s ground state. The vacancy in the 2s state of Li is filled by an electron coming from the surface (either from an Cs adatom or the bulk) whose excess energy is transferred to the Li(2p) electron which, as a consequence, is ejected. This mechanism is also responsible for the rapid singlet to triplet conversion observed when He($2\,^1$S) metastables collide with low work function surfaces [9] (see in particular fig. 18 of ref. [9] with 2s(^{3}S He) and 2s(^{1}S He) replaced by Li(2s) and Li(2p), respectively).

Neglecting again the variation of the electrostatic potential along the surface electrons will be emitted into the vacuum when

$$(E(2s) - \phi) > E(2p). \tag{2}$$

$E(2s)$ and $E(2p)$ are the binding energies of the valence electron in the 2s and 2p state of Li, respectively. Thus electron emission can be observed as long as ϕ remains below about 1.8 eV; this is the case around the minimum of the work function. Of course, AD is not restricted to this small range of work functions, but will occur over a much wider range: the 2p electron will then be transferred to empty states of the solid above the Fermi level [9], but will not be ejected.

Fig. 1 shows indeed the expected behavior: electron emission is mainly limited to the region around the minimum of the work function where $\phi < 2$ eV.

Further support for this view comes from P_e as a function of the oxygen exposure (fig. 3). As can be seen in fig. 2 ϕ drops below 1.8 eV in a narrow range of exposures around 2.2 L. In this region Cs atoms still form the toplayer, and AD can lead to the ejection of electrons.

Again, the electron emission at ϕ values outside the range predicted by eq. (2) may be caused by the variation of the electrostatic potential along the surface.

It is obvious from fig. 1 that AD for $\theta_{Cs} \geq 0.4$ ML is efficient enough to deexcite most of Li(2p) atoms formed by RN.

P_* behaves very similar as a function of the coverage for neutral Li and Li$^+$ projectiles (see also ref. [4]). It was shown in ref. [10] that the crossing seam for electron exchange in Cs$^+$ colliding with Cs–W(110) will be passed several times in a collision at grazing incidence provided that ϕ remains larger than roughly 3.0 eV. Anticipating that this also holds for Li$^+$ collisions it is reasonable to expect that in the region of closest approach of the projectile to

the surface some equilibrium with respect to charge state and excitation has been achieved. On the exit part of the trajectory this distribution of states will be modified by resonant electron exchange and AD independent of the initial charge state of the projectile.

Summarizing, for small Cs-coverages ($\theta_{Cs} < 0.2$ ML) the detected Li(2p) atoms are just those which, after being created by RN into the 2p state, survive deexcitation caused by the Auger process. The coverage dependence of the photon yield would then reflect the increasing probability for AD with increasing Cs coverage.

For larger Cs coverages ionization of a neutral Li projectile seems not to be possible under grazing incidence [10]. Under such circumstances RN into excited states cannot operate at all for neutral projectiles which must be ionized prior to RN into excited states. Thus, the independence of the results on the initial charge state of the projectile is difficult to rationalize on the basis of the model presented in the introduction. The weak, but significant population of higher excited states (table 1) also cannot be explained in this way.

In order to explain Li(2p) excitation for $\theta_{Cs} > 0.2$ ML we use an extension of the current picture for resonant electron transfer at surfaces; it was already employed by us to understand excitation in collisions of neutral Li with cesiated tungsten [4]: projectiles neutralized into the 2s state on their way towards the surface will not be in a pure 2s state when coming close to the surface or on adatom. Their electronic state should rather be described as a superposition of 2s, 2p etc. character. Nonadiabatic effects induced by the motion of the surface-perturbed projectile would prevent that a pure 2s state is recovered after the collision. Quantitative considerations on the basis of this model can be found in ref. [4].

The proposed mechanism could explain the weak, but clearly detectable excitation of higher lying excited states for which at all possible work function values there is no resonance with occupied states in the surface as long as the Doppler shift mechanism is not operative (in our case below 2 keV): only the neutralization into 2s is required before a deformation of the projectile's charge cloud with the admixture of excited states can take place. Some of the atoms in higher states (nl) may suffer deexcitation by RI and also by AD when retreating from the surface. Deexcitation processes will be more efficient than for 2p such that fewer excited atoms survive their retreat from the surface.

Deexcitation of higher excited states by AD could explain part of the emission of electrons observed at $\phi > 1.8$ eV (figs. 1 and 3): for Li(3d) electron emission is possible for $\phi < 3.8$ eV (see eq. (2) with $E(3d)$ replacing $E(2p)$).

Finally the observed independence of the excitation yields on the initial charge state of the projectile also at large Cs coverages can be explained: for $\theta_{Cs} > 0.2$ ML all Li$^+$ ions are already neutralized on their way towards the

surface before the deformation of the projectile's charge cloud which is efficient only near the surface occurs.

4.2. Oxidized cesiated surfaces

By the oxidation procedure documented in fig. 2 we have removed all free electrons in states resonant with the 2p states: above 3 L oxygen exposure the toplayer of the surface is transformed into an oxide layer. The onset of the integration of O_2 molecules into the toplayer manifests itself in the rapid rise of the workfunction from 1.6 to more than 3.0 eV. The UPS and MIES results [8,9] conclusively show that under such conditions the valence electrons of all Cs atoms are involved in the binding of the O_2 molecules. Up to an exposure of about 2 L the Cs atoms still form the toplayer of the surface (see also refs. [8,9]) and RN into 2p followed by efficient AD into 2s can take place. Indeed strong electron emission is observed only as long as the work function of the oxidized Cs layer remains low (see fig. 3).

From the rise of P_* at the exposure where the toplayer is transformed into oxide e.g. where the rise of ϕ occurs and RN into 2p becomes unlikely we conclude two things:

(1) Excitation of the Li(2p) state does not depend upon the possibility of RN of Li^+ into the 2p state, and

(2) processes like AD which would lead to 2p deexcitation take now place with much less efficiency because they require empty states above the Fermi level. Such states will not be available anymore after the oxidation of the Cs toplayer has been completed. This argumentation is entirely analogous to the one given in ref. [9] for the disappearence of the singlet to triplet conversion of $He(2^1S)$ on oxidized Cs films, and oxidized Cs monolayers on a Cu(110) substrate.

We observe reflected Li^+ ions for exposures larger than 4 L which indicates that even the probability for RN into the 2s ground state decreases considerably as compared to a cesiated surface. As for the cesiated surface the projectile looses the memory of its initial charge state almost completely.

We propose that in a first step, during the approach of the ion towards the surface, RN occurs into the 2s state, and, when the work function is low enough, also into the 2p state followed by (2p–2s) deexcitation by AD.

The excitation of the neutralized projectile is then induced by a direct (2s–2p) transition within the neutral atom when it is close to the surface or to an adatom, in particular. This mechanism was already invoked in ref. [2] to calculate the probability for low-energy surface Okorokov excitation. RN into 2p by electrons of the solid seems to be unimportant for the excitation of Li projectiles at small impact energies. Whether the projectile survives in its excited state during its retreat from the surface depends upon the probability for deexcitation by AD as is demonstrated by our results for oxidized cesiated surfaces.

5. Summary

The excitation of Li(2p) in collisions of Li$^+$ ions and Li(2s) atoms with partially cesiated and oxidized cesiated W(110) surfaces has been studied at impact energies below 1 keV under near-grazing incidence.

From the dependence of the (2p–2s) photon yield on the degree of coverage by cesium it is concluded that Auger deexcitation of the 2p state strongly influences the (2p–2s) photon yield. This is confirmed by the study of the electron emission as a function of the cesium coverage.

The results for cesiated surfaces are discussed on the basis of resonant electron exchange between the Li$^+$ projectile and the surface.

The results obtained for oxidized cesiated surfaces can be understood under the assumption that the mechanism for Li excitation under the studied conditions is a direct (2s–2p) transition (without involving electrons from the solid target) with the projectile which was neutralized on its way towards the surface.

Acknowledgement

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