K and *L* x-ray emission from hollow atoms produced in the interaction of slow H-like (I^{52+}) and bare (I^{53+}) ions with different target materials

Jian Sun,[1,*] Hirofumi Watanabe,[1,2] Masahide Tona,[1,2] Tsutomu Watanabe,[1] Nobuyuki Nakamura,[1] Chikashi Yamada,[1] and Shunsuke Ohtani[1,2]

[1]*Institute for Laser Science and Department of Applied Physics and Chemistry, University of Electro-Communications, Chofu, Tokyo, 182-8585, Japan*

[2]*CREST, Japan Science and Technology Agency, Chofu, Tokyo, 182-8585, Japan*

(Received 31 December 2007; published 3 March 2008)

Through the coincidence measurement of x rays with secondary electrons, x-ray spectra, and their yields have been measured in the interaction of H-like and bare highly charged ions (I^{52+} and I^{53+}) with different target materials (Be, C, Cu, and W). *K* x-ray yields are independent of the material, where the *K* vacancies are almost filled through *K* x-ray emission. *L* x-ray yields are affected by the electronic state of the targets and the interacting projectiles, because the states involved in the electron capture depend on the level structure of the projectile and the target and the possible number of electrons captured by the highly charged ions is different depending on the number of electrons in the target states ($2n^2$) which are resonant transferred to the projectile states in the dynamics of below-surface neutralization and deexcitation.

DOI: 10.1103/PhysRevA.77.032901 PACS number(s): 34.35.+a, 32.30.Rj, 79.20.Rf

I. INTRODUCTION

Since the first observation of hollow atoms by Briand *et al.* [1], many scientists have investigated these exotic atoms, which are multiply excited atoms with inner shell vacancies, to understand their formation and deexcitation mechanisms [2–9]. In the interaction of highly charged ions (HCIs) with solid surfaces, the hollow atoms are produced by capturing electrons from the target to Rydberg states. These hollow atoms decay via Auger and radiative transitions. Therefore, measurements of Auger electrons and x-ray spectra have been carried out [2,3]. Briand *et al.* observed the x-ray spectra with a crystal spectrometer for Ar^{17+} ions colliding with several kinds of surfaces [1,6,7]. In the spectra, KL^x x rays due to $2p \rightarrow 1s$ x-ray transitions with x spectator electrons in the *L* shell, were separately identified. In the case of very slow Ar^{17+} ($1-2$ eV/q) collision on H-terminated Si (Si-H) and Au, there is enough time to emit *K* x rays above the surfaces, where KL^1 and KL^2 lines are dominant in the x-ray spectra [2]. Since electrons in high Rydberg states ($n=20-30$) in the hollow atoms take many steps and a long time to decay to the *L* shell, the *L* filling rate by Auger cascading is slower than the *K* filling rate by x-ray transitions from the *L* shell. Therefore *K* x rays are emitted before the *L* shell is filled entirely in the case of the very slow collisions. With increasing the HCI's velocity, the *K* x rays are emitted from the hollow atoms after they penetrated the solid. Below the surface, the close internuclear collisions cause direct electron transfers from the inner shell of the target into the lower shell of HCIs, which is the so-called "side feeding" [8]. Therefore, in the case of Ar^{17+} collisions with kinetic energy of 8.5 keV/amu, the KL^x x rays are emitted with more *L* spectator electrons ($x=5-8$) because the *L* shell filling rate becomes faster [1].

The x-ray emissions are competing with Auger transitions in the filling of inner shells. The fluorescence yield of the y shell filling can be written as

$$\omega_y = \frac{\Gamma_r}{\Gamma_r + \Gamma_A}, \tag{1}$$

here Γ_r is the total radiative (x-ray) transition rate and Γ_A is the total Auger transition rate from upper shells to the y shell. Since the filling probability through Auger transition straightforwardly increases with the number of electrons, ω_y would be smaller with increasing the number of the spectator electrons. As formation and deexcitation mechanisms of hollow atoms were discussed by Briand *et al.* through the observation of x-ray energies in the KL^x spectra, the measurement of x-ray fluorescence yields also gives the information on these mechanisms.

In the neutralization of HCI, the electronic property of the surface layer has been considered to affect the formation mechanism of hollow atoms both above and below surfaces [3,9,10]. The KL^x intensity distribution shows a peaked structure at the satellites having a small number of L^x electrons for a Si-H surface different from that for an Au surface [2,6,7]. The *M* x-ray yields in the Th^{q+} ($7q$ keV) collision with Si, C, Au, and Ag exhibit a dependence on the targets, with the highest *M* x-ray yields for the Si target and the lowest *M* x-ray yields for the Ag target [11], which might be ascribed to the filling rate of the inner shells of the HCI in relation to the number of spectator electrons.

In this paper we show the measurement of x-ray spectra and their yields in the interaction of H-like and bare highly charged iodine ions (I^{52+} and I^{53+}) with different target materials (Be, C, Cu, and W) through the coincidence measurement of x rays with secondary electrons. We also discuss the formation and deexcitation mechanisms of hollow atoms produced in the collisions of HCIs with different target materials.

*sun@ils.uec.ac.jp

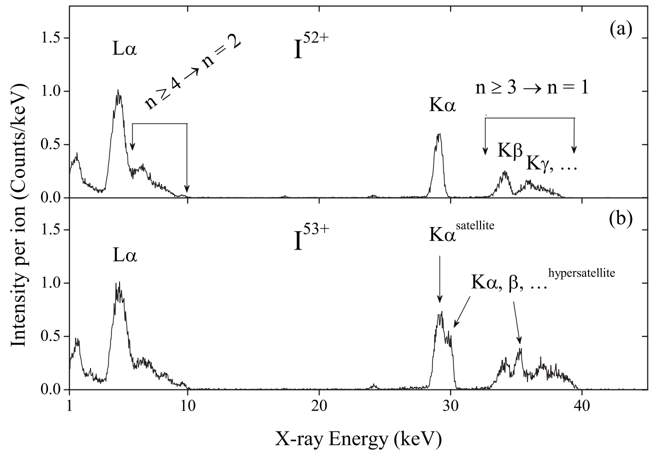

FIG. 1. X-ray spectra for (a) H-like I^{52+} and (b) bare I^{53+} ions impacts on a tungsten (W) surface. The intensity is photon counts emitted into a full solid angle by an incident ion per unit x-ray energy width (keV).

II. EXPERIMENT

The experimental procedure was almost the same as in the previous study [12,13]. Shortly, iodine HCIs were produced by an electron beam ion trap (EBIT), called the Tokyo-EBIT at the University of Electro-Communications [14]. The H-like and bare ions were selected by a sector magnetic analyzer with the kinetic energy of $3.5q$ keV. They were irradiated at normal incidence onto clean surfaces of Be, C (HOPG, highly orientated pyrolytic graphite), Cu, and W, which were prepared by bombardment of 1 μA Ar^+ ion beam with the kinetic energy of 2 keV for several minutes and annealing. Emitted x-ray signals were measured with a Si(Li) detector with active area of $2n^2$ and recorded with a multichannel analyzer (MCA). The Si(Li) x-ray detector was located at 70 mm far from the target on the 60° direction to the HCI beam axis. About 200 secondary electrons are emitted from the target by a single incidence. These burst electrons were accelerated by the −500 V target bias voltage and effectively counted by using an annular-type microchannel plate (MCP) in front of the target with 100% efficiency. These signals were fed to the MCA as gate pulses for the x-ray signals, and also counted as the number of incident ions. The coincidence measurement of x ray with secondary electron emissions could give the x-ray yield per ion. The Si(Li) detector was calibrated with x rays from radioisotopes of ^{55}Fe and ^{241}Am with known energies (^{55}Fe, 5.895 keV; ^{241}Am, 13.812 keV, 26.345 keV, and 59.537 keV).

III. RESULT AND DISCUSSION

X-ray spectra are shown for H-like and bare iodine HCIs (I^{52+} and I^{53+}) incident on the W surface and the HOPG surface in Fig. 1 and Fig. 2, respectively. The x-ray intensity was corrected by using the solid angle, the efficiency of the detector, and also normalized with the number of the incident projectiles and the energy width corresponding to one channel of the MCA. For these two targets the whole spectral structures are similar to each other. Both spectra consist of x-ray emissions of *K*, *L*, and *M* series. In the case of the H-like I^{52+}, the structure from 28 keV to 30 keV is $K\alpha$ ($1s2pnl \rightarrow 1s^2nl$) x rays, which is peaked at 29.1 keV, and spectra above 33 keV are $K\beta$ and $K\gamma$, etc., x rays. The *L* x rays and *M* x rays are separated at around 3.4 keV. For *L* x rays, the main part comes from $L\alpha$ x ray (*M* to *L* transition). The lower shoulder at upper energy is unresolved which consists of $L\beta, \gamma$, etc., x ray ($n \geq 4 \rightarrow n=2$). For bare I^{53+}, since there are two vacancies in the *K* shell, *K* x-ray spectra become complex. For example, the $K\alpha$ x-ray spectra include hypersatellite ($2pnl \rightarrow 1snl$) lines [15].

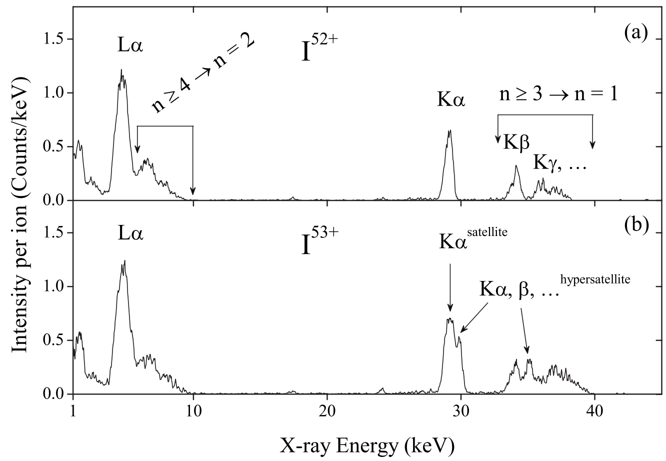

FIG. 2. X-ray spectra for (a) H-like I^{52+} and (b) bare I^{53+} ions impacts on a carbon (HOPG) surface.

The *K* x-ray yields were obtained by integrating from 28 keV to 40 keV. The uncertainty comes from the number of the incident ions, the solid angle, the efficiency of the detector, and the x-ray counts. As shown in Table I, it is found that *K* x-ray yields are 1.00 and 1.01 for H-like I^{52+} collisions with W and HOPG, respectively. For bare I^{53+}, the *K* x-ray yields increase to 1.98 and 2.03 for W and HOPG, respectively. Taking into consideration the fact that there are one and two vacancies in the *K* shell for I^{52+} and I^{53+} ions, these results show that the *K* shell vacancies are almost filled by x-ray transitions, i.e., the fluorescence yield of *K* shell (ω_K) is almost 100%, which is determined to be the ratio of the number of the emitted x rays and the number of the vacancies in *K* shell [16]. A similar result was obtained for Si(111)-H surface by Watanabe *et al.* [12]. Consequently, ω_K is almost 100% and independent of the target materials for H-like and bare iodine ions.

These results can be compared with the case of atomic iodine with one *K* shell vacancy whose ω_K is 88.2(28)%

TABLE I. Total *K* x-ray emission yields for H-like and bare iodine HCIs colliding with C, W and also Si-H as a reference.

	I^{52+}	I^{53+}	
W	1.00 ± 0.06	1.98 ± 0.12	This work
C	1.01 ± 0.06	2.03 ± 0.12	This work
Si(111)-H	1.03 ± 0.06	1.98 ± 0.12	Ref. [12]

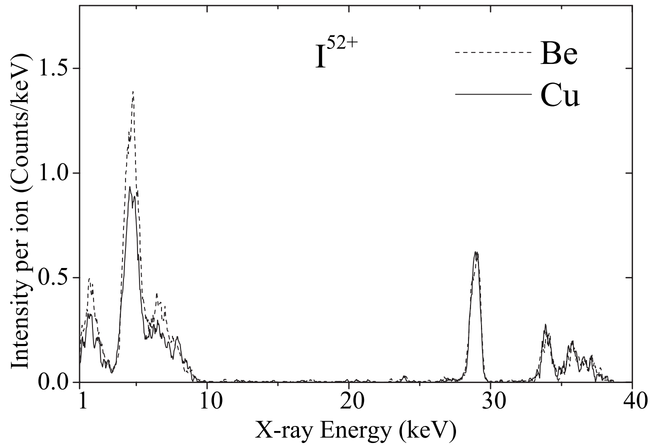

FIG. 3. X-ray spectra emitted in collisions of H-like I^{52+} ion with beryllium (Be) and copper (Cu) surfaces. The intensities are normalized so that the total *K* x-ray yield is 1.

[16]. This difference can be explained by Auger and x-ray transition rates. As seen in the spectra, the *K* x rays are mainly by the decay from the *L*, *M* to *K* shells. The transition rate of $(np)\rightarrow(1s)$ is scaled with ΔE^2 [17] (ΔE is energy difference between initial and final states) and the Auger rates scale approximately with the square number of electrons of upper states [7,18]. The L_{23} to *K* x-ray transition rate is 11.6 fs^{-1} [17] for atomic iodine, while each *KLL*, *KLM*, *KLN* Auger transition rate is slower than 0.2 fs^{-1} [19]. In the case of hollow atoms, however, the *K* x-ray transition rate is faster than 10 fs^{-1} and the Auger transition rate is slower than 0.1 fs^{-1}. Therefore, the filling of the *K* shell vacancies via x-ray transitions is faster than that via Auger transitions, which results in larger ω_K for the iodine HCIs than that for the atomic iodine.

We estimated the ω_K with respect to the number of the *L* spectator electrons (n_L) by using the HULLAC code [20] with neglecting transitions from *M* and higher states in the hollow iodine atom. The ω_K increases from 94% to 100% for KL^8 to KL^1. From our results, we would say that as soon as one electron decays to the empty *L* shell, it will immediately transfer to the *K* shell due to the very high x-ray transition rate. Thereafter, the *L* shell will be populated.

From the spectra of Fig. 1 and Fig. 2, it can be recognized that *L* x-ray yields for HOPG and W are significantly different. The intensities at the $L\alpha$ x-ray peak for HOPG and W surfaces are 1.23 and 1.01 (counts $\text{ion}^{-1}\,\text{keV}^{-1}$) by H-like I^{52+} irradiation, respectively. Figure 3 presents the x-ray yield spectra for H-like I^{52+} impacts on Be and Cu surfaces. The intensities of the spectra were normalized so that the *K* x-ray yield is 1, which is justified by the fact, as shown above, that ω_K is 100% and independent of the target materials. It is clearly seen that the intensities of *L* x rays for Be are larger than that for Cu.

We obtained the *L* x-ray yields by integrating from 3.4 keV to 10 keV, which are shown in Table II. As shown in Table II, the *L* x-ray yields seem to be dependent on the target *Z* (atomic number), i.e., the *L* x-ray yields for Be (Z=4) and HOPG (Z=6) are much larger than those for Cu (Z=29) and W (Z=74). The *L* x-ray yields change from 1.72 (W) to 2.31 (Be).

TABLE II. Total *L* x-ray emission yields from H-like and bare iodine ions impacts on Be, C, Cu, and W surfaces.

	I^{52+}	I^{53+}	
Be	2.31 ± 0.14		This work
C	2.17 ± 0.13	2.35 ± 0.14	This work
Cu	1.75 ± 0.11		This work
W	1.72 ± 0.10	1.87 ± 0.11	This work

According to the classical over barrier (COB) model [4,5], while a H-like I^{52+} HCI approaching a metallic surface with the work function of 5 eV, conduction electrons would be captured to very high Rydberg levels ($n\approx40-50$) starting at about $\sqrt{2q}/W_\phi\approx56$ a.u≈29 Å. The lifetime of the hollow atom ($\sim$10 fs) is longer than its approaching time above the surface ($\sim$5 fs). Then the time above the surface is not enough for subsequent cascading to inner shells of the hollow atom. Hence, most of the vacancies in inner shells such as *K*, *L*, and *M* would survive until it arrives at the surface [7,9]; therefore, most of *K*, *L*, and *M* x rays are emitted below the surface [1]. Highly exited electrons captured above the surface would be mostly peeled off [21] before the projectile penetrates the surface. Further, the target electrons in the inner shells would be transferred to the projectile directly through a near-resonant capture process, which is the so-called "side feeding" [8,21]. These electrons would decay to *M* shells through a few steps of Auger transitions. This

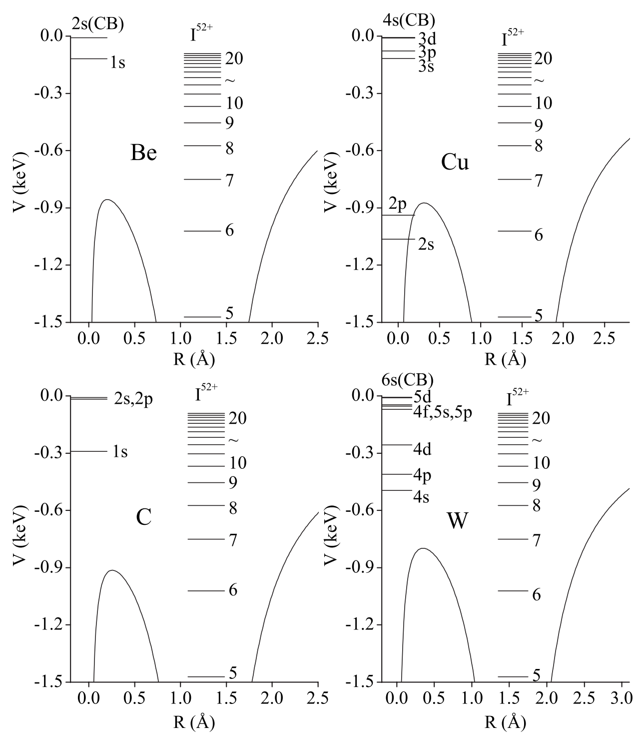

FIG. 4. The level energy matching between the HCI projectile (I^{52+}) and the target atom (Be, C, Cu, and W) together with the Coulomb potential barrier in between them. The position of the projectile exists at the center of the lattice spacing in the target. CB denotes the conduction band.

filling process below the surface and then the production of spectator electrons in a HCI would be strongly related to the electronic structure of the target, which should affect the *L* x-ray yields.

In order to see the energy level matching below the surface, we arrange the energy levels of the target atoms in comparison with those of the projectile. Figure 4 shows a kind of hypothetical potential energy diagram in the COB model between the target atoms [Be, C (HOPG), Cu, and W] and the I^{52+} projectile ion, where the projectile exists at the center of interstitial spacing in the target lattice, interacting with the nearest target atom. In Fig. 4 the Coulomb barriers are also shown, in which the effective nuclear charges estimated by Slater [22] are used for the respective target atoms.

As seen in Fig. 4, 1*s* electrons would be transferred to high Rydberg levels in the projectile from the Be and C target atoms for the side feeding, while, from the Cu target, electrons in the principal quantum number $n=3$ level, and from W electrons in $n=4$ are dominantly captured below the surface, respectively. The electrons captured by the projectile into high Rydberg levels would decay to the inner *M* shell mainly via Auger transitions. However, in this side feeding scheme, the number of those electrons would be different for the different target atoms, since the possible number is considered to be $2n^2$, which is the occupation number of the initial level *n* of the target atom before capturing ($2n^2=2$ for Be and C, $2n^2=18$ for Cu and 32 for W).

Therefore, the number of *M* spectator electrons would be different for the target atoms after cascading in the atomic states of the projectile. As mentioned above, for the Be and C targets there are a small number of *M* spectator electrons, which leads to higher *L* x-ray yields, while for Cu and W the relatively lower *L* x-ray yields were observed due to many *M* spectator electrons in the projectile.

IV. CONCLUSIONS

We investigate the x-ray spectra from the slow hollow atoms produced in the interaction of I^{52+} and I^{53+} with different target materials. We have demonstrated that the fluorescence yield of *K* shell vacancies in the hollow iodine atom is almost 100% which is independent of the target being closely related to the very fast x-ray transition rate to the *K* shell. On the other hand, the *L* x-ray yields are different for the different target materials. This can be attributed to the mechanism of the formation of hollow atom below the surface which is affected by the electronic structure of the target. In the side feeding scheme, the possible number of the electrons captured by the projectile is considered to be $2n^2$, which is the occupation number of the initial level *n* of the target atom before capturing ($2n^2=2$ for Be and C, $2n^2=18$ for Cu and 32 for W).

ACKNOWLEDGMENTS

This work has been supported by the CREST program, "Creation of Ultrafast, Ultralow Power, Super-performance Nanodevices and Systems" in the Japan Science and Technology Agency, and has been performed as an activity under the 21st Century Center of Excellence (COE) Program, "Innovation in Coherent Optical Science" at the University of Electro-Communications.

[1] J. P. Briand, L. de Billy, P. Charles, S. Essabaa, P. Briand, R. Geller, J. P. Desclaux, S. Bliman, and C. Ristori, Phys. Rev. Lett. **65**, 159 (1990).

[2] J.-P. Briand, S. Thuriez, G. Giardino, G. Borsoni, M. Froment, M. Eddrief, and C. Sébenne, Phys. Rev. Lett. **77**, 1452 (1996).

[3] H. Khemliche, T. Schlathölter, R. Hoekstra, R. Morgenstern, and S. Schippers, Phys. Rev. Lett. **81**, 1219 (1998).

[4] J. Burgdörfer, P. Lerner, and F. W. Meyer, Phys. Rev. A **44**, 5674 (1991).

[5] H. Winter and F. Aumayr, J. Phys. B **32**, R39 (1999).

[6] J.-P. Briand, G. Giardino, G. Borsoni, M. Froment, M. Eddrief, C. Sébenne, S. Bardin, D. Schneider, J. Jin, H. Khemliche, Z. Xie, and M. Prior, Phys. Rev. A **54**, 4136 (1996); J.-P. Briand, S. Thuriez, G. Giardino, G. Borsoni, V. Le Roux, M. Froment, M. Eddrief, C. de Villeneuve, B. d'Etat-Ban, and C. Sébenne, *ibid.* **55**, R2523 (1997).

[7] J.-P. Briand, S. Thuriez, G. Giardino, G. Borsoni, M. Froment, M. Eddrief, and C. Sebenne, Phys. Rev. Lett. **79**, 2591 (1997).

[8] L. Folkerts and R. Morgenstern, Europhys. Lett. **13**, 377 (1990); J. Limburg, S. Schippers, R. Hoekstra, R. Morgenstern, H. Kurz, F. Aumayr, and H. P. Winter, Phys. Rev. Lett. **75**, 217 (1995).

[9] F. Aumayr, H. P. Winter, J. Limburg, R. Hoekstra, and R. Morgenstern, Phys. Rev. Lett. **79**, 2590 (1997).

[10] J. W. McDonald, T. Schenkel, A. V. Hamza, and D. H. G. Schneider, Nucl. Instrum. Methods Phys. Res. B **240**, 829 (2005).

[11] G. A. Machicoane, T. Schenkel, T. R. Niedermayr, M. W. Newmann, A. V. Hamza, A. V. Barnes, J. W. McDonald, J. A. Tanis, and D. H. Schneider, Phys. Rev. A **65**, 042903 (2002).

[12] H. Watanabe, S. Takahashi, M. Tona, N. Yoshiyasu, N. Nakamura, M. Sakurai, C. Yamada, and S. Ohtani, Phys. Rev. A **74**, 042901 (2006).

[13] H. Watanabe, J. Sun, M. Tona, N. Nakamura, M. Sakurai, C. Yamada, N. Yoshiyasu, and S. Ohtani, Phys. Rev. A **75**, 062901 (2007).

[14] H. Watanabe *et al.*, J. Phys. Soc. Jpn. **66**, 3795 (1997); F. J. Currell *et al., ibid.* **65**, 3186 (1996).

[15] E. P. Kanter, R. W. Dunford, B. Krässig, and S. H. Southworth, Phys. Rev. Lett. **83**, 508 (1999).

[16] W. Bambynek, B. Crasemann, R. W. Fink, H.-U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. V. Rao, Rev. Mod. Phys. **44**, 716 (1972).

[17] S. T. Manson and D. J. Kennedy, At. Data Nucl. Data Tables **14**, 111 (1974).

[18] S. Winecki, C. L. Cocke, D. Fry, and M. P. Stöckli, Phys. Rev. A **53**, 4228 (1996); K. Suto and T. Kagawa, *ibid.* **58**, 5004

(1998).

[19] M. H. Chen, B. Crasemann, and H. Mark, At. Data Nucl. Data Tables **24**, 13 (1979).

[20] A. Bar-Shalom, M. Klapisch, W. H. Goldstein, and J. Oreg, The HULLAC Package-Computer set of codes for atomic structure and processes in plasmas.

[21] T. Schenkel, A. V. Hamza, A. V. Barnes, and D. H. Schneider, Prog. Surf. Sci. **61**, 23 (1999); L. Folkerts and R. Morgenstern, Z. Phys. D **21**, S351 (1991).

[22] J. C. Slater, Phys. Rev. **36**, 57 (1930).