Dissipation of potential energy through x-ray emission in slow highly charged ion-surface collisions

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X-ray emission yields from highly charged iodine ions incident on a hydrogen terminated silicon surface were measured. It was found that the K shell vacancies were filled through x-ray transitions with the probability of approximately 100%, while only about 20% of L shell vacancies were filled through x-ray transitions and almost all the M shell vacancies were filled nonradiatively. Dissipation of the potential energy E_p of an incident ion through x-ray emissions increases gradually with the number of L shell vacancies and amounts to 10% of E_p for the He-like I^{51+} ion. 30 to 40 % of E_p for I^{52+} and I^{53+} with K shell vacancies was measured to be dissipated mainly by K x-ray emissions.

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Interaction of highly charged ions (HCIs) with solid surfaces has been widely investigated in recent years with fundamental and practical interest [1–6]. A HCI is unique in that it has large potential energy E_p, for example, about 190 keV for I^{53+}, which is defined by the sum of the ionization energies to produce the HCI from a neutral atom, and in that it releases all of E_p in a short time (10 fs) on a nanometer-sized area during the interaction, which leads to dramatic surface effects. In order to obtain detailed understanding of the HCI-surface interaction, it is important to identify and distinguish E_p-dissipation channels and to measure quantitative fractions of distribution to the respective channels. So far, there have been two measurements of the retained fraction of E_p in the surface. Schenkel *et al.* [7] measured the energy deposition in a silicon solid state detector. It was found that 35 to 40 % of E_p of Ne-like Au^{69+} and He-like Xe^{52+} was traced in electronic excitations to produce numbers of electron-hole pairs more than 50 nm deep inside the target. Kentsch *et al.* [8] reported recently that 30 to 40 % of E_p of Ar^{q+} ($q=1-9$) was finally converted into heat in a copper target. In both studies, although the incident charge states of the projectiles were very different, it is interesting that similar and substantial fractions of E_p were measured to be retained in the targets.

A HCI is also unique in its great ability to suck up many electrons from the surface. In slow ($v_{\mathrm{ion}} < 10^6$ m/s) HCI-solid collisions, where most target electrons move faster than the projectile, an approaching HCI captures such active electrons into its high Rydberg states to form the so-called hollow atom. As the hollow atom approaches the surface, it emits Auger electrons and photons, and again receives electrons at the surface. High yields of secondary electrons and photons have been observed [9,10], which might carry away a large fraction of E_p. However, surprisingly, it has been considered that only about 10% of E_p has so far been accounted for in measurements of emitted secondary particles and photons [7]. This might be an underestimated value when a HCI emits high energy photons with large fluorescence yields, which should be a substantial fraction of the E_p dissipation. Actually, the E_p dissipation through high energy photon emissions has not been investigated quantitatively in the interaction of HCIs with very high charge state. This paper reports on systematic measurements of the fractional E_p dissipation through characteristic x-ray emissions from HCIs incident on the solid target.

Iodine HCIs, I^{q+} ($q=34-53$), were produced in an electron beam ion trap (EBIT) at the University of Electro-Communications, called the Tokyo EBIT [11] and extracted with kinetic energy of $q \times 3.5$ keV. After the charge selection by using a sector magnet, I^{q+} ions were introduced to a collision chamber and irradiated onto a target at normal incidence. The target in the present observation was a hydrogen terminated Si(111) crystal which has been used in the course of systematic investigations for the potential sputtering induced by HCI impacts [12,13]. HCIs have been irradiated with the rate of 10^4 cps at the maximum for about 100 h in the course of the previous and present experiments. Typically 10 nm^2 area could be decomposed by an ion incidence. Therefore, the area of 3.6×10^{-4} cm^2 could be affected by the irradiation of HCIs with the maximum dose, which was negligible with respect to that of the sample about 0.25 cm^2. Emitted x-ray signals were observed by using a Si(Li) detector located at 60° to the HCI beam axis with a solid angle of 2.9 msr. The pressure in the collision chamber was kept below 5×10^{-8} Torr during the observation. An individual HCI incidence on the target was monitored with 100% efficiency, by detecting a strong burst of emitted secondary electrons with an annular-type microchannel plate located in front of the target [14]. The number of secondary electron burst signals corresponding to that of incident HCIs was used for the normalization of x-ray emission yields and also used as gate pulses to pick up the true signals from the Si(Li) detector. For x-ray energies between 2 and 20 keV, the detection efficiency of the Si(Li) detector with a Be window of 8 μm thickness is considered to be 100%. Below 2 keV the measured x-ray intensity was corrected with the transmission

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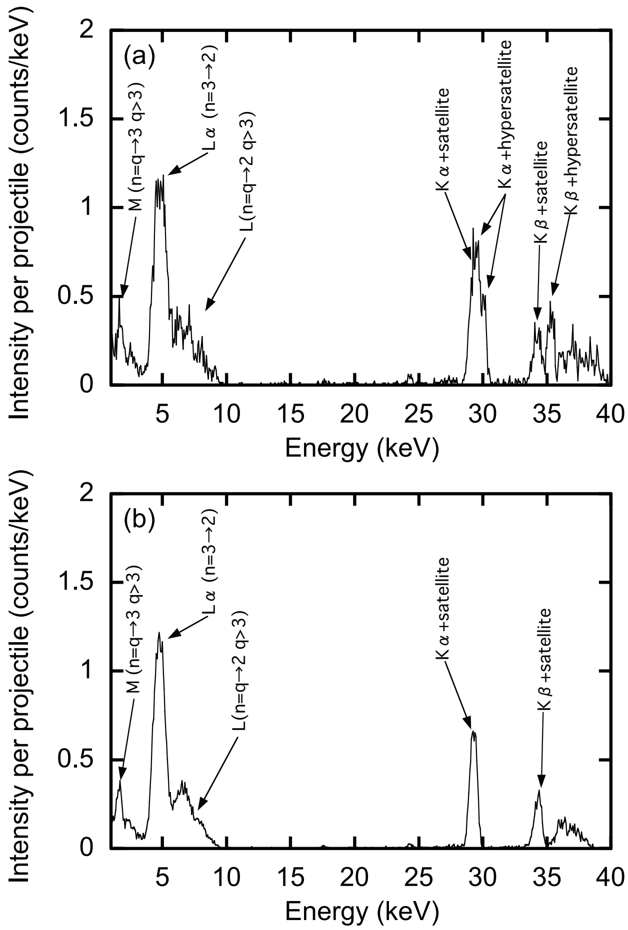

FIG. 1. X-ray spectrum obtained in collisions of bare I^{53+} ions (a) and H-like I^{52+} ions (b) with a hydrogen terminated silicon target, where the intensity is photon counts emitted into a full solid angle by an incident ion per unit x-ray energy width (keV).

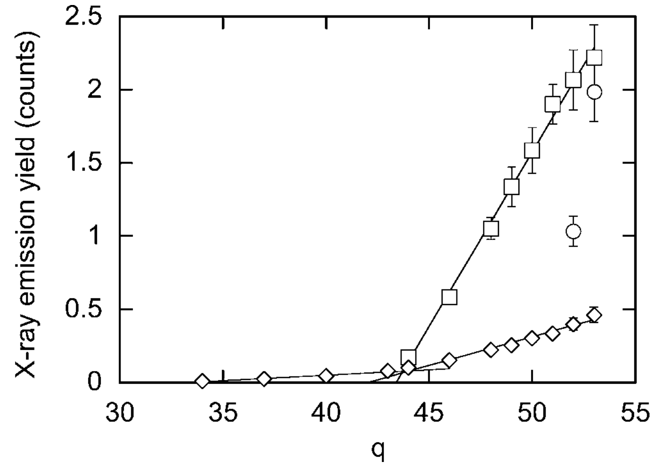

FIG. 2. X-ray emission yields from incident I^{q+} ions as a function of q. Circles are K x rays, squares L x rays, and diamonds M x rays. X-ray emission yield is photon counts emitted into a full solid angle by an incident ion.

coefficient of the Be window. For x-ray energies from 20 to 40 keV, the Si(Li) detector was calibrated by comparing the intensity of a Bremsstrahlung x-ray spectrum produced by a 63 keV electron beam in the EBIT to that measured by a Ge detector with 100% efficiency in this energy range.

Figures 1(a) and 1(b) show typical x-ray spectra observed for the bare I^{53+} ion and the H-like I^{52+} ion incident on the target, respectively. The x-ray energy was calibrated by using emitted x-ray lines from radioisotopes of ^{241}Am and ^{55}Fe. The x-ray intensity was corrected with the detection efficiency including the observation solid angle and normalized with a unit x-ray energy width corresponding to one channel of the multichannel analyzer used in the experiment. The whole spectral structures are similar to those obtained by Briand *et al.* [15] for bare and H-like ions of Fe and Kr incident on metallic targets. In both Figs. 1(a) and 1(b), there are a variety of K series x-ray spectra ($K\alpha, K\beta, K\gamma, \ldots$) in the energy range between 28 and 40 keV. In Fig. 1(b), a clear discrete series of lines is seen for the H-like ion incident on the target. This structure consists of the transition from upper $L, M, N, \ldots,$ shells to the single-vacant K shell (satellite). In Fig. 1(a), additional transitions appear for the bare ion incidence, which correspond to the transition from upper $L, M, N, \ldots,$ shells to the double-vacant K shell (hypersatellite). This rather complex spectral structure for the bare ion results from the sequential filling of the K shell vacancies. Below 10 keV, two series of L and M x-ray spectra are also observed for both the spectra. These are, in general, similar to each other, but the spectrum for the bare ion is slightly broader toward the high energy side. This is the contribution from the ion with double K shell vacancies. All the spectra observed here are considered to be due to x-ray transitions in hollow atomic states which are produced by multielectron capture collisions of I^{53+} and I^{52+} with the surface atoms.

From the measured absolute intensity, the x-ray emission yield was calculated, which is the number of photons emitted into the full solid angle per incident ion. Individual emission yields are shown in Fig. 2 for K, L, M x-ray series as a function of the incident charge state q of I^{q+} (q=34–53). The error bars include uncertainties due to the counting statistics, the calibration of the detection efficiency and of the observation solid angle. It should be noted that the low energy M x-ray emission yields might have additional uncertainties which come from the truncation of the x ray below 1 keV. It should be also noted that the x-ray emission yields are dependent on the number of electrons populated in lower shells, because Auger and radiative rates are sensitive to the number of those electrons. The filling rate to lower shells will be speeded up in the solid, because electrons can be transferred to these shells directly. Therefore there might be non-negligible velocity dependence on the x-ray emission yield of the penetrating ion in the solid. Since the quantitative estimate of the effect is not clear at the moment, the error caused by this effect is not included in the error bar. It can be seen that M x-ray emission yields are small in whole but increase gradually with q from q=34 (K-like ion), in which the slope becomes steeper from q=44 (F-like). A HCI with q higher than 43 has the hole(s) in its L shell. During the interaction, M and the upper shells would be filled rapidly by electron capture processes. The frequency of Auger

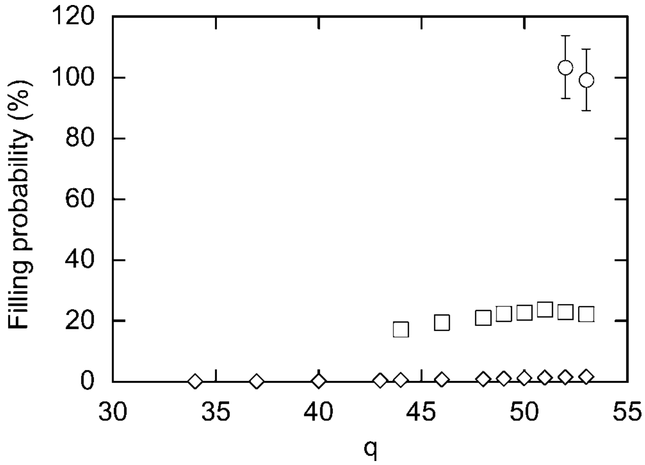

FIG. 3. The filling probability of K, L, and M vacancies by x-ray emissions. The number of x rays emitted in a single ion incidence is divided by the number of the vacancies. Circles are K shell, squares L shell, and diamonds M shell.

processes such as LMM and LMN transitions might increase with $q(\geqslant 44)$ as the number of L holes increases, which produces sequential M shell vacancies and thus increases M x-ray yields, even if the fluorescence yield would be small. The L x-ray appears at $q=44$, which comes from the fact that the x-ray transition is possible for the appearance of one hole in the L shell at the F-like I^{44+} ion. The x-ray emission yields for the L x-ray are larger than those for the M x-ray and increase linearly with q. Similarly the K x-ray can be observed only for the incident HCI with K hole(s), that is, in the present case $q=52$ (H-like) and 53 (bare ion).

Figure 3 shows the number of x rays normalized by the number of the vacancies, which is the filling probability of the shells through x-ray emissions. As shown in the figure, the filling probability of the K shell is estimated to be 100% within the experimental uncertainty. This might not be rather surprising, because the fluorescence yield of the I atom is close to 90% [16]. However, it should be noted that in the present case of the hollow atom the K shell is filled through x-ray transition with the probability of approximately 100%. On the other hand, for the L shell, about 20% of vacancies on average are filled by x-ray transitions, that is to say, about 80% of the L shell is populated through competing nonradiative Auger processes. In the case of the M shell the vacancies are almost filled through Auger processes.

The potential energy dissipated through x-ray emissions by an incident ion was derived by considering the calibrated number and the energy of emitted photons in the measured x-ray spectra for the respective I^{q+} ions. Figure 4(a) shows the dissipated energy as a function of incident charge q, and, as another arrangement of the data, the fraction dissipated to x-ray emission from the potential energy E_p of the incident ion is shown in Fig. 4(b). The dissipated fraction was obtained by dividing the dissipated total emitted x-ray energy by calculated E_p [17]. As seen in Fig. 4(a), the dissipation through x-ray emissions increases with increase of q and the rate of the increase changes at F-like and H-like, which shows the strong dependence on the shell structure of the ions. The importance of x-ray emissions for the E_p dissipation could be seen in the dissipated fraction of E_p in Fig. 4(b). For the ions which have vacancies only in the M shell, the dissipated fractions are almost zero. With increase of the L shell vacancies the fraction increases, which reaches 10% for a He-like ion. Since the K shell vacancies are almost filled through x-ray transitions, the fraction for the ions with K shell vacancies becomes very large and amounts to 30% for the H-like ion and to 40% for the bare ion.

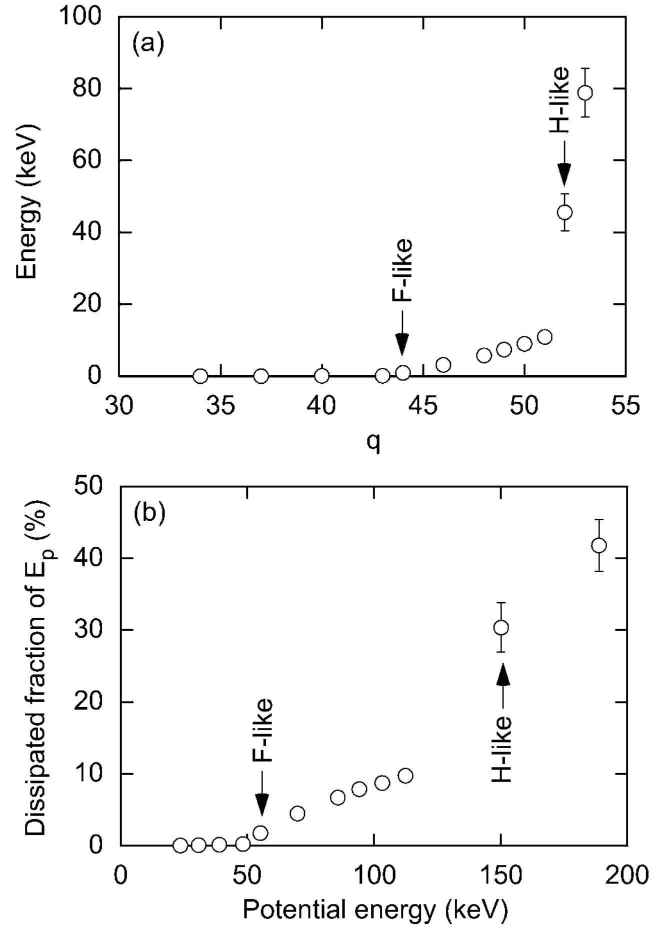

FIG. 4. The potential energy dissipated through x-ray emissions as a function of q (a) and the dissipated fraction of E_p as a function of E_p (b) for the incident I^{q+} ions.

From previous measurements, Schenkel *et al.* [7] estimated that about 10% of E_p could be traced in the emissions of the secondary particles and photons and in the production of plasmons. However, the present measurement shows that 10% of E_p could be dissipated only through x-ray emissions for the He-like I^{51+} ion, the ionic property of which is similar with that of the He-like Xe^{52+} ion used in the study by Schenkel *et al.* [7]. For the He-like ions with Z around 53, 10% of E_p is dissipated through x-ray emission and 40% is deposited to excitations of the solid. It should be noted that the E_p dissipation through x-ray emissions would be more significant for the few electron ions with higher Z, since the x-ray emission yields would increase with Z as the radiative transition rates increase. The present results could not be applicable directly to the result using Ne-like Au^{69+} by Schenkel *et al.* [7] and also to the results using Ar^{q+} ($q=1-9$) by Kentsch *et al.* [8].

As described before, Schenkel *et al.* [7] showed that 40% (48 keV) of the potential energy (E_p=121 keV) of the He-like Xe^{52+} ion was used to produce electron-hole pairs in the silicon solid state detector. This large retained fraction of E_p may contain the contribution of projectile's L x-ray absorbed inside the target, which might not be negligible, because the E_p dissipation through L x-ray emission was measured to be about 12 keV for the incident He-like I^{51+} ion with Z=53, next to Z=54 of Xe. The quantitative contribution is not clear of high energy photons emitted by the projectiles to the elementary excitations in the solid. Therefore, it is interesting to compare the pulse height distributions from a silicon solid state detector responding to the impacts of Ne- to He-like ions (I^{41+} to I^{51+}), with those of H-like to bare ions (I^{52+} to I^{53+}) having the same kinetic energies, which could separate out the effect of the elementary excitations by absorption of high energy photons and also could trace the quantitative contributions of the residual fraction of E_p.

In summary, we have measured the x-ray emission yield and the dissipation of the potential energy E_p of an incident ion through x-ray emission in collisions of I^{q+} (q=34–53) ions with a hydrogen terminated Si(111) surface. The x-ray emission yields clearly depend on the shell structures of the incident ions. K shell vacancies are filled through x-ray transitions with approximately 100% within uncertainties in the present experiment, while the filling of L shell vacancies through x-ray transitions is limited to about 20% of the vacant capacity. Almost all the M shell vacancies are filled through nonradiative transitions. Consequently, the E_p dissipation due to x-ray emissions increases gradually and amounts to 10% of E_p for the He-like I^{51+} ion. For H-like and bare ions with K shell vacancies, the fraction of 30 to 40 % of E_p was measured to be dissipated through high energy photon emissions, in contrast to less than 10% of E_p dissipated through secondary particle and photon emissions, estimated in the previous measurements [7].

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