Crystal Spectroscopy for K_α X-rays from Silicon Bombarded with Protons and Alpha Particles

By

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Abstracts

An automated Bragg spectrometer in which an organic electron multiplier is employed as an X-ray detector has been designed, and the K_α diagram and satellite X-rays from silicon bombarded with hydrogen and helium ions at MeV energies have been analyzed. The procedures of deriving the ionization cross sections from the satellite intensities are described.
The multiple KL^m ionization cross sections are compared with the theoretical binomial distribution, which means a statistical superposition of single ionizations.

1. Introduction

In 1970, Richard and his co-workers[1] first observed broad characteristic X-rays from heavy ion-bombarded targets with the use of a Si(Li) detector, and predicted a mixing of intensive satellite X-rays. In the next year, Knudson *et al.*[2] employed a Bragg spectrometer in order to precisely observe K X-rays from aluminium excited by energetic nitrogen ions, and succeeded in discriminating the satellite peaks. Burch and others[3] have similarly made a high-resolution study for K X-ray satellites from iron bombarded with oxygen ions. Ever since 1971, much research about satellite X-rays has been reported, and the K satellites have clearly been attributed to a K shell plus L shell ionizations of the target atom, denoted as KL^m where m is the L shell ionicity.

A high-resolution spectroscopy for K X-rays from ion-bombarded silicon has been reported by McWherter *et al.*[4] They have studied chemical shifts of X-rays for Si

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and SiO_2 bombarded with 0.8 MeV hydrogen, 3.2 MeV helium, 13.0 and 35.0 MeV oxygen ions. Kauffman and his co-workers[5] have measured the shifts for Si (solid) and SiH_4 (gas) excited by 45 MeV chlorine ions. Dutkiewicz *et al.*[6] have bombarded a SiO_2 target with protons at a fixed energy and discussed the satellite intensities in comparison with multiple ionization. In order to deeply interpret the mechanism of multiple ionization, high-resolution studies for proton- and alpha-induced silicon X-rays at various projectile energies are necessary.

X-ray detectors used in crystal spectrometers are thin-window and flow-mode proportional counters. Since the window foil considerably attenuates low energy X-rays, it becomes difficult to analyze X-rays at less than 2 KeV in energy. To avoid this defect, a vacuum X-ray detector such as a secondary electron multiplier should be applied.

In this paper, therefore, we describe an automated Bragg spectrometer having an organic and vacuum X-ray detector, and report the K_α X-ray spectra for silicon bombarded with hydrogen and helium ions. The derived cross sections for the KL^0, KL^1, KL^2, ··· ionizations are compared with the statistical theory presented by McGuire and Richard.[7]

2. Experimental

A hydrogen (H_2^+) or helium (He^+) ion beam extracted from a 4 MV Van de Graaff accelerator of Kyoto University was analyzed in energy, and well collimated by using a 90° analyzer magnet, a pair of Q-magnets and a defining slit system. The ion beam of 5 mm in diameter was injected into a target chamber and hit on a thick silicon target with a glancing angle of 45°, as shown in Fig. 1. The beam energies were 1.0-2.5 MeV for hydrogen ions and 1.5-2.5 MeV for helium ions, and the beam current was a few μA.

The assemblies of our crystal spectrometer were set in a 16″ vacuum chamber. X-rays emitted from the silicon target were made to pass through a soller slit* (No. 1 : 100 mm in length and 0.15 mm in blade spacing) and hit on an EDDT crystal ($2d$=8.80 Å, 1″×2″ in size) mounted on a rotatable holder. Diffracted X-rays were led into another soller slit *(No. 2 : 30 mm in length and 0.45 mm in blade spacing) and then counted by an X-ray detector. The θ-2θ relation between the crystal and the detector was accomplished by means of a gear system, and a laser beam was employed to align the spectrometer assemblies.

Photon counting was done by using a flexible channel electron multiplier† (chan-

* Manufactured by Rigaku-denki Co. Ltd., Takatsuki, Osaka.

† Manufactured by Matsushita Electric Industrial Co. Ltd., Central Research Laboratory, Kadoma, Osaka.

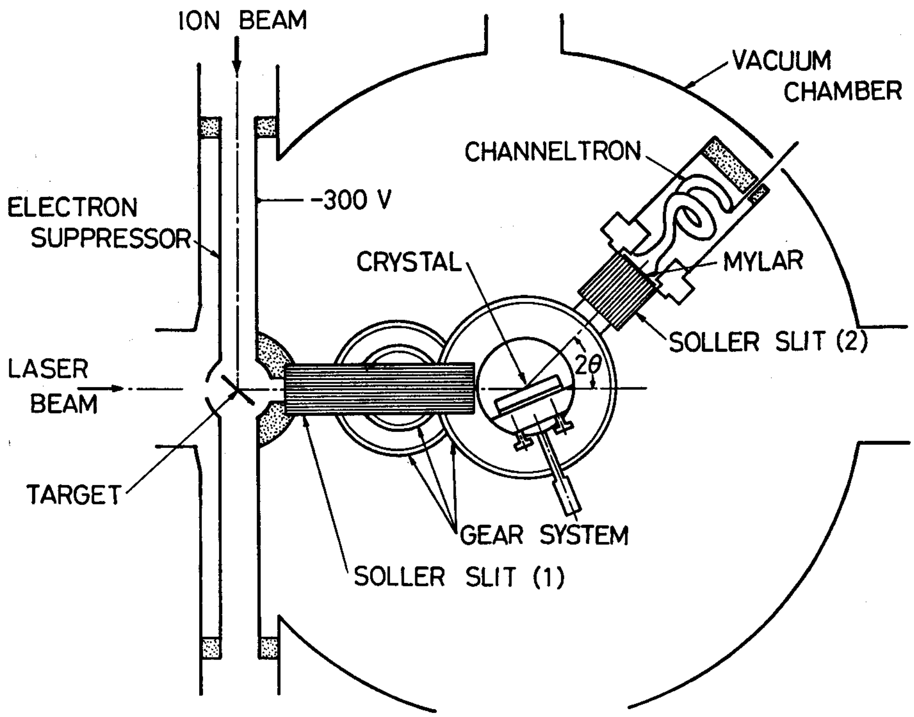

Fig. 1. Assemblies of the crystal spectrometer for analyzing X-rays emitted from ion-bombarded target.

neltron) made of an organic semiconductor. For these several years, we have employed this channeltron for detecting low energy electrons[8] as well as ions[9] and confirmed its reliable performance. Therefore we newly applied it as an X-ray detector, although its absolute counting efficiency against photon energy was hardly determined in the present experimental arrangement. Since the target and the detector were placed in co-vacuum, precaution was necessary to prevent stray electrons from entering the detector. For that reason, formvar film of about 50 $\mu g/cm^2$ in thickness was inserted in front of the channeltron mouse. The output X-ray signals from the detector were amplified, pulse-shaped and then accumulated on a multiscaler.

In order to automatically display the X-ray spectrum, we developed an electronic device. An IC scaling circuit generated one shot pulse after counting every ten to a thousand pulses which were sent from a beam current integrator. The shot pulse triggered a stepping motor which drove the arm of the X-ray detector with $2\theta=0.075°$ per step, while simultaneously one channel of the multiscaler was made to advance. Accordingly, each X-ray spectrum could be obtained by simply giving a start pulse to this auto-scanning system.

3. Results and Discussion

3.1. Thick target X-ray yields for diagram and satellite lines

In Fig. 2(a) and (b) are shown the K_α X-ray spectra for silicon bombarded with H_2^+ and He^+ ions at 2 MeV, respectively. The peak at $2\theta=108.2°$ is the diagram line (KL^0, 1.740 keV) and other peaks correspond to the satellite lines (KL^1, KL^2, KL^3). Assuming that each peak has a gaussian form and applying a least squares fitting, we get the FWHM values for the diagram lines as follows:

3.3 eV for H_2^+ bombardment, 3.6 eV for He^+ bombardment.

These values are somewhat broader than 2.0 eV for the Si-K_α diagram line observed by McWherter *et al.*[4] and the 1.9 eV for Al-K_α diagram line reported by Awaya and others.[10] The poorer resolution of our spectrometer is because the blade spacing of

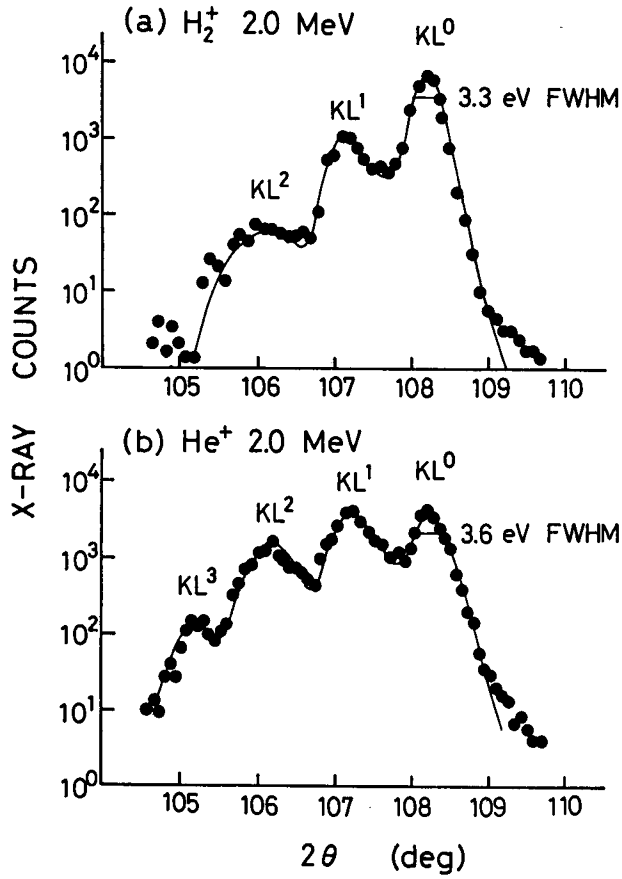

Fig. 2. Yield profiles of diagram and satellite X-rays emitted from silicon bombarded with hydrogen and helium ions at 2.0 MeV.
(a) by H_2^+ ion bombardment, (b) by He^+ ion bombardment.

the second soller slit was as broad as 0.45 mm.

The peak areas in Fig. 2 give the observed yields Y_m ($m=0, 1, \cdots$) for the KL^0, KL^1, $\cdots$ X-rays. In order to derive the thick target yields I_m ($m=0, 1, \cdots$) for X-ray production, corrections are necessary against the absorption of the formvar film, the counting efficiency of the channeltron, the reflection of X-rays on the crystal surface and other attenuations. These estimations were done as described below.

When H_2^+ ions were incident at 2 MeV in energy, the observed count ratio for the Si-K_α and -K_β X-ray groups was $(K_\beta/K_\alpha)_{exp}=0.0249$. On the other hand, the theoretical intensity ratio has been presented by Scofield[11] as $(K_\beta/K_\alpha)_{theo}=0.0171$. The difference between the experiment and the theory is attributable to the overall counting efficiency of the detecting system, including the above attenuations. If we assume that the overall efficiency varies linearly between the K_α ($KL^0=1.740$ keV)

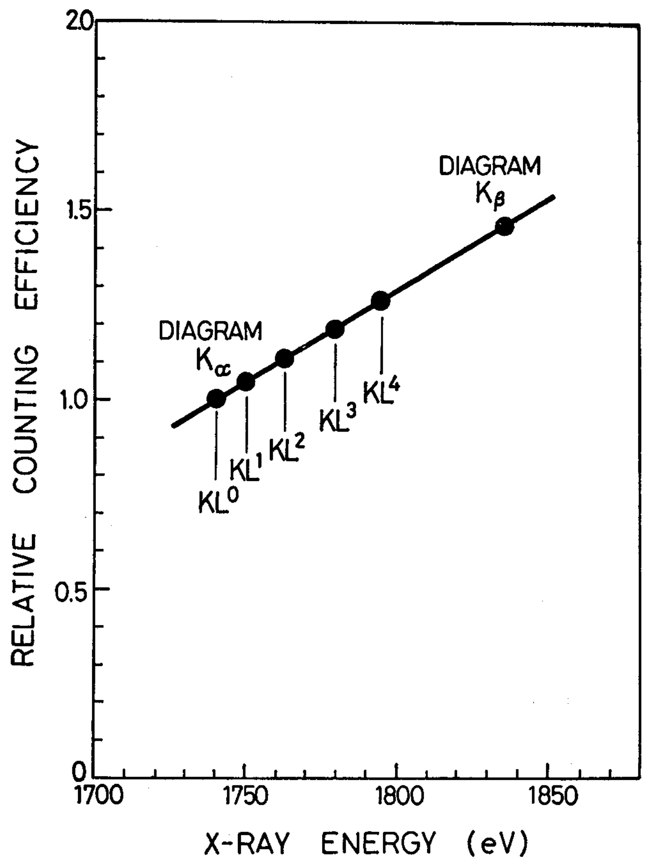

Fig. 3. Relative counting efficiency of the detecting system as a function of the X-ray energy.
The efficiency for the Si-K_α diagram line is taken as unity.

and K_β (KL^0=1.836 keV) diagram lines, the relative correction factors $\eta_m (m=0, 1, 2, 3)$ for the KL^0, KL^1, KL^2 and KL^3 lines can be estimated as shown in Fig. 3, where the factor η_0 for the K_α (KL^0) line is taken as unity. The results are

$$\eta_0=1, \quad \eta_1=1.05, \quad \eta_2=1.11 \text{ and } \eta_3=1.19.$$

3.2. Relative values of X-ray production cross section

The thick target yield I_m for X-ray production has been defined as total X-rays per projectile for a thick target. It is thus given by

$$I_m(E)=Y_m(E)/\eta_m \qquad m=0, 1, 2, 3, \tag{1}$$

where E is the projectile energy. Then the cross section for the X-ray production, denoted as $\sigma_{Xm}(E)$, is derived from the following relationship:

$$\sigma_{Xm}(E)=\frac{1}{N}\left[\frac{dI_m(E)}{dE}S(E)+\mu_m I_m(E)\right] \qquad m=0, 1, 2, 3, \tag{2}$$

where N is the number of target atoms per unit volume, $S(E)$ is the stopping power and μ_m is the self absorption coefficient for the m-th X-rays. The $S(E)$ values for protons and alpha particles are referable to the report of Skyrme[12] and the μ_m to that of Henke.[13] The observed behavior of the thick target yield can be fitted to the following expression:

$$I(E)=a \exp(bE^2+cE), \tag{3}$$

where a, b and c are the coefficients suitable to describe the excitation function. With the aid of eqs. (1) to (3), we derived the relative values for the X-ray production cross section.

3.3. Atomic fluorescence yields for multiply ionized silicon

The atomic fluorescence yields for singly K-ionized atoms, ω_K, have theoretically and experimentally been studied by some workers.[14][15] When an atom is ionized with a K plus mL vacancies, the fluorescence yield ω_{KmL} is expected to increase as the L shell ionicity m increases, because a decrease of L shell electrons reduces the probability of an Auger transition. The ω_{KmL} calculations for some atoms have been done by Larkins[16] and Bhalla *et al.*[17] by applying a Hartree-Fock-Slater method. However, the calculation for silicon has not been reported.

We computed the Si-ω_{KmL} values according to the way similar to Bhalla and others, the results of which are represented in Table I. Figure 4 shows the calculated yields as a function of the L shell ionicity. In the same figure are compared the analytical results of Walters *et al.*,[18] which overestimate our values at higher ionicities. The present calculation seems reasonable if we refer to the values for neon presented by

Table I. Calculated atomic fluorescence yields ω_{K_mL} for multiply ionized silicon.

Vacancy	KL^0	KL^1	KL^2	KL^3
ω_{K_mL}	0.0489	0.0531	0.0562	0.0577

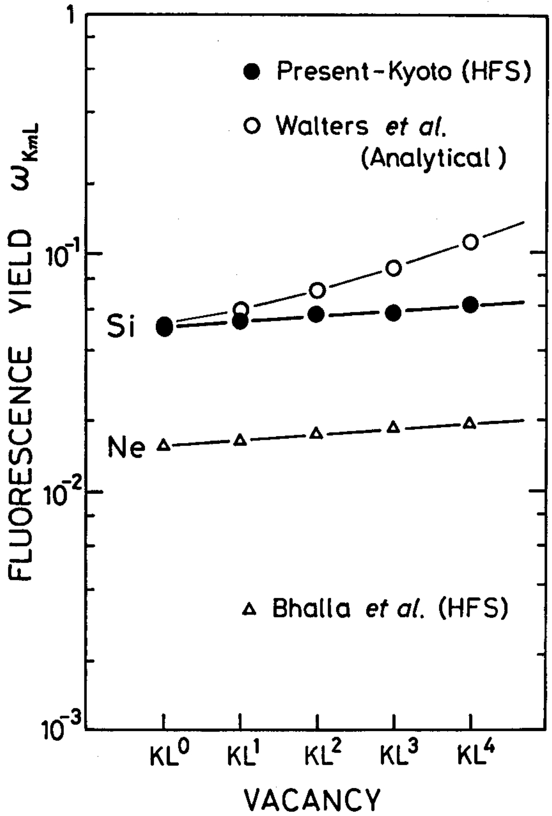

Fig. 4. Calculated atomic fluorescence yields for multiply ionized silicon against multiple vacancy.

Bhalla *et al.*

3.4. Ionization cross sections

By combining $\sigma_{X_m}(E)$ of eq. (2) with ω_{K_mL} described above, the ionization cross section $\sigma_{I_m}(E)$ is given by

$$\sigma_{I_m}(E) = \sigma_{X_m}(E)/\omega_{K_mL} \qquad m=0,\ 1,\ 2,\ 3. \tag{4}$$

Exactly speaking, however, the derived cross sections do not always reflect the initial populations of vacancies. Vacancies initially produced by impact ionization immediately move upward accompanying the L X-ray emission or the L-MM Auger electron emission. This rearrangement effect has been pointed out by Dutkiewicz *et al.*[6] We quantitatively estimated this contribution for multiply ionized silicon according to the procedure similar to them, and found that the necessary corrections for the ionization cross sections stay within 3.5%, which are small enough compared with the experimental uncertainties. The cross section results are tabulated in Table II, where the second and last columns give the scaled relation of $u^2\sigma_I/z^2$ vs $E/\lambda u$ predicted by Garcia.[19]

Table II. Experimental K shell ionization cross sections for silicon bombarded with H_2^+ and He^+ ions and comparison with the BEA theory.

Projectile	Energy E (MeV)	$E/\lambda u$	Cross section σ_I (10^{-20} cm^2)	$u^2\sigma_I/z^2$ (10^{-20}keV$^2\cdot$cm^2)
H_2^+	1.00	0.148	0.23±0.02	0.78
	2.00	0.296	1.4 ±0.10	4.7
	2.50	0.370	1.6 ±0.14	5.4
He^+	1.50	0.111	0.24±0.03	0.20
	2.00	0.148	0.87±0.09	0.74
	2.50	0.185	2.3 ±0.20	1.9

The notaton u is the K shell binding energy and u=1.839 keV, λ is the projectile mass in units of electron mass and z is the projectile atomic number.

In order to obtain the absolute values for the ionization cross section, a Si(Li) X-ray detector was separately used.

3.5. Multiple ionization

According to the binary encounter approximation (BEA) theory proposed by Garcia,[19] the ionization cross section for a single vacancy production is expressed by

$$\sigma_I(E)=\int_0^\infty 2\pi b\ P(V,\ b)\ \mathrm{d}b, \tag{5}$$

where b is the impact parameter, V is the ion velocity and $P(V,\ b)$ is the ionization probability. McGuire and Omidvar[20][21] have precisely calculated the probabilities for K and L shells by applying hydrogen-like wave functions, the forms of $P(V,\ b)$'s resembling gaussian distributions.

McGuire and Richard[7] have regarded the multiple ionization as a statistical superposition of the above single ionization processes. The probability that one of two K shell electrons together with m among L shell electrons are simultaneously ionized from an atom is thus given by

$$P_{K_mL}(b)={}_2C_1P_K(b)[1-P_K(b)]\,{}_8C_mP_L(b)^m[1-P_L(b)]^{8-m} \tag{6}$$

where ${}_iC_j$ are the binomial coefficients and the relation $P_K(b)\ll P_L(b)$ is satisfied in the

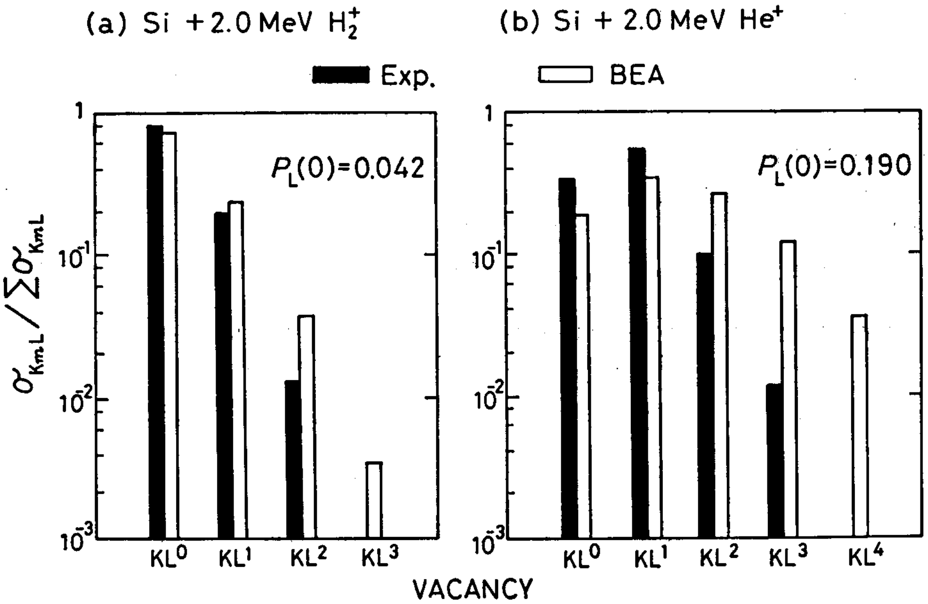

Fig. 5. Experimental and theoretical fractions for multiple ionization cross sections of ion-excited silicon as a function of the multiple vacancy.
(a) by H_2^+ ion impact at 2.0 MeV, (b) by He^+ ion impact at 2.0 MeV.

case of silicon. Since $P_L(b) \approx P_L(0)$ holds within the K shell radius a_K and the integral in eq. (5) is most effective at around $b=a_K$, we have the following approximation:

$$\sigma_{KmL} \approx {}_8C_m P_L(0)^m [1-P_L(0)]^{8-m} \cdot \sigma_K \qquad m=0,\ 1,\ 2,\ \cdots,\ 8. \qquad (7)$$

Expression (7) shows that the multiple ionization spectrum can be reproduced by a binomial distribution.

In Fig. 5(a) and (b), the experimental relative intensities of multiple ionization for silicon bombarded with H_2^+ and He^+ ions at 2 MeV, respectively, are compared with the $\sigma_{KmL}/\sum\sigma_{KmL}$ values calculated from eq. (7). The experimental and theoretical $\sigma_{KmL}/\sigma_{K0L}$ results as a function of the projectile energy are represented in Fig. 6. Both figures reveal that the deviations between experiment and theory increase as the ionicity m becomes larger, and one should rather conclude that only the spectroscopic behaviors resemble each other.

According to the BEA theory, the $P_L(0)$ is uniquely decided when the projectile, its energy and the target atom are given. Then eq. (7) also leads to a unique spectrum for multiple ionization. If a larger $P_L(0)$ value is chosen, the deviations found in Figs. 5 and 6 are reduced. This means that the single ionization probability $P_L(b)$ for silicon predicted by the BEA theory is underestimated, so far as the multiple ionization is concerned. Note that the simple underestimation causes a contradiction with

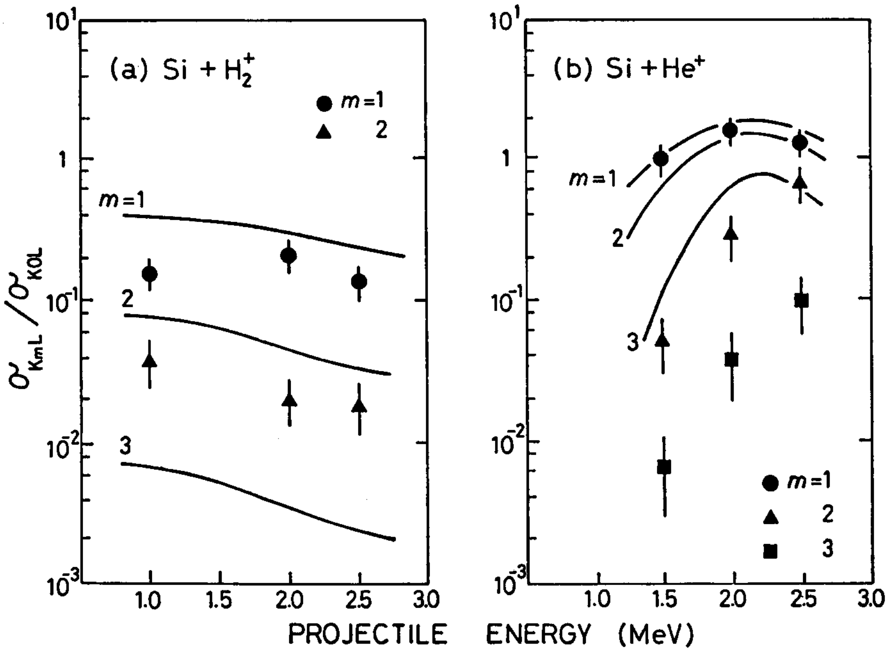

Fig. 6. Observed and calculated ratios of $\sigma_{K_mL}/\sigma_{K0L}$ for ion-bombarded silicon against projectile energy.
(a) by H_2^+ ion incidence, (b) by He^+ ion incidence.

the experimental facts obtained in the single ionization.

Another explanation is possible. Microscopically speaking, the multiple ionization does not always imply a perfect simultaneous phenomenon —— there should be a time-lag between two successive ionizations, but the lag is short enough compared with the travelling time of a projectile in a target atom. Then the probability $P_K(b)$ or $P_L(b)$ should be differently chosen as the ionicity varies, whereby the simple statistical superposition in eqs. (6) and (7) is inappropriate. At the present state of knowledge, however, a quantitative description is quite difficult.

3.6. Concluding remarks

By applying an LSD crystal ($2d=100.3$ Å), we have also measured K X-rays from a graphite target bombarded with hydrogen and helium ions at 2 MeV. There is no clear difference between the observed spectra since both profiles express mono-peaks with the FWHM values of 10-15 eV. The energy of the C-K diagram line (KL^0) is 277 eV. Consequently, the present experiments for silicon and carbon mean that our Bragg spectrometer is applicable for analyzing X-rays of 0.25 to 2 keV in energy, and its automatic mechanism together with the electronic devices work satisfactorily. A better energy resolution would be attained if the second soller slit is improved. An organic semiconductor channeltron placed in co-vacuum with the ion-bombarded target is found to be a useful device for detecting soft X-rays.

In this article are described the procedures in converting the observed satellite X-ray yields to the multiple ionization cross sections, where the correction for the detector sensitivity and the calculation of the atomic fluorescence yields for multiply ionized atom are most important. When the final values are compared with the binomial distribution predicted by the statistical treatment for multiple ionization, considerable deviations are found at higher ionicities.

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References

1) P. Richard, T. I. Bonner, T. Furuta, I. L. Morgan and J. R. Rhodes; Phys. Rev. **A1**, 1044 (1970).
2) A. R. Knudson, D. J. Nagel, P. B. Burkhalter and K. L. Dunning; Phys. Rev. Letters **26**, 1149 (1971).
3) D. Burch, L. Wilets and W. Meyerhof; Phys. Rev. **A9**, 1007 (1974).
4) J. McWherter, D. K. Olsen, H. H. Wolter and C. F. Moore; Phys. Rev. **A10**, 200 (1974).
5) R. L. Kauffman, K. A. Jamison, T. J. Gray and P. Richard; Phys. Rev. Letters **36**, 1074 (1976).
6) V. Dutkiewicz, H. Bakhru and N. Cue; Phys. Rev. **A13**, 306 (1976).
7) J. H. McGuire; Phys. Rev. **A9**, 1374 (1974).
8) H. Hori, N. Maeda, N. Kobayashi and M. Sakisaka; Nucl. Inst. Meth. **130**, 135 (1975).
9) N. Maeda, N. Kobayashi, T. Kusakabe and M. Sakisaka; Bull. Inst. Chem. Res., Kyoto Univ. **57**, 23 (1979).
10) Y. Awaya K. Izumo, T. Hamada and M. Okano; Physics Letters **61A**, 111 (1977).
11) J. H. Scofield; Phys. Rev. **179**, 9 (1969).
12) D. Skyrme; Nucl. Inst. Meth. **57**, 61 (1967).
13) B. C. Henke, R. L. Elgin, R. E. Lent and R. B. Ledingham; Norelco Reporter **14**, 112 (1967).
14) R. W. Fink, R. C. Jopson, H. Mark and C. D. Swift; Rev. Mod. Phys. **38**, 513 (1966), W. Banbynek, 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).
15) E. J. McGuire; Phys. Rev. **A2**, 273 (1970), ibid. **A3**, 716 (1971).
16) E. P. Larkins; J. Phys. B, Atom. Molec. Phys. **4**, 1 (1971).
17) C. P. Bhalla, N. O. Folland and M. Hein; Phys. Rev. **A8**, 649 (1973).

18) D. L. Walters and C. P. Bhalla; Phys. Rev. **A3**, 1919 (1971).
19) J. D. Garcia; Phys. Rev. **A1**, 280 (1970).
20) J. H. McGuire; Phys. Rev. **A9**, 286 (1974).
21) J. H. McGuire and K. Omidvar; Phys. Rev. **A10**, 182 (1974).