OBSERVATION OF TWO-ELECTRON-ONE-PHOTON TRANSITIONS IN SILICON*

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We report on the observation of Kαα X-rays of Si, produced in collisions of 15–28 MeV Si projectiles with various target atoms in the range $Z = 6$ to 29. Energy shifts of X-rays were measured and are compared with theoretical predictions. Cross section ratios for emission of Kαα and Kα radiation are given.

Since the first experimental identification of cooperative two-electron transitions in the K shell of doubly ionized atoms leading to the emission of a single photon, published recently in a paper by Wölfli et al. [1], increasing interest has been devoted to the study of these phenomena. Further experimental work [2–5] has confirmed the existence of these transitions in several atoms. Measured Kαα X-ray energies are found to be in rather good agreement with theoretical calculations [6–9], whereas relative transition rates are still subject of some theoretical controversy [9–11]. A convenient method for study of two-electron-one-photon transitions is the observation of X-rays induced in collisions between heavy ions and atoms because of the relatively large probability for the production of two K shell vacancies in such reactions [12]. We have investigated two-electron–one-photon transitions into K shell of silicon, measuring energies of Kα and Kαα X-rays and production cross sections for various target atomic numbers and several incident silicon projectile energies and present comparison with calculated X-ray energies.

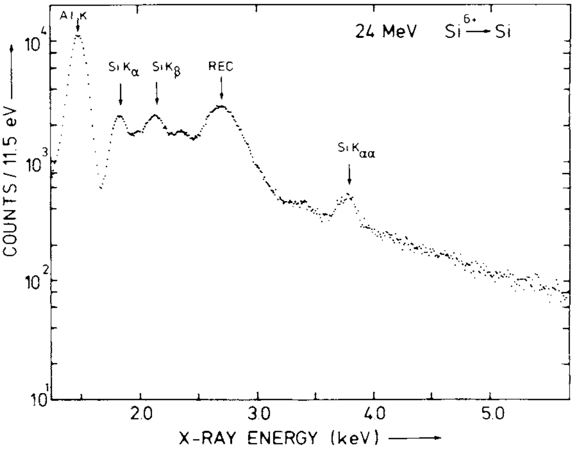

Fig. 1. Observed X-ray spectrum from 24 MeV Si^{6+} bombardment of a 15 $\mu g/cm^2$ Si target. The spectrum is not corrected for absorption, which is mainly due to a 10 μm Al foil. Fluorescence excited absorber X-rays, characteristic Kα and Kβ radiation of Si and X-rays from radiative electron capture into projectile K-shell are clearly visible. The single peak at 3.75 keV is attributed to the two-electron–one-photon transition into the Si K-shell. It is imposed on an almost exponentially decreasing background of non-characteristic radiation.

Experiments were performed with the ^{28}Si beam from the 4 MV Dynamitron-Tandem at Bochum. Thin targets (effective thickness of 20–60 $\mu g/cm^2$) either self-supporting foils (C, Al) or evaporated onto 20 $\mu g/cm^2$ carbon backings (Si, KBr, Ti, Fe, Cu) were bombarded with 15–28 MeV ^{28}Si ions. X-rays emitted from projectiles and target atoms were observed at 90° with respect to the beam line through a 6 μm Mylar window separating the vacuum chamber from a 3 mm × 10 mm^2 Si(Li) detector with an energy resolution of 158 eV at 5.9 keV. For measurement of Kαα intensities, low energy X-rays (mainly Si Kα) were strongly

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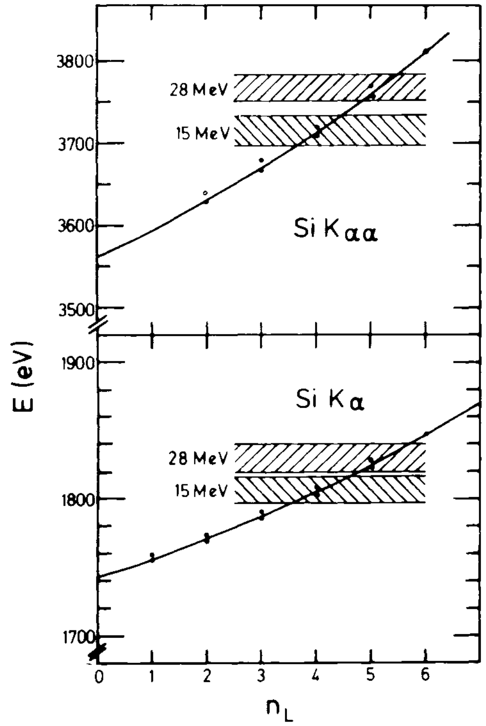

Fig. 2. Comparison of measured Kα and Kαα energies of silicon with calculations using a Dirac–Fock–Slater program [14]. The shaded bands represent experimental X-ray energies for two projectile energies. Calculations were performed for different L-shell defect configurations. Theoretical energies are plotted versus n_L, the number of L shell vacancies accompanying the K-shell vacancies and are connected by solid lines. Different points at the same n_L represent different combinations of subshell holes. Additional M-shell vacancies change the calculated X-ray energies by less than the experimental uncertainties.

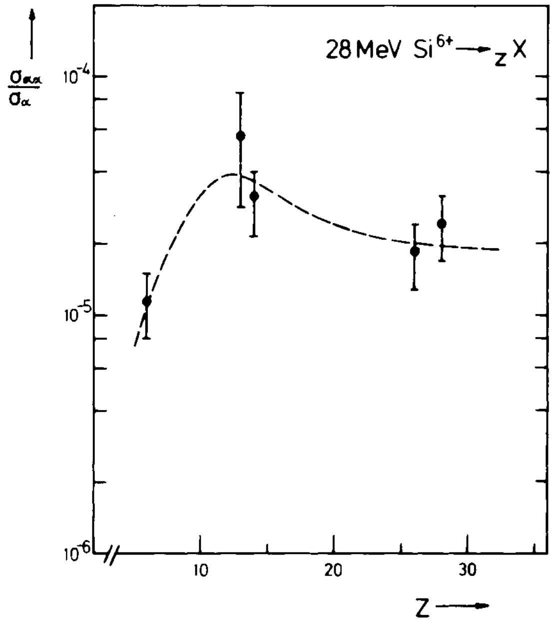

Fig. 3. Ratio of cross sections for production of Si Kαα and Kα X-rays as function of atomic number Z of target atoms. The broken curve is arbitrarily drawn through data points to indicate the gross behaviour of the ratio with Z. Similar dependence is found for the other projectile energies.

suppressed by absorber foils to avoid pile-up effect in the vicinity of the Kαα.line. Projectile and target Kα and Kβ X-ray cross sections were measured separately without absorber foils to reduce necessary correction factors in cross section evaluation. Normalization to incoming particle flux and target thickness was achieved with Rutherford scattered projectiles observed at 30° scattering angle by a surface barrier detector. In fig. 1 is shown an uncorrected X-ray spectrum from the symmetric collision system Si + Si at 24 MeV bombarding energy. The Si Kα and Kβ peaks are well separated due to their large energy shifts of 80 eV and 270 eV respectively. Radiative electron capture [13] is producing the broad peak centred near 2.7 keV. The peak at 3.75 keV is ascribed to the two-electron–one-photon transition in Si atoms with two K-shell vacancies. Its identification is based on the following experimental observations: (i) the peak is observed in bombardment of all targets, except KBr and Ti where characteristic X-rays of target atoms obscure its identification; (ii) pile-up effect could be excluded by measurements with different absorber thicknesses; (iii) the peak is clearly shifted from twice the measured Si Kα energy by a positive amount of 103 ± 10 eV; (iv) the Al target was subsequently checked for the presence of impurities by the method of proton induced X-ray analysis giving an upper limit for the concentration of trace elements with characteristic X-rays in the interesting region of at least two orders of magntidue too low to explain the peak found in the measurements with Si projectiles. The experimental observations are in full agreement with the assumption that the peak is due to the two-electron–one-photon transition.

Energies of Kα, Kβ and Kαα are influenced by vacancies in outer shells [5–9]. Calculations of these energies were performed with a Dirac–Fock–Slater [14] and a Dirac–Fock [15] program for various outer shell vacancy configurations assuming the K$\alpha\alpha$ line as being due to a $1s^{-2} \rightarrow 2s^{-1} 2p^{-1}$ electric dipole transition [2, 7–9]. The two different computation methods lead within less than 7 eV to the same transition energies. In fig. 2 are shown the results for E_α and $E_{\alpha\alpha}$ from calculations as function of n_L, the number of additional L shell vacancies accompanying the 1s vacancies. The experimental values of Kα and K$\alpha\alpha$ energies are in good agreement with $n_L = 5 \pm 1$ at 28 MeV and $n_L = 4 \pm 1$ at 15 MeV. The same n_L are also obtained from calculated and measured Kβ energy shifts. Furthermore the mean charge state of Si ions after penetration of a solid foil, $\bar{q} = 10$ at 28 MeV and $\bar{q} = 8.5$ at 15 MeV [16] is consistent with the obtained n_L if the Si M Shell is assumed to be almost completely stripped. The experimental K$\alpha\alpha$ energy increases by about 60 eV with bombarding energy varying between 15 and 28 MeV, however, the difference $E_{\alpha\alpha}(\exp) - 2E_\alpha(\exp)$ is found to be constant within the experimental uncertainty of ±10 eV, reflecting the similar influence of outer shell vacancies on E_α and $E_{\alpha\alpha}$ [7]. The width of the K$\alpha\alpha$ peak shows no additional broadening as was observed fro neighbouring Kα and Kβ radiation of K and Ti induced by collisions with Si ions. The observed K$\alpha\alpha$ width is completely due to detector energy resolution.

Because all measurements were performed with thin targets, cross sections could easily be deduced from X-ray yields, the known detector efficiency including absorption corrections and solid angles, and the yield of Rutherford-scattered projectiles. Fig. 3 shows cross section ratios for production of K$\alpha\alpha$ and Kα X-rays plotted versus the atomic number Z of the target. The ratio has its maximum of 4×10^{-5} for symmetric collisions and decreases with increasing deviation from symmetry, a behaviour that is to be expected if vacancy production via molecular orbital mechanisms [17] is dominating. Similar observations were reported for other collision systems [3]. The cross section ratio for all investigated collision systems is found to be by a factor of 2 smaller at 15 MeV bombarding energy, whereas the Si K$\alpha\alpha$ cross section itself decreases by one order of magnitude from 28 to 15 MeV. Observation of K$\alpha\alpha$ X-rays in experiments with thin targets offers an excellent method to investigate the mechanism of multiple vacancy creation in inner atomic shells. Further experiments to study in more detail projectile energy and target atomic number dependence of K$\alpha\alpha$ cross sections are now in progress.

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