

Chemical effects on Al $K\alpha$ x-ray satellite structure*

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Using a 38-MeV F beam for vacancy production, we find minimal dependence of the Al $K\alpha$ satellite structure on the total valence electron densities of several different solid environments.

A recent paper¹ reported significant variation of the relative intensities of S $K\alpha$ x-ray satellites produced from bombardment of various solid targets by 2.0-MeV/amu oxygen and neon. The degree of L -shell ionization present at the time of the K transitions was correlated with the total valence electron density of the solid material used as a target, a result which was taken to suggest the importance of interatomic transitions in the outer-shell relaxation occurring in a highly stripped ion to the filling of a K vacancy. We have searched for such an effect in the Al $K\alpha$ satellite structure as reflected in the x-ray emission from various Al compounds and Al metal, here induced by a 38-MeV F beam, and have observed only a slight dependence over electron densities from ~ 0.2 to $\sim 0.56 \text{ \AA}^{-3}$.

The experiment was performed using a F beam obtained from the Stony Brook FN tandem Van de Graaff accelerator. The Al $K\alpha$ x rays emitted from targets of Al, Al_2S_3 , AlN, and Al_2O_3 were detected with a vacuum Bragg curved-crystal spectrometer equipped with an ammonium dihydrogen phosphate crystal. The detecting element was a flow-mode proportional counter operated at 1 atm of P -10 (10% methane-90% argon) and biased to 2000 V. The beam was collimated to 2 mm approximately 9 cm upstream from the targets, which were situated at 45° to the beam axis. The linear signal from the proportional counter was processed through a single-channel analyzer, with a window set on the Al K x ray. Data were accumulated for fixed periods of charge integration and the crystal was stepped automatically by an electronic controller. The resulting multichannel spectrum was stored via a multiscaling analyzer and transferred to magnetic tape for subsequent analysis.

A set of thick targets was assembled in the following ways. An Al slab served as the metal target. The Al_2S_3 and AlN targets were constructed by grinding grains of $\sim 95\%$ and $\sim 97\%$ purities, respectively, into fine powders and compressing them into ~ 1 -mm-thick disks. The Al_2O_3 target consisted of a tantalum backing coated with an oxide layer. The sulfide and nitride disks were

placed in vacuum immediately after fabrication. In order to avoid target deterioration, beam currents were limited to ~ 3 nA with scans of 1 h duration or less. Repeatability of the spectra was excellent. In addition, thin target spectra were taken using a $40\text{-}\mu\text{g}/\text{cm}^2$ Al foil backed by a $20\text{-}\mu\text{g}/\text{cm}^2$ C foil and a $200\text{-}\mu\text{g}/\text{cm}^2$ self-supporting Al_2O_3 foil. Elastic scattering with 2.0-MeV protons revealed negligible contamination of the Al foil by any oxides, e.g., Al_2O_3 .

Thin and thick target spectra are shown in Figs. 1 and 2, respectively. The $K\alpha$ region of about 1490–1550 eV is seen in each case to consist of a group of six distinct peaks corresponding to increasing degrees of L -shell ionization, starting with the diagram or KL^0 line at 1486 eV. The higher energy peaks are denoted by KL^n , i.e., KL^1 , KL^2 , etc., where n indicates initial configurations with a single K vacancy and n L vacancies. The areas obtained from least-squares fitting of the spectra with gaussians superimposed on a linear

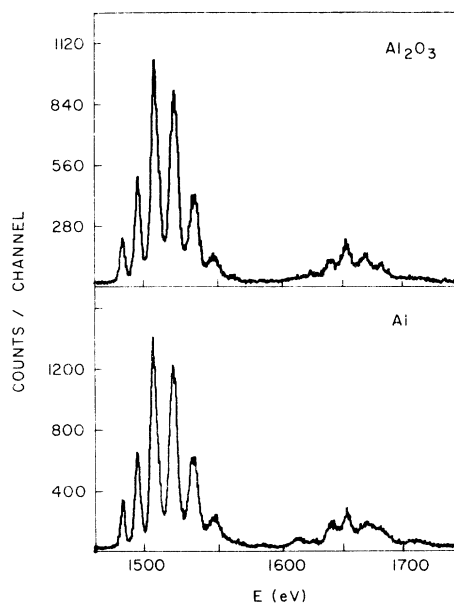


FIG. 1. Thin-target Al_2O_3 and Al K x-ray spectra induced by 38-MeV F.

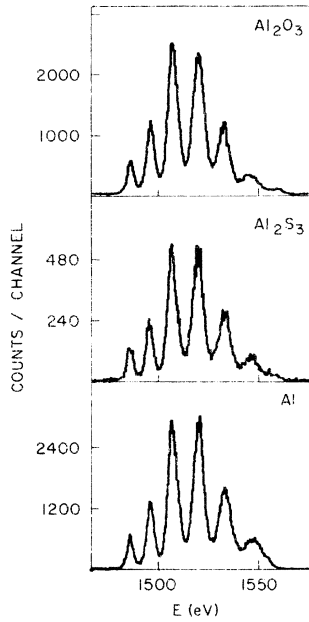


FIG. 2. Thick-target Al, Al_2S_3 , and Al_2O_3 $K\alpha$ x-ray satellite structure.

background are presented in Table I in fractional form. The thin-target spectra exhibit nearly identical $K\alpha$ structure, as do the thick Al and Al_2O_3 spectra. This result is in agreement with previous satellite data² for those two materials induced by 5.0-MeV Ne ions, a regime in which recoil effects might possibly play a role in determining the L vacancy distributions. Such effects are less important in the present situation where a recoil energy of ~ 40 eV, corresponding to an impact parameter about equal to the Al K -shell radius, leads to a displacement of less than 0.4 \AA during a single K vacancy lifetime³ of 2.2×10^{-15} sec.

There is a slight shift downward in the relative intensities of the satellites from the thin targets as compared to the corresponding values for an infinitely thick target of the same material. How-

ever, as has been noted before^{1,4} there is remarkably little difference between them, in spite of the great variance in velocities arising from energy loss by the projectile in the process of slowing down and stopping. Apparently the majority of the target K vacancy production takes place before much energy is lost and the L ionization does not change significantly over that range. The Al_2S_3 and AlN intensities are seen in Table I to lie very close to those for Al.

In the current analysis, we have taken the fluorescence yields ω_n of the satellites to be equal, thereby equating the x-ray distribution with the L vacancy distribution. This procedure is not exact, as calculations⁴ for the Al satellites for ions with filled M shells indicate increasing values for the higher satellites, e.g., $\omega_4 = 1.5\omega_0$ and $\omega_5 = 2.0\omega_0$. However for our purposes the x-ray distribution is a sufficiently good approximation for the vacancy distribution, as our main interest is in changes in the relative intensities with changes in environment. A more serious question is the possibility of changes in the fluorescence yield for a given satellite from one material to another due to the dependence of KLM and KMM Auger rates on the M shell occupancy. The decrease in the total K Auger rate due to a fully vacant M shell would be about 30% for the KL^5 satellite and much less for the lower lines.⁴ The energy centroids of the satellite peaks in our spectra, including the KL^5 group, were found to within ± 0.5 eV of a mean value regardless of material, implying very similar M occupation. The effect on the fluorescence yields due to the specific nature of the bonding in the solids is a topic which constitutes an unresolvable uncertainty in the present experiment. However we note that the trends to be discussed shortly are evident even when KL^4 and KL^5 are excluded from the analysis, thereby lessening the importance of chemical effects insofar as fluorescence yields are concerned.

Following the multiple ionization formulation^{1,5} used in many previous high-resolution analyses, we have extracted an average L -shell ionization probability by fitting the six prominent peaks through KL^5 simultaneously with the binomial expression

$$R_{nL} = C \binom{8}{n} P_L^n (1 - P_L)^{8-n},$$

where R_{nL} is the fractional intensity of the n th satellite and P_L is the probability. We show the P_L values for the various targets in Fig. 3 as a function of the total valence electron density of the material. Over a change in density from 0.18 to 0.56 \AA^{-3} , the probabilities are nearly the same

TABLE I. Relative $K\alpha$ satellite intensities.

	KL^0	KL^1	KL^2	KL^3	KL^4	KL^5
Thin						
Al	0.046	0.116	0.285	0.291	0.185	0.077
Al_2O_3	0.045	0.120	0.295	0.304	0.172	0.063
Thick						
Al	0.040	0.098	0.276	0.301	0.193	0.091
Al_2S_3	0.053	0.099	0.276	0.306	0.184	0.081
AlN	0.044	0.101	0.284	0.310	0.183	0.077
Al_2O_3	0.045	0.112	0.301	0.318	0.163	0.059

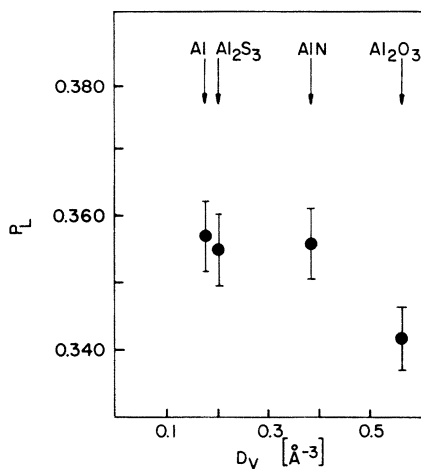


FIG. 3. Extracted L -shell ionization probabilities plotted as a function of total valence electron density.

with only a small dropoff at Al_2O_3 . The error bars represent ranges of P_L which yielded about the same accuracy of fit. The thin-target values, not shown, were even closer in agreement. The previous suggestion¹ that the mobility of the conduction electrons in Al may compensate for the larger density of Al_2O_3 and lead to equivalent spectra is not supported by the present results. The salt Al_2S_3 with about the same density as Al yielded about the same P_L . The lack of change from ~ 0.2

to ~ 0.4 is in contrast to changes of $\sim 10\%$ in the probabilities for the $S K\alpha$ satellites over similar densities. It also departs somewhat from the behavior reported⁶ for the Al $K\alpha$ structure in Al, Al_4C_3 , AlN, and Al_2O_3 induced by 32.4-MeV O, for which Al yielded a noticeably higher ($\sim 5\%$) value of P_L than AlN and Al_2O_3 yielded about the same value as AlN.

Estimates of both the initial configurations created in the collisions and the K and L rates are needed to assess the significance of the data in Fig. 3. An experiment involving gas-solid comparisons of Si $K\alpha$ satellites produced by 30-MeV O bombardment⁷ would indicate that the Al L rearrangement in the present situation is probably shifting the distribution by an average of about one L vacancy. The ratio of single vacancy L rate to single vacancy K rate³ is $\sim (2-3)\%$ depending on weighting of the $2s$ and $2p$ values, compared with a S ratio of $\sim 14\%$. Of course the L relaxation should compete more favorably in a highly stripped ion.^{7,8} However, the fact that Al has a substantially lower single vacancy ratio than S makes it seem reasonable that the density effect may be less. The agreement in the Al_2O_3 comparisons between the low-energy Ne and the present high-energy F experiments is not matched by similar analyses for Si/ SiO_2 , where spectra induced by MeV/amu ions show noticeable differences⁶ and low-energy Ne-induced spectra do not.⁹

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