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Investigation of the Noncharacteristic X-Radiation Band Observed in Ar Bombardment of Various Targets

G. Bissinger*

Rutgers University, New Brunswick, New Jersey 08903

L. C. Feldman

Bell Laboratories, Murray Hill, New Jersey 07974

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The noncharacteristic X-ray band produced by Ar bombardment of various targets has been investigated with a high-transmission window, gas-flow proportional counter, and Si(Li) detectors. We find (i) that the Ar bombardment of Si produces a continuum with monotonically decreasing intensity for increasing energy, extending from $\lesssim 250$ eV to ~ 1.2 keV; (ii) Ar-target interactions and Ar-Ar interactions produce distinctly different spectra; (iii) the band observed in the Ar-C system is inconsistent with a simple quasimolecular picture. The bearing of these results on the quasimolecular description is discussed.

A recent Letter¹ by Saris, van der Weg, Tawara, and Laubert described the observation of a broad band of noncharacteristic x rays at ~ 1 keV produced by low-energy (< 600 keV) Ar bombardment of C, Al, Si, and Fe. This band was explained as being due to a radiative transition during the time of the collision in the quasimolecule formed in an Ar-Ar collision (projectile-implanted Ar), with the projectile carrying an *L*-shell vacancy from a previous interaction. This band was also reported² by Macdonald and Brown who, in addition, bombarded a solid Ar target with 200-keV Ar beams and saw an even broader band whose centroid was at higher energy than those observed with C, Al, and Si targets. These latter authors also observed the band for low doses of Ar, leading them to suggest that Ar-Ar collisions were not the sole source of the band observed with the C, Al, and Si targets; rather, the band was due to Ar-target interactions, with Ar-Ar collisions becoming important only for high doses of Ar.

We report here the results of a further investigation of this band, using higher efficiency Si(Li) detectors and a thin-window proportional counter. The former extends our sensitivity to this band to lower doses of Ar. The latter extends our detection ability to x rays of energies ~ 250 eV; well

below the effective lower-energy cutoff of the Si(Li) detectors used in this and in previous^{1,2} works. In addition, thin targets were used to allow a distinction between Ar-Ar collisions involving implanted argon and Ar-target interactions. Such an experiment permitted a definitive test of the suggestion in Ref. 2.

EXPERIMENTAL

Proton, He⁺, and Ar⁺ beams were provided by two accelerators at Bell Laboratories covering the range from 50 keV to 2 MeV. Both accelerators used a common target chamber which was equipped with an electron suppressor and a cold trap. The Si(Li) detectors (30 mm² × 3 mm and 80 mm² × 3 mm) had 7.6- μ - and 12.7- μ -thick Be windows and were placed perpendicular to the beam, facing directly toward the target at a distance of 5 cm. Resolution of both detectors was 180–200 eV under actual experimental conditions for a 5.9-keV line. The experimental setup is shown in Fig. 1.

The thin-window gas-flow proportional counter³ was positioned on the opposite side of the beam at a distance of about 35 cm from the target. The detector utilized a ~ 3550 - \AA Parylene-C window,⁴

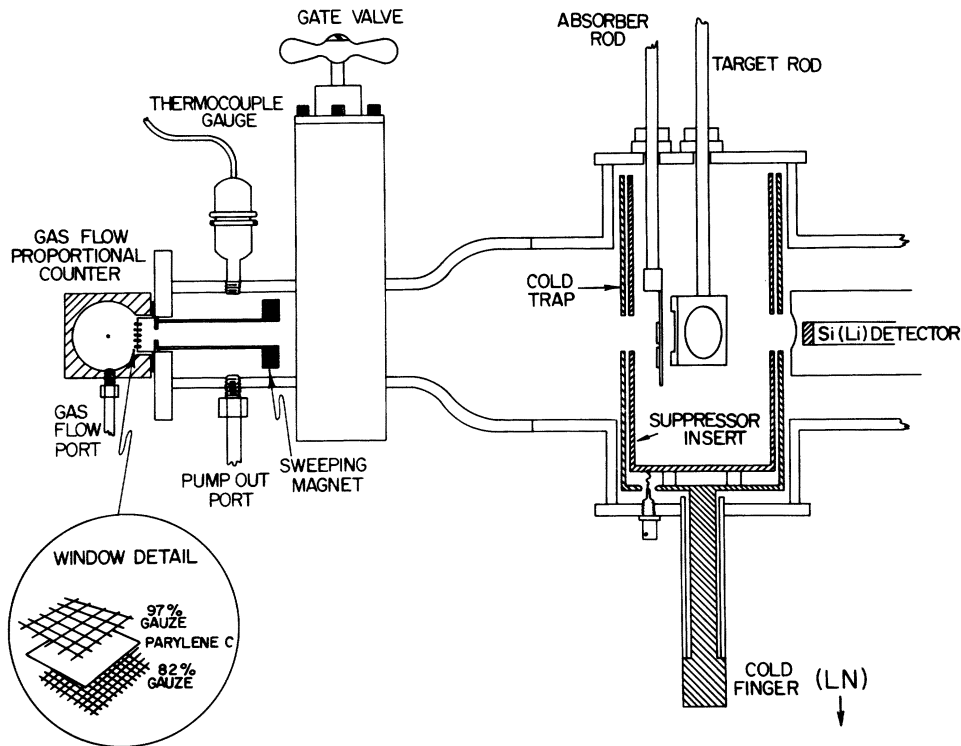


FIG. 1. Experimental setup for x-ray measurements showing proportional and Si(Li) detectors faced onto common target chamber.

sandwiched between 82 and 97% transmission gauzes, with transmission varying from 7.5 to 75% between 100 eV and 1.2 keV. It was possible to interpose absorbers between the target and proportional counter.

The proportional counter was calibrated by exposing targets of C, Al, NaCl, Si, and SiO to 1-MeV proton beams, and then inserting either a 12.7- μ Be or a 25.4- μ polyethylene foil between the target and detector to observe absorber effects. Typical proportional-counter spectra obtained with proton beams on C + Al and SiO targets are shown in Fig. 2. The effective low-energy cutoff is a result of adjusting the detector parameters to achieve a linear energy response from ~ 200 eV to ~ 2 keV. Small gain shifts of $\sim 5\%$ were observed in the spectra—due primarily to gas-pressure variations in the proportional counter. Count-rate-associated gain shifts were minimized by keeping count rates < 500 cps.

The momentum-analyzed beams were focused through a collimator 2×2 mm, 17 cm in front of the target. The current was collected directly from the target holder in the thick-target measurements, and in a Faraday cup 40 cm long, downstream from the target, in the thin-target measurements. The target was placed at a 45° angle to the beam, facing the detector to be used.

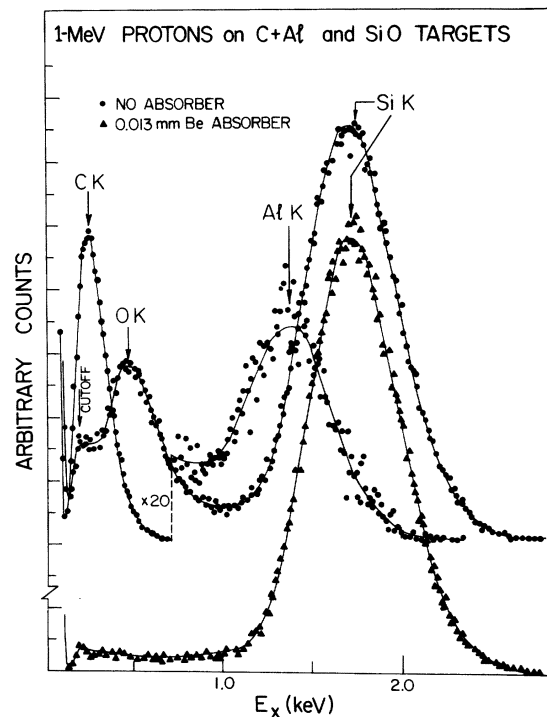


FIG. 2. Proportional-counter spectra obtained for 1-MeV protons on C, Al, and SiO targets. Also shown is a spectrum taken with a SiO target and a 12.7- μ Be absorber between the detector and target.

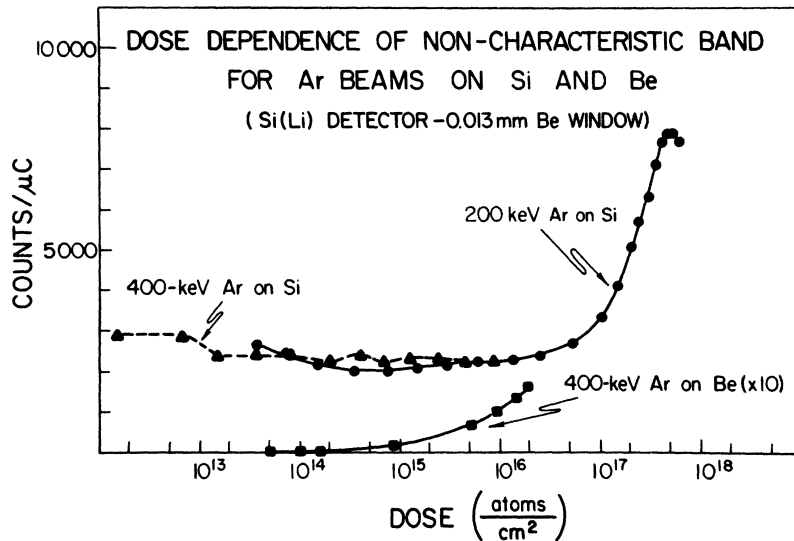


FIG. 3. Dose dependence of band for Ar on Si and Be targets.

The targets were first checked for trace impurities with proton and He beams.

RESULTS AND DISCUSSION

A. Si(Li) Detector Measurements

Investigation with the Si(Li) detector of the band observed in Refs. 1 and 2 gives results in good agreement with those previously reported. The 7.6–12.7- μ Be windows on the Si(Li) detectors used in this experiment permit a considerable improvement in detection efficiency for ~ 1 -keV radiation over those used previously. This has permitted us to extend the Ar dose dependence measurements to doses of $\sim 3 \times 10^{12}$ atoms/cm². In Fig. 3 are presented the dose dependence measurements of the x-ray yield below the Si K line for 200- and 400-keV Ar on Si and 400-keV Ar on Be. The factor-of-4 increase in the ~ 1 -keV band for Ar on Si from low dose to high doses ($\sim 5 \times 10^{17}$ atoms/cm²) is in good agreement with prior results.¹ For doses as low as $\sim 10^{12}$ /cm² the band is observed. This is the case for Ar bombardment of C and Al as well. Bombardment of Be with Ar beams, however, shows no evidence of this band initially. The saturation dose of $\sim 5 \times 10^{17}$ atoms/cm² observed here, combined with the known straggling⁵ of 500 Å for 200-keV Ar on Si, produces Ar densities of $\sim 10^{23}$ atoms/cm³ in the implantation region (assuming the Ar ions do not diffuse from their implant site), which is higher than the Si-atom density, providing a ready explanation for the observed saturation effect.

Further evidence on the source of the band is provided by comparing thick- and thin-target measurements. In Fig. 4 (a) the spectra obtained for 400-keV Ar on thick (low-dosage) and thin

(40- μ g/cm²) Al targets are shown. The target thickness is considerably less than the range of 400-keV Ar in Al; hence the probability of Ar-Ar collisions (with implanted Ar) is negligible. There is no significant difference between the spectra. Similar results (not shown) were obtained using thick and thin (20- μ g/cm²) C targets. The band was observed for Ar beam energies of 300–1800 keV with these thin targets.

The evidence presented here indicates that Ar-target interactions are important contributions to the intensity of the band. This is further demonstrated quite clearly in Fig. 4 (b), which superimposes spectra obtained for Ar on Si at low ($\sim 10^{15}$ -atoms/cm²) and high ($\sim 5 \times 10^{17}$ -atoms/cm²) doses. The band obtained at low doses which must arise from Ar-Si interactions is not only less intense, but also narrower (~ 360 eV) and lower in energy (centroid at ~ 950 eV) than the high-dose band (~ 480 -eV width and 1030-eV centroid) which also includes Ar-Ar interactions. This follows the trend observed in Ref. 2.

Difficulties arise in the interpretation of the Ar-target band as being due to radiative transitions in the transient-projectile-target system. If this were correct, the high-energy cutoff for this band is just that obtained for L x rays from the "combined" Ar + Si(=Ge) system with a 2s binding energy of 1.41 keV. This is consistent with our observations. On the other hand, Ar + C(=Cr) has a 2s binding energy of 0.7 keV, which is not at all in agreement with the "band" observed. Clearly this quasimolecular mechanism is not the *direct* source of the band observed in the Ar + C system. In this case the observed x rays could arise from a carbon-carbon recoil mechanism which would present a cutoff of ~ 1.3 keV (Mg K) consistent with our data.⁸ A band due to a similar mechanism

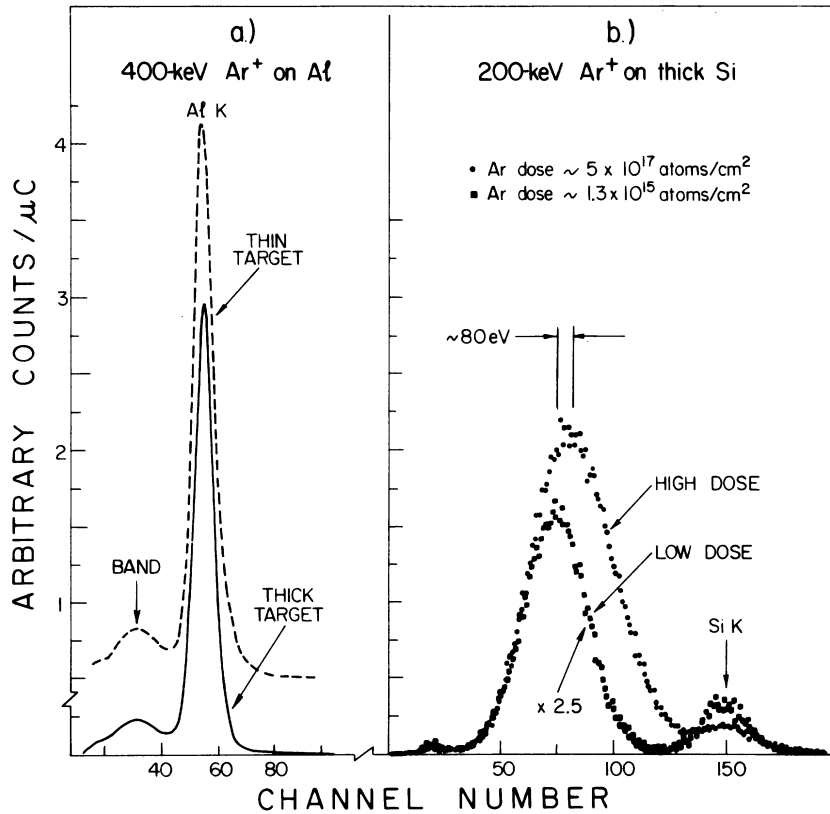


FIG. 4. Spectra obtained with Si(Li) detector: (a) 0.4-MeV Ar on thick and thin Al targets; (b) Ar dosage effect on band.

then would not be expected to be observed in the Ar + Be experiment for one of two reasons: (a) the Be + Be recoil system producing oxygen K x rays cannot be observed with our Si(Li) detection system, or (b) the Ar L vacancy, which may be a necessary initial condition for the x-ray

production, is not produced. This is a result of the fact that the 1s electrons of Be have a smaller binding energy than the 2s and 2p levels of Ar (see Fig. 5 for the Ar-Be and Ar-C correlation diagrams) and thus provide no mechanism for producing L holes in the Ar projectile by the elec-

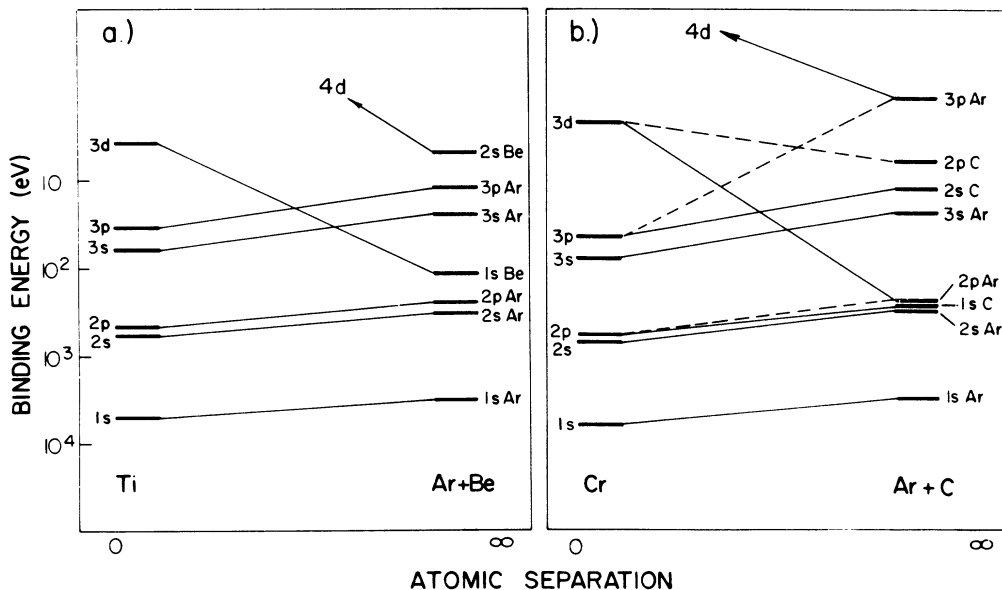


FIG. 5. Correlation diagrams for Ar on Be and C.

tron-promotion scheme (direct Coulomb excitation is expected to be unimportant at these energies).⁷

B. Proportional-Counter Measurements

In an attempt to better understand the entire x-ray spectrum associated with these phenomena we have examined the band using a high-transmission-window proportional counter, since it is clear that the Si(Li) detector Be window is distorting the observed spectra, particularly on the low-energy side. The proportional-counter spectra obtained for 400- and 1000-keV Ar beams on thick Si (low dose) are shown in Fig. 6. These spectra show no evidence of any structure near 1 keV, at either beam energy. Instead they show a broad distribution, monotonically decreasing with increasing x-ray energy. The proportional-counter-window transmission properties, which are relatively constant from 500 eV to 1.3 keV (45–75%, see insert on Fig. 6) indicate that the ~1-keV band observed with Si(Li) detectors is an artifact of these detectors. To demonstrate this more clearly, a 0.5-mil Be foil was interposed between the target and proportional counter. If one keeps in mind the fact that the resolutions are quite different, the spectra obtained from the two detectors are quite similar.

The question then arises whether the broad distribution observed with the proportional counter is indeed that which would be expected for radiative transitions during the Ar-target or Ar-Ar collisions, since the radiated energy will depend on the internuclear separation at the time of the decay. Saris *et al.*¹ advanced arguments concerning this band and noted that not only is the vacancy lifetime much shorter (relative to the collision time) in the united atom (approximately a factor of 10) but also that the fluorescence yields in the united atom are considerably higher ($\sim 10^2$ higher than in Ar). The combined effect might then be sufficient to produce an enhanced intensity at the high-energy end of the band if the essentially geometric reduction in cross section for close collisions does not offset it. The proportional-counter data in Fig. 6 show no evidence for an enhancement strong enough to yield a peak at the high-energy end of the x-ray spectrum.

In conclusion we note the following facts concerning the production of this interesting phenomenon: (i) The x-ray spectrum produced by Ar

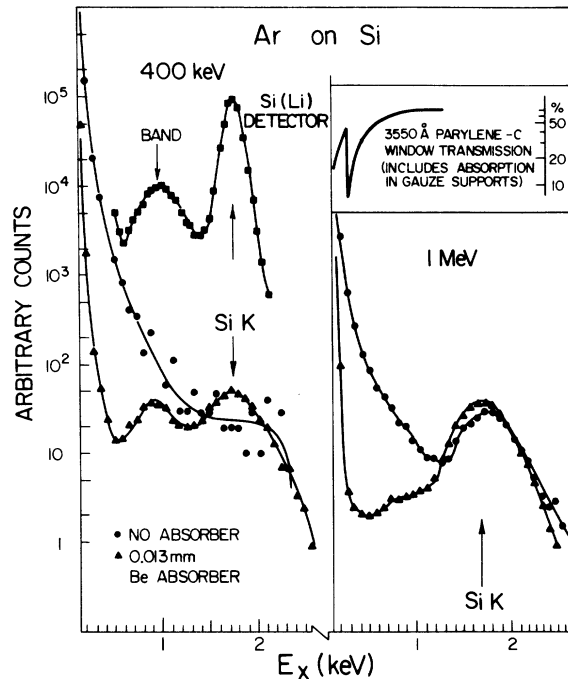


FIG. 6. Proportional-counter spectra obtained with 400-keV and 1.0-MeV Ar on Si. Insert shows window transmission over the 100-eV to 1.2-keV photon energy range. Also shown is a Si(Li) detector spectrum for 400-keV Ar.

bombardment of Si is a continuum with monotonically decreasing intensity for increasing energy, extending from very low energy (≈ 250 eV) to approximately 1.2 keV. (ii) There is a broad spectral distribution associated with Ar-target interactions alone. There is a different broad distribution associated with Ar-Ar interactions. (iii) The band observed in the Ar-C system is not consistent with a simple quasimolecular picture. A recoil mechanism, however, could explain the spectrum. These results indicate that a variety of phenomena are occurring which must be carefully distinguished before a quantitative interpretation can be brought forth.

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