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SUMMARY

The continuing improvement in energy resolution of semiconductor detector X-ray spectrometers has led to interest in the use of these devices at energies less than 2 keV.[1,2] This is an energy region of potential analytical interest since the K X rays of several elements of biological and chemical importance occur at these energies. However, the low X-ray fluorescence yield in low-Z elements, combined with the absorption of low energy X-rays due to the windows of both the detector and the vacuum enclosure, have made work in this energy region impossible with the standard semiconductor detector X-ray spectrometers. In the present work, the X-ray absorption path has been reduced to a minimum to permit low-energy X ray studies of excitation, entry window thickness, detector linearity and resolution. Using electron beam excitation on low-Z targets, we have performed measurements of characteristic K X rays of elements down to and including carbon (277 eV).

EXPERIMENTAL APPARATUS

The modified X-ray spectrometer system used in the measurements is shown schematically in Fig. 1. It consists of an experimental chamber with access ports for mechanical motion feedthroughs, electron-gun electrodes, and vacuum pumpout. A standard "dip-stick" type semiconductor detector spectrometer cryostat projects into the chamber through a vacuum seal as indicated. A two position gate valve is mounted in place of the usual window on top of the detector holder and is actuated by means of a mechanical feedthrough to the outside of the sample chamber. In one position of the gate valve, a 0.005 inch beryllium window isolates the detector from the sample chamber, enabling operation of the chamber at atmospheric pressure without disturbing the detector cryostat. In the other position, a thin self supporting aluminum foil typically of 25 $\mu gm/cm^2$ to 60 $\mu gm/cm^2$ thickness is present. The reasons for this choice of window are twofold:

a) It prevents light from the sample chamber from reaching the detector and generating leakage current. Light is produced both by the filament of the electron-gun and, in some cases, by luminescence in samples exposed to electron or X-ray bombardment. It is difficult to obtain thin foils completely free of pinholes, so it is desirable to limit the amount of light incident on the window by using appropriate baffles and by operating the tungsten filament at low power levels when possible.

b) The foil also serves as a vacuum baffle to isolate the detector cryostat from the relatively poor vacuum in the sample chamber. In practice the detector vacuum chamber is maintained at less than 10^{-7} Torr by a 2 liter/sec ion pump while the sample chamber is maintained in the range of 10^{-5} to 10^{-6} Torr by a diffusion pump. The vacuum isolation is an important feature since material produced by outgassing of the filament or by target heating might deposit on the surface of the cold detector.

The normal pumpout cycle is designed to prevent pressure differentials from rupturing the thin foils. Absorption of low-energy X rays in the thin foils has been calculated[3,4] and is shown in Fig. 2. These curves show that approximately 60% transmission can be obtained at 277 eV using the 25 $\mu gm/cm^2$ window. Also apparent is the transmission discontinuity at the aluminum K-shell absorption edge at 1.56 keV, which must be included in any detailed efficiency analysis.

The detector was 3 mm depth by 5 mm diameter Si(Li) included in a system employing pulsed-light feedback electronics.[5] This choice of electronics was dictated by the requirement of good energy resolution for the wide range of counting-rates obtained with electron excitation. The electronic resolution of the system was 118 eV when using a Gaussian pulse-shape peaking at 35 μs. It is unfortunate that this was not as good as some of our best systems due to microphonics and other factors. Better energy resolution would be very desirable in low-energy work.

EXCITATION OF LOW-ENERGY FLUORESCENCE X RAYS

Fluorescence excitation of the sample by photons produced either by an X-ray tube or by a radioactive source is quite adequate for many applications of semiconductor X-ray spectrometers. However, in the case of light elements, this situation is modified by the difficulty in generating photons of energy low enough to provide efficient excitation. Furthermore, the filling of a K-shell vacancy in the light elements is usually accompanied by the emission of an Auger electron rather than an X ray. The yield of X rays per K-shell vacancy (ω_k) is shown as a function of atomic number in Fig. 3, illustrating the drastic reduction in X-ray yield for the lighter elements.[6] Observation of these elements therefore requires the use of an intense excitation source; for this reason we have chosen to use a small electron-gun of our own design to excite the samples. The cathode of the gun is maintained at ground potential while the sample is held at a positive potential ranging up to 15 keV. The targets are covered by a thin conducting film when necessary to prevent electrostatic charging effects. Operation of the target at a positive potential ensures that no backscattered electrons will reach the detector. The electron optics are designed to produce a diffuse focal spot approximately 5 mm diameter on the sample. A grid in the gun permits continuous control of the electron current up to 50 μamps, and can also be used to pulse the electron beam when such excitation is required.

Unfortunately, electrons striking the sample produce a brehmstrahlung background from zero up to the maximum electron energy which is superimposed on the fluorescent X-ray peaks observed by the detector. Although such parameters as incident electron energy and angle, and emission angles of the X rays can be adjusted to improve the peak to background for a given characteristic X-ray, we have undertaken no extensive studies of these parameters. The design of the chamber is such that it can be adapted for use with a 200 keV proton accelerator. X-ray data obtained with proton excitation should be much improved both because of the reduction in brehmstrahlung background, and also due to the increased efficiency for light element X-ray excitation with heavy particles.[7]

STUDIES OF DETECTOR ENTRY WINDOW

The attenuation of a photon beam before it reaches the sensitive volume of the detector establishes an effective lower limit on the energy of X rays detected. The entry window on the Si(Li) detector used in these experiments consisted of a 200 Å evaporated gold contact plus an unknown thickness of silicon from which incomplete collection of ionization occurs. A detailed characterization of the silicon dead layer is a complex problem but we will consider it to be a discrete layer characterized by its attenuation of incident X rays.

The effect of the silicon window was studied making use of the discontinuity in X-ray absorption which occurs at the K electron binding energy of silicon. This discontinuity produces an abrupt change in absorption cross section of a factor 10.4 giving a very sensitive measure of silicon layers of 0.1 to 1.0 micron. Figure 4 shows data obtained from a continuous brehmstrahlung spectrum generated with a 3.5 keV electron beam striking a Be foil. A discontinuity occurs at the expected energy and exhibits a dependence of bias voltage characteristic of an undepleted silicon window. Comparison of the data with theoretical absorption cross-sections yields a window thickness vs. bias voltage dependence as shown in Fig. 5. The silicon window thickness is observed to be approximately 0.2 microns at the normal operating bias of 500V.

As assessment of the absorption characteristics of the 200 Å gold layer is more difficult for two reasons:

a) The nominal thickness of the layer is comparable to the wavelength of the incident radiation (e.g. 44 Å for Carbon K X rays) making normal absorption calculations of dubious validity.

b) The thickness of the evaporated layer is thought to be extremely non-uniform as indicated by resistance vs. thickness measurements. The resistance exhibits an abrupt decrease when the thickness exceeds 80 Å, indicating that the layer is

being formed by the growth of islands of evaporant which eventually coalesce to form a filamentary net. Electron microscope photographs of the growth of films with similar resistance characteristics indicate large open areas in films of thicknesses approaching 200 Å.[8] This is a fortuitous circumstanced for the detection of low-energy X rays since the absorption of a uniform 200 Å gold layer, assuming the validity of extrapolated attenuation coefficients, would seriously attenuate low-energy X rays from carbon and oxygen.

Figure 6 is a plot of the relative efficiency, assuming absorption only in the silicon window. The figure also shows the energies of characteristic K-X ray lines from some important light elements. As shown in the curves, the silicon window itself does not severely limit measurements for elements of higher Z than carbon but its effect is becoming significant for carbon X rays. An accurate measure of the efficiency of the system at very low energies, including the effect of the gold layer, could not be made due to the difficulty in generating X-ray lines of known relative intensities at these energies.

LINEARITY AND ENERGY RESOLUTION

The response of the spectrometer to fluorescence X rays in the energy range below 2 keV has been measured and the system linearity and energy resolution determined. The maximum observed deviation of

the measured energies from a straight line fitted to the data between 500 eV and 2 keV was less than the minimum estimated error of 4 eV. The RMS deviation over the entire range was less than 3 eV. These limits were established by uncertainty in locating the midpoint of peaks from the graphical spectra and might be reduced by the application of more sophisticated curve fitting techniques; the system non-linearity could be less than the numbers quoted, but we cannot eliminate the possibilities of minor non-linearities[9] below our limits.

Energy-resolution data were also obtained and are summarized in the plot of resolution squared vs. energy given in Fig. 7. The data are reasonably consistent with statistical summing of electronic noise and detector charge fluctuations. The straight line shown in this figure was calculated using the measured electronic resolution of 118 eV, assuming a mean energy per hole electron pair (ε) of 3.81 eV and a Fano factor (F) of 0.132. There appear to be systematic departures from the straight line below 1 keV which can be attributed to the effect of tailing on the peaks. Measurements of the full-width at 1/10 maximum show an increased tailing effect at low energies, and at low bias voltages, suggesting window-related charge collection problems. Typical resolution (FWHM) are 137 ± 3 eV for Al K X rays (1480 eV), and 126 ± 8 eV for oxygen (525 eV).

DETECTION OF CARBON X RAYS

The region below 500 eV warrants separate discussion because of the special techniques that have been used in these experiments to observe X rays in this range, specifically carbon and nitrogen K X rays at 277 eV and 392 eV respectively. As we have already shown, window-entry problems do not prohibit the measurement of these X rays, but the lower limit to detection is set by noise counts at low energies. A resolution of 118 eV (FWHM) is consistent with a noise-tail extending out to about 250 eV, with the rate of noise counts falling very rapidly with increasing energy in this region.

To reduce noise counts by a large factor relative to the fluorescent X-ray signal, we have employed a pulse-excitation source generated by pulsing the grid on the electron-gun, then gating the electronics for only a brief time when we know that fluorescent X-ray pulses would be at their peak value. By sampling the system output only when there is a very high probability of an X ray being present, we increase the ratio of X-ray events to noise pulses by the off-on ratio of the sampling pulses. Preliminary experiments using a 1 μs wide pulse with a 150 μs period have yielded sufficient noise suppression to permit observation of carbon X rays. A more detailed discussion of this technique will be presented in a forthcoming paper.[10] Figure 8 is a spectrum obtained with a graphite target using the pulse technique; we also show an Al_2O_3 spectrum for comparison.

The energy resolution of carbon and nitrogen X rays under the pulsed operation are 136 ± 8 eV and 131 ± 10 eV (FWHM) respectively, which is consistent with our previous discussion of the departure from statistical behaviour at low energy. We have also consistently observed departures from linearity for these X rays, which we have attributed to the detector itself. The carbon peak position, defined as the point midway between the half-height valves, lies approximately at 0.7 of the expected value extrapolated from the linearity curves for higher energy while the nitrogen line is at 0.9 of its expected value. Linearity measurements on the electronics using a pulser exhibit no non-linearity--we have therefore concluded that it is most likely due to charge collection in the detector. At 277 eV approximately 10% of the photoelectric events occur within 2×10^{-6} cm of the surface. 200 Å In addition to being less than the estimated window thickness, this distance is also less than the range for the photoelectrons produced in the silicon. A sizeable fraction of the ionization will therefore be produced outside the intrinsic volume and will not appear in the signal.

Figure 9 is a composite spectrum showing the response of the detector to carbon, nitrogen, oxygen, and fluorine X rays. The targets were graphite, boron nitrite, a mineral sample, and teflon, respectively. It should be pointed out that further improvements in energy resolution, by improvements in electronics and detector window effects, would significantly enhance the sensitivity for very light-element analysis.

As an illustration of the potentialities for fluorescence analysis at these low energies, Figs. 10a and 10b, show spectra obtained by bombardment of mineral targets by 7.5 keV electrons. The minor differences in composition are readily apparent in the two spectra.

CONCLUSION

These measurements show that the combination of electron excitation with a standard semiconductor detector spectrometer makes possible the measurement of characteristic K X rays of low-Z elements including carbon. We have demonstrated that window absorption effects and the poor excitation efficiency of these low-energy X rays do not prohibit their observation. We have not examined the possibilities of quantitative analytical applications of the techniques described here.

ACKNOWLEDGEMENTS

A large number of people have contributed to this program. In particular; D. Landis, D. Malone, W.Searles, H. Sommer and J. Walton have contributed to the experiments, and we have had useful discussions with R. Pehl and W. Hansen. This work was performed as part of the program of the Nuclear Chemistry Division of the Lawrence Radiation Laboratory, Berkeley and was supported by the U. S. Atomic Energy Commission, Contract No. W-7405-eng-48.

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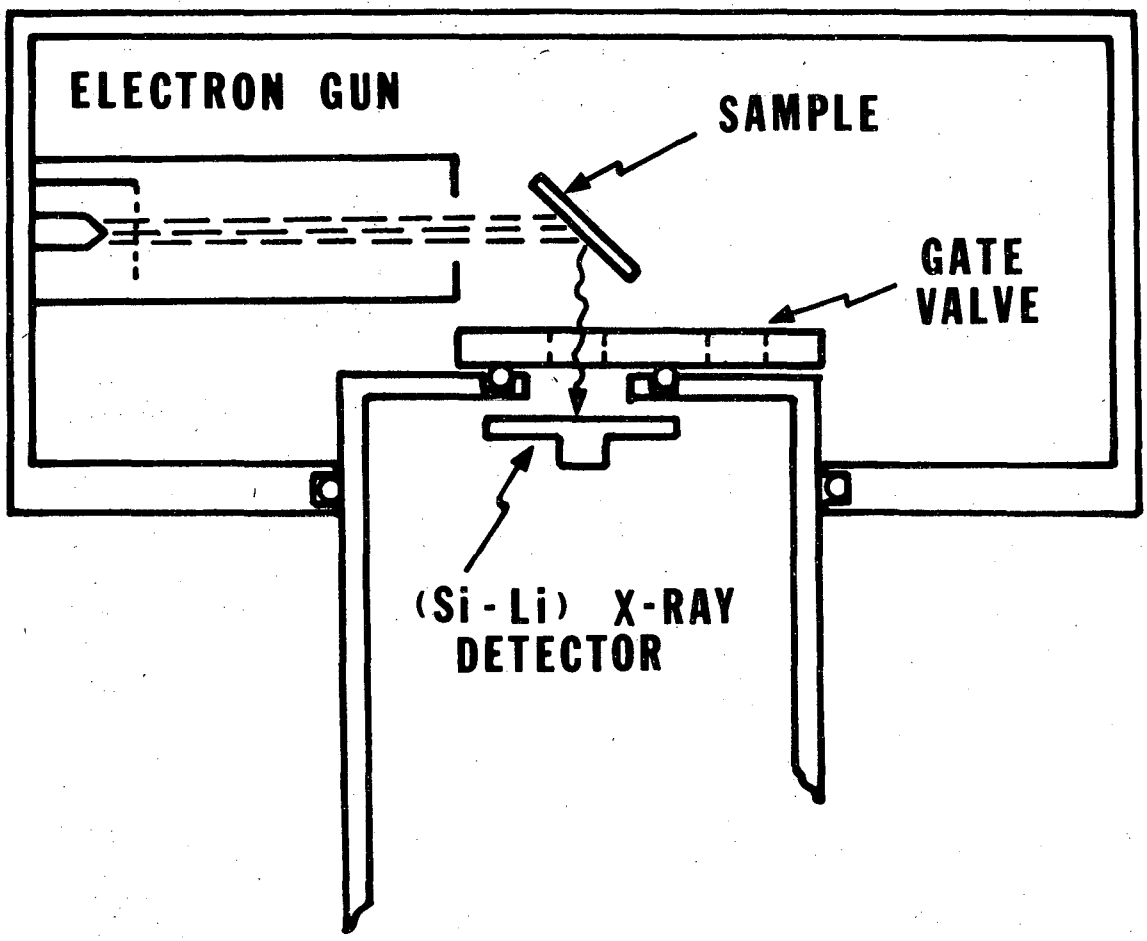

XBL 7010-6814

Fig. 1. Schematic diagram of modified X-ray spectrometer and sample chamber.

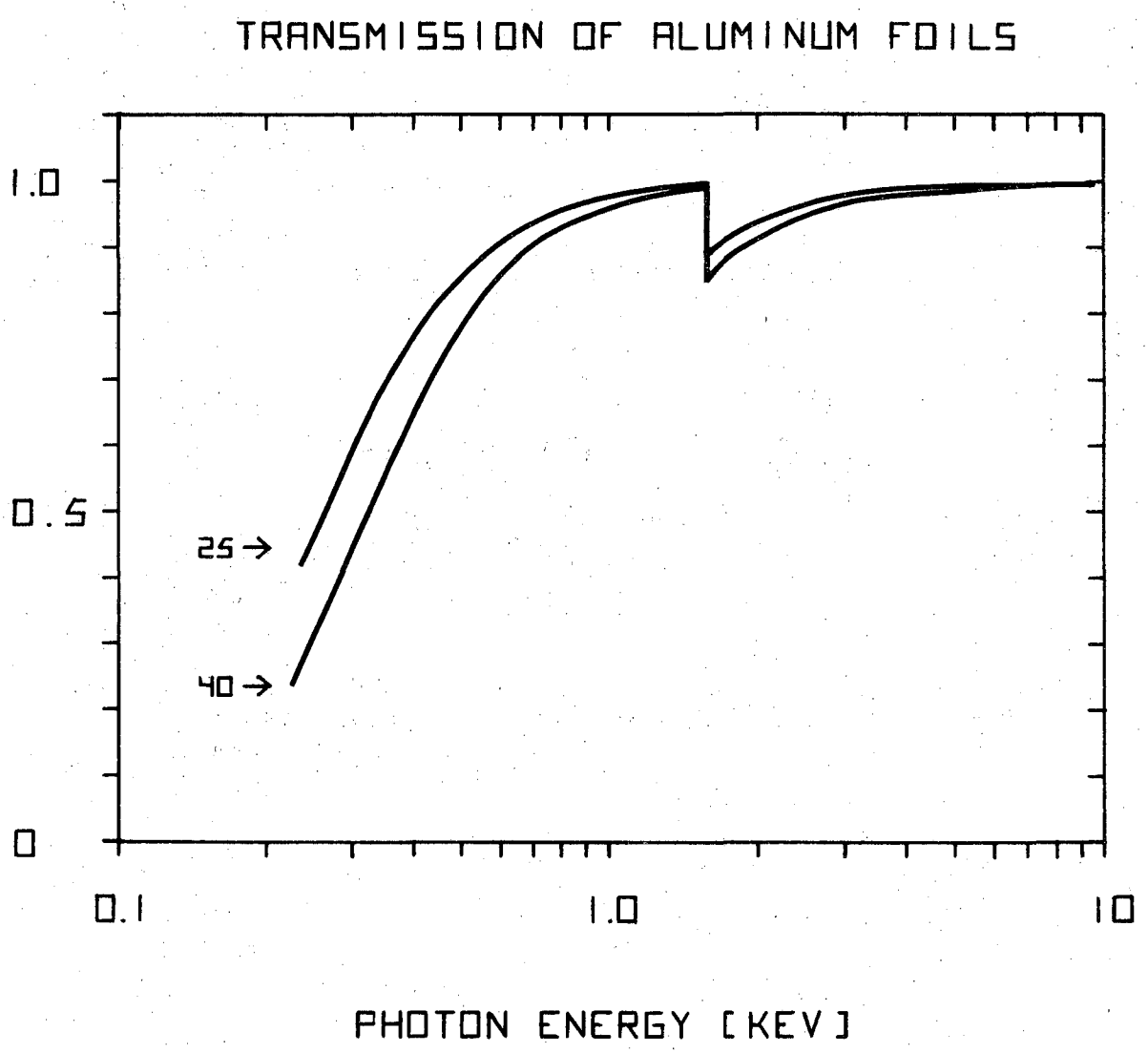

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Fig. 2. Transmission of thin aluminum windows as function of photon energy. Thickness in $\mu gm/cm^2$ is indicated on the plots.

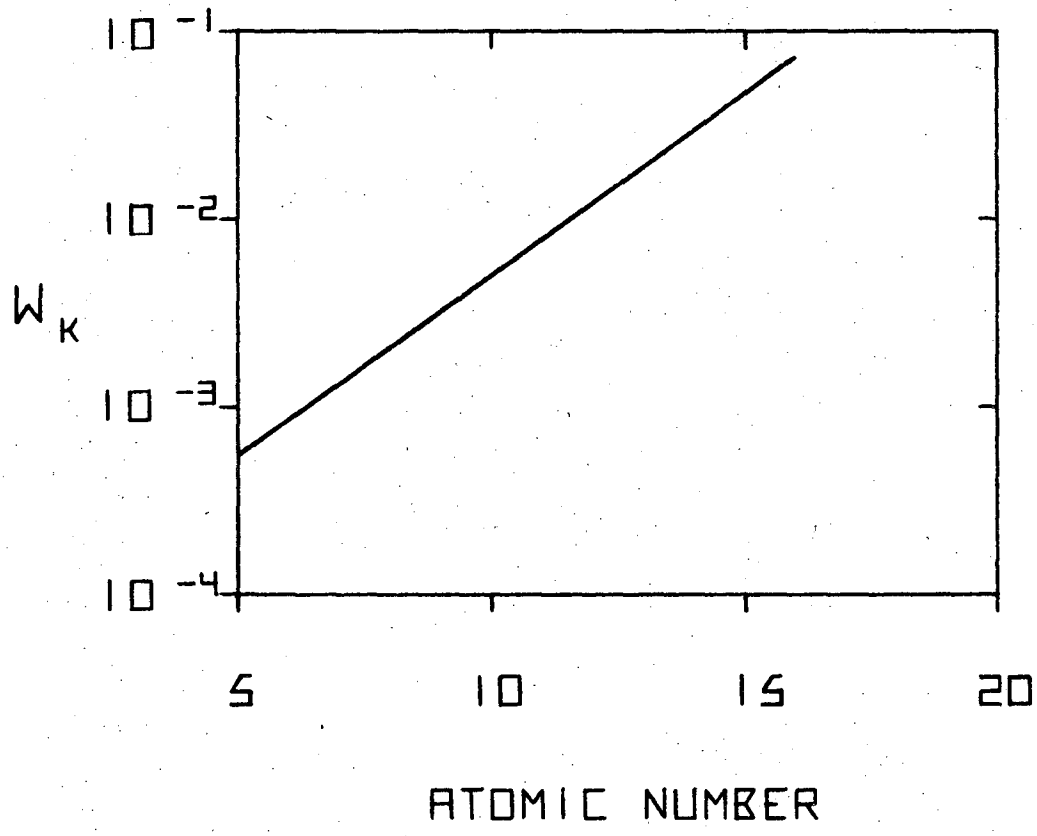

Fig. 3. Fluorescence yield for light elements based on data presented in Ref. #6.

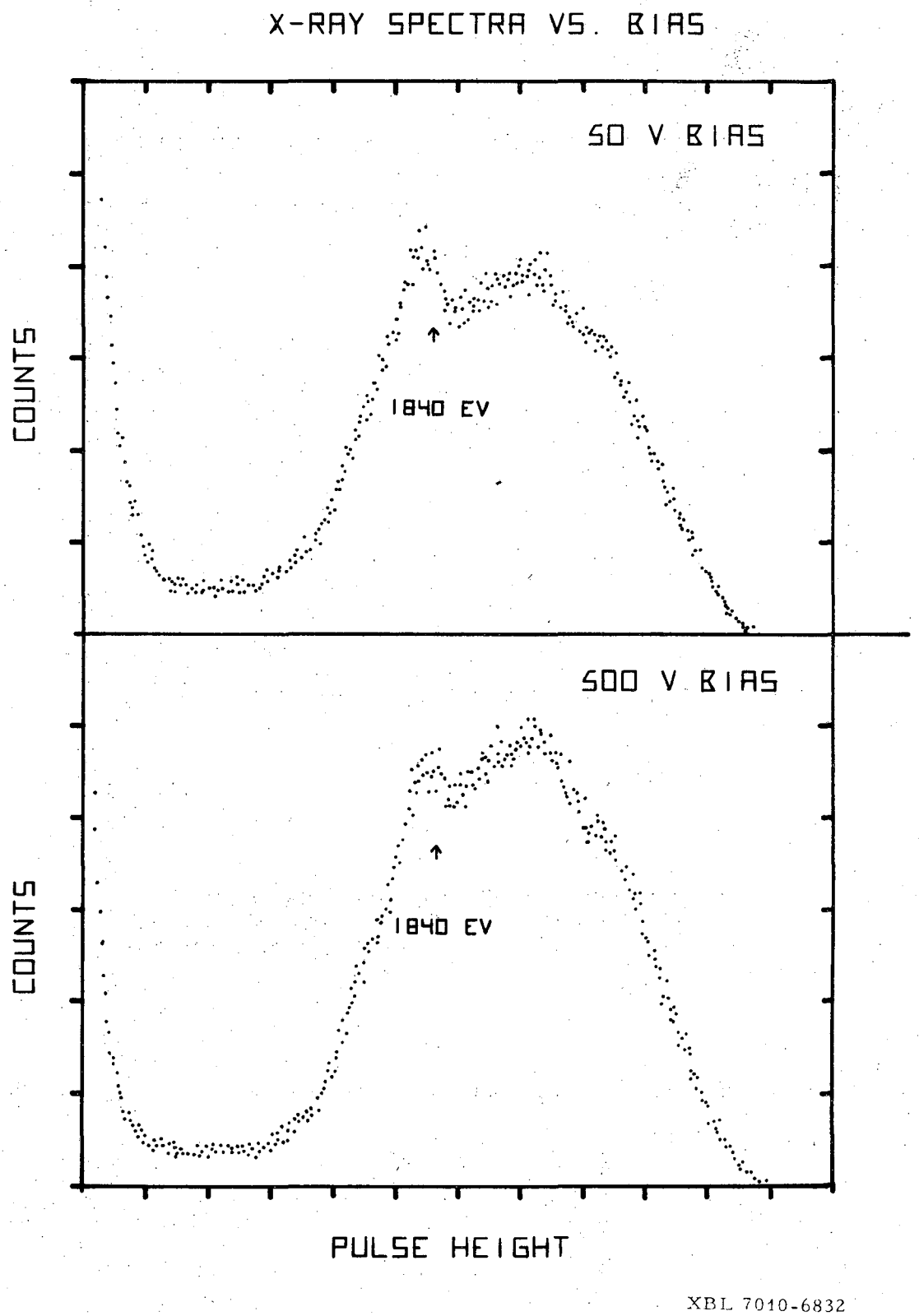

Fig. 4. Detector response to continuous brehmstrahlung spectrum showing discontinuity at the silicon K edge (1840 eV).

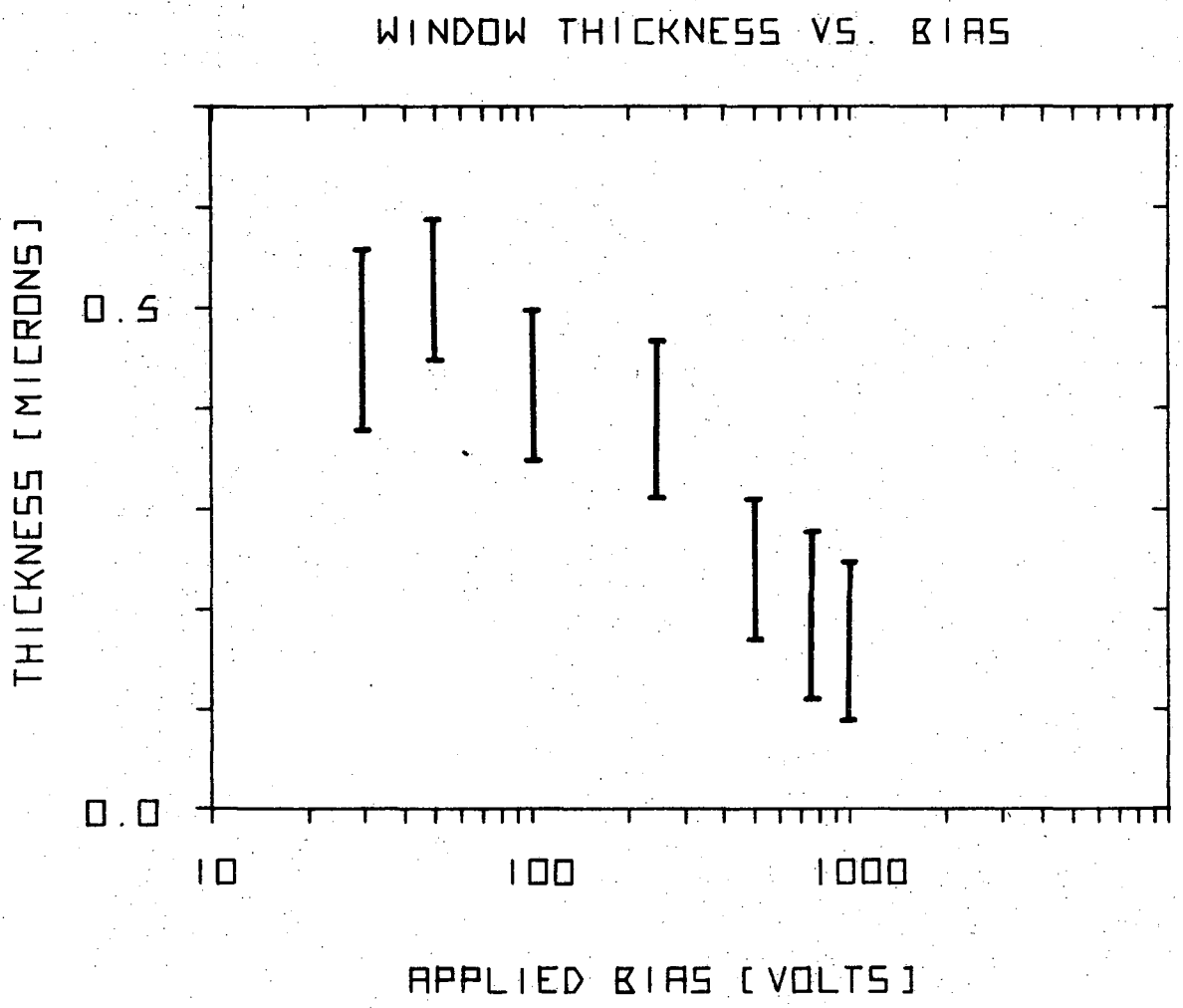

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Fig. 5. Dependence of silicon window thickness on bias voltage as determined from K edge measurements.

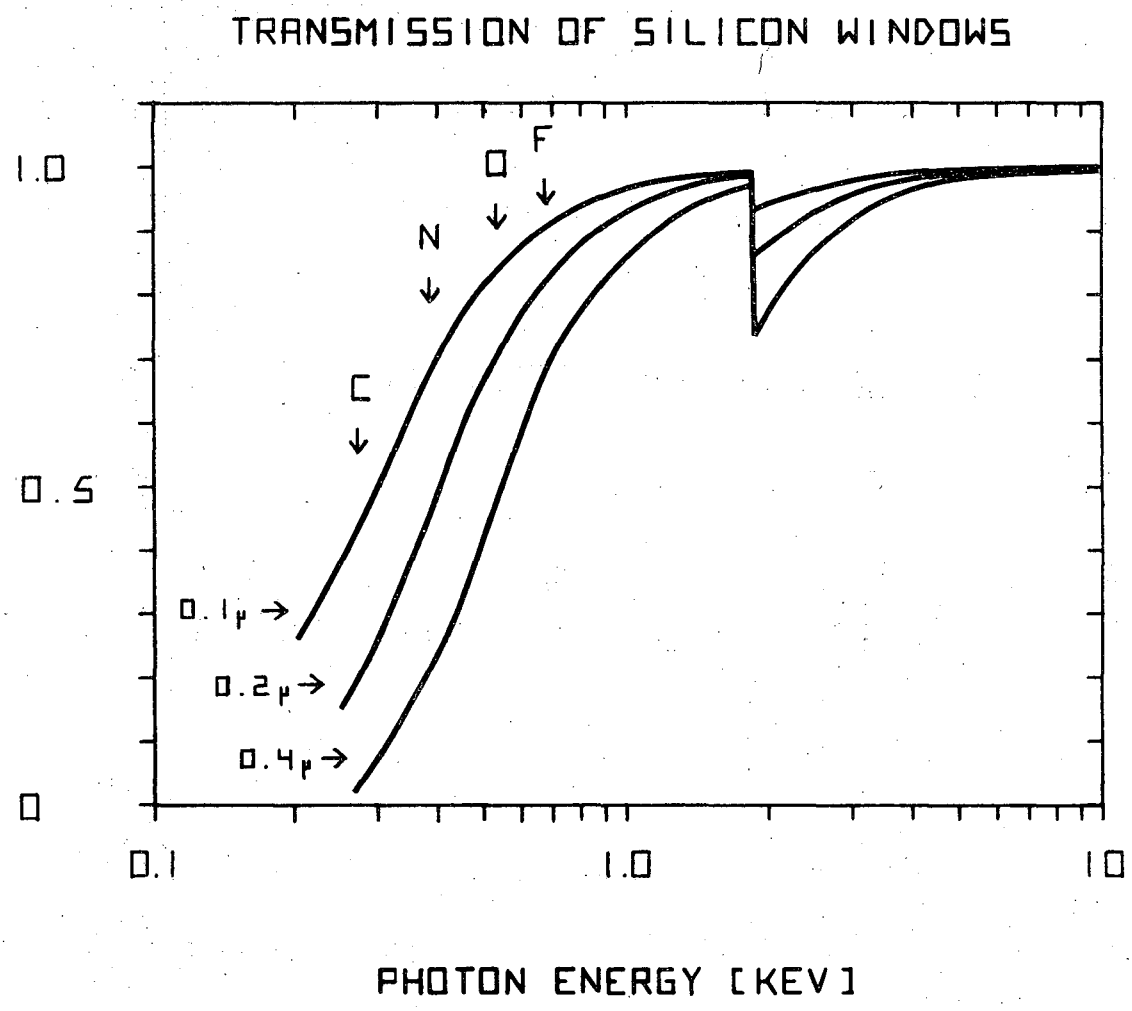

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Fig. 6. Plot of relative efficiency vs. energy corresponding to silicon windows of various thicknesses.

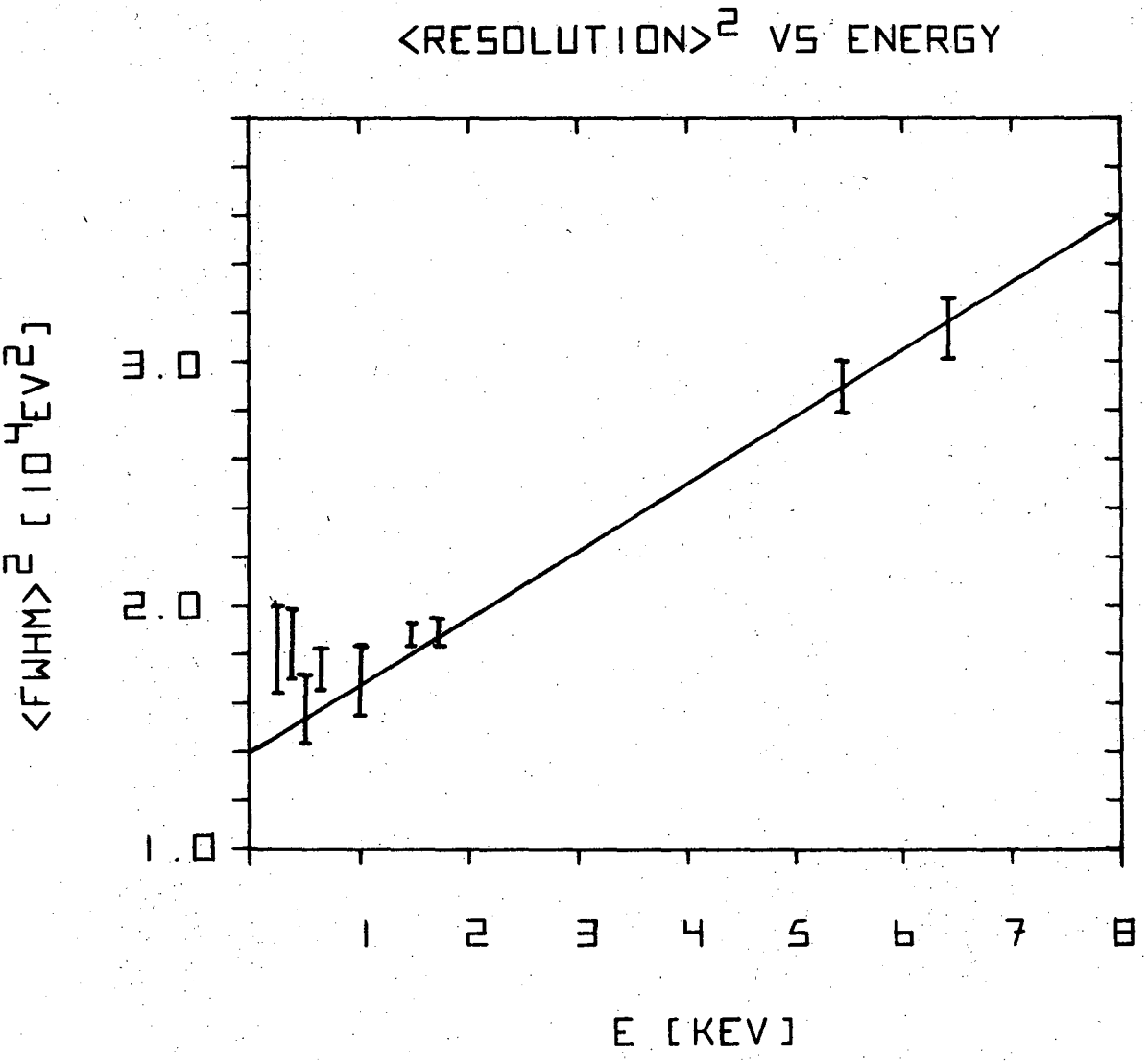

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Fig. 7. Resolution squared vs. energy.

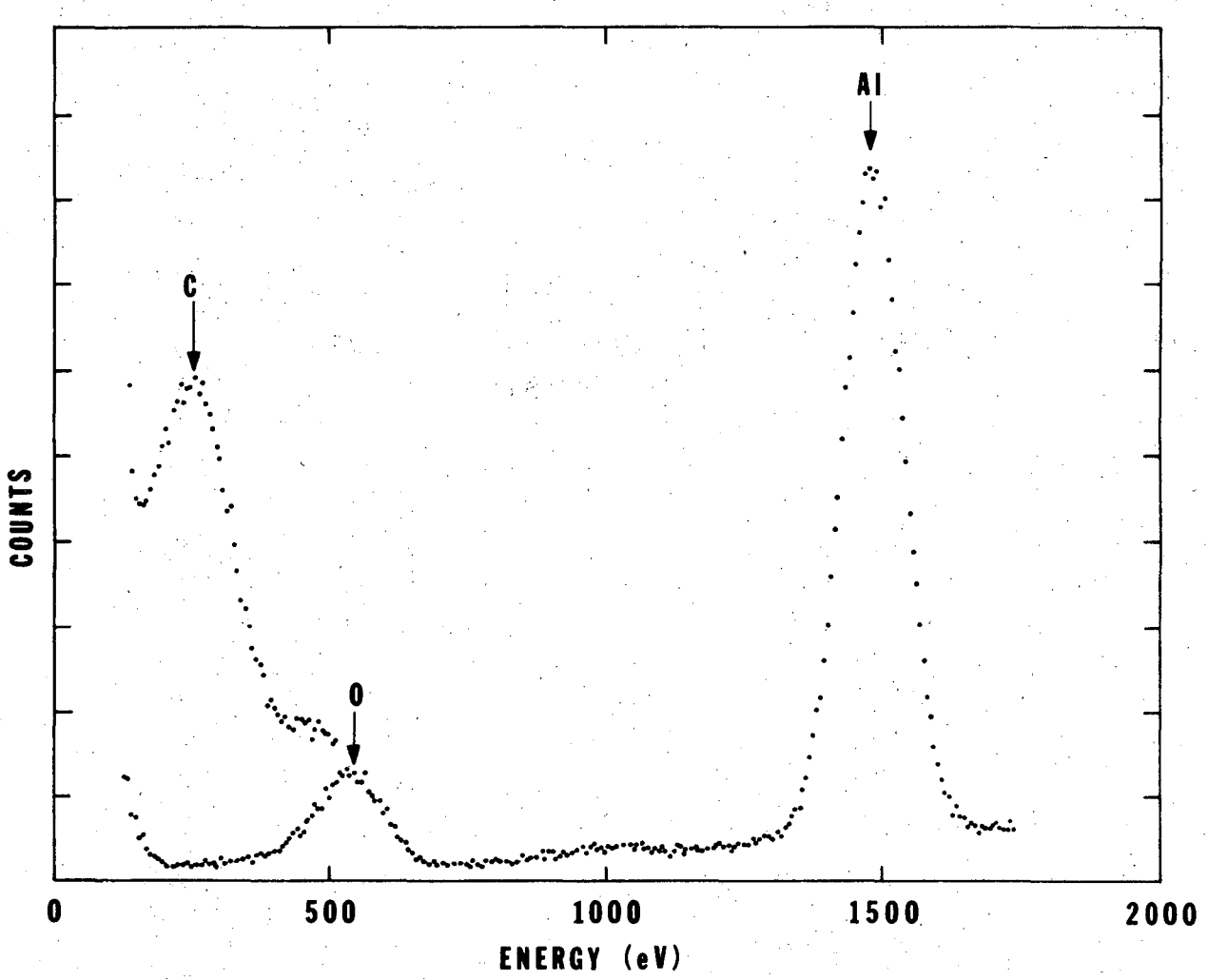

Fig. 8. Spectrum of carbon X rays from graphite target observed in the pulsed mode. An Al_2O_3 spectrum is shown for comparison.

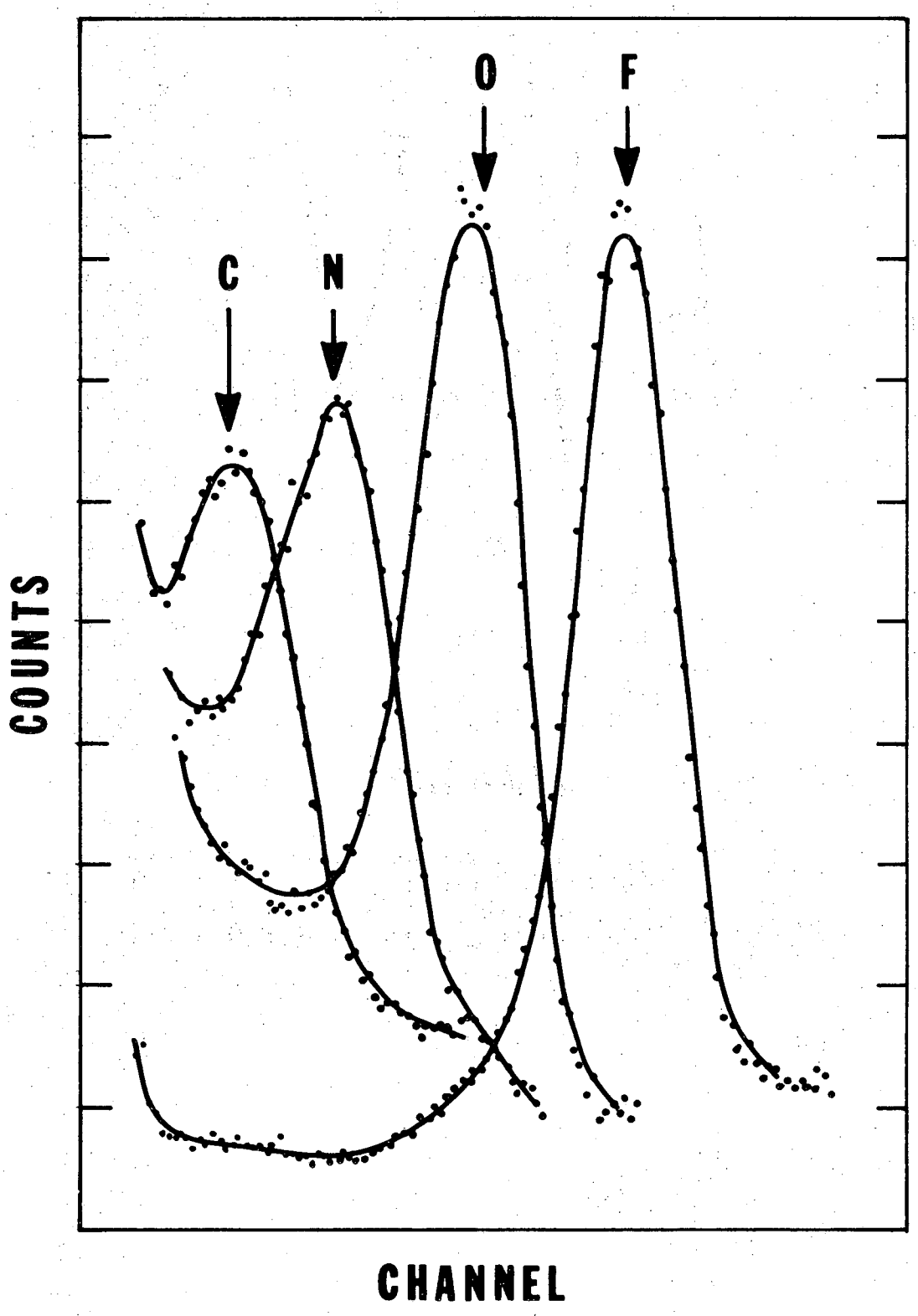

Fig. 9. Composite spectrum of characteristic X ray for light element. Targets were graphite (carbon), boron nitrite (nitrogen), mineral sample (oxygen) and teflon (fluorine).

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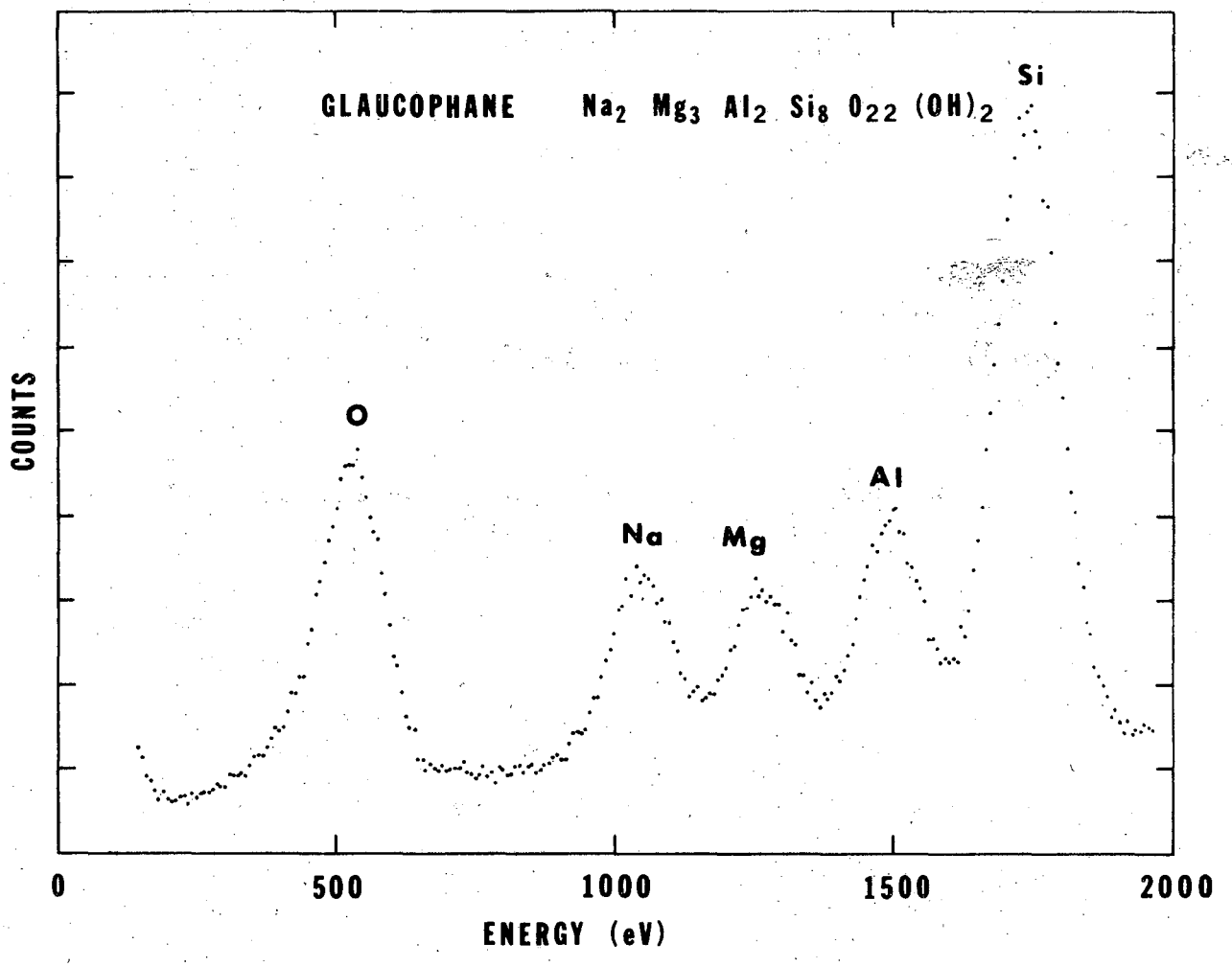

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Fig. 10a. Spectrum of fluorescence X rays from electron excitation of glaucophane. The composition of this mineral sample is indicated on the graph.

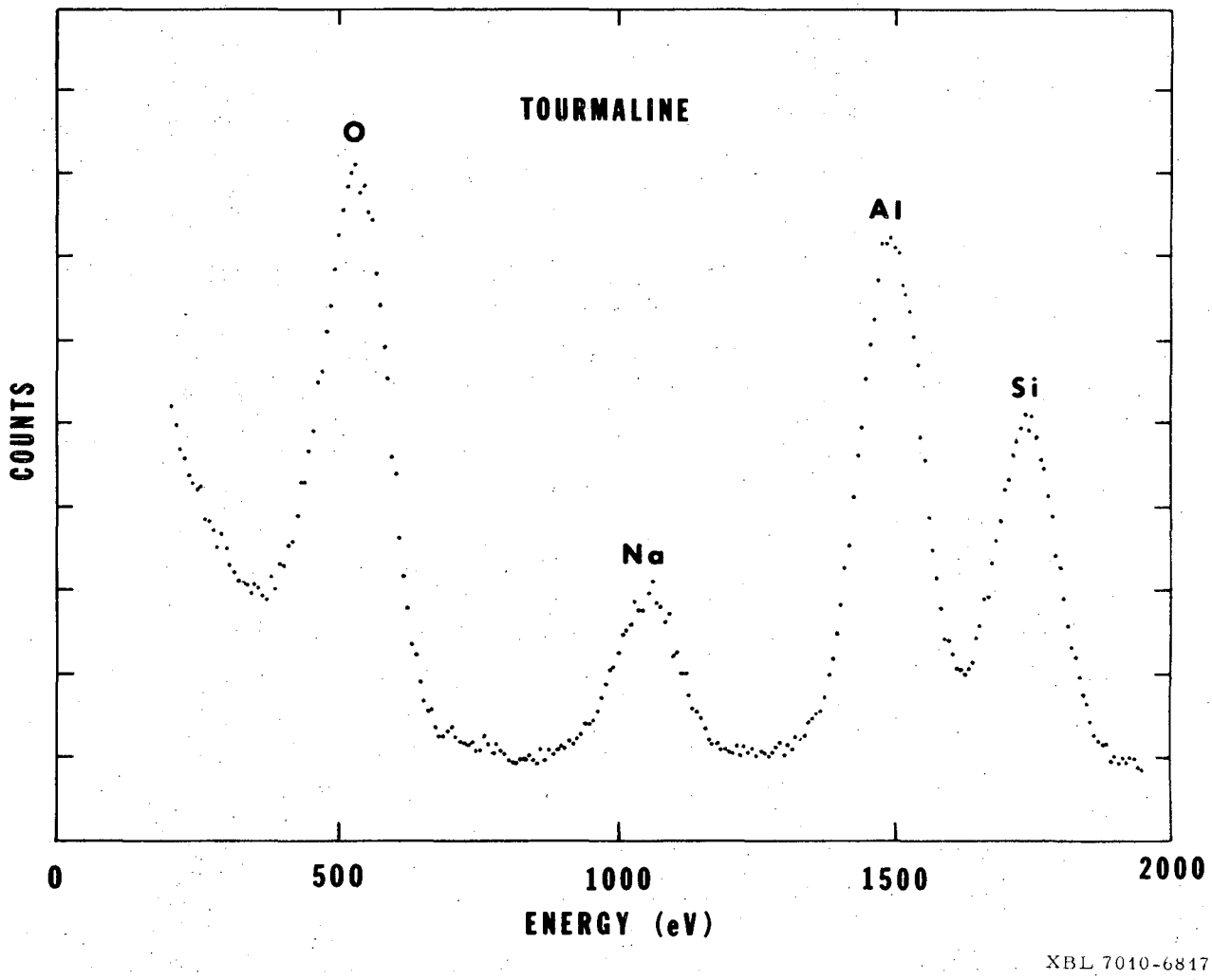

Fig. 10b. Spectrum of X rays from tourmaline sample. The composition is similar to glaucophane except for the absence of magnesium in this sample.

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