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V. 30 PIXE with high-energy Proton Beams

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The detection limit of a trace-element analysis by particle-induced x-ray emission (PIXE) spectroscopy has been defined by $N_p \geq 3\sqrt{N_b}$, where N_p is the counts of a characteristic x-ray peak in an x-ray spectrum and N_b is the background counts under the peak. The continuum X-ray background usually comes from secondary electron bremsstrahlungs (SEB),^{1,2)} radiative ionizations (RI)³⁾ or quasifree-electron bremsstrahlungs (QFEB), nuclear bremsstrahlungs and γ rays from nuclear reactions.⁵⁾ Relative intensities and the spectral shape of these components depend on the projectile particle and energy, and the continuum background, in general increases rapidly with increase in the projectile energy. It has therefore been argued that protons of 1-3 MeV are most favorable to obtain the maximum sensitivity of PIXE.⁵⁾ Though the background increases rapidly with increase in the projectile energy, K x-ray production cross section for medium and heavy atoms increases too. The detection limit is then determined by competition between these two factors. We have studied the detection limit on the basis of the background actually measured at high proton-energy region with our cyclotron facility, where special care has been taken to reduce the background coming from parts other than the target material.⁶⁾

The experiment was performed using a scattering chamber in the third target room. The target room is shielded with a concrete wall of 1-m thickness and also with heavy-concrete blocks of 50-cm thickness. Neither slit nor collimator is used in the target room. The beam spot on the target was less than 3 mm in diameter. After passing through the target, the beam is focused with a pair of quadrupole magnets on the end of a Farady cup, which is 5.6-m distant from the target and is shielded with 1-m thick heavy-concrete blocks. X-rays emitted from the target in the direction of 135° to the beam were measured with an ORTEC Si(Li) detector of energy resolution of 155 eV for 6-keV x-rays. This backward direction was chosen since the secondary-electron bremsstrahlung decreases in this direction because of the relativistic retardation effect¹⁾ and also the quasifree-electron bremsstrahlung shifts to low-energy because of the Doppler effect.⁷⁾ With this experimental setup, it was confirmed that the background measured without target is quite negligible in comparison with continuum x-rays from the target. The target was a selfsupporting Al foil and its thickness was 3.46×10^{17} atoms/cm². Continuum x-ray spectra from this target bombarded with protons of 3, 6, 9 and 12 MeV are shown in Fig. 1, where the abscissa shows the x-ray energy plotted in the atomic number corresponding to the characteristic K x-rays and the ordinate represents the background counts

N_b per microcoulomb of the proton beam under each characteristic K x-ray peak, of which the FWTM was assumed to be $\sqrt{a + bE_x}$ with the x-ray energy E_x , a and b being experimentally determined. As seen in this figure, the continuum background increases and extends to higher energy with increase in the projectile energy. By taking these continuum x-rays actually measured as the background, the detection limits were estimated from the K and L x-ray production cross section.⁵⁾

The detection limits thus obtained for K and L x-ray measurements are shown in Figs. 2 and 3, respectively, where $T_m = 4 m_e E_p / M_p$ represents the maximum energy that can be transferred from a projectile of mass M_p and energy E_p to a free electron of mass m_e . It is concluded from Figs. 2 and 3 that PIXE with 6 ~ 12 MeV protons are preferable for simultaneous multi-element analyses containing $Z = 22 \sim 40$ elements and $Z = 65 \sim 85$ elements by K and L x-ray measurements, respectively. Since, in this proton-energy region, density of the contour lines is low in contrast to the high density in the low energy region, practically high sensitivity of nearly constant can be obtained over a wider range of atomic number. From a practical point of view, it is quite important that a wide range of atomic number can be covered by characteristic K x-ray measurements. These values of detection limit were estimated for a backing of 3.46×10^{17} -atoms/cm²- thick Al foil and change depending on the thickness and atomic number of the backing material. However, the general trend of change with the projectile energy is not so sensitive to the backing so far as it is thin enough. In Figs. 2 and 3, T_m and T_r^D are the maximum energies of the secondary electron bremsstrahlung (SEB)¹⁾ and the quasi-free electron bremsstrahlung (QFEB) taking account of the Doppler effect.⁷⁾ The line V_{dl} shows a locus of the minimum detection limit. In the region between T_r^D and T_m , SEB is the predominant component of the background, and on the right side of T_m , the background comes only from the Compton-scattered γ rays. In the region $E_p < 3$ MeV, the lines T_m and V_{dl} are expected to overlap or to be very close and the error due to background subtraction can not be expected to be small. In the region $E_p > 3$ MeV, however, the line V_{dl} is on the left side of the line T_m and the background spectrum becomes smooth so that it can easily be subtracted. Furthermore, the sensitivity is the highest in this region.

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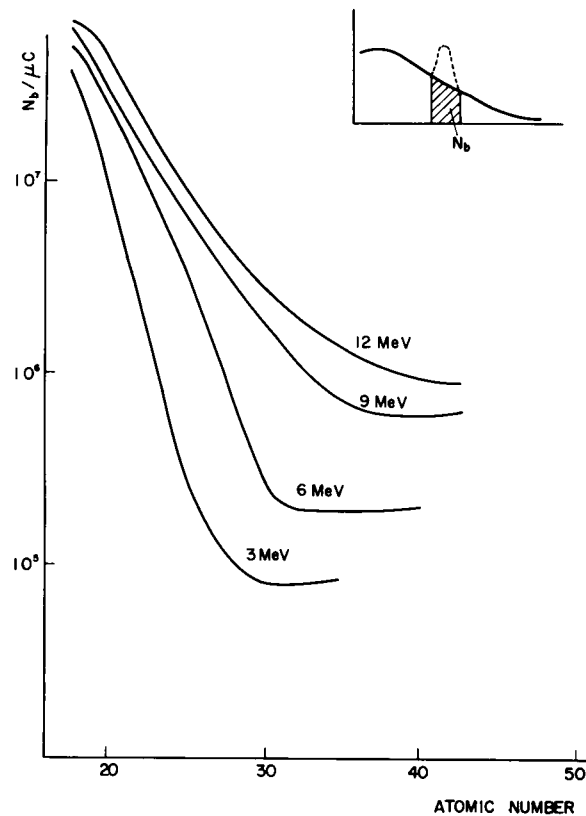


Fig. 1. Continuum X-ray spectra from an Al target of high purity bombarded with 3-, 6-, 9-, and 12-MeV protons. The abscissa shows x-ray energy plotted in atomic number corresponding to its characteristic K x rays. The ordinate represents the continuum x-ray counts -background counts- under each characteristic x-ray peak.

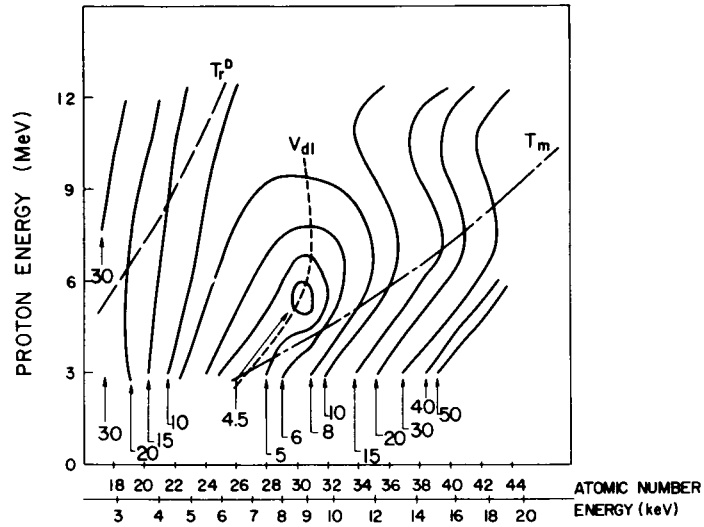


Fig. 2. Contour representation of the detection limit in units of number of atoms (10^{10})/cm² for K x-ray measurements. The lines T_r^D and T_m show the high-energy limits of QFEB and SEB, respectively, and the line V_{dl} is a locus of the minimum value of detection limit.

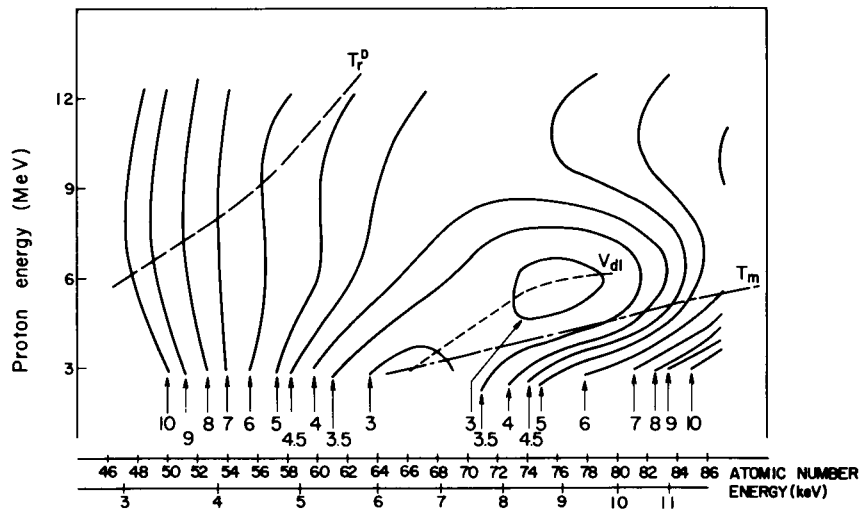


Fig. 3. Same as Fig. 2, except for the L x-ray measurements.