MAX-PLANCK-INSTITUT FÜR PLASMAPHYSIK

GARCHING BEI MÜNCHEN

Energy Distributions of Charged and Neutral Hydrogen Atoms Backscattered from Metal Surfaces Bombarded with 5 to 18 keV Protons

P. Meischner and H. Verbeek

9/18 Januar 1975

Die nachstehende Arbeit wurde im Rahmen des Vertrages zwischen dem Max-Planck-Institut für Plasmaphysik und der Europäischen Atomgemeinschaft über die Zusammenarbeit auf dem Gebiete der Plasmaphysik durchgeführt.

IPP 9/18

P. Meischner
H. Verbeek

Energy Distributions of Charged and Neutral Hydrogen Atoms backscattered from Metal Surfaces Bombarded with 5 to 18 keV Protons.

January 1975

ABSTRACT

Polycrystalline targets of Be, V, stainless steel, Mo, Nb and Ta have been bombarded with protons with primary energies in the range from 5 to 18 keV. The energy distributions of the charged and neutral particles backscattered at 135° with respect to the primary beam direction have been measured between 200 eV and 18 keV. Before measurement, the neutral particles were partly ionized in a calibrated gas stripping cell.

The observed energy distribution of the neutrals has a pronounced maximum between 0.5 and 1 keV whose position does not depend within the experimental error on the primary energy, the target material, or the angle of emergence of the scattered particles. The energy distribution of the charged particles shows a less pronounced maximum between 1 and 1.5 keV for angles of emergence near the normal to the target surface. For more oblique emergence, the distribution becomes flatter and the maximum vanishes for grazing emergence. Only slight differences in the shape of the energy distributions have been observed for different target materials.

The fractional number of charged backscattered particles increases from 3 % at 300 eV to 40 % at 18 keV. In this energy range it decreases slightly as the angle of emergence becomes more oblique.

...2

NOTE

A paper with the above title was presented at the conference on "Surface Effects in Controlled Thermonuclear Fusion Devices and Reactors" at Argonne National Laboratory Jan. 10-12, 1974 and appeared in Journal of Nuclear Materials(1). In this paper the essential results were reported and illustrated by a few typical figures. The purpose of this report is to present more completely the experimental material that was collected during the last 6 months of 1973. For reasons of comprehension the introduction of Ref. (1) is repeated here with only slight modifications.

INTRODUCTION

Knowledge of the energy, angular, and charge distributions of light atoms backscattered from metal surfaces is of importance for the evaluation of the interaction of a hot plasma with the surrounding walls. The energy range up to 20 keV is of special interest since this corresponds to the temperature in a D-T fusion reactor.

For energies below 20 keV it is no longer possible to determine the energy distributions using surface barrier detectors, which are sensitive to charged and neutral particles alike. There are several papers (2 to 6) which report measured energy and angular distributions of the charged component of the backscattered beam. Most of the particles are, however, backscattered as neutrals. In some cases attempts have been made to calculate the energy distribution of the total backscattered intensity (including the neutrals) from the energy spectra of the charged component by using experimental(3, 4) or theoretical data for the ion escape probability. There are, however, considerabl doubts whether the experimental data measured at higher energies and for different materials are applicable to this problem and whether the theoretical values are reliable.

...3

EXPERIMENTAL

The experiments were performed with the BOMBARDON accelerator (7). A neutral particle analyzer designed for measuring ion temperatures in Tokamak experiments by the TFR group in Fontenay-aux-Roses (France) (8) was employed. With this device, the energy distributions of positively charged and neutral particles backscattered at the fixed angle of 135° have been measured. In Fig. 1 a schematic view of the experimental setup is shown. A magnetically selected ion beam impinges on the target in an area of 0.5 mm diameter. The target can be rotated so that the entrance and exit angles with respect to the target normal can be varied. The target may be baked by electron bombardment from the rear. Further cleaning by sputtering with 10 keV Ne^+ ions is possible. From the scattered particles a beam is selected and passed through a set of deflection plates and a stripping cell, then analyzed by a 90° electrostatic cylinder spectrometer, and counted by a channeltron electron multiplier. The scattered beam is confined by the entrance aperture of the stripping cell to a solid angle of 6×10^{-6} sterad. For the analysis of the charged particles the deflection plates were grounded and the stripping cell evacuated. For the neutrals, an electric field of 1.5 kV/cm at the deflection plates eliminated the charged component and quenched the metastables. The neutrals are partly ionized in the stripping cell with 2×10^{-3} torr N_2.

For the conversion of the number of counted particles to the number of neutrals entering the stripping cell, we relied on the calibration curve of the TFR group. The calibration takes into account the stripping efficiency, scattering in the gas cell, and the detection efficiency of the channeltron. The overall efficiency rises from 8×10^{-4} at 200 eV to 3.5×10^{-2} at 3 keV with 2×10^{-3} torr nitrogen in the cell. This overall efficiency is claimed to be accurate within $\pm$ 20 %. We extrapolated this curve up to 18 keV, a procedure which seems to be valid within the errors of $\pm$ 20 %, since the TFR calibration curve follows the trend of corresponding curves of other authors extending to higher energies (9) and the stripping cross

...4

section is monotonically increasing with increasing energy up to 30 keV(10). Below 1 keV, however, the error is larger because our experimental conditions may differ from the calibration conditions. Therefore, the height of the calculated spectra may be uncertain by a factor of two below 1 keV.

The data were collected in the following manner: The primary beam current impinging onto the target was digitized and counted by a preset count number (resembling a certain charge collected on the target) was reached the channel advance of a multiscaler was triggered. The multiscaler counted the number of pulses from the channeltron. The voltage on the analyzer plates was controlled by an analog signal proportional to the channel number. Thus the particle energy transmitted through the analyzer was proportional to the channel number. The spectra were therefore independent of fluctuations in the primary beam current.

From the data collected with the multichannel analyzer and the calibration curve, the energy spectra and the charged fraction of the number of positively charged to the total number of backscattered particles, Q, were calculated using an IBM 360/91 computer.

The shapes of the spectra obtained by using the calibration curve of the TFR group are believed to be correct. The absolute value of the fraction of charged backscattered particles (neglecting negatively charged ions) to the total number is correct within ± 20 %. The relative heights of the curves for different primary energies, different angles of emergence ß, and different materials are correct within counting statistics. The absolute numbers of particles backscattered per primary particle could not be determined in these experiments, since the correct angle of acceptance and the spectrometer function are unknown.

...5

RESULTS

In Fig. 2 a computer plot of the energy distributions of protons and neutral hydrogen atoms backscattered from a Ta sample is shown. The energy spectra are corrected for the resolution of the electrostatic analyzer and normalized to the primary beam current. Since the calibration curve of the neutral particle analyzer used to obtain the neutral spectrum contains the efficiency of the channeltron as measured by Egidi (11), the same efficiency curve was used to obtain the charged spectrum. In all spectra plotted one channel is equal to an energy interval of 75 eV.

The charged fraction $Q = N^+/(N^+ + N^0)$ was calculated for each channel, and is also plotted in Fig. 2.

The spectra shown in Fig. 2 are typical of all spectra measured. They extend from zero energy to a sharp cutoff at $E_0 - \Delta E$, where E_0 is the primary energy and ΔE the elastic energy loss of protons singly scattered from surface atoms to $\vartheta = 135^\circ$. The gradient of the curve at the cutoff is due to the limited resolution of the energy analyzer. The backscattered intensity increases with decreasing energy of the emerging particles. It has a maximum at ≈ 1 keV for the neutrals and ≈ 2.5 keV for the protons. The charged fraction Q increases gradually with increasing energy.

VARIATION OF THE PRIMARY ENERGY

The backscattering spectra shown in Fig. 3 are for Ta bombarded with protons with primary energies $E_0 = 9.2$ to 18.5 keV, and were obtained in the manner described above. The shape of the spectra is essentially the same for all primary energies investigated. The number of particles backscattered from the surface layer, i.e. with the highest energies, decreases with increasing primary energy E_0. This is due to the decrease of the cross section for

...6

nuclear collisions with increasing energy. The number of all backscattered particles integrated over all energies also decreases when E_o is increased. It should be borne in mind that the spectra shown in Fig. 3 are taken for equal numbers of primary particles per channel. Thus a greater number of primary particles were utilized in recording spectra at higher E_o than for lower E_o. Therefore the integrals over all energies of the spectra in Fig. 3 had to be divided by E_o to relate them to equal numbers of primary particles. The relative numbers of backscattered particles for $E_o = 9.2$, 12.3, 15.4, and 18.5 keV are 3.9, 2.7, 1.6, and 1.

When E_o is decreased the slope of the spectra towards the maximum at ≈ 1 keV is increased, and the maximum is more pronounced. The position of the maximum is not affected by E_o, within the experimental error.

In addition, the charged fraction Q shows no dependence on the primary energy.

BOMBARDMENT WITH MOLECULAR IONS

For low primary energies E_o it is advantageous to use H_2^+ beams, because the achievable beam currents are larger. Fig. 4 shows backscattering spectra for Ta bombarded with 12.3 and 18.5 keV H_2^+ molecular ions. The shapes of the spectra are the same as those for bombardment with protons with primary energy $E_o/2$. The molecular ions dissociate into 2 protons at the metal surface, when their binding electron is stripped. Each proton receives half the energy of the molecule. This is clearly shown in Fig. 5 when the backscattering spectra for Nb bombarded with H_1^+ and H_2^+ ions of equal primary energies E_o are compared. The charged fraction Q seemed to be somewhat lower for H_2^+ bombardment in several cases. This effect was not well established by these experiments. It seems to be real, however, since later experiments (reported elsewhere$^{(12)}$) clearly showed that the fraction of the number of negative to the number of positive ions may depend on the primary ion species.

VARIATION OF THE ANGLE OF EMERGENCE ß

With the present experimental setup it was only possible to measure at the scattering angle of $\vartheta = 135^{\circ}$. The angle of emergence ß, however, could be varied by rotating the target. This caused, however, a simultaneous variation of the angle of incidence α (see Fig. 1) with $\alpha = /45^{\circ} - ß/$. The shapes of the backscattering spectra vary considerably with α. The depths from which particles appearing with a certain energy are backscattered depend strongly on the specific combination of α and ß. Since the charged fractions Q are believed to depend on the angle of emergence only, we shall now consider this dependence in greater detail.

In Figs. 6 and 7 the influence of ß on the spectra and the charged fraction Q is shown for backscattering from Nb and Ta. When ß is increased the total backscattering intensity is decreased. A large effect on the shapes of the spectra is seen only on the charged energy distributions, especially at low energies. The resulting decrease of Q with increasing ß i.e. for more grazing emergence is shown in Fig. 8 in one plot for the case of Nb.
A further example of this variation is shown in Fig. 9 obtained using a primary beam of H_2^+ ions incident on Vanadium.

BACKSCATTERING FROM DIFFERENT MATERIALS

The selection of materials investigated was determined by their potential role as a wall material for nuclear fusion devices. In addition, Be was studied as an example of a very low Z material. In Fig. 10 the backscattering of 18.5 keV protons from Be, V, Nb, and Ta at ß = 0 is shown. Fig. 11 shows results obtained for bombardments with H_2^+ ions of 18.5 keV on Be, V, stainless steel, Mo, Nb, and Ta for $ß = 45^{\circ}$. In general the total backscattering intensity is increased with increasing Z. However, no quantitative members are presented here. The influence of the increasing cross section

...8

for nuclear scattering with increasing Z can best be seen from the increase of the backscattering intensity at the surface.

The shapes of the spectra depend also on Z, showing a much more pronounced maximum at low energies for low Z than for high Z.

With the exception of Be, no apparent influence of Z on the charged fraction Q could be observed. The spectra of Be, however, show the presence of some heavier impurities on the surface. This will be discussed in the next paragraph.

INFLUENCE OF SURFACE CONTAMINANTS

All samples were mechanically polished and cleaned with methanol in an ultrasonic chamber. They were heated in situ by electron bombardment of the back surface of the sample up to 1700 °C in the case of V, Mo, Nb and Ta, and to 900 °C in the cases of Be and stainless steel. This removed adsorbed layers from the surfaces and altered considerably the shapes of the spectra at the high energy cutoff, as was observed earlier(5). A further cleaning was in some cas accomplished by sputtering with Ne ions of ≈ 10 keV. Only in the case of Be did this method cause any further alternation of the shapes of the energy spectra. In Fig. 10, an arrow on the Be spectrum indicates the theoretical cutoff at high energies for a clean Be surface. The backscattering intensity with energies above the marked energy is therefore due to impurities. These were identified to be mostly O and some C. After alternating heating and sputtering with Ne ions the Be spectrum no longer showed any remarkable backscattering at energies above the Be-cutoff. This is shown in Fig. 12. A large change ocurred in the charged spectrum and thus in Q, which decreased in the case of the cleaner surface. In the case of stainless steel shown in Fig. 13, the same behaviour of the charged fraction was observed. In this case, however, as with all other metals investigated, the surface cleanliness was not monitored. The surface conditions of V, Nb, and Ta are doubtful. It is well known that

...9

these metals usually have an oxide layer at the surface, and this layer is not easily to be removed. For these cases it therefore cannot be excluded that the observed dependence of Q on the angle of emergence ß is due to such surface layers (13).

OCCURENCE OF NEGATIVE BACKSCATTERED IONS

In a later experiment negative as well as positive ions could be detected (14). Fig. 14 shows an example. In this case the neutral spectrum was obtained from the spectrum of the positively charged particles by using the Q-values from Fig. 8. It is seen that in this case the number of negatively charged particles in less than 5% of the total number at all energies. Thus the error when neglecting the negative ions in the total spectra is less than 5 % at least for Nb.

DISCUSSION

One result of this work is that the energy distributions of the charged and of the neutral particles backscattered in this energy range are quite different. Since the neutrals are the major fraction of all backscattered particles, it is impossible to deduce the energy distributions of all backscattered particles (including the neutrals) from a measurement of the charged particles alone. On the other hand only the total number of backscattered particles can be compared with present calculations.

Another result is that the energy distributions of backscattered hydrogen atoms (including the neutrals) show a pronounced maximum in the range of 500 to 1000 eV independent of the primary energy and the metal investigated. Although the height of this maximum, and its position to some extent, depend on the calibration curve of the neutral particle analyzer, the shape of the curves

...10

is apparently correct, as discussed above. The existence of this maximum yield at such low energies is believed to be an essential feature for these backscattering spectra. Unfortunately no information on the energy range below 300 eV could be obtained from the measurements. Thus it is still an open question whether the backscattered intensity is zero or finite at zero energy. The occurrence of the maximum can be understood qualitatively as follows(15). While a particle is penetrating into a solid its energy decreases, and because of the increasing cross section for backscattering, the backscattering intensity increases at low energies of the emerging particles. At the same time the probability for a second large angle deflection increases, removing particles from the observed direction the maximum is created by the competition of the two processes.

The maximum is also indicated by recent computer simulations(16,17). Fig. 15 shows a comparison of the calculations of Schäffler(16) with our experimental results for 5 keV H^+ backscattered from Nb. In Fig. 16 one result of J. E. Robinson(17) is compared with our experimental spectrum for 18 keV H^+ backscattered from Mo. Both calculations show the maximum observed in these experiments. At higher energies the calculations show comparatively more intensity than observed in the experiments.

The observed charged fractions were not as reproduceably as estimated from counting statistics, as the error bars in Fig. 8 indicate. To clarify this, further measurements on well defined metal surfaces are necessary. The measured charged fractions follow the trend observed at higher energies(18,19) quite well. They also agree with data given by Zaidins(20), and with those of Berkner et al.(21) for D^+ ions when compared at equal velocities. These investigations(18,19) however, show no dependence on the angle of emergence ß.

...11

ACKNOWLEDGEMENTS

The authors are indebted to the PULSATOR group of the IPP for making available the neutral particle analyzer used in these measurements. Thanks are due to R. Behrisch for his interest in and encouragement of this work. We also thank R. Hippele and S. Schrapel for technical assistance and P. Martin for programming the computer. Finally thanks are due to J. W. Müller (ORNL) for a critical reading of the manuscript and correcting the English.

... 12

LITERATURE

/1/ P. Meischner, H. Verbeek, J. Nucl. Mat. 53, 276 (1974)

/2/ E. R. Cawthron, D. L. Cotterel, and M. Oliphant, Proc. Roy. Soc. Lond. A 319, 435 (1970)

/3/ K. Morita, H. Akimume, T. Suita, Jap. Journ. Appl. Phys. 7 , 916 (1968)

/4/ G. H. McCracken, N. J. Freeman, J. Phys. B, 2 , 661 (1969)

/5/ W. Eckstein, H. Verbeek, J. Vac. Sci. Techn. 9 , 612 (1972)

/6/ E. S. Mashkova, V. A. Molchanov, Rad. Eff. 13, 131 (1972)

/7/ W. Eckstein, H. Verbeek, Vacuum 23, 159 (1973), Report IPP 9/7 (1972)

/8/ G. F. Girard, G. Monnom, F. P. G. Valckx (TFR-Group) private communication

/9/ C. F. Barnett, J. A. Ray, Nucl. Fusion 12, 65 (1972)

/10/ S. K. Allision, Rev. Mod. Phys. 30, 1137 (1958)

/11/ A. Egidi, R. Marconero, G. Pizella, Rev. Sci. Instr. 40, 88 (1969)

/12/ S. Datz, W. Eckstein, H. Verbeek (to be published)

/13/ R. Sizmann, private communication

/14/ H. Verbeek to be published

/15/ R. Behrisch, private communication

/16/ H. G. Schäffler, private communication

/17/ J. E. Robinson and S. Agamy, Proc. 5th Intern. Conf. Atomic Collisons in Solids, Gatlinburg, Tenn., Sept. 24-28, 1973, and private communication.

/18/ R. Behrisch, W. Eckstein, P. Meischner, B. M. U. Scherzer, H. Verbeek, Proc. 5th Intern. Conf. Atomic Collisions in Solids, Gatlinburg, Tenn., Sept. 24-28, 1973

/19/ T. M. Buck, L. C. Feldman, G. H. Wheatley, Proc. 5th Intern. Conf. Atomic Collisions in Solids, Gatlinburg, Tenn., Sept. 24-28, 1973

/20/ C. Zaidins cited by I. B. Marion, F. C. Young in Nuclear Reaction Analysis, Amsterdam 1968.

/21/ K. H. Berkner, I. Bornstein, R. V. Pyle, J. W. Stearns, Phys. Rev. A6, 272 (1972)

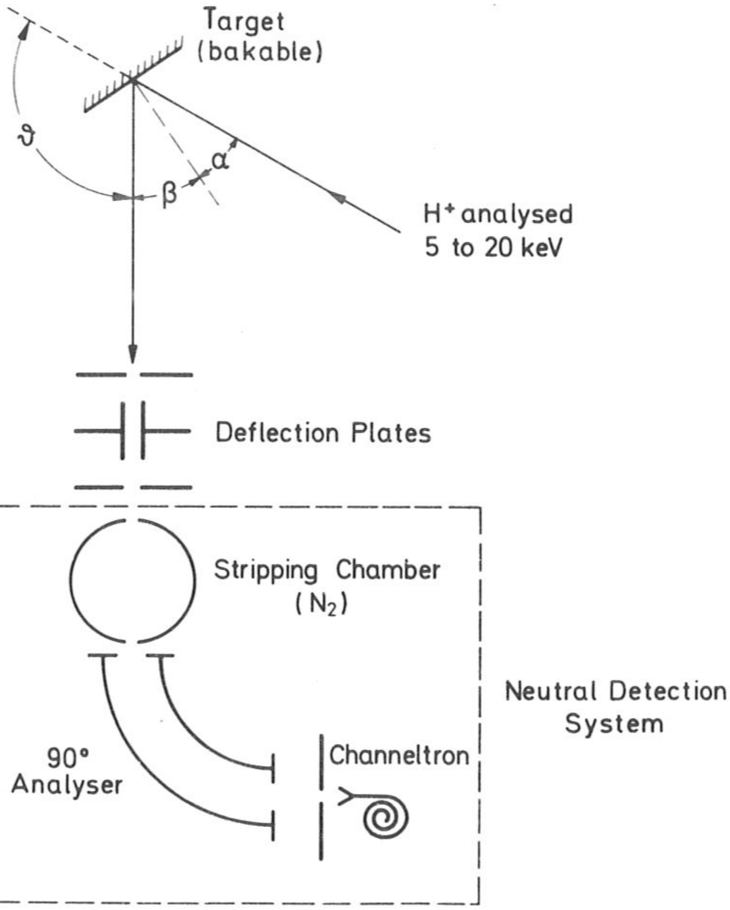

Fig. 1 Experimental set up (schematic)

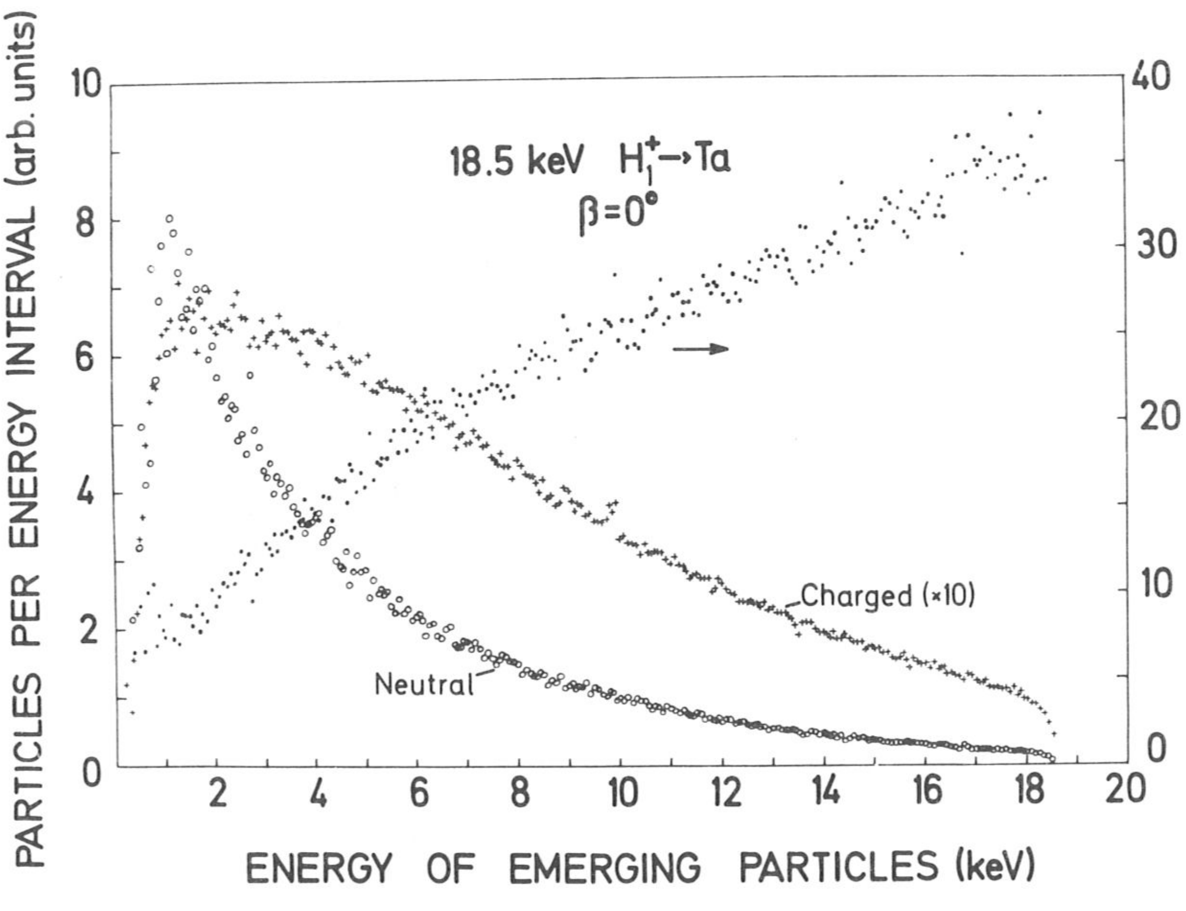

Fig. 2 Energy distributions of neutral (o) and positively charged (+) particles backscattered from Ta when bombarded with 18.5 keV protons. The charged fraction (.) is indicated by the right hand ordinate scale (computer plot).

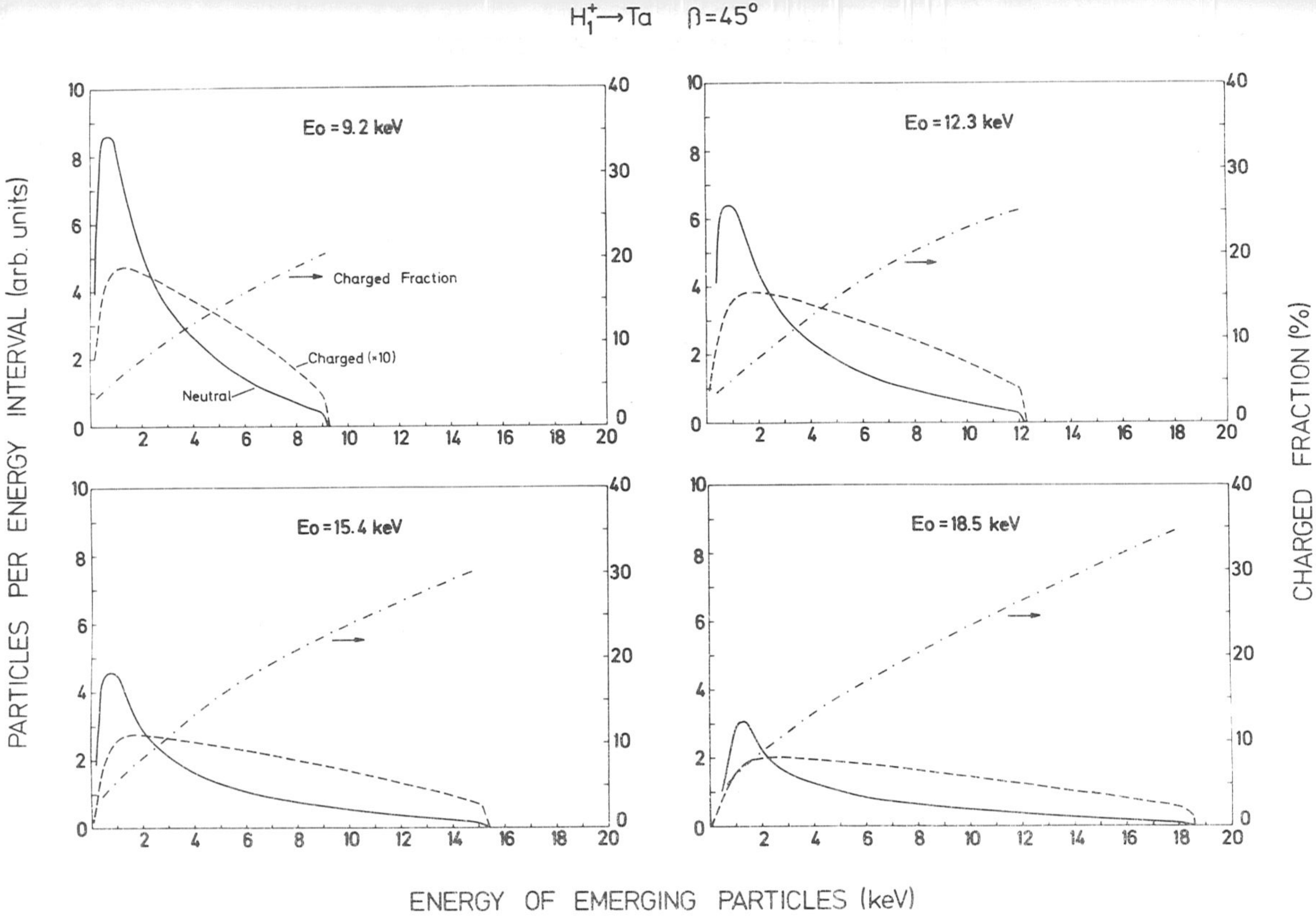

Fig. 3 Charged fraction (- . - .) and energy distribution of charged (- -) and neutral (——) particles backscattered from Ta at ß = 45° for primary energies E_o = 9.2, 12.3, 15.4, and 18.5 keV.

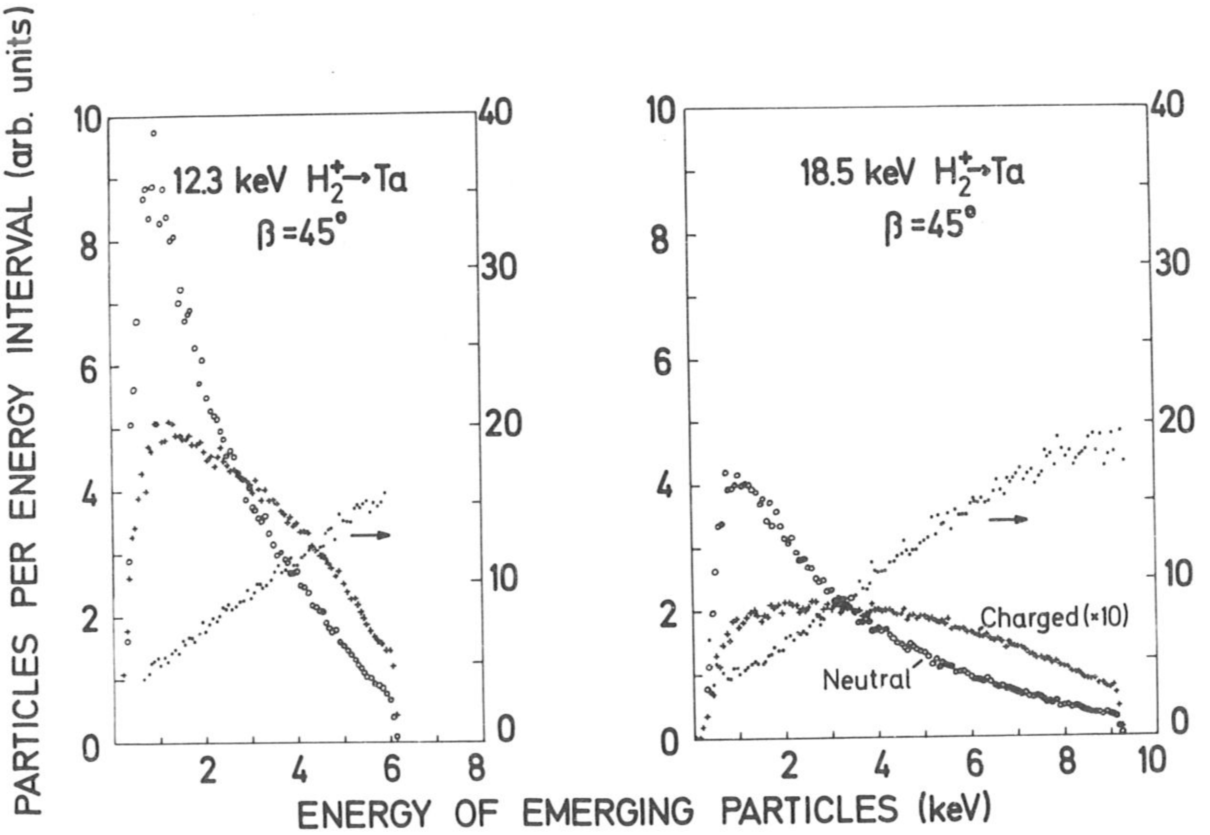

Fig. 4 Energy spectra and charged fractions of particles backscattered from Ta when bombarded with H_2^+ molecules of 12.3 and 18.5 keV (plotted as in Fig. 2).

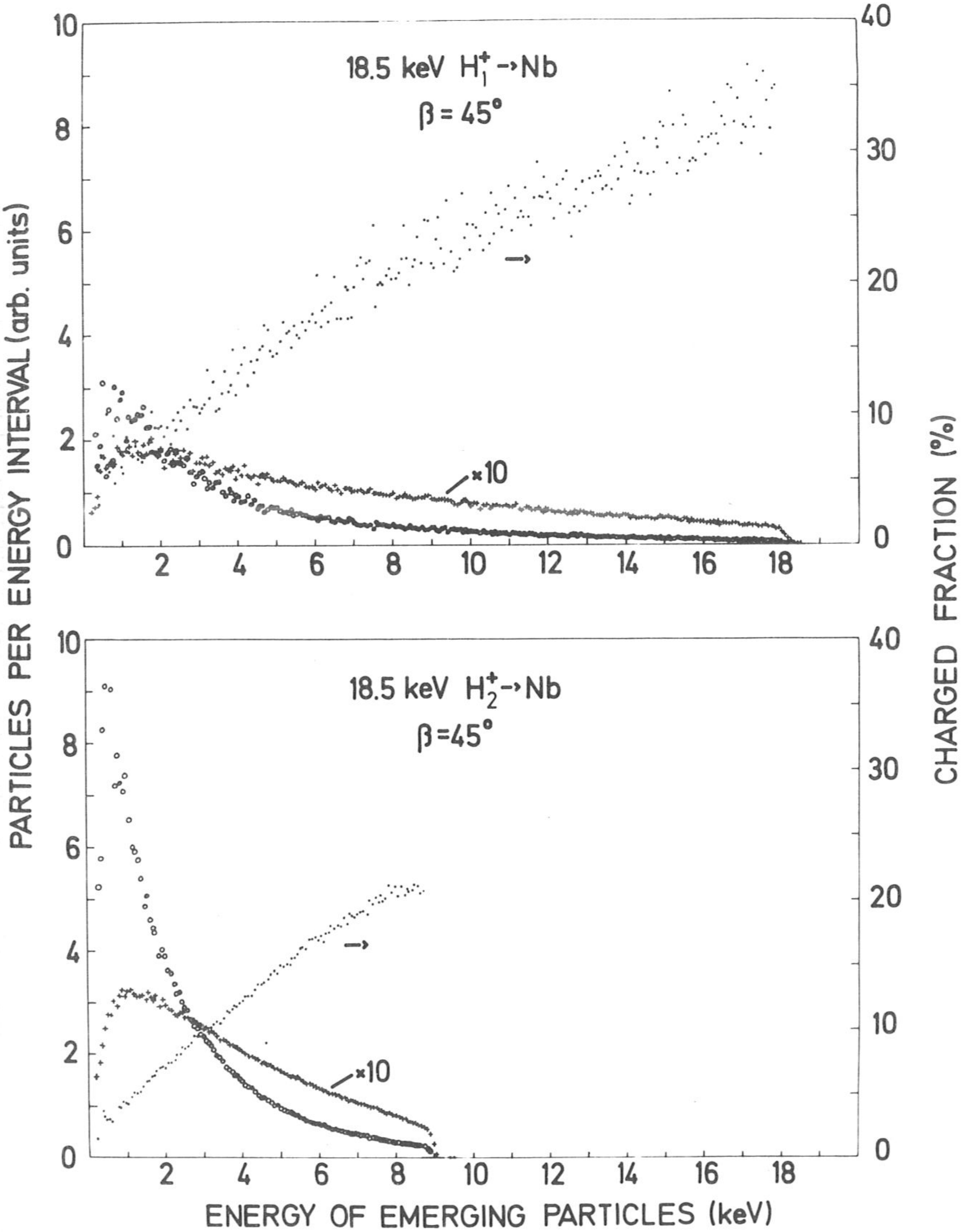

Fig. 5 Energy distributions and charged fractions of particles backscattered from Nb when bombarded with 18.5 keV protons (top) and H_2^+ molecular ions (plotted as in Fig. 2.).

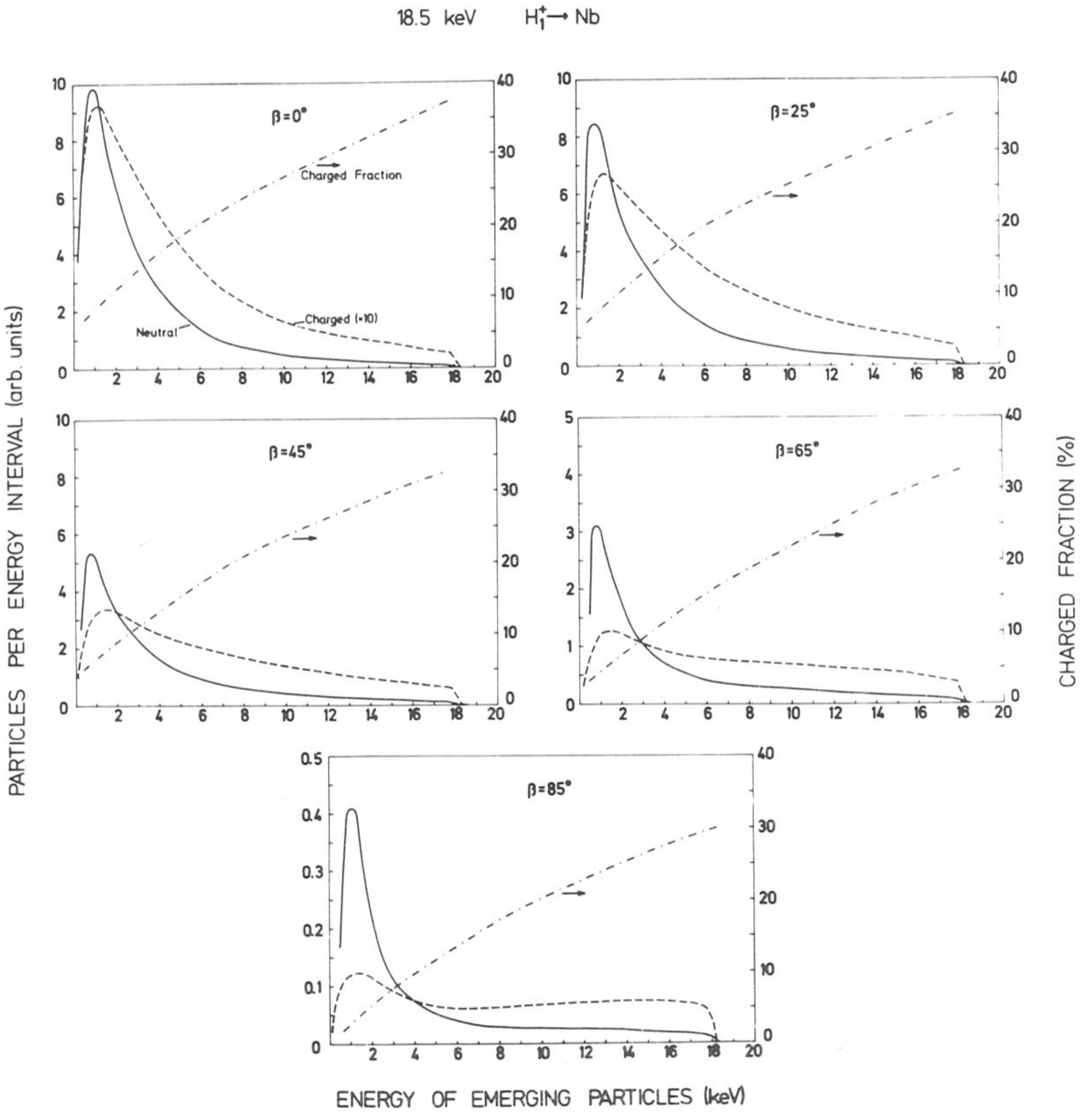

Fig. 6 Energy distributions and charged fractions of particles backscattered from Nb when bombarded with 18.5 keV protons for angles of emergence ß = 0, 25, 45, 65, and 85° (Note different ordinate scales!).

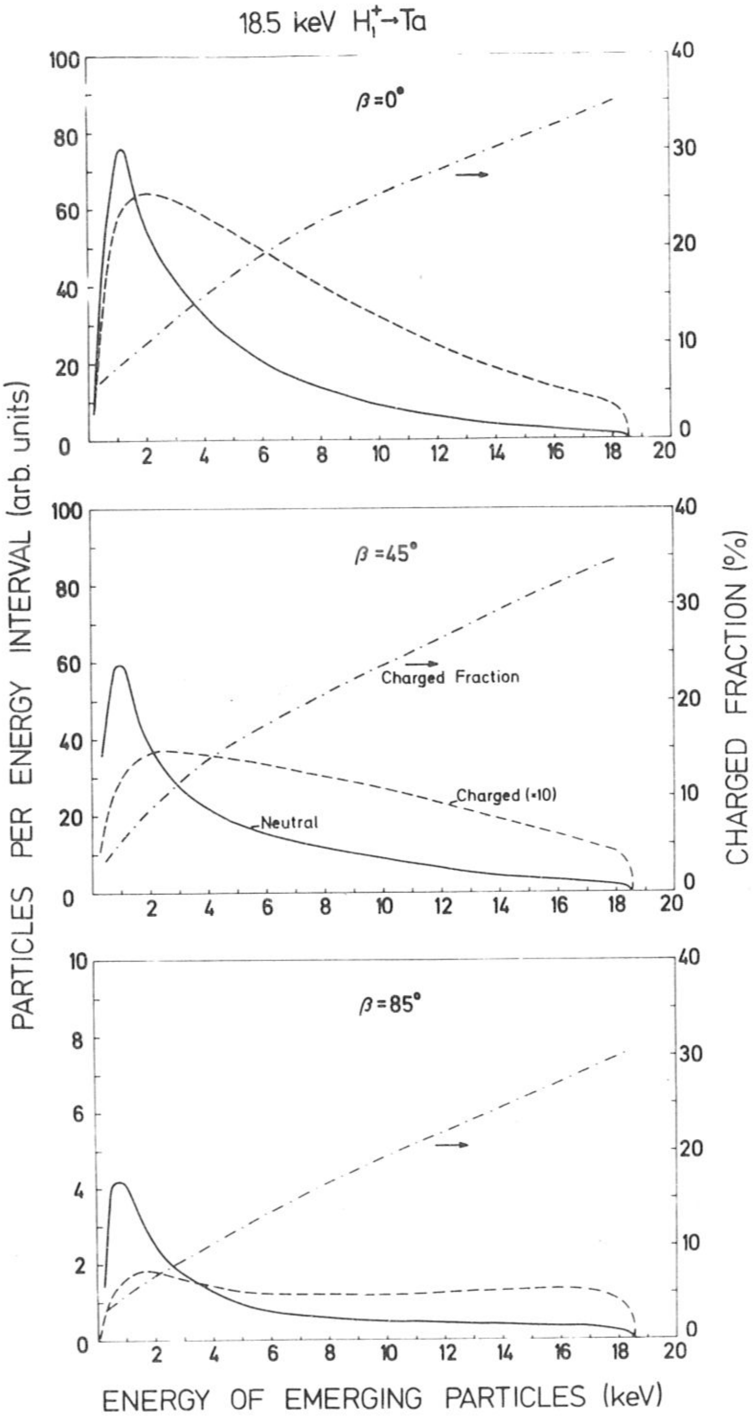

Fig. 7 Energy distributions and charged fractions of particles backscattered from Ta when bombarded with 18.5 keV protons for angles of emergence ß = 0, 45, and 85°. (Note different scales!).

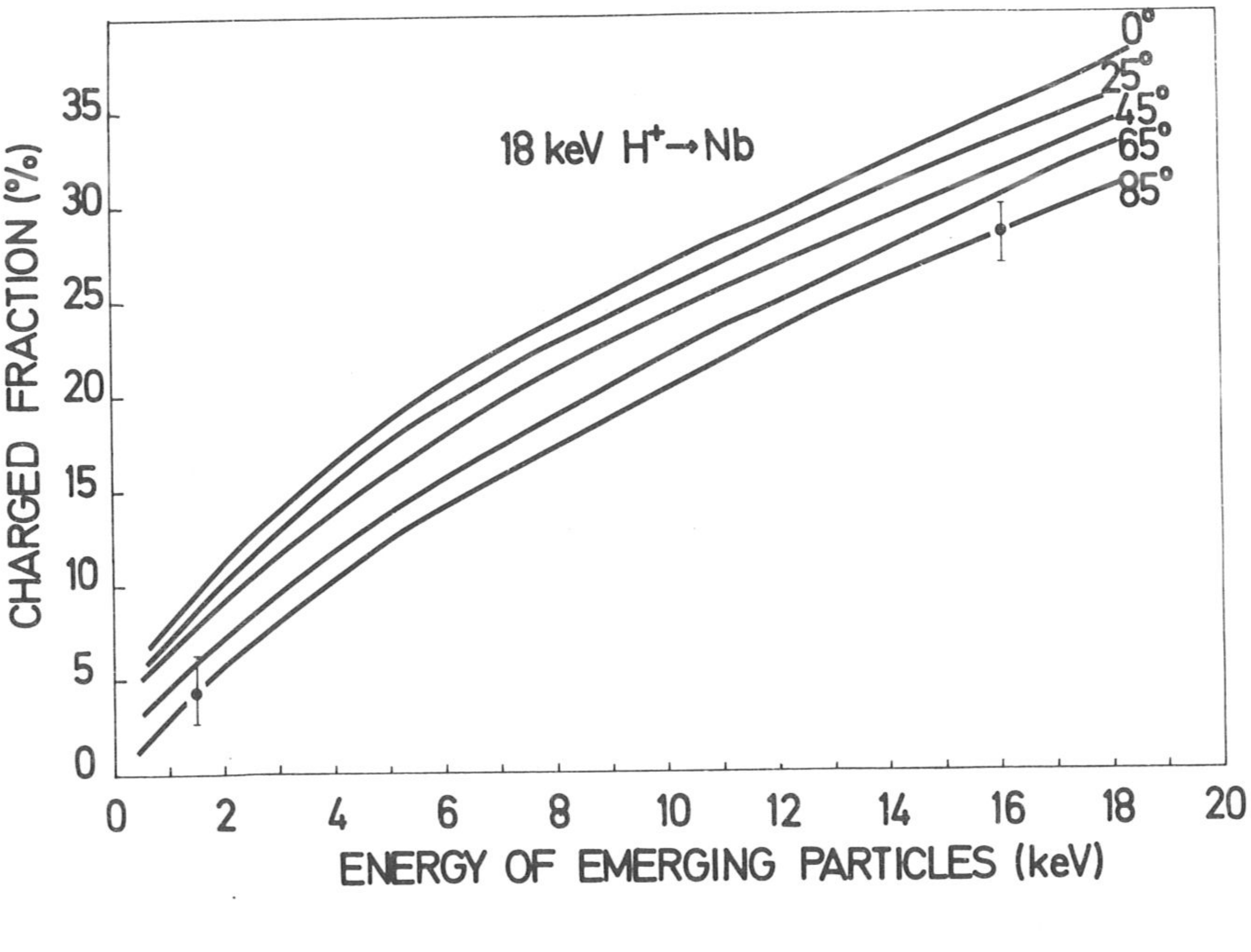

Fig. 8 Charged fraction $Q = N^{+}/(N^{+} + N^{0})$ of particles backscattered at different exit angles ß from Nb bombarded with 18.5 keV protons.

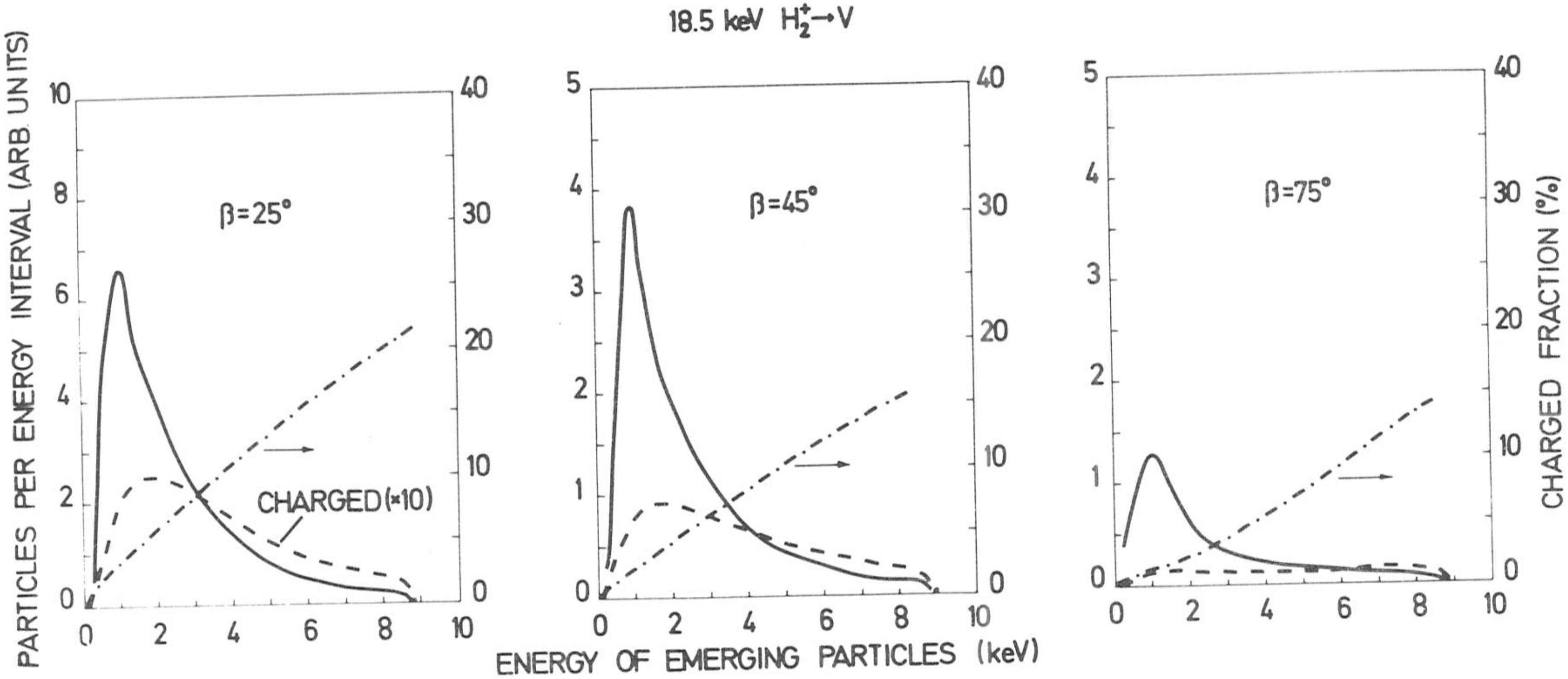

Fig. 9 Energy distributions and charged fractions of particles backscattered from Vanadium when bombarded with 18.5 keV H_2^+ ions for emergence angles ß = 25°, 45°, and 75°.

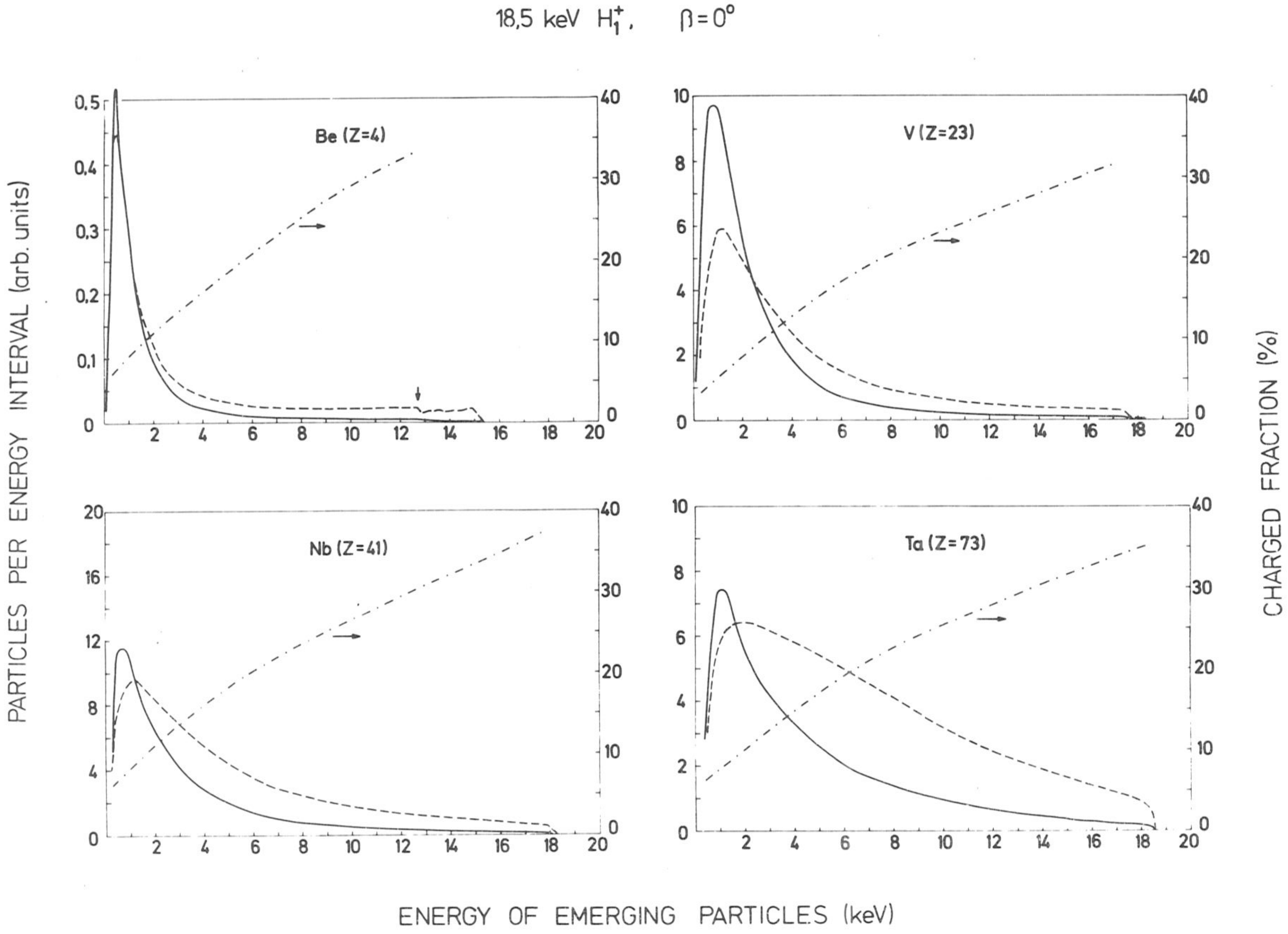

Fig. 10 Energy distribution and charged fractions of particles backscattered from Be, V, Nb, and Ta bombarded with 18.5 keV protons. (Note different ordinate scales!).

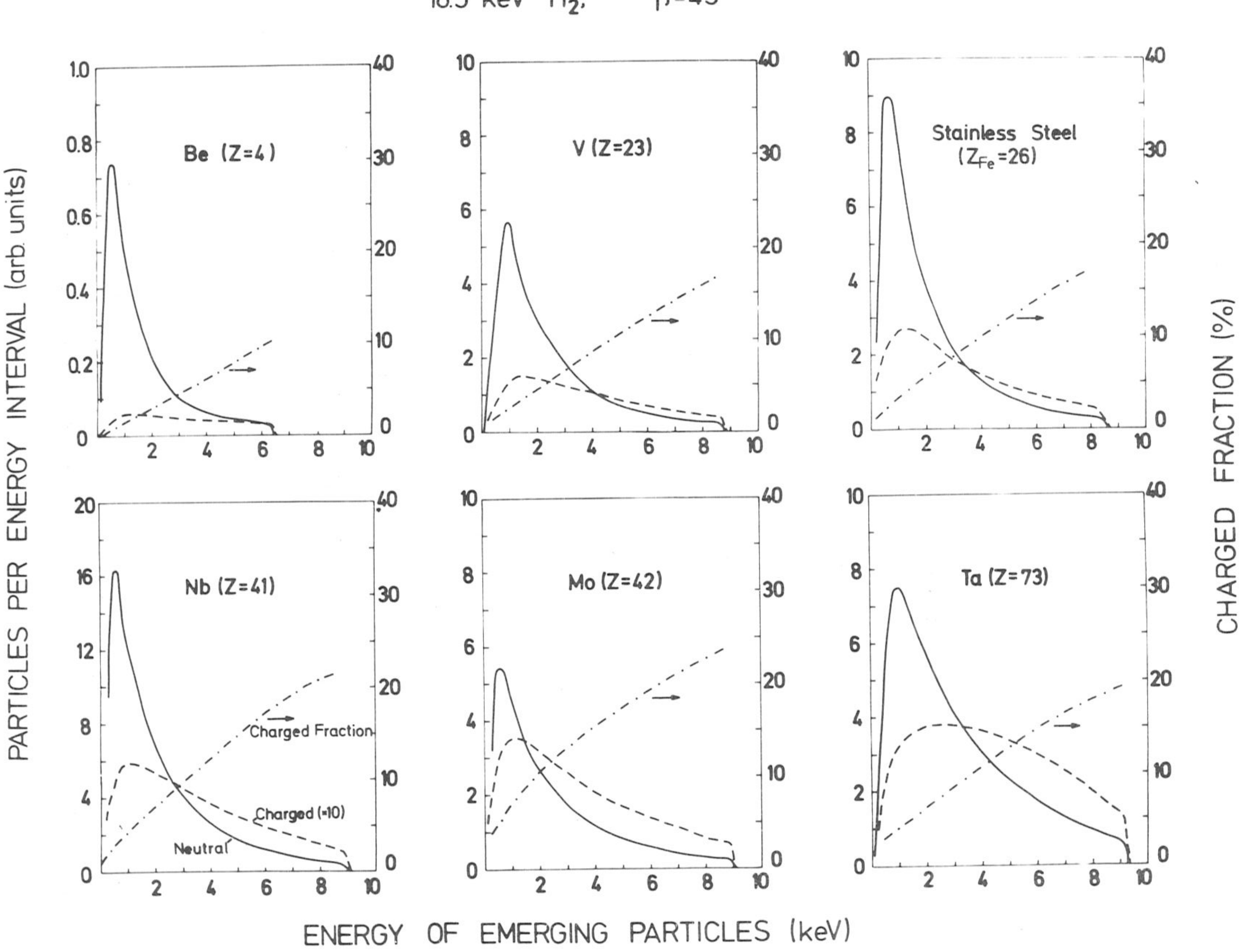

Fig. 11 Energy distributions and charged fractions of particles backscattered from Be, V, stainless steel, Nb, Mo, and Ta bombarded with 18.5 keV H_2^+ ions.(Note different ordinate scales !).

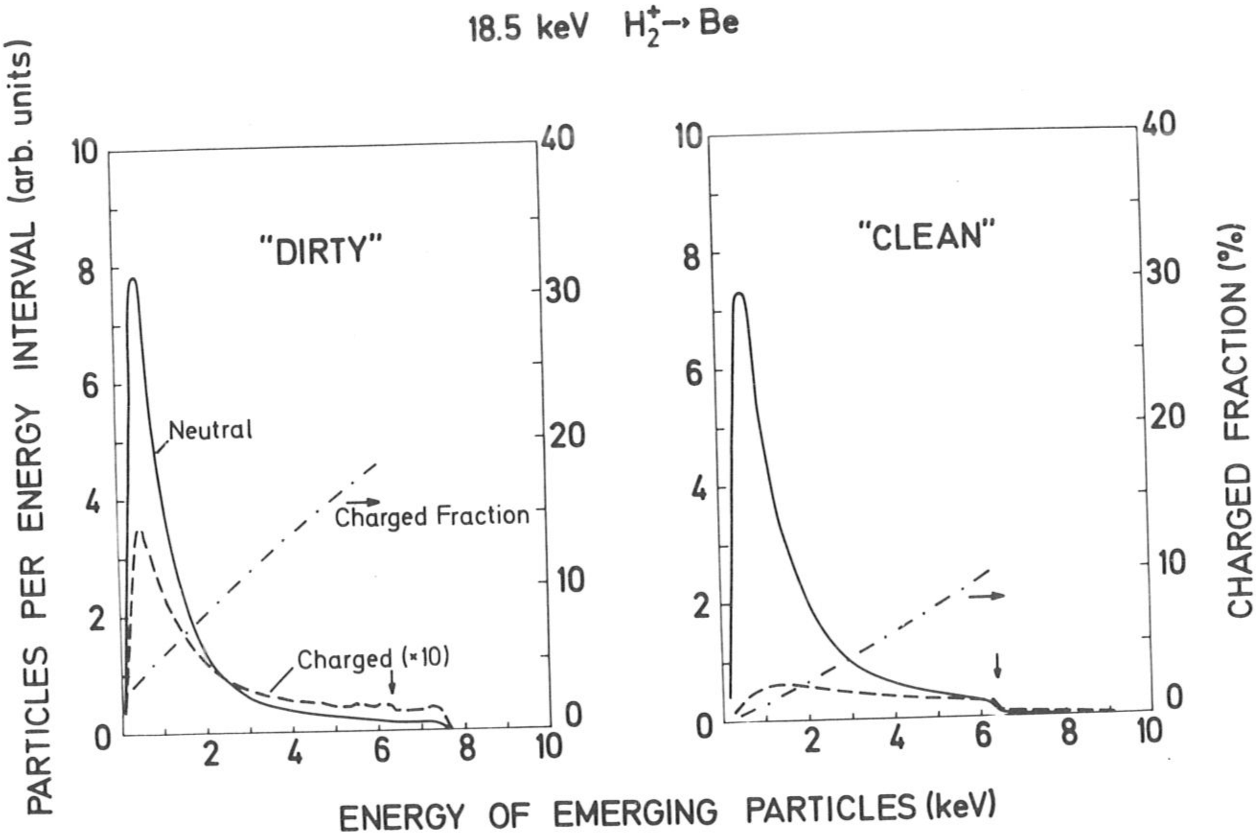

Fig. 12 Energy distributions and charged fractions of particles backscattered from Be bombarded with 18.5 keV H_2^+ ions. Left: after heating to 900°C, right: after alternate heating and sputtering with Ne ions.

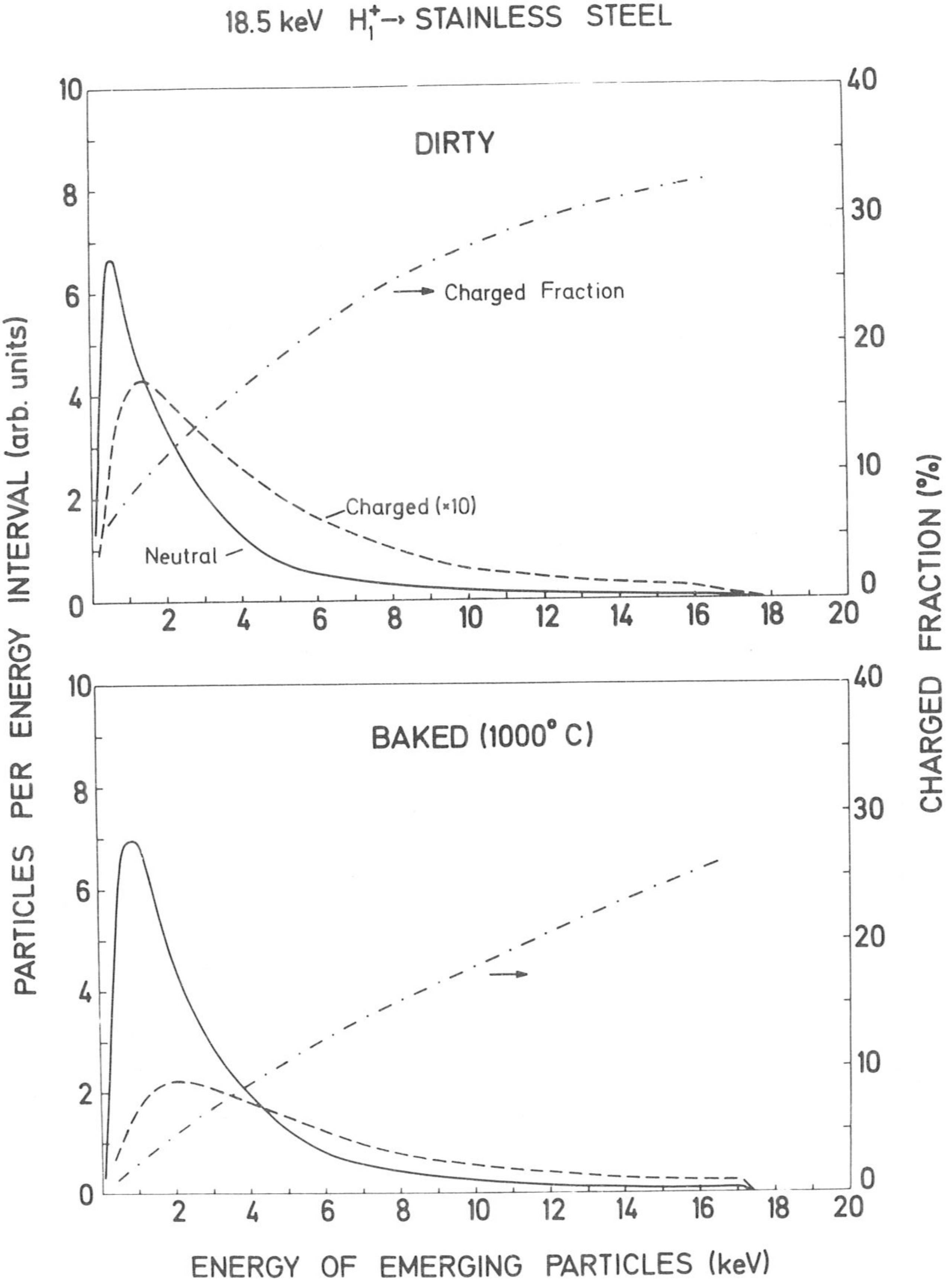

Fig. 13 Energy distributions and charged fractions of particles backscattered from stainless steel bombarded with 18.5 keV protons. Top: as built in, bottom: after heating to 1000°C.

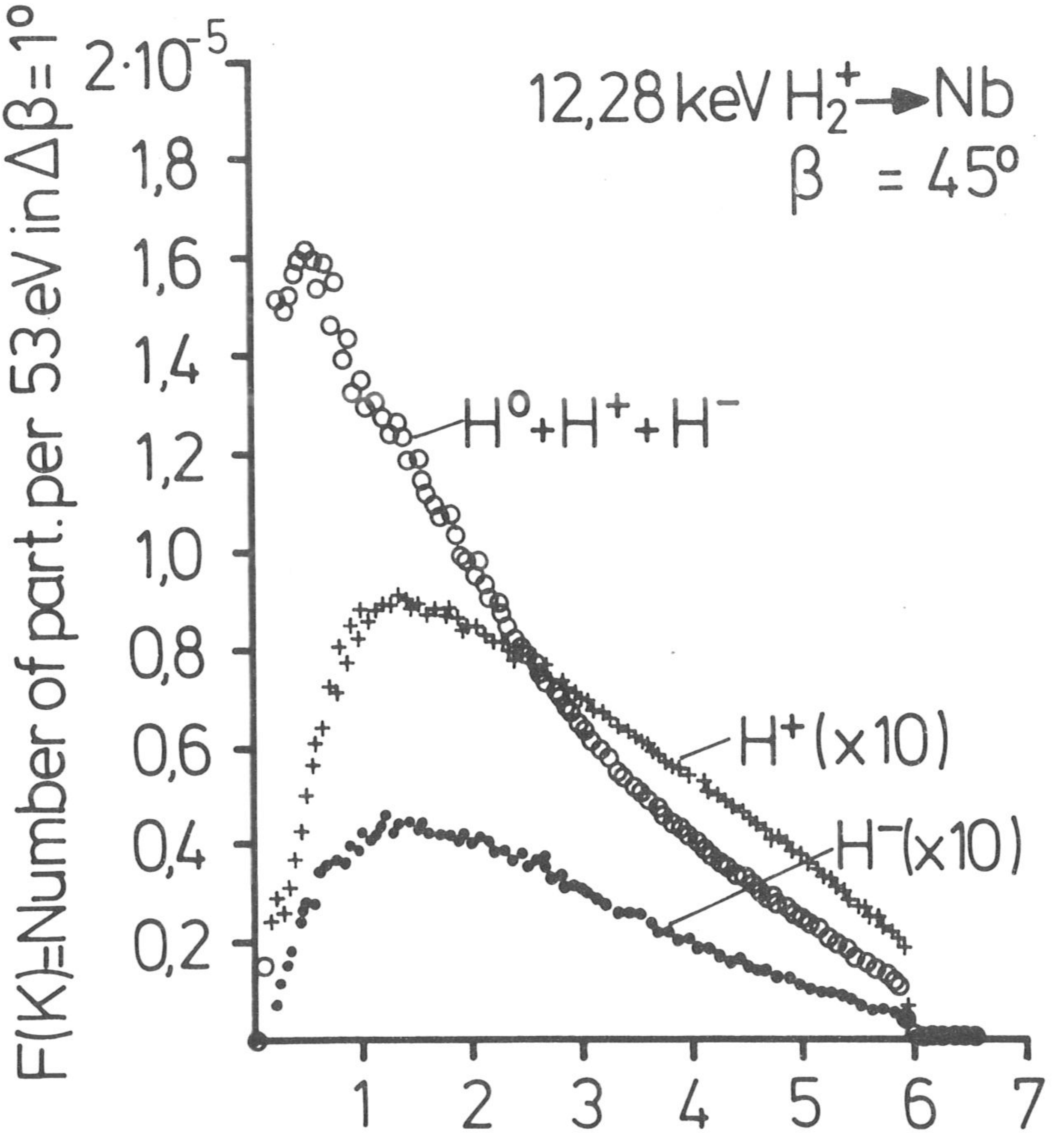

Fig. 14 Energy distributions of positively and negatively charged and the total number of particles backscattered from Nb, when bombarded with 12.28 keV H_2^+ ions.

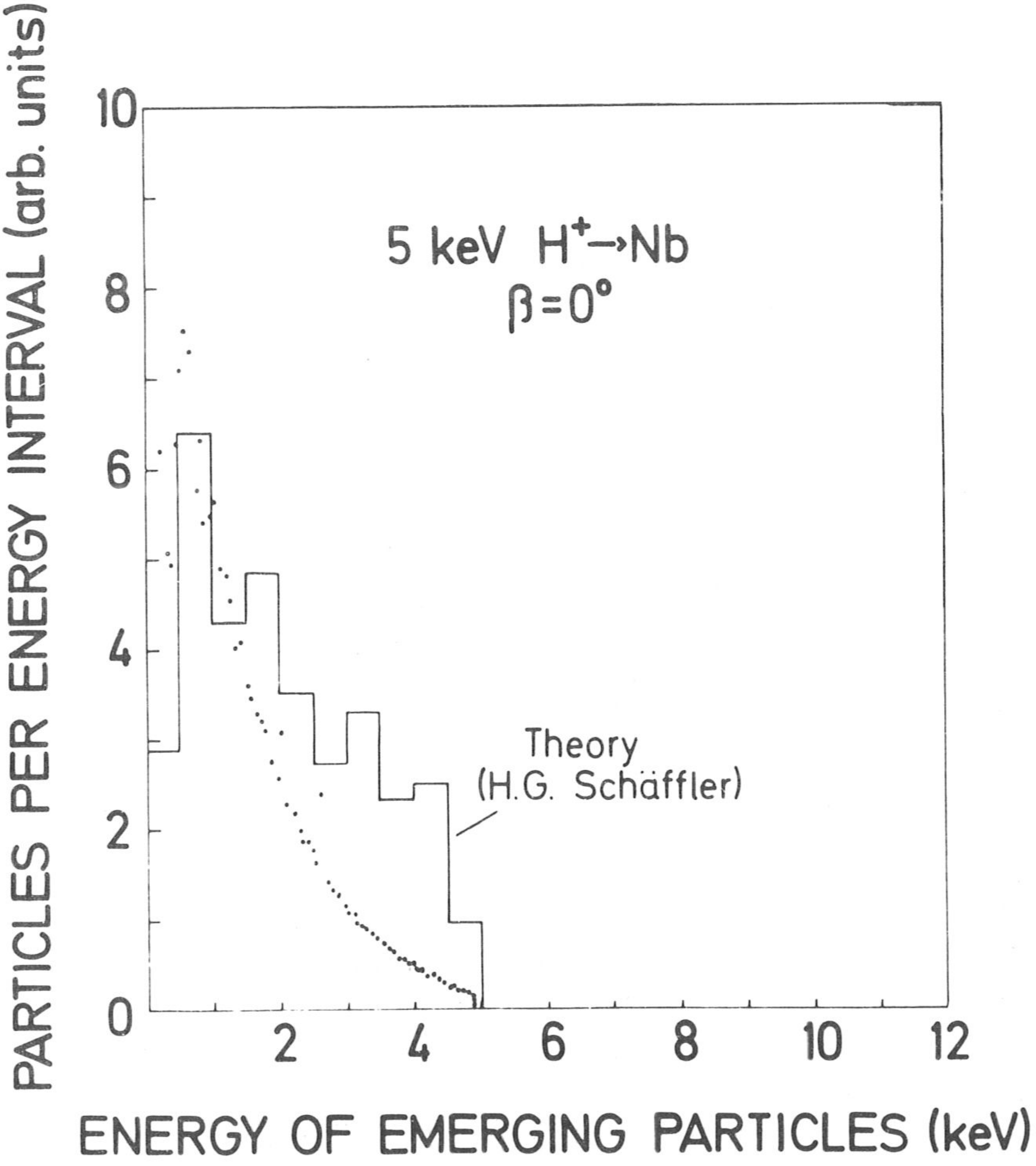

Fig. 15 Comparison of an experimental energy distribution (...) of backscattered particles with the calculations of Schäffler(16).

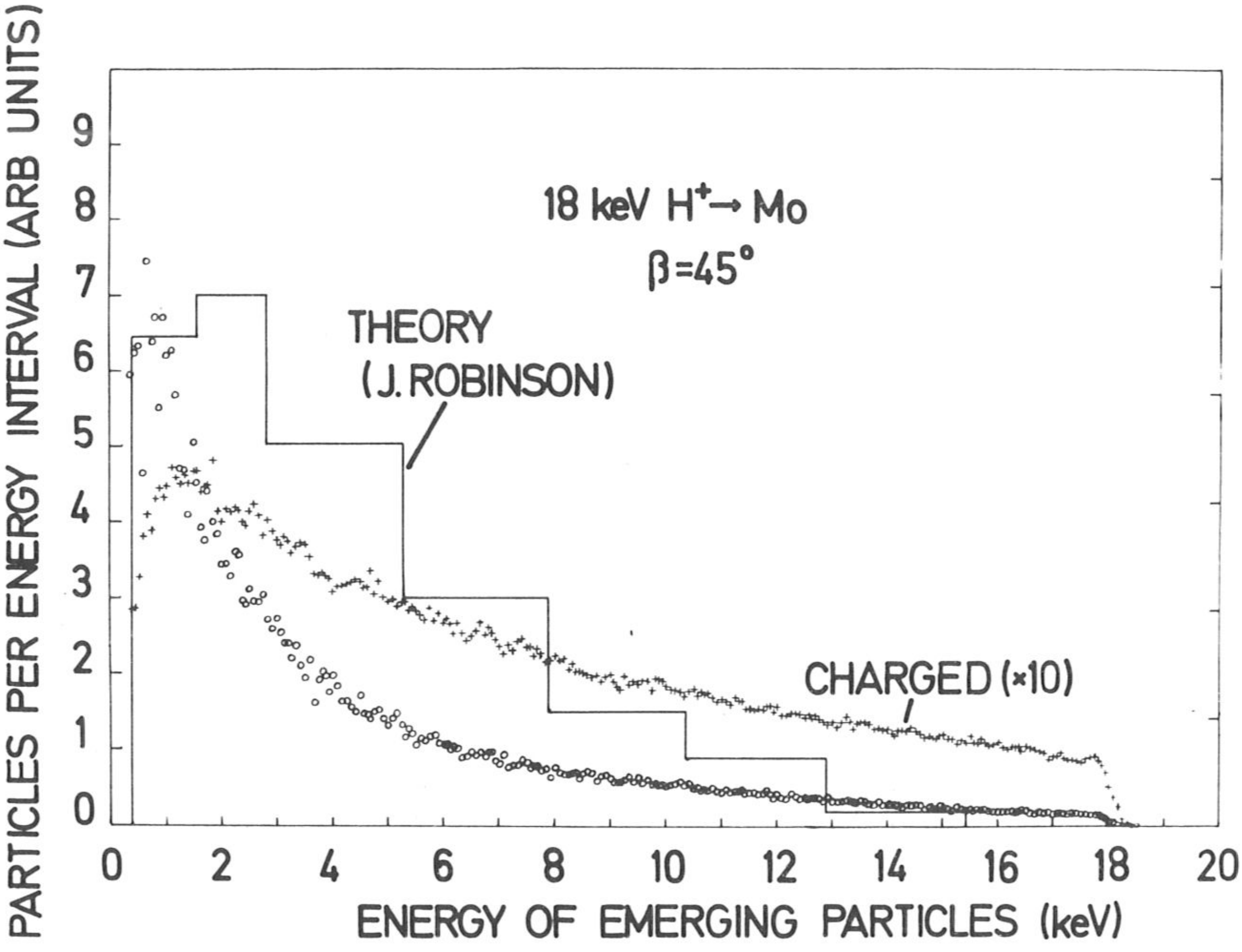

Fig. 16 Comparison of an experimental energy distribution (ooo) of backscattered particles with the calculation of J. E. Robinson(17).

This IPP report is intended for internal use.

IPP reports express the views of the authors at the time of writing and do not necessarily reflect the opinions of the Max-Planck-Institut für Plasmaphysik or the final opinion of the authors on the subject.

Neither the Max-Planck-Institut für Plasmaphysik, nor the Euratom Commission, nor any person acting on behalf of either of these:

1. Gives any guarantee as to the accuracy and completeness of the information contained in this report, or that the use of any information, apparatus, method or process disclosed therein may not constitute an infringement of privately owned rights; or
2. Assumes any liability for damage resulting from the use of any information, apparatus, method or process disclosed in this report.