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SPECTROSCOPY OF QUASIMOLECULAR X RAYS WITH H-LIKE DECELERATED IONS*

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Abstract

The quasimolecular $2p\pi-1s\sigma$ transition energy as a function of the internuclear distance is obtained from the interference structure observed in quasimolecular K-x-ray spectra from low energy H-like projectiles measured at certain impact parameters.

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It is well established that the formation of quasimolecular orbital plays an important role for the excitation process in nearly symmetric ion-atom collisions /1/. The electronic excitation proceeds mainly by electron promotion via coupling of close lying quasimolecular states. The coupling strength is strongly dependent on the energy gap ΔE_{if} between the quasi-molecular orbitals i and f, where ΔE_{if} is generally varying with the internuclear separation (R) of both colliding nuclei. Therefore it is crucial for the theoretical description of the excitation process to have precise information on this energy difference as a function of R. For inner shells in heavy ion-atom collisions there are several ways to determine the quasimolecular binding energies as function of the internuclear distance. Measuring the radiation emitted from the seperated collision partners is one attempt to derive total or differential cross sections for the excitation of different subshells and to compare these values with predictions made by theoretical models[2]. From this comparision thus indirect information on the ΔE_{if} values can be obtained which obviously depends on the accuracy (approximations) of the calculations. Furthermore, the experimental values mostly reflect the occupation long after the quasimolecular formation, when the vacancies decay in the separated collision systems. In addition possible fast cascading processes in outer shells make a clean determination of the primary quasimolecular excitation and therefore of ΔE_{if} rather difficult.

It was expected[3] that the radiation emitted during the collision originating from transitions between quasimolecula states, (quasimolecular radiation: MOR) would give a more direct access to these quasimolecular energy values ΔE_{if} (R) than the radiation from separated systems. In the quasistatic approximation[4] the measured photon energy E_x is assumed to be identic with the binding energy difference ΔE_{if} of the two participating orbitals. Here MO x rays with the energy E_x can be emitted only at given internuclear distance $R(E_x)$. For a fixed impact parameter b, the emission probability $\Delta P_{if}(b,E_x)/\Delta E_x$ per photon energy intervall (assuming that the orbital has <u>one</u> vacancy) is then given by:

$$\frac{\Delta P_{if}(b,E_x)}{\Delta E_x} = 2 \cdot \frac{\Delta R(E_x)}{v_R} \cdot A(E_x) \quad , \qquad (1)$$

where v_R is the radial component of the ion velocity and $\Delta R/v_R$ corresponds to that time intervall, in which photons in the energy range E_x to $E_x - \Delta E_x$ can be emitted. $A(E_x)$ denotes the radiactive transition rate. The factor of two reflects the possibility of a decay both on the incoming as well as on the outgoing part of the trajectory.

In the following we will discuss only transitions into the quasmolecular $f \mathrel{\hat{=}} 1s\sigma$ orbital, the innermost quasimolecular state. ($i = 2p\pi$ or $2p\sigma$). Within the framework of the quasimolecular picture at a given b the photon energy for $1s\sigma$-MOR extends from the $K_{\alpha/\beta}$-line of the separated systems up to the energy difference ΔE_{if} (R_{min}) between the orbitals i and f at the distance of closest approach R_{min}. Using equation (1) and calculated transition rates, $\Delta E_{if}(R)$ could thus be derived from measured continous MOR probabilities. Numerous experiments and theoretical studies of the $1s\sigma$-MOR have shown, however, that the quasistatic approximation is a rather crude one and not appropriate for describing MOR spectra in ion-atom collisions[5].

A more precise investigation of the dynamics of MOR shows, that the photon energy E_x is not anymore identic with $\Delta E_{if}(R)$. Any excess or deficit of x-ray energy can be obtained from the conversion of translational kinetic energy of the two colliding nuclei into photon energy, giving rise to the so called collision broadening. Furthermore, in nearly all experimental investigations so far "lowly charged" projectiles were used where $1s\sigma$ MOR could be emitted only if the $1s\sigma$ vacancy production $a_f(t)=a_{1s\sigma}(t)$ is strongly dependent on the collision time and also rather difficult to calculate. Weisskopf[6)] has shown that the spectrum for such a transition $i \rightarrow f$ can be calculated from the Fourier transform of the time-dependent dipole matrix element $D_{if}(R(t))$ (see also Anholt[7)] 1978).

Taking the dynamics of the collision into account one obtains for the emission probability

$$\frac{\Delta P_{if}(b,E_x)}{\Delta E_x} \sim \left| \int_{-\infty}^{+\infty} dt \cdot a_f(R) \cdot D_{if}(R) \cdot \exp\left(\frac{i}{\hbar} \cdot \int_0^t \left\{ E_x - E_f(R) + E_i(R) \right\} \cdot dt' \right) \right|^2, \quad (2)$$

where $R=R(b,t)$ is dependent on b and t. A detailed description of the dynamical theory is given in ref. 5.

The spectral shape of the $1s\sigma$ MOR for a given impact parameter is in clear contradiction to the quasistatic prediction but in agreement with this dynamical theory. The spectrum extends further than the maximal quasistatic transition energy at the distance of closest approach showing an exponential slope beyond this x-ray energy. The MOR spectrum is bare of any significant structure (see figure 1) and does not allow a reliable assignment between the measured photon energies E_x and $\Delta E_{if}(R)$.

In conclusion from the measured MOR emission probabilities for low charged fast ions, where the 1s vacancy is created at small R in the same collision, no quasimolecular spectroscopy could be performed so far. This result indeed is disappointing after the tremendous amount of work which has been done on this field[5] in the last decade.

There is, however, an access to a reliable quasimolecular spec-copy for the 1sσ orbital, if H-like ions are used as projectiles for these investigations. For H-like projectiles the transition to a given E_x therefore can occure on the incoming as well as on the outgoing part of the trajectory ("way-in" and "way-out").

Both transition amplitudes cannot be distinguished experimentally. Since for such slow heavy-ion collisions both amplitudes have a well defined phase relation they will interfere. For appropriate velocities the experimental spectrum should show a significant interference structure.

This interference structure allows a direct determination of the relevant phase differences which can be used for a determination of the energy gap of the involved orbitals i and f.

In figure 2 the "Two way" MOR decay process is illustrated. As function of R the two relevant quasimolecular orbitals are shown. For low velocities the transition with photon energy E_x will occur at $R_-(E_x,-t_0)$ and $R_+(E_x,+t_0)$. The transition amplitudes on the "way-in" $(-t_0)$ and "way-out" $(+t_0)$ are qualitatively shown in the lower part of the figure 2. With decreasing velocity (see stationary phase approximation)[5,8] photons with E_x will be emitted only in the region around R_- and R_+. The phase difference of the transition amplitude at $-t_0$ and $+t_0$ $\Delta\phi(E_x)$ can be determined from the time evolution of the wave functions of both involved molecular orbitals and of the dipole operator. Using the stationary phase approximation it can be shown, that the coherent addition of these two amplitudes with the phase difference $\Delta\phi(E_x)$ leads to an interference term in the emission probability which causes an oscillation in the shape of the MOR spectrum. This oscillation can be described by[8,9]

$$\frac{\Delta P_{if}(b,E_x)}{\Delta E_x} \sim \cos^2\left(\frac{\Delta\phi(E_x)}{2} - \frac{\pi}{4}\right) \quad , \qquad (3)$$

where the phase difference $\Delta\phi_{if}$ is

$$\Delta\phi_{if}(E_x) = \frac{2}{\hbar}\cdot\int_0^{t_o}(E_x - \Delta E_{if}(R))\,dt' \quad . \qquad (4)$$

The constant phase term $\pi/4$ is explained in ref. [8,9]. From the measured spectra, experimental $\Delta\phi$ values as function of E_x can now be determined. Using equation (4) a simple quantitative relation between E_x and R can now be derived. In equation (4), however, all $R_{min} \leq R \leq R(E_x)$ contribute to the phase integral making the assignment between E_x and $R(E_x)$ rather difficult. However, from the derivative of $\delta\Delta\phi/\delta E_x$ in a simple direct way $E_x = E_x(R) = \Delta E_{if}(R)$ can be determined. This derivative yields

$$\frac{\delta\Delta\phi(E_x)}{\delta E_x} \approx \frac{2}{\hbar}\cdot t_o(E_x) \quad , \qquad (5)$$

where $2 * t_0$ is the transit time along the classical trajectory from the position $-R(E_x)$ to $+R(E_x)$. The transit time can be easily converted into the path length S or into the internuclear separation $R(E_x)$ assuming a Rutherford trajectory. Simple measurements of the oscilliatory structure of the MOR spectra at a given impact parameter b therefore yield the x-ray energy dependence of $\Delta\phi$ and from this straight forward way ΔE_{if} can be determined i.e. spectroscopic information on these transiently formed quasimolecular orbitals can be obtained only on the basis of quantummechanically well established phase relationships. For the observation of at least one or more oscillations in the MOR emission probabilities, H-like projetiles are needed with velocities $v_p \lesssim 0.2 v_K^{UA}$ where v_K^{UA} is the K electron velocity in the united atom (UA) system. Since presently no ion sources for H-like ions ($Z_p \gtrsim 16$) with sufficient intensity in combination with suitable accelerators are available, the only way to produce such beams is the so called accel-stripping-decel technique [10,11,12]. Ions are accelerated to such high velocities that the penetration of a thin C-foil produces a considerable fraction of one-electron ions (H-like). These ions are then decelerated to such low velocities that the above mentioned condition is fullfilled. One of the very well working accel-decel systems is the 4-stage-tandem facility of the Brookhaven Nat.Lab, where the Cl^{16+} on Ar collision system was investigated[10]. The S^{15+} on Ar system was measured with the Tandem - postaccelerator system of the MPI für Kernphsyik in Heidelberg[11]. Very recently the Ge^{31+} on Kr-system was investigated at the Unilac of GSI-Darmstadt[12].

To measure the MOR emission probabilites for a given b, the emitted photon ($\rightarrow E_x$) and the scattered projectile ($\longrightarrow b$) have to be detected in coincidence. The well collimated H-like beam ($\sim 10^8$ ions/sec.) hit a differentially pumped gas target where the pressure was kept low enough to avoid charge exchange of the beam before entering the central target area. The scattered projectiles were detected with a position sensitive parallel plate avalanche detector and the x-rays with a Si(Li)-detector.

The coincidence electronic is presented in ref. 13. The data were collected in " event mode". Because of the low beam intensity and target density the contributions of random coincidences in the MOR regime were negligible. In figure 3 an absorber-corrected coincidence spectra for Cl^{16+} on Ar is shown. In contradiction to figure 1 a clear oscillatory structure is observed. Because of the experimental difficulties (true coincidence rate:one per 10 min) and limited beam time the statistical error could not be any more reduced. For the same system in figure 4 the velocity dependence of this structure is presented. With decreasing velocity $\Delta\phi$ increases and more structure appears, in nice agreement with the scaling of the stationary phase approximation $(\Delta\phi \sim t \sim 1/v_p)$. Even for Ge^{31+} on Kr the oscillatory structure in the emission probability $\Delta P/\Delta E_x$ has been observed[14).

In figure 5 for Cl^{16+} on Ar for 2,5/5/10 and 20 Mev the positions of constructive (maxima) and destructive (minima) interference are shown. They follow nicely the scaling with v_p and b of the stationary phase approximation. Figure 6 depicts for $b \approx 1000$ fm the experimental phases times the velocitiy $(\Delta\phi \cdot v_p)$ for constructive and destructive interference (maxima and minima from figure 5). In agreement with equation (4) they scale well on one common curve. The derivative of this experimental curve yields directly the path length S (E_x). From this function $S=S(E_x)$ the $\Delta E_{if}(R)$ can be immediately determined.

Using only the "clean" experimental information of the maxima and the minima positions we obtain for equation (5) the following approximation

$$\delta \Delta \phi \text{ (max-min)} = \pi = \delta E_x \text{(max-min)} \cdot \frac{2}{\hbar} \cdot t_0(E_x) \quad . \qquad (6)$$

The measured δE_x (max-min) can be taken from figure 5. With these data the corresponding $t_0(E_x)$ values can be calculated and can then be converted into the internuclear separation $R(E_x)$ using a classical Coulomb trajectory. The so obtained $E_x=E_x(R)$ values are presented in figure 7 for Cl^{16+} on Ar. Independent of the projectile velocity they scale all on a common curve in agreement with the theory. The so derived transition energies are compared in figure 7 with calculated MO transition energies, using a two center potential Dirac Fock program for 26 electrons in the Cl - Ar collision system[15]. The solid line represents $2p\sigma - 1s\sigma$ and the dashed line the $2p\pi - 1s\sigma$ transitions. It can be seen that the calculated $2p\pi - 1s\sigma$ transition energies qualitatively agree with the measured R dependence but are slightly above the experimental values, even though the screening is somewhat overestimated. The predicted values for $2p\sigma - 1s\sigma$ transitions are clearly below the experimental values. This result is in agreement with the expected intensity contributions of both transitions into the $1s\sigma$ state[9].

In conclusion for H-like ions S^{15+} and Cl^{16+} on Ar and for Ge^{31+} on Kr, the experimental $1s\sigma$ MOR spectra[9,14] show a clear oscillatory structure originating from the interference of the transiton amplitudes on the incoming and outgoing parts of the trajectory.

From the interference structure information on the phase relationship of inner shell transition amplitudes can be obtained. From these experimentally determined phase differences, quasimolecular

$2p\pi - 1s\sigma$ transition energies could be determined. The method discussed above presents a unique possibility for spectroscopy of quasimolecular orbitals and might be even applicable in super heavy quasimolecules in not too far future.

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E_x and impact parameter b for 90 MeV Ni on Ni. The solid lines are to guide the eyes.

Fig. 2 Illustration of the "two-way" MO decay process with H-like projectiles

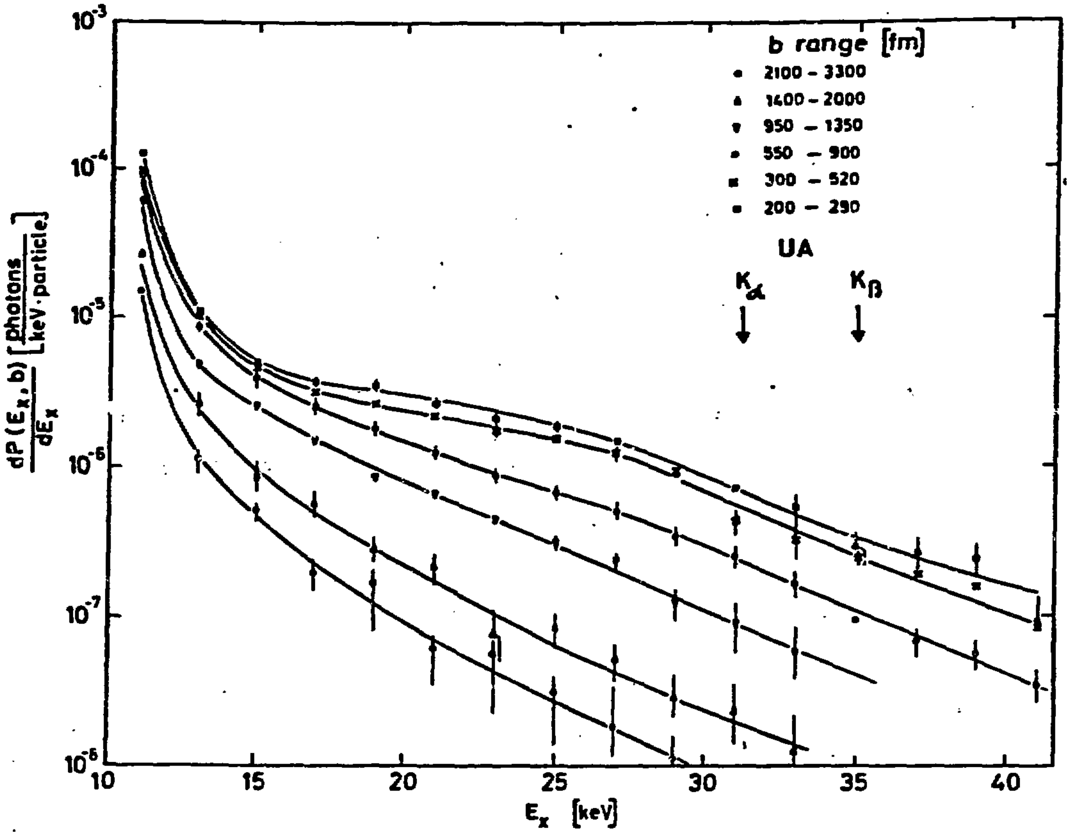
Fig. 3 Coincident absorber corrected $1s\sigma$ MOR-spectra for 2,5 MeV Cl^{16+} on Ar.

Fig. 4 Projectile velocity dependence of absolute $1s\sigma$ MOR-emission probabilities for Cl^{16+} on Ar.

Fig. 5 x-ray energies for constructive (solid lines) and destructive (dashed lines) interference for Cl^{16+} on Ar as function of projectile velocity and impact parameter.

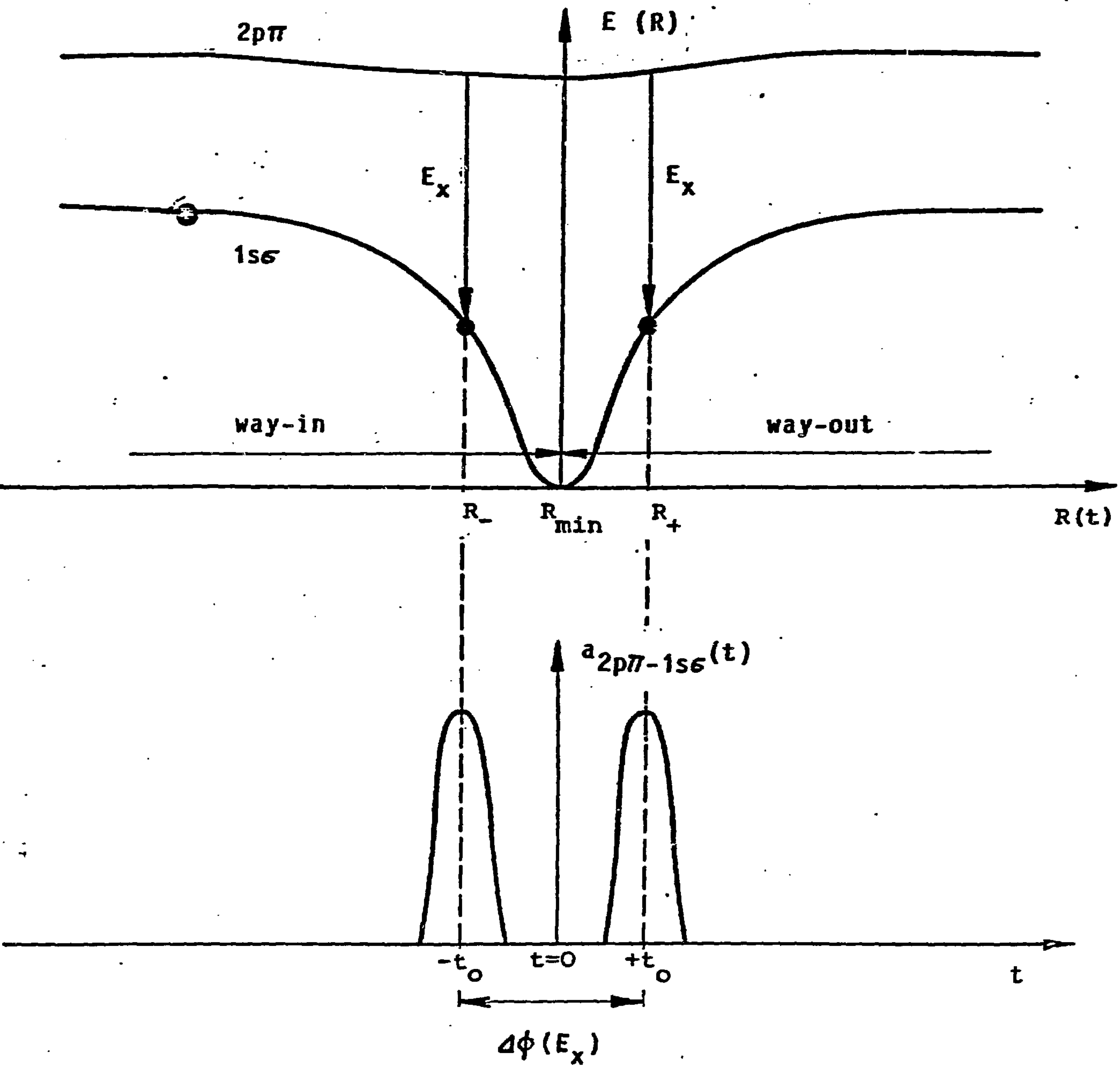
Fig. 6 Experimentally determined phase differences times projectile velocity ($\Delta\phi \cdot v_p$)(solid line). The dashed line represents the derivative of the path length (see text)

Fig. 7 Experimentally from interference structure derived $2p\pi - s\sigma$ energy differences as function of the internuclear separation. The curves represent theoretical calculation of the $2s\sigma - 1s\sigma$ (solid line) and the $2p\pi - 1s\sigma$ (dashed line) energy difference (se text)



b range [fm]
2100 - 3300
1400 - 2000
950 - 1350
550 - 900
300 - 520
200 - 290
UA
K_α
K_β
10^{-3}
10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-8}
$\frac{dP(E_x, b)}{dE_x}\left[\frac{\text{photons}}{\text{keV}\cdot\text{particle}}\right]$
10
15
20
25
30
35
40
E_x [keV]

Fig. 1



2pπ
E (R)
E x
E x
1sσ
way-in
way-out
R_
R min
R+
R(t)
a 2pπ-1sσ(t)
-t o
t=0
+t o
t
Δφ(E x)

Fig. 2

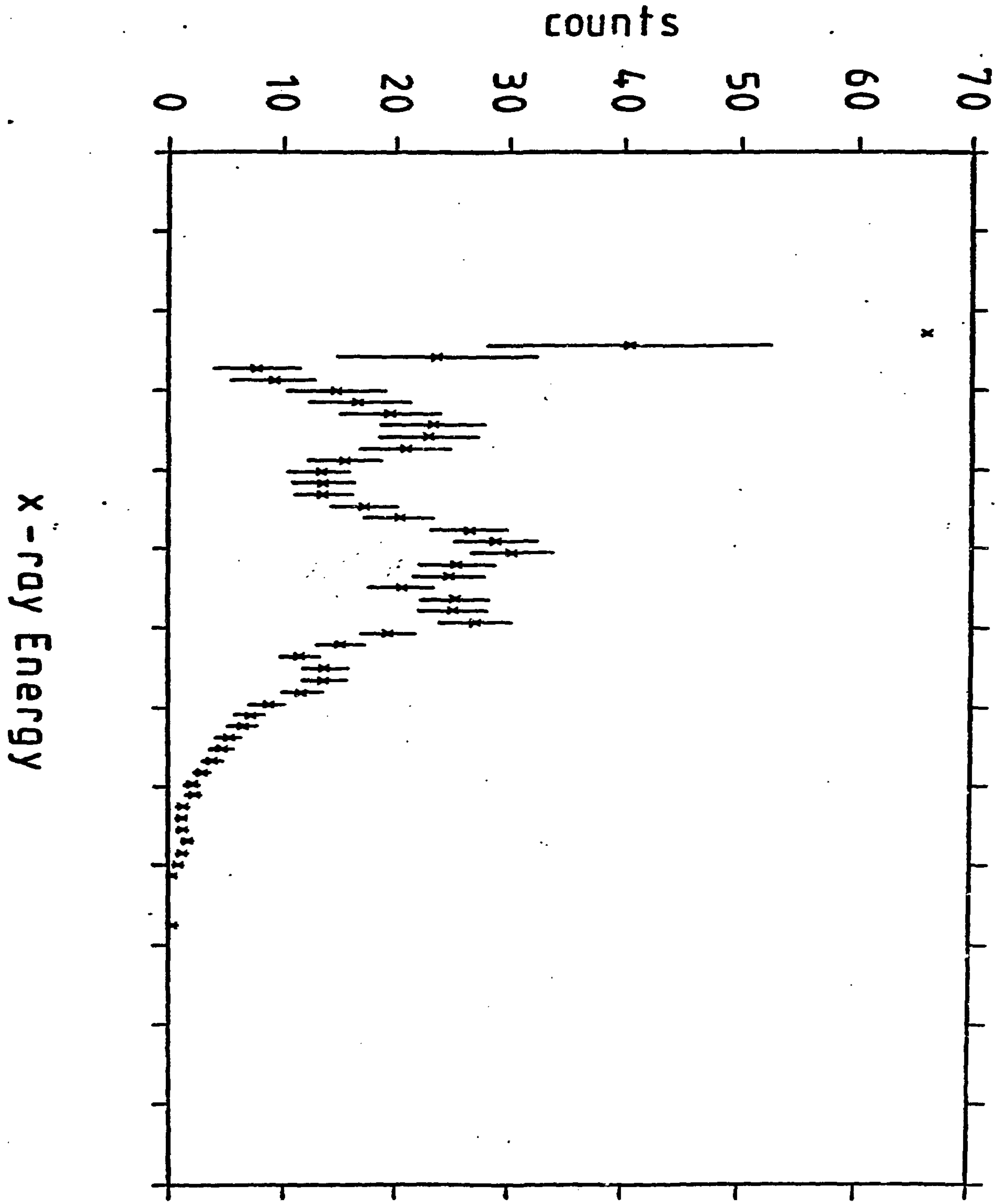
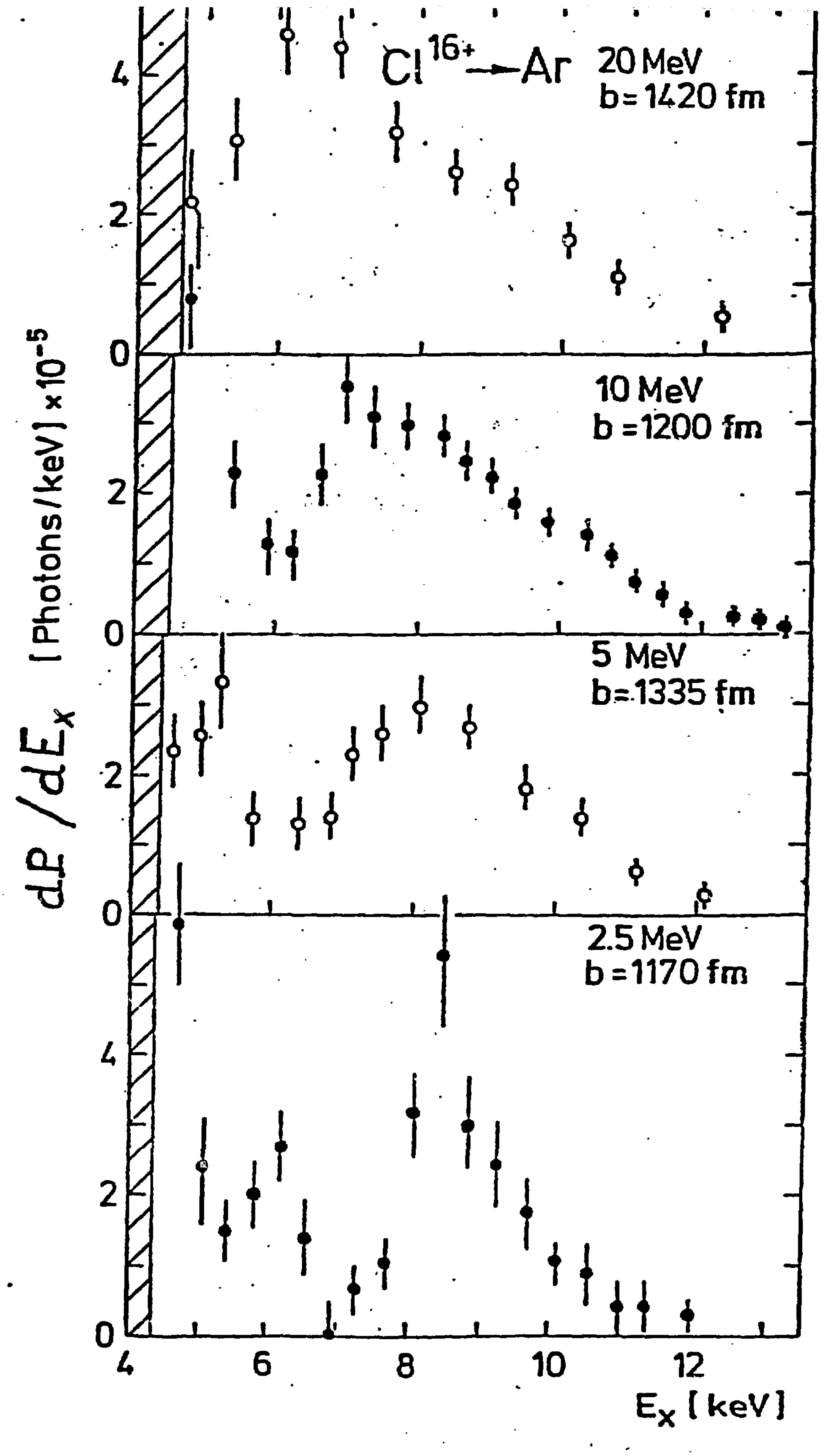


Fig. 3



$Cl^{16+} \to Ar$
20 MeV
b = 1420 fm
10 MeV
b = 1200 fm
5 MeV
b = 1335 fm
2.5 MeV
b = 1170 fm
dP / dE_x [Photons/keV] ×10^-5
E_x [keV]
0
2
4
4
6
8
10
12

Fig. 4

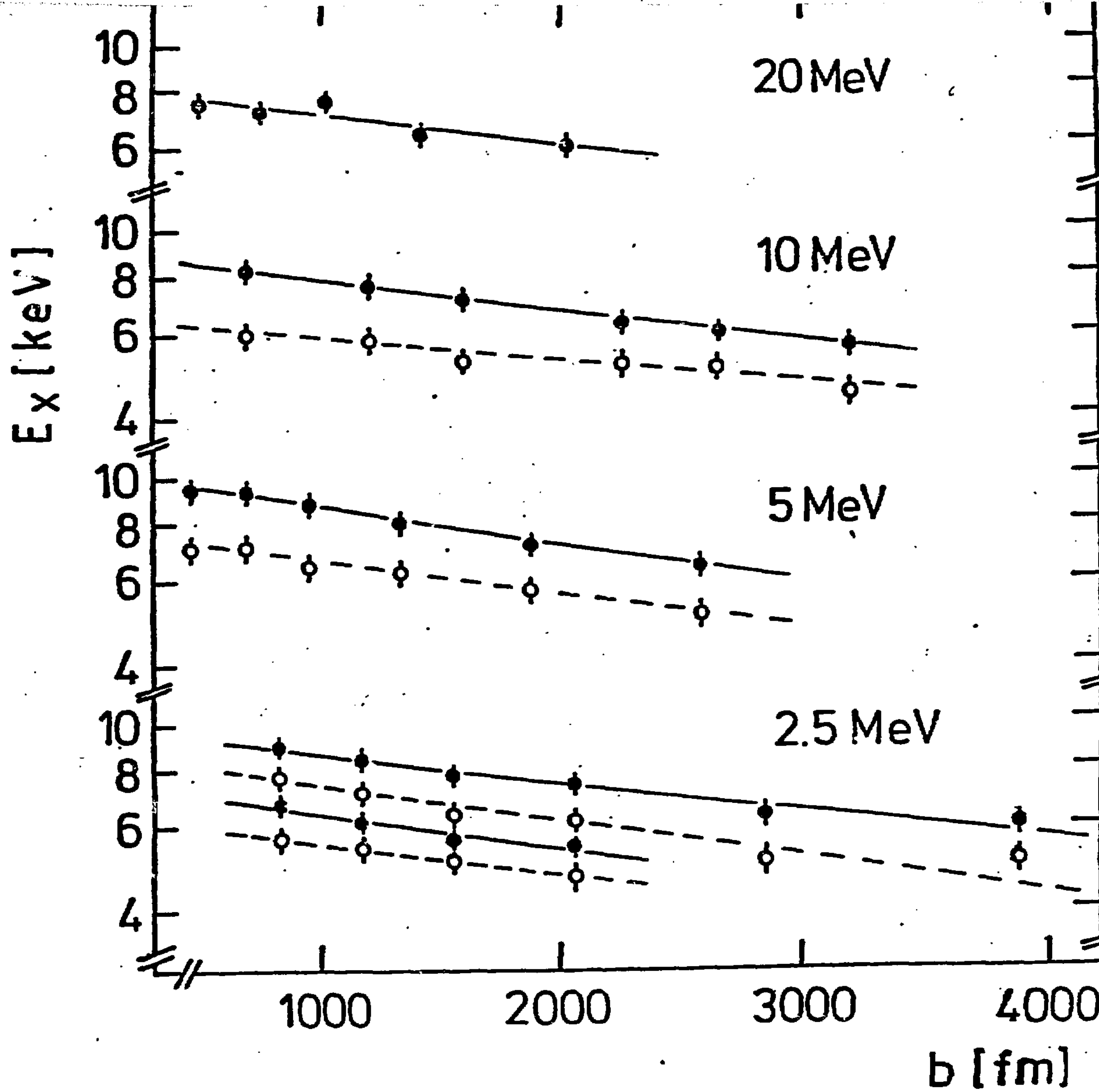
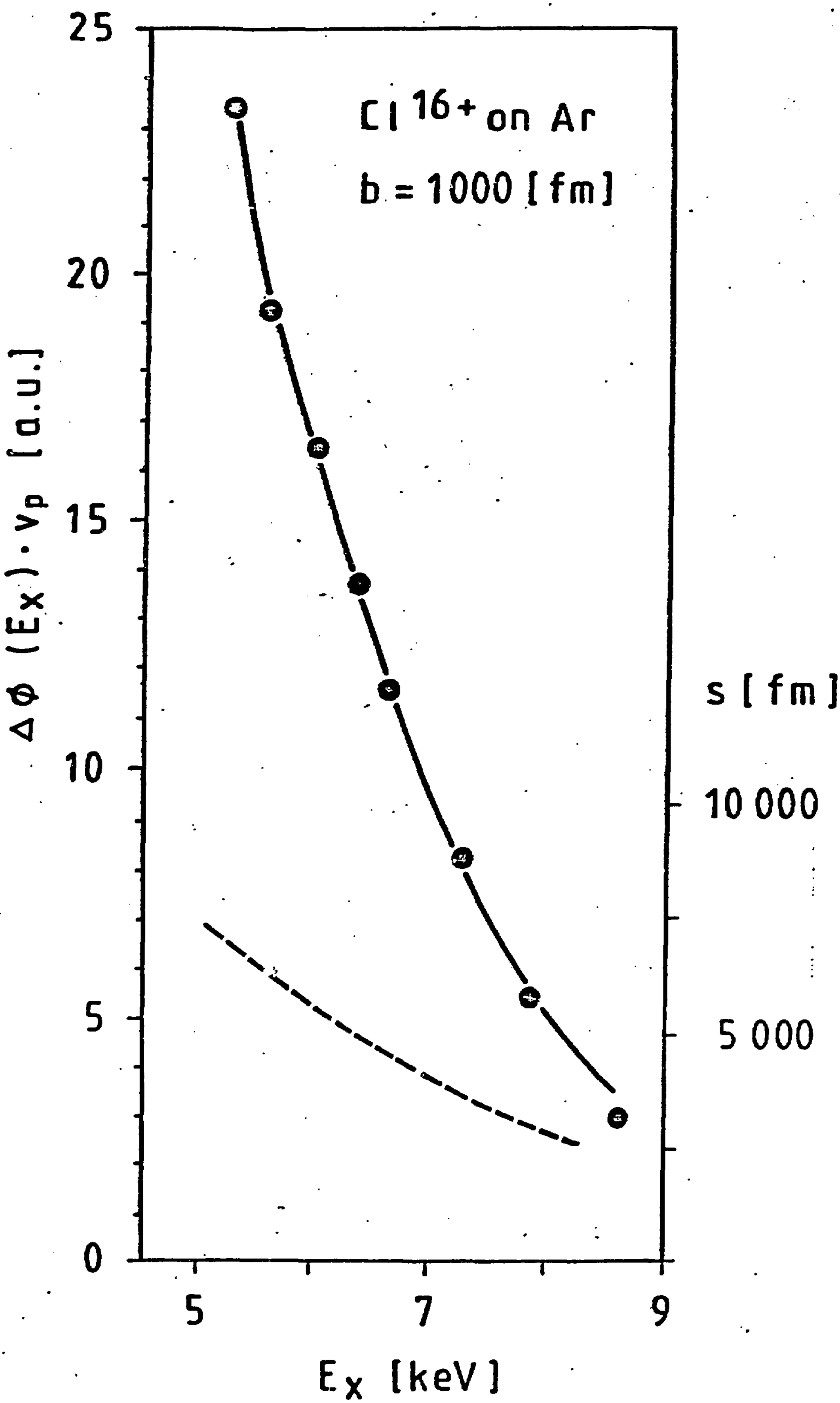


Fig 5



Cl 16+ on Ar
b = 1000 [fm]
25
20
15
10
5
0
ΔΦ (Ex)·vp [a.u.]
s [fm]
10 000
5 000
5
7
9
Ex [keV]

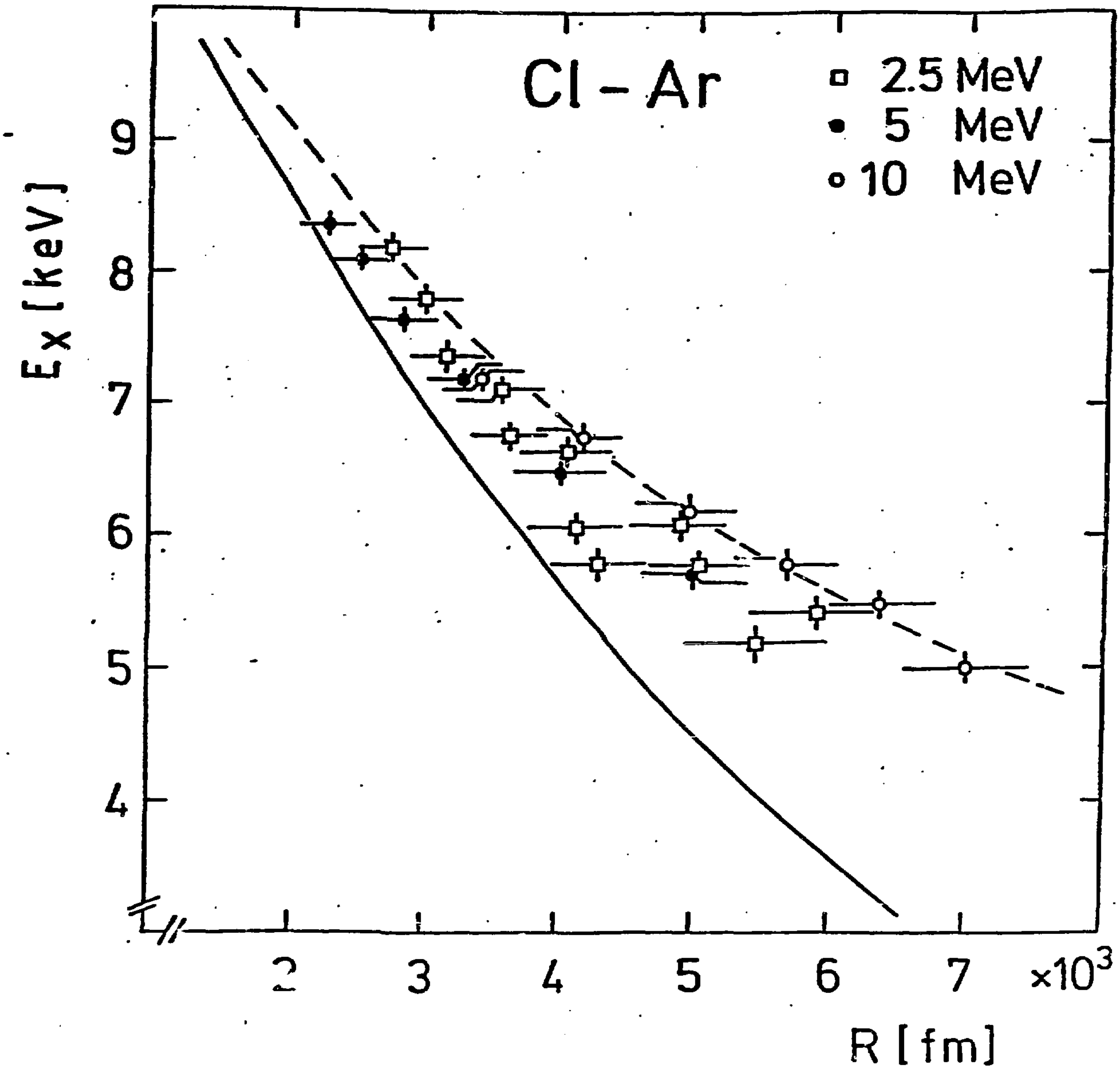


Fig. 7