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Nondissociative and Dissociative Ionization of Molecular Hydrogen by 50 to 2000 keV Antiprotons

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Abstract

A beam of antiprotons with energies between 50 keV and 2 MeV has been used for measurements of nondissociative and dissociative ionization cross sections of H_2. The results are compared to cross sections for equivelocity protons and electrons, and the role of interference effects in two–electron processes is discussed.

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In a series of papers, two-electron processes in collisions between energetic charged particles and He atoms have been discussed (for references, see Pedersen (1990)). These studies took place in order to investigate interference effects in atomic collisions which can be seen in experiments when using equivelocity projectiles of opposite charge. The processes studied were double ionization (DI) (Andersen et al 1989), double excitation (DE) (Pedersen and Hvelplund 1989), and ionization excitation (IE) (Pedersen and Folkmann 1990). It was concluded that when close collisions play an important role as they are thought to do in DI and IE, the cross sections for negative particles are up to a factor of two larger than for equivelocity positive particles. This difference is now understood as an interference effect between different collision mechanisms, and theoretical estimates by Reading and Ford (1987) and Andersen et al (1987) explain the data rather precisely.

In a recent communication (Andersen et al 1990), we reported on the first single-ionization measurements on He by energetic antiprotons. We have found that there is no $\bar{p}/p$ (antiproton/proton) difference at high energies, but that the p cross section is ~20% larger than the $\bar{p}$ cross section around the cross-section maximum (~100 keV). At energies lower than 50 keV, this difference is reversed indicating that the $\bar{p}$ cross section is larger than the p cross section at low collision energies.

Clearly, a natural continuation of the helium experiment would be the study of the most elementary process of atomic hydrogen ionization by antiprotons. Striking differences in comparison with the proton case have been predicted for this cross section, in particular at low energies (Müller-Groeling and Soff 1988, Kimura and Inokuti 1988, Ermolaev 1990). However, experimental constraints led to a first run with a molecular hydrogen target. This system provides in a unique way a study of two-electron processes (dissociative ionization) as well as the simpler (nondissociative) ionization.

It is well known that single nondissociative ionization of H_2 by energetic particles leaves the H_2^+ ion in its $1s\sigma_g$ ground state, whereas dissociative ionization leading to a production of H^+ ions results from a repulsive excited H_2^+ intermediate state (see Fig. 1). In the present work, we have studied the two processes

$$\bar{p} + H_2 \rightarrow \bar{p} + H_2^+ + e^- \quad \text{(nondissociative ionization)} \qquad (1)$$

and

$$\bar{p} + H_2 \rightarrow \bar{p} + H^+ + H + e^- \quad \text{(dissociative ionization)} \qquad (2)$$

In the first process, only one electron is active, whereas in the second, two electrons are active, i.e., it can be interpreted as an IE process.

The above interactions, together with double ionization, have been studied earlier with electrons and positive particles H^+, He^{++}, etc., as projectiles. Edwards et al. (1988) have compared single and double ionization of H_2 by equivelocity electrons and protons. They found that the ratio of single-ionization cross sections by electrons to that by protons is about 0.8 to 1 in the energy interval from 750 keV/amu to 3500 keV/amu, and that the similar ratio for double ionization is around two. They also discussed the ratio R of double-to-single ionization for protons and electrons. The value of R for electrons is about a factor of two larger than that for protons, as follows from the above discussion. This indicates the existence of interference effects in double ionization of H_2 similar to that also observed in He. The absolute value of R, which Edwards et al. (1988) reported to be around $5x10^{-4}$, has recently been questioned by Kossmann et al. (1990). For electron impact around 2 MeV/amu, they found an R value of $\sim 4x10^{-3}$ which is of the same magnitude as the corresponding R value for a He target.

The present experiment was carried out with antiprotons from the LEAR facility at CERN. The experimental arrangement was the same as described in our recent publication (Andersen et al 1990) on single ionization of He. A 5.9 MeV $\bar{p}$ beam was slowed down in a foil, then passed through a 'start' scintillation detector and the H_2 target before being stopped in another scintillator. The ions (H_2^+ and H^+) produced in the H_2-gas target of 5 mTorr were accelerated by an electric field of 800 V/cm towards a time-of-flight (TOF) spectrometer. Their TOF with respect to the antiproton signals was recorded event by event. An ion TOF spectrum is shown in Fig. 2, where the H_2^+ and H^+ peaks are clearly separated. Such spectra normalized to the incoming beam current were obtained between 50 keV and 2 MeV, and the relative ionization probabilities in this energy interval were obtained. From a comparison of the results for dissociative ionization by Shah and Gilbody (1982) and results for double ionization by Kossmann et al (1990), we estimate that double ionization amounts to less than 10% of the H^+ production. Thus the H^+ peak is essentially a signature of dissociative ionization. The results were placed on an absolute scale by normalizing to high-energy proton results for nondissociative ionization by Shah and Gilbody (1982). The same normalizing procedure was chosen in our earlier measurements on helium.

Figure 3 shows our measured cross sections σ_{ni} (reaction (1)) and σ_{di} (reaction (2)), together with the proton results of Shah and Gilbody (1982). The good agreement between $\bar{p}$/p results for nondissociative ionization in the energy interval 400-1500 keV indicates that our normalization procedure is well justified. A similar $\bar{p}$/p agreement was found for single ionization of He in the same energy interval (Andersen et al 1990). At energies between 50 and 400 keV, the proton cross section exceeds that of the antiproton by up to 14%, a number that should be compared to 22% for a He target. No theoretical estimates of this effect exist at present for a H_2 target, but the similar difference is for He explained as a result of target polarization by Fainstein et al. (1987).

The dissociative ionization cross sections were normalized via nondissociative cross sections, but because of lower count numbers and rather large background corrections due to H^+ from water vapour in the rest gas, the experimental uncertainty is ~±30%. At the highest energies, ~500 keV, where we could extract this cross section, we find that the antiproton results exceed the proton results by ~35%, but no conclusion can be drawn due to the experimental uncertainty.

Since nondissociative ionization is a one-electron process and dissociative ionization is a two-electron process, we have formed the ratio $R^*=\sigma_{di}/\sigma_{ni}$ in analogy with the ratio between double- and single-ionization cross sections for He. Figure 4 shows this ratio for e^-, H^+, He^{++}, and $\bar{p}$ projectiles. It is observed that R* behaves in a qualitative way like the ratio between double and single ionization of He, i.e., at high velocities, the ratio is larger for the negative particle than for the equivelocity positive one. This indicates that the two-electron process is also influenced by interference between a process where the projectile interacts only with one of the target electrons, whereas the second electron is excited as a result of electron-electron interaction (TS-1), and a process where the projectile interacts with both target electrons (TS-2). If we assume that these two processes can interfere, the cross section for dissociative ionization attains the value

$$\sigma_{di} = q^2\sigma_I + q^4\sigma_{II} - q^3\cdot 2\sigma_{int} , \qquad (3)$$

where σ_I and σ_{II} are the cross sections for ionization excitation as a result of one and two interactions with the projectile, respectively. σ_{int} is the contribution due to interferences between the two processes.

Under the assumption that $\sigma_{ni}(H^+)=\sigma_{ni}(\bar{p})$ and $\sigma_{ni}(He^{++})=4\cdot\sigma_{ni}(H^+)$, which is valid at 500 keV/amu, we obtain

$$R^*_I = \frac{\sigma_I}{\sigma_{ni}(H^+)} = R^*_{H^+} + \frac{1}{3}R^*_{\bar{p}} - \frac{1}{3}R^*_{He^{++}} \qquad (4)$$

$$R^*_{II} = \frac{\sigma_{II}}{\sigma_{ni}(H^+)} = \frac{1}{2}R^*_{H^+} + \frac{1}{6}R^*_{\bar{p}} - \frac{1}{3}R^*_{He^{++}} \qquad (5)$$

$$R^*_{int} = \sigma_{int}/\sigma_{ni}(H^+) = \frac{1}{4}(R^*_{\bar{p}} - R^*_{H^+}) . \qquad (6)$$

At 500 keV, we obtain (see Fig. 4) the following experimental values:

$$R^*_I = 3.2\text{x}10^{-2} , \quad R^*_{II} = 1.3\text{x}10^{-2} , \text{ and } R^*_{int} = 3.7\text{x}10^{-3} . \qquad (7)$$

We have made theoretical estimates of the values of R^*_I and R^*_{II} by using the procedures developed by Andersen et al. (1987) and Pedersen and Folkmann (1990) for double ionization and ionization excitation of He. The distant collisions were evaluated by means of the Weizsächer-Williams method of virtual quanta by using known cross sections for photon interactions (Kossmann et al 1987, Browning and Fryar 1973, Masuoka 1984). The close collisions were treated by applying the Thomson formula and energy intervals corresponding to Franck-Condon transitions for ionization and excitation (see Fig. 1). The values at 500 keV/amu estimated in this way are $R^*_I=6.4\text{x}10^{-2}$ and $R^*_{II}=3.5\text{x}10^{-2}$. In TS-1, the distant-collision contribution amounts to ~13%, and the maximum value of R^*_{int} is accordingly estimated to be $4.4\text{x}10^{-2}$.

From a comparison between experimental and estimated values of R^*_I and R^*_{II}, it should be noted that the latter overestimates the measured values by a factor of two. For two-electron processes in He, estimated values are in much closer agreement with experimental ones. The measured value of R^*_{int} amounts to only 10% of the estimated maximum value. This indicates that the interference term is not as important for ionization-excitation of H_2 as it is for He, where the measured interference term amounts to 50% of the estimated maximum value.

For a He target, the ratio between ionization excitation and single ionization is found to be ~3 times larger than R* (Pedersen and Folkmann 1990). This difference between results obtained for H_2 and He probably originates from effects caused by the two-center molecule structure of H_2 compared with the one-center atomic structure of He.

Recently, Edwards et al. (1989) measured ionization excitation of H_2 by 1

MeV/amu electrons. From an energy analysis of the H^+ target ion, they extracted information about excited H_2^+ states. They concluded that the dominating processes are excitation to the H_2^+ states $2p\sigma_u$, $2p\pi_u$, and $2s\sigma_g$. The present experimental results lead us to the conclusion that the same intermediate states are formed in $\bar{p}+H_2$ collisions, leading to dissociative ionization.

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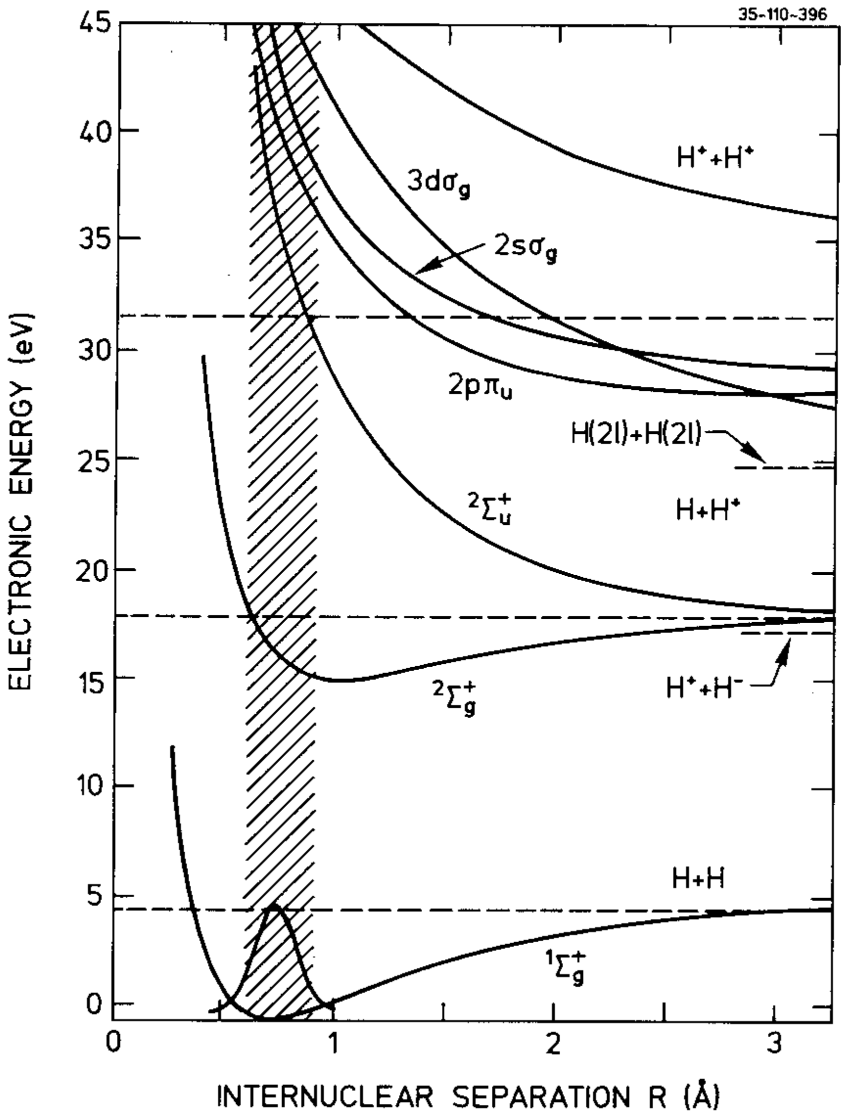
35-110-396
ELECTRONIC ENERGY (eV)
45
40
35
30
25
20
15
10
5
0
INTERNUCLEAR SEPARATION R (Å)
0
1
2
3
H⁺+H⁺
3dσ$_g$
2sσ$_g$
2pπ$_u$
H(2l)+H(2l)
$^2\Sigma_u^+$
H+H⁺
$^2\Sigma_g^+$
H⁺+H⁻
H+H
$^1\Sigma_g^+$

Fig. 1

FIGURE CAPTIONS

Fig. 1. Potential-energy curves of H_2 and H_2^+. For the ground state of H_2, we have indicated the associated vibrational probability, and the hatched area indicates electronic transitions allowed by the Franck-Condon principle.

Fig. 2. Time-of-flight spectrum obtained for 250-500 keV antiprotons colliding with 5 mTorr H_2.

Fig. 3. Cross sections σ_{ni} for nondissociative ionization, upper part, and σ_{di} for dissociative ionization, lower part. Present results for antiprotons, ●. Results for proton impact, —— (Shah and Gilbody 1982).

Fig. 4. The ratio $R^*=\sigma_{di}/\sigma_{ni}$ between dissociative and nondissociative ionization cross sections ●. Results of Shah and Gilbody (1982) for protons, o, and for He^{++}, Δ. Results of Rapp et al (1965) for electrons, Δ.

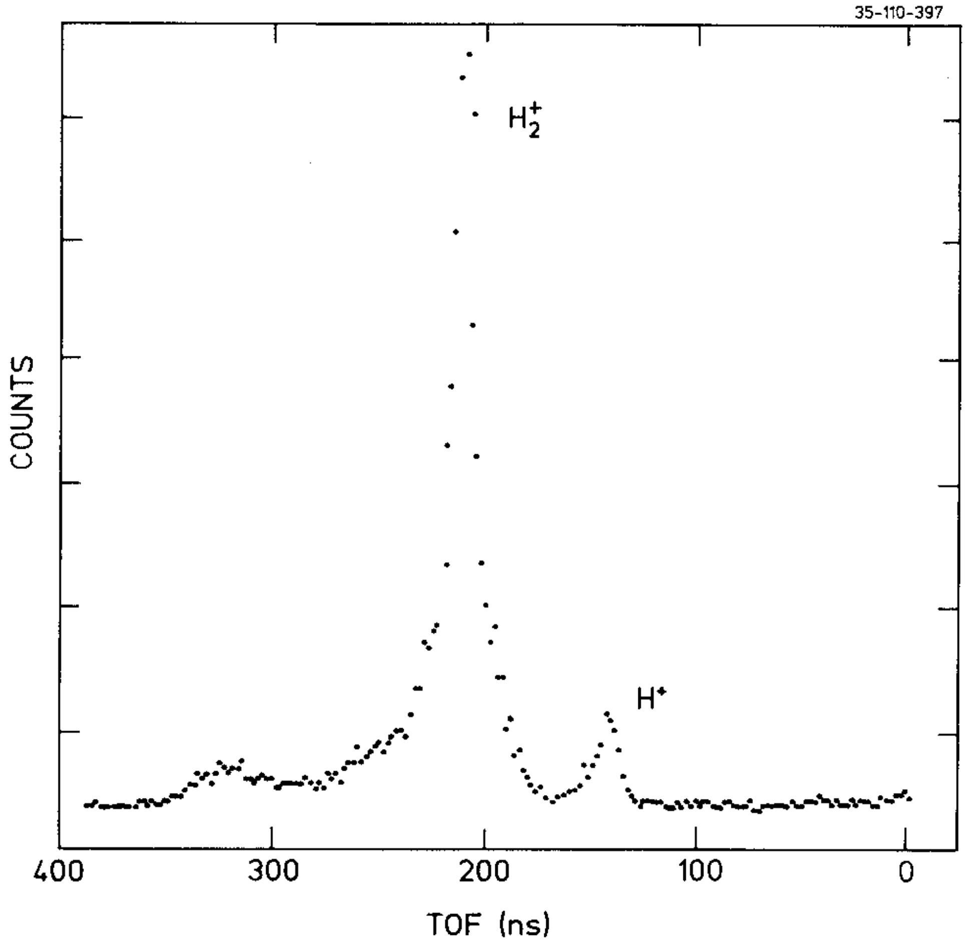

Fig. 2

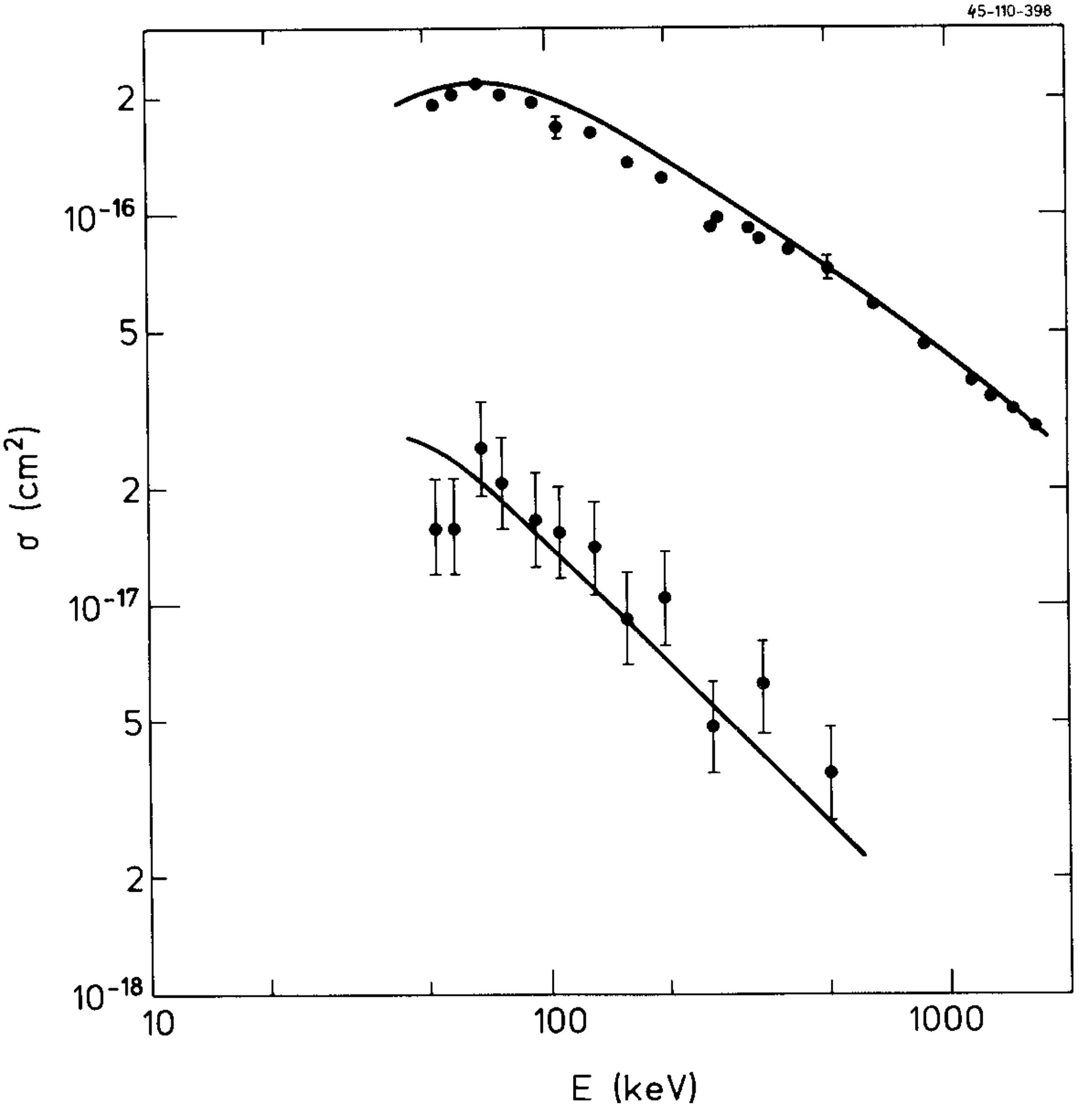

Fig. 3

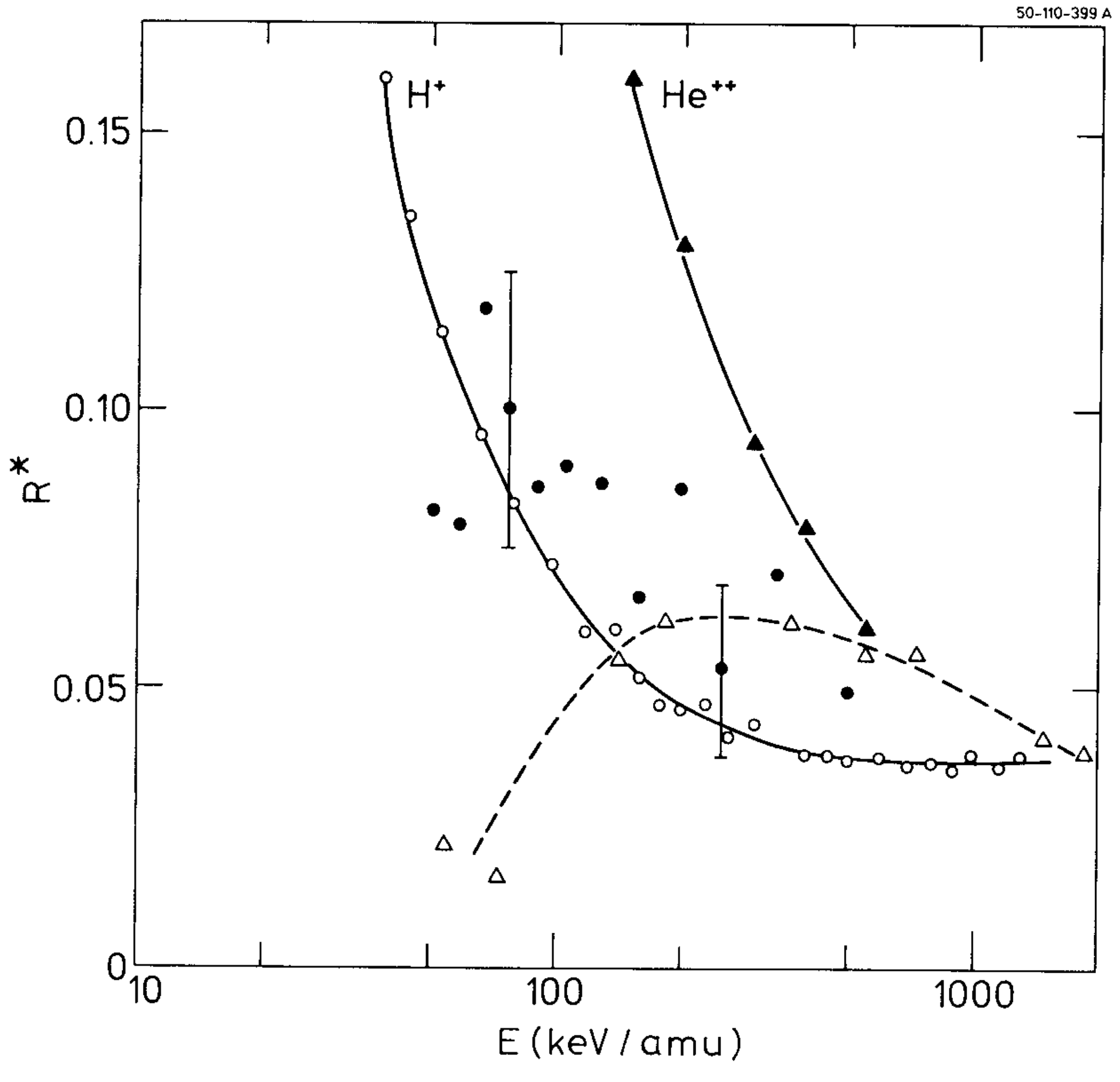

Fig. 4

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