

HYPERTHERMAL BEAMS SPUTTERED FROM ALKALIHALIDE SURFACES

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(Received September 27, 1973)

Velocity selected beams of atoms and molecules have been produced by sputtering from various alkali halide surfaces under 6 keV Ar⁺ ion bombardment. The mass spectrum and the velocity distribution of these particles have been analysed in the range from 0-13000 m/s. In the low velocity part of the spectrum monatomic particles, as well as polyatomic ones have been found. For velocities larger than 3000 m/s the number of sputtered molecules is very small compared to the number of atoms. In addition to typical sputtering velocity distributions, thermal distributions are also found, due to vaporization of the less effectively sputtered atoms from the crystals. Above a few eV the energy distribution of a given alkali or halogen atom is independent of the target from which it is sputtered. There are indications that some sputtered atoms and molecules are formed in long lived excited states.

1 INTRODUCTION

During the last years there has been a growing interest in "hot atom" beams for collision physics and chemistry, as well as for the investigation of surface phenomena. The supersonic gasflow method has an upper limit of about one eV kinetic energy, although by "seeding" energies can be obtained as high as 15 eV, dependent on the mass. The charge exchange method seems to work best above 10 eV. The sputtering method is especially suitable between 1 and 20 eV. Sputtering has been usually limited to the production of alkali beams, mainly because the detection of alkali atoms in the eV range is so easy by high temperature surface ionization. Only recently scattering experiments were reported by Cohen, Young and Wexler¹ with Ba, Ti, Ta and Al beams, obtained by bombardment of Ba, Ti, Ta and Al targets with Ar⁺ projectiles of 9 keV kinetic energy.

A very important category of projectiles for chemical research are the halogens and it is because of that reason that we have tried to develop sputtering of alkali halide crystals (Table I), and to investigate the detection possibilities for halogen atoms in the eV energy range.²

Several investigators have studied certain aspects of the sputtering process of ionic crystals by heavy projectiles. Very few however, measured the neutral particles which usually represent the bulk of the

sputtered material, the reason being that ions are much easier to detect.³⁻⁷

Campbell and Cooper⁸ looked for the neutral and charged sputtering products of:

KBr + Ar⁺, Xe⁺ (25 - 180 eV kinetic energy),

but were only able to detect Br⁻.

Stein⁹ however, detected neutral K from (001) and (011) surfaces from:

KCl + Ne, Ar (400 eV kinetic energy).

He investigated the angular distribution of the sputtered products. Windawi and Cooper¹⁰ recently measured the angular distribution of Cl from the (100) face of:

KCl + Ar⁺ (200 eV kinetic energy).

Navinšek¹¹ determined the total sputtering coefficient S for the (100) surface of many salts:

NaCl, KCl, KBr, LiF + Ar⁺ (2-10 keV kinetic energy).

S proves to be 1.0 for NaCl, 1.9 for KCl, 0.6 for KBr and 2.2 for LiF at an Ar⁺ kinetic energy of 10 keV. It is an important number for those people who want to use the sputtering process to generate hot atom beams for collision research.

Finally Schmidt-Bleek and Datz¹² sputtered AgI and AgBr surfaces by 0.5-7 keV Ar⁺ ion beams and indeed got hot beams of Ag, I and Br atoms and of AgI and AgBr molecules.

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TABLE I
Values of various properties of alkali halide crystals

	$E_b(\text{eV})^{30}$	$E_D(\text{eV})^{31}$	$E_u(\text{eV})^{31}$	$R(\text{Å})^{31}$	crystal type
LiCl		4.9	8.6	2.49	fcc NaCl type
NaCl	2.5	4.2	7.9	2.78	fcc NaCl type
KCl	2.4	4.4	7.2	3.14	fcc NaCl type
RbCl	2.3	4.4	7.0	3.28	fcc NaCl type
CsCl	2.0	4.6	6.6	3.48	prim. cubic CsCl type ^a
KBr	2.3	3.9	6.9	3.29	fcc NaCl type
CsBr	2.3	4.2	6.3	3.63	prim. cubic CsCl type
KI	2.3	3.3	6.5	3.53	fcc NaCl type
RbI		3.3	6.3	3.67	fcc NaCl type

^a For temperatures above 728 K fcc NaCl type.

E_b is the binding energy of a molecule with respect to the surface, E_D the dissociation energy of the molecules in the gas phase, E_u the lattice energy and R the distance of the halogen ion to the nearest alkali ion in the crystal. The lattice energy is the energy required to separate the ions of a crystal to an infinite distance from each other. It is expressed in eV/ion pair.

In this paper we present the results of an analysis of mass spectra and velocity distributions of many atoms and radical groups sputtered from a large variety of ionic targets. Above about 2 to 4 eV this method seems to be suited for the production of well defined atom beams using a mechanical velocity selector. Below this energy the presence of dimers is confusing.

Apart from this application our investigations give information about the sputtering process from salt surfaces bombarded by a beam of 6 keV Ar^+ ions.

2 THE APPARATUS

2.1 General

A schematic view of the apparatus is seen in Figure 1. Particles are sputtered from a target by a 6 keV Ar^+ ion beam. After collimating and velocity selection they can be detected on two hot wires by means of positive and negative surface ionization (PSI and NSI). The composition of the beam can be analysed by means of a home made mass spectrometer [$(m/\Delta m) \approx 20$] provided with an Extra Nuclear electron bombardment ionizer (efficiency 10^{-3} for thermal atoms).

The four compartments containing the ion source, the velocity selector, the detection wires, and the mass spectrometer are differentially pumped, the first two compartments by oil diffusion pumps and the third and fourth by an ion getter pump and by two ion pumps respectively. Under working conditions, the pressure is 10^{-5} torr in the first com-

partment, 10^{-7} in the second one and better than 10^{-7} in the third and fourth compartment.

2.2 Beam Source

The ion source is the same one as used by Politiek *et al.*,¹³ who described it in detail. We put a chopping potential on one of the quadrupole lenses in order to modulate the sputtered beam. As targets various alkali halide (MX) discs are used. They have been pressed at 5000 atm for about five minutes, and are about 2 mm thick. No difficulties have been met in connection with possible charging up by the ion beam.

Possible sputtering products are halogen atoms X, alkali atoms M as well as different molecular species, and positive and negative ions. The positive ions, however, cannot leave the target holder, which has a potential of -6 keV. The negative ions would get an energy of 6 keV. For such fast particles the transmission of the velocity selector is zero. Actually, even without velocity selection, we never found any negative ions in the mass spectrometer.

We also tried sputtering from a Br_2 surface at liquid nitrogen temperature. Although this technique seems to give a larger amount of hyperthermal Br atoms than sputtering from a salt, it was not applied because it is technically more difficult. Furthermore, the large amount of Br_2 in the apparatus is a disadvantage.

2.3 Electron Bombardment Detector

A scale drawing of the ionizer and the additional lens system is shown in Figure 2. In the lens system, the

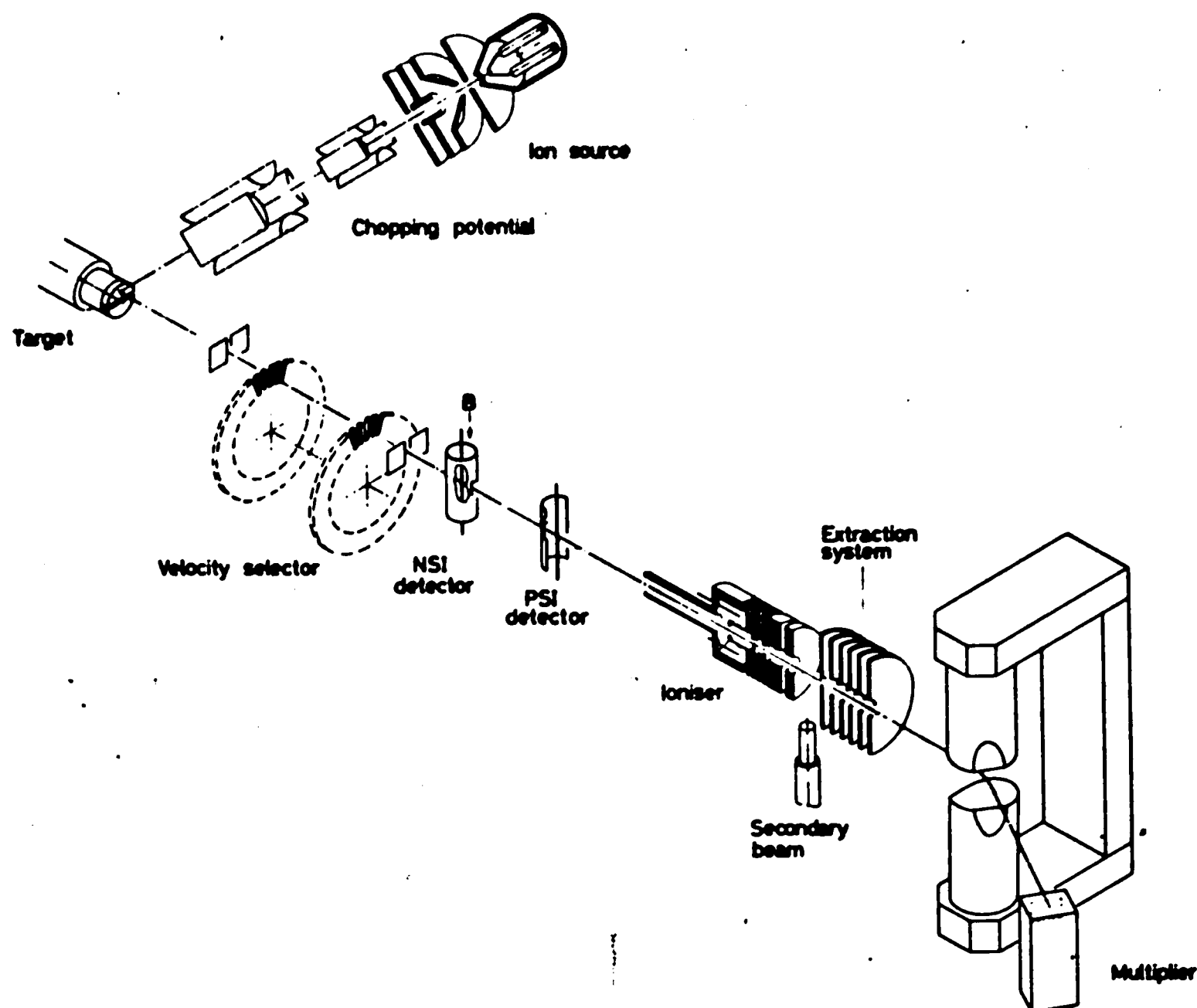


FIGURE 1 Schematic drawing of the apparatus. PSI and NSI stand for positive and negative surface ionization respectively.

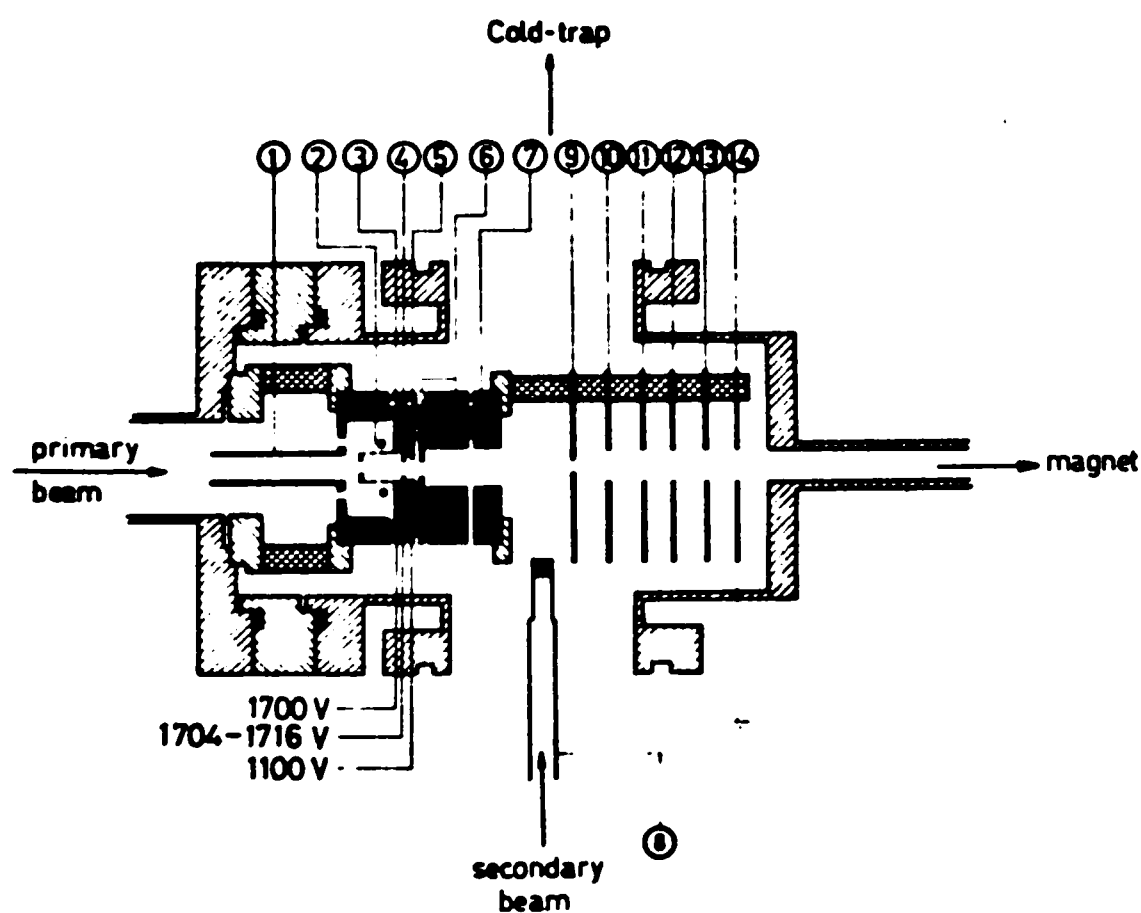


FIGURE 2 Scale drawing of the ionization system to make positive ions. Cylinder 1 prevents penetration of the outer fields; 2 filament; 3 ionization region; 4 extractor; 5, 6, 7 focusing lenses; the electric field from 7 to 12 is homogeneous; 12, 13, 14 form an Einzel lens. The principal voltages are indicated. Extractor 4 is used for discriminating thermal particles against hyperthermal ones at a voltage of +1716 V. The surrounding house is at ground potential. The potential of cylinder 1 and of filament 2 is +1600 V.

positive ions are accelerated with a potential difference of 1700 V, lenses 12, 13 and 14 are used as an Einzel lens. It is possible to discriminate thermal back-

ground particles against hyperthermal ones, if one applies to lens 4 a potential which is positive with respect to that of the ionization region.¹⁴ In our discrimination mode, particles with a kinetic energy higher than 4 eV are not attenuated, while the thermal background decreases by a factor of ten. The flow of the velocity selected beam of about $4 \cdot 10^5$ to 10^7 particles/s gave a mass spectrometer signal of 5 to 400 counts/s dependent on their velocity. Of course, for the measurements of low energy sputtered particles this attenuation threshold has not been applied. Because of the large background signal ($2 \cdot 10^4$ counts/s for I and $5 \cdot 10^5$ counts/s for Cl, if the attenuation threshold is not used), long measuring times were necessary to measure the velocity distributions.

3 RESULTS AND DISCUSSION

3.1 Low Velocity Part of the Spectrum

The velocity distributions up to 4000 m/s of particles sputtered from KI, NaI and CsCl are shown in Figures 3 to 6.

The curves give the signals obtained at the collector of the mass spectrometer. The ionization efficiency of the ionizer is inversely proportional to v ; the velocity selector has a constant resolution

so that the $M_2 X^+$ peak, observed by us, is almost entirely due to $M_2 X_2$ molecules. The X^+ , M_2^+ , X_2^+ and MX^+ peaks are mainly produced from the respective atoms and molecules. The M^+ peak is formed mainly from M atoms, but will also have a contribution from dissociative ionization of MX . The MX^+ peaks found in the mass spectra of $CsCl$ and $CsBr$ are small in comparison with those found with KI and NaI . This is in agreement with the measurements of Berkowitz and Chupka, who find that for KI and for NaI channels (2) and (3) have about the same cross section, while for $CsCl$ and $CsBr$ nearly all molecular ions dissociate.

From these considerations we conclude the following. The velocity distributions of the neutral X , X_2 , MX and $M_2 X_2$ particles are identical to the curves for X^+ , X_2^+ , MX^+ and $M_2 X^+$ respectively, as given in Figures 3, 4, 5 and 6. For alkali atoms this is only true at velocities for which the contribution from MX may be neglected.

The velocity distributions found can be divided into three groups. *First*, there are the atoms, which show a large intensity and are present in the beam to a measurable extent even at very high velocities. *Second*, we have the salt molecules MX and $M_2 X_2$; they have somewhat lower intensity and go to zero much faster with velocity than the atom distributions do. An example of the *third* group is I_2 . This molecule manifests itself only at low velocities. Within the experimental error, the velocity distribution of I_2 which is shown in Figure 4 is Maxwellian with a temperature of 560 K. The velocity distribution of I sputtered from KI shows a sharp maximum on the low velocity side, which can be fitted with Maxwellian distribution of the same temperature of 560 K (Figure 3). I sputtered from NaI also shows a sharp maximum on the low velocity side, which fits a Maxwellian distribution of 450 K (Figure 5). Obviously, these low velocity atomic peaks belong to the third group of thermal particles.

A possible explanation of the thermal peaks can be the fact that Na and K are sputtered more effectively than I under bombardment with Ar^+ ions. The sputtering ratio for a monatomic crystal depends on the energy transfer of the impinging particles, which is proportional to the factor

$$\frac{4m_1 m_2}{(m_1 + m_2)^2} \quad (5)$$

where m_1 and m_2 are the masses of the bombarding particles and of the target atoms.¹⁶ The same relation is expected to influence the ratio in which M

and X atoms are removed from our crystals. We estimate from our ion beam intensity, using (5), that after a few milliseconds of sputtering, the upper layers of the crystal would mainly exist of I atoms if no vaporization would take place. The excess of I will vaporize thermally from the surface. If this is indeed the mechanism of production, the temperatures found are the temperatures of the surface locations from which the particles evaporate. A thermal peak should only arise for the atoms with the smaller sputter ratio. We measured thermal maxima in the velocity distributions of sputtered alkali's from $NaCl$, $CsCl$, $CsBr$ and CsI , under bombardment of Ar^+ . In the case of NaF , KCl , KBr , KI , $RbCl$ and RbI a thermal alkali peak was never observed. These measurements of the thermal alkali distributions were performed with the PSI detector. We have to mention that the height of the thermal peak is not stable, but depends strongly on the place from which sputtering takes place and on the time duration of the bombardment. For $CsCl$ and $CsBr$ for instance, the thermal Cs -peak sometimes vanishes totally. In Figure 6 we clearly see only a higher energy distribution for the measured Cs^+ ions in the mass spectrometer.

In contrast to the molecules M_2 and X_2 , the species MX and $M_2 X_2$ are present at much higher energies than the thermal peaks. So we conclude that they are really a product of direct momentum transfer and not merely products of thermal vaporization.

In Figure 7 the energy distributions $dS/dE(E)$ of $K_2 I_2$, KI and I sputtered from KI are plotted on a double logarithmic scale. These energy distributions are calculated from the Figures 3 and 4, using (1) and assuming that the $K_2 I^+$, KI^+ and I^+ peaks in the mass spectrometer are entirely due to ionization of $K_2 I_2$, KI and I respectively. The energy distributions of the molecules go to zero much more rapidly than those of the atoms. The same is observed for the energy spectra of alkali dimers from polycrystalline alkali targets,¹⁷ and also for charged polyatomic clusters, sputtered from various polycrystalline metal targets.¹⁸⁻²⁰

The formation of dimers can be explained by a simple kinetic model. If a collision cascade arrives at the target surface there is a probability that two particles will leave the surface simultaneously. These particles will in general have different directions and velocities. If however the relative kinetic energy of these particles is lower than the potential energy between them, they will stick together and form a vibrational and rotational excited dimer. Dimers are likely to be formed mainly by sputtering of neigh-

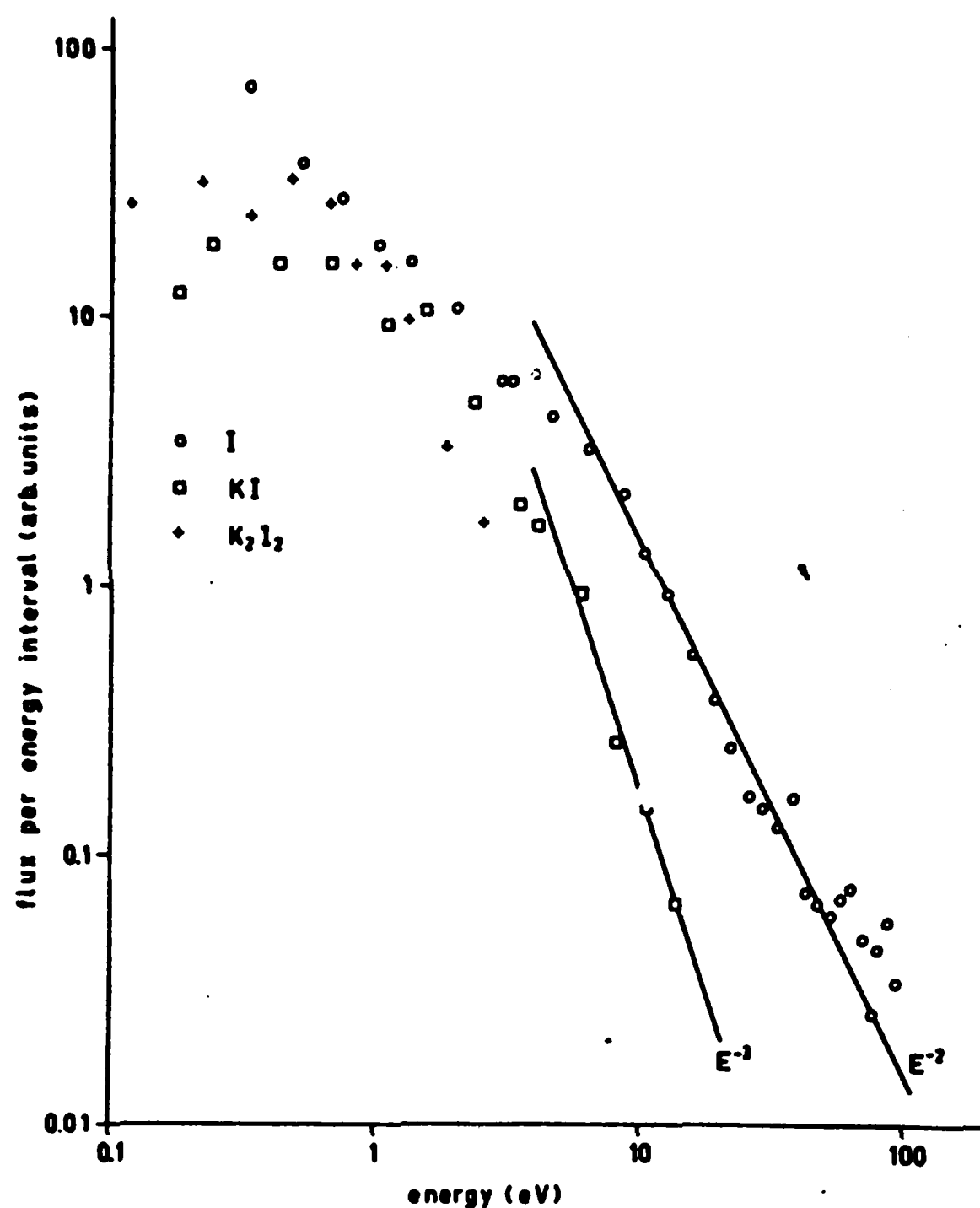


FIGURE 7 The energy distributions of K_2I_2 , KI and I as a function of the kinetic energy.

bouncing particles, because they have the strongest interaction.

One would expect that the formation of dimers decreases rapidly if they have kinetic energies above their dissociation energy. The formation of hyperthermal M_2 and X_2 dimers is unlikely, because the distance between equal particles in an alkali halide lattice is larger than of unequal particles and their interaction in the gas phase is low. The fact that equal M or X ions in the lattice repel each other may also obstruct the formation of hyperthermal M_2 and X_2 molecules.

Calculations on basis of this model give good agreement with the experimental energy distributions of the dimers.²¹ The M_2X_2 molecules are likely to be formed in an analogous way.

3.2 High Velocity Spectrum

In Figures 8, 9 and 10 we make a comparison of the energy distributions $dS/dE(E)$ for halogens and alkalis sputtered from targets with the same mass ratio, e.g. I from KI is compared with Cs from ClCs. The alkalis, for which a much higher precision is obtained, are represented by solid lines. They have been analyzed by means of the PSI detector with a

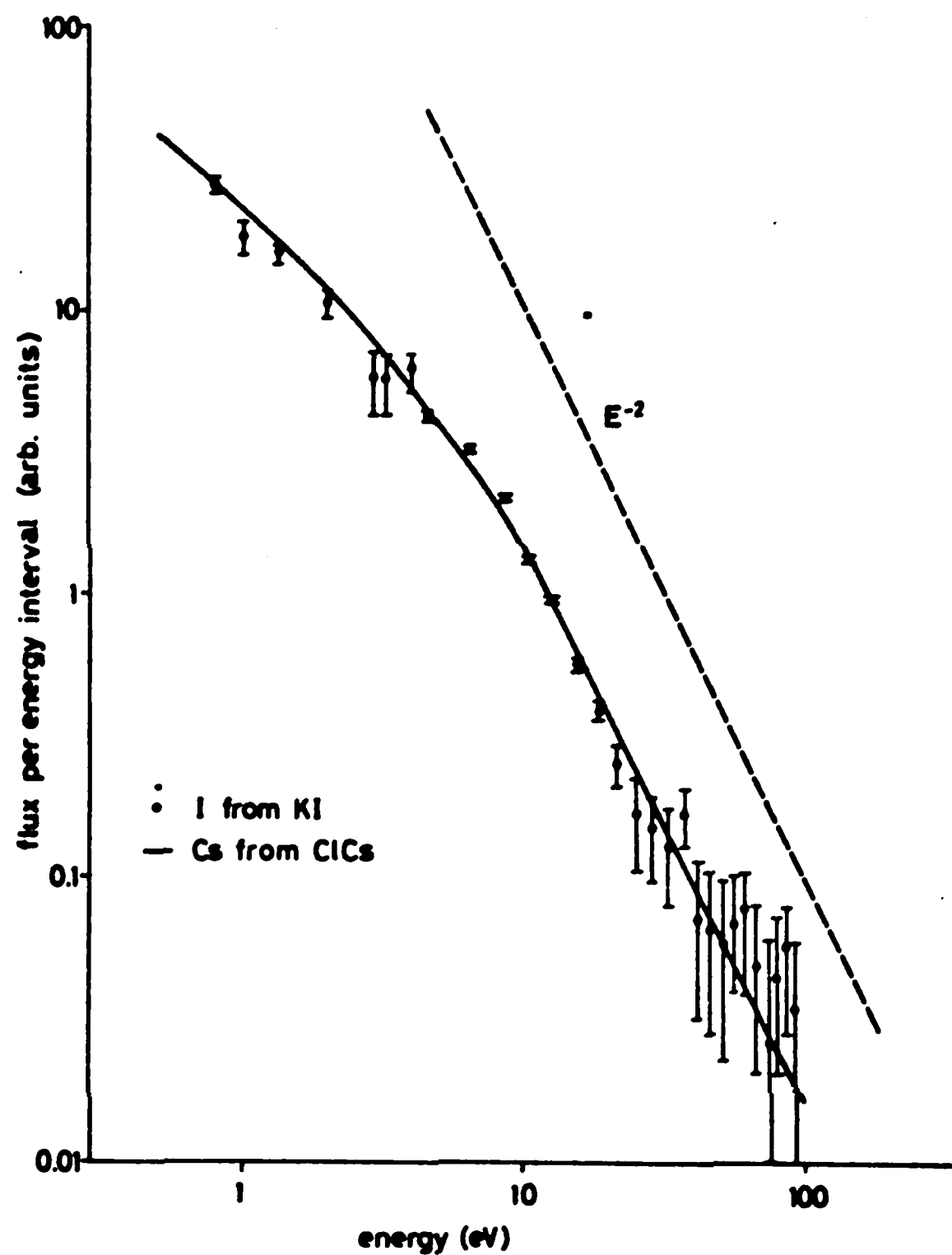


FIGURE 8 The energy distribution of I from a KI target bombarded with 6 keV Ar^+ ions. The solid line represents measurements of Cs from CsCl. The dashed line indicates a slope $\propto E^{-2}$.

Pt 8%W wire. The K fluxes have been corrected with the ionization efficiency values given by Politiek and Los,²² while those for Rb and Cs are uncorrected. The halogens were analyzed with the mass spectrometer. Their absolute intensities therefore cannot be compared directly with the intensities of the alkali's. By vertical shifting the alkali and halogen distributions can be made to coincide in each figure.

Measuring the chlorine component of LiCl, NaCl, KCl, RbCl and CsCl, by means of the NSI detector with a thoriated tungsten wire, we found above 3 eV the same energy distributions (see Figure 11). The same was observed for bromine from CsBr and KBr. The energy distributions of the potassium component of K, KCl, KBr and KI measured with the PSI detector are also equal within a few percent for energies above 5 eV (see Figure 12). Deviations below 5 eV may be partly due to PSI of MX and M_2X_2 molecules in the latter case.

The energy distributions of the atoms in Figures 8, 9 and 10 tend towards E^{-2} as the energy increases. The latter distribution is to be expected from a random collision cascade model.^{23,24} This model

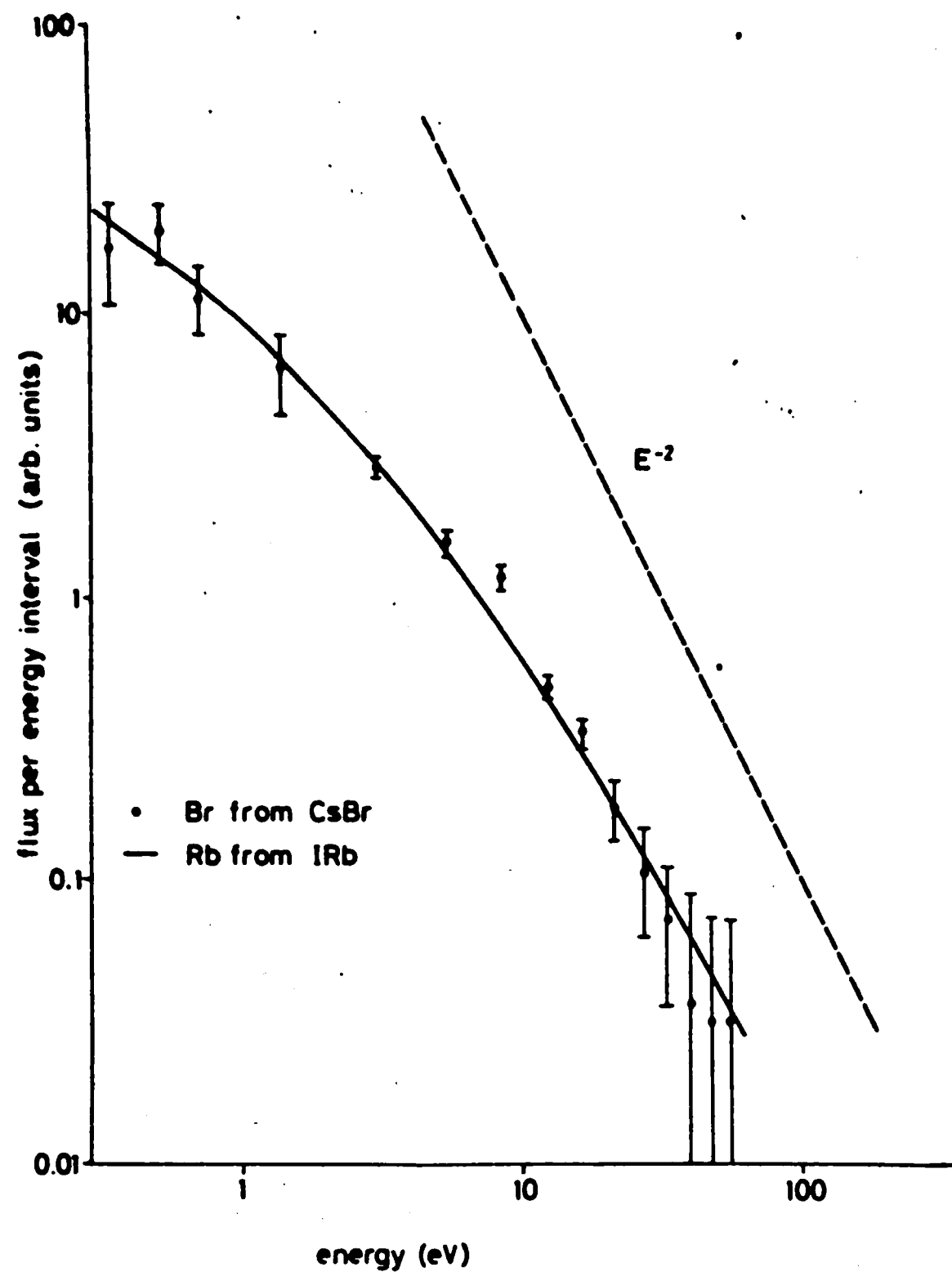


FIGURE 9 The energy distribution of Br from a CsBr target bombarded with 6 keV Ar^+ ions. The solid line represents measurements of Rb from RbI. The dashed line indicates a slope $\propto E^{-2}$.

gives for the energy distributions of the particles outside the surface²³

$$\frac{dS}{dE} \propto E(E + E_b)^{-3} \quad (6)$$

in which E_b is the binding energy of an atom to the surface. Differentiating formula (6) one finds the maximum to occur at an energy of $\frac{1}{2}E_b$. The experimental curves reach a maximum below 0.5 eV, which is in agreement with the low binding energies E_b of 0.3 to 0.7 eV for adsorbed alkali and halogen atoms on alkali halide surfaces.^{25,26}

The energy distributions of Rb, Br, K and Cl atoms in Figures 9 and 10, being mutually equal within the experimental error, deviate slightly from E^{-2} even at higher energies. The value of the exponent is between -1.5 and -1.8 for energies above 10 eV. A similar deviation of the exponent has been reported in the energy distributions of the atoms from polycrystalline Au²⁷ and polycrystalline K.²⁸ The energy distributions of Cs and I in Figure 8 show on the other hand a E^{-2} behaviour between 10 and 100 eV.

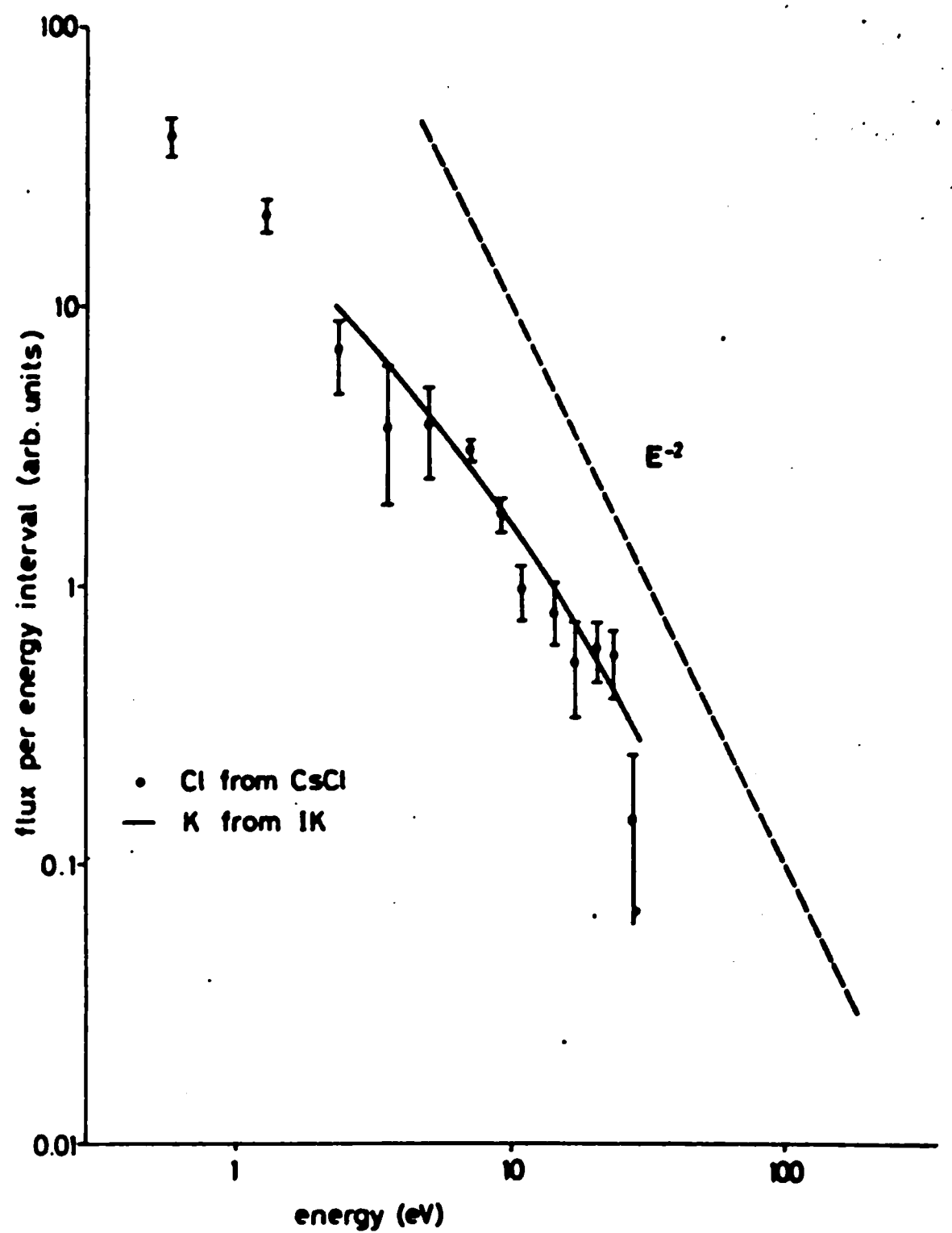


FIGURE 10 The energy distribution of Cl from a CsCl target bombarded with 6 keV Ar^+ ions. The solid line represents measurements of K from KI. The dashed line indicates a slope $\propto E^{-2}$.

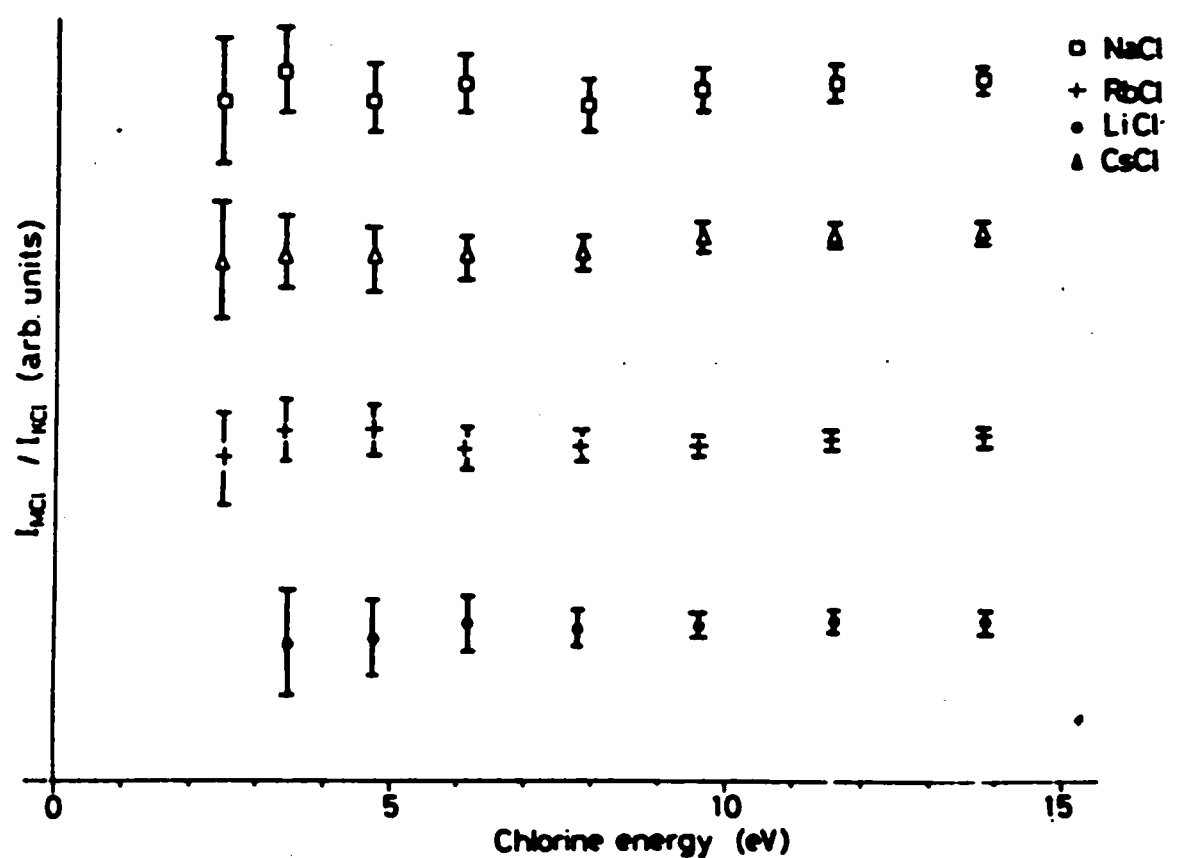


FIGURE 11 The NSI signal of beams sputtered from different chlorine containing targets MCl , divided by the signal from KCl , as a function of the chlorine kinetic energy.

From Figure 11 the most striking conclusion seems to be that Cl sputtered from LiCl, NaCl, KCl, RbCl and the heaviest salt CsCl seems to have exactly the

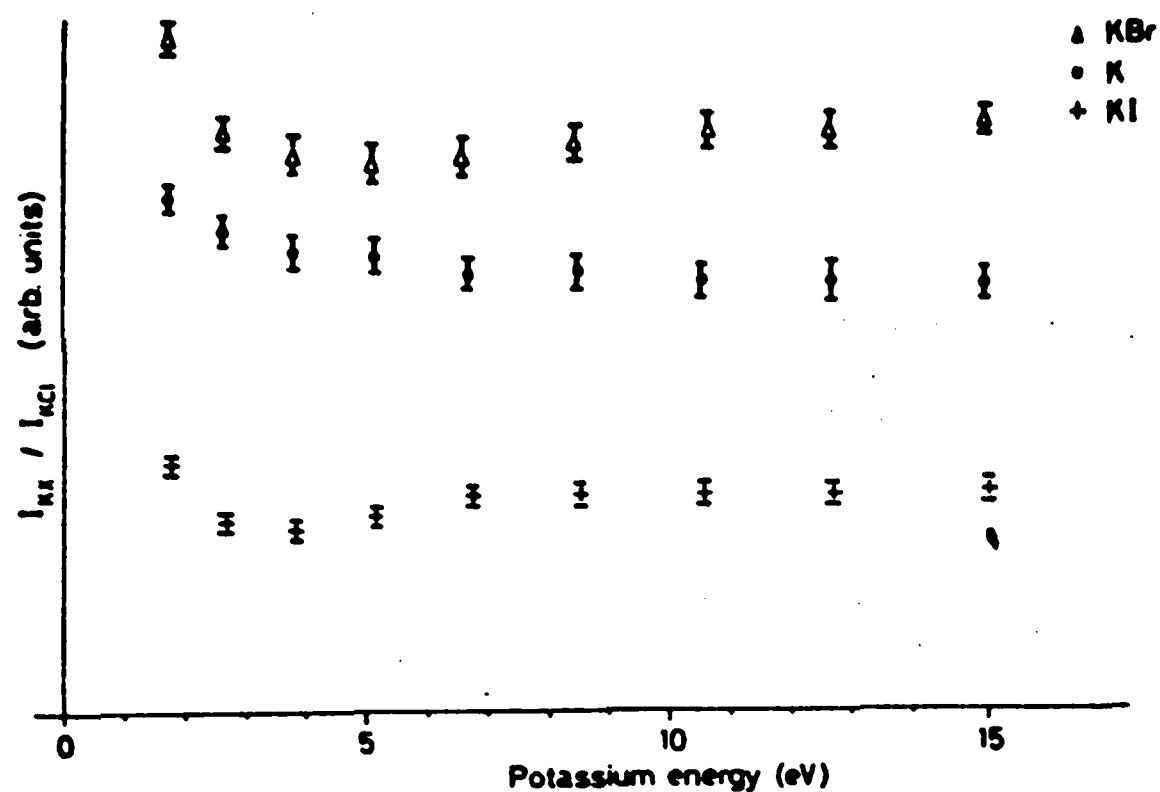


FIGURE 12 The PSI signal of beams sputtered from different potassium containing targets KX, divided by the signal of KCl, as a function of the potassium kinetic energy.

same energy distribution, in spite of the fact that the energy distributions of for instance Cs and K from CsCl and KCl are different, as can be seen from Figures 8 and 10. The energy distribution of K also seems to be independent of the chemical composition of the target. A possible explanation for this effect might be that in the collision cascade the energy distribution of elements like K or Cl mixed with heavy or light elements like Cs, I or Li is hardly affected by the presence of the heavier or lighter particles. In fact, the transfer factor (5) indicates that a collision of K or Cl against an element like Cs, I or Li hardly leads to energy transfer. On the other hand, in a salt like KCl the masses of K and Cl are nearly equal which simply means that in a collision cascade for collision energies larger than say the binding energy in the lattice there will be no difference in the energy distribution functions of these particles. These facts combined with the low surface binding energy of adsorbed atoms apparently lead to equal energy dependences in the range above 5 eV.

From the data in Figures 8, 9, 10, 11 and 12 one gets the impression that it is the energy distribution of the individual atoms in the collision cascades which determines the sputtering energy distributions. Indeed, the equal deviations from the E^{-2} behaviour of the energy distributions of K and Cl sputtered from various ionic targets as well as from the alkali metal makes it unlikely that this is due to contributions like surface deflected recoils as is suggested for Au.²⁷ Moreover, the equality of the energy distributions of K and Cl and of I and Cs sputtered from KI and CsCl indicates that the crystal structure of the targets (being fcc NaCl type and prim. cubic CsCl type respectively) does not play an important role. It is not impossible that the observed difference in the

energy distributions for Cs and I with respect to K and Cl originates from inelastic processes in the collision cascades.

3.3 Metastable Excited Particles

Part of the sputtered products are in metastable states. Evidence for this was found for I and Br atoms and most of the MX molecules, investigating the energy dependence for collisional ionization with heavy particles. For the halogens, this evidence was obtained from measurements on the process $X + C_6H_5NH_2 \rightarrow X^- + C_6H_5NH_2^+$.²⁹ The thresholds for Br and I turned out to be smaller by about 0.4 and 0.9 eV than expected for the ground state atoms. These are just the excitation energies for the $^2P_{1/2}$ state of respectively Br and I. We estimate that about 30 to 50% of the sputtered halogen is formed in the $^2P_{1/2}$ state. This is found from a comparison of the experimental ionization cross sections for Br and I with the expected linear proportionality with the velocity near threshold. For Cl, the excitation energy of this state is only 0.1 eV, which is within the spread of the velocity selector. The measured threshold for ionization was in agreement with the expected one.

The conclusion that *metastable MX molecules* are present in the beam, comes from the following argument: We find M^+ and X^- ions if the sputtered beam is crossed with various secondary beams. Specific experiments were done by sputtering from KCl and CsCl targets with crossing beams of CH_4 , C_6H_{12} (cyclohexane) and $C_6H_5NH_2$ molecules. The results for KCl on $C_6H_5NH_2$ are given in Figure 13,

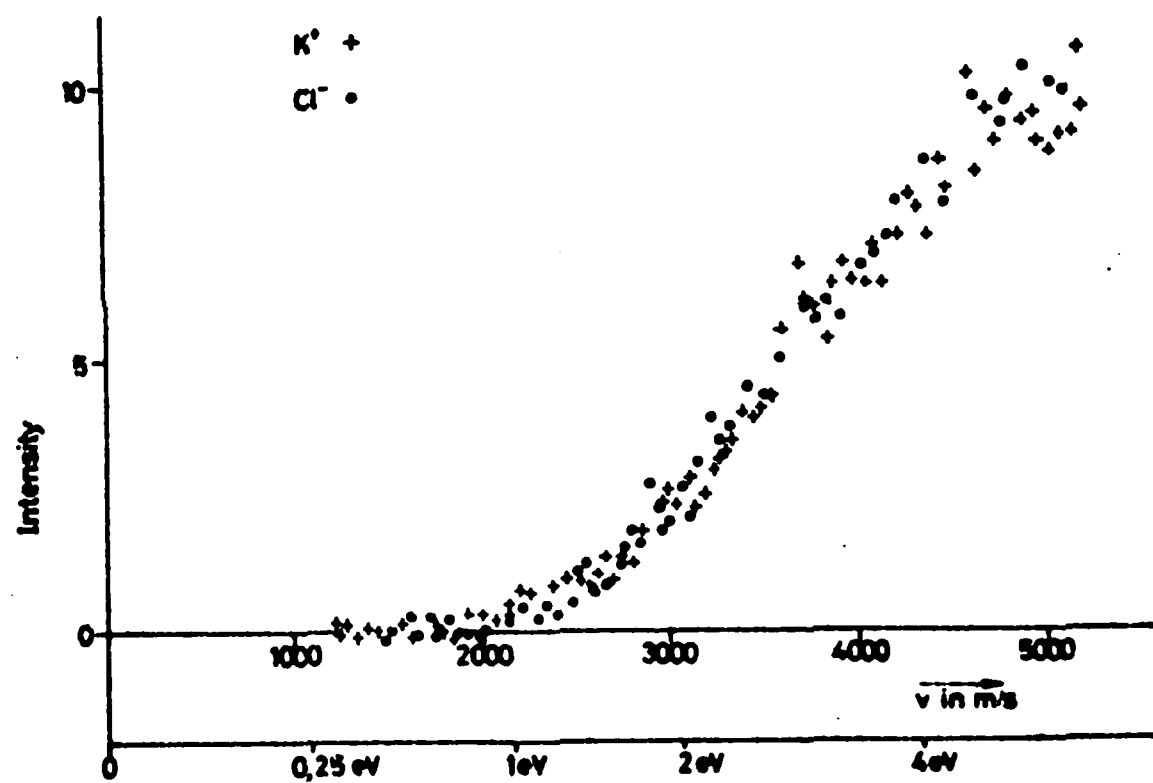


FIGURE 13 The K^+ and Cl^- signals arising from collisions of particles sputtered from a KCl crystal with a crossed beam of aniline molecules ($C_6H_5NH_2$) as a function of the sputtered beam velocity. The kinetic energy in the centre of mass system of KCl and aniline has been indicated.

those for CsCl are similar. These measurements have been performed below the threshold for the reaction $\text{Cl} + \text{C}_6\text{H}_5\text{NH}_2 \rightarrow \text{Cl}^- + \text{C}_6\text{H}_5\text{NH}_2^+$. The M^+ and X^- signals are equal for all velocities which fact is easiest understood if we assume that the process is taking place with the MX constituent of the beam. No sharp threshold has been found for these processes; they take place at centre of mass energies as low as 0.3 eV. Since the energy for dissociation of ground state MX molecules into ground state M^+ and X^- ions is 5.1 eV for KCl and 4.9 eV for CsCl, we have to conclude that at least a fraction of the MX molecules is in an excited state. Nothing is known about the nature of this excitation. The M^+ and X^- signals are proportional to $\exp(-a/v)$, within the experimental error (a is a constant). It is not clear if this exponential behaviour is caused by a collisional effect or if it is a time of flight effect related to the decay of the excited molecules into the ground state. In that case, the lifetime of these molecules would be $5 \cdot 10^{-5}$ s.

ACKNOWLEDGEMENTS

We thank Drs. H. E. Roosendaal for discussions. This work is part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie (Foundation for Fundamental Research on Matter) and was made possible by financial support from the Nederlandse Organisatie voor Zuiver-Wetenschappelijk Onderzoek (Netherlands Organization for the Advancement of Pure Research). One of us (J. G.) thanks the Deutsche Forschungsgemeinschaft for a fellowship.

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