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ENERGY SPECTRA OF ELECTRONS ARISING FROM HEAVY-PARTICLE COLLISIONS IN THE keV REGION

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Synopsis

The energy spectrum of electrons ejected after an inelastic heavy-particle collision is calculated on the assumption that the inelastic energy is statistically distributed among the outer-shell electrons. Together with the measured differential cross section for the inelastic energy this gives an absolute value of the cross section for the production of electrons with a certain kinetic energy. The results are compared with published data of the electron energy spectrum arising from collisions of 100 keV Ar^+ with Ar.

1. *Introduction.* Rudd *et al.*[1]) have measured the absolute differential cross section for the ejection of electrons resulting from collisions of 100 keV Ar^+ ions with Ar atoms, as a function of the electron energy. They noticed a continuous background, superimposed on which structure was visible in the region below 20 eV and between 120 and 220 eV. The high-energy peak was also observed by Snoek *et al.*[2]). The structure is interpreted in terms of the Fano–Lichten electron promotion mechanism[3]) while the background is assumed to originate from collisional ionization as a result of Coulomb interaction. Rudd justifies this interpretation of the continuum by extrapolation of results obtained by comparison of experimental and theoretical electron-spectra in the case of H^+ on H_2 and H^+ on He collisions[1,4]).

Instead of this extrapolation we propose the following model for the calculation of the background. A statistical distribution of excitation energy among the outer-shell electrons is thought to be the basic mechanism[5]). Electrons which get more energy than their ionization potential will be ejected and give rise to the continuum. In ref. 5 two alternative formulae

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are derived predicting the electron spectra. Both models have in common that the residual ion is left in a state of excitation which is stable against further auto-ionization and both are based on a statistical distribution of kinetic energy among the ejected electrons. The two statistical models differ in that one is based on equal *a priori* probability per unit interval of electron energy, while the other is based on equal *a priori* probability per unit volume of momentum space. The first will be referred to as "energy-cell statistics", while the second will be referred to as "phase-space statistics". The number of electrons receiving a kinetic energy in the interval $(E_k, E_k + \mathrm{d}E_k)$, when the excitation energy E_T is distributed among N outer-shell electrons, is given by

$$\frac{\mathrm{d}\mathcal{N}(E_k, E_T, N)}{\mathrm{d}E_k} = \frac{1}{E_1} \frac{\sum_{n=2}^{N} \binom{N}{n} n g^n \left[\frac{(E_T - \epsilon_n - E_k)}{E_1}\right]^{n-2} \Big/ (n-2)!}{\sum_{l=1}^{N} \binom{N}{l} g^l \left[\frac{(E_T - \epsilon_l)}{E_1}\right]^{l-1} \Big/ (l-1)!} \tag{1a}$$

in energy cell statistics and

$$\frac{\mathrm{d}\mathcal{N}(E_k, E_T, N)}{\mathrm{d}E_k} = \frac{1}{E_1} \frac{\sum_{n=2}^{N} \binom{N}{n} n g^n S_{n-1}[(E_T - \epsilon_n - E_k)/E_1]}{\sum_{l=1}^{N} \binom{N}{l} g^l S_l[(E_T - \epsilon_l)/E_1]} \times$$

$$\times S_1\left(\frac{E_k}{E_1}\right) \tag{1b}$$

in phase-space statistics, where ϵ_n is the sum over the first n ionization energies, g is a statistical weight and $S_n(E) = C_n E^{(3n-2)/2}$, where (see ref. 5)

$$C_1 = 1, \quad C_2 = \pi/8, \ \ldots, \ C_{n+2} = 2\pi C_n/[(3n+4)(3n+2)(3n)].$$

It is known from measurements of Kessel and Everhart[6]) and Snoek *et al.*[7]) that the total inelastic energy Q dissipated during collisions with the same collision parameters shows a rather broad distribution around a mean value $\bar{Q}$.

In the model proposed by Everhart and Kessel[8]) a Gaussian distribution in Q is assumed:

$$h = \exp[-(Q - \bar{Q})^2/2a^2]. \tag{2}$$

The half-width, $\sqrt{2}a$, of this distribution at 1/e height is measured as a function of $\bar{Q}$. Their result can be approximated by

$$\sqrt{2}a = 0.260 \times \bar{Q}. \tag{3a}$$

Considering collisions of two identical particles it is reasonable to assume

that each of them, on average, gets half of the total inelastic energy loss:

$$\bar{E}_T = \bar{Q}/2. \tag{4}$$

The distribution in E_T can now be given by

$$h' = \exp[-(E_T - \bar{E}_T)^2/a^2] \tag{3b}$$

with $a = 0.375 \times \bar{E}_T$.

The number of electrons with kinetic energy in the energy range $(E_k, E_k + \mathrm{d}E_k)$ produced during collisions where a mean excitation energy $\bar{E}_T$ is dissipated is given by:

$$\frac{\mathrm{d}\mathcal{N}(E_k, \bar{E}_T, N)}{\mathrm{d}E_k} = \frac{\int \frac{\mathrm{d}\mathcal{N}(E_k, E_T, N)}{\mathrm{d}E_k} \exp[-(E_T - \bar{E}_T)^2/a^2]\, \mathrm{d}E_T}{\int \exp[-(E_T - \bar{E}_T)^2/a^2]\, \mathrm{d}E_T}. \tag{5}$$

For collisions with impact parameters in the interval $(b, b + \mathrm{d}b)$, the mean excitation energy has values in $(\bar{E}_T, \bar{E}_T + \mathrm{d}\bar{E}_T)$. For each $\bar{E}_T$ we can evaluate eq. (5). The absolute differential cross section follows from integration over the impact parameter:

$$\frac{\mathrm{d}\sigma(E_k, N)}{\mathrm{d}E_k} = \int_0^\infty 2\pi b(\bar{E}_T) \frac{\mathrm{d}\mathcal{N}(E_k, \bar{E}_T, N)}{\mathrm{d}E_k} \mathrm{d}b(\bar{E}_T). \tag{6}$$

2. *Calculations.* 2.1. Dealing with the Ar^+ on Ar problem one must calculate the expression (6) for $N = 7$ and $N = 8$ and add the results.

A relation between $\bar{E}_T$ and b is experimentally found by Morgan and Everhart[9]). In eqs. (1a) and (1b) we need, furthermore, the relevant ionization energies. For these we take the values as reported by Lotz[10]).

The solution of eq. (6) is shown in fig. 2. (For energy-cell statistics as well as for phase-space statistics.) It is found that the general shape of the curves is close to the experimental curve measured by Rudd *et al.* and shown here for comparison. The absolute value of the phase-space curve is for low electron energies higher, and for high electron energies lower than the curve which follows from energy-cell statistics.

Both theoretical curves, however, give the absolute value in the right order of magnitude without any fitting or normalization procedure, whatsoever. The strange form of the experimental curve may be due to the fact that the data were obtained from a published graph which was difficult to reproduce.

2.2. Fast electrons. Above 100 eV, the theoretical p.s.s. curve is higher than the experimental one. This can be explained by the fact that in some

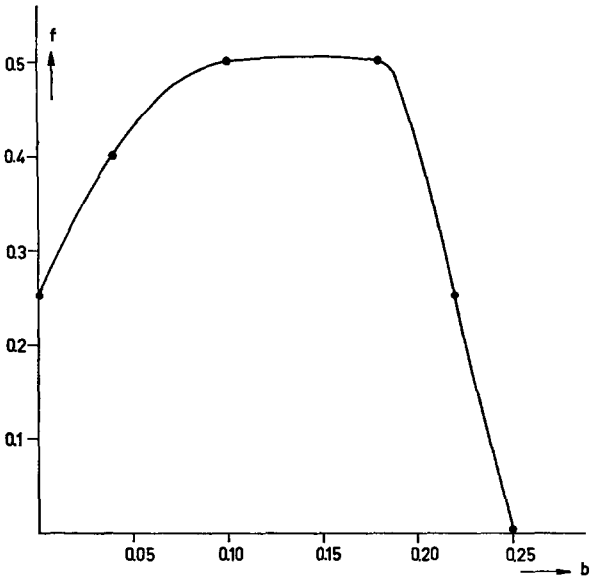

Fig. 1. The probability for the ejection of two fast electrons/mol as a function of impact parameter b in the case of 100 keV Ar^+ on Ar.

of the collisions a large part of the excitation energy E_T can predominantly be carried away by *one* electron. It was shown experimentally that a peak of 200 eV electrons arises in the energy spectrum of electrons ejected during Ar^+ on Ar bombardment[1,2]). There is evidence that collisions with distances of closest approach $\lesssim 0.24$ Å give rise to these fast electrons. In this region an excitation can take place where, by energy-level crossing, one L electron is promoted in an M-shell state as suggested in a paper of Fano and Lichten[3]). They further predict that the vacancy in the L shell, created by the promotion mechanism, will be filled via a two-electron process, in which one electron "falls" into the L-shell vacancy while the second is ejected with an energy of about 200 eV. In our opinion the probability for de-excitation of the L-shell vacancy by this mechanism is smaller than unity. This is already clear from the measurements of Carlson *et al.*[11]) where an L-shell vacancy was created with the help of X rays and the resulting charge-state distribution was studied. To determine the probability for fast-electron production as a function of impact parameter, we used experimental results of Everhart (ref. 8, fig. 5). They measured the mean excitation energy E_T for different ionization processes and compared this with the mean excitation energy as computed by Russek and Meli[5]). For very high excitation energies both values turned out to be the same indicating that the statistical model the collisions can describe in the corresponding impact parameter region satisfactorily. We interpret this as an indication that no fast electron is

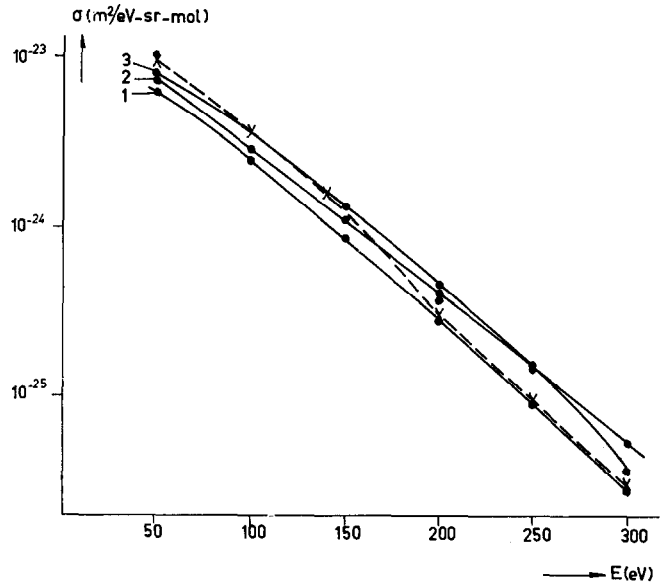

Fig. 2. Experimental[1]) (dashed) and theoretical (solid) electron spectra in the case of 100 keV Ar^+ on Ar.

1. phase-space statistics + fast electron
2. energy-cell statistics
3. phase-space statistics

ejected. On the other hand, for low excitation energies $\bar{E}_T$ the computed values are considerably below the measured ones. Here the interpretation must be that in a fraction of the cases one electron carried 200 eV energy away. The difference between the experimental and theoretical values determines this fraction f. The results obtained with this method are shown in fig. 1. The energy spectrum can now be calculated from eq. (6) with the help of:

$$\frac{\mathrm{d}\mathcal{N}}{\mathrm{d}E_k} = (1 - f(b)) \frac{\mathrm{d}\mathcal{N}(E_k, \bar{E}_T, 8)}{\mathrm{d}E_k} + f(b) \frac{\mathrm{d}\mathcal{N}(E_k, \bar{E}_T - 200, 7)}{\mathrm{d}E_k} +$$

$$+ (1 - f(b)) \frac{\mathrm{d}\mathcal{N}(E_k, \bar{E}_T, 7)}{\mathrm{d}E_k} + f(b) \frac{\mathrm{d}\mathcal{N}(E_k, \bar{E}_T - 200, 6)}{\mathrm{d}E_k}. \quad (7)$$

The resulting electron spectrum using phase-space statistics is shown in curve 1 of fig. 2. As expected the absolute cross section is lower than in the case where no fast electron is considered.

3. *Discussion.* 3.1. Residual excitation energy. The formulae in ref. 5 are such that a value for the residual excitation energy can be dictated. To be consistent with the concept of an auto-ionizing transition, this

must not be greater than the next ionization energy. In the calculation presented in this paper it is taken equal to the next ionization energy since excited states in general have energies rather close to the ionization energy. For instance, if we consider the probability that of four escaping electrons, one electron gets a kinetic energy E_k, we take the energy space available to the 3 other electrons equal to $E_T - E_k - (E_1 + E_2 + E_3 + E_4 + E_5)$; in which E_5 is the approximation here used to the residual excitation energy and E_i is the ith ionization potential. This choice is justified by the fact that most excited states of the residual ion have energies near the ionization potential. Taking on a lower value will shift the energy spectrum to higher energies. For instance, the other extreme assumption; dictating a residual excitation energy equal to zero leads to a horizontal shift of about 18 eV towards higher energies, in the electron energy spectrum.

3.2. Fast electrons. With the help of the function giving the probability for fast electron ejection *versus* impact parameter (fig. 2), one can calculate the absolute cross section for this process using

$$\sigma = 2\pi \int_0^\infty f(b)\, b\, \mathrm{d}b. \tag{8}$$

We find in this way a value of 1.57×10^{-21} m^2/mol.

From the measurements of Rudd one can also calculate this cross section by integrating the area under the high-energy peak. This method yields a value of 2.7×10^{-21} m^2/mol[12]). Given the uncertainty of 50% in this figure both results can be considered to be in the same order of magnitude.

3.3. Comparison with other theories. In a recent paper of Orgutsov *et al.*[13]), a model is proposed to explain the continuous part of the spectrum. They suppose that these electrons arise from ionization *during* pseudo-molecular formation. Because of the continuous change of states during the collision the spectrum is predicted to be continuous. However, this model seems to be in contradiction with the coincidence measurements of Everhart and Kessel[8]), where in general no correlation was found between the charge states of the scattering products. Furthermore, a quantitative description is rather hard to perform while the intermediate states are not well known. So also in this case a statistical setup seems to be the only method of attack. Then one can expect that the result will not differ drastically from ours because the distribution of inelastic energy in 2 successive steps, the first during pseudo-molecular formation ($N = 15$) and the second after separation is roughly the same as statistical distribution in one step.

Rudd *et al.*[1]) do indicate an alternate way of obtaining the continuous part of the spectra. The Born approximation should be used to calculate direct ionization by Coulomb interaction. However, it is doubtful whether a scaling of the problem 100 keV H^+ on H_2 to 100 keV Ar^+ on Ar in this ap-

proximation is valid. On the other hand the statistical model as proposed in this paper cannot be used in the H^+ on H_2 case, because obviously there is no auto-ionizing state which yields two or more electrons. Just one electron will be emitted with a specific energy. The statistical model, however, turns out to describe the heavy-atom collisions satisfactorily thus indicating that here violent collisions contribute mostly to the cross section in the continuum.

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