

**TOTAL DIFFERENTIAL SCATTERING CROSS SECTION OF  $\text{Ar}^+$ -Ar AT 15 TO 400 keV****H. HARTUNG, B. FRICKE, W.-D. SEPP, B. THIES, D. KOLB, D. HEINEMANN***Department of Physics, University of Kassel, D-35 Kassel, FRG*

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We report on the measurement of the total differential scattering cross section of  $\text{Ar}^+$ -Ar at laboratory energies between 15 and 400 keV. Using an *ab initio* relativistic molecular program which calculates the interatomic potential energy curve with high accuracy, we are able to reproduce the detailed structure found in the experiment.

Knowledge of the potential energy surfaces of a molecule is the key to the interpretation of many molecular properties. The same statement holds true for a quasi-molecule formed during a scattering process, where the knowledge of all potential energy curves for all excitations plus knowledge of the connecting matrix elements between these channels would, in principle, allow a complete description of the scattering process.

Because the potential energy curves play such an important role in molecules as well as quasi-molecules, their determination is one of the main theoretical as well as experimental tasks. The theoretical difficulty in calculating a realistic potential energy surface is, of course, the complexity of the many-particle problem which, in addition, has to include relativity for heavy systems. Quantum chemists are now able to determine potential energy surfaces for many systems and many excitations with great accuracy. An almost complete list of references is given in the book by Schaefer III [1]. Their methods, however, are only suitable for small  $Z$  systems and for internuclear distances in the range of chemical distances or larger.

For light as well as heavy systems the region just below chemical distances is a field where our knowledge is still very limited. The aim of this letter is to

determine the accurate ground-state interatomic potential energy curve for the relatively heavy system  $\text{Ar}^+$  on Ar at the internuclear region from chemical distances to about 0.1 au which plays the most important role at scattering energies between 15 and 400 keV.

Up to now the information on the potential energy curves in this region of internuclear distances comes from interatomic screening functions [2-4]. Among the atomic screening functions derived from the Thomas-Fermi (TF) function [5] are the Lenz-Jensen screening function [6,7] and the Moliere function [8]. The last two reproduce experimental results on the average even better than the TF function, because they represent a smooth analytical fit to the TF function for a group of diatomic systems in a large range of interatomic distances. However, they do not reproduce any detail or structure. For all these screening functions the deviations from realistic values become very large in the region of chemical distances.

A number of experimentally measured total differential scattering cross sections exist already for heavy systems [9], which show a lot of unexplained structure. Probably these observed structures reflect the molecular shell structure of the many-electron sys-

tems during heavy-ion scattering [9]. The first quantitative indications that this might be the correct interpretation has been given by Hartung et al. [10] who calculated the Ne-Ne ground-state potential energy curve and obtained a very good agreement with the experimental differential scattering curve. While the influence of inelastic effects thus seems small in the Ne-Ne case, the  $\text{Ar}^+ - \text{Ar}$  situation might differ, as inner shell excitation takes place with high probability [11].

The purpose of this letter is to show that these very first results are indeed not accidental, and that such a system with 35 electrons, already affected by relativistic effects at smaller internuclear distances, can be well described by theory.

The scattering chamber used for the measurements is a modified version of that shown in the paper by Fastrup et al. [11]. All scattered particles are detected without energy analysis in absolute measurements. Laboratory scattering angles are varied from 1 to 10 degrees, and the resolution is about 0.05 degrees. The absolute uncertainty of the measured total differential cross sections is about 8% and the relative uncertainty, comparing various energies and angles, is about 4%.

The theoretical description is based on a fully self-consistent relativistic Dirac-Fock-Slater diatomic molecular code which was developed during the last years. Details can be found in the paper of Sepp et al. [12]. This code has an overall accuracy of  $10^{-5}$  to  $10^{-6}$  which allows a determination of the total energy at every internuclear distance with an accuracy of the order of eV. This approximation is sufficient for the interpretation of the experiment in contrast to chemical problems, where usually a very high accuracy is needed. Further increase of the numerical accuracy within the method used may not be meaningful because the error of the Slater-type approximation of the exchange term in the Fock operator might be larger than the numerical error.

The basis functions used in the calculations are numerical Dirac-Fock-Slater (DFS) atomic wavefunctions of Ar from  $n=1$  to  $n=3$ . In addition 11 wavefunctions from an atomic DFS calculation using the monopole part of the molecular nuclear potential ( $Z=2 \times 18$ ) are added. Furthermore, basis functions are added which result from atomic DFS calculations where the nuclear charge is used as a

parameter such that the resulting wavefunctions have their outermost maxima at a distance which is about 20% larger or smaller, respectively, for the valence electrons in the quasi-molecule. In total we used 50 basis functions. The completeness of this basis was checked by introducing further wavefunctions, but all additions changed the results by very small amounts, only. Special care had to be taken to ensure the orthogonality of the basis set because spurious states originating from the numerical uncertainties in the process can show up sometimes.

As a first result of such calculations we present in fig. 1 the relativistic, adiabatic correlation diagram of  $\text{Ar}^+ - \text{Ar}$ . An empirical correlation diagram of this system [13] has been discussed as an example in many papers. A non-relativistic version using the variable screening method with a Thomas-Fermi potential for the screening has been published by Eichler et al. [14]. Our correlation diagram, fig. 1, confirms that one of the diabatic energy levels correlating to the 2p level of the separated atoms (usually called the  $4f_\sigma$  level) rises steeply through adiabatic pseudocrossings for  $R < 0.7$  au, thus causing the known sudden increase in the inelastic energy loss in this region.

The semi-empirical Lenz-Jensen potential  $V_{\text{aLJ}}$  as parametrized by Loftager et al. [9] is a good first approximation to the scattering potential. This  $V_{\text{aLJ}}$ -potential curve is structureless and smooth and provides a good basis for comparison. We therefore present the scaled difference between our self-consistent field (SCF) result and  $V_{\text{aLJ}}$  in fig. 2. One notices that the calculated realistic potential energy curve contains a lot of structures. The "molecular binding" of the electrons manifesting itself in this structure varies for different internuclear distances. The large minimum at 2.2 au probably reflects the "molecular binding" between the  $n=3$  shells of argon, whereas the minimum at 0.7 au is due to the overlap and rearrangement of the  $n=2$  electrons. The minimum at 0.4 au is probably due to the same effect, namely the  $n=2$  and  $n=1$  electrons forming molecular states thus inducing an increase in binding energy.

With this information on the SCF potential energy curve we performed a classical trajectory calculation to determine the differential elastic scattering cross section  $d\sigma/d\Omega$  as function of the center of mass angle  $\theta$ . To see the effect of the structure in the potential

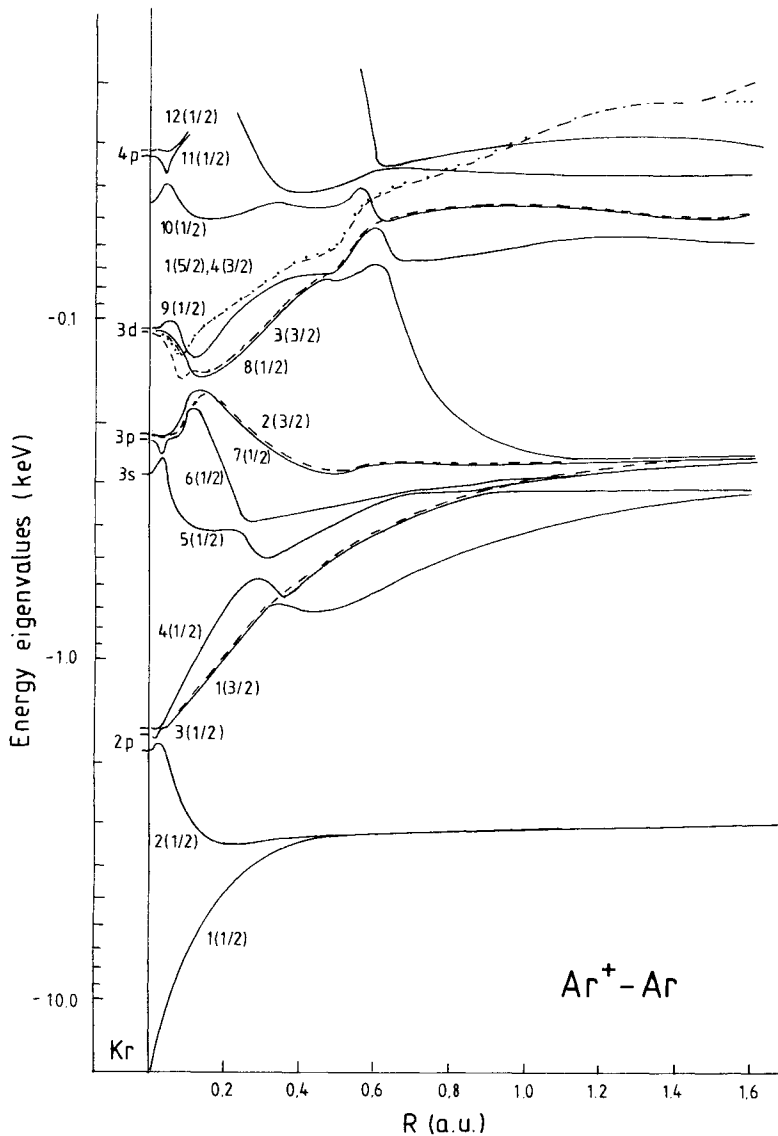


Fig. 1. Relativistic adiabatic correlation diagram of the system  $Ar^+ - Ar$ .

energy curve we also calculated  $d\sigma/d\Omega$  for the Lenz-Jensen potential. The result is shown in fig. 3. The ratio  $(d\sigma/d\Omega)_{SCF}/(d\sigma/d\Omega)_{aLJ}$  is plotted as function of the dimensionless quantity  $s$  from the theory of Lindhard et al. [2]:

$$s = \epsilon \sin(\frac{1}{2}\theta),$$

where  $\epsilon$  is the reduced center of mass energy

$$\epsilon = E_{CM} a / Z_1 Z_2,$$

and  $a$  is the screening length in atomic units

$$a = \frac{1}{3} (3\pi/4)^{2/3} (Z_1^{2/3} + Z_2^{2/3})^{-1/2}.$$

The experimental structures measured for laboratory energies from 15 to 400 keV are reproduced very well by the theoretical ab initio SCF calculation. The maxima in the difference of the potential energy curves in fig. 2 at 1.5, 0.6, and 0.2 au are the reason

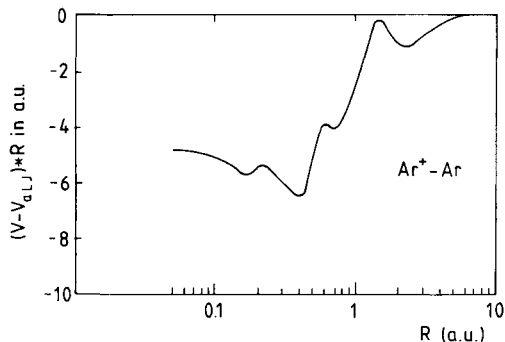


Fig. 2. Difference of the SCF potential energy curve and the average Lenz-Jensen potential  $V_{aLJ}$  by Loftager et al. [9].

for the maxima in the ratio of the differential scattering cross sections of fig. 3. The shape and width of all maxima in the calculated differential cross sections are very sensitive to the form of the potential energy curve. This statement is especially valid for the outermost maximum with  $s$  just below 0.01 which is determined by the form of the maximum in the potential energy curve at 1.5 au. The good agreement between experiment and theory shows the accuracy of both.

Although we know that a lot of electron excitation occurs during the collision and that a spectacular increase in inelastic energy loss (from 200 to nearly 800 eV) takes place around  $R_0 = 0.5$  au, this obviously does not affect the position of the observed maxima. This behaviour is probably caused by the fact in the vicinity of the point where the major part of the deflection occurs the shape and not the absolute value of the potential energy curve mainly determines the position of the maxima. Even if electron excitations are included in the potential energy curve the slope seems to be well described by the ground-state potential energy curve. On the other hand the influence of the electron excitations on the potential energy curve is probably the cause of the slight disagreement between experiment and theory in the region between the maxima in fig. 3.

One remaining question is where do the relativistic effects come in? To answer this, one has to compare the non-relativistic radii with the relativistic ones of the  $n=2$  electrons of argon which cause the peak at  $s=0.5$ . The non-relativistic radii are larger by 4%, this should shift the innermost maximum in the potential energy curve to somewhat larger radii.

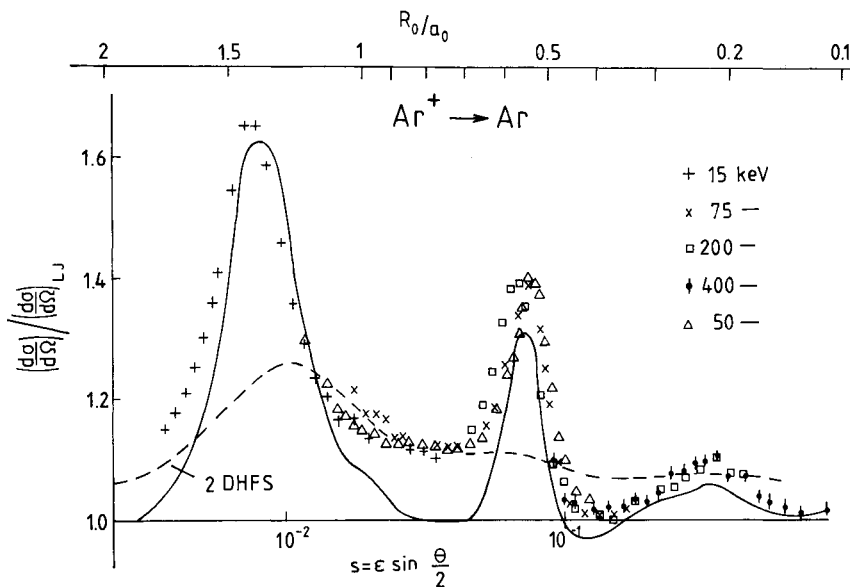


Fig. 3. Experimental total differential scattering cross sections for  $\text{Ar}^+ - \text{Ar}$  scattering at 15 to 400 keV impact energies. The dimensionless quantity  $s$  is explained in the text. The solid curve is the result of the elastic differential cross section calculation with the SCF potential from fig. 2. The 2DHFS curve is a more crude theoretical estimate as explained by Loftager et al. [9]. The experimental as well as the theoretical values are all normalized to the cross section based on the original Lenz-Jensen screening function by Lenz [6] and Jensen [7]. The upper part of the figure gives the distance of closest approach  $R_0$  in au.

In turn this would then shift the peak towards slightly smaller  $s$  values. This effect would, of course, be much more drastic in systems with larger  $Z$ .

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