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Excitation processes in H^--Kr collisions

V. A. Esaulov
Laboratoire des Collisions Atomiques et Moléculaires, Bâtiment 351, Université de Paris-Sud, 91470 Orsay, France

J. P. Grouard, R. I. Hall, and J. L. Montmagnon
Groupe de Spectroscopie par Impact Electronique et Ionique, Université Pierre et Marie Curie, 4 place Jussieu, T12-E5, 75005 Paris, France
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An electron spectroscopy study of H^--Kr collisions in the 20–500-eV collision energy range revealed the excitation of $(2s^2)^1S$, $(2p^2)^1D$, and $(2s2p)^1P$ states of H^- and of the $(5s^2)^2P_{3/2,1/2}$ Kr^- state. The angular distribution of electrons produced from the decay of these states is reported. An anisotropic distribution for the $(2s^2)^1S$ state is observed at these energies as was previously reported for H^- collisions with He, Ar, and H_2 by Risley and co-workers [Phys. Rev. A **9**, 1115 (1974) and IEEE Trans. Nuc. Sci. **NS26**, 1027 (1979)]. Relative cross sections for the excitation of these states are deduced from the angular distributions.

INTRODUCTION

Negative-ion collisions have been studied quite extensively over the past years. Considerable progress has been done in identifying the possible detachment channels and significant progress in the description of direct detachment, i.e., the process

$$A^- + B \rightarrow A + B + e^-$$

has been achieved.[1] On the other hand, excitation processes

$$A + B \rightarrow A^*(A) + B(B^*) + e^-$$
$$\rightarrow A^{-*}(A) + B(B^{-*})$$

have not been the subject of similar attention. On the experimental side there exist some measurements of total cross sections of target atom and stripped projectile atom excitation (optical measurements) as well as some electron spectroscopy studies of autodetaching state production reporting mainly electron spectra and in a few cases electron angular distributions. Total cross sections for autodetaching state production are limited to the H^- case.[2]

There do not appear to exist any reasonably satisfactory theoretical descriptions of excitation processes. Attempts[3,4] have been made to describe these in qualitative terms in the framework of the electron promotion model.[5] However, the applicability of this model to negative-ion collisions does not appear obvious because of the loosely bound nature of the outer negative-ion electron. Indeed a somewhat different model was proposed by Esaulov *et al.*[6] to describe excitation processes in H^- collisions with inert gases (IG). In this model, which appears conceptually compatible with Demkov's[7] zero range potential (ZRP) model for detachment, the outer H^- electron and the H-IG quasimolecular core were described independently. Excitation processes were described in the framework of the molecular orbital (MO) model insofar as the core collision[8] is concerned. The outer electron was then assumed to be either detached or recaptured in the outward leg of the collision. In the latter case production of autodetaching states is possible.

$$H^- + He \rightarrow [H + He \rightarrow H^* + He] + e^-$$
$$\rightarrow H^{-*} + He \, .$$

This model was successful in describing some of the general features, such as summed differential excitation cross sections and total detachment cross sections for H^+ production in collisions with He (Ref. 8) (see Refs. 6 or 1 for details). It does not appear obvious, however, even in the particular case cited above, that the division into an outer independent electron and an independently evolving core is always possible. This assumes that the range of internuclear distances at which excitation processes occur is smaller than that at which detachment occurs, i.e., the outer electron may be considered as being loosely bound and independent. Thus further theoretical work is necessary to understand the details of excitation processes in negative-ion collisions.

In this context detailed experimental data that can provide sensitive tests for theory are necessary. A good example of such data is provided by a study of the production of autodetaching states in H^-—inert-gas collisions initiated by Risley[9] and Edwards.[10] Of particular interest are studies of H^- collisions with Ar, Kr, and Xe, where excitation of autodetaching states of both the projectile and the target is possible and a study of this sharing would be particularly instructive. Such a study was begun by Risley,[11] who measured detached electron energy spectra for H^--Ar, -Kr, and -Xe collisions which display peaks due to projectile and target anion autodetaching states. However, these measurements do not give any idea of the relative magnitude of cross sections for the production of these states and their evolution as a function of collision energy. In the present work we present the first results of such a study of H^--Kr collisions for laboratory impact energies between 20 and 500 eV.

EXPERIMENTAL PROCEDURE

An electron spectroscopy study of the production of autodetaching states was performed on the apparatus previously described by Montmagnon *et al.*[12] Electron spectra were obtained in the 15°–135° angular range. One of the problems that has to be taken into account here concerns the kinematic shift of the peaks in the electron spectra due to the production of H^- autodetaching states. Thus at 500 eV, in the studied angular range, the total shift for the H^- $2s^2\,{}^1S$ line (at the unshifted energy of 9.58 eV) is from 7 to 13 eV. When performing measurements of the relative population of the H^- and Kr^- lines as a function of laboratory ejection angle, the transmission function (i.e., the variation of transmitted electron intensity as a function of electron energy) has to be known. This was determined from a study of $e^- + He$ elastic scattering in the above energy range. A comparison of our experimental data and the theoretical data of Andrick and Bitsch[13] allowed us to determine this function.

The energy resolution of the electron spectrometer was about 50 meV. It should be noted that the H^- lines are broadened by kinematic effects. Peak areas were therefore used to determine the relative magnitude of H^- and Kr^- peaks at each angle.

RESULTS

Spectra

Figure 1 shows a typical electron spectrum for H^--Kr collisions for a 100-eV collision energy and 15° electron ejection angle. As can be seen in the figure, excitation of the Kr^- ${}^2P_{3/2}$,${}^2P_{1/2}$ states and the H^- $(2s^2){}^1S$, ${}^1D$, and ${}^1P$ states is observed. These peaks were observed down to the lowest studied collision energy of 20 eV. No other autodetaching states were observed. The H^- lines are shifted and broadened due to kinematic effects. For small energies and small scattering angles these lines were found to be asymmetric in shape. This asymmetry may be due to interference effects (see, e.g., Ref. 11). Kinematic effects due to scattering into a finite angular cone of the H atoms could also alter the shape of these peaks but this effect is difficult to estimate without the knowledge of this distribution.

Angular distributions

Figure 2 shows a typical angular distribution corrected for kinematic effects and the transmission function of the analyser, and normalized to unity at 80°. Because of kinematic effects, peaks due to decay of the Kr^- and H^- autodetaching states were superposed at some laboratory ejection angles. A simple deconvolution procedure was adopted to determine the area of the Kr^- ${}^2P_{3/2}$ peak, assuming a Gaussian form for it. Because of the very small magnitude of the Kr^- ${}^2P_{1/2}$ peak this was not always possible for this state. The angular distribution for this state could thus not be determined and we have assumed an isotropic distribution (see Ref. 10 for a discussion of the angular distribution for the case of H^--Ar collisions).

The Kr^- ${}^2P_{3/2}$ angular distribution was found to follow a $\cos^2\theta$ dependence. Neglecting H-atom scattering into a finite angular range, this distribution may be expressed[10] in terms of magnetic sublevel populations $P(J,M_J)$ as

$$\sigma(\theta)=\frac{3}{4\pi}\{[P(3/2,3/2)+P(3/2,1/2)/3] + [P(3/2,1/2)-P(3/2,3/2)]\cos^2\theta\} .$$

The dashed line in Fig. 2 corresponds to a least-squares fit to the data giving at 500 eV the ratio of the $M_{J=3/2}$ popu-

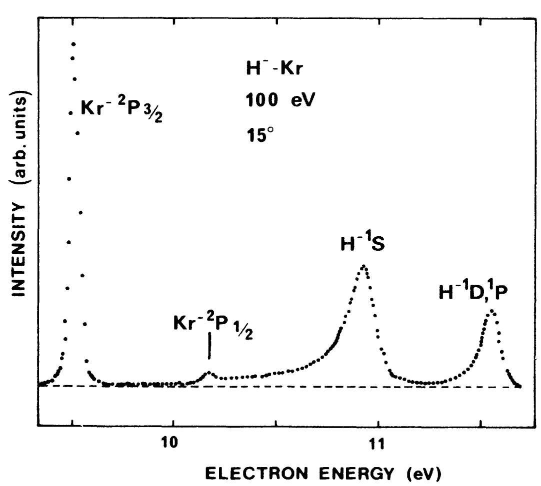

FIG. 1. Electron spectra produced in H^--Kr collisions showing peaks due to autodetaching states of H^- and Kr^-. Note that the H^- peaks are kinematically shifted to higher energies in the laboratory frame of reference.

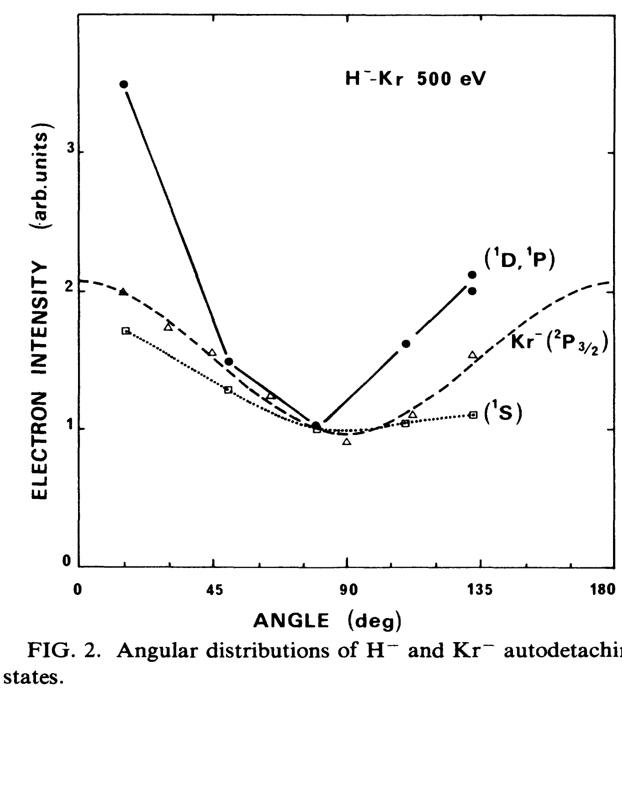

FIG. 2. Angular distributions of H^- and Kr^- autodetaching states.

lation to the $M_{J=1/2}$ population for the $^2P_{3/2}$ state as

$$\frac{^2P(3/2,3/2)}{^2P(3/2,1/2)}=0.29 \ .$$

The angular distributions of the H^- $(2s^2)^1S$ state is found to be anisotropic (forward peaked) at our collision energies. This feature has been previously noted in case of the He, Ar, and H_2 targets[9–11] but remains unexplained.

Cross sections

In order to obtain the population of the $^2P_{3/2}$ state as a function of energy, measurements were performed at 125.3° since here

$$\sigma(\theta)=(2\pi)^{-1}[P(3/2,3/2)+P(3/2,1/2)] \ .$$

This was only possible for energies above 100 eV where this peak was sufficiently resolved from the H^- peak. The resulting cross section is shown in Fig. 3. The cross section for the $^2P_{1/2}$ state was obtained with the assumption of an isotropic distribution as mentioned above. Integration over angle of the angular distribution of the H^- states yields, after normalization to the Kr^- $^2P_{3/2}$ state, the cross section shown in Fig. 3.

On the basis of the available data it appears rather difficult to comment on these cross sections since, in particular, data on related reactions such as

$$H^- + Kr \rightarrow H^- + Kr^*$$

are not available. Knowledge of cross sections for excitation processes in H + Kr collisions would also be useful in order to understand the general characteristics of these collisions and delineate the specific characteristics of the negative-ion system. Extensive photoemission data on H excitation in H-Kr collisions have recently become available,[14] but no data concerning Kr excitation exist. Thus at present one can only make some general basic statements which follow from a quasimolecular model that should be applicable in the case of the neutral system and could also serve as a guideline to understand the anion system in virtue of the discussion of the H^--He collision in Ref. 6.

One can expect that excitation processes will occur as a result of the promotion of the outermost H orbital ($\sigma 1s_H$ or $4f\sigma$) leading to radial- or rotational-coupling-induced transitions to higher-lying orbitals. On the outward leg of the collision rearrangement in the core $[(HKr)^+ \rightarrow H^+Kr$ or $H\text{-}Kr^+(^2P_{3/2,1/2})]$ can lead to the population of either H or Kr excited states. An earlier study of charge exchange in $H^* + Kr$ collisions[15] showed that in the present energy range charge exchange to Kr^+ is quite important and that mainly Kr^+ $(^2P_{3/2})$ production is expected. Hence in our case large cross sections for Kr and Kr^- excited states may be expected and these should mainly correspond to the Kr^+ $(^2P_{3/2})$ core. This is indeed observed in our measurements. In order to understand the shape of the cross sections, as well as the characteristics of the angular distributions, considerable further experimental and theoretical work is necessary.

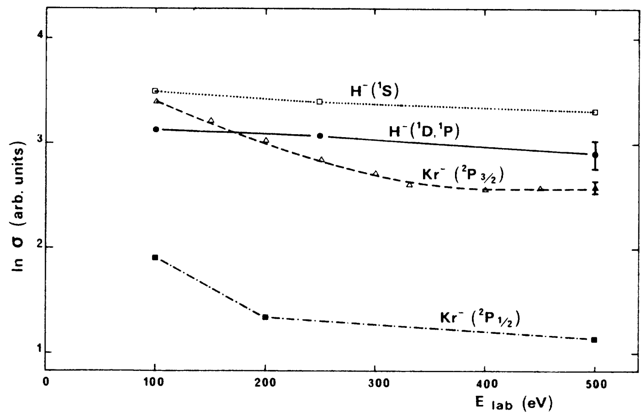

FIG. 3. Relative cross sections for the production of autodetaching states of H^- and Kr^-.

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