Energy-loss scaling in $Li^+ + D_2$ collisions

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$Li^+ + D_2$ collisions are investigated in an energy range from 0.600 to 3.0 keV and for τ (the reduced scattering angle equal to $E\theta$) values out to 2.5 keV deg. The electronically elastic channel is found to be dominant and is studied in detail as a continued test of a scaling law which predicts that a reduced energy loss f should depend only on τ. Earlier results that scaled were particularly useful since they could test the validity of a calculated potential-energy surface. Although we clearly find the scaling to break down at small scattering angles, a related theoretical study shows that the calculated surface may still be tested by the experimentally determined $f(\tau)$. Electronically inelastic processes resulting from singly as well as doubly excited states of the D_2 are also found. The two inelastic processes occur with comparable probabilities which rise to about 0.2 at the largest τ values.

INTRODUCTION

Detailed studies of collisions, particularly in the past 20 years, have given us a basic understanding of the interactions that occur between ions (or atoms) and atomic targets. A number of studies have also addressed problems involving interactions of ions (atoms) with diatomic molecular targets. The present work on $Li^+ + D_2$ at energies in the range of 600 eV to 3.0 keV probes the underlying interactions in the system and, in particular, tests a recently calculated $(LiD_2)^+$ ground-state potential energy surface. The experimental results are presented here and a theoretical analysis[1] is presented in a second paper.

A long-standing major goal of atomic and molecular physics has been a determination of the interparticle potential energies for ion-atom and atom-atom complexes. Experimentally the potential energy as a function of interparticle separation has generally been obtained from measurements of the energy dependence of the total cross section or from the angular dependence of differential cross sections. Similar measurements on ion–diatomic-molecule and atom–diatomic-molecule systems can provide information on the potential-energy surfaces of the triatomic molecule which is formed during a collision. Because of the added complexity introduced by the vibrational and rotational motions, collisions involving molecules are inherently more difficult to understand. As has been shown, however, studies of the collisional excitation of the vibrorotational degrees of freedom of the molecule can provide an alternate approach to testing calculated potential energy surfaces for ion (atom)–homonuclear-diatomic-molecule systems.[2,3]

This study is one in a series initially undertaken[4] to test a scaling law for energy loss in the electronically elastic collision channel. The law was originally derived[5] under restrictive conditions that were applicable to collision systems where purely electronically elastic scattering occurs. In addition, the target molecule was assumed not to vibrate or rotate during the collision, and also, the scattering is only into small angles. The law states that under the conditions assumed, a "scaled energy loss"

$$f = \tfrac{1}{2}(1 + Q/T) \tag{1}$$

is a function of $\tau = E\theta$ only. E is the beam energy, Q is the most probable vibrorotational excitation energy of the homonuclear-diatomic-target molecule and is the difference between ΔE, the measured energy loss, and T, the energy loss for purely elastic scattering.[4] The neon–H_2,D_2 systems met the restrictive conditions and the scaling was verified.[4] A later paper generalized the law[6] but still predicted that the scaled energy loss is a function of τ only. In an investigation of $He + D_2$, strong electronic excitation of D_2 was found even at small angles. The scaling was again obeyed[3] showing that the restriction to electronically elastic collisions was unnecessary. It is important to note that by using the techniques of Snyder and Russek,[2] the experimentally determined $f(\tau)$ tested the validity of the calculated ground-state potential-energy surface. The same potential was used to calculate a differential cross section for elastic scattering which was found to be in good agreement with experiment.[3] In a recent study of $Ar + D_2$ the analysis[7] was extended to include higher-order terms in an expansion of the energy loss. The experimental results again showed scaling even at energies where the vibrational motion of the molecule is non-negligible during the collision. In addition, our findings showed the presence of strong electronically inelastic channels and scattering at large angles in the center of mass. The scaling was found to be valid under conditions that were disallowed in its original derivation.

The present experimental study finds that the energy-loss scaling, which was expected to be valid, breaks down in $Li^+ + D_2$ at small angles. The related theoretical work,[1] however, shows that the ground-state potential-energy surface may still be obtained from the experimentally determined $f(\tau)$ behavior.

EXPERIMENTAL TECHNIQUES

Our studies require measurements of the energy lost, by the Li^+, as a function of scattering angle at a number of incident beam energies. The experimental arrange-

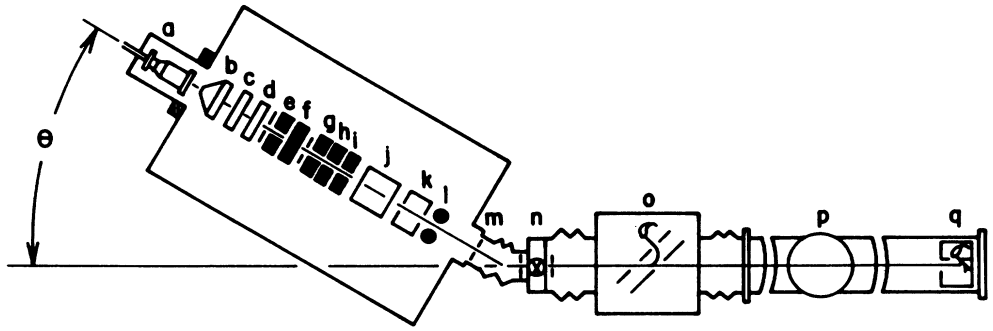

FIG. 1. The experimental arrangement. Ion source (a), extractor and einzel lens system (b, c, and d), deflector plates (e–i), Wein filter (j), charge-exchange cell (k, not used), deflector plates (l, grounded), scattering cell (m), valve (n), electrostatic energy analyzer (o), cryopump (p), and time-of-flight detector (q).

ment used for the present measurements is shown in Fig. 1. Except for several changes, cited below, it has been basically described in Ref. 3. $^7Li^+$ is generated in a spectromat ion source (a, in place of a Colutron used in Ref. 3), extracted, and focused by an einzel lens (b, c, and d). The beam passes through shim fields (e and f) and a collimating hole into a beam "chopping" region (g) used in time-of-flight measurements. The Li^+ passes through additional shim fields (h and i) into a Wien filter (j) for mass identification, and through a charge-exchange cell (k, not used in this work) and two grounded electrodes (l). The beam enters a small cell (m) containing D_2 or He and scatters through an angle θ into the detector chamber. The Li^+ is then energy analyzed by a parallel plate electrostatic energy analyzer (o) having a resolution of 0.5 eV per 1000 eV. The beam is detected by a channeltron electron multiplier. In studies of electron capture where the projectile is neutralized, the Li passes through the analyzer into a flight tube to a detector (q) for energy analysis by time-of-flight techniques. During the course of this study, a 6-in. oil diffusion pump originally on the source chamber was replaced by a 1500 l/sec turbomolecular pump which resulted in substantially improved stability for Li^+ beams.

The basic experimental difficulty involves the measurement of a small projectile energy loss at a scattering angle that must be accurately known. Although the apparatus incorporates a vernier scale which can be read to 1.0 min of arc and which is suitable for the typical "high-resolution" differential cross-section measurement, its use is limited here since even small uncertainties in the scattering angle can give rise to unacceptable errors in the computed f values. In this work the scattering angle is determined from energy-loss measurements on the elastic channel in $Li^+ + He$ collisions. It is well known that in an elastic collision between a projectile of mass m and a target of mass M, an amount of energy

$$\Delta E = (m/M) E \theta^2 \tag{2}$$

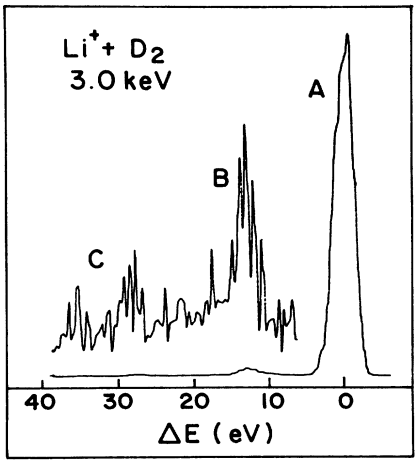

FIG. 2. A typical small-angle energy-loss spectrum. Peak A corresponds to the electronically elastic process. Peak B results from single electron excitation of the D_2 target. Peak C, with a threshold excitation energy near 28 eV, is due to excitation of doubly excited and ionizing states in the target.

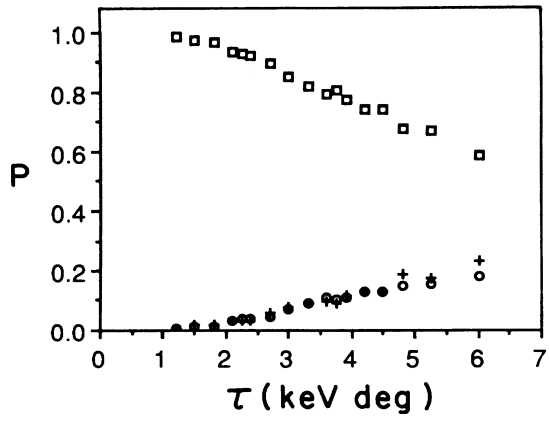

FIG. 3. The probabilities of excitation of the processes corresponding to peaks A (□), B (○), and C (+) at 3.0 keV. The same probabilities are found at 2.0 keV but the data are taken over a smaller range in angle and are not plotted. The probabilities of excitation of the singly and doubly excited states of D_2 are seen to be comparable.

is lost by the projectile of initial energy E in scattering through a small angle θ. By simply measuring E and ΔE for the $Li^+ + He$ collisions and using the known masses of the projectile and target the scattering angle θ can be determined from Eq. (2). The He is then replaced by D_2 target gas, the ΔE for D_2 is measured, and f is determined at the "computed" scattering angle. The data analysis is particularly simple in this case since D_2 and He have the same mass. To take a data point, the "scattering angle" is approximately set using the vernier scale on the apparatus. Since the scattering angle is actually determined from the $Li^+ + He$ measurement a particular angle could not be precisely set prior to acquisition of a data point (it was only determined after the $Li^+ + He$ measurement). Since the angles could not be precisely preset and because of the finite angular resolution ($\cong 0.1$ deg, FWHM) it is necessary to report f values which are averaged over small angular ranges. This results in damping of the rapidly varying angular structure in $f(\tau)$ which is predicted by the theory.[1]

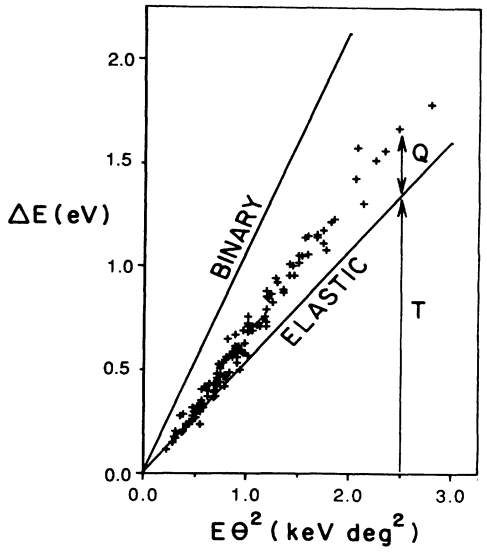

FIG. 4. The quasielastic energy loss ΔE vs $E\theta^2$ for $Li^+ + D_2$ at 1.0 keV. At a given $E\theta^2$ value the difference in energy between the measured ΔE and the elastic limit curve represents the vibrorotational excitation energy Q. The binary limit curve corresponds to energy losses which would result from a collision with one D atom in the molecule.

EXPERIMENTAL RESULTS AND CONCLUSIONS

Our results on $Li^+ + D_2$ show that in the τ region studied the collision is primarily elastic. Figure 2 is a typical small-angle energy-loss spectrum. The peak which is labeled A results from electronically elastic collisions. It is

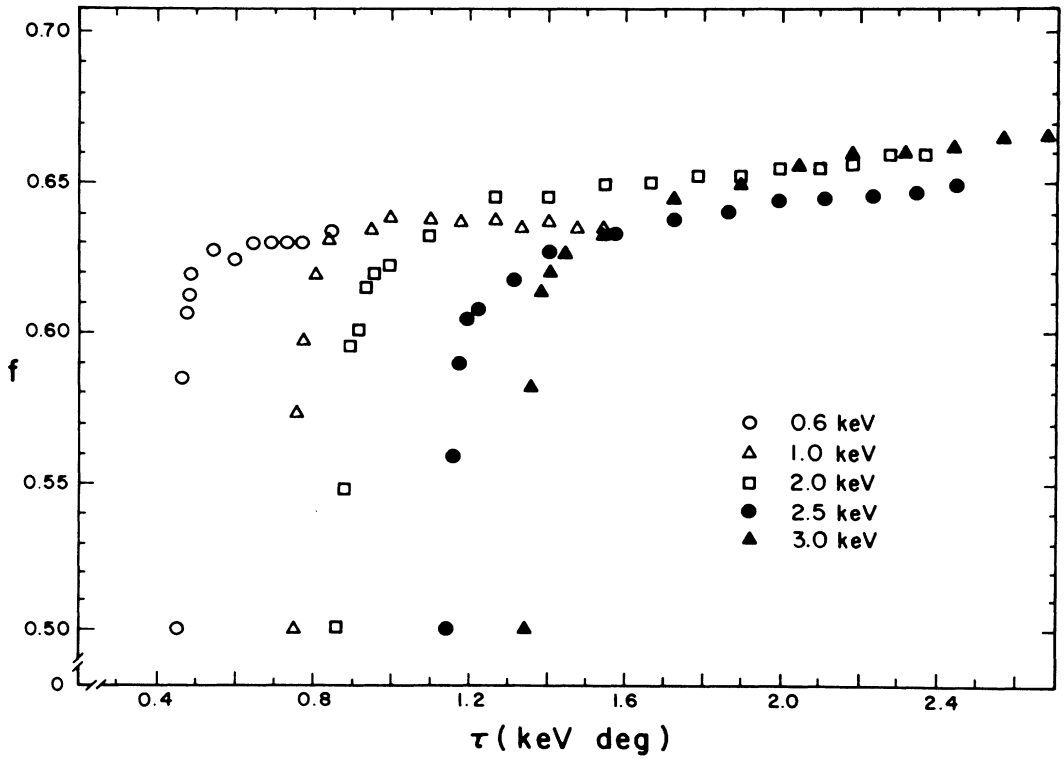

FIG. 5. The averaged scaled energy loss f vs the reduced scattering angle τ for $Li^+ + D_2$ collisions. The data, especially at small τ, cannot be fit by a common curve as required by the scaling law. Energy-loss scaling is seen to break down in this collision.

this process that provides the information on the $(LiD_2)^+$ potential-energy surface and therefore the corresponding channel is studied in detail.

The relative importance of the electronically elastic channel and the inelastic channels, resulting in excitation of the D_2 target, is presented in Fig. 3. Since the possible excited states of D_2 lie too close in energy for a definitive identification we present the probabilities for electronically elastic scattering (A), and for single-electron excitation of D_2 (B), and two electron and ionizing collision processes (C). The energy loss for peak C is consistent with the excitation of doubly excited D_2^{**} in the Franck-Condon region.[8] The excitation of these states is expected from earlier calculations[3] on the isoelectronic $He+D_2$ system. It is interesting to note that the probabilities for excitation of the singly and doubly excited states of D_2 are comparable. Additional measurements, using time-of-flight techniques on $Li^+ + D_2 \rightarrow Li^0$ show the electron capture to be weak and it is not studied in detail. Excitation of Li^+ is also found to be weak.

Figure 4 shows a plot of ΔE versus $E\theta^2$ at a beam energy of 1.0 keV for the electronically elastic channel (A). The data are seen to lie along the "elastic" curve for $E\theta^2 < 0.7$ keV deg^2 indicating that the most probable energy loss results from purely elastic collisions. The elastic curve is plotted using Eq. (2) with a D_2 target mass. For $E\theta^2 > 0.7$ keV deg^2 the data fall above the elastic limit. The scaled energy loss f is defined by Eq. (1) but an equivalent definition of f is simply the ratio of the measured ΔE to the ΔE ($=2T$) value on the "binary" limit curve [plotted using Eq. (2) for a target having a mass of the D atom]. Data that lie along the limiting curves result in $f=0.5$ and $f=1.0$ values for the elastic and binary limits, respectively. The figure also shows the value of Q which represents the most probable vibrorotational excitation energy. The average f versus τ behavior in the energy range from 600 eV to 3.0 keV is plotted in Fig. 5. It is clearly seen that there is a breakdown in the scaling in $Li^+ + D_2$.

The present results show that, unlike the case in $He+D_2$ at comparable energies, the electronically elastic collision channel dominates the scattering in the angular range studied. The expected energy-loss scaling is seen to break down at small τ but within the experimental errors it is found to be valid for $\tau > 1.4$ keV deg. The breakdown is understood in terms of quantization of the energy loss to the vibrational degrees of freedom and will be discussed in a companion paper by Russek *et al.*[1]

ACKNOWLEDGMENTS

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