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Differential measurements on elastic and rotationally inelastic $\text{Ne}^*(^3\text{P}_{0,2})\text{-H}_2$ collisions at thermal energies

V. Bocvarski (*), J. Robert, I. Colomb de Daunant, J. Reinhardt (**) and J. Baudon

Laboratoire de Physique des Lasers (+), Université Paris-Nord, Avenue J.-B. Clément,
93430 Villetaneuse, France

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Résumé. — On décrit une expérience de faisceaux croisés dans laquelle la sélection en vitesse est obtenue au moyen d'une technique de double hachage. La résolution est suffisante pour séparer les collisions $\text{Ne}^*(2p^53s, ^3\text{P}_{0,2})\text{-H}_2$ accompagnées d'excitation rotationnelle, de celles qui sont purement élastiques. On donne les sections efficaces différentielles mesurées à 67 meV, dans le référentiel du centre de masse, pour les collisions élastiques ($J = 0 \rightarrow J = 0$) et rotationnellement inélastiques ($J = 0 \rightarrow J = 2$).

Abstract. — A velocity selected crossed beam experiment, using a double chopping technique, is described. Its resolution is good enough to discriminate rotationally inelastic $\text{Ne}^*(2p^53s, ^3\text{P}_{0,2})$ on H_2 collisions from purely elastic ones. Elastic ($0 \rightarrow 0$) and inelastic ($0 \rightarrow 2$) differential cross sections in the C.M. frame are given, for a relative energy of 67 meV.

1. Introduction.

Up to now, atom-diatom collisions at low energy have been studied in crossed beam experiments, essentially for ground state species, typically rare gas atoms on H_2 , N_2 , CH_4 , ... [1], and for ground state and excited alkali atoms or molecules [2] ⁽¹⁾. The point here is to investigate, by means of the characteristics of the rotational excitation process, the anisotropy of the atom-molecule interaction. At thermal energies (few tens of meV), the vibrational excitation of a light enough molecule is energetically impossible (and negligible as a closed channel). Therefore, a unique potential surface is sufficient to describe the whole scattering process. In such conditions, semiclassical or quantum methods are applicable. As usual, however, a direct inversion of scattering data into potentials is generally impossible, but a comparison of measured and calculated differential cross sections is a test very sensitive to the potentials in the intermediate distance range (typically 5-10 a_0). In some cases, particular features such as a rotational rainbow are observable; they give quasi-direct information about the potential surface [3]. Unfortunately

(*) Permanent address : Institute of Physics, Beograd, Yugoslavia.

(**) Permanent address : LCAM, Bât. 351, Université Paris-Sud, 91405 Orsay Cedex, France.

(+) Laboratoire associé au C.N.R.S., n° 282.

⁽¹⁾ Many other systems have been studied in cell experiments by means of optical methods (e.g. the rotational relaxation and more generally energy transfers involving the rotational degrees of freedom).

only very few atom-diatom potential surfaces have been calculated up to now for electronically excited states (e.g., $\text{He}^*\text{-H}_2$ [4]).

From the experimental point of view, metastable rare gas atoms used as projectiles provide some advantages in crossed-beam differential measurements : they are easily produced by electronic bombardment (with a rather poor efficiency however : a few 10^{-4}), and they are detected with a high efficiency by secondary electronic emission from a surface. Metastable rare gas atoms (heavier than helium) are also interesting fundamentally because of their internal angular momentum ($^3\text{P}_{0,2}$ states), which is expected to enhance the anisotropy of the interaction, and complicates « moderately » — i.e. just the angular algebra — the collision problem. The main experimental difficulty here is to analyse small energy defects, or gains, corresponding to different initial and final rotational states. From this view-point the ideal molecule is of course H_2 , which has a high rotational constant ($B_e = 7.5 \text{ meV}$). However, its small mass, compared to that of neon for instance, leads to other difficulties : the centre-of-mass angular range $0\text{-}180^\circ$ is contracted, in the laboratory frame, into a limited angular domain, which requires a good angular resolution. As will be seen later, this is not a pure inconvenience since it provides a very simple way to investigate large centre-of-mass scattering angles.

We report here measurements of elastic and rotationally inelastic differential cross sections for $\text{Ne}^*(^3\text{P}_{0,2})$ on H_2 collisions at thermal energy ($E \simeq 67 \text{ meV}$).

2. Experiment (Fig. 1).

Metastable neon atoms are produced within a thermal ground-state atom beam by electronic bombardment. The electronic energy has been optimized to the value of 55 eV . The electron-atom interaction region is 2 mm long only, in order to improve the resolution of further time-of-flight measurements. Electrostatic deflecting plates eliminate charged particles from the atomic beam. Previous experiments [5] have shown that $^3\text{P}_2$ and $^3\text{P}_0$ metastable levels are populated proportionally to their statistical weights ($5 : 1$).

The metastable beam is velocity selected by means of a double-chopping technique. After a path l_1 the beam is chopped periodically by a slotted disk. Synchronous photodiode pulses, conveniently delayed, are used to trig a voltage pulse applied to the electron gun. The pulse period is large enough ($T > 1\,000 \mu\text{s}$) to eliminate any ghost velocity in the pulsed metastable beam emerging from the disk. Changing the delay provides an easy way to select any velocity (v_1) within the (modified) Maxwellian velocity distribution of the metastable beam [6], or, as well, to select UV photons produced by the metastable atom source. The width δv_1 of the selected velocity distribution depends on l_1 (133 mm), on the aperture time of the disk slit ($\simeq 10 \mu\text{s}$ at 400 Hz), on the time width of gun voltage pulses ($\simeq 10$ to $15 \mu\text{s}$), on the electron-neon interaction length ($\delta l_1 = 2 \text{ mm}$) and on the angular aperture of the beam ($\delta\theta_1 \simeq 0.7^\circ$ FWHM) in the collision plane, i.e. perpendicularly to the slits. For $v_1 = 1\,000 \text{ m/s}$, the value of δv_1 (FWHM) is 70 m/s (see Fig. 3). At a distance $L_1 = 154 \text{ mm}$ from the electron gun, the metastable atom beam is crossed at right angle by a supersonic beam [7] of hydrogen. The velocity distribution of this beam has been measured by a time-of-flight method. It is centred at $v_2 = 2\,427 \text{ m/s}$ and its width is less than 220 m/s . Scattered metastable atoms are detected by a multichannel plate detector, located at a distance L_2 from the collision volume. The time of flight spectrum is then obtained using a time-to-amplitude converter and a pulse-height analyser.

The measurement of elastic and inelastic differential cross sections is carried out as follows : an initial value v_1 of the metastable atom velocity is selected and a time-of-flight spectrum $n(t)$ is stored at a given laboratory scattering angle θ_L . Standard time-of-flight spectra, $n_0^{e,i}(t)$ are calculated assuming all differential cross sections, in the C.M. frame, equal to unity (i.e. isotropic, velocity independent and identical for all processes) ; $n_0^{e,i}(t)$ contains all instrumental and geometrical factors involved in the relationship between Lab and C.M. differential cross sections, σ^e for elastic collisions ($J = 0 \rightarrow J = 0$), σ^i for rotational excitations ($J = 0 \rightarrow J = 2$, etc...).

Fig. 1. — Experimental set-up. An effusive Ne beam emerging from a multicapillary array (CA) is bombarded by electrons in gun (G); after a path l_1 , it is chopped by a slotted disk (Di); synchronizing pulses, delayed by t_G , trig the electron gun voltage V_a . The inset shows the corresponding time-distance diagram; $l_1 = 133$ mm, $L_1 = 154$ mm; $L_2 = 163$ mm; the time-width of pulses is about $10 \mu\text{s}$ for the disk slits and for the gun voltage pulses. Collimating slits are indicated by S. DE is the detector (multichannel electron multiplier).

Fig. 2. — Newton diagram of the Ne*-H₂ collision. v_1 , $v_2 (= 2\,427 \text{ m/s})$ are initial velocities respectively of Ne* and H₂; OC is the velocity of the centre of mass. Circles e, i and s correspond to elastic (0-0), inelastic (0-2) and superelastic (2-0) collisions.

Fig. 3. — Time of flight distributions at $\theta_L = 0$: (a) pulsed electron gun, without disk ; (b) continuous gun with disk ; (c) double chopping.

The experimental differential cross sections are obtained in relative values (but with a unique arbitrary unit) by fitting $n(t)$ to a linear combination of the n_0^i spectra :

$$n(t) \propto \sigma^e(\theta_{CM}^e) \cdot n_0^e(t) + \sum_i \sigma^i(\theta_{CM}^i) \cdot n_0^i(t) .$$

Because of the mass ratio ($M(\text{Ne})/M(\text{H}_2) = 10$), two values of the final velocity are generally obtained for each process, at least when θ_L lies within the corresponding angular range, as may be seen in the Newton diagram shown in figure 2. It may be observed also that endothermal-inelastic velocities are inbetween the two elastic ones. For a given process (k), the standard time-of-flight spectrum can be written as :

$$n_0^k(t) = \rho(L_1/v_1) \cdot J \cdot \delta\Omega_c^k / \delta\Omega_L \cdot [1 + d_2/(\varepsilon v_1 + \delta l_1)]^{-1}$$

$\rho(L_1/v_1)$ is the time-of-flight distribution in the incident Ne^* beam at point C ; the corresponding (selected) velocity distribution is taken as a Gaussian one, fitted to the experimental distribution

measured at $\theta_L = 0$; $J = \left| \frac{\partial}{\partial t} \left(\frac{l_1}{v_1} \right) \right|$ is the Jacobian of the transformation $v_1 \rightarrow t = L_1/v_1 + L_2/v''$,

where v'' is the final metastable atom velocity ; $\delta\Omega_L$ is the constant solid angle viewed by the detector and $\delta\Omega_c^k$ is the corresponding solid angle in the C.M. frame ; d_2 is the supersonic beam width in the collision volume.

In fact the distribution of (v_r) of the relative velocity is narrow, even when no selection is performed on v_1 : the non-selected v_1 distribution is :

$$\rho_1(v_1) = \text{const.} \cdot (v_1/u)^4 \cdot \exp(-v_1^2/u^2), \quad \text{where } u = 606 \text{ m/s.}$$

This leads to $f(v_r) = \text{const.} \cdot (v_r/u) [(v_r/u)^2 - a^2]^{7/2} \cdot \exp(-v_r^2/u^2)$, where $a = \frac{v_2}{u} = 4.00$. This distribution is centred at $v_r = 2\,666$ m/s, which corresponds to an impact energy of 67.3 meV, and its spread $\Delta v_r/v_r^{\text{max}}$ is about 11 % (FWHM). This makes the present experiment rather restrictive, in as much as a quasi-constant relative velocity is obtained independently of the selected velocity v_1 . On another hand this is also an advantage since by changing v_1 one scans the C.M. scattering angle, keeping the relative velocity approximately constant.

3. Results.

Figure 4 shows a typical experimental time-of-flight spectrum obtained at $\theta_L = 7^\circ$ for a selected primary velocity $v_1 = 1\,054$ m/s. Calculated spectra for elastic (0-0) and endothermal (0-2) processes are also shown. Actually only 0-0 and 0-2 processes are present, at all lab-scattering angles in the range 5° - 10° , and for initial velocities v_1 ranging from 650 m/s to 1 300 m/s. This indicates that the population of excited rotational states in the H₂ beam are negligible or, more precisely, that their contributions to the scattering in the present range are negligible. Other possible collision processes, such as Penning ionization or electronic excitation transfers, with or without dissociation, are not detectable with the present experimental arrangement.

In figure 5, differential cross sections are displayed as functions of θ_c . As explained before, the collision energy is about 67 meV. The elastic differential cross section fastly decreases with angle. A hump, visible at $\theta_c \approx 70^\circ$, is presumably due to a rainbow effect. The purely elastic scattering is governed by that part of the Ne*-H₂ interaction which is independent of the direction of the H₂ molecular axis. From this viewpoint Ne*-H₂ shows similarity with Ne*-He. For this latter system model-potential calculations have been performed [8]. The energy of state $^3\Sigma^+$ is obtained from that of state $X\ ^2\Sigma_{1/2}^-$ of the ionic core (Ne-He)⁺, by adding a repulsive energy term. As the core potential exhibits a deep well (0.77 eV at $2.46\ a_0$) a hump appears in the resulting potential at about $4.5\ a_0$. A similar feature, probably present in Ne*-H₂ at a slightly larger distance, with a magnitude comparable to the present collision energy, could explain the observed rainbow effect.

Fig. 4. — Time of flight spectrum at $\theta_L = 7^\circ$, $v_1 = 1\,054$ m/s. (●) experiment; calculated standard spectra, weighted to fit experimental data : e, e' elastic contributions; i, i' inelastic (0-2) contributions.

Fig. 5. — Differential elastic (0-0) and inelastic (0-2) cross sections in the C.M. frame, at an impact energy of 67 meV ; a unique symbol is used to indicate values obtained from a single time of flight spectrum (similar to that of Fig. 4).

The rotationally inelastic differential cross section is much smaller than the elastic one at small angle. It becomes almost equal to it around $\theta_c = 90^\circ$. The observed oscillatory behaviour cannot be explained simply. The interpretation needs (i) the knowledge of the potential surface, which has not been calculated yet, (ii) a quantum or semi-quantum treatment of the collision itself, in a body-fixed formulation [9].

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