Pure rotational excitation of H_2 at electron impact energies of 3 to 100 eV†

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Cross sections for pure rotational excitation ($J = 1 \rightarrow 3$) in H_2 are reported at electron energies E_0 of 15 eV to 100 eV at scattering angles θ of 115°, 3 to 40 eV at 20°, and at 40 eV at angles between 10° and 135°. It is found that for intermediate E_0 the probability of pure rotational excitation at high θ is greater than that for elastic scattering. Moreover, this probability is found to decrease slowly with E_0, having a value at its peak (at 4 eV) which is only ~20 times that at 100 eV.

I. INTRODUCTION

The study of elastic and rotational excitation in H_2 is important for modeling several different types of plasmas, electron-excitation laser systems, Earth's ionosphere, and the Jovian atmosphere. As the simplest diatomic molecule, it is the most amenable to theoretical study, and can serve as a testing ground for various theoretical models and approximations. Previous theoretical and experimental electron-scattering studies of rotational excitation[1-4] have been confined to incident energies E_0 of less than 12 eV. Rotational excitation by intermediate-energy electrons were, in general, assumed to be insignificant relative to elastic scattering. It is shown in this paper that is not the case; and at large scattering angles θ (which are heavily weighted in momentum-transfer cross sections), the rotational excitation cross section *exceeds* the elastic-scattering cross section at E_0 greater than about 30 eV. Also, the rotational cross sections are found to decrease slowly with E_0, and have a magnitude at their peak at about 4 eV which is only ~20 times that at 100 eV. These observations may account in part for the large population of excited rotational states observed in interstellar H_2.[5] Rotational excitation temperatures are found to exceed the average ambient kinetic temperature of ~80 °K. While effects of optical quadrupole cascading into low rotational states have been considered,[6] the effects of electron-impact excitation of these same levels have thus far not been taken into account.

II. EXPERIMENTAL PROCEDURES

Two different electron-impact spectrometers of the crossed-beam type were used in the present measurements. These spectrometers, details of the electron optics, the method of data acquisition, etc., have been described earlier.[7,8] Briefly, a monoenergetic electron beam of desired energy E_0 is focused onto the target H_2 beam, and the electrons scattered into a small solid angle ($<10^{-3}$ sr) at an angle θ are energy-analyzed and detected by pulse-counting and multichannel scaling techniques. In the measurements reported here E_0 was varied between 3 and 100 eV, and θ between 10° and 135°. The overall resolution through both electrostatic analyzers ranged from 18 to 30 meV (FWHM).

The following precautions were taken to avoid errors in the measurements.

(i) The energy of the incident electron beam was calibrated using the 19.3-eV resonance in He.

(ii) The true zero scattering angle was determined from the symmetry of the scattering intensity corresponding to the $2\,^1P$ excitation in He.

(iii) The incident electron current was monitored by a Faraday cup. It was found that the current was constant during the time of measurements.

(iv) At low angles of scattering, some of the direct incident electron beam gets into the energy analyzer. This direct beam contribution was obtained by evacuating the vacuum chamber to about 1×10^{-7} Torr pressure and measuring the angular distribution of the electrons. For scattering angles of 10° and higher, the effect of direct beam was found to be negligible.

(v) The transmission of the analyzer optics varies with electron energy. In the spectrometers used for these measurements, the lens potentials were varied in such a way that this transmission was constant. In any case, the errors due to the transmission effects are expected to be negligible since the energy loss associated with rotational excitation is small. Typical energy-loss spectra, so obtained, are shown in Figs. 1 and 2.

III. EVALUATION OF DATA

For pure rotational excitation of a homonuclear diatomic molecule from parity conservation the ΔJ = even selection rule applies, where J is the rotational quantum number. Of all these transitions, it has been shown theoretically,[9] and found

experimentally at low impact energies,[4] that the strongest transitions are those with $\Delta J = 2$. The intensity of a given transition at some E_0 and θ will be proportional to $\sigma_{rot}(J \rightarrow J+2) \cdot N_J$, where $\sigma_{rot}(J \rightarrow J+2)$ is the rotational excitation cross section, and N_J the population of the initial J level. The fractional populations N_J/N (where N is the total number of ortho- and para-H_2 molecules) of the various rotational levels may be calculated in a straightforward way. Using a rotational temperature of 300 °K and a rotational constant E_0 of 7.353 meV, these relative populations are 13% ($J=0$), 66% ($J=1$), 12% ($J=2$), 9% ($J=3$), and all higher levels having populations less than 0.5%. It is expected, therefore, that the transition $J=1 \rightarrow 3$ should be the strongest. This was found to be the case in the present work at all energies and angles.

The quantity of interest in the present measurements is the ratio of intensity of the $J=1 \rightarrow 3$ excitation, $I_{rot}(1 \rightarrow 3)$, to that of elastic scattering, I_{el}. The former is proportional to $N_1 \sigma_{rot}(1 \rightarrow 3)$, while I_{el} is proportional to the sum of elastic cross sections associated with the various J levels times their respective populations, that is, to $\sum_J N_J \sigma_{el}(J \rightarrow J)$. Since the elastic cross section $\sigma_{el}(J \rightarrow J)$ is only weakly dependent on J,[10,11] we may assign the same σ_{el} to all J levels, and the desired ratio of cross sections becomes

$$\frac{\sigma_{rot}(1 \rightarrow 3)}{\sigma_{el}} = \frac{I_{rot}(1 \rightarrow 3)}{I_{el}} \frac{N}{N_1} . \qquad (1)$$

IV. RESULTS AND DISCUSSION

The cross-section ratios obtained from the experimental intensity ratios and Eq. (1) were converted to absolute values of $\sigma_{rot}(1 \rightarrow 3)$ by the use of recently measured values of the absolute cross section,[12] $\sigma_{el} + \sigma_{rot}(1 \rightarrow 3)$. The cross-section ratios and absolute values of $\sigma_{rot}(1 \rightarrow 3)$ are shown in Fig. 3 at 40 eV, at angles between 10° and 135°. The same quantities are shown in Fig. 4 at θ of 20° and 115°, in the range $E_0 = 3$ to 100 eV. The errors

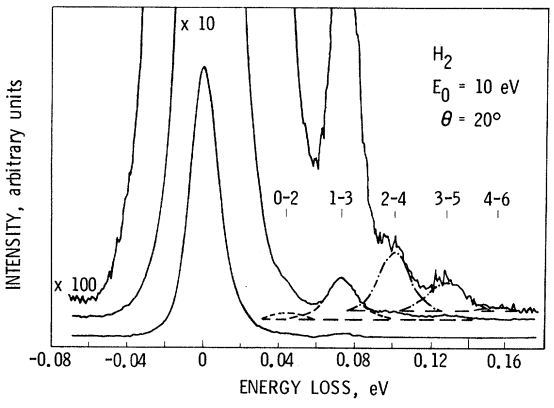

FIG. 1. Energy-loss spectrum of rotational excitation in H_2 at the indicated incident energy and scattering angle. The resolution (FWHM) here is 18 meV.

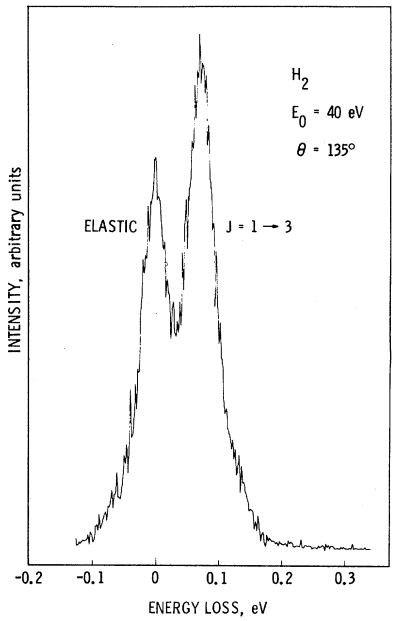

FIG. 2. Energy-loss spectrum at the indicated energy and angle in which the intensity of rotational excitation $J=1 \rightarrow 3$ exceeds the elastic scattering.

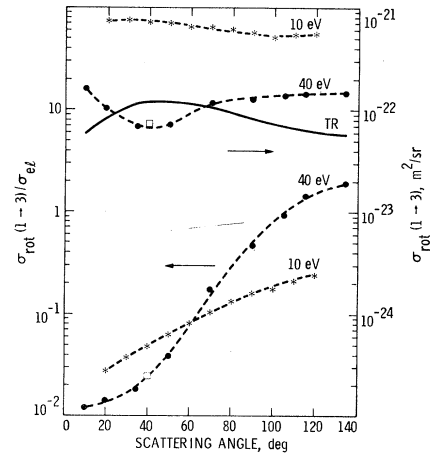

FIG. 3. Lower two curves: the ratios $\sigma_{rot}(1 \rightarrow 3)/\sigma_{el}$ at the indicated energies as read on the left-hand ordinate. Upper two curves: absolute, pure rotational excitation cross sections $\sigma_{rot}(1 \rightarrow 3)$ at the indicated energies as read on the right-hand ordinate. Present measurements are indicated by ● and □ (open and filled symbols refer to different instruments), and those of Ref. 4 by *. The curve TR is obtained from theoretical calculations at 45 eV of Ref. 11.

associated with the ratios in both figures are estimated to be ± 20% at 20° and ± 15% at 115°. The absolute cross sections have an error of ± 25%, which is due to a ± 20% error in the ratio and a ± 15% error in the absolute elastic-plus-rotational cross section.[12]

We have included for the sake of comparison in Fig. 3 the experimental results of Ref. 4 at 10 eV, and unpublished theoretical calculations of Ref. 13 at 45 eV. Several interesting trends emerge in comparing the data at 10 eV and 40 eV. First, while the ratios $\sigma_{rot}(1\rightarrow 3)/\sigma_{el}$ at 10 eV are always less than unity, they *exceed* unity at 40 eV for scattering angles greater than about 105°. Second, the 10 eV angular distribution of $\sigma_{rot}(1\rightarrow 3)$ is nearly isotropic over the entire angular range. At 40 eV, there is a distinct minimum at $\theta \sim 40°$, with a total excursion in $\sigma_{rot}(1\rightarrow 3)$ of a factor of ~2 in the entire angular range.

In Fig. 4, the interesting results in the excitation-function curves are that at low angles (20°) the ratios *decrease* with increasing E_0, while at large angles (115°) the opposite is true, with the ratio becoming larger than unity at $E_0 \geq 30$ eV. Also, one sees in Fig. 4 that the absolute values $\sigma_{rot}(1\rightarrow 3)$ decrease only slowly with E_0 at both high and low angles, and have values at low incident energies which are ~20 times larger than those at 100 eV. The agreement in Fig. 4 of the present measurements with those of Ref. 4 is seen to be quite good. Although rotational excitations corresponding to other than the $J = 1$ to $J = 3$ transition were observed (see Fig. 1), their intensities were much weaker and no reliable quantitative conclusions could be drawn concerning these processes.

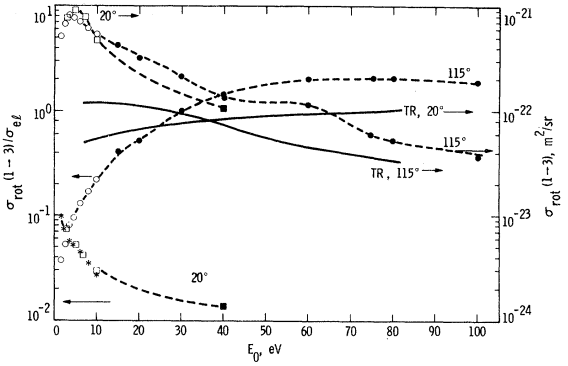

FIG. 4. Curves as read on the left-hand ordinate: the ratio $\sigma_{rot}(1\rightarrow 3)/\sigma_{el}$ at $\theta = 20°$ and 115°; * and ○, Ref. 4; ●, ■, and □, present measurements (open and filled symbols refer to different instruments). Curves as read on the right-hand ordinate: absolute, pure rotational excitation cross sections $\sigma_{rot}(1\rightarrow 3)$. Present measurements are indicated by ● and □ (open and filled symbols refer to different instruments). The curves TR are obtained from theoretical calculations of Ref. 11.

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