$\chi(3413)$, but the fit is of marginal statistical significance. At the 90% confidence level we find that there are no monoenergetic γ rays in the $\psi(3095)$ spectrum between 50 and 1000 MeV with branching fractions larger than 2.0%. Upper limits in other channels are 1.7% for $\chi(3095) \rightarrow \chi(2850)\gamma$,[15] 1.0% for $\psi'(3684) \rightarrow \chi(2850)\gamma$, and 2.5% for $\psi' \rightarrow \chi(3455)\gamma$, all at the 90% confidence level. There is some evidence for this last χ state in the exclusive channel $\psi' \rightarrow \chi(3455)\gamma$, $\chi \rightarrow \psi\gamma$, with a branching-ratio product of $(0.8 \pm 0.4)\%$.[10]

We thank the Stanford Linear Accelerator Center (SLAC), in particular the Experimental Facilities Division, for an extraordinary effort in setting up this experiment in only six weeks. We gratefully acknowledge use of SLAC computing services in the data analysis.

*Work supported by National Science Foundation Grants No. MPS72-04426, No. MPS71-02657, No. MPS 75-19166, and No. GP38611; the U. S. Energy Research and Development Administration; National Aeronautics and Space Administration Grant No. NGR452; and Istituto Nazionale di Fisica Nucleare.

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Isotope Effects in Molecular Scattering by Electrons*

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(Received 17 February 1977)

Recent improvement in electron-beam resolution has made possible the direct study of isotope effects in differential cross sections in H_2 and D_2. The observation of the reduced-mass dependence ($\mu^{-1/2}$) of vibrational ($v = 0 \rightarrow 1$) cross sections and isotopic independence of rotational cross sections at 4 eV has been understood using the frame-transformation theory. Generalization and limitation of the theory are discussed. Specific predictions are made for cross sections in HD, HT, DT, and T_2.

Scientific investigations involving isotopes in molecules have been a subject of current interest in many different fields. Typical examples are interstellar molecules, thermal neutron scattering, laser isotope separation, and the nuclear fusion problem. Of particular importance is the substitution of deuterium or tritium for hydrogen, because the effects are more pronounced as a result of the greater change of the reduced mass. For the nuclear fusion problem one needs explicit cross sections for D_2, DT, and T_2 while most electron impact cross sections[1] have been measured only in H_2.

Conventionally isotopic substitution in molecules

has been used in spectroscopic studies to unravel complex spectra because the isotope shifts in rotational and vibrational energy spacing are well known. In electron-molecule collisions isotope effects in scattering cross sections are not well known and constitute the subject of the present work.

Previous experiments have revealed that the momentum transfer and integrated rotational cross sections in H_2 and D_2 are nearly the same at low energies.[2] We report here a crossed-beam study of the isotope effect in the differential rotational and rotational-vibrational cross sections in H_2 and D_2 at 4 eV where the scattering process is dominated by a broad shape resonance $^2\Sigma_u^+$. The present apparatus is capable of resolving individual rotational transitions in both H_2 and D_2. Thus a *direct* study of the isotope effects in individual cross sections can be made. In this Letter we first present predictions of the isotope effects in various scattering cross sections using the frame-transformation theory.[3] Then we show the experimental data for H_2 and D_2. The good agreement between experimental observations and the theory enables us to predict with confidence the unmeasured results for HD, HT, DT, and T_2 in terms of the known results in H_2 over a broad energy range (1–10 eV). We shall also point out that certain isotope effects such as the dependence on the reduced mass μ of vibrational cross sections and the isotopic independence of rotational cross sections are of general validity. These effects occur in molecules when the scattering processes are characterized by short interaction times,[4] such as broad resonances or potential scattering.

A more precise criterion of "short interaction time" exists in the frame-transformation theory; i.e., the S matrices are entirely determined in the body frame and are independent of the rotational quantum number of the electron-molecule complex. Under these circumstances, the differential scattering cross sections for vibrational excitation ($0 \to v$) have been shown[5] to be

$$d\sigma^v(\Delta j=0)/d\Omega = (4\kappa^2)^{-1}\{|S_{s\sigma}{}^v|^2 P_0 + [S_{s\sigma}{}^v(S_{p\sigma}{}^v + 2S_{p\pi}{}^v)^* + \text{c.c.}]P_1 + |S_{p\sigma}{}^v + 2S_{p\pi}{}^v|^2(\tfrac{1}{3}P_0 + \tfrac{2}{3}P_2) + \bar{f}|S_{p\sigma}{}^v - S_{p\pi}{}^v|^2(\tfrac{2}{3})(P_0 + \tfrac{1}{5}P_2)\}, \quad (1)$$

and

$$d\sigma^v(j \to j+2)/d\Omega = (4\kappa^2)^{-1}(j0, 20|j+2\ 0)^2|S_{p\sigma}{}^v - S_{p\pi}{}^v|^2(\tfrac{2}{3})(P_0 + \tfrac{1}{5}P_2), \quad (2)$$

where κ is the wave vector in units of a_0^{-1}, $(\ldots, \ldots|\ldots)$ denotes the Clebsch-Gordan coefficient, and P_L is the Legendre polynomial.

Isotope effects in scattering cross sections enter via the dependences on $S_{l\Lambda}$ and $\bar{f}$ in Eqs. (1) and (2). The S matrices for various partial waves $(l\Lambda)$ appear in both equations in quadratic form and can be expressed as derivatives of body-frame phase shifts $\delta_{l\Lambda}$ with respect to the internuclear distance R. Specifically for $v=1$, we have

$$S_{l\Lambda} \propto R_S\, d\delta_{l\Lambda}/dR, \quad (3a)$$

where R_S is the scale length. For a harmonic oscillator,

$$R_S = (\hbar/\mu\omega)^{1/2} = [\hbar/\mu'(k/\mu)^{1/2}]^{1/2} = (\hbar^2/\mu k)^{1/4}, \quad (3b)$$

where k, the spring constant, is isotope independent. More generally, previous work[5] shows that (approximately)

$$S_{l\Lambda}{}^v \propto (R_S\, d\delta_{l\Lambda}/dR)^v. \quad (3c)$$

The factor $\bar{f}$ enters only in $\Delta j = 0$ cross sections. It reflects the averaging of the p-wave anisotropic scattering over the rotational states (other terms have unit contribution as a result of this averaging) and is given by

$$\bar{f} = \frac{\sum_j (j0, 20|j0)^2 p_j}{\sum_j p_j}, \quad (4)$$

where the population of the rotational state j is given by

$$p_j = (2j+1)g(j)\exp[-j(j+1)B/KT]. \quad (5)$$

In Eq. (5), K is the Boltzmann constant and T, the temperature, is measured to be 300°K. The rotational constant B and the nuclear spin statistical weight, $g(j)$, are isotope dependent. These and other basic parameters for the various isotopes in molecular hydrogen are listed in Table I.

In summary, the theory predicts that isotopic substitution produces two distinct effects in differential cross sections. One, due to the reduced mass as Eqs. (3) show, is that all differential cross sections are multiplied by the factor $\mu^{-v/2}$, where v is the final vibrational level. The other, due to the factor $\bar{f}$, enters via the isotope effects

TABLE I. Parameters for the isotopes of molecular hydrogen: reduced mass, μ; rotational constant, B; nuclear spin statistical weight, g_j; and population factor, $\bar{f}$.

	μ (nuclear mass)	B (meV)	g_j Even	g_j Odd	$\bar{f}$[a]
H_2	1/2	7.33	1	3	0.326
D_2	1	3.67	2	1	0.252
T_2	3/2	2.44	1	3	0.302
HD	2/3	5.50	1	1	0.270
HT	3/4	4.89	1	1	0.273
DT	6/5	3.05	1	1	0.276

[a] At high temperatures, $\bar{f}$ approaches 0.25 for all isotopes.

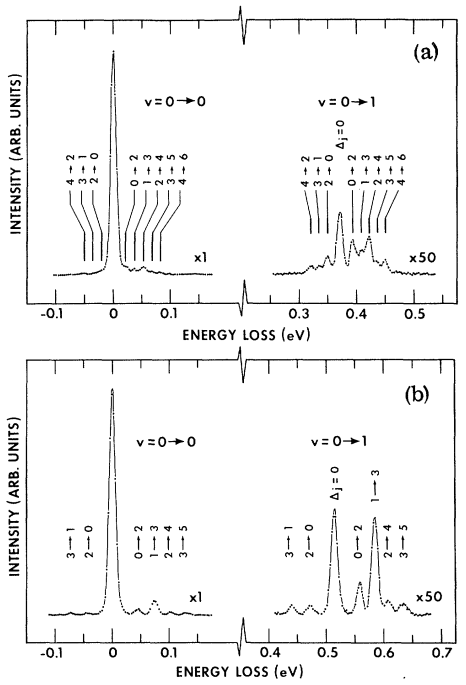

FIG. 1. (a) Intensity of electron scattering in D_2 plotted against energy loss measured at 90° and at a fixed incident energy of 4 eV. Vibrational and rotational transitions are identified. (b) Same in H_2.

in rotational-state populations (different nuclear spin statistical factors and rotational constants) and it affects only the elastic and the pure vibrational cross sections.

The above theoretical predictions are tested against the crossed-beam measurements on differential rotational and vibrational cross sections in H_2 and D_2. Previous work[1] in H_2 has already shown good agreement between experimental results and theory for both the energy dependence (1–10 eV) and the angular dependence (up to $v=3$ level). The present experiment chooses an incident energy of 4 eV (the center of the $^2\Sigma_u^+$ shape resonance) at 90° for testing isotope effects, on the basis of signal-to-noise ratio considerations.

The crossed-beam electron impact spectrometer used in the present study has been previously described.[6] The apparatus consists of an electrostatic hemispherical monochromator, a molecular beam which crosses the incident electron beam, and a hemispherical energy analyzer to examine the scattered electrons. With improved control of the electron optics and the shielding of stray fields in the collision region, we have been able to achieve an energy resolution of 11.5 meV (full width at half-maximum in the energy-loss peaks) for the present isotope studies. The electron-beam current at this resolution is about 1×10^{-10} A. The temperature for both the H_2 and the D_2 molecular beams is estimated to be 300°K.

Figure 1(a) shows the energy-loss spectra in D_2 and Fig. 1(b) the corresponding results in H_2. Results are shown for both $v=0\rightarrow0$ and $v=0\rightarrow1$ vibrational transitions. The separation of the main peaks ($\Delta j=0$) in D_2 is smaller than that in H_2 in accordance with the well-known vibrational spacing dependence on $\mu^{-1/2}$. Profiles of the rotational transition peaks are dictated primarily by the rotational-energy-spacing dependence on μ^{-1} and by the rotational-level populations given by Eq. (5).

We will now explicitly examine the isotope effects on each type of differential cross section at 90°, and discuss the corresponding experimental results. The situation for the rotational inelastic cross sections given by Eq. (2) is simpler and will be discussed first. From earlier discussions, the D_2/H_2 cross-section ratio is expected to show a $\mu^{-v/2}$ dependence for all values of v. Specifically this ratio for the pure rotational transition ($v=0$) is 1.0, and for the vibrational-rotational transition ($v=1$) it is 0.71. These ratios are in good agreement with the experimental observations as shown in Table II.

Consider next the rotational elastic cross sections. At 90° both the second and the third terms in Eq. (1) vanish, and the isotope effects are particularly simple. For the elastic process the first term dominates and contributes about 97% to the cross sections, and so there is no observable isotopic effect as is observed in the experimental result in Table II. On the other hand, the pure vibrational process ($v=1$) is dominated by

TABLE II. Ratio of D_2 to H_2 cross section at 90° and 4 eV.

Cross sections	Theory[a]	Experiment[b] $\Delta j=0$	$j=0$	$j=1$	$j=2$	$j=3$
Rotational ($v=0\rightarrow 0, j\rightarrow j+2$)	1.00		0.96	1.10	0.80	1.15
Vibration-rotational ($v=0\rightarrow 1, j\rightarrow j+2$)	0.71		0.72	0.79	0.72	0.78
Elastic ($v=0\rightarrow 0, \Delta j=0$)	1.00	1.05				
Pure vibrational ($v=0\rightarrow 1, \Delta j=0$)	0.56	0.55				

[a]Estimated accuracy is 1%.

[b]Except for the elastic result, all others are measured as ratios to the elastic cross sections. The estimated errors are 5% for the elastic, and about 10% for the other measurements.

the $^2\Sigma_u^+$ resonance, and the first term becomes negligible. Therefore the fourth term in Eq. (1) shows that the D_2/H_2 cross-section ratio should be $(1/\sqrt{2})\bar{f}(H_2)/\bar{f}(D_2)$, which has a value of 0.56. Again it is in excellent agreement with the experimental value of 0.55 in Table II.

The above comparison shows that the experimental results for all differential cross sections are consistent with the theory in regard to isotope effects. The cross sections for T_2, HD, HT, and DT can be calculated with the aid of Table I and Eqs. (1), (2), and (3).

For practical applications, vibrational cross sections are usually measured without the resolution of rotational states. Under such circumstances it can be shown that the vibrational cross sections (summed over final and averaged over initial rotational states) are given by Eq. (1) with $\bar{f}=1$. Hence the vibrational cross section ($v=0\rightarrow 1$) behaves as $\mu^{-1/2}$, or more generally the cross section ($v=0\rightarrow v$) behaves as $\mu^{-v/2}$ for both the differential and the integrated cross sections. This effect could be of interest for electron impact on other molecules, e.g., hydrocarbon molecules.

Finally we wish to point out that the present theory on isotope effects applies only to potential and broad resonant scattering. The case of narrow resonances requires further study.

This paper is dedicated to the late G. J. Schulz for his never ending enthusiasm in science and his constant encouragement and assistance to young colleagues. We would like to acknowledge A. Herzenberg for many illuminating discussions and J. Kearney for his technical assistance on many aspects of the present work.

*Work supported by the National Science Foundation and the U. S. Army Research Office.

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