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Charles E. Johnson

July 1973

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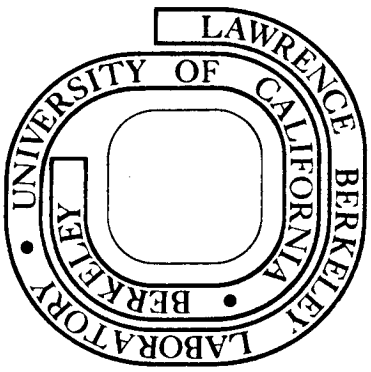

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Quenching of the $c^3\Pi_u$ Metastable State of H_2 and D_2 by an Electric Field*

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July 1973

ABSTRACT

The quenching of the v = 0 $c^3\Pi_u$ metastable state of H_2 and D_2 by a static electric field has been measured using the time-of-flight technique. Neutral ground-state molecules effusing from a source slit are immediately excited to the $c^3\Pi_u$ metastable state by a pulse of antiparallel magnetically focused electrons. After passing through a uniform electric field region 0.5 m long, the velocity distribution of this thermal beam of metastable molecules is then preferentially detected at the end of the time-of-flight region, 1.825 m from the electron gun. The number of $c^3\Pi_u$ molecules arriving at the detector in specific velocity intervals with the electric field off is compared to the number with the field on to determine the quenching rate ($=kE^2$). The result for the quenching constant k is the same for both para- and ortho-H_2, as well as for D_2, and with E in kV/cm is $k = 8.6 \pm 0.3$.

The time-of-flight technique previously employed to measure[1] the quenching of the 2^1S_0 metastable state of helium by a static electric field has been used to investigate the quenching of the $c^3\Pi_u$ metastable state of H_2 and D_2. The $v = 0$ $c^3\Pi_u$ state, approximately 12 eV above the ground state, decays by a combined magnetic-dipole electric-quadrupole transition to the repulsive $^3\Sigma_u^+$ state; the natural lifetime[2] is 1.02 ± 0.05 msec. However, the presence of a sufficiently strong electric field may, through admixtures of higher lying $^3\Sigma_g^+$ states, quench the $c^3\Pi_u$ state and yield a shorter lifetime.

A complete description[1] of the apparatus, data-collection scheme, and time-of-flight data analysis has been reported. The experiment is based on the time-of-flight technique where the metastable molecule is assumed to leave the metastable state only by radiative decay, either natural or electric-field induced, as it drifts through the time-of-flight region. Neutral, ground-state hydrogen (or deuterium) molecules effuse from a source slit and are immediately excited by a 100 eV pulse of magnetically focused electrons. The metastable beam is then collimated while passing through a buffer chamber where a weak electric field of about 100 V/cm quenches H (or D) atoms in the $2^2S_{\frac{1}{2}}$ metastable state, eliminating them from the beam. The $c^3\Pi_u$ metastable molecules next pass through the main electric-field plates, 50.8 cm long, and are finally detected at the end of the time-of-flight region, 182.5 cm from the electron gun. The detector is a solid copper target and

0 0 0 3 8 3 4 7 3 0

intercepts the metastable molecules which survive the flight through the static electric field. An electron multiplier collects the secondary electrons which are ejected from the copper surface by the metastable molecules.

The data-taking and timing aspects of the experiment are controlled by an on-line computer. An example of the data collected with the static electric field both on and off is shown in Fig. 1(a), and represents about 10^6 separate data collection sweeps during a total collection time of 12 h. The electron gun is pulsed on only during channel 0 and counts are then collected into 99 channels, not all of which are shown; all channel widths are equal to 12.75 μsec. After a few hundred data collection cycles with the electric field off (on) , the field is switched on (off) and the same number of cycles repeated. To allow the electric field to stabilize, there is about a 1 sec waiting period before data collection resumes after the voltage is switched.

The number of metastable $c^3\Pi_u$ molecules with initial velocity distribution $n_o(v)$ which are counted by the detector at time t is

$$N(v) = \int_{\text{surface}} \varepsilon \, n_o(v) \, e^{-\gamma t} \, dS$$

where ε is a detector efficiency factor; the exponential factor allows for the possibility of decay with the electric field either on or off. If L is the length of the electric field region while D is the total time-of-flight distance, then the effective decay

rate is $\gamma = \gamma_o + (L/D)\ k\ E^2$ where γ_o is the natural decay rate.

After assuring that n_o is uniform across the beam, the time and velocity dependence can be extracted from the integral. Then, taking a ratio of electric-field-on counts to electric-field-off counts for molecules of the same velocity, we obtain

$$R(t) = \frac{N_{on}(v)}{N_{off}(v)} = e^{-(L/D)kE^2t}$$

where t is the time of flight. This ratio is not only independent of the initial velocity distribution $n_o(v)$ but also of the detector efficiency factor ε. The natural logarithm of the ratio R is $\ln (R) = -(L/D)\ k\ E^2t$. The calculation of ln (R) for several different velocity intervals of the time-of-flight distribution, combined with a measurement of the two distances L and D, and the electric field E, allows a determination of the quenching constant k from the slope [$= -(L/D)\ k\ E^2$] of a straight line, least-squares fitted to a plot of ln (R) versus the time of flight t. A sample plot is shown in Fig. 1(b).

The result for the quenching constant of the $c^3\Pi_u$ metastable state of H_2 in a static electric field is $k = 8.6 \pm 0.3$; electric fields equal to both 14.14 kV/cm and 20.00 kV/cm were used. The error is mostly statistical, and to within this error, the quenching constant is the same for both para- and ortho-H_2, as well as for D_2. These experimental results disagree with the theoretical estimates of Freis and Hiskes[3], who obtained different quenching rates for

para- and ortho-H_2. However, in their calculation only mixing with the $2s\sigma\ ^3\Sigma_g$ state was considered. Actually, the electric field mixes a large number of other excited states with the $c^3\Pi_u$ state as well, and they may contribute significantly to its quenching. For example, when compared to the 2^1P state, the higher lying (n>2) ^1P states of He contribute almost 25% to the quenching constant[1,4] of the 2^1S_0 state.

References

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Figure Caption

Fig. 1. (a) Time-of-flight distributions. The upper $e\,^3\Pi_u$ distribution is with the electric field off, while the lower is with the field on; the decrease in height represents quenching by the electric field. (b) The ratio of the two distributions vs time-of flight is a straight line on a logarithmic plot. The quenching constant k is obtained from the slope of the least-squares fitted straight line; the fit only includes points which have counts equal to at least 10% of the number in the peak of the distribution.

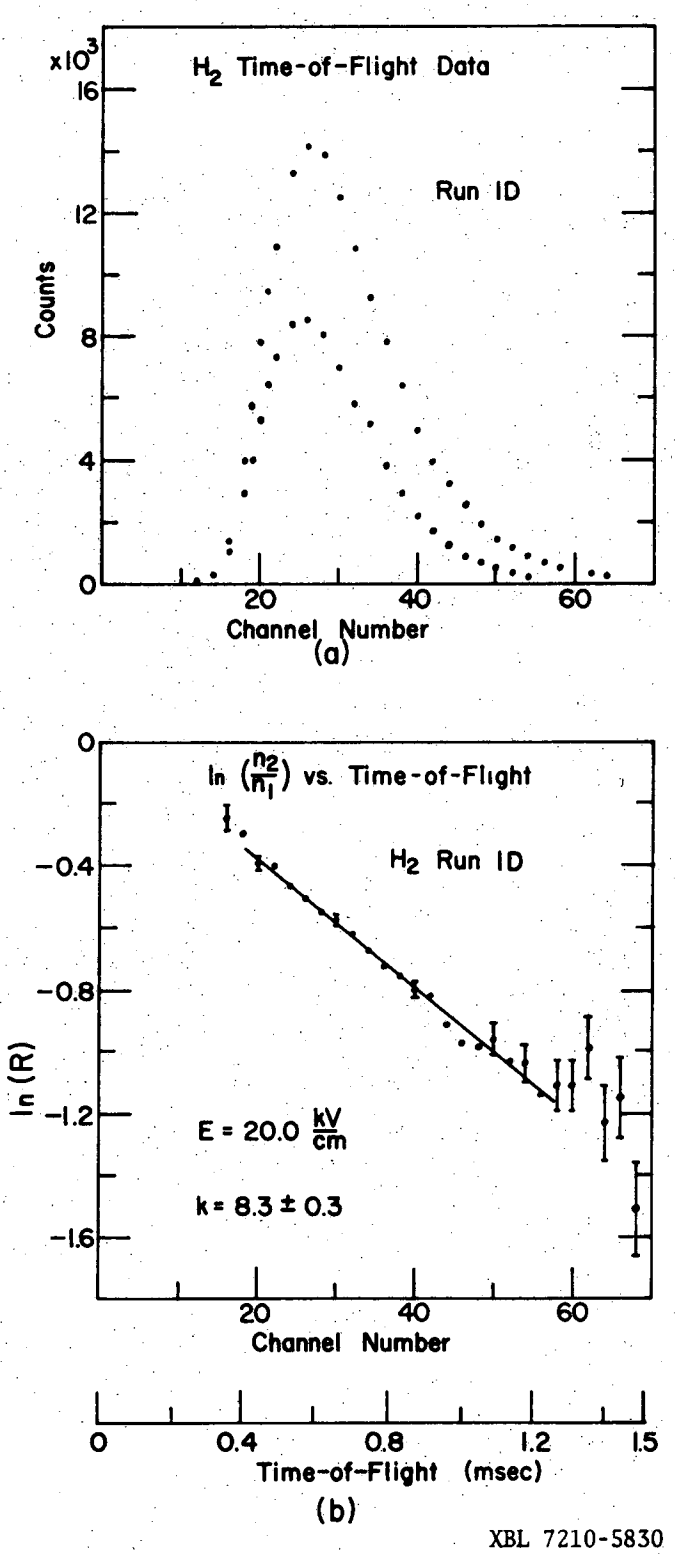
x10^3
H2 Time-of-Flight Data
Run ID
Counts
16
12
8
4
0
20
40
60
Channel Number
(a)
ln (n2/n1) vs. Time-of-Flight
H2 Run ID
E = 20.0 kV/cm
k = 8.3 ± 0.3
ln (R)
0
-0.4
-0.8
-1.2
-1.6
20
40
60
Channel Number
0
0.4
0.8
1.2
1.5
Time-of-Flight (msec)
(b)
XBL 7210-5830

Fig. 1

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