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$^2S_{1/2} \rightarrow {}^2D_{5/2}$ Double-Quantum Transition in H, $n = 3$*

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We have used rf techniques and a fast hydrogen beam to observe in zero magnetic field the two-quantum transition $^2S_{1/2} \rightarrow {}^2D_{5/2}$ in the $n = 3$ state of atomic hydrogen. The possibility of using this and related transitions for a high-precision measurement of the fine-structure constant is discussed.

Measurements of the hydrogen fine structure give the most theoretically unambiguous determination of the fine-structure constant α.[1] In conventional fine-structure measurements on hydrogenic atoms, the lifetimes of the states determine the linewidth of the transitions and limit the precision of the measurements.[2] Multiphoton transitions which cross a particular n-level manifold present the possibility of observing lines with a significantly higher ratio Q of the transition frequency to the natural linewidth and thus improving the precision of the measurements.[3] Figure 1 shows the energy levels in the $n = 3$ manifold of atomic hydrogen. The value of Q is 400 for the $3^2S_{1/2} \rightarrow 3^2D_{5/2}$ double-quantum transition. This is 4 times larger than for conventional measurements (e.g., $3^2S_{1/2} \rightarrow 3^2P_{3/2}$).[4]

In the presence of an rf electric field the perturbing Hamiltonian is given by

$$\mathcal{H}' = eE_0 z \cos\omega t.$$

There is no direct coupling between the $3^2S_{1/2}$ and $3^2D_{5/2}$ states. There is, however, a second-order coupling through virtual transitions to the $3^2P_{3/2}$ state, which becomes resonant when $2\hbar\nu = E(^2D_{5/2}) - E(^2S_{1/2})$. In the neighborhood of this resonance, the coupling between S and D can be approximated by the effective potential

$$V_{DS}^{\text{eff}} = \tfrac{1}{2}(eE_0)^2 \cos 2\omega t \sum_P \frac{\langle D|z|P\rangle\langle P|z|S\rangle}{(E_S - E_P + \hbar\omega)},$$

where the sum is taken over the intermediate P states.[5,6] The lifetimes of the various states are taken into account via the usual Bethe-Lamb prescription $E \rightarrow E + i\Gamma/2$. The transition probabilities and line shapes can be calculated just as for conventional electric-dipole rf resonance.

A schematic diagram of the apparatus is shown in Fig. 2.[7] It is basically a device to produce a

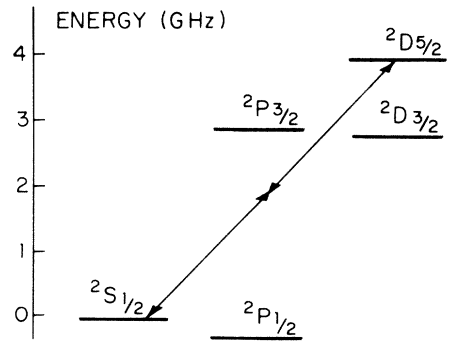

FIG. 1. Level diagram of hydrogen, $n = 3$, showing the two-photon transition. The states are coupled through virtual transitions to the $^2P_{3/2}$ state.

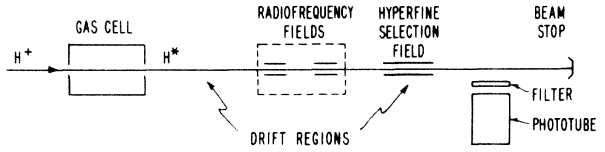

FIG. 2. Schematic diagram of the apparatus.

beam of fast hydrogen atoms with a substantial population in the $3^2S_{1/2}$ state, and to monitor the population of that state as a means of detecting resonance depopulation by rf electric fields in the intermediate region. The excited hydrogen atoms are produced by neutralization of a fast (100 keV) proton beam in a charge-exchange cell. Because of its long lifetime (160 nsec), the $3^2S_{1/2}$ dominates the $n=3$ population remaining after a short drift region. The detector is a photomultiplier–interference-filter combination which monitors the Balmer-α photons emitted in the natural decay of the $3^2S_{1/2}$ state.

In the intermediate region the atoms pass through an rf interaction region which consists of two identical parallel-plate transmission lines which are excited in phase by rf fields. When the amplitudes of these rf electric fields are held constant at approximately 20 V/cm and the frequency of the fields is varied in the neighborhood of 2000 MHz, the S-state depopulation signal shown in Fig. 3(a) is observed. The complexity of this curve is due to the superposition of several hyperfine components and can be considerably reduced by the addition of a continuous hyperfine-state–selection rf field prior to the phototube in the second drift region shown in Fig. 2. If the frequency of this field is chosen to preferentially quench the $3^2S_{1/2}, F=1$ state by mixing it with the very short-lived (5.4 nsec) $3^2P_{1/2}$ state, the overall detection efficiency for the $F=1$ state is reduced and the Balmer-α photocurrent is due primarily to the $3^2S_{1/2}, F=0$ state. The resultant signal which is shown in Fig. 3(b) is thus mainly due to the $3^2S_{1/2}, F=0 \rightarrow 3^2D_{5/2}, F=2$ transition. The signal is a superposition of a Rabi line shape, a Ramsey separated oscillatory field pattern, and a flat background due to nonresonant quenching.

Two features of this transition unique to its two-quantum character were observed. First, the transition amplitude was found to be proportional to the square of the rf power. Second, the inversion of the interference term in the separated oscillatory field pattern [the "wiggles" of Fig. 3(b)] was observed to occur when the relative

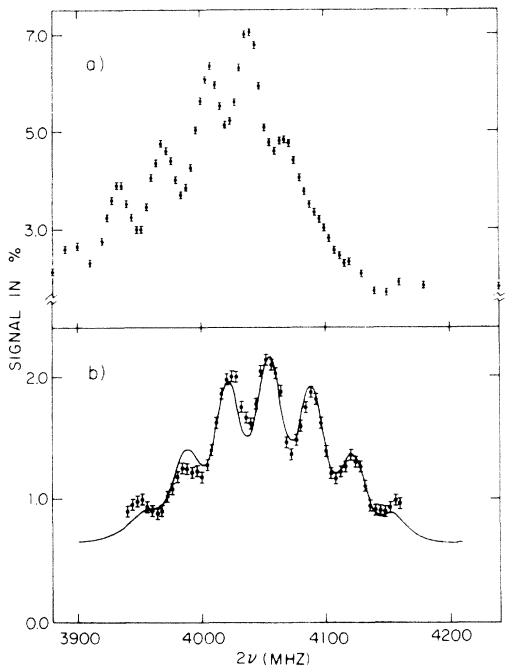

FIG. 3. Line profiles of $^2S_{1/2} \rightarrow {}^2D_{5/2}$ double-photon resonance (a) without hyperfine selection, (b) with selection of the $F=0, {}^2S_{1/2}$ state. The solid curve in (b) is a computer fit using two-level theory.

phase of the two interaction fields was 90° rather than the usual 180°.

The smooth curve shown in Fig. 3(b) is a fit by a simple two-level line shape. The free parameters were the center frequency, the amplitudes of the Rabi and Ramsey line shapes, a flat background, and the residual contribution of the competing hyperfine components. The fit indicates that the hyperfine-state selection is not complete; this produces the residual asymmetry. After correction for small (~1 MHz) rf Stark shifts the center frequency is in good agreement with the known $^2S_{1/2} \rightarrow {}^2D_{5/2}$ splitting. The 16-MHz width (in 2ν) of the central peak is due to the time spent in between the two rf fields and is somewhat greater than the 10-MHz natural linewidth. The agreement between the experimental and theoretical line profiles is good.

At present we do not have rf equipment with which to shift reliably the relative rf phase between 0 and 90° and thereby use the separated-oscillatory-field technique to narrow the line. Judging from our experience with the $^2S_{1/2} \rightarrow {}^2P_{1/2}$ Lamb shift in the $n=2$ state of hydrogen, we could, conservatively, reduce the linewidth to $\frac{1}{3}$ the natural linewidth and thus obtain a line Q of

1200.[8] A measurement to 1 part in 10^3 would then give a value of α with a precision of 4.2 in 10^7. This is significantly more precise than the present 1.6 in 10^6 determination of α from measurements using the ac Josephson effect.

The determination of the fine structure of the He^+ ion gives an even more fertile field for the multiple-quantum–transition technique. In helium there is no hyperfine structure and one can envision using a multiple-quantum transition in the $n=6$ state with a Q of 10^4 to 10^5 for determination of the fine-structure constant.

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[1]Indirect measurements of the fine-structure constant such as the hydrogen hyperfine structure, the muonium hyperfine structure, the helium fine structure, the electron $g-2$, and the ac Josephson effect all have possible ambiguities in their interpretation.

[2]For a review of conventional measurements see B. N. Taylor, W. H. Parker, and D. N. Langenberg, Rev. Mod. Phys. 41, 375 (1969).

[3]Measurements of related high-Q fine-structure transitions through level-crossing techniques have been reported by T. G. Eck and R. J. Huff [Phys. Rev. Lett. 22, 319 (1969)] and by H. J. Beyer *et al.* [Phys. Rev. Lett. 28, 263 (1972), and J. Phys. B: Proc. Phys. Soc., London 4, L129 (1971)]; M. Glass-Maujean, Opt. Commun. 8, 260 (1973).

[4]For a recent discussion of the possibility of high-resolution double-quantum laser spectroscopy in hydrogen see D. E. Roberts and E. N. Fortson, Phys. Rev. Lett. 31, 1539 (1973).

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[6]M. Silverman, thesis, Harvard University, 1973 (unpublished).

[7]The apparatus and technique are similar to that described in C. W. Fabjan and F. M. Pipkin, Phys. Rev. A 6, 556 (1972).

[8]S. R. Lundeen *et al.*, Bull. Amer. Phys. Soc. 18, 1500 (1973).

Observation of Momentum Transfer in Rotationally Inelastic Molecular Collisions of CO_2 with H_2, He, CO_2, and Kr†

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We present an experimental technique enabling a direct observation of the momentum transfer distribution generated by rotationally inelastic molecular collisions of CO_2 with H_2, He, CO_2, and Kr. The observations indicate that the rotational transition results primarily from peripheral collisions which are effective in transferring angular momentum, but which communicate a relatively small linear momentum transfer. The *mean* change in the z component of velocity is presented for CO_2-H_2 collisions.

We report initial experimental observations of the momentum-transfer distribution generated by rotationally inelastic molecular collisions of CO_2 with H_2, He, CO_2, and Kr. The general process under examination is

$$CO_2(j,\vec{v})+M \rightleftharpoons CO_2(j',\vec{v}')+M, \quad (1)$$

where the pair $(j,\vec{v})$ labels, respectively, the rotational quantum number and velocity of the CO_2 molecule, and M represents the appropriate collision partner. Previous studies examining the velocity dependence of collisions of atomic and molecular systems have involved molecular-beam experiments,[1] coherent spectroscopy,[2] treatment of collisional effects on spectral line profiles,[3,4] determination of infrared pressure-broadening coefficients,[5] the role of velocity cross relaxation on the Lamb dip,[6] and two-photon, double-resonance studies of collisionally redistributed populations.[7] In the technique de-