and approximately 0.5 watt. Figure 1(a), with the microwave absent, indicates clearly the point of discharge termination with time, there being more momentary emission from the afterglow than from the main discharge. Figure 1(b), with the microwave present, shows the effect of quenching at a point on the anode side of the most intense negative glow, while in the wake of the microwave a greater emission is noted ("saving-up" effect[2]). Figure 1(c) shows the effect of enhanced emission in the presence of the microwave, observed at a point on the cathode edge of the negative glow.

A broad flat minimum in the magnitude of the quenched radiation with increase of microwave energy incident presumably indicates an almost complete halting of observable recombination before further excitation by the microwave is appreciable. Thus the residual light is probably due to a normal discharge excitation process, perhaps a result of the "intermediate" electron group. The light that can be quenched and attributed to recombination is experimentally found, with increasing incident microwave energy, to approach the $-\frac{3}{2}$ power law[4] intrinsic in the recombination coefficient. Temperature of the "ultimate" group of electrons in this discharge type has been found by independent Langmuir probe measurements to be approximately 1000°K, which is conducive to strong recombination. A measurement based on the saving-up effect and a knowledge of absolute electron density allows calculation of the recombination coefficient, α—preliminarily $\sim 10^{-8}$ cm^3/ion sec. These indications of recombination in the helium negative glow are consistent with the hypothesis of dissociative recombination[4] and give support to the early measurements of Dewey.[5]

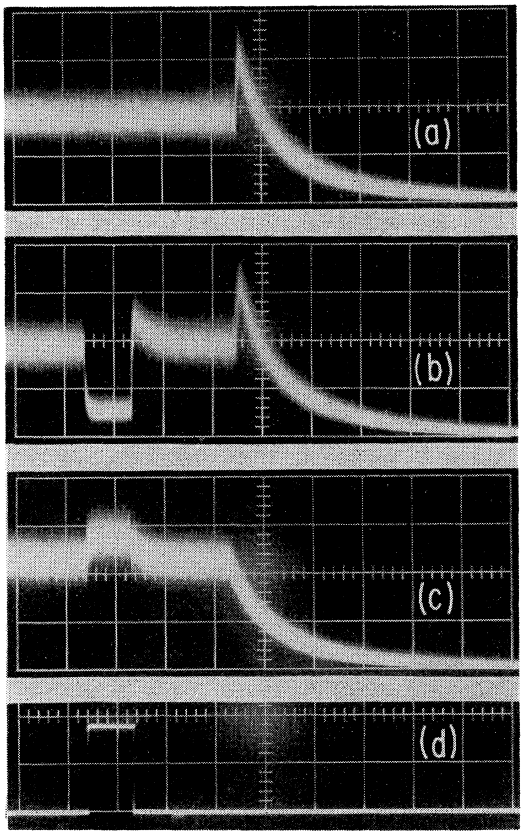

FIG. 1. Quenching and enhanced light emission by a microwave pulse from the negative glow of a cold-cathode dc discharge in helium. (See text for details.)

Similar discharges in neon, argon, and xenon in the pressure range 1–10 mm Hg gave no quenching effect with application of the microwave field, only enhanced emission. Neon with small admixtures of argon gave a small magnitude of quenching at a point close to the anode side of the most intense negative glow, but at no other point in the negative glow or Faraday dark space. Preliminary spectral separation in helium by filters showed the microwave-induced quenching to be an equal percentage of the total quenchable light throughout the visible, but the light attributed to discharge or microwave excitation to be predominantly from the red portion of spectrum.[6]

The afterglows, upon interruption of all the above indicated discharges, gave an almost complete quenching effect, but the maximum incident microwave energy gave enhanced emission (less quench), as for the case of the continuous discharge.

I am indebted to Dr. John A. Bradshaw for many helpful discussions.

[1] Goldstein, Anderson, and Clark, Phys. Rev. **90**, 487 (1953).
[2] C. Kenty, Phys. Rev. **32**, 624 (1928).
[3] R. G. Fowler, *Handbuch der Physik* (Springer-Verlag, Berlin, 1956), Vol. 22, pp. 227–229, 245, 247.
[4] L. B. Loeb, *Handbuch der Physik* (Springer-Verlag, Berlin, 1956), Vol. 21, p. 493.
[5] Jane M. Dewey, Phys. Rev. **32**, 918 (1928).
[6] M. Druyvesteyn, Z. Physik **57**, 292 (1929).

High-Temperature Molecular Beam Microwave Spectrometer*

ALLEN K. GARRISON† AND WALTER GORDY
Department of Physics, Duke University, Durham, North Carolina
(Received August 26, 1957)

MOLECULAR beam spectrometers have the advantage that both Doppler and pressure broadening can be largely avoided. They are especially attractive for high-temperature measurements where these broadening factors become very significant and where the difficulties of heating an entire microwave absorption cell with its sealed windows become increasingly difficult. The sensitive and intricate molecular-beam electric resonance method[1] cannot generally be applied in the upper frequency microwave region. Furthermore, the electric resonance method as usually applied gives the product of the dipole moment, μ, and spectral constant, B, although in some instances[2] (for

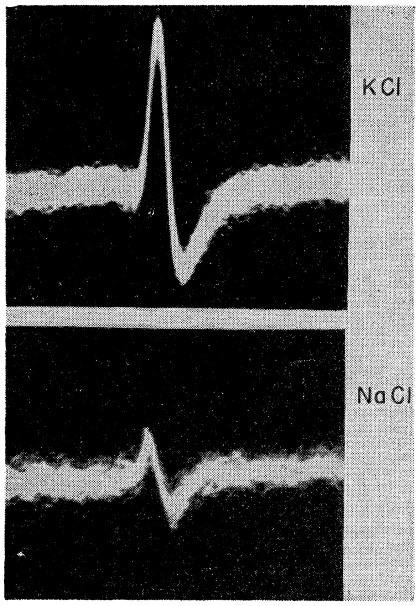

FIG. 1. Upper curve is the $J=12\rightarrow13$ rotational transition of KCl^{35} at 3.00-mm wavelength (99 931.418 Mc/sec). Lower curve is the $J=6\rightarrow7$ rotational transition of $NaCl^{35}$ at 3.30-mm wavelength (91 172.36 Mc/sec). Both curves were obtained with the molecular beam cell shown in Fig. 2.

KCl, KBr, and RbCl) it has been possible to measure the separation of the lower rotational levels and thus obtain B unambiguously.

Simple calculations indicate that one should be able to detect directly microwave power absorbed through rotational transitions in high-temperature molecular beams. Several years ago unsuccessful attempts were made by Luck and one of us (W. G.) to do this.[3] In view of Fig. 1 obtained with a crystal video receiver from molecules simply sprayed across the radiation path, as indicated by Fig. 2, it is now apparent that the earlier experiment could have been successful. Nevertheless, no one else has to our knowledge in the intervening years succeeded in detecting the power absorption by molecular beams at elevated temperatures, although the M.I.T. group[4] has observed absorption by NH_3 beams at ordinary temperatures and the Columbia[5] group has detected stimulated emission from NH_3 beams.

To favor our chances of success we took advantage of the increasing absorption of molecules with increasing frequency. Our experiments were made in the 3-millimeter wave region with the fourth harmonic energy from a K-band klystron with the harmonic generator previously described.[6] From the strength of the signals obtained here it is evident that detection can also be

TABLE I. Observed frequencies (in Mc/sec) of the $J=12\rightarrow13$ transition of KCl^{35}.

	$v=0$	$v=1$
KCl^{35}	99 931.418±0.016	99 318.168±0.016

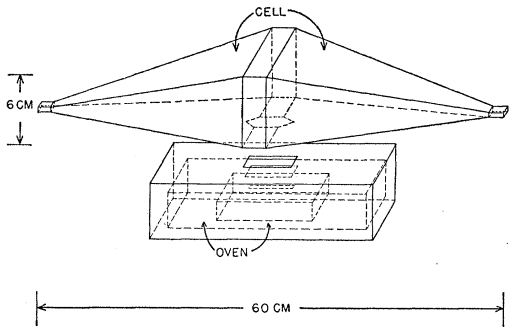

FIG. 2. Sketch of high-temperature molecular beam cell for the shorter millimeter wave region.

made in the centimeter wave region. Much of the more important work left to be done at high temperatures is in the millimeter or submillimeter regions where the rotational spectra of such light diatomic molecules, LiF, LiD, NaH, KH, can be reached and where the centrifugal stretching effects of the heavier molecules can be accurately measured. However, the molecules already investigated with ordinary hot cells in the centimeter region can now be remeasured to advantage with the high resolving power provided by the molecular beam spectrometer. For example, the error limits quoted, ±0.20 to ±0.75 Mc/sec, for the centimeter wave lines measured with the Columbia[7] hot cell are an order of magnitude greater than those of the 3-millimeter wave KCl transitions in Table I. The width of the KCl lines between half-power points measured here for the $J=12\rightarrow13$, $v=0$, line of KCl at 100 kMc/sec is only 60 kc/sec. This width, which would correspond to 15 kc/sec at the fundamental klystron frequency, is the order of 100 times less than that obtainable with the usual hot cell. This width can be reduced further by better collimation of the molecular beam, by use of a high-fidelity amplifier, and by stabilization of the klystron frequency. The nuclear quadrupole broadening of the principal KCl line at this frequency is only about 10 kc/sec. More details will be given in a later publication.

We are greatly indebted to C. F. Luck from whose earlier attempts to detect microwave absorption from molecular beams much was learned about how to make the present experiments work.

* This research was supported by the U. S. Air Force through the Air Force Office of Scientific Research of the Air Research and Development Command.

† Danforth Foundation Fellow.

[1] H. K. Hughes, Phys. Rev. **72**, 614 (1947); J. W. Trischka, Phys. Rev. **74**, 718 (1948).

[2] Lee, Fabricand, Carlson, and Rabi, Phys. Rev. **91**, 1395, 1403 (1953). J. W. Trischka and R. Braunstein, Phys. Rev. **96**, 968 (1954).

[3] C. F. Luck and W. Gordy, unpublished work described in a review by J. M. Mays, Ann. N. Y. Acad. Sci. **55**, 789 (1952).

[4] H. R. Johnson and M. W. P. Strandberg, Phys. Rev. **86**, 811 (1952).

[5] Gordon, Zeiger, and Townes, Phys. Rev. **95**, 282 (1954).

[6] W. C. King and W. Gordy, Phys. Rev. **93**, 407 (1954).

[7] Honig, Mandel, Stitch, and Townes, Phys. Rev. **96**, 629 (1954).

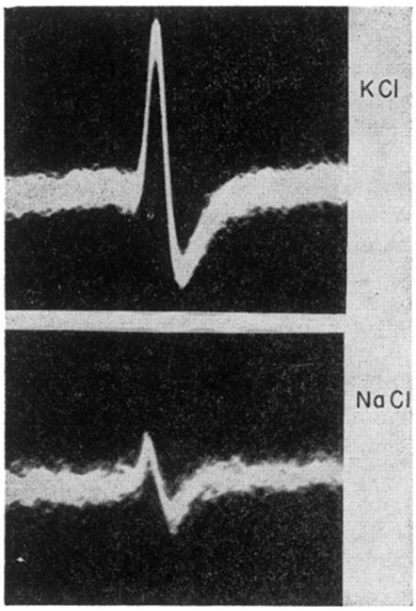

FIG. 1. Upper curve is the $J=12\rightarrow13$ rotational transition of KCl^{35} at 3.00-mm wavelength (99 931.418 Mc/sec). Lower curve is the $J=6\rightarrow7$ rotational transition of $NaCl^{35}$ at 3.30-mm wavelength (91 172.36 Mc/sec). Both curves were obtained with the molecular beam cell shown in Fig. 2.