Collision Cross Section of Slow Electrons and Ions with Cesium Atoms*

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Microwave interferometry is adopted to measure the effective-collision cross section for momentum transfer $\bar{Q}_m$ of thermal electrons (of mean energies of ~0.06 to 0.071 eV) with cesium atoms in the afterglow of pure cesium and helium-cesium discharges. The momentum-transfer cross section $\bar{Q}_m$ is found to be best represented by $1.61\times10^{-10}\,T_e^{-1}-9.63\times10^{-12}\,T_e^{-1/2}+2.03\times10^{-13}$ cm^2 in the temperature range of approximately 450 to 550°K. The energy dependence of the elastic electron-cesium-atom collision probability for momentum transfer is determined to be $P_m=997\,u^{-1}-4810\,u^{-1/2}+7230$ cm^{-1}, where u is the energy of the electron volts. This shows a smooth tendency to join Brode's data at higher electron energies. Mobilities of Cs^+ ions in helium and in cesium have been determined from the helium-cesium mixture experiments in the same temperature range. They are $\mu(Cs^+ \text{ in He})=18.5\pm0.5$ cm^2/V-sec and $\mu(Cs^+ \text{ in Cs})=0.4\pm0.05$ cm^2/V-sec referred to standard gas density (i.e., 2.69×10^{19}/cm^3).

INTRODUCTION

A RATHER thorough study (such as mobility of electrons, recombination processes, spectral intensity distribution, etc.) of the positive column of cesium discharges has been made by Boeckner and Mohler[1] and by Mohler.[2] Nevertheless, the knowledge of the elastic electron-atom collision cross section at low energies (below 0.3 eV)[3] and ion mobilities at low E/p have been lacking until recently.[4] The present experiment extends the electron-atom collision cross-section measurements to the thermal energy range (450 to 544°K) and the cesium ionic mobility to $E/p\sim0.1$ V/cm-mm Hg or less.

EXPERIMENTAL APPARATUS

Microwave techniques[5] are used to measure the electron density and electron collision frequency (for momentum transfer) in the afterglow of a pulsed dc discharge established in pure cesium and in cesium-helium mixtures. The duration of the breakdown voltage pulse is of the order of 7 μsec, the repetition frequency of the pulse is ~31 cps. The discharge tube is made of thin wall (0.7 mm) Pyrex tubing (22 mm o.d., and 72 cm long) with 6 cm tapering to a point at each end. The tube is housed coaxially in a 1-in. ×1-in. square waveguide, which is connected to the standard X-band waveguide through two 6-in. tapering sections. Tantalum electrodes are used for the discharge tube. A schematic diagram of the microwave circuitry used is shown in Fig. 1. A 5-μsec, low-power (~7-μW), 8647-Mc/sec probing signal is launched at times in the afterglow and measurements of phase shifts and attenuations are made.

The cesium used is obtained from the Bram Metallurgical Chemical Company claimed to be 99.9% pure, the impurities being mainly sodium and potassium. Small capsules of cesium are made for the present experiments through vacuum distillation in a standard ultra-high vacuum system.[6] A capsule is then installed on the system in a side tube connected to the discharge tube. The system is then baked at 400°C for more than 24 h. An ultimate vacuum of the order of 4×10^{-10} mm Hg is attained. The cesium capsule is opened by a breaker (a short, soft, steel bar sealed in an evacuated glass tubing) and the discharge tube is then sealed off. The temperature of the discharge tube is then brought

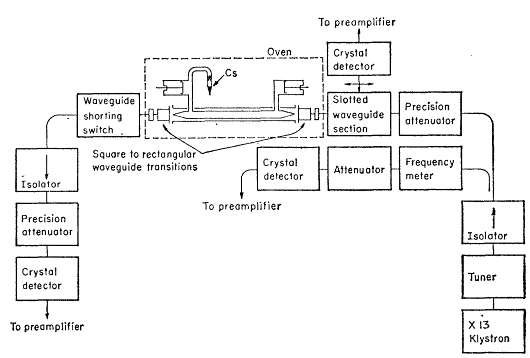

Fig. 1. Schematic diagram of the microwave circuitry.

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[1] C. Boeckner and F. L. Mohler, Bur. Standards J. Research **10**, 357 (1933).

[2] F. L. Mohler, Bur. Standards J. Research **9**, 493 (1932); **10**, 771 (1933); **16**, 227 (1936); **17**, 45, 849 (1936).

[3] R. B. Brode, Revs. Modern Phys. **5**, 257 (1933).

[4] R. M. Kushnir, B. M. Palyukh, and L. A. Sena, Bull. Acad. Sci. U.S.S.R., Phys. Ser. **23**, 995 (1959). R. K. Flavin and R. G. Meyerane, Jr., *Symposium on Thermionic Power Conversion at Colorado Springs, May 14–17, 1962* [Pergamon Press, Inc., New York (to be published)]; D. Roehling, *ibid.*

[5] M. A. Biondi and S. C. Brown, Phys. Rev. **75**, 1700 (1949); L. Goldstein, M. A. Lampert, and R. H. Geiger, Elec. Commun. **243**, (1952); J. M. Anderson and L. Goldstein, Phys. Rev. **100**, 1037 (1955); S. Takeda and E. H. Holt, Rev. Sci. Instr. **30**, 722 (1959); C. L. Chen, C. C. Leiby, and L. Goldstein, Air Force Cambridge Research Center Scientific Report No. **7**, AFCRC-TN-59-597, 1959 (unpublished).

[6] D. Alpert, J. Appl. Phys. **24**, 860 (1953).

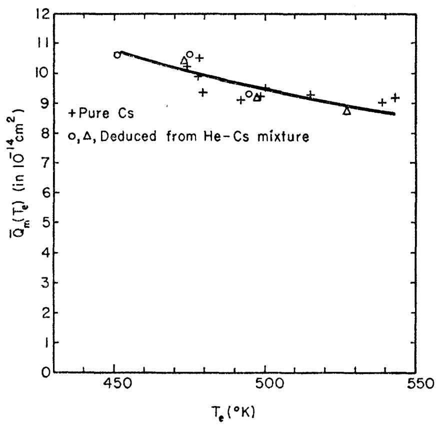

FIG. 2. Momentum-transfer cross section of electrons with cesium atoms. The circular and triangular points are deduced from helium-cesium mixture experiments with helium partial pressures of 1.82 and 3.5 mm Hg, respectively, referred to 0°C. The solid curve is calculated from $Q_m(v)=98.7\ v^{-2}-8.04\times10^{-6}\ v^{-1}+2.03\times10^{-13}$ cm^2, where v is the electron velocity in cm per sec.

up to the desired value by the oven control. The temperature of the cesium capsule and of the waveguide are monitored constantly by five copper-constantan thermocouples. The vapor pressure p (in mm Hg) of Cs is calculated from[7]

$$\log_{10}p=-4075/T+11.38-1.45\log_{10}T,$$

where T is the temperature of the cesium reservoir in degrees Kelvin. Two ~1.5-mil mica sheets are placed on both sides of the discharge tube in the waveguide to cut down any convection and guarantee uniformity of temperature in the discharge tube. By this means, the temperature variation of the discharge tube is kept within 1°C.

MEASUREMENTS IN PURE CESIUM

It can be shown[8] that the complex electrical conductivity σ_c of a plasma is given by

$$\sigma_c=-\frac{ne^2}{3m}\int\frac{v\partial f_0/\partial v}{\nu+j\omega}d^3v, \qquad (1)$$

where n is the electron density, e is the electron charge, m is the electron mass, and v is the electron velocity. f_0 is the zeroth order of a spherical harmonic expansion of the electron-velocity distribution function and is assumed Maxwellian. $\nu=NQ_m(v)v$ is the momentum-transfer collision frequency of the electrons with the neutrals in the plasma. N is the neutral gas density and $Q_m(v)$ is the momentum-transfer collision cross section defined as

$$Q_m(v)=\int(1-\cos\theta)I(\theta,v)d\Omega, \qquad (2)$$

where θ is the scattering angle of the electrons, and $I(\theta,v)$ is the differential-scattering cross section.

For $\omega^2\gg\nu^2$ (i.e., low pressure and high frequency), Eq. (1) becomes

$$\sigma_c=-\frac{ne^2}{3m\omega^2}\int(\nu-j\omega)v\frac{\partial f_0}{\partial v}d^3v=\frac{ne^2}{m\omega^2}(\nu_{\text{eff}}-j\omega), \qquad (3)$$

where ν_{eff} is the effective electron collision frequency and can be shown to be

$$\nu_{\text{eff}}=\tfrac{4}{3}N\bar{Q}_m\langle v\rangle, \qquad (4)$$

for f_0 to be Maxwellian. The average velocity of the electrons $\langle v\rangle$ is given by

$$\langle v\rangle=(8kT/\pi m)^{1/2}. \qquad (5)$$

Here k is the Boltzmann constant and T is the electron temperature. $\bar{Q}_m$ is the effective-collision cross section for momentum transfer of the electrons with the neutrals and is given by

$$\bar{Q}_m=\frac{1}{8}\left(\frac{m}{kT}\right)^3\int_0^\infty Q_m(v)v^5\exp\left(-\frac{mv^2}{2kT}\right)dv. \qquad (6)$$

$\bar{Q}_m$ as calculated from the measured ν_{eff} as a function of temperature is shown in Fig. 2. Data were taken generally 1 to 2msec after termination of the excitation pulse (i.e., in the afterglow). The electrons are assumed to have relaxed back to the gas temperature at times that the measurements were made. This is justified through comparison of the mobility data (Cs^+ ions in Cs) as calculated from the measured characteristic ambipolar diffusion time constants with those deduced from the helium-cesium mixture experiments (see Sec. V). Within experimental errors, they are generally the same. The fast cooling of the electrons in the afterglow of pure cesium discharge is attributed to the phenomenon of "diffusion cooling."[9]

The collision probability for momentum transfer is defined as

$$P_m=NQ_m/p_0, \qquad (7)$$

where p_0 is the gas pressure in mm Hg referred to 0°C.

MEASUREMENTS IN CESIUM-HELIUM MIXTURES

In order to insure that the electrons have relaxed back to the gas temperature at times in the afterglow that the data were taken, helium gas of known density was admitted to the discharge tube.[10] Since

[7] C. J. Smithells, *Metals Reference Book* (Interscience Publishers, Inc., New York, 1955), 2nd ed., Vol. II, p. 613.

[8] H. Margenau, Phys. Rev. **69**, 508 (1946).

[9] M. A. Biondi, Phys. Rev. **93**, 1136 (1954).

[10] The temperatures of the electron gas in the present experiments are assumed to be equal to those of the gas at times in the afterglow (0.8 to 5 msec) the measurements are made. The effectiveness of helium as a recoil gas in bringing down the electrons from the high temperature in the active discharge to the gas temperature has been checked in the case of He-Xe mixtures by comparing the noise power emitted from the decaying plasma with a calibrated standard noise source in conjunction with a maser. It is shown that the electrons relax back to the gas temperature shortly (within 200 μsec) after termination of the breakdown voltage pulse.

$\nu = \sum_i N_i Q_{m_i} v$, where i is the index representing different species of gas molecules in the discharge tube, it can easily be shown that

$$\nu_{\mathrm{eff}'} = \sum_i g_i \nu_{\mathrm{eff}\,i'}, \tag{8}$$

where the primes refer to the quantity per unit pressure and g_i is the fractional concentration of the ith species of gas molecules provided $\omega^2 \gg \nu^2$. In our case, the effective electron-cesium collision frequency for momentum transfer is calculated from the measured $\nu_{\mathrm{eff}'}$ by subtracting the electron-helium part using existing data.[11] $\bar{Q}_m$ for electron-cesium so calculated is shown in Fig. 2. The value for $\bar{Q}_m$ for electron-helium is taken to be 5.3×10^{-16} cm^2 and has been shown to be independent of the electron energy from about 0.04 eV to about 2eV.[11]

Theoretically, it is required to know the functional dependence of $Q_m(v)$ over the entire velocity spectrum in order to be able to calculate $\bar{Q}_m$ according to Eq. (6) and compare it with the experimentally determined one. This is not available at the present. Experimentally, $Q_m(v)$ could be determined if enough data of $\bar{Q}_m$ as a function of temperature were available. A least-squares fit to the present experimental points of $\bar{Q}_m$ by a polynomial of the form

$$Q_m(v) = A/v^2 + B/v + C \tag{9}$$

gives

$$A = 98.7\ \mathrm{cm}^4/\mathrm{sec}^2, \quad B = -8.04\times10^{-6}\ \mathrm{cm}^3/\mathrm{sec},$$

and

$$C = 2.3\times10^{-13}\ \mathrm{cm}^2.$$

Here v is the electron velocity in cm/sec. It is felt that this simple polynomial approximation is adequate because of the following reasons: (1) In the temperature range (450 to 550°K), the fraction of electrons possessing an energy higher than 0.5 eV is very small and the effect of these electrons on $\bar{Q}_m$ is negligible. (2) The fluctuation of the data (due mostly to the temperature drifts of the order of one degree in the experiment) prevents us from doing anything more meaningful.

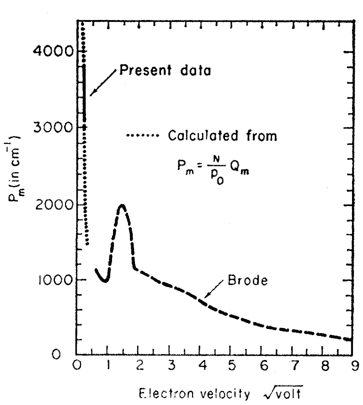

FIG. 3. Collision probability of electrons with cesium atoms.

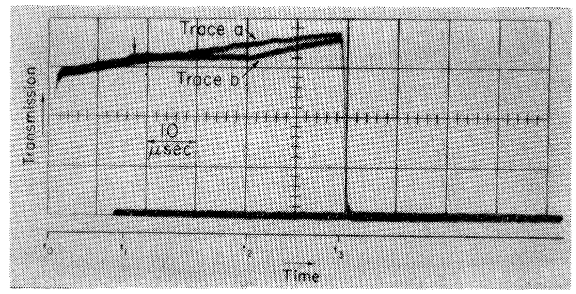

FIG. 4. Microwave cross modulation in helium-cesium mixture.

The velocity dependence of the electron collision probability for momentum transfer P_m calculated according to Eq. (7), together with the existing data by Brode,[3] is shown in Fig. 3. The extrapolated curve beyond the thermal range (approximately 0.06 to 0.071 eV) of the present experiment, according to Eqs. (9) and (7), exhibits a smooth tendency to join Brode's data at high energies.

To exhibit experimentally this strong velocity dependence of the electron-cesium-atom collision probability, we have performed microwave cross modulation[5,12] experiments in the decaying plasma established in helium-cesium mixture. The electron temperature dependence of $\bar{Q}_m$ for electron and cesium atoms as calculated from Eqs. (6) and (9) is

$$\bar{Q}_m = 1.61\times10^{-10} T_e^{-1} - 9.63\times10^{-12} T_e^{-1/2} + 2.03\times10^{-13}\ \mathrm{cm}^2.$$

If the contribution of electron-ion collisions to the measured effective electron collision frequency is negligible as in the present experiment, it can be shown immediately from Eqs. (4) and (8) that the effective electron collision frequency in helium-cesium mixture, in the temperature range of present interest, is

$$\nu_{\mathrm{eff}} = \tfrac{3}{4}(8k/\pi m) \times [(5.3\times10^{-16}[p_0(\mathrm{He})/p_0(\mathrm{Cs})] + 2.03\times10^{-13}) T_e^{1/2} + 1.61\times10^{-10} T_e^{-1/2} - 9.63\times10^{-12}] N_{\mathrm{Cs}}, \tag{10}$$

where $p_0(\mathrm{He})$ and $p_0(\mathrm{Cs})$ are the partial pressures of helium and cesium gases referred to 0°C. N_{Cs} is the cesium gas density in numbers per cubic centimeter. The effective electron collision frequency, as given by Eq. (10), has a minimum value at an electron temperature of

$$T_e = \frac{1.61\times10^{-10}}{5.3\times10^{-16}[p_0(\mathrm{He})/p_0(\mathrm{Cs})] + 2.03\times10^{-13}}\ {}^\circ\mathrm{K}. \tag{10a}$$

Since the attenuation of microwave traversing through a weakly ionized gas is proportional to ν_{eff}, the transmitted wave should exhibit a maximum value at the temperature given by Eq. (10a) if T_e is changed and

[11] L. Gould and S. C. Brown, Phys. Rev. **95**, 897 (1954); J. M. Anderson and L. Goldstein, *ibid.* **102**, 933 (1956); J. L. Pack and A. V. Phelps, *ibid.* **121**, 798 (1961).

[12] L. Goldstein, J. M. Anderson, and G. L. Clark, Phys. Rev. **90**, 151 (1953).

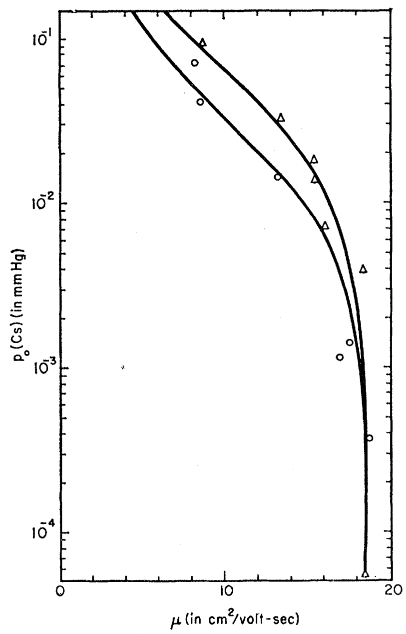

FIG. 5. Measured cesium ionic mobility (referred to standard gas density, i.e., $2.69\times10^{19}/\text{cm}^3$) in helium-cesium mixtures as a function of cesium partial pressure. The circular and triangular points are those with helium partial pressures of 1.82 and 3.5 mm Hg, respectively. The solid curves are calculated from Blanc's law with $\mu(\text{Cs}^+ \text{ in He})=18.5$ cm²/V-sec and $\mu(\text{Cs}^+ \text{ in Cs})=0.4$ cm²/V-sec.

passes through this value. This is shown typically in Fig. 4. In this case, a rectangular, 9410-Mc/sec, 10-μW sensing wave pulse of 60-μsec duration $[t_0t_3]$ is propagating through the decaying plasma 700 μsec after termination of the breakdown voltage pulse ($\sim$10 μsec). At times shortly after introducing the sensing wave (about 15 μsec), a rectangular, 8650-Mc/sec, $\sim$120-mW, $\sim$20-μsec duration disturbing wave pulse $[t_1t_2]$ is launched into the plasma. A fraction of this energy is absorbed by the electron gas, and its temperature is hence increased. The change of ν_{eff} and, hence, the real part of the electrical conductivity of the plasma, is then sensed by the sensing wave. The sensing wave is picked up at the far end of the discharge tube through a ferrite isolator and two microwave filters. The disturbing wave is rejected by the filter and is absorbed by the ferrite isolator. The traces shown in Fig. 4 are double exposures of the oscilloscope traces. Traces a and b are the transmitted sensing wave without and with the disturbing wave present. It is predicted from Eq. (10a) for this particular case $[p_0(\text{He})=1.82$ mm Hg and $p_0(\text{Cs})=0.071$ mm Hg$]$ that the minimum value of ν_{eff} should occur at $T_e=745$°K while the background gas temperature is 495°K. A bump is observed (as shown by an arrow in Fig. 4) shortly after initiation of the disturbing microwave pulse. This confirms qualitatively the predicted pattern of electromagnetic wave interaction with a plasma established in a helium-cesium mixture and is a further evidence that we do have a decaying plasma in which electrons have already been thermalized with the background gas at times in the afterglow studied.

MOBILITY OF CESIUM IONS

By applying Blanc's law[13] which states that the reciprocal of the mobility in a binary mixture should be a linear function of the concentration of either of its constituents, i.e.,

$$\frac{1}{\mu}=\frac{g_1}{\mu(\text{Cs}^+ \text{ in He})}+\frac{g_2}{\mu(\text{Cs}^+ \text{ in Cs})}, \qquad (10)$$

the mobilities of cesium Cs^+ ions in helium $\mu(\text{Cs}^+ \text{ in He})$ and in cesium $\mu(\text{Cs}^+ \text{ in Cs})$ can then be determined. g_1 and g_2 are the fractional concentrations of helium and of cesium, respectively. The mobility of Cs^+ ions in the mixture μ is calculated from the time constant of electron-density decay curve by

$$D_a=\Lambda^2/\tau_a\simeq 2D_+, \qquad (11)$$

and

$$D_+/\mu=kT/e. \qquad (12)$$

Here D_a and D_+ are the ambipolar and ionic diffusion coefficients, respectively. Λ is the characteristic diffusion length of the discharge tube and τ_a is the measured characteristic ambipolar diffusion time constant. From the best fit to the experimental points (see Fig. 5), we get $\mu(\text{Cs}^+ \text{ in He})=18.5\pm0.5$ cm²/V-sec and $\mu(\text{Cs}^+ \text{ in Cs})=0.4\pm0.05$ cm²/V-sec.

DISCUSSION

The elastic-collision cross section of low-energy electrons with heavy atoms is difficult to compute theoretically, and only very few calculations have been performed.

Robinson[14] has calculated $Q_m(v)$ for cesium by using a polarization potential in addition to a scattering potential constructed from a Slater-orbital-type wave function. His preliminary analysis reproduces the shape of the cross section versus energy curves at high energies but disagrees considerably with the present experiment at low energies. Recently, Phelps[15] has calculated the electron collision frequency with cesium atoms from Mohler's data[2] and found $\nu/N_{\text{Cs}}\simeq1.6\times10^{-6}$ cm³/sec-atom for electrons of $\sim$0.22 to 0.40 eV. We arrive at an expression

$$\nu/N_{\text{Cs}}=1.34\times10^{-4}T_e^{-1/2}-8\times10^{-6}+1.66\times10^{-7}T^{1/2} \text{ cm}^3/\text{sec-atom}$$

from Eq. (9). This yields $\nu/N_{\text{Cs}}=1.95\times10^{-6}$, 1.76×10^{-6}, and 1.66×10^{-6} cm³/sec-atom for electrons

[13] L. B. Loeb, *Basic Processes in Gaseous Electronics* (University of California Press, Berkeley, 1955), Chap. 1.
[14] L. B. Robinson, Aerospace Corporation, Report No. TDR-594 (1203-01)TR2, 1961 (unpublished).
[15] A. V. Phelps (private communications).

at 450, 500, and 550°K, respectively, and in fair agreement with each other.

As to the mobility of Cs^+ ions in helium, the polarization force is believed to be the dominating interacting force at such low energies. Langevin's theory[16] (to this limit) gives a value of 15.8 cm^2/V-sec in contrast with 18.5±0.5 cm^2/V-sec determined by the present experiment. Tyndall *et al.*[17] used shutter methods to determine the mobilities of alkali ions in helium and found a value of 18.4 cm^2/V-sec for Cs^+ ions in helium, in excellent agreement with the present experiment.

[16] P. Langevin, Ann. chim. et phys. **5**, 245 (1905).

[17] A. M. Tyndall, *The Mobility of Positive Ions in Gases* (Cambridge University Press, Cambridge, New York, 1938).

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Hyperfine Structure of the Hydrogen Molecule in its Metastable $^3\Pi_u$ State*

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The hfs of the hydrogen molecule in its metastable excited state $^3\Pi_u$ is calculated theoretically using Amemiya's wave function. The theoretical separation of the $F=3$ and $F=2$ states is 618 Mc/sec, and that for the $F=2$ and $F=1$ states is 412 Mc/sec, for $K=1$, $J=2$; these separations are to be compared with the observed values of 707.55 Mc/sec and 462.44 Mc/sec, respectively. The theoretical second-order correction is found to explain the deviation of the observed values from the interval rule.

INTRODUCTION

THE hyperfine structure of the hydrogen atom in its ground state produces a resonance at 1420 Mc/sec. Observation of this radio-frequency line by means of radio telescopes gave us extensive data on the distribution of hydrogen atoms in space.[1] This line, however, is the only spectral line observed by radio telescopes so far. Since there exist so many hydrogen atoms in space, one expects that there must be some hydrogen molecules in space also. Bates[2] calculated the cross section of formation of molecules from atomic hydrogen by collision and recently McCrea and McNally[3] estimated the rate of molecular formation at the surface of interstellar dust. The existence of molecular hydrogen, however, cannot be detected by using a radio telescope directly, since the ground state of the neutral hydrogen molecule does not have an appreciable hfs splitting. The difference between atomic and molecular hydrogen is due to the fact that in atomic hydrogen the spin of the electron produces a strong magnetic field at the center to produce a large interaction with the proton magnetic moment, while molecular hydrogen in its ground state cannot produce such a strong magnetic field since the resulting electronic spin and orbital angular momenta are both zero.

There are, however, possibilities of having an appreciable hfs if one goes to the ionized hydrogen molecule H_2^+. One of the present authors (M. Mizushima) did some calculations[4] on this molecule ion, and Burk[5] and Dalgarno *et al.*[6] published calculations of the same nature independently. In this case the resulting electronic spin is 1/2 which produces a strong magnetic field for protons to couple. The other possibility is the neutral hydrogen molecule in its excited $^3\Pi_u$ state. In this excited state the finite electronic angular momenta can produce a large magnetic hfs. This particular state, $^3\Pi_u$, happens to be a metastable state since the only other triplet state below this state is a $^3\Sigma_u^+$ state which cannot be combined with our state by an electric dipole transition. The $^3\Sigma_u^+$ state, to which our $^3\Pi_u$ state goes by a forbidden transition, is a dissociative state. The molecule in the $^3\Pi_u$ state, therefore, decays into atomic hydrogen through the $^3\Sigma_u^+$ state.

Lichten[7] first showed the metastability of this $^3\Pi_u$ state in his molecular beam experiment. His later experiment[8] showed that some states, including the states we are considering in this paper, have a lifetime of 0.1 to 0.5 msec, and the hfs of the $K=1$, $J=2$ state is such that

$$(F=3)\leftrightarrow(F=2)=707.55\ \text{Mc/sec},$$
$$(F=2)\leftrightarrow(F=1)=462.44\ \text{Mc/sec}. \quad (1)$$

Fontana[9] recently calculated the fine structure of this state by using an approximate wave function.

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[1] For example, see J. L. Pawsey and R. N. Bracewell, *Radio Astronomy* (Oxford University Press, New York, 1955).

[2] D. R. Bates, Monthly Notices Roy. Astron. Soc. **111**, 303 (1951).

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[4] M. Mizushima, Astrophys. J. **132**, 493 (1960).

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[6] A. Dalgarno, T. N. L. Patterson, and W. B. Somerville, Proc. Roy. Soc. (London) **A259**, 100 (1961).

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[8] W. Lichten, Phys. Rev. **126**, 1020 (1962).

[9] P. R. Fontana, Phys. Rev. **125**, 220 (1962).

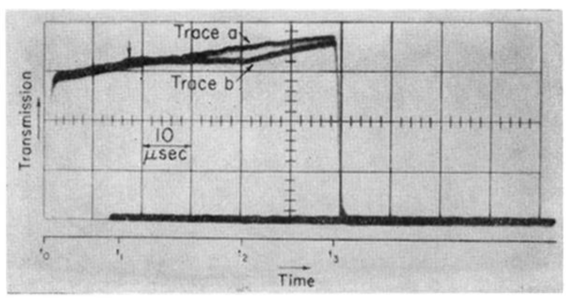

FIG. 4. Microwave cross modulation in helium-cesium mixture.