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Deflection of CsF molecules by resonant inhomogeneous electric fields*

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An experiment that tests a novel method for deflecting molecular beams using inhomogeneous resonance fields is described. The idea is easily understood in classical terms. Imagine that the rotating electric dipole moment of a polar diatomic molecule is placed in an electric field. In a static field the $\vec{\mu}\cdot\vec{E}$ interaction averages to zero due to the rotation of the molecule. If, however, the field rotates at the same frequency as the molecule, the field and molecule remain aligned and the $\vec{\mu}\cdot\vec{E}$ interaction does not average to zero. If the field is spatially inhomogeneous, then there is a net force $-\nabla(\vec{\mu}\cdot\vec{E})$ on the molecule. This force can be used in making a molecular beam deflector. Resonance deflection of molecules in the $J=0$ and 1 rotational states of a molecular beam of CsF has been observed. The deflection is produced by passing a collimated molecular beam of CsF through the center of a TE_{111} microwave cavity which is oscillating at the $J=0\rightarrow1$ transition frequency, 11.019 GHz. The deflection of the $J=0$ and 1 molecules can be detected as a decrease in the undeflected on-axis beam or as an increase in the off-axis beam. The theory of the effect as well as potential applications are also discussed.

INTRODUCTION

Since the Stern-Gerlach experiment, molecular-beam-deflection techniques have played an ever-expanding role in atomic and molecular physics. A wide variety of methods have been developed for deflecting and focusing beams using both magnetic and electric fields. An excellent review of these methods is given by Ramsey.[1] We discuss here a method for deflecting a rotating dipole using an inhomogeneous field which is rotating at the same frequency as the dipole. Specifically, we have used an oscillating electric field which acts on the rotating electric dipole moment of CsF to deflect a molecular beam of CsF. An analogous experiment using magnetic fields and the magnetic moment of ground-state potassium has been done by Bloom *et al.*[2]

The concept is easily understood in classical terms. Let us represent the molecule as a dumbbell, with oppositely charged ends, tumbling end over end at angular frequency ω. Let us assume that the molecule is rotating so that the dipole moment μ has a projection in the x direction given by

$$\mu_x=\mu\cos\omega t. \tag{1}$$

Since force on a dipole is given by

$$\vec{F}=-\nabla(\vec{\mu}\cdot\vec{E}), \tag{2}$$

then if we put the dipole of Eq. (1) in a static field the force oscillates at angular frequency ω and, to first order, has a time average of zero. However, consider what happens in an oscillating field. For simplicity let us assume that the inhomogeneous electric field and gradient are both in the x direction and write

$$E_x=E(x)\cos(\omega t+\phi), \tag{3}$$

where ϕ is the phase angle between the field and the dipole. Now if we compute the time average force $\langle F_x\rangle$ on the dipole of Eq. (1) produced by the field of Eq. (3) we find

$$\langle F_x\rangle=-\tfrac{1}{2}\mu\frac{dE}{dx}\cos\phi. \tag{4}$$

Depending on the phase angle ϕ between the field and the dipole, the time average deflecting force is either in the $+x$ or $-x$ direction with a maximum value $F_{\max}$ of

$$F_{\max}=\mu\frac{dE}{dx}. \tag{5}$$

At this point it is worth noting the difference between this method and the static electric deflection method first developed by Hughes[3] and Trishka.[4] The static electric deflection process is based on an effect which is second order in the applied field and consequently requires a strong field as well as a strong gradient. In contrast, the resonant deflection technique is a first-order effect which only requires a strong gradient.

The quantum-mechanical treatment of this phenomenon differs only in a few respects: the rotating dipole is replaced by the electric dipole transition-matrix element, the rotation frequency is replaced by the transition frequency between two quantum levels, and the projection of $\vec{\mu}$ on $\vec{E}$ is not free to take all projection angles but is quantized into various projected components parallel and antiparallel to $\vec{E}$. The states which are deflected are symmetric and antisymmetric combinations of the two stationary states involved in the transition. The correspondence between the rotating dipole and a transition-matrix element suggests the possibilities of this method. A beam of any atom or molecule having allowed electric or magnetic dipole transitions can be deflected. Consider a few examples. With microwave transitions virtually any pair of rotational states of a polar molecule can be selectively deflected; whereas with static electric fields only a few low rotational states can be deflected. If we used infrared radiation instead of microwave radiation, we could drive the vibrational transitions of a molecule to make a vibrational state selector.

We have observed the resonance deflection of molecules in the $v=0$, $J=0$, and $J=1$ states of CsF produced by passing a molecular beam of CsF through an inhomogeneous electric field in a microwave cavity. The microwave cavity was tuned to the CsF $v=0$, $J=0\rightarrow 1$ rotational resonance frequency of 11.019 GHz. The results are qualitatively in agreement with the predictions of a two-level theory given below. We have observed the deflections, and the linewidth increases with the electric field. However, the deflection signal and the linewidth are both smaller than predicted. We have considered several possible explanations for these discrepancies—unknown microwave power losses, collisional dephasing, and the possibility of admixing higher rotational states. The discrepancies are not clearly resolved at present, suggesting that more work on the problem is needed. Theoretical work will be needed to provide a more detailed understanding of the deflection process. Experimentally there seem to be two fruitful avenues for future research. One is to do similar microwave rotational resonance experiments using more sophisticated molecular-beam sources, such as a nozzle-beam source, to increase the size of the resonantly deflected beam and facilitate a more detailed investigation of the effect. The second is to extend the technique to new spectral regions such as the infrared and visible.

In the following sections we shall give a quantum-mechanical derivation for a simple two-level system, discuss the experiment and its results, and finally discuss a few possible causes for some discrepancies between our calculations and observations as well as potential applications of the technique.

TWO-LEVEL THEORY

By treating the problem as a two-level quantum system we can develop expressions not only for the deflections but also for the line shape of the deflection as a function of the frequency of the applied field. Bloom and Erdman used a Fourier transform method to treat the case when the frequency of the applied field is exactly resonant with the transition frequency.[5] Here we shall outline a similar approach extended to the off-resonant case. In essence the method is to Fourier transform the spatial wave function to determine the change in momentum transverse to the molecular beam's direction produced by the field. Implicit in this is the assumption that the deflecting force is small so that there is no transverse displacement in the region of the applied field.

Let us assume that the molecular beam is traveling in the z direction, that the oscillating field is in the x direction, and its amplitude is a function of x. Let us assume the two quantum levels have the orthonormal states Ψ_+ and Ψ_-, with energies $\hbar W_+$ and $\hbar W_-$. The wave function for the system is then given by

$$\Psi(x,t)=f(x)[F_+(t)e^{-iW_+t}\Psi_+ + F_-(t)e^{-iW_-t}\Psi_-]; \quad (6)$$

$f(x)$ gives the spatial distribution of the beam in the x direction (transverse to the beam's direction of travel). The spread in x momenta is given by $\Phi(k,t)$, the Fourier transform of $f(x)$,

$$\Phi(k,t)=\frac{1}{(2\pi)^{1/2}}\int_{-\infty}^{\infty}\Psi(x,t)e^{ikx}\,dx. \quad (7)$$

Initially all the x dependence of Eq. (7) is in f, so let us define $g(k)$ as the Fourier transform of $f(x)$,

$$g(k)=\frac{1}{(2\pi)^{1/2}}\int_{-\infty}^{\infty}f(x)\,e^{ikx}\,dx; \quad (8)$$

$g(k)$ is maximum at $k=0$ and its width reflects the angular divergence of the molecular beam. The oscillating field, given by $E(x)\cos\omega t$, couples the two states via the matrix element ω_1, where

$$\omega_1=\frac{\langle\Psi_+|\vec{\mu}\cdot\vec{\mathrm{E}}(x)|\Psi_-\rangle}{\hbar}\cos\omega t.$$

Although we have not written it explicitly, ω_1 is a function of x, and it is this dependence which gives rise to the deflection.

The general problem of a strong resonant or nearly resonant oscillating field interacting with a two-level system has been investigated extensively by Autler and Townes.[6] In the resonance deflection experiment the applied field is sufficiently weak so that $\omega_1 \ll W_+ - W_-$ (Autler and Townes's "weak-field" case). This allows us to make the rotating-field approximation, that is, we can replace the oscillating $\cos\omega t$ with the resonant term of its equivalent-rotating and counter-rotating form $\frac{1}{2}(e^{i\omega t}+e^{-i\omega t})$, ignoring the antiresonant term. The Schrödinger equation is then soluble exactly and the antiresonant field leads to a second-order shift in the resonance frequency, the Bloch-Siegert effect.[1] Using the wave function of Eq. (6) and making the rotating-field approximation, the time dependence of the Schrödinger equation is given by the two coupled equations

$$\dot{F}_\pm(t) = (-i\omega_1/2)e^{\mp i\omega t}\, e^{\pm\omega_0 t}\, F_\mp(t), \tag{9}$$

where $\omega_0 = W_+ - W_-$.

Following the method of Ramsey[1] these may be solved to give

$$\begin{aligned} F_+(t) &= e^{-i\Delta\omega t/2}(Ae^{i\Omega t/2} + Be^{-i\Omega t/2}) \\ F_-(t) &= e^{i\Delta\omega t/2}(Ce^{i\Omega t/2} + De^{-i\Omega t/2}) \end{aligned} \tag{10}$$

where $\Delta\omega = \omega - \omega_0$,

$$\Omega = (\omega_1^2 + \Delta\omega^2)^{1/2},$$

and A, B, C, and D are given by the initial conditions. Initially $F_+(t)$ and $F_-(t)$ are not functions of x so that if $F_+(0) = Q_+$ and $F_-(0) = Q_-$ then

$$\Psi(x, 0) = f(x)[Q_+\Psi_+ + Q_-\Psi_-] \tag{11}$$

where

$$|Q_+|^2 + |Q_-|^2 = 1.$$

Applying these initial conditions to Eqs. (9) and (10) yields

$$\begin{aligned} A &= \frac{\Omega + \Delta\omega}{2\Omega}Q_+ - \frac{\omega_1}{2\Omega}Q_-, \quad B = \frac{\Omega - \Delta\omega}{2\Omega}Q_+ + \frac{\omega_1}{2\Omega}Q_-, \\ C &= \frac{\Omega - \Delta\omega}{2\Omega}Q_- - \frac{\omega_1}{2\Omega}Q_+, \quad D = \frac{\Omega + \Delta\omega}{2\Omega}Q_- + \frac{\omega_1}{2\Omega}Q_+. \end{aligned} \tag{12}$$

In principle, the momentum distribution can be found from the Fourier transform of the wave function. The wave function implied by Eq. (10) is given by

$$\begin{aligned} \Psi(x, t) = f(x)[&(Ae^{i\Omega t/2} + Be^{-i\Omega t/2})e^{-i\Delta\omega t/2}e^{-iW_+ t}\,\Psi_+ \\ &+ (Ce^{i\Omega t/2} + De^{-i\Omega t/2})e^{i\Delta\omega t/2}e^{-iW_- t}\,\Psi_-], \end{aligned} \tag{13}$$

which is not particularly easy to transform. It is the x dependence of ω_1 and hence of Ω that produces the deflections, so let us replace Ω by

$$\Omega = \Omega|_{x_0} + \left.\frac{d\Omega}{dx}\right|_{x_0}(x - x_0) = \Omega_0 + \Gamma x \tag{14}$$

where x_0 is the value of x at which the molecular beam passes through the deflection region and $\Gamma = (d\Omega/dx)_{x_0}$.

In Eq. (13) Ω enters in the exponents and through A, B, C, and D. The most sensitive terms for the Fourier transforms are the exponentials, so if we ignore the x dependence of A, B, C, and D, and take the Fourier transform of the result using Eq. (14) for Ω, we have

$$\begin{aligned} \varphi(k, t) = [&Ae^{i(\Omega_0/2 - \Delta\omega/2 - W_+)t}\,g(k + \tfrac{1}{2}\Gamma t), \\ &+ Be^{-i(\Omega_0/2 + \Delta\omega/2 + W_+)t}\,g(k - \tfrac{1}{2}\Gamma t)]\,\Psi_+, \\ &+ [Ce^{i(\Omega_0/2 + \Delta\omega/2 - W_-)t}\,g(k + \tfrac{1}{2}\Gamma t) \\ &+ De^{-i(\Omega_0/2 - \Delta\omega/2 + W_-)t}\,g(k - \tfrac{1}{2}\Gamma t)]\,\Phi_-. \end{aligned} \tag{15}$$

Taking the momentum distribution of Eq. (15) yields

$$\begin{aligned} |\varphi(k, t)|^2 = &[|A|^2 + |C|^2]\,|g(k - \tfrac{1}{2}\Gamma t)|^2 \\ &+ [|B|^2 + |D|^2]\,|g(k + \tfrac{1}{2}\Gamma t)|^2. \end{aligned} \tag{16}$$

The physical interpretation of Eq. (16) is that after a time t in the deflecting field the beam is split into two oppositely deflected components having transverse x momenta $\pm\frac{1}{2}\hbar\Gamma t$. The two beam components are the symmetric and antisymmetric combinations of the Ψ_+ and Ψ_- states.

The main features of the frequency dependence of the deflection can be seen by neglecting the small dependence of A, B, C, and D on $\Delta\omega$ and considering only the dependence of Γ on $\Delta\omega$. Since the transverse momentum gained after a time t in the deflection field is given by $\frac{1}{2}\hbar\Gamma t$, the magnitude of the force is given by $\frac{1}{2}\hbar\Gamma$.

$$\frac{\hbar\Gamma}{2} = \frac{1}{2}\frac{d\Omega}{dx} = \frac{\hbar\omega_1}{2(\omega_1^2 + \Delta\omega^2)^{1/2}}\frac{d\omega_1}{dx}. \tag{17}$$

At resonance, this reduces to the classical expression of Eq. (5). We can rewrite this expression as

$$\frac{\hbar\Gamma}{2} = \frac{\hbar f(\omega)}{2}\frac{d\omega_1}{dx}, \tag{18}$$

where

$$f(\omega) = \frac{\omega_1}{(\omega_1^2 + \Delta\omega^2)^{1/2}}, \tag{19}$$

and contains the line-shape information. This line shape is essentially the square root of the familiar Lorentzian line seen in transition probabilities. This difference occurs because the deflecting force depends on the transition dipole matrix element rather than its square as does a transition probability. At resonance the full width at half-maximum (FWHM) of the angular frequency scan is given by $2(3)^{1/2}\omega_1$. In frequency units then

$$\text{FWHM} = 2(3)^{1/2}\omega_1/2\pi. \tag{20}$$

Using Eq. (18) we may now compute the deflection produced in molecules in a thermal beam. Assuming the beam has z momentum p_z and velocity v_z, then at resonance the angular deflection γ produced in the x direction is given by

$$\gamma = \frac{p_x}{p_z} = \frac{\hbar\tau\, d\omega_1/dx}{2p_z}, \tag{21}$$

where τ is the length of time the molecule spends in the deflection region. If the deflection region is of length l and the molecular mass m, then the angular deflection γ_a of molecules traveling at the most probable velocity α is given by

$$\gamma_\alpha = \frac{\hbar\, d\omega_1/dx l}{2m\alpha^2} = \frac{\hbar\, d\omega_1/dx l}{4kT}, \tag{22}$$

where k is Boltzmann's constant and T is the beam temperature.

APPLICATION TO CsF

The relevant molecular constants for CsF are listed in Table I.

The rotational transitions are electric dipole transitions between the $J=0$ and $J=1$ rotational states so that ω_1 is given by

$$\omega_1 = (\mu E/\hbar)\langle J=0, m=0|\cos\theta| j=1, m=0\rangle. \tag{23}$$

We have found that a convenient conversion factor to remember is

$$\frac{\mu E}{h} = 0.503\ \mu(\text{debye})E(\text{V/cm}), \tag{24}$$

thus ω_1 can be expressed in frequency units as

$$f_1(\text{MHz}) = \frac{\omega_1}{2\pi} = 0.290\ \mu(\text{debye})E(\text{V/cm}). \tag{25}$$

The weak-field requirement is that f_1 be less than the rotational resonance frequency of 11.019 GHz.

Until this point we have ignored the hyperfine structure of the rotational states. If f_1 is small compared to the hyperfine interval, then each hyperfine component must be treated separately. If on the other hand f_1 is considerably greater than the hyperfine interval, the rotational moment is coupled more strongly to the external field than to the cesium nucleus so that the rigid-rotor states are good quantum states and the hyperfine structure may be ignored.

In sum, the requirements of f_1 are that 0.3 MHz $\ll f_1 \ll 11$ GHz. This implies that a microwave field strength must lie in the range 0.1 V/cm $\ll E \ll 10^3$ V/cm.

TABLE I. Relevant molecular constants for CsF.

Molecular constant	Symbol	Value
Electric dipole moment	μ	7.87 debye [a]
Rotational constant	B	5527.34 MHz [a]
Vibrational constant	ω_e	352 cm^{-1} [b]
$J=1$ hyperfine interval	W_{HF}	0.3 MHz [c]

[a] A. Honig, M. Mandel, M. L. Stitch, and C. H. Townes, Phys. Rev. 96, 629 (1954).

[b] S. E. Veazey and W. Gordy, Phys. Rev. 138, A1303 (1965).

[c] T. C. English and J. C. Zorn, J. Chem. Phys. 47, 3896 (1967).

To calculate the expected deflections for molecules at the most probable beam velocity, we can rewrite Eq. (22) using Eq. (23) as

$$\gamma_\alpha = \frac{\mu\, dE/dx}{4(3)^{1/2}kT}\, l = \frac{h[d(\mu E/h)/dx]}{4(3)^{1/2}kT}\, l. \tag{26}$$

This can be evaluated for CsF using our apparatus parameters of $l=20$ cm and $T=1000$°K to be

$$\gamma_\alpha = 5.5\times 10^{-7}\frac{dE}{dx}\ (\text{V/cm}^2). \tag{27}$$

According to Eq. (35), 9 watts of microwave power produces a gradient of 4.3×10^2 V/cm^2. Substituting this into Eq. (22) we find that $\gamma_\alpha = 2.4\times10^{-3}$ rad. As the beam divergence is 2×10^{-4} rad, this large a deflection for molecules with the most probable velocity suggests that nearly all of the molecules in the $J=0$, $m=0$ and $J=1$, $m=0$ states will be deflected out of the beam.

On this basis we can estimate our signal to be the fraction of the beam in the $J=0, m=0$ and $J=1$, $m=0$ rotational states and the $v=0$ vibrational state. At a temperature of 1000°K the fraction of the beam in these states is 7.6×10^{-5}. If we use the electrostatic low-J-state selector, the fraction of the beam which we expect to see is reduced to 3×10^{-5}.

From Eqs. (20) and (24) we can see that the FWHM of the deflection linewidth is given by

$$\text{FWHM}(\text{MHz}) = 1.006\mu\ (\text{debye})E(\text{V/cm}). \tag{28}$$

To exactly represent the line shape we should average over the cross section of the molecular beam and along its path through the microwave cavity. Since we do not know the position of the molecular beam relative to the cavity to better than a beam diameter, it seems to us that using some average field sampled by the molecular beam should be sufficiently accurate. If the beam center is off the cavity center line by a distance x_0, then a reasonable value for the average field sampled by the beam is $x_0\,(dE/dx)$, the field at the center of the molecular beam.

Using this for the average field we can rewrite Eq. (28) as

$$\text{FWHM}(\text{MHz}) = 1.006\mu\ (\text{debye})x_0(\text{cm})\frac{dE}{dx}\ (\text{V/cm}^2). \tag{29}$$

When the beam is nominally on the cavity centerline, the field at the center of the molecular beam is nominally zero so we shall simply assume a misalignment of half a beam diameter.

EXPERIMENT

The basic design of the experiment is quite simple. Figure 1 shows a plan drawing of the apparatus, which is divided into a source, deflection region, and detection region. The deflection and detection regions are maintained as a single vacuum chamber operating in the range $(0.4-2)\times10^{-7}$ Torr. The source chamber has a separate vacuum pump and operates from pressures $(1-4)\times10^{-7}$ Torr. Both chambers are pumped by oil diffusion pumps with refrigerated baffles, and the flanges are sealed with copper or aluminum gaskets.

The source is a conventional tube oven. It is made of 0.015-cm-wall stainless-steel tubing with a 0.030-cm-diam hole in the side through which the beam effuses. The tube is filled with the beam material, and the ends are crimped. The tube is resistively heated by passing up to 50 A at 2 V through it. Typical operating temperatures are ~1000°K.

The detector is a 0.003-cm-thick and 0.030-cm-wide tungsten ribbon that is aligned vertically. Before using the filament, we coat it with tungsten using tungsten hexacarbonyl to reduce its background noise, following the procedure of Greene.[7] A movable horizontal 0.030 cm slit in front of the detector gives the detector a 0.030 cm square active area. A Bendix electron multiplier is used to amplify the ion current from the detector.

The microwave power is provided by a Varian X-13 klystron, which is then amplified by a traveling-wave tube amplifier. Frequencies are measured with a Hewlett-Packard X532 A wavemeter. To enhance the signal-to-noise ratio we square-wave modulated the microwave power using a resistance-card chopper and used phase-sensitive detection.

The unique part of the apparatus is the microwave cavity that deflects the beam. To see how we arrive at our design, it is helpful to consider the requirements that the deflector must satisfy. It must have a high electric field gradient in the beam region to produce a sizeable deflection. For a deflector 20 cm long, the gradient must be 10^3 V/cm^2. At the same time, the field must be low enough to satisfy the $\mu E \ll B$ criterion to avoid mixing the rotational states.

Our solution is to use a TE$_{111}$ cavity 1.97 cm $\times$1.97 cm$\times$20 cm long. As shown in Fig. 2, the molecular beam travels along the z axis. The electric fields in the cavity are given by

$$\begin{aligned} E_y &= -E_0\cos(\pi x/b)\sin(\pi y/b)\cos(\pi z/d), \\ E_x &= E_0\sin(\pi x/b)\cos(\pi y/b)\cos(\pi z/d), \end{aligned} \qquad (30)$$

where b is the width of the cavity in the x and y directions and d is the length in the z direction. Along the z axis this satisfies the requirements of a high gradient and a low field. The cavity is designed to operate in the neighborhood of 11 GHz, corresponding to $\frac{1}{2}\lambda_0 = 1.4$ cm.

As mentioned earlier a length of about 20 cm is convenient for the deflector. In making a cavity this long for the 11-GHz radiation, one would expect to have about 14 lobes of the standing wave in the z direction. Since the phase of the electric field would reverse 180° in going from lobe to lobe, the beam deflections produced by successive lobes would cancel. To circumvent this we have used a waveguide that is very near cutoff so the guide half-wavelength is 20 cm, producing only one lobe in the z direction. In addition, using a cavity instead of a transmission line allows us to use the available microwave power much more efficiently. The electric field and gradient are proportional to the square root of the Q of the cavity, which we measured to be 2000. The electric field amplitude E_0 in terms of the microwave input power may be expressed as[8]

$$E_0 = (4QP/\pi\epsilon f b^2 d)^{1/2}, \qquad (31)$$

where P is the microwave input power, Q is the Q of the cavity, ϵ is the permittivity of free space, and b and d are the dimensions of the cavity. For the values $Q = 2000$, $\epsilon = 9\times10^{-14}$ farad/cm, f

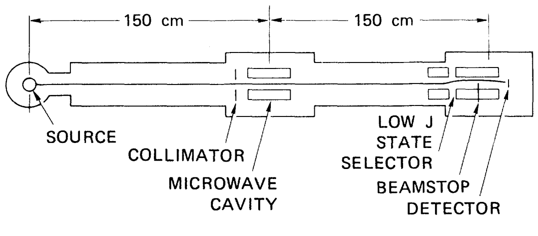

FIG. 1. Schematic diagram of the apparatus.

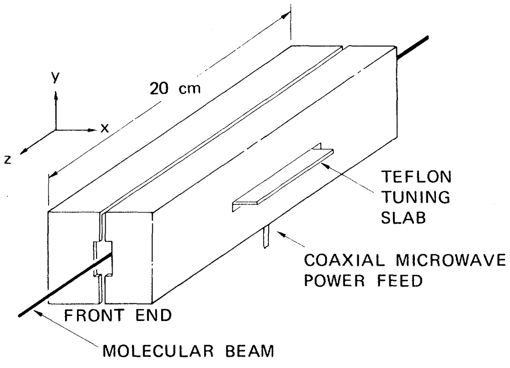

FIG. 2. Perspective view of the TE$_{111}$ cavity.

$=1.1\times10^{10}$ Hz, $d=20$ cm, and $b=2.0$ cm, Eq. (31) reduces to

$$E_0(\mathrm{V/cm^2})=146P^{1/2}(\mathrm{watts})\,. \tag{32}$$

The value of the derivatives of the field in the vicinity of the z axis is

$$\nabla E=\frac{dE_x}{dx}=\frac{dE_y}{dy}=\frac{\pi E_0\cos\pi_z z}{bd}\,. \tag{33}$$

If we use Eq. (33) and average along the z axis we find the average x and y field gradients along the z axis to be

$$\nabla E(\mathrm{V/cm^2})=141P^{1/2}(\mathrm{watts})\,. \tag{34}$$

This value of ∇E may be substituted in the equations of the theory section. Note that along the z axis the components of the field are given by

$$\begin{aligned} E_x&=\nabla E\,x\,,\\ E_y&=\nabla E\,y\,. \end{aligned} \tag{35}$$

Tuning the cavity is quite straightforward. The waveguide is split along the vertical $x=0$ plane. As the two halves are moved apart, the TE_{111} mode oscillation frequency can be varied from 11.2 GHz to below 10 GHz. Fine tuning is accomplished by the symmetric insertion of Teflon tuning slabs in the $y=0$ plane. This tuning is effective over a 100-MHz range. The waveguide is fed by an E probe in the center of the cavity, as shown in Fig. 2.

The cavity has resonances for modes other than the TE_{111} mode. How do we ascertain that we are using the correct one? Higher TE_{11n} modes are present, but all are at higher frequencies than the TE_{111} mode, the nearest being 80 MHz away. Consequently TE_{11n} modes are not a problem. The biggest problem is the TM_{111} mode, which in a perfect rectangular cavity is exactly degenerate with the TE_{111} mode. Fortunately, in machining our cavity it was necessary to leave a radius on the inside corners that splits the TE_{111}, TM_{111} degeneracy by 25 MHz, which is more than adequate to separate the two modes. Another factor in our favor is that the E-field probe couples much better to the TE than to the TM mode, so that even at its resonance the TM_{111} mode is only weakly excited. To verify that we had correctly identified the modes, we used several checks. First, we put an attenuating wooden rod down the z axis of the waveguide. This did nothing to the TE mode but quenched the TM mode, which has an E-field along the z axis. Then we inserted the dielectric tuning slabs into the cavity, shifting the TE mode 100 MHz and the TM mode less than 5 MHz. On the basis of these tests we feel confident that we are, in fact, using the correct mode.

Most of the molecular beam consists of molecules in high rotational states that are useless to us and are the source of virtually all the beam noise. We therefore decided to put a dc state selector in front of the detector to remove these high-J states.[3] The state selector is an electrostatic deflector consisting of an A field, a B field, and a beam stop. It uses a two-wire field geometry, and the deflections are produced in the x direction only by the polarization induced in the molecules by the high electric field. The higher rotational states have lower polarizabilities and hence are deflected only a negligible amount. The deflecting fields bend the trajectories of $J=0$ molecules around a stop and back to the original beam line. The high-J molecules are virtually undeflected and hit the stop. The unique feature of the selector is that the $J=0$ beam leaving the deflector has the same spatial profile as the entering beam in spite of its Maxwellian velocity distribution.

The design of the state selector was dictated by the character of the beam. The undeflected beam has no transverse momentum in the x or y directions and a modified Maxwellian velocity distribution in the z direction. It also has a width of 0.05 cm in the x and y directions. The main concern is not to enlarge the beam in the static deflection process. Since there is no force in the y direction, there is no expansion in that direction.

To avoid expansion in the x direction, we chose the parameters in the following way. Both the A and B fields have the same cross section and applied voltage, 5000 V, and hence the same deflecting force. The length of the B field $l_B=(1+\sqrt{2})l_A$, the length of the A field. This choice ensures that any molecule leaving the B field comes back to the same transverse position x at which it entered the A field, independent of its velocity in the z direction. The stop is located a distance l_A along the B field which is where the maximum deflection occurs. The configuration of the low-J-state selector is shown in Fig. 3.

The velocity distribution shows up in two ways. First, the molecules acquire a spread of transverse velocities v_x, depending upon their values of v_z. Since the detector is sensitive only to position and not to velocity, the transverse velocity has no effect. Second, the maximum transverse deflection depends on v_z. The maximum distance any molecule is deflected is inversely proportional to the square of its velocity, v_z. The stop is located at the point of maximum deflection for all the molecules, a distance l_A along the B field.

With a voltage of 5 kV on the deflectors, 80% of the molecules in the $J=0$ state are deflected farther than the 0.020 in. the stop is inserted. Unfortunately $J=1$, $m=0$ molecules are deflected the wrong direction and do not pass through the state selector so that the over-all transmission for molecules available for resonant deflection is 40%. However, since only 0.5% of the straight-through beam is transmitted by the state selector, the ratio of the signal-to-background beam is improved by eighty.

RESULTS

Most of our results were obtained using a flop-out method; that is, we monitored the full beam and detected a decrease in beam current when the molecules were deflected by the microwave field. If we used the electrostatic low-J-state selector, we found the flop-out scheme to be the most sensitive method for detecting the small deflections of the beam. Flop-in measurements were done by replacing the collimator hole by a 0.050-cm-diam wire stop to block the straight-through beam. The detector was on axis and detected an increase in the beam current.

Microwave-frequency measurements were made with a precision of ± 1 MHz. In taking the data, the microwave power reflected back from the cavity was minimized at each frequency. Checking the power coupled out of the cavity with a weakly coupled pickup probe showed a variation of less than 20% from 10.970 GHz to 11.080 GHz.

Figures 4 and 5 show data which represent the average of several runs. Typically the data collected had signal-to-noise ratios between 1 and 3 using averaging times of about 2 min. The most obvious feature of our results is that the deflection signal is resonant at the frequency predicted by microwave spectroscopy. However, the magnitude of the observed signals is distinctly smaller than what we had expected. Measured at the electrometer input a typical set of values for the undeflected beam and deflected beam was 3×10^{-8} and 3×10^{-13} A. The ratio of the two is 10^{-5} which is a factor of 3 lower than had been expected. This reduction in signal is most likely due to a smaller than anticipated deflection. When higher-order TE_{11n} modes were used, the detected signal was reduced considerably, as expected.

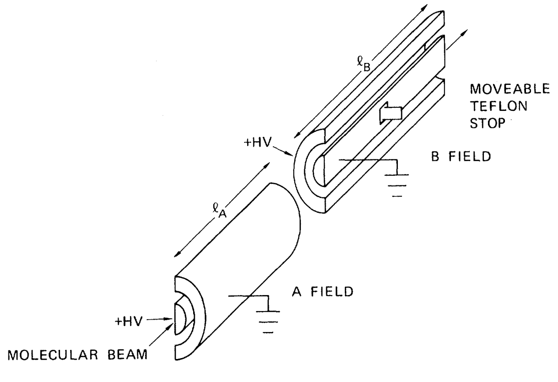

FIG. 3. Electrostatic low-J-state selector. $l_A=8.6$ cm. $l_B=21.0$ cm.

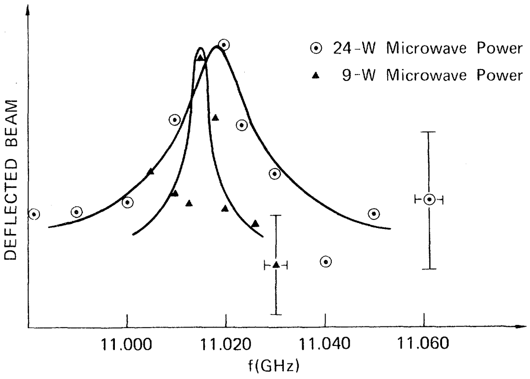

FIG. 4. Observed resonances for CsF $v=0$, $J=0\to 1$ resonance at microwave powers of 9 W (△) and 24 W (○). The molecular beam is aligned along the center of the waveguide.

The linewidth observations show a qualitative agreement with the theory in that both raising the microwave power and moving the cavity so the beam passes through a region of high field broaden the line as expected. Quantitatively there are serious discrepancies between the observed and predicted linewidths. Using Eq. (30) we can calculate the expected FWHM for the resonances of Fig. 4, which were taken with the molecular beam nominally on the cavity center line, we shall assume a misalignment of half the beam width, which is about as close as we can hope to align

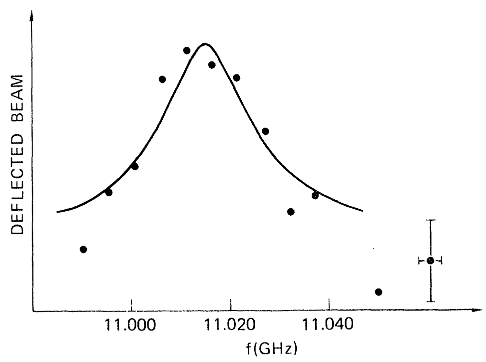

FIG. 5. Resonance observed with the waveguide translated so that the beam passes 0.043 cm from the center line of the waveguide. The microwave power is 9 W.

the beam and cavity. The observed and calculated linewidths are presented in Table II. These observed linewidths are about a factor of five narrower than were predicted. This result is discussed below.

In the process of setting up this experiment we made measurements of the attenuation of the beam by background gas scattering. If we write the intensity of a collimated beam as

$$I = I_0 e^{-kp} \tag{36}$$

where I_0 is the zero pressure beam intensity, p is the pressure in Torr, and k is an attenuation constant, we can conveniently express the scattering in terms of k. Our measured value of k is $2.4\times 10^{+6}$ Torr^{-1}. As the beam path is 300 cm long, this implies a total scattering cross section of 2000 Å^2. Since the scattering occurs over the whole length of the apparatus, there is no unique scattering angle, so we did not attempt to make any measurements of the angular dependence of the scattering. However, our observations indicate that most of the scattering was into angles less than a milliradian.

DISCUSSION

Although resonance deflection of molecular beams has been demonstrated, some questions remain in relating the simple two-level theory to the observations. Some remarks are in order about means of improving the observed signal and about research areas where this technique may prove useful.

There appear to be two possible explanations for the observation of smaller linewidths and signals than we had expected. Either the two-level theory overstates the effective forces for this effect or the effective microwave deflecting field in the resonant cavity is a factor of 4 to 5 less than our measurements indicate. Neither of these is particularly attractive.

According to the calculation, 9 W of microwave power should be adequate to deflect all but the fastest $J=0$, $m_j=0$, and $J=1$, $m_j=0$ molecules out of the beam, in which case raising the microwave power to 24 W should produce only a marginal increase in signal. In fact, raising the power to 24 W roughly doubled the signal suggesting that we may be losing some of the microwave power. A power reduction of roughly 13 dB would be necessary to reduce the linewidth and deflecting force by a factor of 5. However, the total power loss due to line losses and impedance mismatches is measured to be less than 2 dB. There is also the remote possibility that in spite of the indications to the contrary we are in fact driving the TM_{111} mode rather than the TE_{111} mode. Thus, it seems unlikely that a lower microwave power is the sole source of the discrepancy.

We considered several other effects which could conceivably lead to the observed results. These were dephasing collisions with the background gas, dissociation[9] of the CsF molecular beam or the formation of dimers,[10] or coupling to other states of CsF induced by the high rf fields. None of these appear to contribute substantially.

The low signal-to-noise ratio of the deflected molecules constituted one of the major experimental difficulties. This could be improved significantly if a nozzle beam for alkali halides were used. The rotational temperature could be reduced by about 10^2 and the flux increased. This would lead to an increase in the deflected beam of 10^3 or more, giving a vastly improved signal-to-noise ratio. With such a beam source, molecular beams with selected vibrational and rotational levels could be produced by the resonance deflection technique.

In the Introduction we mentioned that any atoms or molecules with allowed electric or magnetic dipole transitions could be deflected. Since the effect does not depend on the frequency of the transition, there is a wide range of possibilities. Let us consider two examples. First, one could deflect an atomic beam using an allowed electronic transition. This of course has already been done in the radiation-pressure experiments.[11] However, radiation-pressure experiments are limited to absorbing one photon per lifetime of the excited state, but dynamic deflection is a coherent process of stimulated absorption and emis-

TABLE II. Comparison of calculated and observed linewidths.

Figure	Beam path (cm)	Power (W)	FWHM (observed) (MHz)	FWHM (calculated) (MHz)
4	$x_0 = 0.015$	9	8	52
4	$x_0 = 0.015$	24	21	84
5	$x_0 = 0.043$	9	28	150

sion and consequently is not limited by the lifetime of the excited states. One of the obvious applications of this is to isotope separation.

A second possibility is that of deflecting vibrational states of polar molecules using the infrared vibrational transitions. This is particularly interesting since at present there is no way of procucing vibrationally state selected molecular beams. The ability to do so would be invaluable in many chemical reation studies. The results of many of these investigations imply a strong vibrational state dependence, which would be investigated in detail using a vibrational state selector. Deflecting vibrational states could also be used for isotope separation.

While both the visible and infrared spectral regions are intriguing, it is worth mentioning that the technical problems would be formidable. For example, it is not obvious how to arrange a laser beam to produce a continuous field gradient along a sufficient length of the molecular-beam path to produce an observable deflection. However, if the technical problems can be overcome, resonant deflection should prove to be a very useful technique in these spectral regions.

CONCLUSION

The physical process of resonance deflection with inhomogeneous electric fields has been demonstrated, but the actual results of the experiment did not agree exactly with a two-level analysis. A more thorough understanding of the interaction between the molecule and the strong oscillating field may explain the observations. If the signal strength could be improved significantly by use of nozzle beams to improve the partition function or by use of stronger sources, useful beams of state-selected molecules could be achieved. Our modest success with the rather difficult rotational selection process encourages us to believe that resonance deflection may become a useful experimental technique.

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