Observation of two-zone Ramsey fringes using laser Stark spectroscopy of a molecular beam

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(Received 16 January 1986)

Well-resolved Ramsey fringes have been demonstrated using laser Stark spectroscopy of CH_3F in a molecular beam with two interaction regions. Fringe separations as low as 155 kHz were obtained. The interaction regions were produced by masks containing slits placed in the Gaussian-profile beam of a cw CO_2 laser. Population inversion resulting from a frequency sweep associated with the curvature of the slit-diffracted wave fronts has limited the observations at high laser-power values. An imaging system has been shown to eliminate this effect. Experiments with three and five laser-molecular beam crossings were also conducted to demonstrate the feasibility of using multiple interaction regions as a tool for high-resolution spectroscopy. The results have been modeled using the optical Bloch equation for a two-level system. Good agreement with the experiment has been obtained.

INTRODUCTION

For many years, the two-interaction-zone Ramsey method[1] of exciting an atomic beam has been used to define the standards for frequency and time. A microwave transition in cesium has been used to obtain a fractional frequency stability of $\simeq 1\times 10^{-14}$.[2] Recently, several theoretical and experimental papers[3-11] have suggested that improved frequency standards may be obtained by using the Ramsey method on infrared and visible transitions. Because of the spectral line narrowing that can be obtained with the Ramsey technique, there is also interest in using the method as a high-resolution spectroscopic tool. The object of this paper is to indicate how Ramsey fringes may be obtained in the infrared spectral region, using laser Stark spectroscopy.

Fluorescence detection has been used for optical Ramsey fringe experiments. However, because of the long infrared excited-state lifetimes, the efficiency of the fluorescence detection method in the infrared region is quite low. The cryogenic bolometric method of detecting the energy absorbed by a molecular beam through infrared transitions[12,13] provides a very sensitive alternative and has recently been used to observe both Rabi oscillations and Ramsey fringes in SF_6 (Refs. 11 and 14) and Rabi oscillations in fluoromethane (CH_3F).[15] The use of the laser Stark spectroscopic technique in conjunction with the bolometric method allows many molecules with permanent electric dipole moments to be investigated using the Ramsey technique. CH_3F is one of the most-studied molecules in the infrared region[16-19] and its low J,K states which are abundant in a supersonic molecular beam,[20] are easily Stark tuned into resonance with CO_2 laser lines. Thus it was natural to extend our coherent excitation studies[15] to Ramsey fringes using this molecule.

EXPERIMENT

The apparatus used in this experiment is the same as that already described.[15] A linewidth of 215 kHz [full width at half maximum (FWHM)] was obtained for the $Q(1,1)$-$1\leftarrow 0$ transition of the ν_3 band of CH_3F where the limitation was essentially transit time broadening given by the laser beam spot size ($w_0=3.29$ mm) and the mean molecular speed ($v=1590$ m/s). The line shape of the power spectrum of the Gaussian laser beam at low power as experienced by a molecule orthogonally crossing it may be shown to have a width of ≈ 180 kHz FWHM. This is found by calculating the Fourier transform of the laser electric field as a function of time as experienced by the molecule passing through (Appendix). This homogeneous linewidth was increased by placing slits in the laser beam, while the geometrically limited residual Doppler broadening was reduced to $\simeq 50$ or 150 kHz by reducing the width of the molecular beam detector aperture to 100 or 300 μm. The reduced interaction time resulted in a broader homogeneous line shape for the signal from a single field region. The Ramsey technique employing a second crossing was then used to demonstrate the recovery of the lost resolution. To accomplish this, one of a set of masks which had two 1-mm-wide slits separated nominally by 3, 5, and 10 mm was placed in the center of the Gaussian laser beam about 15 cm from the molecular beam. The slits were oriented normal to the molecular beam.

In principle it is possible to use a molecular beam of greater divergence and thereby obtain a larger signal if one uses at least three laser-beam crossings. The intermediate counterpropagating beams "refocus" the dephasing molecular dipoles in the manner of spin-echo experiments.[3,4,6,11] The degree of collimation in our experiment was enough to obtain good fringe contrast with two crossings only. Crossing orthogonality ensured that the mean Doppler shift of the line was less than $\approx 10\%$ of linewidth. This was accomplished by the superposition of signals produced by incident and retroreflected laser beams. This ensured that we were tuned to line center of the Stark-shifted transition. The retroreflected laser beam was subsequently eliminated. We also performed three- and five-crossing experiments in order to study how the fringe resolution varies with the number of crossings.

RESULTS

To obtain a spectrum, the Stark field was scanned while monitoring the bolometer signal. The results for the $Q(1,1)$-$1 \leftarrow 0$ transition and two laser crossings are shown in Fig. 1. The frequency scale was determined by applying a radiofrequency field in addition to the Stark field. The resulting satellite spectral peaks are separated from their parent peaks by the applied frequency.[15] Ramsey fringes with spacings of 582, 351, and 155 kHz were obtained for the 3-, 5-, and 10-mm nominal slit separations, respectively. The actual line shape at low power, as for the single-beam case, may be found by calculating the Fourier transform for the laser electric field as experienced by the molecules. The result is a fringe spacing of $\Delta f = 1/T$ (Appendix), where T is the transit time between crossings. In the experiments of others already referred to[3,4,6,11] where one or more intermediate counterpropagating laser crossings were used, the fringe spacing may be shown to be $1/2T$ where T is the time between *adjacent crossings.*

Both the Stark field inhomogeneity and molecular velocity spread are effective in dephasing the molecular dipoles in the time interval between crossings. Therefore, the fringe contrast decreases with increasing separation going from 0.78 contrast in the 3-mm case to 0.33 in the 10-mm case.

A study of the Ramsey signal as a function of laser power was undertaken using the slits separated by 5 mm. The result, shown in the progression of signals in Fig. 2, indicates that with increasing laser power, the central Ramsey fringe oscillates from zero through a maximum (near inversion) and back to zero again. Additional laser power over a certain limit does not cause the central fringe to oscillate further as it might be expected. This phenomenon may be understood in terms of the power dependence of the signal produced using a single 1-mm-wide slit with the Stark field tuned to line center. Figure 3 shows the result for a slit offset by ≈ 2.5 mm from the center of the Gaussian laser-beam profile. The offset corresponds to the position of one of a pair of slits separated by ≈ 5 mm. A centered slit was observed to produce similar effects on the diffraction pattern and the molecular excitation. It is known[14,15,21] that Doppler frequency sweeps in the molecular rest frame resulting from wave-front curvature lead to an interaction which, assuming sufficient field strength, merely inverts a two-level system. Diffraction from a slit aperture in the laser beam causes wave-front curvature. The calculated diffraction pattern

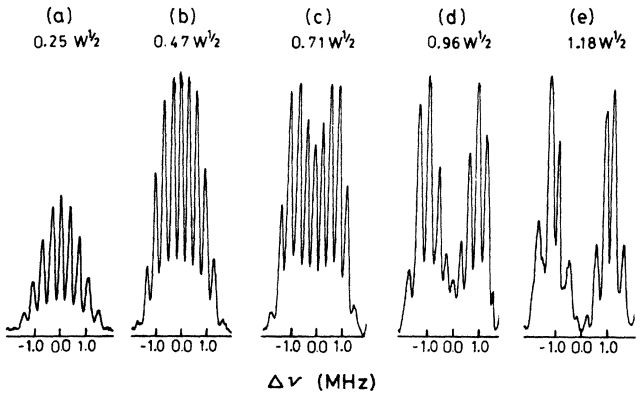

FIG. 2. Ramsey fringe progression for two 1-mm-wide slits separated by $\simeq 5$ mm, 15 cm from the molecular beam. Square root laser power used for (a)—(e) is given in each case. Laser field greater than 1.18 $W^{1/2}$ did not result in the central Ramsey fringe returning to inversion as expected.

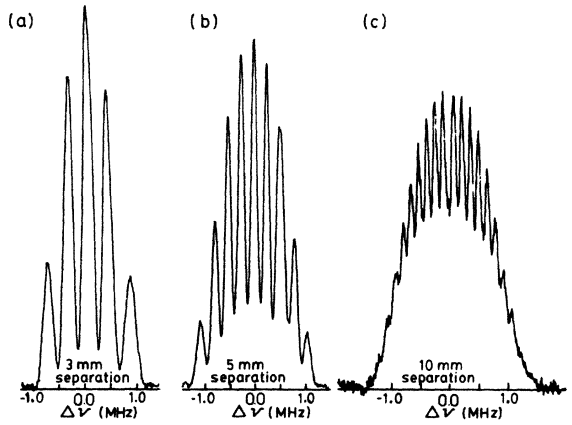

FIG. 1. Ramsey fringes produced using two slits with different nominal slit separations placed 15 cm from the molecular beam. (a) 1-mm-wide slits separated by $\simeq 3$ mm. Laser power used was 0.028 W. Measured fringe separation is 582 kHz. (b) 1-mm-wide slits separated by $\simeq 5$ mm. Laser power used was 0.124 W. Measured fringe separation is 351 kHz. (c) 1-mm-wide slits separated by $\simeq 10$ mm. Laser power used was 0.926 W. Measured fringe separation is 155 kHz.

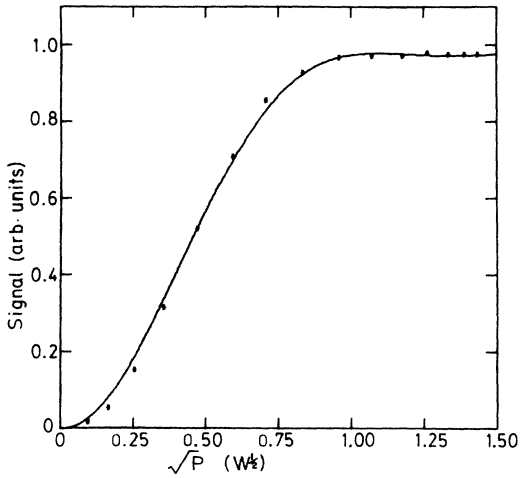

FIG. 3. Single slit, 1 mm wide, offset $\simeq 2.5$ mm from the center of the Gaussian laser profile, 15 cm from the molecular beam was used to produce the data (***). Superimposed on the experimental points are the results of a simulation (——) based on the Bloch equation and diffraction calculations described in the text. P is the laser power. Data and simulation are scaled to have the same asymptote. No Rabi oscillations are observed; inversion of the system is seen instead.

at the molecular beam is shown in Fig. 4. Because of these effects, the system achieves inversion for sufficiently high laser power, but further increase of power causes no change at line center. A second such laser crossing at line center (corresponding to the central Ramsey fringe) brings the molecules back to the ground state again. This results in zero signal for the central Ramsey fringe. Increasing the laser power further leaves the signal essentially zero. Ideally, one would wish to have the molecular beam cross the laser beam in a region where the wave fronts are plane. However, since the slits cannot be placed between the Stark plates close to the molecular beam, the effects of diffraction are unavoidable. Thus, an attempt was made to reconstruct at the molecular beam the wave fronts which existed at the slits. This involved two lenses in the unit power telescope configuration shown in Fig. 5(a). The first test of this system involved imaging a single slit, offset by 2.5 mm from the center of the Gaussian laser-beam profile, to recover Rabi oscillations. Figure 5(b) shows that the oscillations were obtained using this arrangement.

It has been suggested[22–24] that coherent excitation from a sequence of equidistant equal-action interaction regions will produce fringes which are more distinctly separated as the number of regions is increased. The resulting spectrum will resemble the diffraction pattern from a grating

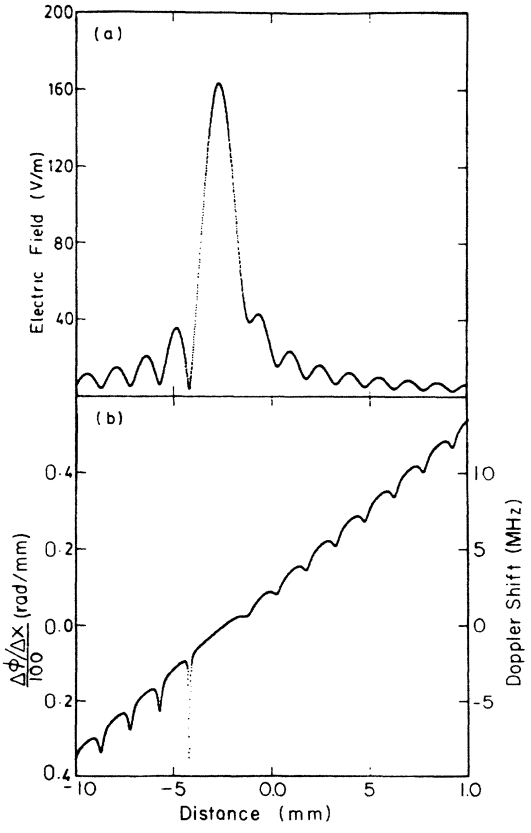

FIG. 4. Electric field amplitude (a) and associated phase angle increments (b) for the laser beam at the molecular beam after diffraction by a single offset slit. A Doppler frequency scale is provided for $v = 1590$ m/s.

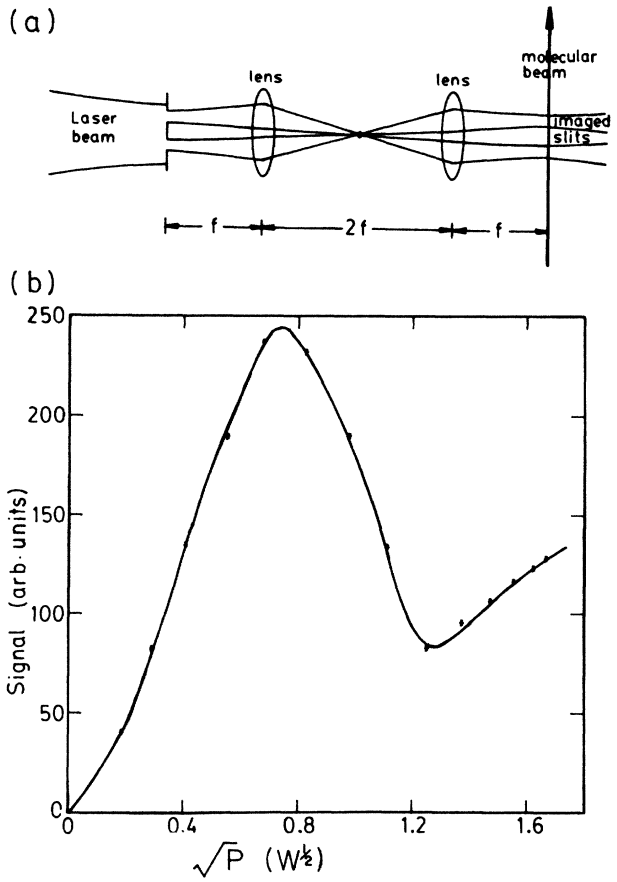

FIG. 5. (a) Imaging system used to image the slits onto the molecular beam is shown. Focal length of the ZnSe lenses used was 25 cm. (b) Single slit, 1 mm wide, offset $\simeq 2.5$ mm from the center of the Gaussian laser profile was used to produce this signal. P is the laser power. Imaging of the slit recovers the Rabi oscillations.

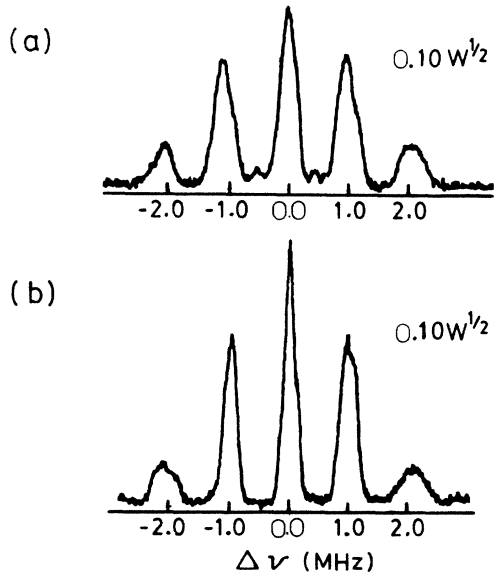

FIG. 6. Multicrossing signals are shown. Slits were placed 15 cm from the molecular beam with slit widths chosen as described in the text. (a) Three-slit signal giving 307 kHz FWHM for the central fringe. (b) Five-slit signal giving 192 kHz FWHM for the central fringe.

the apertures of which correspond with the interaction regions. This result is potentially useful for high-resolution spectroscopy because the multiple crossings will reduce the number of fringes observed, leaving gaps between the "diffraction orders" where closely spaced frequencies may be resolved. The technique and the expected spectrum are described in Ref. 24. Masks containing three and five slits of varying widths with 1.5-mm separation between adjacent slits were constructed such that over the Gaussian laser profile, each slit would transmit the laser power necessary to provide the same action integral. Spectra resulting from the use of these masks are shown in Fig. 6. The FWHM for the three-slit spectrum is 307 kHz, while the five-slit spectrum has a FWHM of 192 kHz. These spectra clearly indicate the line narrowing and the missing diffraction orders produced by this technique. They were taken at a laser-power level well below that necessary to produce inversion.

MODELING

Simulation of the experiment is based on the optical Bloch equation with relaxation neglected as in a previous paper.[15] This equation describes the motion of a pseudospin vector $\mathbf{R}$ in response to a driving vector $\boldsymbol{\beta}$ such that

$$\dot{\mathbf{R}}=\mathbf{R}\times\boldsymbol{\beta}\ . \tag{1}$$

The vector $\boldsymbol{\beta}$ is defined, using the usual Cartesian unit vectors, as

$$\boldsymbol{\beta}=\frac{\mu\mathscr{E}}{\hbar}\hat{\mathbf{x}}-(\omega-\omega_l-\omega_d)\hat{\mathbf{z}}\ , \tag{2}$$

where μ is the transition dipole moment, $\mathscr{E}$ is the laser field amplitude, ω is the transition frequency, ω_l is the laser frequency, and ω_d is the Doppler shift due to wave-front curvature, all in rad/s. The difference in the present application is that diffraction of the laser beam by the slits is considered. The well-known Fresnel-Kirchhoff diffraction integral is solved numerically to give the laser electric field amplitudes (real and imaginary parts) at discrete points along the molecular beam. The phase differences between adjacent points for the diffracted light is found by the trigonometric relation for the difference between the $\tan^{-1}(\mathscr{E}_{\rm im}/\mathscr{E}_{\rm re})$ for adjacent discrete points. These phase differences are converted into a time-dependent frequency which is the Doppler shift, given by

$$\omega_d=\Delta\phi/\Delta t=v\Delta\phi/\Delta x\ . \tag{3}$$

This is the frequency used in (2) to obtain $\boldsymbol{\beta}$. The diffraction pattern at a distance of 15 cm from a single slit of 1 mm width offset by 2.5 mm in the laser beam is shown in Fig. 4. This figure shows the modulus of the laser electric field, Fig. 4(a), and the rate of change of phase with position, as well as the corresponding Doppler shift for a molecule crossing orthogonally at 1590 m/s. The mean wave-front curvature due to diffraction is ≈ 16 cm. Equation (1) is solved numerically using a unitary matrix transformation[25] for each discrete step in the passage through the field.

The energy deposited per molecule in the beam by the laser is $\frac{1}{2}\hbar\omega(1+R_z)$, where R_z is the component of $\mathbf{R}$ along the z axis in the space in which $\mathbf{R}$ moves. The detector signal is therefore proportional to $1+R_z$. This quantity has been calculated for several laser-power values for single-slit diffraction (a 1-mm slit offset by 2.5 mm from the center of the Gaussian profile) with the slit 15 cm from the molecular beam. The calculated curve is compared with the experimental data in Fig. 3 where the data have been scaled so that the experimental asymptote and the simulated one are the same. The agreement between experiment and simulation indicates that diffraction is the cause of the inversion process.

A comparison between a simulated spectrum and experimental data for two 1-mm-wide slits separated by 5 mm and placed 15 cm from the molecular beam is shown in Fig. 7(a). In order to produce the maximum on-resonance signal, a laser power of 53 mW was used in the experiment. However, it was found that inversion was reached in the simulation with just 28 mW. The data have been scaled so that the intensities of the central fringes are the same. Aside from the discrepancies in the laser power necessary to achieve the maximum signal, there is also a discrepancy in the contrast which, in the simulation, is far too good. Both of these discrepancies have their origin in the fact that no damping mechanisms were included in the calculations. It is known[1,4,7] that the velocity distribution and the residual Doppler effect will reduce fringe contrast. As well, it has been shown[15] that averaging over the vertical profile of the molecular beam also affects the signal. Because the computer time required to simulate a Ramsey signal was quite large (computation was performed on a DEC VAX/780) and introducing damping mechanisms increased the calculation time drastically, the effects were phenomenologically represented by convolut-

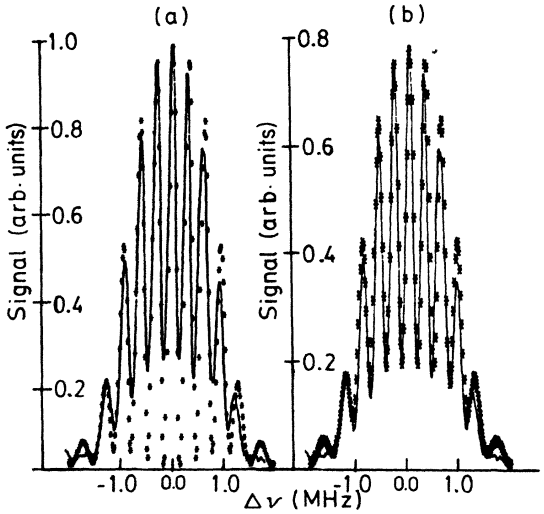

FIG. 7. (a) Simulated Ramsey spectrum for a laser power of 28 mW is shown (***) for two 1-mm-wide slits with a 5 mm separation, 15 cm from the molecular beam. Superimposed on the simulation are data (——) for a laser power of 53 mW, scaled so that the intensity of the central fringes are the same. (b) Simulation of (a) is convoluted with a Lorentzian line shape with a FWHM of 52 kHz to reproduce the contrast of the experimental data. Data (——) are scaled in intensity and superimposed on the simulation (***).

ing a Lorentzian-line profile with the undamped simulation. The width of the Lorentzian line was adjusted until the experimental fringe contrast was reproduced. The result of a calculation of this type is shown in Fig. 7(b), where the experimental data of Fig. 7(a) have again been superimposed on the simulation so that the intensities of the central fringes are the same. A Lorentzian width (FWHM) of 52 kHz was found to reproduce the contrast. The intensities of the individual peaks and their positions also agree quite well.

The three- and five-slit data were also modeled without damping although the convolution with a Lorentzian line shape was not performed. Figure 8 shows how the simulations compare with the experimental data. Once again the data have been scaled and superimposed on the simulations. In Fig. 8(a), laser power of 24 mW was used in the experiment and 33 mW in the simulation. Laser power of 9 mW was used for the experimental data of Fig. 8(b) with 10.4 mW in the simulation. In both cases the peak positions and linewidths are in fair agreement but as in the case of the two-slit simulations, there are discrepancies in the peak intensities and the laser power required because no damping mechanisms were included. However, both the experiment and the simulations indicate the possible usefulness of the technique for resolving closely spaced transitions.

SUMMARY

The results given in this paper show that by using the laser Stark spectroscopic technique in conjunction with cryogenic bolometric detection of a molecular beam, Ramsey fringes with good contrast may be obtained using just two interaction regions. Crossing separation of up to 10 mm obtained by using two slits in the laser beam have been used to achieve fringe resolution of $\simeq$155 kHz which is better than that obtained from using the entire Gaussian beam. Multicrossing experiments have shown the spectral line narrowing and the presence of gaps between peaks that result. Both the Ramsey experiment and the multicrossing results indicate possible application of this technique in high-resolution infrared spectroscopy. At present the experimental linewidth is limited by a contribution of $\simeq$50 kHz Doppler width due to molecular beam divergence, 40 kHz or less due to laser instability[26] and $\simeq$20 kHz due to Stark field inhomogeneity.[15]

The optical Bloch equation and calculations of the diffraction caused by the slits placed in the laser beam have been used to model the experiment. The agreement is good. Simulated fringe positions, separations, and line shapes are in agreement with experiment but there is some discrepancy in the laser power required to produce the patterns. This discrepancy is probably due to the absence of any damping mechanisms in the model. The experimental observation of rapid passage effects caused by diffraction has also been confirmed by modeling. The success of the theory used to model the experiment shows that the interaction of the molecules with the laser beam under the present experimental conditions is well understood.

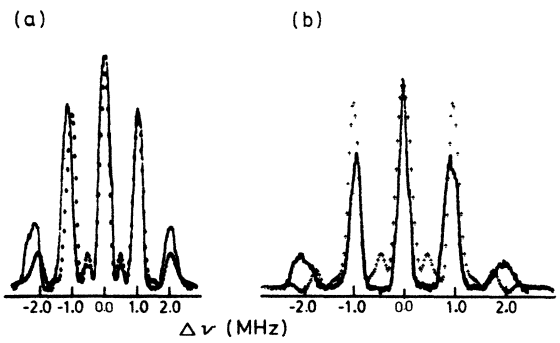

FIG. 8. Comparison of three- and five-slit multicrossing simulations (* * *) with experimental data (——). The slits were located 15 cm from the molecular beam. Simulations and the data are scaled so the intensity of the central peaks are the same. (a) Laser power of 33 mW was used in the calculation while 24 mW was used in the three-slit experiment. (b) Laser power of 10 mW was used in the simulation while 9 mW was used in the five-slit experiment.

ACKNOWLEDGMENTS

The authors acknowledge funding for this project provided by the Natural Sciences and Engineering Research Council of Canada. One of us (A.G.A.) would also like to thank the Natural Sciences and Engineering Research Council (NSERC) for additional support. The members of the Centre for Molecular Beams and Laser Chemistry participate in the Guelph-Waterloo Program for Graduate Work in Physics[2] (GWP) and the Guelph-Waterloo Centre for Graduate Work in Chemistry[2] (GWC).

APPENDIX

The power spectrum for a Gaussian laser beam as experienced by a molecule crossing it orthogonally at its waist is given by

$$G(\omega)=|g(\omega)|^2 , \tag{A1}$$

where

$$g(\omega)\propto\int_{-\infty}^{\infty}\exp[-(x^2+y^2)/w_0^2+i(kz-\omega_l t)]\times\exp(i\omega t)dt . \tag{A2}$$

It may be noted that we take the laser-beam direction as z, and if we take the molecular beam direction as x, we may substitute $x=vt$. The result is given by

$$G(\omega)\propto\exp[-(\omega-\omega_l)^2 w_0^2/2v^2] \tag{A3}$$

which has a width about the laser frequency ω_l of $(8ln2)^{1/2}v/w_0$ in rad s^{-1} (FWHM).

The above calculation may be made for two laser crossings separated by a time T. If we assume that the two crossings are similar and represented by $f(t)$ and $f(t+T)$, we do not need to know the Fourier transform of the individual crossing; we may represent it as $g_1(\omega)$. We have

$$g(\omega) \propto \int_{-\infty}^{\infty} [f(t)+f(t+T)]\exp(i\omega t)dt$$
$$\propto \int_{-\infty}^{\infty} [\phi(\tau)\exp(i\omega t)dt + f(t')\exp(i\omega t')\exp(-i\omega T)dt'] , \quad \text{(A4)}$$

where $t'=t+T$. Thus,

$$g(\omega) \propto g_1(\omega)+g_1(\omega)\exp(-i\omega T) , \quad \text{(A5)}$$

which gives

$$G(\omega) \propto |g(\omega)|^2 \propto |g_1(\omega)|^2 \cos^2(\omega T/2) , \quad \text{(A6)}$$

which has maxima at $\omega T/2=N\pi$ where $N=0,\pm1,\pm2$, etc. This gives the separation of the maxima of $\Delta\omega=2\pi/T$ or $\Delta f=1/T$.

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