SUPERSONIC BEAM SPECTROSCOPY OF LOW J TRANSITIONS OF THE ν_3 BAND OF SF_6: RABI OSCILLATIONS AND ADIABATIC RAPID PASSAGE WITH A CW LASER ☆

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The P(3) and P(4) manifolds of the ν_3 band of SF_6 have been observed in a supersonic beam with a bolometric detection. The influence of the laser beam divergence on the excitation efficiency has been studied. Rabi oscillations are observed when the wavefront is flat in the interaction region whereas only adiabatic rapid passage occurs when the molecules see a curved wavefront.

We have applied the cryogenic bolometer method of detection of vibrationally excited molecules in supersonic beams [1] to the spectroscopy of SF_6 in the 10 μm spectral region. The P(3) and P(4) manifolds of the ν_3 band which are in good coincidence respectively with the R(10) line of the N_2O laser and the P(16) line of the CO_2 laser [2] appeared to us as the most suitable choice for such an experiment, given the low rotational temperature in a supersonic beam. Fig. 1 is the schematic diagram of the experiment and fig. 2 shows the bolometer signal corresponding to the A_2 and F_2 components of the P(3) manifold when a helium beam seeded with 7% of SF_6 is illuminated by the N_2O laser. The comparison with a room temperature saturation spectrum confirms the assignment of these low J transitions. Recently we have obtained similar results with a waveguide CO_2 laser for the A_1, F_1 and E components of the P(4) manifold respectively at 228.154, 245.080 and 257.116 MHz from $Q(38)E^0$ [2].

We have used the P(3) A_2 line to perform a detailed quantitative study of the influence of the interaction geometry on the linewidth and excitation efficiency. The laser beam divergence has been varied by tuning the position of the second lens of a telescope. Fig. 3 shows the bolometer signal as a function of this position for three laser intensities. For these experiments the laser frequency was locked to the center of the line using saturation spectroscopy in an auxiliary cell. Because of the Doppler detuning only a fraction of the molecular beam velocity distribution along the optical axis interacts with the light. The signal is proportional to the width of the hole burnt in this distribution. This width is dominated by transit effects (including amplitude and phase modulation in a curved gaussian beam). The signal is thus minimum for minimum transit broadening, that is when the telescope focusing is perfectly adjusted and it reaches a maximum when the molecular beam and laser beam divergences are matched.

To account for the experimental data of fig. 3 we

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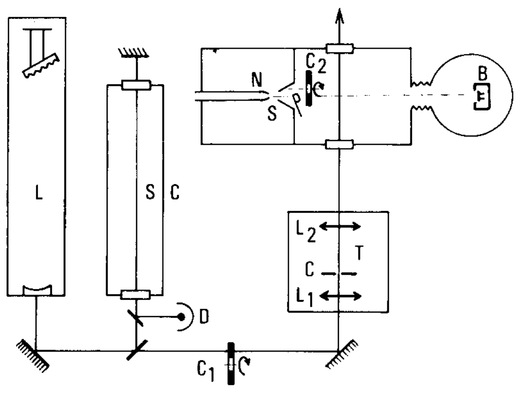

Fig. 1. Schematic diagram of the experiment. The molecular beam machine consists of two separate chambers each with its own diffusion pump. Gases are expanded through the 90 μm diameter nozzle (N) followed by a 0.5 mm diameter skimmer (S) into the second chamber equipped with a liquid-N_2 trap (Combined pumping speed of 1200 l/s). The operating pressures in the two chambers are respectively a few 10^{-4} Torr and a few 10^{-6} Torr. A beam flag and a chopper (C_2) are used for full beam intensity measurements. The bolometer (B) located 50 cm from the nozzle is mounted in contact with the cold surface of a liquid He Dewar. Its responsivity is 7×10^{-3} VW^{-1} and the RMS noise at 4.2K is 100 nV $Hz^{-1/2}$. The cw N_2O laser (L) is a conventional low pressure laser. Saturated absorption in an auxilliary cell (SC) is used to control the frequency tuning or to lock this frequency to the center of any observed resonance. The laser beam chopped by C_1 at a frequency around 30 Hz is expanded with a telescope (T) and spatially filtered with a pinhole (C) before its interaction with the molecular beam. The focal length of the two lenses of the telescope (L_1 and L_2) are respectively F_1 = 10 cm and F_2 = 33 cm.

found it necessary to use a non-perturbative calculation of the interaction between the molecules and the gaussian laser beam based on the equations of reference [4]. This theory shows that, in the strong field regime, the transition probability averaged over the

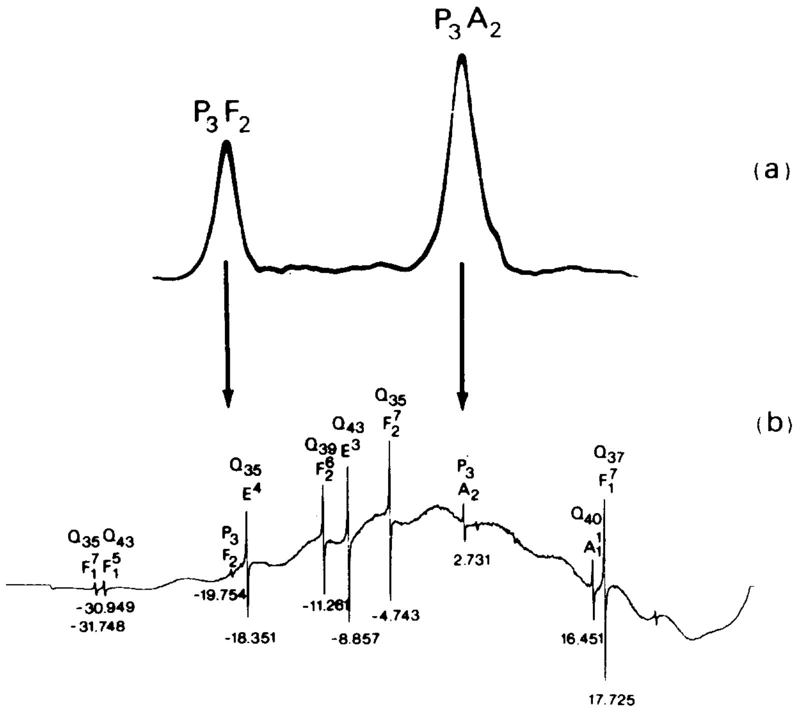

Fig. 2. SF_6 spectra obtained with the R(10) N_2O line; (a) supersonic beam (7% SF_6 in helium, source stagnation pressure 1.8 bar), (b) saturation spectrum in a room temperature cell. Frequencies are in MHz from the N_2O line center [2]. The absolute frequency of Q(37) F_1^7 is 28414593720 kHz [3].

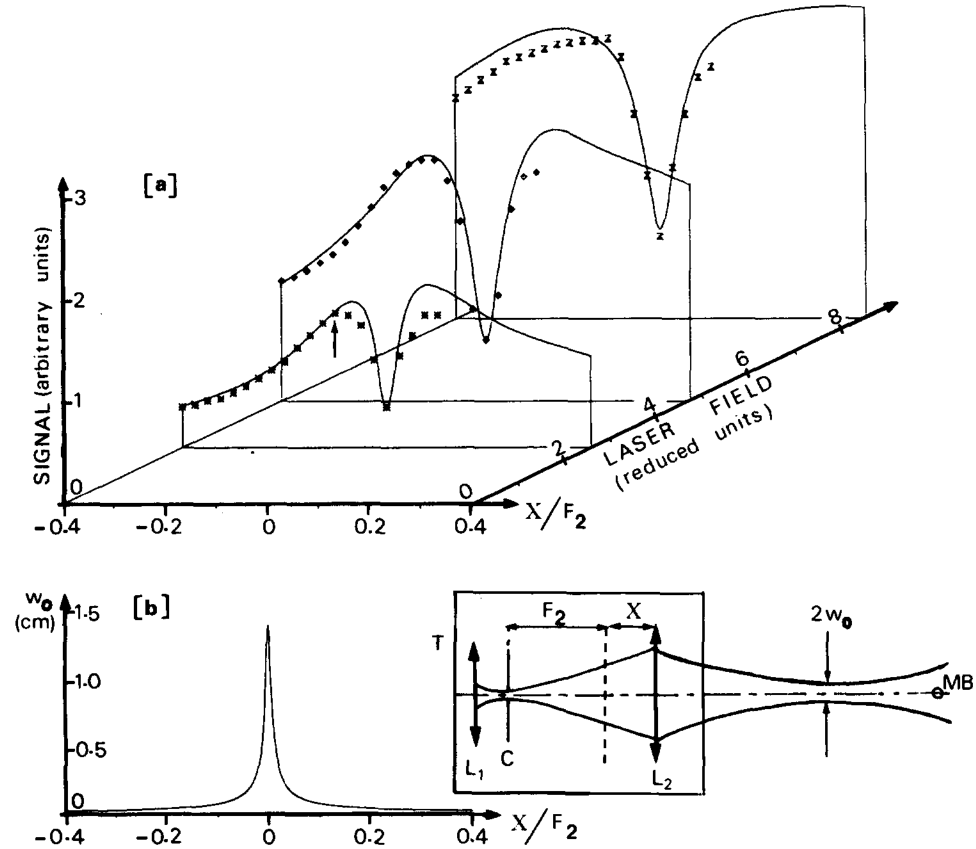

Fig. 3. (a) Excitation efficiency as a function of the laser focusing (position of the second lens of the telescope in units of focal length F_2 = 33 cm with respect to focal position) for laser powers respectively equal to 1.8, 6. and 19.4 mW. The only adjustable parameter of the set of calculated curves (solid lines) has been the signal amplitude for the point indicated by the arrow.
(b) Gaussian laser beam 1/e radius w_0 as a function of the laser focusing.

velocity distribution undergoes Rabi oscillations only when the laser field has minimum curvature in the interaction region for reasons to be discussed below.

To demonstrate these oscillations with a good enough signal-to-noise ratio we had to increase the number of interacting molecules without introducing curvature that is when the telescope is afocal and for this we modified the telescope to reduce the beam waist radius to 3 mm when located on the molecular beam (in fig. 1, the focal length F_2 was changed to 6.7 cm).

In fig. 4a we give experimental evidence that such oscillations do occur when the c.w. laser field strength is varied. The oscillations disappear for highest fields owing to the transverse field distribution and to the existence of three different Clebsch-Gordan coefficients in the dipole moment and are well represented by the theory.

When the laser beam waist is slightly offset from the molecular beam, i.e. when the molecules see curved wave-fronts, the Rabi oscillations disappear as illustrated in fig. 4b. The reason is that, as they travel across a curved gaussian beam, molecules see a linear sweep of the instantaneous frequency which induces a rapid adiabatic passage [5] if the field is strong enough. This rapid adiabatic passage inverts the medium without possibility for a complete Rabi precession as illustrated on the pseudo-spin [6] trajectories given in fig. 5. This doubles the available signal from the case where populations would be simply equalized. A detailed description of the coherent interaction between a gaussian laser beam and a supersonic molecular beam will be presented in a further paper.

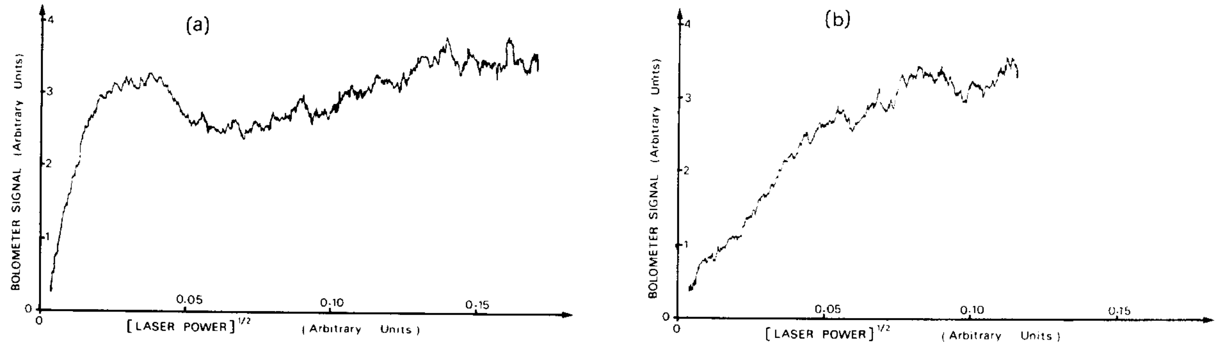

Fig. 4. (a) Rabi oscillation of the holometer signal as a function of the laser field. The laser is locked to the P(3) A_2 line center and the laser beam waist ($w_0 \approx 3$ mm) is accurately set on the molecular beam. The horizontal scale is roughly in $(\mathrm{Watt})^{1/2}$. (b) Same as (a) but the laser beam waist is slightly offset from the molecular beam (the second lens of the telescope was moved by 0.5 mm with respect to (a).

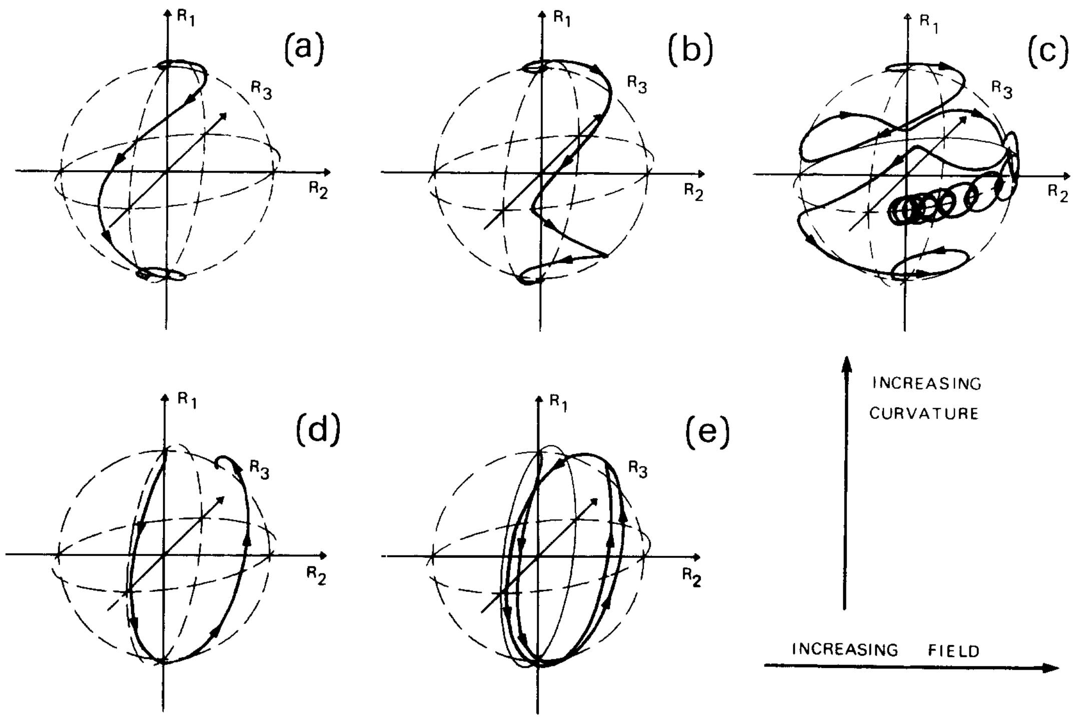

Fig. 5. Pseudo-spin trajectories in a gaussian laser beam calculated from the equations of ref. [4] with a predictor-corrector method. $R_1 = \rho_{bb} - \rho_{aa}$, $R_2 = 2\mathrm{Re}\,\rho_{ba}$ and $R_3 = 2\mathrm{Im}\,\rho_{ba}$. For (a), (b) and (c) the radius of curvature in units of half confocal parameter $2R/b = 3.55$ and $w^2/w_0^2 = 11.48$. For (d) and (e), $2R/b = 4.25$ and $w^2/w_0^2 = 1.06$. The Rabi frequency in units of reduced transit time $(\mu E_0/2\hbar)\,(w_0/u)$ is respectively 1.8 for (a) and (d), 3.4 for (b) and (e) and 39 for (c). For (d) and (e) the curvature is small enough for Rabi precession to occur. For (a), (b) and (c) adiabatic rapid passage inverting the populations takes place; the only effect of a field increase is to complexify the trajectory between the two poles.

A quantitative understanding of the excitation efficiency should be useful to most physical chemistry studies combining molecular beams and lasers. Furthermore the possibility to produce vibrationally excited SF_6 in states of well-defined symmetry should be specifically of interest to reactive scattering studies with this molecule.

References

[1] T.E. Cough, R.E. Miller and G. Scoles, Appl. Phys. Lett. 30 (1977) 338.
[2] A. Van Lerberghe, S. Avrillier and Ch.J. Bordé, IEEE J. Quantum Electron. 14 (1978) 481;
C. Salomon, A. Van Lerberghe and Ch.J. Bordé, to be published.
[3] A. Clairon, A. Van Lerberghe, C. Salomon, M. Ouhayoun and Ch.J. Bordé, Optics Comm. 35 (1980) 368.
[4] Ch.J. Bordé, J.L. Hall, C.V. Kunasz and D.G. Hummer, Phys. Rev. 14 (1976) 236.
[5] A. Abragam, Principles of nuclear magnetism (Oxford University Press, New York, (1961).
[6] R.P. Feynman, F.L. Vernon and R.W. Hellwarth, J. Appl. Phys. 28 (1957) 49.