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OBSERVATION OF OPTICAL RAMSEY FRINGES IN THE 10 µm SPECTRAL REGION USING A SUPERSONIC BEAM OF SF_6 (*)

Ch. J. Bordé, S. Avrillier, A. Van Lerberghe, Ch. Salomon, D. Bassi (**) and G. Scoles (**) (+)

Laboratoire de Physique des Lasers (Associé au C.N.R.S., L.A. 282), Université Paris-Nord, Avenue J.B. Clément, 93430 Villetaneuse, France

(**)Istituto per la Ricerca Scientifica e Tecnologica and Unità CNR-GNSM, Dipartimento di Fisica, Università di Trento, 3805 Povo (TN), Italia.

Abstract : We report a first observation of optical Ramsey fringes in the 10 µm spectral region using a supersonic seeded beam of 7% SF_6 in He, illuminated by a CO_2 laser in spatially-separated field zones. We have used either three standing waves or four travelling waves and obtained highly contrasted fringes with a 23 kHz half-width corresponding to a 5 mm distance between zones.

Laser excitation of the vibrational energy of molecules in a beam can be conveniently detected with a cryogenic bolometer [1] and a demonstration of this technique in the case of the ν_3 mode of SF_6 excited by CO_2 or N_2O lasers has been recently given [2] . With this equipment the spatial analog of coherent transient effects such as the Rabi oscillations of the transition probability and the adiabatic rapid passage were shown to occur respectively with plane and curved wavefronts. In an attempt to investigate the potential use of this method for very high resolution spectroscopy and optical frequency standards we have made a preliminary experiment to detect the Ramsey fringes associated with saturation spectroscopy in an interaction geometry comprising three or four field zones [3-14].

For this experiment we used the P(4) F_1 and E components of the ν_3 band of SF_6 which can be reached with a waveguide CO_2 laser oscillating on the P(16) CO_2 line at 10.55 µm. To control the frequency of this laser we locked it, with a tunable frequency offset, to a conventional reference laser locked to the Q(45) F_2^7 SF_6 line. The beam from the waweguide laser was spatially filtered and magnified to have a waist of w_0 = 6 mm. In the case of illumination by this single beam the resulting width (FWHM) of the observed line was a combination of transit broadening and residual first-order Doppler effect along the optical axis and amounted to 300 kHz.We used the Rabi oscillation to set the laser beam waist precisely on the molecular beam [2]. Four oscillations of the signal could be observed with a 40% contrast with successive minima obtained for a total power of $\sim$1,4,9 and 16 mW.

To obtain fringes ,part of the laser beam was intercepted before the interaction region by a screen which transmitted the light only through 1 mm wide slits. Two different geometries were used in these experiments. In the first one, three equidistant standing waves were generated by three equidistant slits of 5 mm separation together with a corner cube placed on the other side of the molecular beam to

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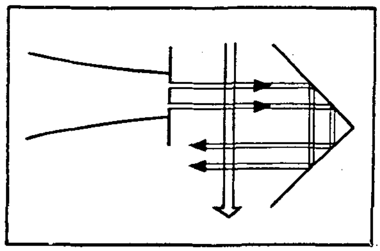
(+)Permanent address : Physics and Chemistry Department, University of Waterloo, Waterloo, Ontario, Canada, N2L 3G1.

retroreflect the light back through the slits. In the second geometry, only two of the previous slits were illuminated. An offset between the center of the slits and the center of the corner cube generated two counter-propagating sets of travelling waves with a 5 mm distance between adjacent co-propagating waves of each set. The spacing between the two sets can be arbitrary and was actually 10 mm in this experiment. Highly contrasted fringes have been obtained in both cases and as an example the figure displays the signal corresponding to the four travelling waves case.
Use of purely travelling waves to obtain optical Ramsey fringes has been suggested by the analysis of references [4-5,10] and we have here a first demonstration of this possibility together with the Ca beam experiment of Helmcke et al.[14] . The broad pedestal has a width ∿1.4 MHz corresponding to the transit broadening width across a single zone. The fringes themselves have a 23 kHz width (HWHM) consistent with a ∿930 m/sec peak velocity of the SF_6 molecules [15].



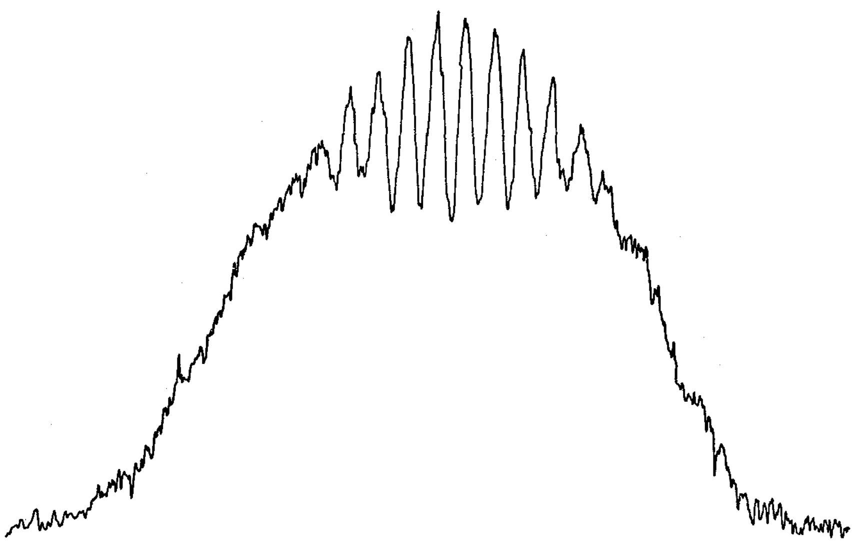


Figure 1. :

Ramsey pattern obtained for $P(4)F_1$ SF_6 line with four travelling waves (interaction geometry illustrated by the inset). The horizontal scale is linear in frequency and one fringe period corresponds to 92.5 kHz. The total laser power before the slits was 18 mW. The signal was recorded in a single one minute sweep with a 0.1 second time constant and a 30 Hz modulation frequency of the laser amplitude.

We have the choice between various mathematical tools for the theoretical description of these experiments. The approaches which have been used in the past are either perturbative calculations using density matrix diagrams or numerical treatments of the density matrix equations in the strong field case [4,5] . Another powerful tool is the 2x2 matrix formalism presented at this conference [16] . Since it applies very well to the present experiment it is worth giving here a brief outline of this theory.

The evolution operator for the two-component spinor $\begin{pmatrix} b \\ a \end{pmatrix}$ describing a two-level system is written :

$$\mathcal{U}(t,t_o) = \mathcal{T}\exp\left\{\int_{t_o}^{t}\left(\frac{H(t')}{i\hbar} - \frac{\Gamma}{2}\right) dt'\right\}$$

where $H = \begin{pmatrix} E_b & 0 \\ 0 & E_a \end{pmatrix} + V(t)$ and $\Gamma = \begin{pmatrix} \gamma_b & 0 \\ 0 & \gamma_a \end{pmatrix}$ are easily expanded on the basis of the Pauli matrices $I, \vec{\sigma}$.

The time-ordering operator $\mathcal{T}$ can be ignored either in the case of travelling waves with constant fields in each zone or in the case of standing waves with arbitrary transverse dependence of the fields.

In the first case the interaction hamiltonian V(t) is time-independent in a rotating frame (if we make the rotating wave approximation) :

$$-\frac{\mu E}{2}\begin{pmatrix} 0 & \exp(\pm ikz+i\varphi) \\ \exp(\mp ikz-i\varphi) & 0 \end{pmatrix}$$

The evolution operator reduces to a simple 2x2 matrix :

$$\begin{pmatrix} A & Be^{\pm ikz} \\ Ce^{\mp ikz} & D \end{pmatrix} = \exp\left\{\left(-i\,\frac{E_a+E_b}{2\hbar} - \frac{\gamma_{ba}}{2}\right)(t-t_o)I\right\}\left[I\cos\frac{\Omega(t-t_o)}{2} + i\,\frac{\vec{\Omega}.\vec{\sigma}}{\Omega}\sin\frac{\Omega(t-t_o)}{2}\right]$$

where $\vec{\Omega} = \left(\frac{\mu E}{\hbar}\cos(kz+\varphi),\ \mp\frac{\mu E}{\hbar}\sin(kz+\varphi),\ \Omega_o \mp kv_z + i(\gamma_b-\gamma_a)/2\right)$
is the effective field vector and with

$$\gamma_{ba} = (\gamma_b + \gamma_a)/2 \quad , \quad \Omega_o = \omega - \omega_o \quad , \quad \Omega^2 = \vec{\Omega}^2$$

With four travelling waves and molecules initially in state a, we obtain the final two-component spinor by simple multiplication of matrices :

$$\begin{pmatrix} A_4 & B_4e^{-ikz} \\ C_4e^{ikz} & D_4 \end{pmatrix}\begin{pmatrix} e^{i(\Omega_o+kv_z)T/2} & 0 \\ 0 & e^{-i(\Omega_o+kv_z)T/2} \end{pmatrix}\begin{pmatrix} A_3 & B_3e^{-ikz} \\ C_3e^{ikz} & D_3 \end{pmatrix}\begin{pmatrix} e^{ikv_z(t-T)} & 0 \\ 0 & e^{-ikv_z(t-T)} \end{pmatrix}$$

$$\begin{pmatrix} A_2 & B_2e^{ikz} \\ C_2e^{-ikz} & D_2 \end{pmatrix}\begin{pmatrix} e^{i(\Omega_o-kv_z)T/2} & 0 \\ 0 & e^{-i(\Omega_o-kv_z)T/2} \end{pmatrix}\begin{pmatrix} A_1 & B_1e^{ik_z} \\ C_1e^{-ik_z} & D_1 \end{pmatrix}\begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

where T is the time of flight between the first two and last two field zones. For the sake of simplicity we have taken $\gamma_a = \gamma_b$ and we have ignored the time of flight between the two central zones but we have written explicitly the change of rotating frame between these two counter-propagating fields.

The part of the final upper level population representing the fringe pattern is simply :

$$bb^{*} = \left(A_4B_4^{*}A_3C_3^{*}A_2B_2^{*}B_1D_1^{*}e^{2i\Omega_oT} + A_4B_4^{*}B_3D_3^{*}C_2D_2^{*}B_1D_1^{*}e^{2i\Omega_oT} + \text{c.c.}\right)\exp(-\,2\gamma_{ba}T)$$

The Doppler phase cancels out because it reverses in the second dark zone. (It is easy to follow the corresponding trajectory of the pseudo-spin and this will be illustrated in another paper). The factors multiplying the oscillating terms $\exp\pm 2i\Omega_o T$ have to be numerically integrated over the v_z distribution to obtain the exact shape of the fringes envelope. Let us finally point out that when the field phases cancel out (which is the case in our experiment) the central fringe corresponds to a negative contribution.

In the case of standing-waves we cannot completely remove the time dependence of the interaction hamiltonian which is written (in the rotating frame at ω and in the molecular frame) :

$$V = -\mu E \cos(kz + kv_z t + \varphi)\ U(\vec{r} + \vec{v}t)\ \sigma_x$$

where $U(\vec{r})$ is the transverse dependence of the field in each zone.
The only possibility to reduce the total hamiltonian to an operator commuting with itself at different times is to neglect the σ_z term during the interaction with the field. The evolution operator is then simply :

$$I\cos\Phi + i\sigma_x \sin\Phi$$

$$\text{with} \qquad \Phi = \frac{\mu E}{\hbar}\int_{-\infty}^{+\infty} U(t)\cos(kv_z t + kz + \varphi)\,dt$$

The time evolution of the two-component spinor is again described by the product of matrices corresponding to the five zones and the final result for the oscillating part of bb* has the following form already derived by Dubetsky [6] with a different approach :

$$bb^* = \frac{1}{4}\exp(-2\gamma_{ba}T)\ \cos 2\Omega_o T\ \sin 2\Phi_3\ \cos 2\Phi_2\ \sin 2\Phi_1$$

This formula displays the oscillation character but, unfortunately, does not have the simplicity of the travelling wave case since further integration on both z and v_z are required to obtain the signal.

A detailed comparison of the observed line profiles with calculated ones is presently under way. The predicted apparent splitting of the fringe pattern in a strong field has been observed in the three-slit experiment and is also being investigated.

We expect now to be able to increase significantly the resolving power with Gaussian zones separated by much larger distances using the corner cube techniques described in [17] . Let us finally point out that a major advantage of supersonic beams for an optical frequency standard is the good monochromaticity in velocity space yielding a well-defined second-order Doppler shift.

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