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# 2.3-kHz two-photon Ramsey fringes at 30 THz

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This paper reports observations of Ramsey fringes on a 10.6-$\mu$m two-photon transition of $SF_6$. We used two interaction zones separated by 8 cm and observed fringes of frequency 2.3 kHz. The experimental apparatus might be adapted to increase the resolution by an order of magnitude. The present 10-$\mu$m secondary frequency standards based on the $CO_2$ laser locked onto a saturated absorption resonance of $OsO_4$ reach an accuracy of a few $10^{-13}$. This work contributes to the current attempts to build a new generation of frequency standards with a potential accuracy in the $10^{-14}$–$10^{-15}$ range. [S1050-2947(99)50608-4]

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The $CO_2$ laser locked onto a saturated absorption resonance of $OsO_4$ provides a secondary frequency standard in the 10-$\mu$m region, with an accuracy between 50 Hz and 1 kHz [1,2] and a stability better than the hydrogen maser for averaging times less than 100 s [3,4]. To improve this performance the well-known strategy is to record narrower and narrower lines; since most of the systematic errors are proportional to the linewidth of the frequency reference, any increase in the experimental resolution will result in an increase in the ultimate accuracy of the frequency standard.

Recently, we explored the possibility for high-resolution spectroscopy of the slow molecule detection technique. Since the usual cooling techniques are not possible for molecular references in the infrared (IR), it consists in recording the signal coming solely from the slow molecules, without any cooling of the gas. This method was demonstrated for saturated absorption in methane [5,6] and in $OsO_4$ [7] and for a two-photon resonance in $SF_6$ [8], leading to linewidths of the order of 100 Hz. Alternatively to cell spectroscopy, a well-known technique to reduce the transit broadening is to use a molecular beam and to apply the method of separated fields. This method has been employed for $SF_6$ using a saturated absorption resonance [9]. This paper presents some preliminary results of narrow Ramsey fringes obtained using a two-photon Doppler-free absorption.

The main point of Ramsey spectroscopy [10] is to provide a signal the linewidth of which is no longer limited by the transit time through the laser beam. For this purpose, the single interaction zone is replaced by two spatially separated zones, with a fixed relative optical phase. When a molecule has passed through the first zone of interaction it is in a coherent superposition of lower and upper levels. The coherence precesses freely between two zones. In the second zone the molecule is either excited or deexcited, depending on the relative phase between the excitation field and the coherence. Thus, fringes develop in the excitation probability versus the laser frequency, and their spacing depends on the transit time between the two zones. The resolution is then no longer limited by the transit time associated with one beam.

In the optical domain, beam divergence dictates that this method must be associated with a sub-Doppler technique in order to avoid scrambling of the fringe pattern. The use of saturated absorption and a three- or four-zone configuration [11,9] imposes severe conditions on parallelism and equidistances which, in practice, limit the distance between zones and, finally, the ultimate resolution. By contrast, in the case of two-photon spectroscopy only two zones are required, each comprising a standing wave, and the Doppler shift is compensated in each one [12]. The only experimental condition is that the relative phase between two zones is fixed, which is easily fulfilled by generating both standing waves inside the same Fabry-Perot cavity. The experimental signal should exhibit fringes of spacing half the inverse of the mean transit time between two zones [12–14]. The side fringes will be attenuated due to the longitudinal velocity dispersion, and the entire pattern superimposed on the broader two-photon signal arising from the absorption in one single zone.

A complete treatment of energy and momentum conservation for the configuration with two spatially separated zones reveals a new subtlety. The fringes must then be interpreted as arising from the change of momentum of the molecules along the laser beam [15].

The experiment was performed on the $P(4)\ E^0$ transition of the $2\nu_3$ band of $SF_6$, first measured by Linskens *et al.* [16], who also pointed out its potential as a two-photon candidate. Figure 1 shows the structure of the three levels involved, with a detuning from the intermediate level of 76 MHz. The two-photon resonance is composed of four main hyperfine components spanning 40 kHz, with the following relative amplitudes and positions: 0.68 at −11.78 kHz, 1 at 0 kHz, 0.46 at +7.43 kHz, and 0.24 at +17.06 kHz [8,17].

10-kHz two-photon Ramsey fringes were already observed on this line elsewhere [18], with a resolution limited by the hyperfine structure and the laser linewidth. We plan to reach a much higher resolution, and have constructed an apparatus with an adjustable interzone distance to give fringes from 2 kHz to 50 Hz. Figure 2 displays the experimental setup.

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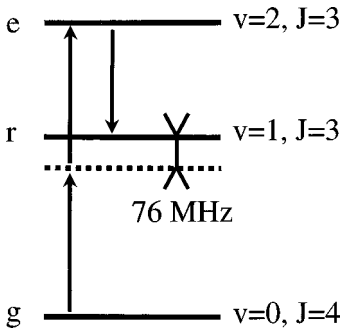


FIG. 1. Schematic of the three levels involved in the $P(4)$ $E^0$ resonance.

The $CO_2$ lasers emit in the 10-$\mu$m region, the first is frequency-locked to a strong saturation line of $OsO_4$, and the beat note between this laser and the second laser is then phase-locked to a tunable synthesizer. Thus, the stability acquired by the $OsO_4$ lock is transferred to the second laser, which is tunable around each emission line of $CO_2$. The laser frequency is then shifted by two acousto-optic modulators to reach either the two-photon resonance (transition $e$-$g$ in Fig. 1) or the upper one-photon resonance (transition $e$-$r$). The stability characteristics were already described in detail [3]: the laser linewidth is of the order of 6 Hz full width at half maximum, which is $2.10^{-13}$ in relative value, while the Allan variance reaches 0.1 Hz ($\Delta\nu/\nu = 3.5\times10^{-15}$) for an averaging time of 100 s. The reproducibility was estimated to be 10 Hz. This stability performance ensures that our experimental resolution will not be limited by the laser itself.

The molecular-beam apparatus employs a pure $SF_6$ supersonic beam to gain two main advantages. First, an enhancement of the population in the $J=4$ level arises from the rotational cooling of the supersonic expansion. Second, the velocity is lower than with the standard He-seeded beam, so the transit time is longer. The vacuum apparatus is 3 m long with two chambers separated by a skimmer. Efficient low-noise operation is ensured by diffusion pumps, an ionic pump and a cryogenic trap. The mean velocity is typically 370 m/s with a longitudinal dispersion of 70 m/s, for a nozzle pressure of $5\times10^5$ Pa. The beam divergence is of the order of 30 mrad, depending on the nozzle-skimmer distance. The total flux in the $J=4$ level is a few $10^{12}$ molecules/s, and the measured rotational temperature is less than 30 K.

A key point of the experiment is the generation of the two phase-coherent standing waves in the four-mirror folded Fabry-Perot cavity. The mirrors are mounted on an independent structure made from invar and graphite that is mechanically isolated from the vacuum chamber by graphite wool padding. The mirror mounts can be translated along the structure to adjust the interzone distance. The cavity is symmetric; the coupling mirrors are plano-concave with a 50-m curvature, while the folding mirrors are plane. The beam radius is almost constant inside the cavity and is 3 mm for a 10-cm interzone spacing. The reflectivity of each mirror is 99.8%, leading to a finesse of 500. Each concave mirror is mounted on a piezoelectric transducer (PZT) to modulate or adjust the cavity length, while the transmitted signal allows the cavity resonance to be locked onto the laser frequency.

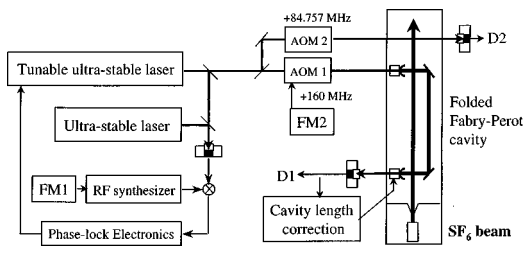


FIG. 2. Experimental scheme for two-photon Ramsey fringes: AOM, acousto-optic modulator; FM, frequency modulation.

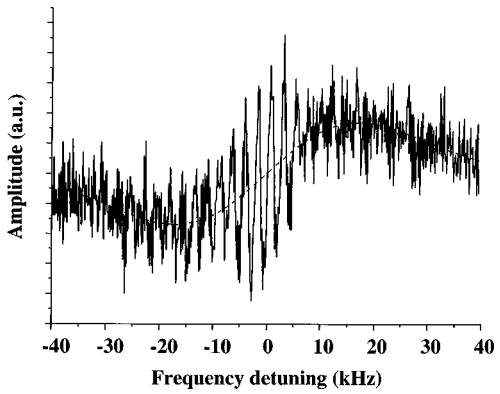


FIG. 3. Two-photon absorption signal detected on the cavity transmission beam. Experimental conditions: FM applied on the laser PZT at 1.012 kHz, depth 1 kHz, 20 mW in the cavity, pure $SF_6$ beam at $5\times10^5$ Pa, accumulation time 10 s/point. Data are fitted with the derivative of a Gaussian.

Fringes are recorded on the transmission of the cavity, detection channel D1 in Fig. 2, to take advantage of the enhancement of the signal contrast due to this cavity. For low-noise detection purposes the laser frequency is modulated through the synthesizer driving the phase-lock loop. Alternatively, the signal can be read on the molecular beam itself by probing its absorption on the upper one-photon transition, using rapid adiabatic passage with an auxiliary beam, detection channel D2 in Fig. 2. If the modulation depth is small enough, the laser frequency modulation can be applied via the radio frequency driving the acousto-optic modulator (AOM) 1. In the latter method, the auxiliary beam is not modulated. The signal arises from a modulation transfer from the molecular beam to the laser beam, and thus there is no background.

Figure 3 displays a two-photon spectrum. The central part exhibits Ramsey fringes with more than ten oscillations, which are superimposed on a broader signal arising from the two-photon absorption in a single zone. The background signal fits very well with the derivative of a Gaussian, peak-to-peak width of 34 kHz, which indicates that the interaction time is limited by the transit time. The width is a convolution of the transit line shape, peak-to-peak width of 28 kHz, with the hyperfine structure. Figure 4 displays the central part of the spectrum, corresponding to the main hyperfine component. It is fitted with a sine superimposed on a slope, resulting in a fringe frequency of 2.3 kHz and a relative determination of the central frequency with better than 10 Hz precision, limited by the signal-to-noise ratio (SNR). The absolute frequency agrees with the previous measurement [8]. The contributions of other hyperfine components, while

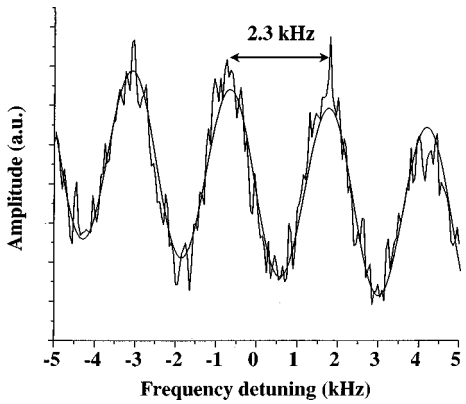


FIG. 4. Two-photon Ramsey fringes for the central hyperfine component detected on the cavity transmission beam. Experimental conditions same as for Fig. 3. Data are fitted with a sine superimposed on a slope.

evident, are less visible and the positions are consistent with the values given above.

Both spectra were recorded on the transmission of the cavity with a laser FM at 1.012 kHz at a depth of 1 kHz. The optimal power in the folded standing wave was about 20 mW, which corresponds roughly to a $\pi/2$ pulse for the two-photon transition. Multiple scans were co-added with a total accumulation time per point of 10 s for both spectra.

Since we are using FM at low frequency, the amplitude of the fringes should be greatly enhanced compared to the two-photon absorption background. This is obviously not the case, and from a comparison with a line shape simulation we deduced that the fringes arise from roughly one order of magnitude less molecules than the two-photon absorption signal. For the central component, only a few $\times 10^{10}$ molecules per second cross the two zones and are able to contribute to the interference signal. Considerable room for signal improvement is thus available.

Fringes recorded by detection channel D2, Fig. 2, using rapid adiabatic passage exhibited a rather better SNR, although the signal itself was reduced. Significantly, we succeeded in recording the fringes with a 512-Hz FM of 500 Hz depth applied through the AOM 1, which opens the possibility of doubling the interzone separation. The SNR is limited by the technical intensity noise of the auxiliary beam. The signal will be further improved if we can actively reduce this noise or, alternatively, if we store the probe beam for rapid adiabatic passage in an optical resonator.

In this paper, we present our first results in the 10-$\mu$m region using a molecular beam and Doppler-free two-photon Ramsey spectroscopy. Our spectra exhibit a 2.3-kHz spacing, which improves by a factor of 5 the previous results obtained with a saturated absorption resonance of $SF_6$ [9]. The fringes arise from a 8-cm interzone separation, which demonstrates the experimental advantage of a two-photon configuration. The achieved resolution compares very well with that obtained in the optical domain using Ramsey spectroscopy as, for example, the 1-kHz spacing using Ca at 657 nm in a magneto-optical trap with time-separated fields [19] or the 5-kHz spacing of the two-photon $1S$-$2S$ transition at 243 nm in hydrogen [20].

Using the same transition with detection of slow molecules in a cell [8] we achieved a linewidth of 280 Hz half width at half maximum (HWHM), to be compared with the present 575-Hz linewidth (HWHM) of the fringes. But the present setup has the potential to increase this resolution by an order of magnitude. Thus the Ramsey method seems promising because there is considerable room for both signal improvement and noise reduction. Concerning the metrological features, the systematic effects are either very small or can be easily and precisely measured or calculated, with the possible exception of the blackbody radiation shift [21]. This system is thus a very serious candidate for a frequency standard in the infrared region with a potential accuracy in the $10^{-15}$ range.

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