RAMSEY FRINGES IN SATURATION SPECTROSCOPY

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Provided one has been able to develop/acquire frequency and intensity stabilized sources of coherent radiation, there remain two main problems in the pursuit of narrow lines: transit time broadening and Doppler broadening. The first-order Doppler effect may be essentially eliminated by various techniques: saturated absorption, two-photon spectroscopy, highly collimated beams, ... Transit time (interaction time) broadening is more difficult to eliminate. Still, it has recently proved possible to resolve the radiative recoil-induced doublets in the three main hyperfine components of methane at 3.39 \( \mu m \). This high resolution (2 parts in \( 10^{11} \)), derived from an external absorption cell with a 30 cm aperture, nevertheless remained 2 orders of magnitude removed from the natural lifetime line width. Larger cells and associated optics being cumbersome, one is induced to find alternative schemes to reduce transit time broadening and so approach natural line width resolution.

An attractive solution to this transit time problem in saturated absorption was recently studied by Ye.V. Baklanov and his coworkers [1]. A beam of quantum absorbers could be sent transversely through three consecutive standing wave light fields equally separated in space. The resulting absorption/oscillation line shape would be an interference fringe pattern. This is the optical analog of the well-known Ramsey fringes [2] which are routinely utilized in rf spectroscopy of atomic and molecular beams. In these interference methods the spectral resolution is limited by the travel time between radiation zones rather than by the transit time through each zone.

The need for three radiation zones with saturated absorption at optical wavelengths should be explained, especially when one remembers that two zones sufficed in the microwave case. (Also only two interaction regions are needed with two-photon spectroscopy at optical wavelengths, cf. papers by Salour \textit{et al.} and Hänsch \textit{et al.} in this volume.) In the rf and microwave region, Ramsey used the interference resulting from the interaction of a quantum absorber with two separated radiation fields. After being initially prepared by the first interaction region, the atomic polarization phase evolves during the dark interzone interval at a rate which, in general, differs from that of the driving fields. The effect of the second radiation field depends on the phase of that radiation relative to the phase of the quantum system's oscillation, thus bringing a frequency tuning sensitivity dependent on the

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interzone transit time. However, the immediate synthesis of Ramsey's idea and saturated absorption in the optical domain meets with difficulties related to the shortness of the wavelength in comparison with the dimensions of the interaction region. Even in a collimated atomic beam there will be a spread of the residual Doppler velocity projections on the direction of light wave propagation.

Consider a beam of quantum absorbers crossing a collimated laser beam of waist size $w_0$. Near perpendicular incidence there is a narrow angular slice, $\delta \theta = \lambda/3w_0$, within which the absorbers experience no progressive phase shift. That is, for absorbers within this slice, the transit-time broadening exceeds the residual Doppler broadening. Adding a second optical interaction zone a distance $L$ downstream does not lead to strong Ramsey fringes since the angular slice defined by the first interaction maps into a large extension, $\Delta \lambda = L \cdot 60 = L \lambda/3w_0$, at the second beam. The condition for increased resolution by the Ramsey interference, $L/w_0 > 1$, is just the same condition that the dipoles originating at one spatial position in light zone 1 will be spread out several wavelengths along beam 2. This results in a spatial averaging of the Ramsey effect since the quantum systems with the same interzone transit time phase evolution experience different phases of the second driving field dependent on their spatial entry position into the second zone.

Baklanow and his colleagues [1] have drawn attention to the spatial-modulation aspects of these interactions and have introduced the idea of a third equally spaced interaction zone as a method to recover the Ramsey fringes. However, as a way to see the physical effects -- and especially to appreciate the phase relationships -- it is more convenient to assume we have four interaction zones. Basically, in saturation spectroscopy, the narrow resonances arise from four conceptually-separate time-ordered interaction processes: lower state population $P_L$ through dipole $\rightarrow$ upper state population $P_U$, $P_U$ through dipole $\rightarrow$ $P_L$ and $P_U$. Each process will be proportional to the electric field at the interaction point, and the system response will therefore acquire a phase associated with the electric field. Thus the atomic system will sequentially interact at four space time points with four running wave fields, which may be given in the laboratory frame as

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\begin{align*}
\epsilon_1 &= E_1 \text{ e}^{i(\omega t_1 - k z + \phi_1)} \\
\epsilon_2 &= E_2 \text{ e}^{i(\omega t_2 - k z + \phi_2)} \\
\epsilon_3 &= E_3 \text{ e}^{i(\omega t_3 + k z + \phi_3)} \\
\epsilon_4 &= E_4 \text{ e}^{i(\omega t_4 + k z + \phi_4)}
\end{align*}
$$

The space-time points $z_i$, $t_i$ are related by the absorber's free-flight trajectory. In the first interzone darkness the dipole prepared by the first interaction will precess at its own natural frequency $\omega_0$ and decay with a dipole decay rate $\gamma_0$. In the second interzone space the excited state population will only decay, with the population decay rate $\gamma_p$. In the third interzone space the system again carries a dipole moment, and precesses and
decays as before. Assuming the interzone distances to be $L$, $aL$, $bL$ respectively leads to the following expression for the total phase of the Ramsey signal:

\[ e^{i\phi_{\text{total}}} = e^{-ikv_x \frac{L}{v}(b-1)} \times e^{-i(\omega - \omega_0) \frac{L}{v}(b+1)} \times e^{i(\phi_1 - \phi_2 + \phi_3 - \phi_4)} \]

\begin{align*}
\text{Doppler} & \quad \text{Phase} & \quad \text{Detuning} & \quad \text{Phase} & \quad \text{Cavity} \\
\text{Phase} & \quad \text{Phase} & \quad \text{Phase} & \quad \text{Phase} & \quad \text{Phase} \\
\text{Dipole} & \quad \text{Population} & \quad \text{Decay} & \quad \text{Population} & \quad \text{Decay} \\
\text{Decay} & \quad \text{Decay} & \quad \text{Decay} & \quad \text{Decay} & \quad \text{Decay}
\end{align*}

For typical atomic systems of interest in connection with Ramsey fringes the lower level may be long-lived, so the dipole decay rate, $\gamma_{ab}$, and the upper level population decay rate, $\gamma_p$, are related by $\gamma_{ab} = (1/2)\gamma_p$. Considering that even a collimated beam contains a distribution of $v_x$ values, one can conclude that the Ramsey fringes are observable only for the case $b=1$, that is, when the dipole free precession distances are equal. One can also show that the fringes arise only for the case of two separated interactions with parallel-running beams followed by two separated interactions with oppositely-running beams.

It is clear that the spectral resolution is not enhanced by the second dark space, although $a \neq 0$ is sometimes a useful condition experimentally.

In the three-beam geometry two interactions occur in the central region. One may make connection with the usual saturation spectroscopy description by noting that the high resolution provided by Ramsey interference is necessary to define a narrow Bennett hole in the saturation process and is also necessary in the probe interrogation process. Thus two (equal) dark spaces are necessary.

Such saturated absorption optical Ramsey fringes were observed recently in our laboratory [3]. The experimental setup is shown in Fig.1. A fast ($v/c = 10^{-7}$) monovelocity ($\Delta v/v = 10^{-6}$) beam of metastable $^{85}\text{Rb}$ beam is efficiently produced by charge exchange of a 5-50 keV Ne$^+$ beam focused through a Na oven. This metastable atomic beam sequentially interacts with three spatially-separated, standing-wave light beams from a single mode frequency stabilized dye laser. The 588 nm $^1\text{S}_2 \rightarrow 2\text{P}_2$ transition is excited, and the fluorescence emission of the $2\text{P}_2 \rightarrow ^1\text{S}_2$ at 660 nm is detected with excellent signal-to-noise ratio with an appropriately filtered photomultiplier. $^{85}\text{Rb}$, having zero nuclear spin, is free of hyperfine structure, thus allowing a clear interpretation of our results.

We recall that the fringes are produced by the transport of phase information between the separated radiation zones by the atom's freely precessing induced dipole polarization. It is necessary then that the dipole "lives" between zones. In our experiments, the common spatial separation of the light fields was $\sim 5$ mm while the dipole decay length for a typical beam energy of 20 keV was $\sim 16$ mm.

The experimentally observed fluorescent profiles are shown in Fig.2. Curve a shows most of the beam Doppler profile, the saturated absorption dip, and the fringes due to the atom's interaction with three equally spaced
Fig.1 Schematic of experiment. The three standing-wave interaction regions are formed by two well-corrected cat's-eye retroreflectors. Typical values are the following: \( i(\text{Ne}^+) = 3 \mu \text{A} \), \( V = 20 \text{ kV} \), and the laser power is 50 mW. Fluorescence signals \( \sim 10^8 \) photons/sec reach the multiplier through the f/2 collection optics and filter.

Fig.2 Fluorescence signals. Curve a, most of beam Doppler profile (full width at half-maximum of 1.41 GHz) showing saturation dip and Ramsey fringes; \( V = 19.5 \text{ kV} \). Fringe contrast =3.8%. Curve b, saturation dip observed with two separated laser beams, \( V = 19.5 \text{ kV} \). Curves c and d, Ramsey fringes and saturation dips at \( V = 35 \text{ kV} \). Curve c, three beams; and curve d, four beams.
standing-wave radiation zones. In the inset, we compare the signals of three different light beam geometries. Curve b is the fluorescent profile produced when the Ne beam interacts with only two standing waves. Consistent with the theoretical ideas discussed above, no fringes result due to the interaction with two separated radiation fields. Curve c shows the emission profile when the atomic beam interacts with three equally spaced standing waves: one easily observes the optical Ramsey fringes. Their expected form is essentially a cosine multiplied by the saturated absorption envelope [1,4]. Adjacent fringe separations, Δν, were measured as a function of atomic beam velocity, v, and interzone separation, L. The expected relation Δω = 2πΔν = πv/L, Δν = v/2L, was verified to within the experimental precision of 10%. We note that the periodic phase shift between adjacent fringes will be developed by a smaller frequency interval if the interzone transit time is increased (larger L and/or smaller v).

The fringes produced with four equally spaced light beams are shown in curve d. The interzone separation was 2/3 that of the three light beam cases with the result that the fringe frequency separation is 3/2 the previous value (the atomic beam velocity remained the same).

Symmetric fringes result when the spatial phases in the three zones are such that they appear to be samples of a large planar wave front. This condition is intrinsically provided by the opposition of two correctly focused cat's-eye retroreflectors, as may be verified by applying Fermat's principle to the optical system illustrated in Fig.1. In our experiment the cat's-eye retroreflectors were correctly focused using an auxiliary interferometer. As may be seen in Fig.2, the fringes were symmetric, as expected.

It is interesting to consider shifting the phase of the inner zone relative to the outer ones, for example, by defocusing the cat's-eye retroreflector (i.e., we want the spatial phases to appear to be samples of a large curved wavefront). For a phase shift of 0 < φ < π in the central zone we would expect to obtain a Ramsey pattern which is asymmetric and shifted in frequency, analogous to the asymmetric line shapes obtained with curved wavefronts in the cell experiments of Ref. 5. However in the present experiments we employ standing waves in each zone, and so do not distinguish in the first two interactions which running beam is playing the role analogous to the saturating beam in the cell experiments. Thus we expect to have two overlapping Ramsey contributions of essentially equal size where one corresponds to a blue-shifted Ramsey fringe system, the other to red-shifted fringes. Their sum, as detected by fluorescence from the third zone and beyond will show essentially no asymmetry or shift in the remaining Ramsey structure. In the idealized case of standing waves, composed of equal intensity counter running beams, the fringe pattern will remain symmetrical for all values of the spatial phase difference, only the intensity of the Ramsey pattern will have a cosine dependence on this phase. In particular, for a phase shift of 180° the fringes will be inverted. This cosine behavior was first obtained by Baklanov et al. [1].

It is attractive to consider use of an electro-optic phase modulator to switch the cavity phase between 0° and 180°. The fringe signal recovered by a lock-in amplifier may decrease -- even to zero -- but will not essentially shift in frequency with variations of the additive phase delay induced into one zone (for example, by temperature variations of the electro-optic modulator). This property will clearly be of fundamental utility in the use of optical Ramsey techniques for precision spectroscopy.
In exploring the use of defocusing the cat's eye to provide a stable phase offset between the beams, we found the three-beam configuration to be particularly phase stable: no significant Ramsey fringe shape changes occurred over any reasonable focus range. With a slight lateral offset of one cat's eye, one can obtain four standing-wave radiation zones. This configuration was found suitable for providing usable phase shifts with reasonable defocusing. The fringe signals in this case can be shown to arise from the above-described three-zone interaction sequences in radiation zones 1, 2, 3 and 2, 3, 4. Another term of about the same resolution is contributed by one interaction in each zone, with an excited state population prepared in zone 2 and traveling as a population to zone 3. Figure 3a shows the four-zone Ramsey fringes with the cat's eye properly focused. For some defocus distance of the cat's eye, the phase of the central zone is appropriate to give essential cancellation of the Ramsey fringe signals, as may be seen in Fig. 3b. With the four-zone geometry, the cancellation condition is somewhat complicated and occurs for a phase shift \( \angle \Phi = \pi/2 \) where the contributions of the 1, 2, 3 and 2, 3, 4 fringes just cancel the 1, 2, 3, 4 fringes. Finally, with further defocusing, the central fringe becomes inverted as shown in Fig. 3c. Since the phase offsets for the two types of fringes are not the same (differing by a factor of 2), it is reasonable that a significant loss of the fringe contrast occurs.

We plan to explore several phase shift/fringe modulation techniques to further elucidate the behavior of the optical Ramsey effect, especially in connection with bias-free line center algorithms for precision spectroscopy and optical frequency standards applications.

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Fig. 3 Four-zone Ramsey fringes with different cavity phase shifts. Top: Correct focus, negligible phase shift. Middle: Somewhat defocused, \( 0 < \Phi \leq \pi/2 \). The two systems of fringes essentially cancel. Lower: Increased defocusing, \( \pi/2 < \Phi \leq \pi \). The fringes are inverted. See text for discussion of contrast.
There are two distinct, important advantages of the nonlinear separated interaction Ramsey method when compared to other nonlinear techniques. First, most of the quantum absorber's phase evolution occurs in the absence of the driving field, therefore power broadening is minimized. Secondly, we note that three-zone coherent interaction occurs for all the absorbers in the angular slice $\Delta \theta \sim \lambda/3w_0$ allowed by the transit time frequency uncertainty of one zone. If the radiation field filled the entire effective aperture 2L, only those particles in the angular interval $\Delta \theta' \sim \lambda/3L$ would contribute. Thus, we can achieve significant line narrowing without loss of signal-to-noise ratio.

To summarize, we have observed optical "Ramsey" fringes with spatially separated light beams and nonlinear absorption. Symmetry of the fringes is controlled by the phase relationships between the light beams. The fringes can be highly symmetric if one uses interferometric quality cat's-eye retroreflectors. With the distinct advantages of high contrast and a very sharp spectral feature (ultimately only lifetime limited), this method offers the possibility of significant improvement in optical spectroscopy and frequency metrology. An immediate candidate which should show high resolution with good signal-to-noise ratio is the calcium intercombination line $1S_0 \rightarrow 3P_1$ at 657 nm with a natural line width limit of 400 Hz. Finally, we note that these fringes have been computer synthesized with the high intensity theory [6] (program SHAPE) for both the case of separated Gaussian beams and for the case of three zones induced on a single Gaussian beam by using an annular aperture. The aperture-induced fringes were first observed by one of us (J.C.B.) in an external CH$_4$ cell at 3.39 µm.

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References