SATURATED ABSORPTION OPTICAL RAMSEY FRINGES

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Abstract

Recently, optical Ramsey interference fringes within the saturated absorption profile of a neon atomic beam were demonstrated using three spatially separated radiation fields.[1] Briefly, this method utilizes the interference which arises from the sequential interactions of the induced dipole of the quantum absorbers with the separated radiation zones in order to achieve line narrowing. The natural extension of this technique to long lived atomic and molecular absorbers is considered of important value not only in wavelength and frequency metrology but also in high resolution optical spectroscopy. A prominent candidate, which we have chosen to study, is the calcium ${}^1S_0 \leftrightarrow {}^3P_1$ (657 nm) transition[2] which has a 410 Hz natural linewidth. We will discuss the theory of saturated absorption optical Ramsey fringes and the distinct advantages of this method in comparison to the usual saturated absorption techniques. We will review the experimental neon results and present the initial results of our calcium studies.

Introduction

Classical and quantum interference has proven to be an important, productive source of information in many areas of physics. Examples in optical physics include light-light interference, such as multiple slit interference, and light-quantum system interference, such as quantum beat spectroscopy or coherent transient otpical spectroscopy. In that which follows we discuss the results and some of the possible uses of the latest, novel optical interference experiment - optical Ramsey fringes.[1,3-6] The immediate importance of this technique owes to the improvement in spectroscopic resolution. In addition the space-time separation of system-field interactions should allow quantitative experimental verification of different theoretical concepts.

Initial conceptual and experimental development of separated space-time interactions of quantum system with field was done by Ramsey[3] with microwave sources. The interference, which resulted when a beam of absorbers interacted with two spatially separated radiation fields, produced line narrowing. The straightforward extension of this method to optical frequencies was not feasible due to the very short optical wavelengths. Baklanov and co-workers[4,5] proposed three equally spaced radiation zones as an experimentally practical way to produce the optical interference analog of Ramsey's microwave experiment. This theoretical concept was further developed by C. Bordé[6] to include realistic beam geometries and the importance and effect of the relative radiation phases. A natural extension of Bordé and colleagues' earlier comprehensive study of the saturated absorption lineshape[7] permits the correct prediction of the saturated absorption 'interference' lineshape[6-8] and a possible interpretation of the physical process.

Theoretical Concepts

In most spectroscopy experiments, the radiation field extends more or less uniformly throughout the volume in which move the absorbers to be studied. Ramsey[3] was the first to point out that this may not be the most advantageous method to apply the oscillating driving field; that useful absorption profiles could be obtained if the amplitude and phase of the radiation field were non-uniform throughout this region. Particularly useful is the arrangement that would allow independent phase evolution of oscillation of quantum system and radiation field between interactions with the driving radiation field. One can imagine various experimental schemes which would produce this effect of field off, field on, field off... Simplest, perhaps, is a beam of quantum absorbers intercepting spatially separated radiation fields. That this would provide high resolution information about the energy levels of the absorber is probably not immediately clear. However, using perturbation theory, or Fourier analysis, to calculate the lineshape which results from an absorber's interaction with spatially separated radiation zones (in actuality, space separation only serves to produce temporally separated, independent interactions of absorber and field), gives an absorption profile dependent on the absorber's natural transition frequency with spectral width determined by the time between the radiation regions, rather than the transit time through each region. To better see this, we note that the interaction of absorber and field in the first radiation beam produces a coherent superposition of upper and lower states. This results in a dipole which oscillates at the natural resonance frequency in the field free region between the radiation zones. The effect of the second light field depends on the phase of the radiation relative to the absorber's oscillations, so that the absorbers passing through this field will either be further excited or returned to the ground state by stimulated emission. By these arguments, it is clear that the quantum absorption transition probability is dependent not only on the frequency of the driving field but also on the phase evolution difference of quantum system and field between zones. The phase evolution difference is proportional to the interzone transit time.

As mentioned, the extension of Ramsey's spatially separated radiation field method from microwave to optical frequencies meets with difficulties related to the shortness of the wavelength in comparison with the dimensions of the interaction region. Even in a well collimated atomic beam, there will remain a small but finite spread of transverse velocities. This means a spread of absorber velocity projections on the direction of light wave propogation. At optical frequencies, these projected velocities give rise to important Doppler frequency shifts which serve to 'wash out' the Ramsey interference fringes. To better appreciate the relevance of these effects, consider a beam of quantum absorbers crossing a transverse laser beam of waist size w_o. Near perpendicular incidence there is a narrow angular slice, $\delta\theta \sim \lambda/3w_o$, within which the absorbers experience a progressive phase shift $\leq \pi/2$. 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 Ramsey fringes since angular slice defined by the first interaction maps into a large spatial extension, $\Delta z = L\cdot\delta\theta \sim L\lambda/3w_o$, at the second beam. 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.

Baklanov and his colleagues[4,5] proposed nonlinear, saturation spectroscopy <u>and</u> three equally spaced interaction zones as a possible method to circumvent the above described spatial modulation effects and recover the optical Ramsey interference fringes. Bergquist et.al.[9] discuss ed saturation spectroscopy with four spatially separated interaction zones as a way to better see the physical effects and, most importantly, to appreciate the system-field phase relationships. As will be apparent from the following review of that discussion, the physics relevant to the understanding of the interference fringes is the same for both the 3-zone and 4-zone cases.

Saturation spectroscopy is essentially a two step process -- selection of one velocity group of absorbers, followed by a 'probe' of this group. Associated with each process are two time ordered system-field interaction processes; the first interaction produces a coherent superposition of upper and lower states, i.e., a dipole (or off diagonal matrix element, $\rho_{\alpha'\alpha}$); the second interaction gives a corresponding population (or diagonal matrix element, $\rho_{\alpha\alpha}$). Each interaction is dependent on the amplitude and phase of the radiation field at the interaction point, which gives a corresponding driving field phase dependence to the quantum system.

Consider, then, a quantum absorber's sequential interactions at space time points z_j, t_j with four spatially separated mutually parallel running wave fields, with one interaction per radiation field (and where $\hat{z}_j \,||\, \vec{k}$). For simplicity in the present discussion, we assume the running waves to be plane waves. Thus, at these interaction points, the fields may be represented as

$$\varepsilon_j^{\pm} = E_j e^{i(\omega t_j \pm k z_j + \phi_j)} + \mathrm{C.C.}$$

where ε_j^{-} is used for the first pair of interaction and ε_j^{+} for the second pair. In the absence of collisions, the space time points are related by the absorber's velocity and velocity projection onto the radiation axis. Between radiation zones, the dipole prepared by the first and third interactions will precess at the natural resonance frequency ω_o and decay with a dipole decay rate $\gamma'_{\alpha\alpha}$. In the interzone space between the second and third radiation fields, the prepared population will decay, with population decay rate $\gamma_{\alpha\alpha}$. Assuming the interzone distances to be L, aL, and bL, respectively, leads to the following expression for[9] the total phase, ϕ_T, of the Ramsey signal:

$$e^{i\phi_T} = e^{-i(\omega-\omega_o)L/_v(b+1)} \quad e^{i(\phi_1-\phi_2+\phi_3-\phi_4)}$$

Detuning Phase   Cavity Phase

$$\times e^{-\gamma'_{\alpha\alpha}(L/_v)(b+1)} \quad e^{-\gamma_{\alpha\alpha}L/_v a}$$

Dipole Decay   Population Decay

$$\times e^{-ikv_z L/_v(b-1)}$$

Doppler Phase

Since v_z is not equal to zero, even for well collimated beams, $b \neq 1$ produces a v_z-space (or equivalently z-space) modulation of the Ramsey signal. Thus, Doppler velocity, v_z, integration will average the Ramsey signal to zero for $b \neq 1$ (and $L/w_o \geq \sim 2$). We can see, however, that the Ramsey signals would be observable as a function of detuning $(\omega-\omega_o)$ for b=1, that is, when the dipole free precession distances are equal. It is also clear that the spectral resolution is not improved by the second interzone separation aL. Indeed, the Ramsey signal can be only attenuated by the population decay for $a \neq 0$. If we set a=0, the three zone geometry discussed by Baklanov is recovered. Since the dipole free precession distances must be equal, two interactions must occur in the central radiation zone.

The above arguments, which lead to the requirement of three equally spaced zones (or 4, with the proviso that the dipole precession distances be equal) in order to benefit from Ramsey interference at optical frequencies, should be only partially persuasive. The choice of the directions of each running wave field is not arbitrary, but to this point we have not shown why. 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, can be quantitatively understood with density matrix formalsim. Briefly stated, in the rotating wave approximation, only those terms in which the atomic and field 'phasors' rotate together are kept. Thus, each pair of interactions, population to dipole, dipole to population, which contributes to the absorption signal, must occur with parallel running waves with but one choice for the sign of the frequency of the driving field. Additionally, to obtain the line narrowing afforded by nonlinear saturated absorption, the probe process interactions occur in parallel running waves, which are antiparallel to the first two running waves.

We note that, in general, the optical Ramsey fringes will be displaced due to the cavity phase factor, $\exp i(\phi_4^- - \phi_3^- + \phi_2^+ - \phi_1^+)$, which involves the fixed phases of the four fields. If, instead, we use a folded standing wave, with equal intensity in the counterrunning beams, then we have a second contribution to the power absorption signal where the field phase factor is the complex conjugate of the first contribution, i.e.,

$$(\phi_4^- - \phi_3^- + \phi_2^+ - \phi_1^+) = -(\phi_4^+ - \phi_3^+ + \phi_2^- - \phi_1^-).$$

Thus, the asymmetric or shifted part of each contribution will cancel. It is to be understood that this is strictly true only if the counterrunning beam intensities are equal. More precisely, Bordé[8] has calculated the purely oscillating part of the power absorption lineshape for the special case of three equally spaced, Gaussian, standing wave radiation beams, with equal relaxation constants, $\gamma_\alpha = \gamma_{ab}$, and with equal beam waists, $w_i = w_o$, and found that the fringes are proportional to the analytic expression

$$\exp\left(-(\omega-\omega_o)^2 w_o^2/v_r^2\right) \cos\left([\omega-\omega_o]\left[2v_x d/w_o^2 - \gamma_{ba}\frac{w_o}{v_r^2}\right]\right) \cos\phi$$

where $v_r^2 \equiv v_x^2 + v_y^2$

and $\phi \equiv \phi_3^+ - \phi_2^+ + \phi_2^- - \phi_1^-$

The above formal solution similarly determines that for standing waves composed of equal intensity counterrunning beams, the asymmetric contributions cancel with the result that the fringe pattern is symmetrical for all values of the spatial field phases. Only the <u>intensity</u> of the Ramsey pattern will have a cosine dependence on the value of the net field phase. This is an important observation both for spectroscopy applications and for frequency standards work.

Experimental Results

Initial experimental investigations of optical Ramsey fringes were done by one of us (JCB)[10] with a precision frequency controlled infrared spectrometer and methane as the quantum absorber. This work was quickly followed by investigations by Bergquist, Lee and Hall, in the visible with neon as the quantum absorber.[1,9]

The Ne experimental setup for the detection of Ramsey fringes in the visible is summarized below. A fast ($v/c \sim 10^{-4}$) beam of metastable $^1S_5$ ^{20}Ne atoms was efficiently produced by charge exchange of a 5-50 keV Ne$^+$ beam focused through a Na oven. This metastable atomic beam sequentially interacted with three spatially-separated, standing-wave light beams from the single mode probe-dye laser. The dye laser was frequency stabilized to a tunable transfer cavity. The common spatial separation of the light fields was ~ 5mm while the dipole decay length for a typical beam energy of 20 KeV was ~ 16mm. The 588 nm, Ne, $^1S_5 \rightarrow {}^2P_2$, transition was excited, and the fluorescence emission of the $^2P_2 \rightarrow {}^1S_2$ at 660 nm was detected with excellent signal-to-noise ratio with an appropriately filtered photomultiplier. ^{20}Ne, having zero nuclear spin, is free of hyperfine structure, which allowed a clear interpretation of the results.

The experimentally observed fluorescent profiles are reproduced in Fig. 1. 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 standing-wave radiation zones. In the inset, we compare the signals of two 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 earlier, 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 form appears to be essentially a cosine multiplied by the saturated absorption envelope which theory predicted for a single v velocity class. Adjacent fringe separations, $\Delta\nu$, were measured as a function of atomic beam velocity, v, and interzone separation, L. The expected relation

$\Delta\nu = v/2L$, was verified to within the experimental precision of $\leq$ 10%,

Early Calcium Results

With the enormous improvement in the reduction of transit time broadening afforded by the optical Ramsey technique it was desirable to investigate long-lived atomic absorbers. This task was undertaken, not only as a precision measurement experiment, but also to determine the feasibility of the optical Ramsey fringe method as a visible frequency/wavelength standard. Subsequently we chose to study calcium, in particular the $^1S_o - {}^3P_1$ intercombination line at 657 nm[11]. The prominent features[2] of this transition include no hyperfine structure, a non-degenerate ground state, and a long lifetime (.39 ms). This gives a natural linewidth of 410 Hz or an ultimate fractional linewidth of 1×10^{-12} (with expected reproducibility accuracy 10^{-15})!

Figure 2 is a block diagram representation of the experimental setup. A Ca atomic beam, from a resistively heated oven, sequentially interacts with three spatially separated standing wave light beams from a single mode dye laser. The dye laser was frequency locked to an actively stabilized and tunable transfer cavity.[12] The servoed instantaneous linewidth of our laser is less than 750 Hz (DC drifts on a resolution scale of a few part in 10^{11} remain an experimental difficulty for this style spectrometer).[13] The common spatial separation of the light fields is variable from 0 to ~ 2 cm. Perturbations of the line shape by extraneous magnetic fields are eliminated by using a field of $2\text{-}3\times10^{4}$ Tesla (a few Gauss) in the interaction region. This splits the $\Delta m_J = \pm1$ components away from the observed $\Delta m_J = 0$ component, which has no first order Zeeman effect and whose second order shift is only about 10^{8} Hz/$_T2$ (1Hz/$_G2$). The signal is obtained by observing the fluorescence from the $^3P_1$ state downstream from the interaction region. Currently we use a five cm diameter cathode photomultiplier located approximately 20 cm downstream and in close proximity to the calcium beam. With this detection scheme, we estimate that we collect about one percent of the emitted photons; even so, useful S/N ratios are obtained. More efficient fluorescence detection systems are possible.[14] Remember that symmetric fringes result if the counterrunning beam intensities are equal or if the net field phase is zero. The last condition is satisfied 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 (to help visualize this properly, imagine a point light source at the focus of an achromatic lens -- the image is a plane wave). In the experiment, the cat's-eye retroreflectors were correctly focused using an auxiliary interferometer.

A representative derivative fluorescent profile (phase sensitive detection) of the three zone optical Ramsey fringes in calcium is shown in Fig. 3. The common radiation zone separation in this case was ~ 2.2 mm, with a radiation spot size of ~ 1 mm. the power of the input radiation beam was approximately 1 m W. The observed linewidth is approximately 100 kHz. We have seen linewidths as narrow as 50 kHz and expect soon to resolve the recoil doublet structure of this line.[15]

Conclusion

To summarize, optical Ramsey interference fringes with spatially separated light beams and nonlinear absorption have been observed. An outline of a phase evolution argument has been presented in order to better understand the physical process of Ramsey interference fringes. The fringes can be highly symmetric if one uses interferometric quality cat's-eye retroreflectors and/or equal intensity counterrunning light beams. We reiterate that this method permits line narrowing without the degradation of signal to noise inherent in the usual (single radiation zone) saturated absorption spectroscopy. Additionally power broadening and/or shift can be minimized. With this important advantage of high contrast and a very sharp spectral feature, we have demonstrated the possibilities of significant improvement in optical spectroscopy and frequency metrology. Finally, the Ramsey method promises to be of significance for the full realization of calcium as a frequency/wavelength standard.

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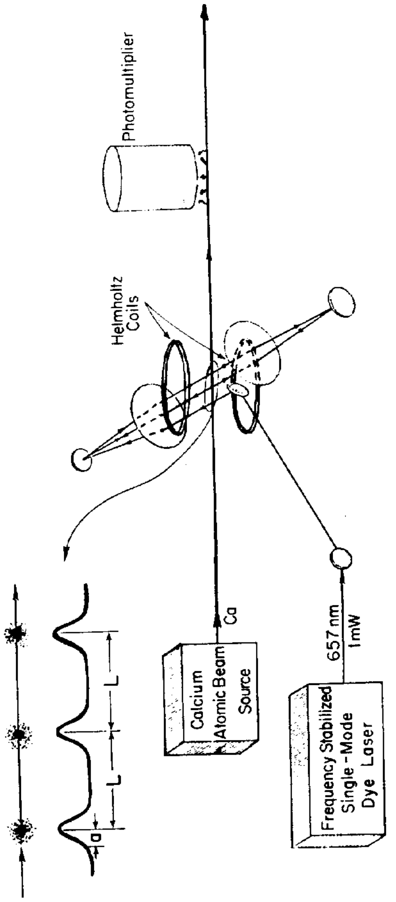

Fig. 2. Schematic of experiment. The three standing wave interaction regions are formed by two well corrected cat's eye retroreflectors. The laser power is ≲ 1 mW. Detection of the fluorescence signals by the photomultiplier occurs ~ 25 cm downstream from the interaction regions.

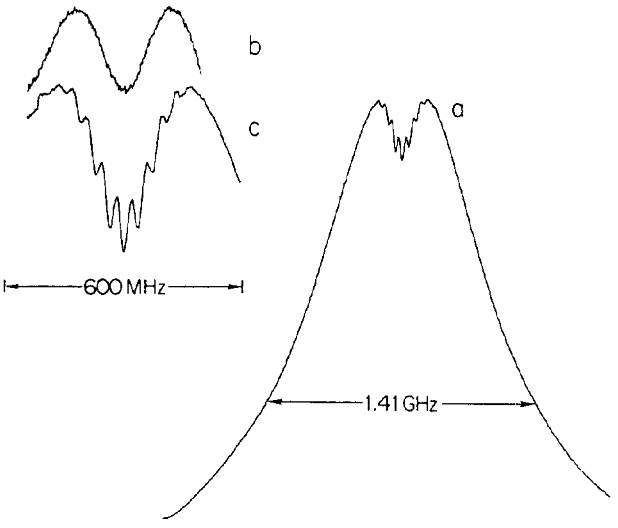

FIGURE 1. Fluorescence signals. Curve a, most of beam Doppler profile (full width as half-maximum of 1.41 GHz) showing saturation dip and Ramsey fringes, v=19.5 kV. Fringe contrast~ 3.8%. Curve b, saturation dip observed with two separated laser beams, v = 19.5 kV. Curve c, saturation dip and Ramsey fringes observed with three equally spaced laser beams, v = 35 kV.

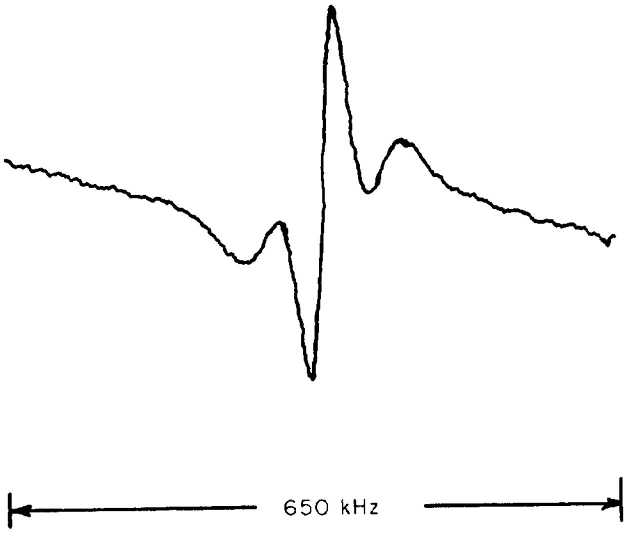

FIGURE 3. Derivative spectrum of Ca atomic beam Ramsey fringes.