Pulse train single-photon induced optical Ramsey fringes

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The fluorescence signal from an atom or molecule following its single-photon near-resonant interaction with a train of N phase-coherent Gaussian pulses is calculated within the rotating-wave and first-order Magnus approximations. The Gaussian line shape will be flanked by Ramsey fringes whose widths are inversely proportional to N, provided the time delay of the train is an integral multiple of the carrier period. The signal intensity is proportional to N^2.

I. INTRODUCTION

Ramsey[1–3] conceived the ingenious idea of using two spatially separated radio-frequency fields in order to realize a considerable reduction of time-of-flight broadening in Rabi-type molecular beam experiments. The first field, through its coherent near-resonant interaction with the molecules in the beam, induces a dipole moment whose phase depends on the interaction time and the detuning of the field from the molecular transition frequency. On exiting from the first-field zone, the molecular dipole precesses in the field-free region at the transition frequency. Upon entering the second field, the dipole has accumulated a phase change which depends on the mean molecular velocity and the distance separating the two fields. The interaction between the dipole and the second field depends on their relative phases; if the two are in (out of) phase the molecules absorb (emit) radiation. Measured as a function of the field frequency, the observed fluorescence signal exhibits an interference pattern, the so-called Ramsey fringes. The fringe pattern depends on the dephasing time (coherence lifetime) of the molecular excitation.

The extension of Ramsey's idea to the optical domain[4] requires more than the mere replacement of the radio-frequency fields by two phase-coherent laser fields, basically because the dephasing time of an optical transition is usually very short. Several methods have been developed which overcome this difficulty[5] and allow ultranarrow Ramsey fringes to be observed. One of these methods[6] uses saturation spectroscopy with spatially separated fields but introduces a third interaction zone downstream at an extension equal to that between the first and second fields. The energy absorbed in the third field is available from third-order perturbation theory. A variant[7] of this method dispenses with the third field and instead monitors there the continuous coherent radiation which is due to polarization transfer. Another method[8–10] is based on Doppler-free two-photon spectroscopy; use of two photons with oppositely directed wave vectors causes the cancellation of the first-order Doppler effect thereby avoiding the phase dependence on the transverse velocity component. In the experiments of Baklanov and co-workers[8] two separated interaction zones are present and the two-photon transition probability for excitation of the upper level after having passed through both fields is available within the rotating-wave approximation (RWA). The transition probability is also known[10] within first-order perturbation theory for the conditions that apply to Salour's experiments,[9,10] where instead of having the laser excite a beam at two separate spatial points, two time-delayed laser pulses are each used to induce Doppler-free two-photon transitions in the same group of atoms in a vapor cell. The Ramsey fringes depend on the phase correlation of the two exciting pulses; if the pulses are not phase-correlated no fringes are observed. Also, if the time delay exceeds the coherence lifetime so that the atoms dephase before the arrival of the second pulse, the fringes disappear. Teets, Eckstein, and Hansch[11] have described the Doppler-free two-photon excitation of atoms with a train of phase-coherent light pulses, originating by multiple reflections of a single laser pulse inside an optical resonator with partially transmitting mirrors. Additionally, within second-order perturbation theory, Teets, Eckstein, and Hansch provide an expression for the slowly varying amplitude describing two-photon excitation of the final state as the product of a factor containing the entire dependence on the laser frequency and a sum over the amplitude and phases which depends only on the transition frequency, the pulse delay time, and the reflectivities of the resonator mirrors. Typically the pulse durations and delay times in the experiments of Salour[9,10] and Teets, Eckstein, and Hansch[11] were of the order of a nanosecond.

With the technology for the generation of extremely-short-duration pulses now available,[12] the associated modulating envelope varies rapidly with the period of oscillation of the laser field and should be explicitly included in efforts to simulate laser pulse-atom (molecule) interactions on these times scales. The idealized transform-limited output from a perfectly mode- and phase-locked laser consists of a train of Gaussian-amplitude sinusoids. Recent advances[13] in techniques for the optical cooling and trapping of atoms could make their high-resolution spectroscopy—with greatly reduced Doppler-broadening effects—a reality. The possibility of observing single-photon induced optical Ramsey fringes prompted the calculation herein of the fluorescence spectrum of a two-level system following its interaction with a train of Gaussian-modulated short-duration phase-coherent pulses. The calculations are carried out in atomic units ($\hbar=1$) and within the RWA (Ref. 14) and the first-order Magnus approximation.[15] The RWA involves the omission of all rapidly oscillating contributions to the transition rates while the Magnus approximation is a unitarity-preserving perturbative approach to the calculation of transition amplitudes.

II. CALCULATION

The train of N Gaussian-shaped phase-coherent pulses, each with the mean carrier frequency ω_0, is represented by

$$E(t)=\sum_{k=0}^{N-1} E_k^0 \exp[-(t-kT)^2\pi/\tau_k^2]\cos[\omega_0(t-kT)] \ . \tag{1a}$$

E_k^0 is the amplitude, τ_k is the epochal duration, and $\phi_k=-k\omega_0 T$ is the phase of the kth pulse. Contiguous pulses are separated by a time delay T. Clearly $E(\pm\infty)=0$ and $E(kT)\simeq E_k^0$ for $T\gg\tau_k$, $k=0,\ldots,N-1$; the condition $T\gg\tau_k$ ensures that the epochal and nonoverlapping nature of the pulses preclude causality difficulties. Also, $NT\ll\Gamma^{-1}$ where Γ is the rapid dephasing rate of the optical transition. The Fourier spectrum of $E(t)$ at frequencies $\omega\sim\omega_0$ is

$$\mathcal{F}(E(t))=\sum_{k=0}^{N-1} \tfrac{1}{2}E_k^0\tau_k \exp[-(\omega-\omega_0)^2\tau_k^2/4\pi+ik\omega T] \ , \tag{1b}$$

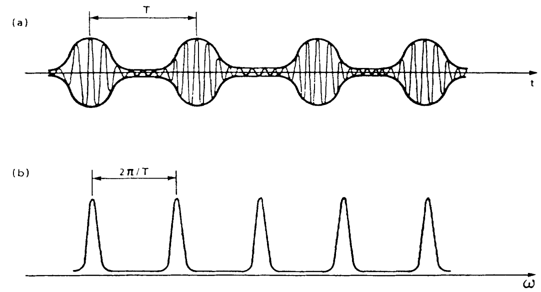

FIG. 1. (a) Series of time (T) -delayed laser pulses and (b) the frequency spectrum of the train of pulses.

a superposition of Gaussians with amplitudes $\frac{1}{2}E_k^0\tau_k$, full width at half maximum (FWHM) spectral bandwidths $\Delta\omega_k=4(\pi\ln 2)^{1/2}/\tau_k$, and phases $ik\omega T$. The frequency spectrum for $\omega\sim\omega_0$ appears as in Fig. 1, consisting of a series of equally spaced spikes with a peak separation $\sim 2\pi/T$.

In the RWA, wherein[14] one retains only the proresonant phasor of each pulse, the interaction-picture state amplitudes $b_1(t)$ and $b_2(t)$ of a two-level system dipole interacting with $E(t)$, which is arbitrarily polarized along the z axis, evolve in accordance with the equation of motion

$$i\frac{\partial}{\partial t}\begin{bmatrix} b_1(t) \\ b_2(t)\end{bmatrix} = -\tfrac{1}{2}\mu \sum_{k=0}^{N-1} E_k^0 \exp[-(t-kT)^2\pi/\tau_k^2] \begin{bmatrix} 0 & \exp[-i(\tilde{\omega}_{21}t+k\omega_0 T)] \\ \exp[i(\tilde{\omega}_{21}t+k\omega_0 T)] & 0 \end{bmatrix} \begin{bmatrix} b_1(t) \\ b_2(t)\end{bmatrix} , \tag{2}$$

where $\mu=\langle 1|\mu_z|2\rangle=\langle 2|\mu_z|1\rangle$, with μ_z being the z component of the dipole moment vector, is the electric dipole transition matrix element coupling the states $|1\rangle$ and $|2\rangle$ which are of opposite parity and $\tilde{\omega}_{21}=\omega_{21}-\omega_0$ is the frequency offset from the level separation ω_{21}. The first-order Magnus[15] unitary asymptotic solution to (2), which derives from assuming that the coefficient matrix in (2) commutes with itself at all times, is

$$\begin{bmatrix} b_1(+\infty) \\ b_2(+\infty)\end{bmatrix} = \exp\left[\sum_{k=0}^{N-1} A_k \exp(-\tilde{\omega}_{21}^2\tau_k^2/4\pi) \begin{bmatrix} 0 & i\exp(-ik\omega_{21}T) \\ i\exp(ik\omega_{21}T) & 0\end{bmatrix}\right] \begin{bmatrix} b_1(-\infty) \\ b_2(-\infty)\end{bmatrix} , \tag{3}$$

where

$$A_k=\tfrac{1}{2}\mu E_k^0 \int_{-\infty}^{\infty} dt \exp[-(t-kT)^2\pi/\tau_k^2]=\tfrac{1}{2}\mu E_k^0\tau_k$$

is the "area" of the kth pulse. For the initial conditions $b_1(-\infty)=1$ and $b_2(-\infty)=0$, the single-photon induced transition probability for excitation from $|1\rangle$ to $|2\rangle$ is given by

$$|b_2(+\infty)|^2=\sin^2\left[\left|\sum_{k=0}^{N-1} A_k \exp(-\tilde{\omega}_{21}^2\tau_k^2/4\pi) \times \exp(ik\omega_{21}T)\right|\right] , \tag{4a}$$

where we have used the identity

$$\exp\begin{bmatrix} 0 & ix \\ ix & 0\end{bmatrix} = \begin{bmatrix} \cos(x) & i\sin(x) \\ i\sin(x) & \cos(x)\end{bmatrix} .$$

The argument of the term on the right in (4a) is a superposition of N Gaussians, each being modulated by a phase factor which is explicitly dependent upon the delay time T. For a train of identical pulses each of area A and duration τ, (4a) reduces to

$$|b_2(+\infty)|^2=\sin^2[2^{-1/2}A\exp(-\tilde{\omega}_{21}^2\tau^2/4\pi) \times\csc(\tfrac{1}{2}\omega_{21}T)\sin(\tfrac{1}{2}N\omega_{21}T)] \ . \tag{4b}$$

Since $\sin(x)\simeq x$ for $x\ll 1$, then if A is sufficiently small, (4b) further reduces to

$$|b_2(+\infty)|^2=\tfrac{1}{2}A^2\exp(-\tilde{\omega}_{21}^2\tau^2/2\pi) \times\csc^2(\tfrac{1}{2}\omega_{21}T)\sin^2(\tfrac{1}{2}N\omega_{21}T) \ . \tag{4c}$$

This is the key result representing the fluorescence signal from $|2\rangle$ which we now proceed to analyze.

III. DISCUSSION

The factor $A^2\exp(-\tilde{\omega}_{21}^2\tau^2/2\pi)$ on the right in (4c) contains the dependence of $|b_2(+\infty)|^2$ on the laser parameters and is independent of N and T; it is equivalent to the transition probability following the initial pulse as given in first-order perturbation theory on use of the RWA. The trigonometric factor on the right in (4c) incorporates the explicit dependence of $|b_2(+\infty)|^2$ on N and T. The transition probability $|b_2(+\infty)|^2$ is of period $\Delta\omega_{21}=2\pi/T$ in the variable ω_{21}. Figure 2 displays $|b_2(+\infty)|^2$ as a function of ω_{21} for the particular case where $N=2$. Generally, on the interval $[0,2\pi/T]$ in the variable ω_{21}, the trigonometric factor on the right in (4c) consists of N peaks which occur with amplitudes N^2 at the end points, so that the signal intensity is proportional to N^2, and with amplitudes $\csc^2[(2l-1)T\pi/2N)]$ at the points $(2l-1)\pi/NT$ for $l=2,\ldots,N-1$, respectively. Since the only variation in $|b_2(+\infty)|^2$ with ω_0 comes from the Gaussian factor on the right in (4c) then as ω_0 is varied, with ω_{21} and T held fixed, no Ramsey fringes appear. This is because as ω_0 varies successive pulses are dephased in the amount $\exp(i\omega_0 T)$ with respect to each other and the trigonometric factor in (4c) merely scales the Gaussian signal. However, by starting out with a single transform-limited pulse one can generate[9,16] from it a train of replica pulses which will be phase locked provided the delay time T is an integral multiple of the carrier optical period $2\pi/\omega_0$. Thus, as ω_0 varies so also does T with which the Gaussian line shape of FWHM bandwidth $2(2\pi\ln 2)^{1/2}/\tau$ is accompanied by Ramsey fringes as displayed in Fig. 2, but with ω_0 and ω_{21} interchanged on the horizontal axis. Clearly the widths of the fringes decrease as N increases. It is interesting to note that as N becomes very large the trigonometric factor in (4c) averages to $\frac{1}{4}\csc^2(\omega_{21}T/2)$, as does the corresponding factor in expression (4) of Teets, Eckstein, and Hansch[11] in the limit of negligible atomic relaxation and unit mirror reflectivities. In fact, if the summation in expression (3) of Teets, Eckstein, and Hansch[11] is truncated after N terms one obtains $[1-2(R_1R_2)^N\cos(N\omega_{21}T)+(R_1R_2)^N]/[1-2R_1R_2\cos(\omega_{21}T)+(R_1R^2)^2]$, where R_1 and R_2 are the mirror reflectivities, as the factor corresponding to the trigonometric factor in (4c) and to which it reduces when $R_1=R_2=1$.

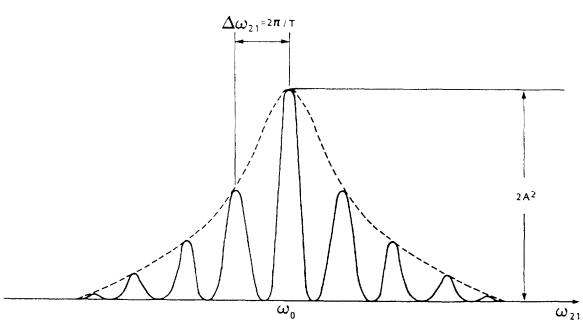

FIG. 2. Single-photon fluorescence signal following excitation by two time (T) -delayed phase-coherent Gaussian pulses as a function of the transition frequency ω_{21}.

The frequency spectrum of the train of identical pulses for $\omega\sim\omega_0$ is of the form

$$|\mathscr{F}(E(t))|$$

$$=\tfrac{1}{2}E^0\tau\exp[-(\omega-\omega_0)^2\tau^2/4\pi]\,|\csc(\tfrac{1}{2}\omega T)\sin(\tfrac{1}{2}N\omega T)|$$

and, like $|b_2(\infty)|^2$, it has a Gaussian-modulated sawtooth structure with a peak tooth gap $2\pi/T$ in the variable ω. It is the sawtooth profile that scans over the resonance $\omega_0\sim\omega_{21}$. The spectral resolution of the Ramsey fringes is limited by the bandwidth of the sawteeth and not by the bandwidth of the pulse envelope. Now, the bandwidth of the Ramsey fringes and the bandwidth of the sawteeth are both smaller than $2\pi/NT$ but since the modulating Gaussian of the fluorescence signal has a smaller (by a factor of $\frac{1}{2}$) bandwidth than that of the pulse envelope, the fringes will be narrower than the sawteeth and therefore within resolution. Detection of pulse train single-photon induced optical Ramsey fringes continues to hinge on the elimination of inhomogeneous broadening, especially Doppler broadening. While it is true that recent advances[13,17] in optical cooling of atoms can be used to greatly reduce Doppler effects, they are not totally eliminated due to the finite, albeit small, root-mean-square velocities of atoms in "optical molasses." If the residual Doppler broadening exceeds the natural linewidth Γ then clearly the Ramsey fringes will not be observed; even if it does not, it can broaden the fringes to the extent that they lie beyond the resolution of the underlying sawteeth of the exciting pulse train. One possible way of assuring that this does not occur is to choose N and/or T to be sufficiently large, but consistent with the necessary requirement that $NT\ll\Gamma^{-1}$.

Elsewhere[18] we have investigated the photodynamics of a two-level system which is orientationally polarized by a strong static electric field. Because of its finite electrical anharmonicity, the system was shown to have acquired the ability to absorb many photons from a continuous-wave (cw) or Gaussian-pulsed laser through the participation of intermediate virtual states. In either case the fluorescence signal is accompanied by satellite fringes flanking the usual Lorentzian or Gaussian line shape. In contrast to Ramsey fringes, the oscillatory sidebands are asymmetrically distributed about the multiphoton resonances. The fringed line shape for both sources is attributed to the essential transparency of the static field-oriented system to the oscillating field at prescribed intensity-dependent frequencies; for the cw source these frequencies occur when there is a coincidence of the laser-dressed states with the undressed stationary states of the aligned system while for the finite-duration pulsed source these frequencies occur whenever its effective area vanishes or is a multiple of π.

IV. CONCLUSION

In summary, we have derived (4c) within the RWA and the first-order Magnus approximation as the phase-coherent Gaussian pulse train single-photon generalization of Salour's expression[19] (25) for two-photon excitation by a pair of time-delayed square transform-limited pulses. Provided the delay time of the train is chosen to be an integral multiple of the carrier period, the fluorescence signal will be accompanied by Ramsey fringes whose widths are inversely proportional to the number of pulses in the train. Also, the signal intensity is enhanced by the square of the number of pulses in the train.

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