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General theory of laser-induced quantum beats. I. Saturation effects of single laser excitation

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A general theory of quantum beats induced by pulsed-laser excitation is presented. The theory accounts for laser saturation effects such as stimulated emission from the excited atomic state during passage of the light pulse and light-induced atomic-level displacements. Under certain experimental conditions saturation may result in a modification, sometimes even in a regeneration, of the beat amplitudes.

I. INTRODUCTION

The application of tunable dye lasers to high-resolution atomic and molecular spectroscopy has led to new spectroscopic techniques that substantially reduce or entirely avoid the Doppler broadening of spectral lines. Laser-induced quantum-beat spectroscopy[1] is one of these techniques that is particularly promising for the study of highly excited atomic and molecular systems (Rydberg systems).

Succinctly described, the quantum-beat effect is an amplitude modulation of the resonance fluorescence of an atom or molecule suddenly prepared in a coherent superposition of excited states. The beats are a consequence of the interference between the different quantum-mechanical transition amplitudes governing the decay of the system from the excited superposition state to some final lower state(s). A single atom or molecule is therefore capable of manifesting this interference phenomenon. Under suitable experimental conditions fluorescence from an ensemble of such systems will also exhibit these modulations; i.e., there will be no averaging out in time of the single-system beat pattern. This effect has been produced by a wide variety of excitation mechanisms (pulsed optical excitation, electron bombardment, collisional excitation with a thin foil, etc.) and has been observed in decays from a variety of excited states (fine-structure beats, hyperfine-structure beats, Zeeman beats, etc.).[2]

The quantum-beat frequencies correspond to the energy intervals (i.e., the classical Bohr precession frequencies) of the excited states comprising the superposition state, and therein lie the advantages of the quantum-beat effect for high-resolution spectroscopy. (i) The technique is conceptually simple and versatile; once excited, an atom "rings" out its characteristic spectrum; no scanning of static or rf fields is necessary. (ii) The observed quantum-beat signal is that of a freely decaying system; however intense the excitation may be it does not affect the atomic evolution during the measurement time; the measured frequencies therefore need no correction for signal broadening or shifting.

With the addition of high-power pulsed-dye lasers as the source of excitation, other experimental advantages become immediately apparent. (iii) The wide range over which such lasers can be tuned makes possible the excitation of a very large number of uv, visible, and ir transitions. (iv) The high power of pulsed lasers makes it possible to saturate even weakly allowed transitions. (v) Two or more lasers used sequentially can reach by a stepwise process excited states of the same parity as the ground state.

The last three advantages make it possible for an experimenter to select virtually any Rydberg state of interest or study with facility an entire series of Rydberg states.

There is also the theoretical advantage that the quantum-beat signal from a system prepared by optical excitation can be completely determined by mathematical analysis (in contrast, for example, with the beam-foil quantum-beat signal for which details of the collisional interaction, still unknown, would need to be furnished). This complete theoretical description is necessary for both optimum experiment design and signal interpretation.

Since there is a close analogy between the phenomenon of quantum beats and other manifestations of atomic resonance fluorescence, the theoretical methods applied in the past to the treatment of double-resonance or level-crossing experiments are applicable to the description of quantum beats.[3] In fact, the theory of quantum beats is implicitly contained in the classic work on dispersion theory by Breit.[4] Specific calculations of the quantum-beat signal have subsequently been performed for optical excitation of atomic and molecular systems

under different experimental conditions.[5] In all these calculations it was assumed that the optical excitation was too weak to saturate the atomic transition during the pulse duration. This is called the weak-pumping approximation. · Looked at from a different perspective, these are all first-order perturbation theories in the sense that the atomic system interacts at most once with the exciting radiation, i.e., the atom absorbs a single photon during the passage of the optical pulse. Thus it is also known as the linear absorption approximation.

While valid for classical light sources, the linear absorption approximation is no longer *a priori* justified when high-power pulsed lasers are used as the light source. In the latter case the intensity can be sufficient to saturate completely the atomic transition during the very brief pulse duration, thereby giving rise to higher-order effects. First, during the passage of the light pulse the atom will be driven back and forth between the ground and excited states by processes of photon absorption followed by stimulated emission. Second, if the exciting light beam has an asymmetric spectral profile or is not centered exactly on the transition of interest, it will displace the atomic energy levels. These processes can modify the amplitude and phase of the ensuing quantum beats.

In this article and the following one we present a general theory of laser-induced quantum beats including the effects of laser saturation, echelon excitation by several lasers, and simultaneous application of external static fields.

This theory is valid within the relatively nonrestrictive limitation of a broad-band pulse excitation. This latter condition is realized in most quantum-beat experiments. It permits the theory to be formulated within the framework of the classical optical pumping cycle first developed by Barrat and Cohen-Tannoudji[6] for weak-light sources and subsequently generalized by others[7] for continuous lasers.

In this first article, which is divided into two main parts, we consider the theory of quantum beats induced by a single-laser excitation in the absence of external static fields. The first part is devoted to a description of the time evolution of the excited atomic systems and a derivation of the quantum-beat signal. To make the physical interpretation of this calculation more apparent we examine in the second part the effects specifically tied to saturation for the simple case of a three-level atom.

In the following article the theory is generalized to include echelon excitation by more than one laser and the addition of external fields. These more complex quantum-beat experiments whose results are completely contained in the theory to be presented permit one to infer not only energy-level separations, but other important parameters characterizing Rydberg states such as g factors, polarizabilities, and the magnitude and sign of internal interaction parameters.

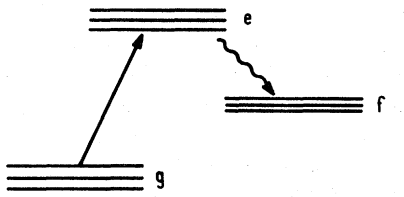

FIG. 1. Representative three-level atomic system with sublevels. Laser excitation induces transitions from g to e. Modulated fluorescence occurs in spontaneous decay from e to f. Beat frequencies are determined by energy level separations within the e multiplicity.

II. GENERAL THEORY OF SINGLE LASER-INDUCED QUANTUM BEATS

A. Determination of signal in terms of the excited-state density matrix

Consider an atomic system composed of three groups of states $|g\rangle$, $|e\rangle$, and $|f\rangle$ (see Fig. 1), and let P_g, P_e, and P_f be the respective projection operators onto these states. In a quantum-beat experiment the atoms are excited from $|g\rangle$ to $|e\rangle$ by means of a light pulse; after the passage of the pulse one observes the fluorescence emitted with a given polarization $\hat{\epsilon}_d$ in the transition $|e\rangle$ to $|f\rangle$. The duration of the light impulse T must be small in comparison with the excited-state lifetime $1/\Gamma_e$ in order that the preparation of the excited states and the detection of the decay radiation be well separated in time (see Fig. 2).

The existence of a multiplicity in the levels of

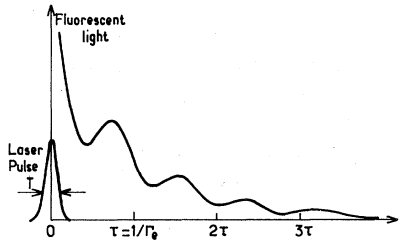

FIG. 2. The laser-pulse duration is short in comparison to the excited-state lifetime. Thus the detected signal is well separated in time from the exciting radiation.

the state $|e\rangle$ is essential to the experiment since the observed beats result from interference of the quantum-mechanical transition amplitudes for spontaneous emission out of the sublevels of the excited state. The existence of structure in the lower states $|g\rangle$ and $|f\rangle$ is not essential to the appearance of single-atom quantum beats.[8] To be complete, we mention that with the framework of QED there is the possibility of quantum beats originating from lower-state splitting if there is an interference of decay amplitudes from two or more atoms.[9] This is to be contrasted with the phenomenon under discussion manifested by individual atoms.

Although not essential to the appearance of quantum beats, the orientation or alignment of the lower-state multiplicities can affect the phase and amplitude of the beats. The energy resolution of the detected radiation, and hence the number of final states entering into the decay process, may also affect the beat signal. For these reasons we treat the most general case of three states decomposed into sublevels.

The signal observed in a quantum-beat experiment is the intensity $I(\hat{\epsilon}_d)$ of the light spontaneously emitted in a given direction with polarization $\hat{\epsilon}_d$. This intensity can be simply expressed in terms of the density matrix of the atomic system $\sigma(t)$ and a detection operator $\theta_{\rm det}(\hat{\epsilon}_d)$ as follows:

$$I(\hat{\epsilon}_d) = K\,\mathrm{Tr}[\sigma(t)\theta^{\dagger}_{\rm det}(\hat{\epsilon}_d)]\,, \tag{1}$$

with

$$\theta_{\rm det}(\hat{\epsilon}_d) = P_e\hat{\epsilon}_d\cdot\vec{\mathrm{D}}P_f\hat{\epsilon}^{*}_d\cdot\vec{\mathrm{D}}P_e\,. \tag{2}$$

The constant K depends on the solid angle of acceptance of the detector, the distance of the detector from the decaying atoms, and other geometric factors. As a scaling factor it does not affect the form of the quantum-beat signal and will henceforth be disregarded. $\vec{\mathrm{D}}$ is the angular component of the electric dipole operator $\vec{\mathfrak{D}}$ of which D_r is the radial part:

$$\vec{\mathfrak{D}} = D_r\vec{\mathrm{D}}\,.$$

In determining the time evolution of the atomic system, it is convenient to eliminate from the equations of motion the explicit appearance of the field-free Hamiltonian $\mathcal{H}_0$ by means of a transformation into the interaction representation. The transformed density matrix is

$$\tilde{\sigma}(t) = e^{+i\mathcal{H}_0 t/\hbar}\sigma(t)e^{-i\mathcal{H}_0 t/\hbar}\,. \tag{3}$$

$\tilde{\sigma}(t)$ is independent of t before and after the passage of the light pulse. Before the optical excitation the system is characterized by the density matrix

$$\sigma^{-} = \tilde{\sigma}(-\infty)\,,$$

which depends on the initial conditions such as initial orientation or alignment in the state $|g\rangle$. After the optical excitation the system is characterized by the density matrix

$$\sigma^{+} = \tilde{\sigma}(+\infty)\,.$$

The effect of the laser pulse is entirely known if one can determine the time evolution of σ^{-} into σ^{+}. This will be discussed in Sec. II B.

A useful expression for the quantum-beat signal is obtained by substituting for $\sigma(t)$ its equivalent in terms of $\tilde{\sigma}(t)$. The finite lifetime of the excited states can be adequately accounted for phenomenologically by a factor $e^{-\Gamma_e t}$, where Γ_e is a diagonal matrix whose elements are the inverse lifetimes (decay rates) of the levels in the excited manifold. This procedure is justified by QED[10] and is applicable even if the atomic system is subjected to external fields.[11] The time evolution of the density matrix (projected onto the manifold of excited states) is therefore given by

$$\begin{aligned}\sigma_e(t) &= P_e\sigma(t)P_e \\ &= P_e e^{-i\mathcal{H}_0 t/\hbar}\sigma^{+}e^{+i\mathcal{H}_0 t/\hbar}P_e e^{-\Gamma_e t}\,,\end{aligned} \tag{4}$$

and leads to an optical signal

$$I(\hat{\epsilon}_d) = \mathrm{Tr}\{P_e e^{-i\mathcal{H}_0 t/\hbar}\sigma^{+}e^{+i\mathcal{H}_0 t/\hbar}P_e\theta^{\dagger}_{\rm det}(\hat{\epsilon}_d)\}\,e^{-\Gamma_e t}\,. \tag{5}$$

From the mathematical form of Eq. (5) one clearly sees that superposed on the exponential decay are modulations of the fluorescence at the various Bohr-precession frequencies of the excited state. The amplitude of each Fourier component of the beat signal depends on the elements of the matrices $\theta_{\rm det}$ and σ^{+}. The elements of $\theta_{\rm det}$ are easily determined once one has decided upon the spectral content and polarization of the detected radiation. The elements of σ^{+}, however, must be obtained by solution of the optical-pumping equations generalized to include pulsed-laser excitation. We consider these equations in Sec. II B.

B. Equations of optical pumping for an atomic system excited by a pulsed laser

The equations of optical pumping were originally established by Barrat and Cohen-Tannoudji for the case of excitation by a weak broad-band continuous light source.[6] Other authors have subsequently generalized these equations to include continuous pumping by intense laser excitation.[7] In all these theories the different parameters characterizing the excitation rate (pumping time) are constants. It is nevertheless possible to describe within the

framework of the optical-pumping equations the effects on an atomic system of pulsed-laser excitation. The various pumping times then become functions of time and are inversely proportional to the instantaneous intensity of the light pulse evaluated at the frequency $\Omega_0 = \omega_{eg}$ (the Bohr-precession frequency characterizing the ground-state–excited-state separation).

In what follows we present the optical pumping equations generalized to pulsed-laser excitation and discuss the physical significance of the resulting terms.

Let σ_e and σ_g be the partial-density matrices obtained by projecting σ onto the manifold of excited and ground states, respectively,

$$\sigma_e = P_e \sigma P_e , \quad \sigma_g = P_g \sigma P_g .$$

The system of equations coupling σ_e and σ_g can be written as

$$\frac{d\sigma_e}{dt} = \frac{-i}{\hbar}[\mathcal{H}_0, \sigma_e] + \frac{1}{T_p(t)} P_e \hat{\epsilon} \cdot \vec{D} P_g \sigma_g P_g \hat{\epsilon}^* \cdot \vec{D} P_e - \frac{1}{2T_p(t)} [P_e \hat{\epsilon} \cdot \vec{D} P_g \hat{\epsilon}^* \cdot \vec{D} P_e, \sigma_e]_+ - i \Delta E(t) [P_e \hat{\epsilon} \cdot \vec{D} P_g \hat{\epsilon}^* \cdot \vec{D} P_e, \sigma_e] , \tag{6a}$$

$$\frac{d\sigma_g}{dt} = \frac{-i}{\hbar}[\mathcal{H}_0, \sigma_g] - \frac{1}{2T_p(t)} [P_g \hat{\epsilon}^* \cdot \vec{D} P_e \hat{\epsilon} \cdot \vec{D} P_g, \sigma_g]_+ + i \Delta E(t) [P_g \hat{\epsilon}^* \cdot \vec{D} P_e \hat{\epsilon} \cdot \vec{D} P_g, \sigma_g] + \frac{1}{T_p(t)} P_g \hat{\epsilon}^* \cdot \vec{D} P_e \sigma_e P_e \hat{\epsilon} \cdot \vec{D} P_g , \tag{6b}$$

with

$$\frac{1}{T_p(t)} = \frac{\pi}{\hbar^2} \mathcal{F}(t, \Omega_0) |(D_r)_{eg}|^2 , \tag{7a}$$

$$\Delta E(t) = \frac{1}{\hbar} |(D_r)_{eg}|^2 \mathcal{P} \int_{-\infty}^{\infty} \frac{\mathcal{F}(t, \omega)}{\omega - \Omega_0} \, d\omega \tag{7b}$$

($\mathcal{P}$ signifies the Cauchy principal value),

$$\mathcal{F}(t, \omega) = \int \langle \mathcal{E}(t) \mathcal{E}^*(t - \tau) \rangle \, e^{i\omega\tau} d\tau ,$$

where

$$(D_r)_{eg} = \langle e | D_r | g \rangle ,$$

$\vec{\mathcal{E}}(t) = \mathcal{E}(t)\hat{\epsilon}$ is the electric field of the exciting radiation, and $\langle\ \rangle$ signifies a time average.

$T_p(t)$ is the excitation rate or pumping time and is a measure of the ground-state lifetime. $\Delta E(t)$ is a laser-induced energy-level displacement. The function $\mathcal{F}(t, \omega)$ is the spectral density of the electric field, i.e., the Fourier transform of the electric field autocorrelation function.

The equations governing the time evolution of the density matrix, Eqs. (6a) and (6b), are valid as long as

$$T \gg 1/\Delta \equiv \tau_c , \tag{8a}$$

$$T_p \gg 1/\Delta , \tag{8b}$$

where Δ is the spectral width of the pulse and T is the pulse duration. The inverse of the spectral width is the correlation time of the field, a measure of the time interval over which the field phase is well defined. The physical significance of Eq. (8b) is that the phase of the incident optical pulse undergoes many random fluctuations over the period of time required to pump the atom out of its ground state. Equation (8b) also places an upper limit on the pulse intensity beyond which the Eqs. (6) may no longer be valid. The above conditions are not too restrictive and are generally satisfied in standard quantum-beat experiments. If, for example, one uses a N_2 laser-pumped dye laser of a few hundred watts peak power, the various time parameters are, for a well allowed pumping transition, of the order

$$T \simeq 10^{-9}\ \text{sec} > T_p(t) \simeq 10^{-10}\ \text{sec} > \tau \simeq 10^{-11}\ \text{sec} .$$

In deriving the density-matrix equations, Eq. (6), we have also made the assumption that the Bohr precession frequencies in the states $|e\rangle$ and $|g\rangle$ are small in comparison to Δ. This assumption is not essential; it simplifies the analysis, however, by leading to a unique pumping time and level shift for all the sublevels of a given manifold. The added complication of T_p and ΔE varying with the sublevels can be easily incorporated into the theory.

The physical significance of the terms appearing in Eqs. (6a) and (6b) is clear. The first term of the equations represents the free evolution in time of the two states concerned. The terms depending only on the elements of ground-state density matrix σ_g represent the effects of optical absorption. This process populates the excited state at a rate proportional to $1/T_p(t)$ with an angular anisotropy given by the product of operators

$$P_e \hat{\epsilon} \vec{D} \cdot P_g \sigma_g P_g \hat{\epsilon}^* \vec{D} \cdot P_e .$$

It also serves to depopulate the ground state at a rate proportional to $-1/2T_p(t)$ and displace the different sublevels of $|g\rangle$ by an amount $\Delta E(t)$. The terms depending only on the elements of the excited-state density matrix σ_e represent the effects of stimulated emission. This process is symmetric to that of absorption; it serves to populate the ground states and to depopulate and displace the excited states.

No partial-density-matrix elements of the form σ_{eg} or σ_{ge} appear in Eq. (6). These optical coherence terms which represent a macroscopic dipole-

moment precessing at optical frequencies vanish when averaged over a time interval long in comparison to the correlation time of the field τ_c. From Eq. (8) we see that all relevant time parameters are much longer than τ_c for broad-band excitation.

We consider next the application of the generalized density-matrix equations to a simple atomic system in order to see clearly the effects of laser saturation on the quantum-beat signal.

III. EFFECTS OF LASER SATURATION ON THE QUANTUM-BEAT SIGNAL OF A THREE-LEVEL ATOMIC SYSTEM

The system under consideration consists of three states, a nondegenerate ground state $|g\rangle$ and two excited states $|e_a\rangle, |e_b\rangle$ separated by an energy interval $\hbar\omega_{ab}$. (See Fig. 3.) Both excited states have a lifetime $1/\Gamma_e$. The matrix elements for optical excitation with polarization $\hat{\epsilon}$ are given by

$$a \equiv \langle e_a|\hat{\epsilon}\cdot\vec{D}|g\rangle\,, \quad b \equiv \langle e_b|\hat{\epsilon}\cdot\vec{D}|g\rangle\,.$$

Likewise, the matrix elements for spontaneous radiation with polarization $\hat{\epsilon}_d$ are given by

$$\alpha \equiv \langle e_a|\hat{\epsilon}_d\cdot\vec{D}|g\rangle\,, \quad \beta \equiv \langle e_b|\hat{\epsilon}_d\cdot\vec{D}|g\rangle\,.$$

To simplify the analysis we let a, b, α, and β be real.

The detection operator, Eq. (2), expressed in a matrix representation takes the form

$$\theta_{\text{det}} = \begin{pmatrix} \alpha^2 & \alpha\beta \\ \alpha\beta & \beta^2 \end{pmatrix}\,.$$

The density matrix of the atomic system has four independent elements which we label as follows:

$$\begin{aligned} &x \equiv \sigma_{gg}\,, \\ &y_a \equiv \langle e_a|\sigma_e|e_a\rangle\,; \quad y_b \equiv \langle e_b|\sigma_e|e_b\rangle\,, \\ &z \equiv \langle e_a|\sigma_e|e_b\rangle = \langle e_b|\sigma_e|e_a\rangle^*\,. \end{aligned} \tag{9a}$$

The initial conditions characterizing the system before the laser excitation are

$$\begin{aligned} &x^- = \tilde{x}(-\infty) = 1\,, \\ &y_a^- = y_b^- = z^- = z^{*-} = 0\,. \end{aligned} \tag{9b}$$

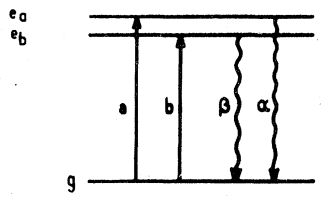

FIG. 3. Simple nondegenerate three-level system. The excited-state density matrix depends on transition-matrix elements a and b. The detection matrix depends on elements α and β.

The quantum-beat signal, Eq. (5), is a function of the density-matrix elements $y_a^+, y_b^+, z^+, z^{*+}$ which characterize the state of the atom after passage of the pulse and takes the form

$$I = [\,\alpha^2 y_a^+ + \beta^2 y_b^+ + \alpha\beta(z^+ + z^{*+})\cos\omega_{ab}t\,]\,e^{-\Gamma_e t}\,. \tag{10}$$

To calculate I explicitly we must first solve Eqs. 6(a) and 6(b) to determine the time evolution of the density-matrix elements. For the system under discussion the equations reduce to

$$\begin{aligned} \frac{dy_a}{dt} = \frac{1}{T_p(t)}&\left(a^2x - a^2y_a - \frac{abz}{2} - \frac{abz^*}{2}\right) \\ &- i\Delta E(t)ab(z^* - z)\,, \end{aligned} \tag{11a}$$

$$\begin{aligned} \frac{dy_b}{dt} = \frac{1}{T_p(t)}&\left(b^2x - b^2y_b - \frac{abz}{2} - \frac{abz^*}{2}\right) \\ &- i\Delta E(t)ab(z - z^*)\,, \end{aligned} \tag{11b}$$

$$\begin{aligned} \frac{dz}{dt} = \frac{1}{T_p(t)}&\left(abx - \frac{ab}{2}y_a - \frac{ab}{2}y_b - \frac{a^2+b^2}{2}z\right) \\ &- i\omega_{ab}z - i\Delta E(t)\,[\,ab(y_b - y_a) + (a^2 - b^2)z\,]\,. \end{aligned} \tag{11c}$$

If we assume weak pumping ($T/T_p \ll 1$) and short pulses ($\omega_{ab}T \ll 1$) we can neglect in the right-hand side of the above equation all terms involving the elements of $\tilde{\sigma}_e$. This is the linear-absorption approximation. The equations can then be immediately integrated to give σ_e^+, the density matrix after the pulse passage:

$$\sigma_e^+ = \begin{pmatrix} y_a^+ & z^+ \\ z^{*+} & y_b^+ \end{pmatrix} = k_0(+\infty)\begin{pmatrix} a^2 & ab \\ ab & b^2 \end{pmatrix}\,, \tag{12a}$$

where

$$k_0(+\infty) = \int_{-\infty}^{\infty} \frac{dt'}{T_p(t')}\,. \tag{12b}$$

The quantum-beat signal in the linear-absorption approximation is

$$I_0 = k_0(+\infty)[\,\alpha^2a^2 + \beta^2b^2 + 2\alpha\beta\,ab\cos\omega_{ab}t\,]\,e^{-\Gamma_e t}\,. \tag{13a}$$

The signal shows the appearance of beats at the frequency ω_{ab} with a modulation depth η_0 given by

$$\eta_0 = 2\alpha\beta\,ab/(\alpha^2a^2 + \beta^2b^2)\,. \tag{13b}$$

The above equations can be applied to the case of Zeeman quantum beats for a $J=0$ to a $J=1$ transition when both the excitation and detection radia-

tion are polarized perpendicular to the external static magnetic field. Only the sublevels $J=1$, $m_J=\pm 1$ are excited and the signal in the linear-absorption approximation is given by Eq. (13) with $a=b$ and $\alpha=\beta$. This leads to a modulation depth of 100%.[12]

In what follows we consider the solution to Eqs. (11a)–(11c) for different experimental conditions in which the effects of saturation are present during the passage of the pulse and compare the true quantum beat signal I to the linear-absorption signal I_0 in order to evaluate the importance of these effects.

A. Saturation in the case of a short-pulse excitation

Assume the pulse duration T to be very short compared to the Bohr precession period $1/\omega_{ab}$ and the effects of light shifts to be negligible:

$$\omega_{ab}T \ll 1, \quad (\Delta E)T \ll 1 .$$

We can therefore drop from Eqs. (11a)–(11c) terms in which ω_{ab} and ΔE appear. The resulting differential equations, with the initial conditions, Eq. (9b) taken into account, can be solved exactly to yield

$$\sigma_e(t) = k(t)\begin{pmatrix} a^2 & ab \\ ab & b^2 \end{pmatrix} \tag{14a}$$

where

$$k(t) = \frac{1}{2(a^2+b^2)}\left(1 - e^{-\int_{-\infty}^{t} \frac{2(a^2+b^2)}{T_p(t')}\,dt'}\right). \tag{14b}$$

The matrix σ_e^+ which characterizes the system after the passage of the pulse is

$$\sigma_e^+ = \begin{pmatrix} y_a^+ & z^+ \\ z^{*+} & y_b^+ \end{pmatrix} = k(+\infty)\begin{pmatrix} a^2 & ab \\ ab & b^2 \end{pmatrix}, \tag{14c}$$

and leads to the quantum-beat signal

$$I = k(+\infty)(a^2\alpha^2 + b^2\beta^2 + 2\alpha\beta ab\cos\omega_{ab}t)e^{-\Gamma_e t}. \tag{15}$$

If the pumping is weak, $k(+\infty)$ is equal to $k_0(+\infty)$ as is easily verified by expanding to first order the exponential in Eq. (14b). If the pumping is very strong, $k(+\infty)$ reduces to $\frac{1}{2}(a^2+b^2)^{-1}$ and is independent of $T_p(t)$. From Eqs. (15) and (13) we see, however, that, regardless of the pumping strength, the signals I and I_0 are proportional. In particular, the modulation depth is totally independent of the saturation parameter

$$S = T/T_p .$$

The physical significance of this result is easy to understand. The evolution of the system can be described as a sequence of absorption and stimulated emission processes taking place on the average every T_p seconds. In the weak-pumping approximation $T \ll T_p$ ($S \ll 1$), one considers only the first absorption process which, starting from the ground state $|g\rangle$, creates the excited-state populations y_a and y_b and the coherence z proportional to a^2, b^2, and ab, respectively. For an intense pulse $T \gg T_p$ ($S \gg 1$), many such sequences of absorption and stimulated emission can occur during the passage of one pulse. Stimulated emission destroys y_a, y_b, and z in the same proportions as absorption creates them, and therefore cannot change the relative proportions of the excited-state populations and coherence. The modulation depth of the beat signal is therefore unaltered. Only the overall efficiency of excited-state preparation, contained in the factor $k(+\infty)$, depends on the pulse intensity.

For a system with a more complex level structure than that of the three-level atom envisioned above, the effects of saturation can change the form of the quantum-beat signal. As an example, consider an atomic system with a pair of ground states, each one coupled by the polarization $\hat{\epsilon}$ and $\hat{\epsilon}_d$ to a distinct pair of excited states (Fig. 4). That is, $|g\rangle$ is coupled to $|e_a\rangle$ and $|e_b\rangle$ by $\hat{\epsilon}$ and $\hat{\epsilon}_d$ with excitation matrix elements a and b and detection matrix elements α and β, respectively. Likewise, $|g'\rangle$ is coupled to $|e_a'\rangle$ and $|e_b'\rangle$ by the excitation and detection matrix elements a', b' and α', β', respectively.

Each of the two subsystems envisioned is analogous to the three-level system originally treated. Since these subsystems are uncoupled, the expression for the quantum-beat signal is obtained by adding the contributions corresponding to each subsystem:

$$I = k(+\infty)(\alpha^2a^2 + \beta^2b^2 + 2\alpha\beta ab\cos\omega_{ab}t)\,e^{-\Gamma_e t} + k'(+\infty)(\alpha'^2a'^2 + \beta'^2b'^2 + 2\alpha'\beta' a'b'\cos\omega_{a'b'}t)e^{-\Gamma_e t}, \tag{16}$$

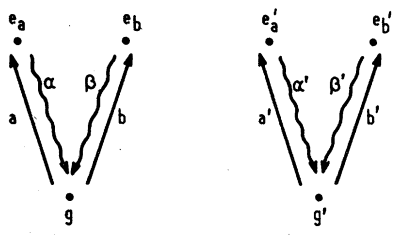

FIG. 4. Complex six-level system composed of two uncoupled three-level systems. If corresponding transition-matrix elements in each three-level system are unequal, the quantum-beat line shape depends on the saturation $S = T/T_p$.

where $k(+\infty)$ is given by Eq. (14b) and $k'(+\infty)$ by a similar expression involving a', b' instead of a, b.

In the weak-pumping limit both $k(+\infty)$ and $k'(+\infty)$ are equal to $k_0(+\infty)$. For strong pumping they are, respectively, equal to $\frac{1}{2}(a^2+b^2)^{-1}$ and $\frac{1}{2}(a'^2+b'^2)^{-1}$. If $a^2+b^2 \neq a'^2+b'^2$, then the ratio $k(+\infty)/k'(+\infty)$, and therefore the modulation depth of the two components at frequencies ω_{ab} and $\omega_{a'b'}$, depend on the saturation parameter T/T_p.

In general, this saturation effect is very small. For many atomic systems the exact calculation of the modulation depth of each spectral component differs by only a few percent from the results of the weak-pumping approximation. Figure 5 illustrates this point in the case of quantum beats observed from sodium Rydberg states[13] where the following parameters are applicable:

$$a^2=\alpha^2=2\times10^{-2},\quad a'^2=\alpha'^2=0.222\times10^{-2},$$

$$b^2=\beta^2=8\times10^{-2},\quad b'^2=\beta'^2=12\times10^{-2},\quad \omega_{ab}=\omega_{a'b'}.$$

The two signals I and I_0, given by

$$I_0=(1+0.1760\cos\omega_{ab}t)e^{-\Gamma_e t}\ \text{(no saturation)},$$

$$I=(1+0.1956\cos\omega_{ab}t)e^{-\Gamma_e t}\ \text{(with saturation)},$$

and shown, respectively, on Figs. 5(a) and 5(b), are barely distinguishable.

B. Saturation in the case of a long-pulse excitation

1. Weak-pumping limit

We consider first the results of the weak-pumping approximation as a basis for comparison with the somewhat surprising results of saturation predicted by the general theory. If the length of the pulse is no longer negligible in comparison to the excited-state Bohr precession periods ($\omega_{ab}T \geq 1$) then Eq. (11c) for the coherence z in the weak-pumping limit becomes

$$\frac{dz}{dt}=\frac{ab}{T_p(t)}-i\omega_{ab}z\ . \tag{16a}$$

This can be directly integrated to give

$$z(t)=ab\int_{-\infty}^{t}dt'\ \frac{e^{-i\omega_{ab}(t'-t)}}{T_p(t')}\ , \tag{17a}$$

which leads to

$$z^+=\bar{z}(\infty)=ab\int_{-\infty}^{+\infty}dt'\ \frac{e^{-i\omega_{ab}t'}}{T_p(t')}\ . \tag{17b}$$

The populations y_a^+, y_b^+ are, in the weak-pumping limit, independent of ω_{ab} and are therefore still given by Eq. (14c). The quantum-beat signal in the linear approximation has the form

$$I_0(\omega_{ab})=[k_0(+\infty)(\alpha^2a^2+\beta^2b^2)+2k_{\omega_{ab}}(+\infty)\alpha\beta\,ab\cos\omega_{ab}t\,]e^{-\Gamma_e t}\ , \tag{18a}$$

with

$$k_{\omega_{ab}}(+\infty)=\int_{-\infty}^{+\infty}dt'\ \frac{e^{-i\omega_{ab}t'}}{T_p(t')}\ , \tag{18b}$$

which is the Fourier transform of the pulse profile at the beat frequency ω_{ab}. [Note: $1/T_p(t)$ is proportional to the pulse intensity $\mathcal{F}(t,\Omega_0)$.] For the time-independent or "dc" component of the beat signal, we see that $k_0(+\infty)$ is the Fourier transform at frequency 0.

The modulation depth, now a function of ω_{ab}, is

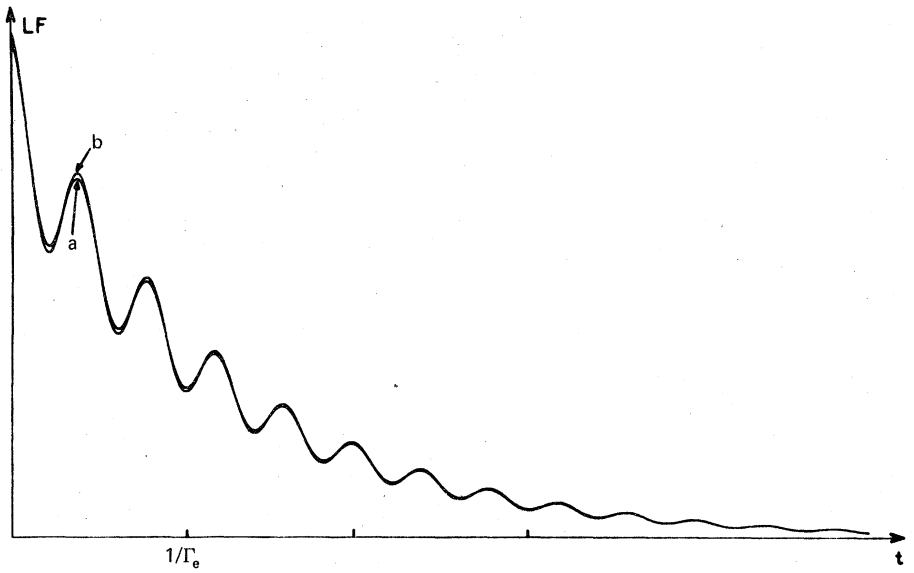

FIG. 5. Theoretical quantum-beat signal for the six-level system of Fig. 4: (a) in the limit of weak pumping; (b) in the limit of strong pumping. Matrix elements are chosen to be $a^2=\alpha^2=2.0\times10^{-2}$, $b^2=\beta^2=8.0\times10^{-2}$, $a'^2=\alpha'^2=0.222\times10^{-2}$, $b'^2=\beta'^2=12.0\times10^{-2}$, with $\omega_{ab}=\omega_{a'b'}$.

given by

$$\eta = \eta_0 \frac{k_{\omega_{ab}}(+\infty)}{k_0(+\infty)} \quad . \tag{18c}$$

From the form of Eq. (18b) we see that $k_{\omega_{ab}}(+\infty)$, and therefore the modulated component of the signal, decreases as ω_{ab} becomes of the same order or larger than $1/T$. The contributions to the signal from each differential time interval dt' destructively interfere when spread over a total interval T greater than that of the period of the beats one is trying to observe. This property is well known and imposes an upper limit of $\sim 1/T$ to the frequency of beats that can be observed.

In the case of intense pulse excitation this conclusion is no longer necessarily true.

2. *Strong-pumping limit*

If the linear approximation is no longer valid and the pulse duration no longer negligible, the entire set of coupled equations (11a)–(11c) must be solved as they stand. Let us disregard for the moment the light shifts and therefore set $\Delta E = 0$. This is always possible if one consider a symmetric excitation profile centered on the optical-transition frequency. This simplification still does not permit the eigenvalues of the determinant associated with the system of equations (11) to be cast into a simple, physically interpretable analytic form. For this case we have assumed a Gaussian pulse profile

$$\frac{1}{T_p(t)} = \lambda_1 \exp\left(-\frac{t^2 \ln 2}{(T/2)^2}\right),$$

and determined the solution to Eqs. (11) by computer. The parameter λ_1 determines the saturation parameter

$$S = \int_{-\infty}^{+\infty} \frac{dt}{T_p(t)} \quad .$$

The excitation and detection-matrix elements are $a = b = \alpha = \beta = 1$ and an energy-level separation was chosen for which $\omega_{ab} T = 5$. Figure 6 (lower trace) shows the signal $I(t)$ for a saturation parameter

$$S = \int \frac{dt}{T_p(t)} = 0.1 \quad .$$

For this case of weak pumping the modulation depth is small as predicted by the analytical solution of Sec. III B 1. Figure 6 (upper trace) shows the signal $I(t)$ for the very substantial saturation parameter $S = 4000$. The result is surprising in that the modulation depth of the beats is important even though the pulse duration is large in comparison to the period of the beats.

This effect can be understood qualitatively in terms of a random-walk problem. We have previously described the time evolution of the atomic system as a succession of absorption and stimulated emission processes each occurring on the average every T_p seconds. When the atom is in the excited state, the coherence z precesses at the frequency ω_{ab}. This precession is interrupted by a stimulated emission-process coupling z to the ground-state population x. When a subsequent absorption process occurs it renews the excited state (both population and coherence) and the precession of the coherence can now take place either in the original sense (x coupled to z) or in the opposite sense (x coupled to z^*). During the passage of the pulse the number of elementary absorption and emission processes is of the order T/T_p. During each of these processes the variation of the coherence phase (i e., the rotation angle of the analogous classically precessing dipole) is of the order $\omega_{ab} T_p$. (We assume the saturation is sufficiently large that $\omega_{ab} T_p \ll 1$.) The phase dispersion during the passage of the pulse is the root mean square of these individual phase variations:

$$(\Delta\phi)^2 = (\omega_{ab} T_p)^2 (T/T_p) = \omega_{ab}^2 T_p T \, . \tag{19}$$

In order for the pulse to generate a substantial coherence in the excited state the following condition must be satisfied:

$$(\Delta\phi)^2 = \omega_{ab}^2 T_p T \leq 1 \, . \tag{20}$$

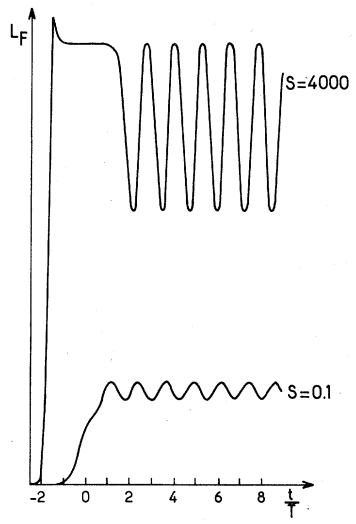

FIG. 6. The quantum-beat signal as a function of the saturation $S = T/T_p$ for a simple three-level system in the limit of long-pulse duration. Theoretical parameters are $a = b = \alpha = \beta = 1$; $\omega_{ab} T = 5$; $\Gamma_e = 0$.

This is equivalent to

$$S = T/T_p \geq (\omega_{ab}T)^2 \ . \tag{21}$$

Hence even if the pulse duration is long ($\omega_{ab}T \gg 1$), as long as Eq. (21) is satisfied it is possible to observe quantum beats in the fluorescence signal.

Looking at this from a different perspective, the effect of saturation is to slow down the coherence precession rate, and thereby prevent the destructive interference from washing out the beats during the passage of the pulse. After the passage of the pulse the precession resumes its normal rate, and the modulation of the fluorescence occurs at the frequencies characterizing the energy-level structure of the field-free atom.

To our knowledge no experimental observation of saturation regeneration of quantum beats has yet been observed because of the high value of the saturation parameter S required ($S>100$). However, the effect of saturation-induced slackening of the coherence precession has been observed in other types of optical-pumping experiments as, for example, in the Hanle effect.[14]

C. The effect of light shifts

We consider as a final case the effects on the quantum-beat signal which may be produced by light-induced energy-level displacements during the optical excitation. In order to isolate this effect from the previously studied saturation effects tied to pulse length, we assume at first that the free precession of the atom during the pulse passage is negligible ($\omega_{ab}T \ll 1$).

It may seem plausible that if the light shifts are large and different for each sublevel of the excited state, the dephasing of the atomic coherence during the passage of the pulse would lead to the disappearance of the quantum beats. This reasoning is deceptive, however. One can show rigorously that regardless of the magnitude of the displacement, and even if $(\Delta E)T \gg 1$, there is no effect of the light shifts on the signal for the assumed experimental conditions. This result follows from integrating the density matrix equations, Eqs. (11a)–(11c) in which $\omega_{ab}=0$. The solution y_a^*, y_b^*, z obtained for $t=+\infty$ is independent of ΔE.

The above result can be understood from a consideration of the symmetry of the original density matrix equation (6a). The light shifts in the excited state are described by an effective Hamiltonian

$$\Delta E P_e \hat{\epsilon}\cdot\vec{D} P_g \hat{\epsilon}^*\cdot\vec{D} P_e \ ,$$

which has exactly the same structure as the product of operators

$$(1/T_p) P_e \hat{\epsilon}\cdot\vec{D} P_g \sigma_g P_g \hat{\epsilon}^*\cdot\vec{D} P_e$$

that govern the preparation of the excited state by the light pulse *if the ground state is isotropic* (i.e., σ proportional to P_g). The atomic system is therefore prepared by absorption of an optical photon in an eigenstate of the effective Hamiltonian governing level displacement. In other words, the optical excitation can only effect a longitudinal pumping (i.e., create a population) in the basis of eigenstates of the displacement operator. In no case can it prepare atomic coherences which evolve in time at frequencies of the order of $\Delta E/\hbar$ during the passage of the pulse, even if $(\Delta E)T$ is large. Therefore saturation effects tied to light shifts are in this case rigorously null.

The above reasoning is no longer valid if $\omega_{ab}T \geq 1$. It is then necessary to consider the contributions of both the atomic Hamiltonian and the effective light-shift Hamiltonian to the evolution of the excited state during the pulse passage. The effective Hamiltonian no longer imposes its direction of quantization. The quantization axis for energy eigenstates depends on the relative magnitudes of ω_{ab} and ΔE and, in addition, varies in time. If the atomic system is more complex than the one studied here and is characterized by a nondiagonal ground-state density matrix, then the pumping operator $P_e \hat{\epsilon}\cdot\vec{D} P_g \sigma_g P_g \hat{\epsilon}^*\cdot\vec{D} P_e$ need no longer have the same symmetry as the effective light-shift Hamiltonian. The optical excitation could then effect a transverse pumping (i.e., create coherence terms) during the passage of the pulse even for $\omega_{ab}=0$.

Those cases for which light shifts play a role in the determination of the quantum-beat signal are generally complicated situations in which other saturation effects are involved as well. The manifestation of these effects generally requires particular experimental conditions (such as light pulses with strong nonresonant spectral components) not likely to be realized in an actual quantum-beat experiment.

IV. CONCLUSION

We have presented a general theory of laser-induced quantum beats that takes into account nonlinear atom-field interactions such as stimulated emission from the excited state or light-induced level shifts. Detailed examination of these saturation effects has shown them to be in general very small. Two effects in particular were pointed out: (a) A short excitation ($\omega_{ab}T \ll 1$) by an intense pulse ($T/T_p \gg 1$) can modify the amplitudes of the beat components; and (b) a long excitation ($\omega_{ab}T \gg 1$) by a very intense pulse ($T/T_p \gg \omega_{ab}^2 T^2$) can restore the beats which would be averaged out

under weak pumping. The latter effect has not yet been observed. For most quantum-beat experiments the effects of saturation are negligible and the preparation of the excited atomic states can be adequatley determined within the weak pumping or linear approximation.

In the following article we generalize our calculation to include the preparation of excited atoms by sequential laser excitation and the effects of external static fields.

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