Quantum beating in uv radiation generation by ultrashort laser pulses via four-wave mixing

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We show that quantum beating appears in the intensity of Stokes and uv radiation generated via hyper-Raman scattering (HRS) and four-wave mixing (FWM), respectively, when a large number of close-lying upper states of atoms or molecules is excited by two-photon interaction with consecutive ultrashort laser pulses. The quantum beating at frequencies correlated with the energy difference between atomic states results from a Ramsey-like interference of atomic wave packets created by the pump and probe laser fields. We reveal that the destructive interference between HRS and FWM is superimposed on the wave packet interference, leading to substantial enhancement of the beating fringe visibility. We examine the effect of laser phase fluctuations and show that they strongly limit the number of beating modes. An analytical solution of transient Maxwell-Bloch equations is obtained, and the oscillatory dependence of uv emission on the delay between two laser pulses is revealed, allowing atomic wave packets to be detected optically. A comparison to available pump-probe experiments is made.

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I. INTRODUCTION

The excitation of atoms and molecules to a desired coherent superposition state is an urgent need for efficient implementation of chemical and biological processes and for quantum control of molecular dynamics in these processes [1,2]. The time scale of the latter is usually in the femtosecond range, which stipulates the use of ultrashort laser pulses with a comparable or smaller duration to excite a medium very rapidly. As a result, laser fields having a wide spectrum can simultaneously excite a large number of close-lying upper levels of atoms or molecules, thus forming a coherent superposition of two or more atomic eigenstates. The spatial probability distribution of such atomic wave packets (WPs) is strongly localized as compared to the classical size of atoms or the internuclear distance in molecules. Therefore, study of the temporal evolution of WPs provides the possibility of retracing the chemical processes, particularly of observing directly the dissociation of the molecules or formation of chemical bonds between them [3]. Substantial progress toward this goal has been made in past decades. The technique of quantum beating (QB) has been employed in pump-probe experiments, where the atomic system is excited and probed by two laser pulses at different times. First, the pump laser pulse generates a WP in an atom or molecule, which is then observed by the second pulse coming after a certain delay time τ_d. Due to the Ramsey-type interference [4] of the two atomic WPs generated by the pump and probe pulses, periodic change or QB appears in the τ_d dependence of the detected signal. It is worth noting that, using the fact that the frequency of QB is proportional to the difference between the energies of excited states, the QB technique has been applied successfully for measurement of small frequency shifts, in particular of the fine and hyperfine splitting of high-lying atomic levels [5].

In early experiments, a photoionization signal has been used for detection of WPs by the QB technique [4,6–8]. However, for well-known reasons, the temporal resolution and the efficiency of this mechanism are low. A much higher efficiency can obviously be achieved when the WPs are detected by a coherent narrowband signal, for example, by Stokes light generated via stimulated electronic Raman scattering (SERS) of ultrashort laser pulses. Recently, such experiments were carried out in potassium and rubidium vapors [9–11], where QB has been observed in the intensities of Stokes and ultraviolet (uv) fields generated on the basis of hyper-Raman scattering (HRS) and four-wave mixing (FWM) [12], respectively. In [11] the QB in the Stokes signal at a wavelength $\lambda \sim 4-8$ μm was used for detection of the motion of atomic fragments in the dissociation of diatomic molecules. Further experiments in this field are in progress [13,14]. Meanwhile, the theory of quantum beating in HRS and FWM is still under development. Furthermore, the first analytical results for a simpler case of SERS have been published only recently [15].

In the present paper, we study theoretically the QB in the intensities of the Stokes and uv fields generated upon two-photon excitation of multilevel atoms by fs laser pulses (Fig. 1). Many quantum systems such as Rydberg atoms and diatomic molecules have an energy structure of this type. We show that in the case of ideally Fourier-transform-limited pulses the spectrum of the QB contains all frequency modes corresponding to various energy differences between excited atomic eigenstates. However, the phase fluctuations of laser fields wash out many of these contributions retaining only the modes with frequencies not exceeding the inverse duration of laser pulses. We show also that the destructive interference between HRS and FWM [16] cooperates with the WP interference strongly increasing the quantum beating visibility, thus producing a well-defined fringe pattern.

The paper is organized as follows. In the next section we present the atomic system and derive the basic equations for the time evolution of the atomic state amplitudes and for the fields generated in the medium. On the basis of the mathematical formalism developed in the Appendix at the end of the paper, we present in Sec. III the analytical solution of these equations. Here we discuss the interplay between the

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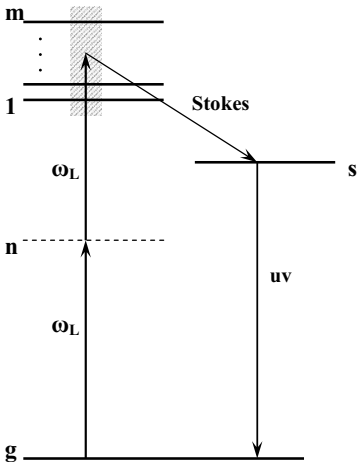

FIG. 1. Level scheme of atoms illustrating two-photon excitation of a manifold of upper structure containing m atomic eigenstates and generation of the Stokes and uv fields on the basis of hyper-Raman scattering and four-wave mixing, respectively.

two interference effects mentioned above, also taking into account the effects of phase fluctuations of the laser fields. In Sec. IV we discuss the features of QB in the intensity of the uv field and compare the results of numerical calculations with the experimental data. Our conclusions are summarized in Sec. V.

II. THE MODEL OF THE ATOM AND EQUATIONS OF MOTION

We consider two-photon interaction of the atoms having a level configuration shown in Fig. 1 with two consecutive ultrashort laser pulses, whose spectrum is wide enough to overlap all states in the excited manifold, $\Gamma \geqslant \omega_{m,1}$, and is centered near the middle of the manifold structure. Here Γ is the spectral width of the laser fields and $\omega_{m,1}$ is the frequency splitting between the extreme states m and 1. Hence, upon interaction of the atoms with each of the pulses, all the states $j=1,\ldots,m$ within the manifold are excited simultaneously, thus producing an atomic WP, whose properties depend on the laser phase coherence. In what follows, we neglect the Doppler broadening because it is small as compared to Γ.

The field amplitudes of the pump $E_p(z,t)$ and probe $E_{pr}(z,t)$ pulses, propagating along the z axis with wave vectors $k_p=k_{pr}=k_L$ and carrier frequency $\omega_p=\omega_{pr}=\omega_L=k_Lc$, are represented in the form

$$E_{p,pr}(z,t)=\mathcal{E}_{p,pr}(z,t)\exp[i(k_Lz-\omega_Lt)],$$

where the time dependence of $\mathcal{E}_{p,pr}(z,t)=\mathcal{E}_{p,pr}(t-z/c)$ is determined by the pulse shapes, while the maximum of the probe field $\mathcal{E}_{pr}(z,t)$ is shifted with respect to that of $\mathcal{E}_p(z,t)$ by the delay time τ_d.

The interaction of the atom with the laser fields is determined by their Rabi frequencies at the corresponding transitions

$$\Omega_{p,pr}^{(j)}=\frac{r_j}{\hbar}\mathcal{E}_{p,pr}^2, \tag{1}$$

where

$$r_j=\sum_n \frac{\mu_{jn}\mu_{ng}}{\hbar(\omega_{ng}-\omega_L)} \tag{2}$$

is the matrix element for two-photon transition $g\rightarrow j$ from the ground state $|g\rangle$ with ω_{nl} the frequency of atomic transition $n\rightarrow l$ and μ_{nl} the dipole moment between a pair of states n and l.

The total Stokes field, which is generated simultaneously on the transitions $j\rightarrow s$, and uv radiation emitted on the transition $s\rightarrow g$ are defined as

$$E_{S,uv}(z,t)=\mathcal{E}_{S,uv}(z,t)\exp[i(k_{S,uv}z-\omega_{S,uv}t)],$$

with the carrier frequencies ω_S and ω_{uv} and the projection $k_{S,uv}=\vec{k}_{S,uv}\hat{e}_z$ of the wave vectors $\vec{k}_S$ and $\vec{k}_{uv}$ on the z axis, respectively.

In a frame rotating with the laser and Stokes field frequencies, the Hamiltonian of the system is given by

$$H=-\hbar e^{2ik_Lz}\sum_{j=1}^{m}(\Omega_p^j+\Omega_{pr}^j)\sigma_{jg}e^{i\Delta_jt}-e^{ik_Sz}\mathcal{E}_S\sum_{j=1}^{m}\mu_{js}\sigma_{js}e^{i\Delta_jt}$$
$$-e^{ik_{uv}z}\mu_{sg}\mathcal{E}_{uv}\sigma_{sg}+\text{H.c.}, \tag{3}$$

where $\sigma_{ij}=|i\rangle\langle j|$ are the atomic operators and $\Delta_j=\omega_{jg}-2\omega_L=\omega_{js}-\omega_S$ is the two-photon detuning of laser fields from the $g\rightarrow j$ transition, as well as the detuning of the Stokes field from the $s\rightarrow j$ transition. We assume that the spectrum of uv radiation is localized at the exact resonance $\omega_{uv}=\omega_{sg}$.

The state of the atom is described by the wave function

$$\psi(z,t)=\sum_i C_i(z,t)|i\rangle, \tag{4}$$

where $C_i(z,t)$ is the amplitude of the ith atomic state satisfying the equation

$$\dot{C}_i(z,t)=-\frac{i}{\hbar}\sum_k H_{ik}C_k(z,t). \tag{5}$$

We neglect the laser field depletion and suppose that the Stokes field, although being stimulated, is not so strong as to change markedly the populations of the excited states. This means that only the amplitude of the Raman state $|s\rangle$ depends on the propagation distance. Then, using the slowly varying envelope approximation, we obtain the following propagation equations for the Stokes and uv field amplitudes:

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\mathcal{E}_S(z,t)=2i\pi\frac{\omega_S}{c}\sum_{j=1}^{m}P_S^{(j)}(z,t), \tag{6}$$

$$\left(\frac{\partial}{\partial z}+\frac{1}{c}\frac{\partial}{\partial t}\right)\mathcal{E}_{uv}(z,t)=2i\pi\frac{\omega_{uv}}{c}e^{i\Delta kz}P_{uv}(z,t), \tag{7}$$

where the polarizations $P_S^j(z,t)$ at the Stokes transitions $j\rightarrow s$ and $P_{uv}(z,t)$ at the uv frequency are given by

$$P_S^j(z,t) = \mu_{js} N C_j(t) \tilde{C}_s^*(z,t) e^{-i\Delta_j t}, \tag{8}$$

$$P_{uv}(z,t) = \mu_{gs} N \tilde{C}_s(z,t) \tilde{C}_g^*(t), \tag{9}$$

with N the atomic number density and $\Delta k = 2k_L - k_S - k_{uv}$ the wave-vector mismatch. Here the propagation phase factors are included in the ground and Raman state amplitudes by $\tilde{C}_g = C_g e^{2ik_L z}$ and $\tilde{C}_s = C_s e^{ik_S z}$.

Under the adopted approximations we obtain the following equations for the atomic state amplitudes:

$$\dot{\tilde{C}}_g(t) = i \sum_{j=1}^{m} (\Omega_p^{j*} + \Omega_{pr}^{j*}) C_j(t) e^{-i\Delta_j t}, \tag{10}$$

$$\dot{C}_j(t) = i(\Omega_p^j + \Omega_{pr}^j) \tilde{C}_g(t) e^{i\Delta_j t}, \tag{11}$$

$$\dot{\tilde{C}}_s(z,t) = \frac{i}{\hbar} \mathcal{E}_S^*(z,t) \sum_{j=1}^{m} \mu_{js} C_j(t) e^{-i\Delta_j t} + \frac{i\mu_{sg}}{\hbar} \mathcal{E}_{uv}(z,t) \tilde{C}_g(t) e^{-i\Delta k z}, \tag{12}$$

which are subject to the initial conditions $C_g(-\infty)=1$ and $C_l(-\infty)=0$, $l \neq g$.

Equations (6)–(12) describe the evolution of the system in the field of the two laser pulses. Our task is to find the intensity of uv light as a function of time delay τ_d after switching on the probe pulse. This problem will be solved in the following section.

III. SOLUTION OF THE BASIC EQUATIONS

Here we obtain the general solution of Eqs. (6) and (7) accounting for the phase fluctuations of laser fields. We note the complexity of the density matrix approach, which does not provide an analytic solution for the field amplitudes; therefore it is convenient to first find this solution in terms of the atomic state amplitudes and then to express the final expressions through the elements of density matrix, which can then easily be averaged over the laser fluctuations.

In wave variables z and $\tau = t - z/c$ Eqs. (6), (7), and (12) are reduced to

$$\frac{\partial \mathcal{E}_S}{\partial z} = 2i\pi N \frac{\omega_S}{c} f(\tau) \tilde{C}_s^*, \tag{13}$$

$$\frac{\partial \mathcal{E}_{uv}}{\partial z} = 2i\pi N \frac{\omega_{uv}}{c} g^*(\tau) \tilde{C}_s, \tag{14}$$

$$\dot{\tilde{C}}_s = \frac{i}{\hbar} f(\tau) \mathcal{E}_S^* + \frac{i}{\hbar} g(\tau) \mathcal{E}_{uv}, \tag{15}$$

where

$$f(\tau) = \sum_{j=1}^{m} \mu_{js} C_j(\tau) e^{-i\Delta_j \tau}, \tag{16}$$

$$g(\tau) = \mu_{sg} \tilde{C}_g(\tau), \tag{17}$$

and the phase-matching condition $\Delta k = 2k_L - k_S - k_{uv} = 0$ is assumed to be satisfied in the medium. The solution of Eqs. (13)–(15) with the boundary condition $\mathcal{E}_S(0,\tau) = \mathcal{E}_S(\tau)$ is found in the Appendix, and for the uv field has the form

$$\mathcal{E}_{uv}(z,\tau) = -qzg^*(\tau) \int_{\tau_d - T/2}^{\tau} d\tau' f(\tau') \mathcal{E}_S^*(\tau') I_1(\psi) \psi^{-1}(\tau',\tau), \tag{18}$$

where $q = 4\pi N \omega_{uv}/c\hbar$, $I_1(x)$ is the modified Bessel function of the first order, and

$$\psi(\tau',\tau) = \left[2qz \int_{\tau'}^{\tau} \left(\frac{\omega_S}{\omega_{uv}} |f|^2 - |g|^2 \right) d\tau'' \right]^{1/2}. \tag{19}$$

Note that Eq. (18) displays nonvanishing generation of uv light even in the limit $\tau \to \infty$. This is because, after the laser pulses have propagated through the medium, a coherent polarization is retained at the frequency ω_{uv}. In practice, however, due to dephasing of the atoms, this polarization decays rapidly via fast exponential law, immediately stopping the uv generation. Therefore, the time integration in Eq. (18) must be limited to the interval $\tau_d - T/2 \leqslant \tau \leqslant \tau_d + T/2$ centered around time τ_d with length equal to the probe pulse duration, $T \ll \tau_d$. This relaxation is not incorporated in our calculations, so that we have to introduce the upper cutoff of integration in Eq. (18) by hand.

We do not present the solution for the Stokes field, which is not relevant here, but we note that it is easily found from Eqs. (A2) and (A4).

In the absence of an input Stokes signal, the Stokes field is generated from spontaneous noise or quantum fluctuations with a δ correlated amplitude $\mathcal{E}_S(\tau)$,

$$\frac{c}{2\pi} \langle \mathcal{E}_S(\tau) \mathcal{E}_S^*(\tau') \rangle = \frac{J_{sp}}{\Gamma_s} \delta(\tau - \tau'), \tag{20}$$

where $J_{sp}(\omega_S) = \hbar \omega_S^3 \Delta\Omega \Delta\nu_S / 2\pi^2 c^2$ is the intensity of spontaneous noise determined as the intensity of a light containing one photon in each mode within the Stokes linewidth $\Delta\nu_S = \Gamma_S / 2\pi$ and in a small solid angle $\Delta\Omega$ in the forward direction [12].

Then using Eq. (20), for the uv intensity we get

$$I_{uv} = q^2 z^2 \frac{J_{sp}}{\Gamma_s} |g(\tau)|^2 \int_{\tau_d - T}^{\tau} d\tau' |f(\tau')|^2 I_1^2(\psi) \psi^{-2}(\tau',\tau). \tag{21}$$

Finally, this expression must be averaged over laser phase fluctuations that is easily performed, if one notes that only the amplitudes $C_j(t)$ of the excited atomic states depend on the laser phase through the two-photon Rabi frequencies, as is seen from Eqs. (10)–(12). So we arrive at the final result by replacing everywhere the quantity $|f(\tau)|^2$ by its averaged value $\langle |f(\tau)|^2 \rangle$. In what follows, we will show how QB arises in the latter thanks to the exponential factors $\exp(-i\Delta_j \tau)$ in Eq. (16).

Before proceeding to these calculations, we find it worthwhile to review the solution of Eq. (21) for different gain regimes. It is clear that the Bessel function $I_1(\psi)$ in the integrand of Eq. (21) is responsible for stimulated gain in uv emission. However, by analyzing the structure of ψ we recognize that the gain is generally possible only if the condition

$$\omega_S\langle|f(t)|^2\rangle > \omega_{uv}|g(t)|^2 \tag{22}$$

holds in a sufficiently large interval of the laser pulses. Indeed, in the alternate case $I_1(\psi)$ is replaced by the ordinary Bessel function $J_1(|\psi|)$, which vanishes as z increases, and the uv radiation remains at the spontaneous noise level. This is a well-known effect of self-induced suppression of HRS caused by destructive interference between HRS and FWM [16]. The effect has been confirmed in a number of experiments with alkali-metal vapors [17]. The condition (22) restricts the laser intensity from below and increases the uv generation threshold. To overcome this constraint, it is enough usually to use a sufficiently strong pumping. However, in our case this does not always guarantee a substantial gain of Stokes and uv fields, since the condition (22) can still be violated periodically in dependence on time delay τ_d. This happens at the values of τ_d where $\langle|f(t)|^2\rangle$ reaches its minima (dark fringes) due to interference between the two atomic WPs created by the pump and probe laser pulses. This means that instead of the intuitively expected QB patterns that could be observed being originated by only the first term $\langle|f(t)|^2\rangle$ in the integrand in Eq. (18) (see, for example, the discussion in [9]), the real picture is much more complicated. To make this point more transparent, let us recall that in the high-gain limit, when $\psi(\tau',\tau)\gg 1$, which corresponds to $\omega_S\langle|f(t)|^2\rangle \gg \omega_{uv}|g(t)|^2$, $I_1^2(x)$ has an asymptotic form $e^{2x}/2\pi x$. Then we can approximate $\psi(\tau',\tau)\sim\psi(\tau_d-T,\tau)=\psi_d(\tau)$ which yields

$$I_{uv}=\frac{\omega_{uv}^2 Nz}{\hbar c\omega_S}\frac{J_{sp}}{\Gamma_s}|g(\tau)|^2\frac{e^{2\psi_d}}{\psi_d}, \tag{23}$$

showing that the factor $e^{2\psi_d}/\psi_d$, being a periodic function of τ_d with the same frequency as $\langle|f(t)|^2\rangle$, exponentially amplifies the maxima of the fringe function (bright fringes) in QB and, thereby, appreciably increases the visibility of the oscillations. Furthermore, whenever $\langle|f(t)|^2\rangle$ attains its minima at corresponding values of τ_d, two regimes are realized depending on whether $1>\psi(\tau',\tau)>0$ or $\psi(\tau',\tau)<0$. In the first case $I_1(x)/x\sim 1$ and the uv field experiences albeit small, but nonvanishing gain, while for negative $\psi(\tau',\tau)$ it is emitted only spontaneously, as is shown above. Along with Eq. (23), this dramatic reduction of uv intensity demonstrates that two interference effects, the destructive one between HRS and FWM and the other between the two atomic WPs, are superimposed, enhancing the contrast of QB practically up to 100%. Figure 2 depicts an illustration of this effect for a model atom having two eigenstates in the upper manifold. Which of the two regimes is realized depends evidently not only on the laser intensity but more essentially on the ratio of dipole matrix elements of Stokes and uv transitions, as is evident from Eq. (22). Despite the complexity of this dependence, it allows us to understand the features of the fringe pattern in uv generation at different transitions when the rest of the parameters are the same.

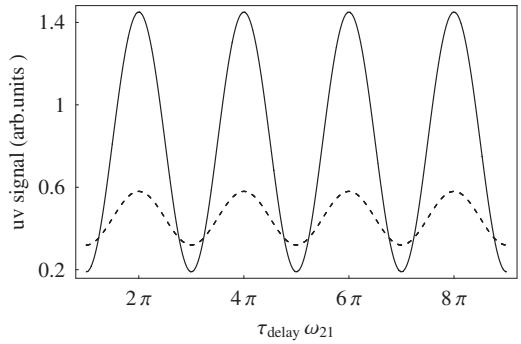

FIG. 2. uv signal obtained from Eq. (21) as a function of time delay τ_d for a model atoms having two upper eigenstates with the energy difference $\hbar\omega_{21}$ (solid). The parameters are chosen such that at the minima of $|f(t)|^2$ the condition (22) is violated, while at the maxima the exponential gain (23) takes place. The QB in $|f(t)|^2$ is shown by the dashed line.

We now return to calculations of QB in $\langle|f(\tau)|^2\rangle$ starting from the solution of Eqs. (10) and (11). We are interested in this solution for the time after probe pulse switched on. We consider first the Fourier-transform-limited laser pulses with duration $T\sim\Gamma^{-1}$. For this case the solution to Eqs. (10) and (11) is easily found in three steps. First, these equations are integrated over the time of interaction with the pump pulse, where to a good approximation the exponential factors can be disregarded because $\Delta_j T\ll 1$. Then the amplitudes $\tilde{C}_g(\tau)$ and $C_j(\tau)$ evolve freely up to $\tau\sim\tau_d$ and at the last step they are obtained from the same Eqs. (10) and (11) now describing the interaction with the probe pulse. Eventually, we find

$$\tilde{C}_g(\tau)=\cos[\theta_p(\infty)+\theta_{pr}(\tau)], \tag{24}$$

$$C_j(\tau)=i\frac{\Omega_{pr}^j}{\Omega_{pr}}\sin[\theta_p(\infty)+\theta_{pr}(\tau)], \tag{25}$$

where $\theta_{p,pr}(\tau)$ are the areas of the laser pulses

$$\theta_{p,pr}(\tau)=\int_{-\infty}^{\tau}\Omega_{p,pr}dt \tag{26}$$

with $\Omega_{p,pr}=\sqrt{\Sigma_{j=1}^N|\Omega_{p,pr}^j(t)|^2}$. Here, we have taken into account that the pump and probe pulses have the same temporal shape, so that the ratios $\Omega_p^j/\Omega_p=\Omega_{pr}^j/\Omega_{pr}$ are equal. It is easy to verify that $|C_g(\tau)|^2+\Sigma_{j=1}^m|C_j(\tau)|^2=1$. This reflects the fact that the weak generation of Stokes field does not affect the population of the upper levels.

Upon substituting Eq. (25) into Eq. (21) and taking into account that for $\tau\simeq\tau_d\gg T$ one can replace $\Delta_j\tau$ by $\Delta_j\tau_d$, $\langle|f(\tau)|^2\rangle$ can be written as

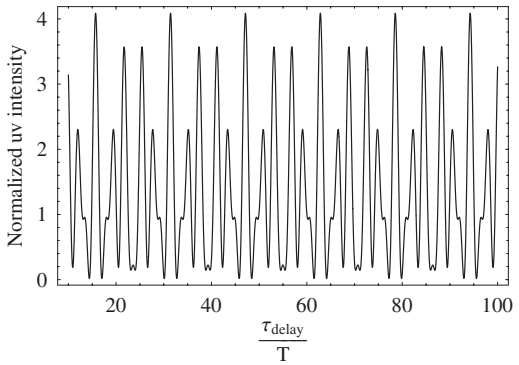

FIG. 3. QB in uv signal in the case of two-photon excitation of three upper atomic levels by Fourier-transform-limited laser pulses. All three QB modes are present.

$$\langle |f(\tau)|^2 \rangle = |f(\tau)|^2 = \sin^2[\theta_p(\infty) + \theta_{pr}(\tau)] \left(\sum_{j=1}^{m} \mu_{js}^2 \frac{|\Omega_{pr}^j(\tau)|^2}{\Omega_{pr}^2(\tau)} + 2\,\mathrm{Re} \sum_{j<k}^{m} \mu_{js}\mu_{ks} \frac{\Omega_{pr}^j(\tau)\Omega_{pr}^{k*}(\tau)}{\Omega_{pr}^2(\tau)} \exp(i\omega_{kj}\tau_d) \right). \tag{27}$$

We observe that $|f(t)|^2$ contains all possible beating modes with frequencies ω_{kj} that produce a very complicated QB pattern in the uv intensity after the time integration in Eq. (21). As an example, we show in Fig. 3 the QB in the uv signal in the case of excitation of three upper levels of the atoms, which differs substantially from the simple picture reported in the experiment with Rb atoms [9], where four atomic states in the upper manifold get populated by two-photon excitation. This discrepancy emerges because the laser phase fluctuations, which are typical for ordinary ultrashort lasers, essentially influence the QB, washing out the contribution of many beating modes.

To solve the problem allowing for laser field fluctuations we have to employ the density matrix approach instead of using Eqs. (10)–(12), which are no longer applicable. From Eq. (16), in terms of the density matrix elements $\rho_{jk}=C_jC_k^*$ averaged over the laser phase fluctuations, $\langle |f(\tau)|^2 \rangle$ is represented as

$$\langle |f(\tau)|^2 \rangle = \sum_{j=1}^{m} \mu_{js}^2 \langle \rho_{jj}(\tau) \rangle + 2\,\mathrm{Re} \sum_{j<k}^{m} \mu_{js}\mu_{ks} \langle \rho_{jk}(\tau) \rangle \exp(i\omega_{kj}\tau). \tag{28}$$

Again, the second term in Eq. (28) being proportional to the atomic coherence ρ_{jk} between the upper levels gives rise to QB. The equations for ρ_{jk} are derived from Eqs. (10) and (11) and have the form

$$\dot{\rho}_{jj} = 2\,\mathrm{Im}[(\Omega_p^{j*} + \Omega_{pr}^{j*})\rho_{jg}e^{-i\Delta_j\tau}], \tag{29}$$

$$\dot{\rho}_{jg} = i(\Omega_p^j + \Omega_{pr}^j)e^{i\Delta_j\tau} - i\sum_{k\neq j}^{m} (\Omega_p^k + \Omega_{pr}^k)\rho_{jk}e^{i\Delta_k\tau}, \tag{30}$$

$$\dot{\rho}_{jk} = i(\Omega_p^j + \Omega_{pr}^j)\rho_{gk}e^{i\Delta_j\tau} - i(\Omega_p^{k*} + \Omega_{pr}^{k*})\rho_{jg}e^{-i\Delta_k\tau}, \tag{31}$$

with $\rho_{jg}=C_j\tilde{C}_g^*$, $\rho_{jj} \ll \rho_{gg} \sim 1$. We separate the phase ϕ of the pump and probe lasers

$$\mathcal{E}_{p,pr}(z,t) = |\mathcal{E}_{p,pr}(z,t)|e^{i\phi}, \quad \Omega_{p,pr} = G_{p,pr}e^{2i\phi},$$

and transform ρ_{jg} to new variables

$$\rho_{jg} = \sigma_{jg}e^{2i\phi}. \tag{32}$$

The phase in an ordinary laser undergoes free diffusion according to the equation of motion [18]

$$\dot{\phi}(t) = \varphi(t), \tag{33}$$

where $\varphi(t)$ is a δ-correlated Langevin-noise operator,

$$\langle \varphi(t)\varphi(t') \rangle = \Gamma\delta(t-t') \tag{34}$$

with diffusion coefficient given by the laser linewidth Γ, which is much larger than the inverse pulse duration, $\Gamma T \gg 1$; it is also assumed again that $\Gamma > \omega_{m1}$.

Then Eq. (30) is modified to

$$\dot{\sigma}_{jg} = -2i\varphi\sigma_{jg} + i(G_p^j + G_{pr}^j)e^{i\Delta_j\tau} - i\sum_{k\neq j}^{m} (G_p^k + G_{pr}^k)\rho_{jk}e^{i\Delta_k\tau}. \tag{35}$$

The mean value of the first term on the right-hand side of this equation can be obtained by its formal integration and substituting the correlation function Eq. (34). We obtain

$$\begin{aligned} i\langle \varphi(\tau)\sigma_{jg}(\tau) \rangle &= -2\int_{-\infty}^{\tau} d\tau' \langle \varphi(\tau)\varphi(\tau')\sigma_{jg}(\tau') \rangle \\ &\simeq -2\int_{\infty}^{\tau} d\tau' \langle \varphi(\tau)\varphi(\tau') \rangle \langle \sigma_{jg}(\tau') \rangle \\ &= -\Gamma\langle \sigma_{jg}(\tau) \rangle. \end{aligned} \tag{36}$$

Thus the equation for $\langle \sigma_{jg}(\tau) \rangle$ is reduced to

$$\langle \dot{\sigma}_{jg}(\tau) \rangle = -2\Gamma\langle \sigma_{jg}(\tau) \rangle + i(G_p^j + G_{pr}^j)e^{i\Delta_j\tau} - i\sum_{k\neq j}^{m} (G_p^k + G_{pr}^k)\langle \rho_{jk} \rangle e^{i\Delta_k\tau}. \tag{37}$$

Using $\Gamma T \gg 1$, we can neglect the time derivative on the left-hand side of Eq. (37) and obtain

$$\langle \sigma_{jg}(\tau) \rangle = \frac{i}{2\Gamma} \left((G_p^j + G_{pr}^j)e^{i\Delta_j\tau} - \sum_{k\neq j}^{m} (G_p^k + G_{pr}^k)\langle \rho_{jk} \rangle e^{i\Delta_k\tau} \right). \tag{38}$$

Substituting $\langle \sigma_{jg}(\tau) \rangle$ into Eqs. (29) and (31), we derive equations for the mean values of the populations $\langle \rho_{jj}(\tau) \rangle$ and atomic coherence $\langle \rho_{jk}(\tau) \rangle$ in first order of the small parameter $G_{p,pr}/\Gamma$,

$$\langle \dot{\rho}_{jj}(\tau) \rangle = \frac{1}{\Gamma}(G_p^j + G_{pr}^j)^2, \tag{39}$$

$$\langle \dot{\rho}_{jk}(\tau)\rangle = \frac{1}{\Gamma}(G_p^j + G_{pr}^j)(G_p^k + G_{pr}^k)e^{i\omega_{jk}\tau}. \tag{40}$$

Equation (40) is very important, as it demonstrates that the atomic coherence ρ_{jk} differs from zero only between the upper states j and k, whose frequency splitting satisfies

$$\omega_{jk}T \leqslant 1. \tag{41}$$

In the opposite case, the right-hand side of Eq. (40) undergoes rapid exponential oscillations that nullify $\langle \rho_{jk}(\tau)\rangle$ after integration of Eqs. (40) over the interaction time with the laser pulses. No such suppression could occur in the previous case of Fourier-transform-limited pulses, for which $\omega_{jk}T < \omega_{m1}T < 1$.

Thus, the solution for populations $\langle \rho_{jj}(\tau)\rangle$ and nonvanishing atomic coherence $\langle \rho_{jk}(\tau)\rangle$ is readily found from Eqs. (39) and (40) as

$$\langle \rho_{jj}(\tau)\rangle = \frac{1}{\Gamma}[\eta_{p,jj}(\infty) + \eta_{pr,jj}(\tau)], \tag{42}$$

$$\langle \rho_{jk}(\tau)\rangle = \frac{1}{\Gamma}[\eta_{p,jk}(\infty) + \eta_{pr,jk}(\tau)], \tag{43}$$

with

$$\eta_{\alpha,jk}(\tau) = \int_{-\infty}^{\tau} d\tau' G_\alpha^j(\tau')G_\alpha^k(\tau'), \quad \alpha = p, pr. \tag{44}$$

Upon substituting this solution into Eq. (28), we finally obtain for $\tau \sim \tau_d \gg T$

$$\langle |f(\tau)|^2\rangle = \frac{1}{\Gamma}\left(\sum_{j=1}^{m} \mu_{js}^2[\eta_{p,jj}(\infty) + \eta_{pr,jj}(\tau)] + 2\sum_{j<k}^{m}{}' \mu_{js}\mu_{ks}[\eta_{p,jk}(\infty) + \eta_{pr,jk}(\tau)]\cos(i\omega_{kj}\tau_d)\right), \tag{45}$$

where the notation $\Sigma_{j<k}^{m'}$ indicates that the summation is running over the states for which the condition (41) is satisfied and, thereby, in Eq. (28) one may replace $\exp(i\omega_{jk}\tau)$ by $\exp(i\omega_{jk}\tau_d)$. The final step is the time integration in Eq. (21), which is performed numerically for the given shapes of laser pulses.

IV. COMPARISON WITH EXPERIMENT: DISCUSSION

Now, with the solution (45), we are able to calculate with the help of Eq. (21) the uv signal for a realistic situation and to compare the results with the existing data. The sample is chosen to be Rb vapor with the ground state $5S_{1/2}$, from which four upper states $9d$, $11s$, $10d$, and $12s$ are simultaneously excited by broadband laser pulses with light wavelength $\lambda \simeq 620$ nm and linewidth $\Gamma/2\pi c \sim 500$ cm^{-1}. The state $5P_{3/2}$ serves as an intermediate one giving the main contribution to the two-photon excitation, while the uv generation may occur at different $nP \rightarrow 5S(n=7-11)$ transitions. Note, however, that the $11P \rightarrow 5S$ transition is well suited for comparison with the experiment, since precisely in this case the "cleanest" signal has been achieved in Rb vapor [9]. Using the parameters pulse duration $T \simeq 100$ fs, peak intensity of Gaussian laser pulses $I_L \sim 10$ GW cm^{-2}, and atomic number density $N \sim 10^{17}$ cm^{-3}, we have calculated the intensity of the uv emission near the $11P_{3/2} \rightarrow 5S_{1/2}$ ($\lambda_{UV} \sim 311$ nm) line as a function of the time delay τ_d between the pump and probe laser pulses depicted in Fig. 4. This picture reproduces very well the main features of the observed QB pattern. First, in accordance with our calculations only two beating modes with frequencies $\omega(12s\text{-}10d)/2\pi c \sim 67$ cm^{-1} and $\omega(11s\text{-}9d)/2\pi c = 95$ cm^{-1}, which satisfy the condition (41), must be taken into account in Eq. (45). This prediction is confirmed by the experiment. Second, the oscillations in the uv signal have a periodicity $T_{os} \sim 350$ fs, which coincides with the measured value and corresponds well to the frequency $\omega(11s\text{-}9d)$. It is worth noting that this result demonstrates both the validity and correctness of our theory. The point is that one would intuitively expect that the mode $\omega(12s\text{-}10d)$ must prevail in the QB because the contribution of atomic coherence $\rho_{11s,9d}$ in $\langle |f(\tau)|^2\rangle$ [Eq. (45)] is suppressed compared to that of $\rho_{12s,10d}$ by a factor ~ 1.5, which is merely the ratio of the corresponding dipole matrix elements. However, just due to the term $\rho_{11s,9d}$ the minima of $\langle |f(\tau)|^2\rangle$ are reduced so much (note that the two contributions $\rho_{11s,9d}$ and $\rho_{12s,10d}$ have the same sign) that the gain condition (22) is violated, resulting in vanishing dark fringes with periodicity T_{os} in the uv signal (see Fig. 4). Thus, the destructive interference between HRS and FWM not only affects the visibility of the fringe pattern, but also determines the prominent mode in the uv spectrum to be $\omega(11s\text{-}9d)$ instead of $\omega(12s\text{-}10d)$, in full agreement with the experimental observations. Note that the decay time for the coherence of the upper levels due to the dephasing of atoms amounts to tens of picoseconds. So QB can be observed, in practice, for long delay times $\tau_d \gg T_{os}$.

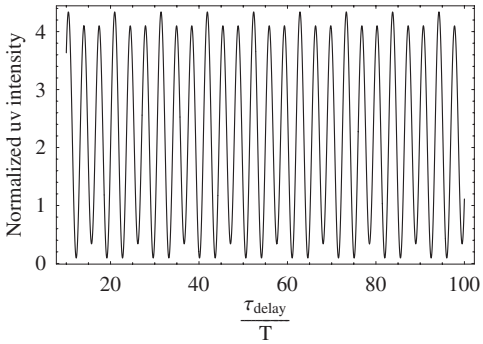

FIG. 4. Theoretical curve for uv signal generated near the $11P_{3/2} \rightarrow 5S_{1/2}$ line of Rb as a function of the time delay τ_d between the pump and probe laser pulses. In numerical simulations, the parameters of the experiment in Rb vapor [9] have been used (see the text).

The next question is the dependence of the uv signal on the laser intensity I_L. As numerical calculations show, the parameters used in the experiment [9] are such that even at the maxima of $\langle |f(\tau)|^2\rangle$ the exponential gain (23) is unattainable, so that to a good approximation the peak uv intensity can be represented in a simple form, $I_{UV}^{max} \sim \int d\tau' |f(\tau')|^2$,

showing, along with Eqs. (44) and (45), its quadratic variation with I_L, which has been detected experimentally.

Many other questions are beyond the scope of this paper and will be addressed elsewhere. We wish, however, to point out an important role of the plasma being created in the medium by a two-photon resonant three-photon ionization. The quasielectrostatic field of the plasma can strongly mix the atomic s and p (or d and p) states, which opens a new way for uv generation again via FWM, where the plasma field plays the same role as the Stokes field in the previous case. This possibility can be tested experimentally by monitoring the appearance of new modes in the Fourier spectrum of the QB corresponding to the s-p or d-p frequency differences. We foresee a new interference effect between the two FWM processes. What dominates in the uv generation depends evidently on the strength of the plasma field. Moreover, as the latter varies sinusoidally in time with the frequency ω_{pl} of plasma oscillations, we predict a periodic transformation of the QB modes from the $(s$-$d)$ to the $(s$-$p)$ modes and back. Clearly, because $\omega_{pl} \ll \omega_{s\text{-}d}$ this exchange between the modes will take place on a much larger time scale compared to that of quantum beating.

Another factor influencing the QB properties is the polarization of the laser fields. In our model, it was assumed that both the pump and probe fields have the same polarization. In the case of different polarizations, they excite different magnetic sublevels of the upper states, which can significantly change the QB pattern in the uv signal. This dependence on the polarization can be used for selective spectroscopic studies of the upper-lying levels, in particular, for the measurement of the oscillator strengths of the corresponding transitions. Of particular interest is the coherent violation of the superposition of the upper states owing to interaction with some control field connecting one of these states with an auxiliary level. In the presence of the control field, the QB, obviously, is strongly modified and even absent under certain conditions, which provides the possibility of controlling the coherent excitation of highly excited atomic and molecular levels and, in other cases, ensuring their selective excitation by femtosecond laser pulses.

V. CONCLUSIONS

In this paper, we have developed the theory of QB in uv generation via FWM in pump-probe experiments. We have presented a detailed analytical study of the system. Comparison of the results with experimental data demonstrated the ability of our theory to reveal the key features of the QB observed in Rb vapor. One conclusion that emerges from this study is that the visibility of beating fringes increases remarkably due to destructive interference between the HRS and FWM, which, in addition, determines the dominant mode of the QB. Another finding is that the laser phase fluctuations strongly limit the number of QB modes. With the assistance of the destructive interference effect, this allows one to create and study the atomic coherence between selected upper levels of the atoms or molecules, which otherwise is very difficult, if not impossible. Subsequent presentations will discuss these questions; the influence of the plasma and the effect of laser field polarization will also be analyzed and the results of detailed numerical simulations will be presented.

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APPENDIX

In this appendix we obtain the solution to Eqs. (13)–(15) with the initial and boundary conditions $\tilde{C}_s(z,-\infty)=0$, $\mathcal{E}_S(0,\tau)=\mathcal{E}_S(\tau)$, and $\mathcal{E}_{\mathrm{uv}}(0,\tau)=0$. We introduce the function $U(z,\tau)=\int_0^z \tilde{C}_s(z',\tau)dz'$. Then from Eq. (15) one obtains

$$\frac{\partial^2 U(z,\tau)}{\partial z \partial \tau} = \frac{i}{\hbar} g(\tau)\mathcal{E}_{uv}(z,\tau) + \frac{i}{\hbar} f(\tau)\mathcal{E}_S^*(z,\tau). \tag{A1}$$

The Maxwell equations (13) and (14) are integrated to the form

$$\mathcal{E}_S(z,\tau) = 2i\pi N \frac{\omega_S}{c} f(\tau)U^*(z,\tau) + \mathcal{E}_S(\tau), \tag{A2}$$

$$\mathcal{E}_{uv}(z,\tau) = 2i\pi N \frac{\omega_{uv}}{c} g^*(\tau)U(z,\tau). \tag{A3}$$

Substitution of these solutions into Eq. (A1) yields

$$\frac{\partial^2 U}{\partial z\, \partial \tau} = \frac{2\pi N}{c\hbar}(\omega_S|f|^2 - \omega_{uv}|g|^2)U + \frac{i}{\hbar} f(\tau)\mathcal{E}_S^*(\tau).$$

With the initial values $U(z,-\infty)=U(0,\tau)=0$, this equation has the solution

$$U(z,\tau) = 2z\int_{-\infty}^{\tau} F(\tau')I_1(\psi)\psi^{-1}(\tau',\tau)d\tau' \tag{A4}$$

where

$$F(\tau) = \frac{i}{\hbar} f(\tau)\mathcal{E}_S^*(\tau),$$

$$\psi(\tau',\tau) = 2\left(\frac{2\pi N}{c\hbar} z\int_{\tau'}^{\tau} (\omega_S|f|^2 - \omega_{uv}|g|^2)d\tau''\right)^{1/2}$$

and $I_1(\psi)$ is the modified Bessel function of the first order.

[1] H. Rabitz, R. de Vivie-Riedle, M. Motzkus, and K. L. Kompa, Science **288**, 824 (2000).
[2] J. L. Herek *et al.*, Nature (London) **417**, 533 (2002).
[3] G. Alber and P. Zoller, Phys. Rep. **199**, 231 (1991).
[4] R. R. Jones, C. S. Raman, D. W. Schumacher, and P. H. Bucksbaum, Phys. Rev. Lett. **71**, 2575 (1993).
[5] J. A. Gallas, G. Leuchs, H. Walther, and H. Figger, Adv. At. Mol. Phys. **20**, 413 (1985).
[6] A. ten Wolde, L. D. Noordam, A. Lagendijk, and H. B. van Linden van den Heuvell, Phys. Rev. Lett. **61**, 2099 (1988).
[7] J. A. Yeazell, M. Mallalieu, J. Parker, and C. R. Stroud, Phys. Rev. A **40**, 5040 (1989).
[8] L. D. Noordam, H. Stapelfeldt, D. I. Duncan, and T. F. Gallagher, Phys. Rev. Lett. **68**, 1496 (1992).
[9] H. C. Tran, P. C. John, J. Gao, and J. G. Eden, Opt. Lett. **23**, 70 (1998).
[10] D. Felinto, L. H. Acioli, and S. S. Vienna, Opt. Lett. **25**, 917 (2000).
[11] A. A. Senin *et al.*, Chem. Phys. Lett. **381**, 53 (2003).
[12] Yu. P. Malakyan, Sov. J. Quantum Electron. **15**, 905 (1985).
[13] Z. H. Lu *et al.*, IEEE J. Sel. Top. Quantum Electron. **10**, 159 (2004).
[14] C.-J. Zhu, Y. Xiao, A. A. Senin, J. Gao, J. G. Eden, T. S. Varzhapetyan, and D. H. Sarkisyan, Phys. Rev. A **75**, 053405 (2007).
[15] A. Gogyan and Yu. Malakyan, Opt. Spectrosc. **101**, 751 (2006).
[16] Yu. P. Malakyan, Opt. Commun. **69**, 315 (1989).
[17] Q. H. F. Vrehen and H. M. Hikspoors, Opt. Commun. **18**, 113 (1976); D. Cotter, D. Hanna, W. Tuttlebee, and M. Yuratich, *ibid.* **22**, 190 (1977); M. A. Moore, W. R. Garrett, and M. G. Payne, *ibid.* **68**, 310 (1988); M.-H. Lu and Yu mei Liu, Appl. Phys. B **57**, 167 (1993).
[18] W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973).