Bichromatic field propagation in a resonant medium: Floquet analysis

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Abstract. We study the propagation of a bichromatic field in a resonant medium. The two modes of the incoming fields are pulsed and delayed with respect to each other. It is shown that in the course of the propagation, new Raman sidebands will be generated. The achievable frequency spacing between the sidebands is determined from experimental data. A numerical example is shown for realistic physical parameters.

1. Introduction

Resonant nonlinear optics (RNLO) is an excellent concept to control the electric susceptibility of a medium by light. In RNLO the light fields propagating in a medium are resonant or nearly resonant with some atomic transitions. The atoms of the medium not only mediate the interaction between the field modes, but form a compound quantum system with the photons, hence a previously unimaginable range of phenomena emerge. In the heart of this new class of processes there is the quantum coherence and quantum interference, which can be used to control the propagation of light pulses in the underlying media. Moreover, the efficiencies of the processes are increased dramatically compared to the conventional nonlinear optical processes with far off-resonant fields. The pioneer work in the domain of RNLO was electromagnetically induced transparency (EIT), which relies on the destructive quantum interference between two atomic transition paths: an initially absorbing medium becomes transparent for a weak probe field due to the interaction with a strong control field [1, 2, 3]. Based on EIT a number of interesting and important applications have been developed, our present work is related to nonlinear field generation [4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14], and pulse propagation in multistate systems such as in a double-lambda [15, 16]. For recent review on the EIT process and related applications see [17].

The first experiments have been realized in hot/cold gases, and in BEC. However, doped single crystals proved to be as good as or even superior medium to gases for RNLO applications. The doping atoms play the role of the active media, which are embedded into some host material. The host material can be either a glass or a crystalline structure. As for the doping atoms, one of the most often used group of elements are the rare-earths. Their relevant properties are the following: the atomic density in the host crystal may reach $10^{18}$ – $10^{20}$ cm$^{-3}$; the radiative lifetime of the atomic levels is of the order of a milli-second, while the homogeneous linewidth
is in the kHz range; the inhomogeneous linewidth may be several GHz; the power of the control lasers should be chosen much smaller than in gases because of the low damage threshold of the host media. However, by means of sophisticated pumping techniques, a subset of doping atoms with the same transition frequency can be selected, and excellent experimental results can be obtained, e.g. photon storage times greater than a second [18]. Several RNLO processes have already been successfully realized in rare-earth doped single crystals, such as EIT [19]; coherent Raman beats [20]; slow down of light pulses in a solid [21, 22, 23]; quantum storage of light pulses [18, 21, 24].

In a series of works [25, 26, 27], Raman-sideband generation has been developed and experimentally realized, where a strong, bichromatic field interacted in an off-resonant Raman-configuration with the atoms of the medium. As a result, several Raman-sidebands have been generated, leading to a sequence of short laser pulses.

In this paper we consider the propagation of a strong, bichromatic laser pulse which interacts resonantly with the ground and excited states of the atoms of a medium. The dynamics of the light-atom interaction is studied with the aid of the Floquet-analysis. The condition of Raman-sideband generation is presented for delayed incoming pulses. The derived Maxwell-Scrodinger equations are solved numerically and an example is shown for Raman-sideband generation in a system with realistic physical parameters.

The paper is organized as follows: in Section 2 we present the model system and derive the equations which govern the time evolution of the atoms and the propagation of the fields. The summary of the Floquet analysis of the atomic dynamics is given in Section 3. The propagation of the field modes is discussed in Section 4, where a numerical example is shown as well. The results are summarized in Section 5.

2. The model system

We consider the propagation of an initially bichromatic field interacting with an ensemble of effective two-state atoms. The coupling configuration is shown in figure 1. It is assumed that the field components are nearly resonant with the atomic transition. Furthermore, the fields are pulsed, with pulse length much shorter than the energy and coherence relaxation times of the atoms. Incoherent line broadening is not taken into account. Under these circumstances, the time evolution of the atoms is described by the Schrödinger equation

\[ \frac{i\hbar}{\partial t} \langle \psi | = H | \psi \rangle, \]

where the Hamiltonian using the rotating wave approximation with respect to the atomic transition frequency \( \omega_{eg} = (E_e - E_g)/\hbar \), is given by

\[ H = \hbar \omega_{eg} |e\rangle\langle e| - \frac{1}{2} (d_{eg} E^{(+)}(t, z) |e\rangle \langle g| + H.c.), \]

where H.c. denotes the Hermitian-conjugate of the previous term, \( d_{eg} = \langle e | d | g \rangle \) is the matrix element of the dipole operator between the excited and ground states, and \( E^{(+)}(t, z) \) denotes the positive-frequency part of the electric component of the applied oscillating electromagnetic field, \( E^{(-)} = (E^{(+)})^* \). Initially, the field consists of two frequency components

\[ E^{(+)}(t, z) = \sum_{l=-1,0} \mathcal{E}_l(t, z) e^{i(k_l z - \omega_l t)}, \]

where \( \omega_l \) and \( k_l = \omega_l / c_n \) are the carrier frequency and the wave number of the field components, respectively. Here \( c_n \) is the speed of light in the medium \( c_n = c_{vac} / n \), with \( n \) being the refractive
index of the host medium. We ignore the small variation of \( n \) which results from dispersion. In vacuum \( n = 1 \), whereas in a crystal \( n \) is the refractive index of the crystal without the atoms participating in the RNLO process. The slowly varying envelope of the field components is denoted by \( E_l(t,z) \).

The field is fed at \( z = 0 \) into the medium. Then it propagates a distance \( Z \), while it interacts nearly resonantly with the atoms comprising the medium. The wave propagation is described by the equation

\[
\sum_l k_l \left( \frac{\partial}{\partial z} + \frac{1}{c_n} \frac{\partial}{\partial t} \right) E_l(t,z) e^{i(k_l z - \omega_l t)} = -i \frac{\mu_0}{2} \frac{\partial^2}{\partial t^2} P^{(+)}(t,z),
\]

which is valid in the slowly varying envelope approximation [28], i.e. \( |\partial^2 E_l(z,t)/\partial z^2| \ll |k_l\partial E_l(z,t)/\partial z| \) and \( |\partial^2 E_l(z,t)/\partial t^2| \ll |\omega_l\partial E_l(z,t)/\partial t| \). Here and in the following we exploit the relation \( \left[ \partial/\partial z + (1/c_n)\partial/\partial t \right] \exp(i[k_lz - \omega_l t]) = 0 \). The positive frequency part \( P^{(+)}(t,z) \) of the microscopic polarization is given by

\[
P^{(+)}(t,z) = 2N d_{ge} \varrho_{eg}(t,z),
\]

where \( N \) is the density of the atoms participating in the process, \( \varrho_{eg}(t,z) \) is the coherence between the excited and ground states of the atoms.

We assume that the polarization of the medium can be expanded into a harmonic series

\[
P^{(+)}(t,z) = \sum_l P_l(t,z)e^{i(k_l z - \omega_l t)}.
\]

Then the time-derivative in the r.h.s. of equation (4) can be easily evaluated. In leading order in \( \omega_l \) one finds

\[
\sum_l k_l \left( \frac{\partial}{\partial z} + \frac{1}{c_n} \frac{\partial}{\partial t} \right) E_l(t,z) e^{i(k_l z - \omega_l t)} = i \sum_l k_l^2 P_l(t,z)e^{i(k_l z - \omega_l t)} \frac{2\varepsilon_0 \varepsilon_r}{\varepsilon_r},
\]

where \( \varepsilon_r = n^2 \) is the relative permittivity. In order to obtain a simpler expression we introduce the frequency differences \( \Delta_l \) through the relation

\[
\omega_l = \omega - \Delta_l,
\]
It turns to be convenient to introduce a retarded frame \( \tau = t - z/c_n \) and \( \zeta = z \). Then in equations (10) the exponents are transformed as \( \exp(i\Delta_l(t - z/c_n)) \rightarrow \exp(i[\Delta_l\tau + \theta_l]) \), where \( \theta_l \) defines an initial phase. Hence for frozen field amplitudes the Hamiltonian in equation (14) is time periodic with two frequencies, the two field-components has a beat frequency (frequency difference) \( \delta = \omega_0 - \omega_{-1} = \Delta_{-1} - \Delta_0 \). Later we will see that useful results can be obtained when the beat frequency has the same magnitude as the Rabi frequencies of the input fields, \( \delta \approx \max_l \{ |\Omega_l(t, z)| \} \ll \omega_{eg}, \) \( (l = 0, -1) \), where the Rabi frequencies are defined as \( \Omega_l(t, z) = -d_{eg}E_l(t, z)/\hbar \). In such a case it is natural to introduce the strong-field dressed Hamiltonian (the so called Floquet or quasienergy Hamiltonian) [29]. Following the derivation in [30], we treat the initial phases \( \theta_l \) as dynamical variables acting on the Hilbert space of 2\( \pi \)
periodic functions $L_2(\theta_l/2\pi)$. In the rotating wave approximation, an effective reduced Hilbert space $K_{\text{eff}}$ can be defined as a tensor product of the Hilbert space of the atoms $\mathcal{H}_0 \equiv \mathbb{C}^2$ and the Hilbert space associated with the classical fields, $K_{\text{eff}} = \mathcal{H}_\lambda \otimes L_2(\eta/2\pi)$, where $\eta = \theta_0 - \theta_{-1}$. The effective Floquet Hamiltonian $K_R$ acting on $K_{\text{eff}}$ is given by

$$K_R = -i\hbar\delta \frac{\partial}{\partial \eta} + \frac{\hbar}{2} \begin{pmatrix}
0 & \sum_{l=-1,0} \Omega_l e^{i\eta} \\
\sum_{l=-1,0} \Omega_l e^{-i\eta} & 2\Delta
\end{pmatrix},$$

where we choose $\omega = \omega_0$ for the reference frequency, corresponding to $\Delta = \Delta_0$. This effective model is valid only if the two frequencies $\omega_0$ and $\omega_{-1}$ are different. The derivative term represents the relative number of photon pairs, one $\omega_0$ photon minus one $\omega_{-1}$ photon. Thus the absorption of one “effective photon” of frequency $\delta$ corresponds to the absorption of one photon of frequency $\omega_0$ and emission of an other photon of frequency $\omega_{-1}$.

The basic properties of the effective Hamiltonian of equation (15) can be understood by determining numerically its quasienergy (eigenenergy) surfaces as a function of $\Omega_{-1}$ and $\Omega_0$. A section of the hierarchy of the computed quasienergy surfaces is shown in figure 2. There are two families of periodic eigenvalues and eigenvectors for frozen $\Omega_{-1}$ and $\Omega_0$: $\lambda_{n;k,-k} = \lambda_{n,0,0}(\hat{\Omega}) + \hbar k\delta$, and $|n;\hat{\Omega},k,-k\rangle = |n;\hat{\Omega},0,0\rangle \exp(i\eta)$, $(n = e, g)$ with $k \in \mathbb{Z}$, and $\hat{\Omega} = [\Omega_{-1},\Omega_0]$. The label $n = e, g$ indicates that the dressed eigenstate families converge to either the state $|g;\hat{0},k,-k\rangle \equiv |g\rangle \exp(i\eta)$ or $|e;\hat{0},k,-k\rangle \equiv |e\rangle \exp(i\eta)$ in the limit of $\hat{\Omega} \to \hat{0}$. Any two neighboring surfaces have points of contact that are conical intersections. All intersections are located either along the line $\Omega_{-1} = 0$ or $\Omega_0 = 0$, corresponding to the situation where only one of the laser fields is interacting with the atom.

For time dependent Rabi-frequencies $\hat{\Omega}(t)$ the system evolves along a trajectory in the quasienergy space. Let us assume that the system is prepared initially in the state $|g;\hat{0},0,0\rangle$. If $\hat{\Omega}(t)$ is a smooth, slowly varying function of time, then the system evolves adiabatically, which is manifested in the quasienergy space as a trajectory which lays in the quasienergy surfaces. For sufficiently large amplitude of one of the components of $\hat{\Omega}(t)$ while the other one is kept zero, the trajectory will cross the intersections of the quasienergy surfaces. In figure 2, we draw a trajectory which results from the interaction with two delayed, sine-square shaped pulses. The peak Rabi frequencies are so large that the system crosses two intersections and evolves to the state $|g;\hat{0},2,-2\rangle$. Using the Landau-Zener analysis it can be shown that the intersections of the quasienergy surfaces are true crossings, i.e. in the adiabatic limit the system evolves from one quasienergy surface to the other one. As a result, the system may traverse several quasienergy surfaces leading to the absorption/emission of several ”effective photons”.

If adiabaticity is violated in the course of the dynamics then the trajectory in the quasienergy space will not lay in the quasienergy surfaces. Consequently, the state vector of the system is not a single eigenstate of the effective Hamiltonian, rather a superposition of several eigenstates.

4. Propagation of the bichromatic field

In order to consider the propagation of an incoming bichromatic pulse in a medium consisting of an ensemble of two-state atoms, we have to evaluate the induced polarization of the atoms defined in equation (5). We have shown in the previous section that when the interaction between a bichromatic light pulse and the atoms is in the strong-field regime, the atomic dynamics can be described conveniently in an effective Floquet space. The position-dependence of the atomic excitation is taken into account by assigning position-dependence to the atomic state vector. The state vector in the retarded frame is defined as

$$|\psi(\tau,\zeta)\rangle_R = \sum_k (c_{g,k}(\tau,\zeta)|g\rangle + c_{e,k}(\tau,\zeta)|e\rangle) e^{ik\eta},$$

(16)
Figure 2. (color online) Quasienergy surfaces of the effective Floquet-Hamiltonian. The trajectory starts from the state $|g;0,0,0\rangle$ and terminates in the state $|g;0,2,−2\rangle$.

in the Floquet space, where the state vector is expanded in the basis $\{|n\rangle \exp(i\eta)\}_{n=g,e; k \in \mathbb{Z}}$. It is transformed back to the atomic Hilbert space by replacing the variable $\eta$ with $\delta t + \eta_0$, the resulting state is denoted by $|\psi(\tau,\zeta)\rangle$. Then the required $\rho_{eg}(\tau,\zeta)$ is given by

$$
\rho_{eg}(\tau,\zeta) = \langle e|\psi(\tau,\zeta)\rangle \langle \psi(\tau,\zeta)|g\rangle = \sum_{l} \left( \sum_{k} c_{e;k}(\tau,\zeta)c_{g;k+l}(\tau,\zeta)^* \right) e^{-i\delta \tau} \equiv \sum_{l} \rho_{eg}^{(l)}(\tau,\zeta) e^{-i\delta \tau}.
$$

(17)

For smooth probability amplitudes $c_{e;k}(\tau,\zeta)$ and $c_{g;l}(\tau,\zeta)$, the coherence $\rho_{eg}(\tau,\zeta)$ has discrete frequency components with amplitudes $\rho_{eg}^{(l)}(\tau,\zeta)$, that we call partial coherences, which are separated by $\delta$. The probability amplitudes are smooth if the characteristic time of their variation is much longer than $\delta^{-1}$. Inserting equation (17) into equation (5), we can observe that the polarization induced by a bichromatic field may involve several higher harmonics of the fundamental frequency $\delta$. In fact, the partial coherence $\rho_{eg}^{(l)}(\tau,\zeta)$ in equation (17) is proportional to the polarization component $P_l$ of equation (10b). Therefore, in the course of the propagation of the bichromatic field, new field components may be generated, which are higher harmonics of the fundamental frequency $\delta$. The generated field is of the form of equation (10a), where the summation for $l$ goes from minus infinity to plus infinity in principle. Then we have to change the effective Floquet Hamiltonian in equation (15) since due to propagation, there emerge several
field components with frequency separation of \( \delta \), which interact with the atoms.

Let us consider a multimode field given by

\[
E(\theta + \omega t) = \sum_{l=-L}^{L} \mathcal{E}_l \cos(\omega_l t + \theta_l),
\]

(18)

where \( \omega_l = \omega_0 + l\delta \), \( l \in [-L, L] \), the limits for the summation \(-L, L\) are chosen for later convenience, they can be arbitrary. The analogous Hamiltonian to the one in equation (2) for a two-level system interacting with the multimode field of equation (18), reads

\[
H(\theta + \omega t) = \begin{bmatrix} 0 & V(\theta + \omega t)^* \frac{\hbar}{\omega_{eg}} \\ V(\theta + \omega t) \frac{\hbar}{\omega_{eg}} & 0 \end{bmatrix},
\]

(19)

where

\[
V(\theta + \omega t) = -d_{eg} E(\theta + \omega t),
\]

(20)

where the rotating wave approximation has not been applied yet. The Floquet Hamiltonian associated with the Hamiltonian of equation (19) is given by

\[
K = -i\hbar \omega_0 \frac{\partial}{\partial \theta} + \begin{bmatrix} 0 & V(\theta)^* \frac{\hbar}{\omega_{eg}} \\ V(\theta) \frac{\hbar}{\omega_{eg}} & 0 \end{bmatrix}.
\]

(21)

We define a rotating wave coordinate system by means of the unitary matrix

\[
R = \begin{bmatrix} 1 & 0 \\ 0 & e^{-i\theta_0} \end{bmatrix}.
\]

(22)

Then the transformed Floquet Hamiltonian \( R^\dagger K(\theta) R \) takes the form

\[
K_R = -i\hbar \omega_0 \frac{\partial}{\partial \theta} + \begin{bmatrix} 0 & V(\theta)^* e^{-i\theta_0} \frac{\hbar}{\omega_{eg} - \omega_0} \\ V(\theta) e^{i\theta_0} \frac{\hbar}{\omega_{eg} - \omega_0} & 0 \end{bmatrix}.
\]

(23)

The coupling terms are given by

\[
V(\theta) e^{i\theta_0} = \frac{1}{2} d_{eg} \sum_{l=-L}^{L} \mathcal{E}_l \left( e^{i(\theta_l + \theta_0)} + e^{i\eta_l} \right),
\]

(24)

where \( \eta_l = \theta_0 - \theta_l \). In terms of the variables \( \{\eta_l\} \) the rotating wave multimode Floquet Hamiltonian equation (23) is given by

\[
K_R = -i\hbar \omega_0 \frac{\partial}{\partial \eta_a} - i\hbar \delta \sum_l (-l) \frac{\partial}{\partial \eta_l} + \frac{\hbar}{2} \left[ \begin{array}{cc} 0 & \sum_l \mathcal{E}_l e^{-i\eta_l} \\ \sum_l \mathcal{E}_l e^{i\eta_l} & 2\Delta \end{array} \right],
\]

(25)

where \( \eta_a = \theta_0, \mathcal{E}_l = -d_{eg} \mathcal{E}_l / \hbar, \) and \( \Delta = \omega_{eg} - \omega_0 \). The first derivative term commutes with all the other terms, producing only a global phase and can thus be omitted. The second one looks like a directional derivative: the gradient \( \partial_{\eta_l} \) is taken in the direction \(-l\). We can define the new variable \( \eta \) through the relation

\[
\frac{\partial}{\partial \eta} = -\sum_l l \frac{\partial}{\partial \eta_l}.
\]

(25)
The derivative of $\eta$ with respect to $\eta$ is given by
\[
\frac{\partial}{\partial \eta} \eta_l = - \sum_j j \frac{\partial}{\partial \eta_j} \eta_l = -l .
\] (26)

It follows that
\[
\eta_l = -l \cdot \eta + c_l,
\] (27)

where $c_l$ are irrelevant constants, hence can be omitted. Then the rotating wave Floquet Hamiltonian in the RWA reads
\[
K_R = - i \hbar \frac{\partial}{\partial \tau} + \frac{\hbar}{2} \begin{bmatrix} 0 & \sum \Omega_l^* e^{i \eta_l} \\ \sum \Omega_l e^{-i \eta_l} & 2 \Delta \end{bmatrix}.
\] (28)

This Hamiltonian has exactly the same form as that of equation (15), the only difference is that now the summation goes from $-L$ to $L$. The state vector of equation (16) is equivalent with $\vec{c}(\tau, \zeta) = [c_g(\tau, \zeta), c_e(\tau, \zeta)]^T$ in the basis $\{|n\} \exp(ik\eta)\}_{n=g,e,k \in \mathbb{Z}}$. The equation of motion for $\vec{c}(\tau, \zeta)$ is given by
\[
i \hbar \frac{\partial}{\partial \tau} \vec{c}(\tau, \zeta) = K(\tau, \zeta)\vec{c}(\tau, \zeta),
\] (29)

where $K(\tau, \zeta)$ is the matrix representation of $K_R(\eta)$ in equation (28).

It follows from equations (5), (6), (10b), and (17) that the polarization components $P_l(\tau, \zeta)$ are given by
\[
P_l(\tau, \zeta) = 2N d_{ge} \phi_{eg}^{(l)}(\tau, \zeta).
\] (30)

Using this result, the Maxwell equation (11) can be decomposed as
\[
\frac{\partial}{\partial \zeta} \Omega_l(\tau, \zeta) = -i \alpha \phi_{eg}^{(l)}(\tau, \zeta),
\] (31)

where $\alpha = N k |d_{eg}|^2 / \hbar \varepsilon_0 \varepsilon_r$. The propagation of a bichromatic pulse is obtained by solving simultaneously the equations (29) and (31). If the partial coherences $\phi_{eg}^{(l)}(\tau, \zeta)$ on the rhs of equation (31) has an appreciable value for $l \neq -1,0$, then new field modes will be generated. Initially the atoms are in the ground state $c_g(0, \zeta) = 1$, all other amplitudes $c_{g,k}(0, \zeta)$ are zero. Therefore, for a large value of the partial coherence $\phi_{eg}^{(l)}(\tau, \zeta)$ it is necessary that the products $c_{e,k}(\tau, \zeta)c_{g,k-l}(\tau, \zeta)$, $l \neq 0$ become non-zero, and interfere constructively. Our purpose is to show that it is possible to generate new field modes in this setup, we made several numerical experiments to point this out.

Initially only the Rabi frequencies $\Omega_l(\tau, 0)$ associated with the field modes $\mathcal{E}_{-1}(\tau, 0)$ and $\mathcal{E}_0(\tau, 0)$ are non-zero. At the input of the medium the envelope of the Rabi frequencies are chosen as
\[
\begin{align*}
\Omega_{-1}(\tau, 0) &= \Omega_{-1} \sin^2 \left( \frac{\pi (\tau - \tau_{12})}{T} \right) , \quad (32a) \\
\Omega_0(\tau, 0) &= \Omega_0 \sin^2 \left( \frac{\pi \tau}{T} \right) , \quad (32b)
\end{align*}
\]

where $\tau_{12}$ describes a time delay between the two peaks of the two frequency components. In our numerical examples we choose the parameters of the praseodymium doped crystal $Y_2SiO_5:Pr^{3+}$. The dipole moment for the strongest transition $^{3}H_4(\pm \frac{5}{2}) \leftrightarrow {}^{1}D_2(\pm \frac{5}{2})$ of the $Pr^{3+}$ ions is $d_{eg} = 2.6 \times 10^{-26} C \mu m$. The population relaxation time is $T_1 = 165 \mu s$, whereas the coherence
relaxation time is $T_2 = 111\mu s$, [31]. We ignore hyperfine splitting and inhomogeneous broadening in our model calculations, and treat the $\text{Pr}^{3+}$ ions as ideal two-state systems. In figure 2 we used the pulse-shapes of equation (32) to compute the trajectory. In order to have two crossings in the course of the evolution, we have chosen $\Omega_l \equiv \max_t\{\Omega_l(t)\} = 2.5\delta$, $(l = -1,0)$ for the input fields. As for the other parameters, we have used $N = 5 \times 10^{18}\text{cm}^{-3}$, $\lambda_{\text{vac}} = 0.605\mu m$, and $n = 1.8$, $\Delta = -\delta/2$.

To determine the possible range of the peak input field Rabi frequencies $\Omega_l$, $(l = -1, 0)$, and $\delta$ we have to take into account the following factors: $\Omega_l$ should be a few times larger than $\delta$ in order to have some crossings in the quasienergy space. The upper bound of $\Omega_l (\mathcal{E}_l)$ is determined by the damage threshold of the medium and the available laser power. We use $\Omega_l = 25\text{MHz} (l = -1,0)$ in our model calculations, which are achievable values in $\text{Y}_2\text{SiO}_5:\text{Pr}^{3+}$. We select $\delta = 10\text{MHz}$ for the beat-frequency. The pulse-length $T$ should be so large that the product $N = \delta \times T$ is of the order of a few hundreds in order to have adiabatic evolution [30]. Adiabaticity ensures that the time of the transition under consideration, $T \ll T_2$. From these considerations it follows that a coherent evolution requires that the relation $N \ll \delta \times T_2$ should be fulfilled. For the parameters of our system we find that $N$ could be a few hundreds, in agreement with the condition for adiabaticity we have seen earlier. In practice it proved to be a reasonable choice for the time delay $\tau_{12}$ between the two components of the incoming bichromatic field the one-third of the sine-square pulse length, $\tau_{12} = T/3$. In the numerical simulations we chose $T = 21\mu s$, so that the total duration of the excitation process was $28\mu s$ at the input of the medium.

There is only one free parameter left: the length $Z$ of the sample. In the numerical evaluation of equation (31) it is useful to introduce the phase-shift $\xi = \alpha\zeta$. In this way the equation becomes more universal. In turn, the length of the sample should also be scaled, hence the domain of integration is given by $\xi \in [0, \alpha Z]$. We conclude that the propagation of the bichromatic field leads to the same result if we vary together $\alpha$ and $Z$ in such a way that $\alpha Z = \text{const}$. For the parameters given above $\alpha = 2.08855 \times 10^5\text{cm}^{-1}$. We choose $Z = 0.25\text{cm}$ as the length of the sample.

We have solved numerically the pair of equations (29) and (31) using the parameters defined above. In figure 3 we have plotted the normalized Rabi frequencies $|\Omega_l(\tau, Z)/\Omega_l|$, $(l = -8 \ldots 7)$ as a function of the scaled time $\delta \times \tau$. It can be seen that the initial modes become significantly distorted and new modes are generated. The zeroth order mode is switched on at $\delta \times \tau = 0$. This pulse has suffered some delay. The fringes starting around $\delta \times \tau \approx 40$ results from Rabi oscillations between the states $|g;\Omega, 0, -0\rangle$ and $|e;\Omega, 0, -0\rangle$. The second incoming pulse is switched on at $\delta \times \tau = 70$. After this time instant, new field modes, Raman sidebands are generated as a result of the strong modulation of the coherence $\rho_{eg}(\tau, Z)$. The spectrum of the total outgoing field is shown in figure 4, whereas the time evolution of the coherence $\rho_{eg}(\tau, \zeta)$ is shown in figure 5. These latter two plots are obtained from a direct time domain simulation, i.e. by solving the equations (11) and (13) in the retarded frame. It can be seen that due to the reshaping of the pulses (see figure 3), a small amount of coherence is left in the system at $\zeta = Z$, which means that the atoms continue to radiate. The emitted field is damped, the damping rate is determined by the $T_1$ and $T_2$ times of the atoms.

5. Summary
We have studied the propagation of a pair of bichromatic pulses in a resonant medium. We have shown using the Floquet-technique, that if the two incoming pulses are delayed and the Rabi-frequencies are a few times larger than the beat frequency between the incoming modes, then new Raman sidebands will be generated in the course of the propagation. The coherence time of the atomic system limits the timescale of the process, which in turn determines the maximal
Figure 3. (color online) The time evolution of the normalized Rabi frequencies $|\Omega_l(\tau, Z)/\Omega_0|$, as a function of the scaled time $\delta \times \tau$. For reference, in the top-left panel we plot the initial envelopes of $\Omega_{-1}(\tau, 0)$ and $\Omega_0(\tau, 0)$ with dash-dot line.
beat frequency between the incoming pulses, $T_2 \delta \gg T \delta = \text{few hundred}$. In the numerical example we have chosen $\delta = 10 \text{MHz}$, however, the previous relation suggests that all results remain valid for scaled total time $T/q$ and beat frequency $\delta \times q$. Therefore, if in a system the Rabi frequency can be as large as a few gigahertz, then the beat frequency can also be in the gigahertz range. As a result, the spacing between the generated Raman sidebands will be in the gigahertz domain.

**Acknowledgments**

We acknowledge the support from the French Agence Nationale de la Recherche (Project CoMoC) and from the European Marie Curie Initial Training Network Grant No. CA-ITN-214962-FASTQUAST. Z.K. is grateful for the hospitality at the University Bourgogne. Z.K. acknowledges the support of the Bolyai Program of the Hungarian Academy of Sciences.

**References**

[1] S.E. Harris, J.E. Field, and A. Imamoglu 1990 *Phys. Rev. Lett.* **64** 1107
[2] K.-J. Boller, A. Imamoglu, and S.E. Harris 1991 *Phys. Rev. A* **66** 2593
[3] M.M. Kash, V.A. Sautenkov, A.S. Zibrov, L. Hollberg, G.R. Welch, M.D. Lukin, Y. Rostovtsev, E.S. Fry, and M.O. Scully 1999 *Phys. Rev. Lett.* **82** 5229
[4] M. Jain, H. Xia, G.Y. Yin, A.J. Merriam, and S.E. Harris 1996 *Phys. Rev. Lett.* **77** 4326
[5] S.E. Harris and M. Jain 1997 *Opt. Lett.* **22** 636
[6] W. Harshawardhan and G.S. Agarwal 1998 *Phys. Rev. A* **58** 598
[7] L. Deng, M.G. Payne, and R.W. Garrett 1998 *Phys. Rev. A* **58**, 707
[8] E. Paspalakis, N.J. Kylstra, and P.L. Knight 2002 *Phys. Rev. A* **65** 053808
[9] E. Paspalakis and Z. Kis 2002 *Phys. Rev. A* **66** 025802
[10] E. Paspalakis and Z. Kis 2002 *Optics Letters* **27** 1836
[11] Z. Kis and E. Paspalakis 2003 *Phys. Rev. A* **68** 043817

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**Figure 4.** (color online) The frequency spectrum of the total field at the output of the medium.
Figure 5. (color online) The time evolution of the atomic coherence $\rho_{eg}(\tau,\zeta)$ as a function of the scaled time $\delta \times \tau$. In the upper panel $\zeta = 0$, whereas in the lower one $\zeta = Z$. 
[12] V.A. Sautenkov, C.Y. Ye, Y.V. Rostovtsev, G.R. Welch, and M.O. Scully 2004 Phys. Rev. A 70 033406
[13] A. Eilam, A.D. Wilson-Gordon, and H. Friedmann 2006 Phys. Rev. A 73 053805
[14] A. Eilam, A.D. Wilson-Gordon, and H. Friedmann 2007 Opt. Commun. 277 186
[15] M.D. Lukin, P.R. Hemmer, M. Löffler, and M.O. Scully 1998 Phys. Rev. Lett. 81 2675
[16] E.A. Korsunsky and D.V. Kosachiov 1999 Phys. Rev. A 60 4996
[17] M. Fleischhauer, A. Imamo glu, and J.P. Marangos 2005 Rev. Mod. Phys. 77 633
[18] J.J. Longdell, E. Fraval, M.J. Sellars, and N.B. Manson 2005 Phys. Rev. Lett. 95 063601
[19] K. Ichimura, K. Yamamoto, and N. Gemma 1998 Phys. Rev. A 58 4116
[20] A. Louchet, J.S. Habib, F. Bretanaker, F. Goldfarb, I. Lorgeré, and J.-L. Le Gouët 2007 J. Luminescence 127 89
[21] A.V. Turukhin, V.S. Sudarshanam, M.S. Shahriar, J.A. Musser, B.S. Ham, and P.R. Hemmer 2002 Phys. Rev. Lett. 88 023602
[22] E. Kuznetsova, O. Khararovsky, P. Hemmer, and M. O. Scully 2005 Phys. Rev. A 66 063802
[23] Q. Sun, Y.V. Rostovtsev, J.P. Dowling, M.O. Scully, and M.S. Zubairy 2005 Phys. Rev. A 72 031802(R)
[24] S.A. Moiseev, V.F. Tarasov, and B.S. Ham 2003 J. Opt. B: Quantum Semiclass. Opt. 5 S497
[25] S.E. Harris and A.V. Sokolov 1997 Phys. Rev. A 55 R4019
[26] S.E. Harris and A.V. Sokolov 1998 Phys. Rev. Lett. 81 2894
[27] A.V. Sokolov, D.R. Walker, D.D. Yavuz, G.Y. Yin, and S.E. Harris 2000 Phys. Rev. Lett. 85 562
[28] L. Allen and J.H. Eberly 1987 Optical Resonance and Two-Level Atoms (New York: Dower Publications, INC.)
[29] J.H. Shirley 1965 Phys. Rev. 138 B979
[30] S. Guérin, I.P. Yatsenko, and H.R. Jauslin 2001 Phys. Rev. A 63 R031403
[31] R.W. Equall, R.L. Cone, and R.M. Macfarlane 1995 Phys. Rev. B 52 3963