Enhanced four-wave mixing via elimination of inhomogeneous broadening by coherent driving of quantum transition with control fields

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Abstract

We show that atoms from wide velocity interval can be concurrently involved in Doppler-free two-photon resonant far from frequency degenerate four-wave mixing with the aid of auxiliary electromagnetic field. This gives rise to substantial enhancement of the output radiation generated in optically thick medium. Numerical illustrations addressed to typical experimental conditions are given.

PACS number(s): 42.50.Gy, 42.65.Dr, 42.65.Ky

Strong optical resonances inherent to free atoms and molecules are negated by the fact that only small fraction of the species can be concurrently resonance coupled in warm bulk gases. This is because of Maxwell distribution of the Doppler shifts of their resonances. Doppler-free (DF) coupling is usually achievable under equal frequency counter-propagating weak waves in two-photon-resonant ladder schemes. Because of the phase matching requirements, such schematic can not be implemented for far from frequency degenerated four-wave mixing (FWM). Second, the conditions of intermediate one-photon quasi-resonance can be very seldom satisfied in this case. Third, DF coupling vanishes with the growth of not only frequency difference but with strengths of the coupled fields too due to ac-Stark effect. Forth, DF coupling can not be routinely achieved in Raman schemes, like considered in this paper, because of the inherent frequency difference. Fifth, DF absorption does not indicate readily achievable DF FWM polarization in general case. Moreover, it is not obvious that increased FWM polarization would result in enhanced output of generated radiation, because of accompanying increased absorption.

This paper is aimed at demonstrating that the outlined limitations in optical physics can be removed by making use quantum coherence processes induced by an auxiliary intense control field. Considered effects lead to concurrent contribution of atoms from a wide velocity interval to the induced resonance and to eliminating it’s Doppler broadening under moderate light intensities. We investigate two-photon-resonant Raman-type FWM, controlled by an
auxiliary driving field. The explicit formulae for power-dependent absorption/gain indices and for nonlinear FWM susceptibilities, accounting for interplay of power and Doppler shifts of the resonances and illustrating the major idea of the proposed method are derived. In order to avoid accompanying population transfer, which would complicate the formulae and would mask the major effect under consideration, the field coupled to the ground state is assumed to be weak.

A possible achievement of sub-Doppler resolution using intense control field was shown in [1, 2, 3, 4, 5]. We propose a novel scheme, which enables to control FWM coupling with the aid of auxiliary electromagnetic (EM) field, taking no part in the FWM process itself. Accompanying increase of absorption of the fundamental radiation is considered too. As the outcome, substantial enhancement in quantum conversion efficiency in optically thick Doppler-broadened medium is shown. Numerical illustrations are given for the model, relevant to the FWM experiments with sodium dimer vapors [6]. As a matter of fact that detuning from the intermediate resonance are larger than the Doppler width of the transition and the populated ground level is coupled to weak fields only, none of the CPT or EIT effects, usually employed for the enhancement of resonant FWM [7], are involved in the proposed technique.

Consider FWM process $\omega_1 - \omega_2 + \omega_3^+ = \omega_S \equiv \omega_4$ and transition configuration (fig.1a), similar to those studied in the experiments [6]. However, in our case the EM radiation $E_3(t, z)$ consists of two components: weak $E_3^+(\omega_3^+)$ and counter propagating strong one $E_3^-(\omega_3^-)$. Their frequencies can be the same or different. Strong radiation $E_2$ and weak $E_1$ co-propagate in the same direction as $E_3^+$. Only lower level remains populated, because $E_1$ is assumed so weak, that the populations can not be driven. Density matrix equations in the interaction representation
are:

\[ L_{01} \rho_{01} = i \{ \rho_{00} V_{01} + \rho_{02} V_{21} \}, \]
\[ L_{03} \rho_{03} = i \{ \rho_{00} V_{03} + \rho_{02} (V_{23}^+ + V_{23}^-) \}, \]
\[ L_{02} \rho_{02} = i \{ \rho_{01} V_{12} + \rho_{03} (V_{32}^+ + V_{32}^-) \}, \]
(1) (2) (3)

where \( L_{ij} = \partial / \partial t + v \nabla + \Gamma_{ij}, V_{ij} = G_{ij} \exp \{ i (\Omega t - k z) \}, \quad V_{23}^\pm = G_{23}^\pm \exp \{ i (\Omega^\pm t \mp k^\pm z) \}, \)
\[ G_{ij} = -E_d d_{ij} / 2 \hbar, \quad G_{23}^\pm = -E_{23}^\pm d_{23} / 2 \hbar \] are coupling Rabi frequencies, \( \Omega_i \) - are corresponding resonance detunings (e.g., \( \Omega_1 = \omega_1 - \omega_{01} \)), \( \Gamma_{ij} \) - homogeneous half-widths of the transitions.

As follows from (1) - (3), induced atomic coherence \( \rho_{02} \) gives rise to the components in polarizations, responsible for novel effects in absorption and generation of the radiations under consideration. In the lowest order on the strength of the weak fields solution of the equations (1) - (3) can be found in the form:

\[ \rho_{02} = r_{02}^{(1)} \exp \{ i [(\Omega_1 - \Omega_2) t - (k_1 - k_2) z] \} + r_{03}^{(4)} \exp \{ i [(\Omega_4 - \Omega_3^+) t - (k_4 - k_3^+) z] \} + r_{02}^{(4)} \exp \{ i [(\Omega_4 - \Omega_3^-) t - (k_4 + k_3^-) z] \}, \]
\[ \rho_{03} = r_{03} \exp \{ i (\Omega_4 t - k_4 z) \} + \tilde{r}_{03} \exp \{ i (\Omega_4 t - k_4 z) \} \]
\[ \rho_{01} = r_{01} \exp \{ i (\Omega_1 t - k_1 z) \} + \tilde{r}_{01} \exp \{ i (\Omega_1 t - k_1 z) \}, \]
(4) (5) (6)

Equations for the density-matrix amplitudes become algebraic. With aid of solution for \( \tilde{r}_{03}, r_{01} \) and \( r_{03} \) expressions for the susceptibilities, dressed by the strong fields \( E_2 \) and \( E_3^- \), can be routinely obtained and presented as:

\[ \chi_4^{(3)} = -i \frac{d_{01} d_{12} d_{23} d_{30} / 8 \hbar}{P_{01} P_{02} (P_{03}^+ + |G_{23}^-|^2 / P_{02})}, \]
\[ \chi_{10} = \frac{\chi_1 (\Omega_1)}{\chi_{30}} = \frac{\Gamma_{03} P_{03} P_{02} + |G_{23}^-|^2}{P_{01} P_{03} P_{02}}, \]
\[ \chi_4 (\Omega_4)/\chi_{30} = \frac{P_{03} P_{01} (P_{02}^- + |G_{12}^-|^2 / P_{03} + |G_{12}^-|^2 / P_{01})}{P_{03}^+ P_{01} (P_{02} + |G_{23}^-|^2 / P_{03} + |G_{23}^-|^2 / P_{01})}, \]
\[ \chi_4 (\Omega_4) = \frac{\chi_1 (\Omega_1)}{\chi_{30}} = \frac{P_{03} P_{01} (P_{02}^- + |G_{12}^-|^2 / P_{03} + |G_{12}^-|^2 / P_{01})}{P_{03}^+ P_{01} (P_{02} + |G_{23}^-|^2 / P_{03} + |G_{23}^-|^2 / P_{01})}, \]
(4) (5) (6)

where \( \chi_0 \) and \( \chi_4 \) are corresponding resonant values under the strong fields being turned off, \( P_{01} = \Gamma_{01} + i (\Omega_1 - k_1 v), P_{02}^- = \Gamma_{02} + i (\Omega_1 - k_1 v), P_{03} = \Gamma_{03} + i (\Omega_4 - k_4 v), P_{03}^- = \Gamma_{03} + i (\Omega_4 - k_4 v), P_{02} = \Gamma_{02} + i (\Omega_4 - k_4 v), \)
\[ P_{02}^- = \Gamma_{02} + i (\Omega_4 - k_4 v), P_{02} = \Gamma_{02} + i (\Omega_4 - k_4 v), P_{03} = \Gamma_{03} + i (\Omega_4 - k_4 v), P_{03}^- = \Gamma_{03} + i (\Omega_4 - k_4 v), \]
\[ P_{02} = \Gamma_{02} + i (\Omega_4 - k_4 v), P_{02}^- = \Gamma_{02} + i (\Omega_4 - k_4 v), \]
\[ v \) is projection of atom velocity on \( z \). Difference between \( k_1 \) and \( \tilde{k}_1 \) as well as between \( k_4 \) and \( k_S \) is neglected here.

With account of absorption but neglecting depletion of fundamental radiations due to FWM conversion, reduced equation for \( E_4 \) can be written as:

\[ dE_4 (z) / dz = i 2 \pi k_4 \chi_4^{(3)} / E_1 (0) E_2^* E_3 \exp (-i \Delta K z), \]
(7)

where \( \Delta K = K_4 - K_1 + K_2^* - K_3^+ \), \( K_j = k_j - i \alpha_j / 2 \) - are complex wave numbers, \( \alpha_j \) - power-dependent absorption indices. Quantum conversion efficiency \( (QCE) \) of \( E_1 \) into \( E_4 \) along the medium \( \eta_4 (z) \) is given by the expression: \( \eta_4 (z) = (\omega_1 / \omega_4) |E_4 (z) / E_1 (0)|^2 \exp (-\alpha_4 z). \)

From (7) one obtains:

\[ \eta_4 = \frac{\omega_1}{\omega_4} \frac{|2 \pi \chi_4^{(3)} E_2 E_3|^2}{|\Delta K|^2} \exp (-\alpha_4 z) \exp (-i \Delta K z) - 1 |^2. \]
(8)
Pre-exponential factor can be expressed over Rabi frequencies, reduced nonlinear susceptibility and absorption indices considered below, ratios of the transition widths and $|d_{03}|^2/|d_{01}|^2$. The last factor is proportional to the ratio of the spontaneous relaxation rates. Thus $QCE$ can be found as absolute value, dependent on the optical thickness of the medium.

The major physics underlying the proposed technique is as follows. Modulation of the atomic wave-functions by the driving fields gives rise to the Autler-Townes splitting, which exhibits itself in our case as resonance shift. Besides intensities, the later depends on detunings of the driving fields and consequently – on the atomic velocities. It turns out that under appropriate intensities the resonances of atoms at different velocities can be shifted to the approximately same position. To illustrate that, consider one-photon detunings, substantially greater than corresponding Doppler HWHM. Then the resonance power-shift factors in (4) - (6) can be presented as: $|G|^2/P \approx (1 + ikv/p)|G|^2/p$, where $p$ is corresponding factor $P$ at $v = 0$. This shows possible control of the resonance Doppler shifts through the power shifts. More details can be found in [1, 5]. In the same way a factor in the denominators of (4), (5), indicating dressed two-photon resonance, can be presented as:

$$
\tilde{P}_{02} \approx \tilde{\Gamma}_{02} + i\tilde{\Omega}_{02} - i\left\{ (|G_{12}|^2/\Omega_1^2)k_1 + \left(1 + \frac{|G_{23}|^2}{(\Omega_4^2)^2}\right)(k_1 - k_2) - \frac{|G_{23}|^2}{(\Omega_4^2)^2} k_3 \right\}v,
$$

where $\tilde{\Gamma}_{02}$ and $\tilde{\Omega}_{02}$ give half-width and position of the induced resonance. As follows from (8), under proper choice of detuning, relative propagation direction and intensity of the control field $E_{3}^-$, all Doppler shifts can be compensated by the power shifts in a such way, that dependence on $v$ vanishes in the given linear on $v$ approximation. This indicates trapping of all atoms, independent of their velocities in DF dressed two-photon resonance. It is seen, that as a matter of fact that $k_2 < k_1$, elimination of Doppler broadening is not possible in the schematic under consideration with the aid of only driving field $E_{2}^-$. However it becomes possible with an auxiliary counter-propagating control field $E_{3}^-$, which does not contribute directly in FWM because of phase mismatch. The equations (4), (5) show similar behavior of absorption index and nonlinear susceptibility near induced resonance.

While approaching closer to the intermediate resonances, required intensities become lower, but relative contribution of the neglected terms, proportional to the higher orders on $k_i v/\Omega_i$, grows. This leads to decrease of the coherently coupled velocity interval.

The discussed outcomes can be illustrated with the numerical model of sodium dimer transitions [1]: $\lambda_{01} = 661$ nm, $\lambda_{12} = 746$ nm, $\lambda_{23} = 514$ nm and $\lambda_{03} = 473$ nm. Corresponding homogeneous half-widths of the transitions are 20.69, 23.08, 18.30 and 15.92 MHz, Doppler HWHM – 0.678, 0.601, 0.873 and 0.948 GHz.

Figure 2 depicts contribution of molecules at different velocities to the absorption index $\alpha_1(\omega_1) \sim Re\{\chi_1/\chi_01\}$ and to nonlinear susceptibility (trivial Maxwell envelopes are removed), while conditions of elimination of Doppler broadening are fulfilled. The figure shows potentials of coherent coupling of molecules from wide velocity interval compared with the width of the
Figure 2: Velocity distribution of the squared modulus of $FWM$ nonlinear susceptibility ((a)) and absorption index at $\omega_1$ ((c)) in Doppler-free resonance; (b) and (d) – corresponding velocity-averaged sub-Doppler resonances (scaled to the corresponding value at the frequency of control-field induced resonance but under $E_3^{-}\left(\omega_3^{-}\right) = 0$). $u$ - thermal velocity, $\Delta_1$ - Doppler HWHM of the transition 01. The insets – same functions at $E_3^{-}\left(\omega_3^{-}\right) = 0$. Detunings and intensities are the same as in fig. 1b.

Maxwell distribution, unlike the case in the absence of the control field. This gives rise to strong sub-Doppler resonances. Figure 3 shows modification of the effects while tuning closer to the intermediate resonances. Despite the growth of absorption $\alpha_1$, the proposed manipulating results in substantial increase in output of generated radiation at $\omega_S$ (fig. 1 (b) and (c)).

In conclusion, we show that substantial enhancement in nonlinear-optical response of a Doppler broaden medium can be achieved by coherent driving of quantum transitions so that molecules from wide velocity interval become trapped to one and the same dressed two-photon Doppler-free resonance. The required intensities can be decreased by tuning driving frequencies closer to one-photon resonances, while the coupled velocity interval decreases too. For ladder-type schemes, where Doppler-broadening of two-photon resonances is much larger compared to Raman-like schemes, the considered effects are even more pronounced.

The authors thank B.Wellegehausen for encouraging discussions. This work was supported
Figure 3: Velocity distribution of the squared modulus of $FWM$ nonlinear susceptibility ((a)) and absorption index at $\omega_1$ ((c)) in Doppler-free resonance; (b) and (d) – corresponding velocity-averaged sub-Doppler resonances (scaled to the corresponding value at the frequency of control-field induced resonance but under $E_{3}^{\omega_3}(\omega_3) = 0$). The insets: same profiles at $E_{3}^{\omega_3}(\omega_3) = 0$. Detunings and intensities are the same as in fig. 1c.

in part by the Krasnoyarsk Regional Science Foundation, by the Grant 97-5.2-61 in Fundamental Natural Sciences and by the Russian Foundation for Basic Research (Grant 99-02-39003).

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