Strong nonlinear regime
of resonant four-wave mixing in a gas

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Abstract

The explicit solution is obtained for four-wave mixing $\omega_4 = \omega_1 - \omega_2 + \omega_3$ of two strong fields $\vec{E}_1, \vec{E}_3$ and two weak fields $\vec{E}_2, \vec{E}_4$ in a four-level system with the large Doppler broadening. The resonance of the mixing coefficient dependence on intensity is found around $\vec{E}_1\vec{d}_1 = \vec{E}_3\vec{d}_3$, where $\vec{d}_{1,3}$ are the dipole moments of corresponding transitions. The effect is interpreted as an intersection of quasi-energy levels. Up to 6 peaks appear in the dependence of conversion coefficient on the detuning of the probe field $\vec{E}_2$. An unexpected additional pair of peaks is a consequence of averaging over velocities and disappears at low temperature. The results allow us to interpret saturation behavior in recent experiments on the mixing in sodium vapor.

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Four-level system is a promising object for resonant optics and spectroscopy owing to a
great variety of nonlinear effects. They are nonlinear interference, inversionless gain, re-
sonance refraction, electromagnetically induced transparency, optically induced energy-level
mixing and shifting, population redistribution etc (see [1,2] and citations therein). Recent ex-
periments on continuous four-wave frequency mixing of Raman type with sodium molecules
in a heat pipe [3,4] gave interesting dependencies of generated wave power on frequencies
and intensities of the incident waves. In particular, the dependence of output power on the
first strong field intensity was saturated in experiment [3] in thin media, whenever that on
the third wave intensity demonstrated the linear growth. The measurements were taken at
large Doppler broadening while the nonperturbative theory is proposed [5,6] for atoms at
rest. The saturation of four-wave mixing efficiency as a function of intensity was observed
in optically thick lead vapor interacted with megawatt pulses [7]. In case the Rabi splitting
exceeded the inhomogeneous width.

Development of nonperturbative theory from the mathematical point of view involves the
solution of the set of 16 algebraic equations for steady-state elements of atomic density matrix
for four-level system. The problem is only of analyzing the resultant awkward expression
and to average it with Maxwellian distribution over velocities. In present paper we study the
particular case of alternate two strong and two weak fields interacting with 4-level system
having some symmetry. The 4th degree equation can be reduced to biquadratic one, then
the integration is possible analytically.

A simple explicit formula for nonlinear susceptibility at zero frequency detuning displays
the resonant behavior as a function of a strong field at fixed another strong field. We
interpreted it as the coherent effect owing to the intersection of quasi-energy levels. The
susceptibility as a function of weak field frequency $\omega_2$, has 6, 4 or 3 peaks. The profile also
displays the important role of the Rabi splitting.

Let us consider the conversion of two strong incident waves $\vec{E}_{1,3}$ resonantly interacting
with opposite transitions $gl, mn$ and the weak field $\vec{E}_2$ near the resonance with transition
$gn$ into the 4-th output wave $\vec{E}_4$, inset of Fig. 1. The electric field in the cell is
\[
\vec{E}(\vec{r}, t) = \sum_{\nu=1}^{4} \vec{E}_\nu \exp \left( i \omega_\nu t - i \vec{k}_\nu \vec{r} \right),
\]
(1)
where $\vec{E}_\nu$ is the amplitude of $\nu$-th field, $\omega_\nu, \vec{k}_\nu$ are the frequency and wavevector. The strong
fields are also near resonance, $\omega_1 \simeq \omega_{gl}, \omega_3 \simeq \omega_{mn}$, where $\omega_{ij} = (E_i - E_j)/\hbar$ are transition
frequencies between energy levels $E_i$ and $E_j$. The intensity of the 4-th wave, that appears
during the process of mixing, is also being small. Its frequency and wavevector satisfy the
phase matching condition
\[
\omega_4 = \omega_1 - \omega_2 + \omega_3, \quad \vec{k}_4 = \vec{k}_1 - \vec{k}_2 + \vec{k}_3.
\]
(2)

Maxwell equation for the output wave can be reduced to
\[
\frac{d\vec{E}_4}{dx} = -\frac{2\pi i \omega_{ml} \vec{d}_{ml}}{c} \langle \rho_{ml} \rangle,
\]
(3)
where $x$ is the coordinate, $\vec{d}_{ml}$ is the matrix element of the dipole moment operator $\vec{d}$, $c$ is the
speed of light, $\rho_{ml}$ is the coherence at transition $ml$, angular brackets denote the averaging
over velocity distribution. We should calculate $\rho_{ml}$ as a function of input amplitudes $\vec{E}_{1,2,3}$, their wavevectors $\vec{k}_{1,2,3}$ and frequency detuning of the weak field.

For this end we solve the equation for Wigner’s atomic density matrix (see [8])

$$
\left( \frac{\partial}{\partial t} + \vec{v} \nabla + \gamma_{ij} \right) \rho_{ij} = q_j \delta_{ij} - i[\hat{V}, \rho]_{ij},
$$

where $\vec{v}$ is the atomic velocity, $\gamma_{ij}$ are relaxation constants, $q_j = Q_j \exp(-i\vec{v}^2/v_n^2)/v_n^3 \pi^{3/2}$ is the Maxwellian excitation function, $\hat{V} = -\vec{E}(\vec{r}, t)\vec{d}/2\hbar$ is the operator of interaction, $i, j = m, n, g, l$.

To the zeroth approximation we can neglect both the weak fields $\vec{E}_{2,4} \to 0$. The set boils down to finding out populations $\rho_j \equiv \rho_{jj}$ and coherences $\rho_1 \equiv \rho_{gl} \exp(-i\omega_1 t + i\vec{k}_1 \vec{r})$, $\rho_3 \equiv \rho_{mn} \exp(-i\omega_3 t + i\vec{k}_3 \vec{r})$ of a pair of separated two-level systems. The solution is written as

$$
\rho_1 = \frac{iG_1N_1\Gamma_{\ast}}{\Gamma_{s1}^2 + \Omega_{\nu}^2},
$$

$$
\rho_2 = \frac{N_2 - N_3}{2|G_2|^2 \gamma_1 \gamma_2} \frac{1}{\Gamma_{s1}^2 + \Omega_{\nu}^2} \rho_1,
$$

$$
\rho_3 = \frac{iG_3N_3\Gamma_{\ast}}{\Gamma_{s3}^2 + \Omega_{\nu}^2},
$$

$$
\rho_4 = \frac{N_4 - N_3}{2|G_4|^2 \gamma_3 \gamma_4} \frac{1}{\Gamma_{s3}^2 + \Omega_{\nu}^2} \rho_3,
$$

where $N_j = q_j/\gamma_{jj}, j = m, n, l, g$ are the unperturbed populations, $N_1 = N_i - N_g$, $N_3 = N_n - N_m$ are the population differences at “strong” transitions, $\gamma_1 \equiv \gamma_{gl}$, $\gamma_3 = \gamma_{mn}$ are their homogeneous width, $G_1 = \vec{E}_1\vec{d}_{gl}/2\hbar$, $G_3 = \vec{E}_3\vec{d}_{mn}/2\hbar$ are the Rabi frequencies, $\Omega_{\nu} = \Omega_\nu - \vec{k}_\nu \vec{v}$ is the Doppler-shifted detuning $\Omega_1 = \omega_1 - \omega_{gl}$, $\Omega_3 = \omega_3 - \omega_{mn}$, $\Gamma_\nu = \gamma_\nu + i\Omega_{\nu}$,

$$
\Gamma_{s1}^2 = \gamma_1^2 + 2|G_1|^2 \gamma_1 (\gamma_1^{-1} + \gamma_2^{-1}) \gamma_2,
$$

$$
\Gamma_{s3}^2 = \gamma_3^2 + 2|G_3|^2 \gamma_3 (\gamma_3^{-1} + \gamma_4^{-1}) \gamma_4
$$

are the homogeneous widths including the power broadening.

Weak fields with amplitudes $G_2 = \vec{E}_2\vec{d}_{gm}/2\hbar$, $G_4 = \vec{E}_4\vec{d}_{ml}/2\hbar$ lead to appearance of cross-coherence between levels belonging to the opposite two-level systems $\rho_2 \equiv \rho_{gm}$, $\rho_4 \equiv \rho_{ml}$ at the allowed transitions, as well as at the forbidden transitions $\rho_5 \equiv \rho_{gm}$, $\rho_6 \equiv \rho_{ml}$. To the first order one can neglect the influence of these fields to the populations. The set of 4 algebraic equations appears for the nondiagonal matrix elements:
\[ \begin{align*}
\Gamma_2 \rho_2 - iG_1 \rho_0^* + iG_3 \rho_5 &= -iG_2 (\rho_g - \rho_n), \\
\Gamma_4 \rho_4^* + iG_3^* \rho_0^* - iG_1^* \rho_5 &= iG_4^*(\rho_m - \rho_l), \\
\Gamma_5 \rho_5 - iG_1 \rho_4^* + iG_3^* \rho_2 &= iG_2 \rho_3^* - iG_4^* \rho_1, \\
\Gamma_6 \rho_6^* + iG_3 \rho_4^* - iG_1^* \rho_2 &= -iG_2 \rho_1^* + iG_4^* \rho_3.
\end{align*} \] (7)

Here \( \gamma_2 \equiv \gamma_{gn}, \gamma_4 \equiv \gamma_{ml} \) are the constants of relaxation of the coherence at the allowed transition, \( \gamma_5 \equiv \gamma_{gm}, \gamma_6 \equiv \gamma_{nl} \) are the constants for forbidden transitions, \( \Omega'_5 = \Omega'_4 - \Omega'_6, \Omega'_0 = \Omega'_1 - \Omega'_2 \) are frequency detunings at these transitions.

The solution of Eq. (7) for the nondiagonal element at output transition \( ml \) can be presented as

\[ \rho_4^* = -i\beta_4 G_1^* G_2 G_3^* - i\alpha_4 G_4^*. \] (8)

During the initial step of the mixing the generated field is small, \(|G_4| \ll |G_2|\), that enables one to neglect the absorption \( \alpha_4 \) and to find the coefficient \( \beta_4 \) only. We found intensity of output wave within the thin medium approximation by the integration of Eq. (3) from \( x = 0 \) to the length of cell \( L \)

\[ I_4(L) = \frac{2\pi^2 \omega_{ml} L}{c^2 h^3} \left| \langle \beta_4 \rangle (d_{gl}^* e_1)(d_{gn}^* e_2) \times (d_{mn}^* e_3)(d_{ml}^* e_4) \right|^2 I_1 I_2 I_3, \] (9)

where \( e_\nu \) is the polarization of \( \nu \)-th wave, \( I_\nu \) is its intensity. We find coefficient \( \beta_4 \) comparing Eq. (7) to solution of the form (8):

\[ \beta_4 = \frac{1}{D} \left( (\Gamma_5 + \Gamma_6^*) (\rho_g - \rho_n) - \frac{\rho_1^*}{iG_1} (|G_1|^2 - |G_3|^2) \right. \]

\[ \left. - \Gamma_2 \Gamma_5 - \frac{\rho_5^*}{iG_3} (|G_3|^2 - |G_1|^2 - \Gamma_2 \Gamma_6^*) \right), \] (10)

Here elements \( \rho_g, \rho_n, \rho_1, \rho_3 \) are defined by Eq. (3), (4), the determinant of set (7) is

\[ D = \Gamma_2 \Gamma_5 \Gamma_4^* \Gamma_6^* + (|G_1|^2 - |G_3|^2)^2 \]

\[ + \frac{1}{2} (|G_1|^2 + |G_3|^2)(\Gamma_2 + \Gamma_4^*)(\Gamma_5 + \Gamma_6^*) \]

\[ - \frac{1}{2} (|G_1|^2 - |G_3|^2)(\Gamma_2 - \Gamma_4^*)(\Gamma_5 - \Gamma_6^*), \] (11)

the polynomial of 4th degree in velocity. The averaging of coefficient \( \beta_4 \) over velocity is possible by the residues theory for the Doppler limit \( v_T \to \infty \).

To examine the intensity dependence of coefficient \( \beta_4 \) let us consider the case of equal relaxation constants of the levels \( \gamma_j = \gamma, j = g, l, m, n \), excitation of the lower level only \( Q_j/\gamma = N_0 \delta_{jl} \), the resonant strong field detunings \( \Omega'_1/k_1 = \Omega'_3/k_3 \ll v_T \), and equal wavenumbers of both weak fields \( |k_2 - k_4| \ll (k_2 k_4 k_5 k_6)^{1/4} \). The last condition is natural in down-conversion scheme. In view of phase matching condition (2) it is felt that the weak field detunings depend on single parameter \( \Omega: \Omega_2 = k_2 \Omega_1/k_1 + \Omega, \Omega_4 = k_4 \Omega_1/k_1 - \Omega \). Within the
assumptions one can also see that \( k_4 = k_2, k_6 = k_5 \). If all the wavevectors are parallel, then the expression for \( \langle \beta_4 \rangle \) assumes a simple form

\[
\langle \beta_4 \rangle = \frac{N}{\sqrt{\pi}v_T} e^{-\Omega_R^2/k_5^2v_T^2} \int_{-\infty}^{\infty} \frac{C(x)}{D(x)\Gamma_{s1}^2 + k_1^2x^2} dx,
\]

(12)

\[
C(x) = 4|G_1|^2iz + (\gamma - ik_1x) \times
\]

\[
[|G_1|^2 - |G_3|^2 - (\gamma - i(k_2x - \Omega)(\gamma - i(k_5x - \Omega))].
\]

Here \( x = \vec{k}_2\vec{v}/k_2 - \Omega_1/k_1, z = \Omega - i\gamma, \Gamma_{s1}^2 = \gamma^2 + 4|G_1|^2 \) is the saturated width. Determinant \( D(x) \) turns to be a function of \( x^2 \)

\[
D(x) = \kappa^4x^4 - 2\kappa^2x^2\Delta_1 + \Delta_2^2, \quad \kappa = \sqrt{k_2k_5}
\]

(13)

\[
\Delta_1 = (\mu^2/2 - 1)z^2 - |G_1|^2 + |G_3|^2, \quad \mu = k_1/\kappa \geq 2,
\]

\[
\Delta_2 = \left[z^2 - (|G_1| - |G_3|)^2\right] \left[z^2 - (|G_1| + |G_3|)^2\right].
\]

The detuning dependence of \( |\Delta_2| \) takes the minimal values at

\[
\Omega = \pm |G_1| \pm |G_3|.
\]

(14)

It is a consequence of the level splitting by the strong driving field. Note that at \( |G_1| = |G_3| \) two points of minimum merge together. The reason is equal Rabi splitting for each level.

The simple form of the determinant (13) allows calculating mixing coefficient explicitly

\[
\langle \beta_4 \rangle = \frac{\sqrt{\pi}}{\kappa v_T \Gamma_{s1}^2 + \Gamma_{s1}R\mu + \Delta_2\mu^2} \left[ \frac{\gamma + iz\mu^2}{R} \right.
\]

\[
+ \frac{4iz|G_1|^2 + \gamma(z^2 + |G_1|^2 - |G_3|^2)}{\Delta_2} \left( \frac{1}{R} + \frac{\mu}{\Gamma_{s1}} \right), \quad (15)
\]

where \( R = \sqrt{2(\Delta_2 - \Delta_1)}, \Re R > 0 \). The branch of two-valued function \( \Delta_2 \) should be chosen according to the following rules

\[
\Re \Delta_2 < 0 \text{ at } P_+ < |\Omega|, \quad \Re \Delta_2 \geq 0 \text{ at } |\Omega| \leq P_-,
\]

\[
\text{sign}(\Im \Delta_2) = \text{sign} \Omega \text{ at } P_- < |\Omega| \leq P_+,
\]

where \( P_\pm = ||G_1| \pm |G_3||. \)

The mixing coefficient \( |\langle \beta_4 \rangle|^2 \) calculated from Eq. (15) is plotted in Fig. 1 (a) as a function of detuning \( \Omega \). The coefficient has 4 peaks at points given by (14). At equal distances between quasi-energy levels \( |G_1| = |G_3| \), two central peaks coalesce in the center \( \Omega = 0 \), Fig.1 (c). Except of the zeros of \( \Delta_2 \), zeros of \( R(\Omega) \) may add two peaks near the center, Fig. 1 (b). The additional central peaks are absent for motionless atoms since only four transitions are possible between two pairs of splitted quasi-energy sublevels. These peaks are contrast at \( G_1 > G_3 \gg \gamma \) and disappear at \( |G_1/G_3| < \mu/\sqrt{\mu^2 - 1} \).

The value \( |\langle \beta_4 \rangle|^2 \) at the exact resonance \( \Omega_\nu = 0, \nu = 1, \ldots, 4 \) is shown in Fig. 2 as a function of \( |G_1|^2 \). The sharp peak at \( |G_1| = |G_3| \) confirms the qualitative interpretation
of the effect as the intersection of quasi-energy levels. Inset in Fig. 2 illustrates why the maximal conversion occurs when the Rabi splitting in opposite two-level systems are equal. Here the cross-transition from the upper sublevel of level \( m \) to the upper sublevel of level \( n \) has the same frequency as the transition between their lower sublevels. In this case only 3 resonances remain in the spectrum, Fig. 1 (c), with the overpowering maximum in the center. The resonance condition \(|G_1| = |G_3|\) brings the maximum conversion efficiency in the intensity dependence.

The splitting effect is evident from experimental results on resonant four wave mixing in Na2 [34]. The main feature is the saturation of output power as a function of one strong field. The conditions of experiment [3] are generally satisfy the above model: (1) down-conversion level scheme \( \omega_4 < \omega_3 \) (see inset, Fig. 1) with \( k_1v_T = 7.0 \cdot 10^9 \text{ s}^{-1} \), \( k_2v_T = 6.5 \cdot 10^9 \text{ s}^{-1} \), \( k_3v_T = 5.2 \cdot 10^9 \text{ s}^{-1} \), \( k_4v_T = 5.7 \cdot 10^9 \text{ s}^{-1} \); (2) all incident waves 1,2,3 are generated by external lasers; (3) the region of interaction is short enough (nearly 1 cm), the model of thin media can be treated; (4) estimated level parameters are \( N_l \sim 10^{12} \text{ cm}^{-3} \Rightarrow N_n \sim 10^{11} \text{ cm}^{-3} \Rightarrow N_g, N_m \), \( \gamma_m \simeq \gamma_l \sim 2 \cdot 10^8 \text{ s}^{-1} \), \( \gamma_n \simeq \gamma_l \sim 2 \cdot 10^7 \text{ s}^{-1} \). Slightly noncollinear geometry (mixing angle \( \theta \sim 10^{-2} \)) leads to an effective broadening \( \Delta \omega \sim kv_T \cdot \theta \sim 10^8 \text{ s}^{-1} \). Another factor is usual jitter of laser frequencies, especially for dimer and dye lasers, \( \Delta \omega \sim (2 \pm 4) \cdot 10^8 \text{ s}^{-1} \). Thus, the effective value \( \gamma = (3 \pm 6) \cdot 10^8 \text{ s}^{-1} \) seems reasonable; (5) the maximal field values estimated from the focusing geometry \(|G_1|_{\text{max}} \sim 10^9 \text{ s}^{-1} \), \(|G_2|_{\text{max}} \sim 2 \cdot 10^8 \text{ s}^{-1} \), \(|G_3|_{\text{max}} \sim 5 \cdot 10^8 \text{ s}^{-1} \) nearly correspond to the condition of two strong fields.

The resonance condition \(|G_1| = |G_3|\) may result in peaks as in dependence \( \beta_4(I_1) \), as in dependence \( \beta_4(I_3) \). If \(|G_1|_{\text{max}} > |G_3|_{\text{max}}\), the peak is seen only in \( \beta_4(I_1) \). The width of the peak is determined by the decay rate \( \gamma \). Since in the experiment \( \gamma \sim |G_3|\), the peak is wide, Fig. 2(b), and gives a smooth saturation curve \( I_4(I_1) \), Fig. 2(c). According to this consideration the saturation of \( I_4(I_1) \) in the experiment (boxes in Fig. 2) is observed at \(|G_1| > |G_3|\) and there is no saturation for \( I_4(I_3) \). Note that such behavior was observed for different values of \( I_2 \) varied by one order. Under the opposite experimental condition \(|G_1|_{\text{max}} < |G_3|_{\text{max}}\) the dependencies \( I_4(I_1) \) and \( I_4(I_3) \) change their behavior in agreement with the consideration.

Thus, the model explains quantitatively the main features of the measured saturation curves. To observe the sharp resonances arising from Rabi splitting the stabilization of laser frequencies seems to be important. To increase the efficiency of conversion into the 4th wave it is necessary to tune up the laser frequencies to corresponding peaks. The optimum at \( \Omega_v = 0 \) corresponds to equal Rabi frequencies \(|G_1| = |G_3|\).

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LIST OF CAPTIONS

Fig. 1. Conversion coefficient $|\langle \beta_4 \rangle|^2$ (arb. units) as a function of detuning $\Omega$ of the second field at $|G_1| = 1$, $|G_3| = 0.5$, $k_1v_T = 7.0$, $k_2v_T = 6.9$, $\gamma = 0.2$ (a), $\gamma = 0.02$ (b), and $\gamma = 0.02$ at $|G_1| = |G_3| = 0.5$ (c) (all frequencies are in ns$^{-1}$). Inset is the level diagram of four-level system interacting with two strong driving fields at the opposite transitions (solid arrows) and two weak fields (wavy arrows). Dotted lines show the forbidden transitions.

Fig. 2. Conversion coefficient $|\langle \beta_4 \rangle|^2$ (arb. units) vs $|G_1|^2$ at $|G_3| = 0.5$, $\Omega = 0$, $k_1v_T = 7$, $k_2v_T = 6.5$: $\gamma = 0.06$ (a), $\gamma = 0.6$ (b), and $|G_4|^2$ vs $|G_1|^2$ at $\gamma = 0.6$ (c). The parameters for (b), (c) correspond to experiment, all frequencies are in ns$^{-1}$. Boxes denote the experimental points from [3]. The inset illustrates the Rabi splitting of dressed states.
