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EFFECTS OF TWO STRONG FIELDS IN RESONANT FOUR-WAVE MIXING

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

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Four-level systems are promising objects for resonant optics and spectroscopy owing to the great variety of nonlinear effects. These include nonlinear interference, inversionless gain, resonance refraction, electromagnetically induced transparency, optically induced energy-level mixing and shifting, population redistribution, etc. (see [1, 2] and citations therein). Recent experiments on continuous resonant four-wave frequency mixing of the Raman type with sodium molecules in a heat pipe [3, 4] gave interesting behavior of the generated wave power as a function of the frequencies and intensities of the incident waves. In particular, the dependence of the output power on the intensity of the first strong field was found to saturate in an experiment on down-conversion [3], while the dependence on the intensity of the third wave exhibited linear growth. The measurements were taken at large Doppler broadening, whereas the nonperturbative analytical theory was intended [5, 6] for atoms at rest.

From the mathematical standpoint the development of a nonperturbative theory involves the solution of a set of 16 algebraic equations for the steady-state elements of the atomic density matrix for the four-level system. The problem is only to analyze the resulting awkward expression and to average this expression over a Maxwellian velocity distribution. In the present paper we study the particular case of two strong and two weak fields interacting with a four-level system having some symmetry. The fourth degree equation can be reduced to a biquadratic one, and then the integration can be done analytically. Fig.1 (*b*).

Let us consider the conversion of two strong incident waves $\mathbf{E}_{1,3}$ resonantly interacting with opposite transitions gl, mn and the weak field $\mathbf{E}_2$ near the resonance with the transition gn into the fourth output weak wave $\mathbf{E}_4$ (inset of Fig. 1). The electric field in the cell is

$$\mathbf{E}(\mathbf{r}, t) = \sum_{\nu=1}^{4} \mathbf{E}_\nu \exp\left(i\omega_\nu t - i\mathbf{k}_\nu \cdot \mathbf{r}\right), \qquad (1)$$

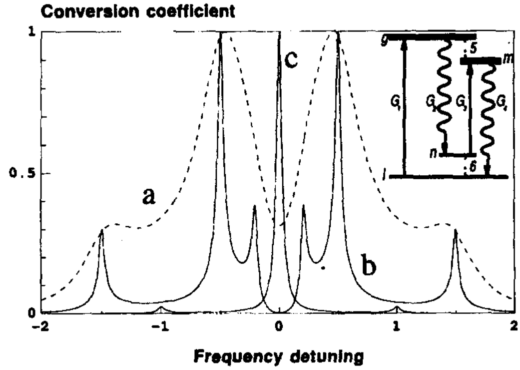

Fig. 1. Conversion coefficient $|\langle\beta_4\rangle|^2$ (arbitrary units) as a function of the detuning Ω of the second field at $|G_1| = 1$, $|G_3| = 0.5$, $k_1 v_T = 7.0$, $k_2 v_T = 6.9$, $\gamma = 0.2$ (*a*), $\gamma = 0.02$ (*b*), and $\gamma = 0.02$ at $|G_1| = |G_3| = 0.5$ (*c*) (all values are in ns^{-1}). The inset shows the level diagram of a four-level system interacting with two strong driving fields at opposite transitions (solid arrows) and with two weak fields (wavy arrows). The dotted lines show the forbidden transitions.

where $\mathbf{E}_\nu$ is the amplitude of the ν-th field, and $\omega_\nu, \mathbf{k}_\nu$ are the frequency and wave vector. The index ν numbers the transitions, $\nu = 1, 2, 3, 4$. Detunings $\Omega_1 = \omega_1 - \omega_{gl}$, $\Omega_2 = \omega_2 - \omega_{gn}$, $\Omega_3 = \omega_3 - \omega_{mn}$, $\Omega_4 = \omega_4 - \omega_{ml}$ are assumed to be small, $\omega_{ij} = (E_i - E_j)/\hbar$ are the transition frequencies between energy levels E_i and E_j. The indices $i, j = m, n, g, l$ denote the energy levels. The frequency and wave vector of fourth wave satisfy the phase-matching condition $\omega_4 = \omega_1 - \omega_2 + \omega_3$, $\mathbf{k}_4 = \mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3$.

The Maxwell equation for the output wave can be reduced to

$$\frac{d\mathbf{E}_4}{dx} = -\frac{2\pi i \omega_{ml} \mathbf{d}_{ml}}{c} \langle \rho_{ml} \rangle, \tag{2}$$

where x is the coordinate along $\mathbf{k}_4$, $\mathbf{d}_{ml}$ is the matrix element of the dipole moment operator $\hat{\mathbf{d}}$, c is the speed of light, ρ_{ml} is the coherence at the transition ml, and the angle brackets denote averaging over the velocity distribution. We should calculate ρ_{ml} as a function of the input amplitudes $\mathbf{E}_{1,2,3}$, their wave vectors $\mathbf{k}_{1,2,3}$, and the frequency detunings $\Omega_{1,2,3}$.

With this goal we solve the equation for Wigner's atomic density matrix

$$\left(\frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla + \gamma_{ij}\right) \rho_{ij} = q_j \delta_{ij} - i[\hat{V}, \hat{\rho}]_{ij}, \tag{3}$$

where $\mathbf{v}$ is the atomic velocity, γ_{ij} are relaxation constants, $q_j = Q_j \exp(-\mathbf{v}^2/v_T^2)/v_T^3 \pi^{3/2}$ is the Maxwellian excitation function, and $\hat{V} = -\mathbf{E}(\mathbf{r}, t) \cdot \hat{\mathbf{d}}/2\hbar$ is the interaction operator.

To the zeroth approximation we can neglect both the weak fields $\mathbf{E}_{2,4} \to 0$. The problem boils down to finding the populations $\rho_j \equiv \rho_{jj}$ and coherences $\rho_1 \equiv \rho_{gl} \exp(-i\Omega_1 t + i\mathbf{k}_1 \cdot \mathbf{r})$, $\rho_3 \equiv \rho_{mn} \exp(-i\Omega_3 t + i\mathbf{k}_3 \cdot \mathbf{r})$ of a pair of separated two-level systems. The solution for a two-level system under strong field is well-known (see [7]).

Weak fields with amplitudes $G_2 = \mathbf{E}_2 \cdot \mathbf{d}_{gn}/2\hbar$, $G_4 = \mathbf{E}_4 \cdot \mathbf{d}_{ml}/2\hbar$ give rise to cross-coherence between levels belonging to the opposite two-level systems at the allowed transitions, $\rho_2 \equiv \rho_{gn} \exp(-i\Omega_2 t + i\mathbf{k}_2 \cdot \mathbf{r})$, $\rho_4 \equiv \rho_{ml} \exp(-i\Omega_4 t + i\mathbf{k}_4 \cdot \mathbf{r})$, and at the forbidden transitions, $\rho_5 \equiv \rho_{gm} \exp(-i\Omega_5 t + i\mathbf{k}_5 \cdot \mathbf{r})$, $\rho_6 \equiv \rho_{nl} \exp(-i\Omega_6 t + i\mathbf{k}_6 \cdot \mathbf{r})$, where $\Omega_5 = \Omega_1 - \Omega_4$, $\Omega_6 = \Omega_1 - \Omega_2$, $\mathbf{k}_5 = \mathbf{k}_1 - \mathbf{k}_4$, $\mathbf{k}_6 = \mathbf{k}_1 - \mathbf{k}_2$. To first order one can neglect the influence of these fields on the populations ρ_j and cohereces $\rho_{1,3}$. A set of 4 algebraic equations for the off-diagonal matrix elements appears:

$$\Gamma_2 \rho_2 - iG_1 \rho_6^* + iG_3 \rho_5 \quad = \quad -iG_2(\rho_g - \rho_n),$$

$$\begin{aligned}\Gamma_4^*\rho_4^* + iG_3^*\rho_6^* - iG_1^*\rho_5 &= iG_4^*(\rho_m - \rho_l), \\ \Gamma_5\rho_5 - iG_1\rho_4^* + iG_3^*\rho_2 &= iG_2\rho_3^* - iG_4^*\rho_1, \\ \Gamma_6^*\rho_6^* + iG_3\rho_4^* - iG_1^*\rho_2 &= -iG_2\rho_1^* + iG_4^*\rho_3.\end{aligned} \tag{4}$$

Here $G_1 = \mathbf{E}_1 \cdot \mathbf{d}_{gl}/2\hbar$, $G_3 = \mathbf{E}_3 \cdot \mathbf{d}_{mn}/2\hbar$ are the Rabi frequencies, $\Gamma_\nu = \gamma_\nu + i\Omega'_\nu$, $\gamma_1 \equiv \gamma_{gl}, \gamma_3 = \gamma_{mn}$, $\gamma_2 \equiv \gamma_{gn}, \gamma_4 \equiv \gamma_{ml}$ are the constants for relaxation of the coherence at the allowed transition, $\gamma_5 \equiv \gamma_{gm}, \gamma_6 \equiv \gamma_{nl}$ are the constants for forbidden transitions, $\Omega'_\nu = \Omega_\nu - \mathbf{k}_\nu \cdot \mathbf{v}$ is the Doppler-shifted detuning.

The solution of Eq. (4) for the off-diagonal element at transition ml can be written as

$$\rho_4^* = -i\beta_4 G_1^* G_2 G_3^* - i\alpha_4 G_4^*. \tag{5}$$

In the thin-medium approximation the generated field is small, $|G_4| \ll |G_2|$, so that one may neglect the absorption α_4 and find the coefficient β_4. We found the intensity of the output wave by integrating Eq. (2) from $x = 0$ to the cell length L:

$$I_4(L) = \left|\frac{2\pi^2\omega_{ml}L}{c^2\hbar^3}\langle\beta_4\rangle(\mathbf{d}_{gl}\cdot\mathbf{e}_1)(\mathbf{d}_{gn}\cdot\mathbf{e}_2)(\mathbf{d}_{mn}\cdot\mathbf{e}_3)(\mathbf{d}_{ml}\cdot\mathbf{e}_4)\right|^2 I_1 I_2 I_3, \tag{6}$$

where $\mathbf{e}_\nu$ is the polarization of the ν-th wave, and $I_\nu = c|E_\nu|^2/8\pi$ is its intensity. We find the coefficient β_4 by comparing Eq. (4) to solution of the form (5):

$$\beta_4 = \frac{1}{D}\left((\Gamma_5 + \Gamma_6^*)(\rho_g - \rho_n) - \frac{|G_1|^2 - |G_3|^2 - \Gamma_2\Gamma_5}{iG_1^*}\rho_1^* - \right.$$

$$\left. - \frac{|G_3|^2 - |G_1|^2 - \Gamma_2\Gamma_6^*}{iG_3^*}\rho_3^*\right). \tag{7}$$

Here elements $\rho_g, \rho_n, \rho_1, \rho_3$ are solutions for separated two-level systems. The determinant D of set (4) is a polynomial of fourth degree in the velocity. The averaging of the coefficient β_4 over velocity can be done by using the residue theorem in the Doppler limit $k_\nu v_T \gg |G_\nu|, |\Omega_\nu|, \gamma$.

To examine the intensity dependence of the coefficient β_4 let us consider the case of equal relaxation constants ($\gamma_{ij} = \gamma$), excitation of the lower level only, detunings of the strong field such that $\Omega_1/k_1 = \Omega_3/k_3 \ll v_T$, and equal wave numbers of the two weak fields. The condition $k_2 = k_4$ (and therefore $k_5 = k_6$) seems realistic for the down-conversion experiment of Ref. [3], where the difference of the wave numbers of the weak fields was about 10%. One can ignore the difference $k_2 - k_4$ provided that $|k_2 - k_4| \ll (k_2k_4k_5k_6)^{1/4}$. In view of the phase-matching condition it is reasonable that the weak field detunings depend on a single parameter Ω: $\Omega_2 = k_2\Omega_1/k_1 + \Omega, \Omega_4 = k_4\Omega_1/k_1 - \Omega$. If all the wave vectors are parallel, then the expression for $\langle\beta_4\rangle$ assumes a simple form:

$$\langle\beta_4\rangle = \frac{N}{\sqrt{\pi}v_T}e^{-\Omega_1^2/k_1^2v_T^2}\int_{-\infty}^{\infty}\frac{C(y)}{D(y)}\frac{dy}{\Gamma_{s1}^2 + k_1^2y^2}, \tag{8}$$

$$C(y) = 4|G_1|^2 iz + (\gamma - ik_1y)\left[|G_1|^2 - |G_3|^2 - (\gamma - i(k_2y - \Omega)(\gamma - i(k_5y - \Omega))\right].$$

Here $y = \mathbf{k}_2\cdot\mathbf{v}/k_2 - \Omega_1/k_1$, $z = \Omega - i\gamma$, $\Gamma_{s1}^2 = \gamma^2 + 4|G_1|^2$ is the saturated width, $N = Q_l/\gamma$ is the unperturbed population. The determinant $D(y)$ turns out to be a function of y^2

$$D(y) = \kappa^4y^4 - 2\kappa^2y^2\Delta_1 + \Delta_2^2, \tag{9}$$

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

$$\Delta_1 = (\mu^2/2 - 1)z^2 - |G_1|^2 + |G_3|^2, \quad \mu = \frac{k_1}{\kappa} = \left[\frac{k_2}{k_1}\left(1 - \frac{k_2}{k_1}\right)\right]^{-1/2} > 2.$$

The limiting case $\mu \to \infty$ corresponds to a quasi-degenerate four-level system $k_{5,6} \to 0$. The opposite limit $\mu \to 2$ means $k_3 \to 0$. The detuning dependence of $|\Delta_2|$ takes its minimum values at

$$\Omega = \pm|G_1| \pm |G_3|. \tag{10}$$

This is a consequence of the level splitting by the strong driving field. Note that at $|G_1| = |G_3|$ the two minima merge together. The reason is the equal Rabi splitting for each level.

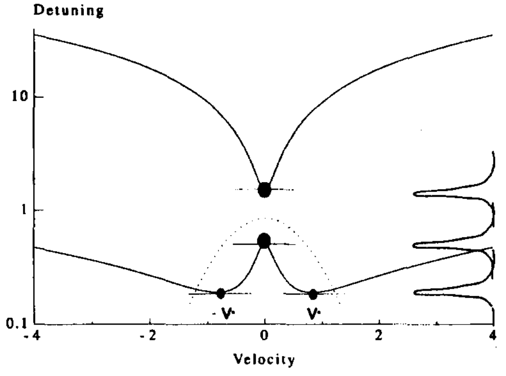

Fig. 2. Two positive solutions $\Omega^{(1,2)}(y)$ of the equation $D(y,\Omega) = 0$ as a function of y/v_T. The other two zeros are symmetric: $\Omega^{(3)} = -\Omega^{(1)}, \Omega^{(4)} = -\Omega^{(2)}$. The Maxwellian distribution is shown by the dotted line

The simple form of the determinant (9) allows calculating the mixing coefficient (8) explicitly,

$$\langle\beta_4\rangle = \frac{\sqrt{\pi}}{\kappa v_T}\frac{Ne^{-\Omega_1^2/k_1^2 v_T^2}}{\Gamma_{s1}^2 + \Gamma_{s1}R\mu + \Delta_2\mu^2}\times$$

$$\times\left[\frac{\gamma + iz\mu^2}{R} + \frac{4iz|G_1|^2 + \gamma(\mu^2 z^2/2 - \Delta_1)}{\Delta_2}\left(\frac{1}{R} + \frac{\mu}{\Gamma_{s1}}\right)\right], \tag{11}$$

where $R = \sqrt{2(\Delta_2 - \Delta_1)}$, $\Re R > 0$. The branch of the double-valued function Δ_2 should be chosen according to the following rules:

$$\Re\Delta_2 < 0 \text{ at } P_+ < |\Omega|, \quad \Re\Delta_2 \geq 0 \text{ at } |\Omega| \leq P_-, \quad \text{sign}(\Im\Delta_2) = \text{sign}\,\Omega \text{ at } P_- < |\Omega| \leq P_+,$$

where $P_\pm = ||G_1| \pm |G_3||$.

The mixing coefficient $|\langle\beta_4\rangle|^2$ calculated from Eq. (11) is plotted in Fig. 1*a* as a function of the detuning Ω. The coefficient has 4 peaks at points given by (10) as for motionless particles. At equal distances between quasi-energy levels $|G_1| = |G_3|$ the two central peaks coalesce at the center $\Omega = 0$ (Fig.1*c*). Besides the zeros of Δ_2, zeros of $R(\Omega)$ may add two peaks near the center (Fig. 1*b*) arising from averaging over velocities. To interpret two ***additional*** peaks let us plot the two positive zeros $\Omega^{(1,2)}(y)$ of D as a function of velocity y (Fig.2). The two negative zeros are located symmetrically about

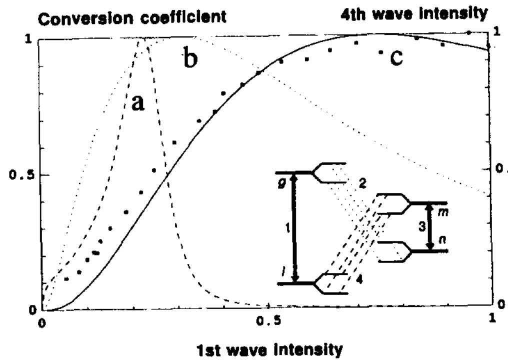

Fig. 3. Conversion coefficient $|\langle\beta_4\rangle|^2$ (arbitrary units) versus $|G_1|^2$ at $|G_3| = 0.5$, $\Omega = 0$, $k_1 v_T = 7$, $k_2 v_T = 6.5$: $\gamma = 0.06$ (a), $\gamma = 0.6$ (b); intensity I_4 (arb. units) versus $|G_1|^2$ at $\gamma = 0.6$ (c). The parameters for (b) and (c) correspond to experiment; all values are in ns^{-1}. The boxes denote the experimental points from Ref. [3]. The inset illustrates the Rabi splitting of dressed states.

the y axis. The *return points*,[1)] where the derivative $\Omega^{(1,2)\prime}(y)$ equals zero, are places of minimum variation of the eigenfrequencies. They therefore give the main contribution to the integral over velocity. The integration over each neighborhood adds one sharp peak in the spectrum, as shown schematically at the right. The two upper return points denoted by big gray circles are located at zero velocity. Two additional return points, shown by tight black circles, appear at finite velocity and correspond to the additional peak. The return points can be found analytically from the conditions

$$\frac{dz}{dy} = 0, \quad D(y,z) = 0, \quad \frac{\partial D}{\partial y} + \frac{\partial D}{\partial z}\frac{dz}{dy} = 0. \tag{12}$$

At $\gamma = 0$ this gives four solutions (10) at $y = 0$, namely, $z = \pm|G_1| \pm |G_3|$. At real $y = \pm\sqrt{\Delta_1}/\kappa$ there are two additional solutions

$$z = \pm 2\sqrt{\frac{|G_1|^2}{\mu^2} - \frac{|G_3|^2}{\mu^2 - 4}}. \tag{13}$$

The coefficient Δ_1 becomes positive at $|G_1/G_3| \geq \mu^2/(\mu^2 - 4) = k_1^2/k_3^2$; otherwise the return point vanishes, and with it the additional peak.

The value $|\langle\beta_4\rangle|^2$ at exact resonance ($\Omega_\nu = 0$, $\nu = 1, \ldots, 4$) is shown in Fig. 3 as a function of $|G_1|^2$. The sharp peak at $|G_1| = |G_3|$ confirms the qualitative interpretation of the effect as a crossing of quasi-energy levels. The inset in Fig. 3 illustrates the case where the cross-transition from the upper sublevel of level g to the upper sublevel of level n has the same frequency as the transition between their lower sublevels. Concurrently, the same resonance is achieved for transition $m - l$. In this case only 3 peaks remain in the spectrum (Fig. 1c), with a predominant maximum in the center. The crossing condition $|G_1| = |G_3|$ brings about the maximum conversion efficiency in the intensity dependence.

The splitting effect is evident from experimental results [3, 4] on resonant four-wave mixing in Na_2. The main feature is saturation of the output power as a function of one of the strong fields. The experimental conditions of Ref. [3] generally satisfy the above model: (1) a down-conversion level scheme $\omega_4 < \omega_1$ (see inset, Fig. 1) with $k_1 v_T = 7.0\ \mathrm{ns}^{-1}$, $k_2 v_T = 6.5\ \mathrm{ns}^{-1}$, $k_3 v_T = 5.2\ \mathrm{ns}^{-1}$, $k_4 v_T = 5.7\ \mathrm{ns}^{-1}$; (2) the interaction region is short enough (about 1 cm) that the model of thin media can be employed; (3) the estimated level

[1)] or "return frequencies," as they are called in the theory of three-level system with large Doppler width [8] parameters are $N_l \sim 10^{12}$ cm$^{-3} \gg N_n \sim 10^{11}$ cm$^{-3} \gg N_g, N_m$; $\gamma_m \simeq \gamma_g \sim 0.2$ ns^{-1}, $\gamma_n \simeq \gamma_l \sim 0.02$ ns^{-1}. A slightly noncollinear geometry (mixing angle $\theta \sim 10^{-2}$) leads to an effective broadening $\Delta\omega \sim kv_T \cdot \theta \sim 0.1$ ns^{-1}. Another factor is the usual jitter of laser frequencies, especially for dimer and dye lasers, $\Delta\omega \sim 0.2 \div 0.4$ ns^{-1}. Thus, the effective value $\gamma = 0.3 \div 0.6$ ns^{-1} seems reasonable; (4) the maximum field values estimated from the focusing geometry. $|G_1|^{\max} \sim 1$ ns^{-1}, $|G_2|^{\max} \sim 0.2$ ns^{-1}, $|G_3|^{\max} \sim 0.5$ ns^{-1}, nearly *correspond to the condition of two strong fields.*

The resonance condition $|G_1| = |G_3|$ may result in peaks in both $\beta_4(I_1)$ and $\beta_4(I_3)$. If $|G_1|^{\max} > |G_3|^{\max}$, the peak is seen only in $\beta_4(I_1)$. The width of the peak is determined by the decay rate γ. Since in the experiment $\gamma \sim |G_3|^{\max}$, the peak is wide (Fig. 3b) and gives a smooth saturation curve $I_4(I_1)$ (Fig. 3c) in agreement with the experimental data (boxes in Fig. 3). The same time there is no saturation for $I_4(I_3)$ both in theory and experiment. Under the opposite experimental condition, $|G_1|^{\max} < |G_3|^{\max}$ (Ref. [4]), the behavior of $I_4(I_1)$ and $I_4(I_3)$ changes.

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

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