Doppler-free spectroscopy and wave front rotation in the parametric interaction of nonmonochromatic waves

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A method is proposed for determining the natural widths of Doppler-broadened two-photon transitions by using an excitation whose spectral width greatly exceeds the Doppler width of the transition. The possibility for the effective rotation of the emission wave front with a large spectral width is shown.

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1. Two-photon transitions free of Doppler broadening are usually observed in the field of two colliding waves of identical frequencies. It is easy to show that in the general case the resonance width of two-photon absorption in nonmonochromatic fields is determined only by the spectral width of the emission, and information concerning the transition width is lost. Because of the properties of parametric nonlinear interactions in generating sum and difference frequencies in gases in a field of unidirectional nonmonochromatic waves under the condition of two-photon resonance, the transformation coefficient is inversely proportional to the product of the emission spectral width and the Doppler width of the two-photon transition.[1,2]

In recent years great interest has been drawn to possible wave front rotation for electromagnetic waves in the degenerate four-photon parametric interaction of colliding waves under conditions of two-photon resonance.[3] It will be shown in this work that in this interaction scheme the transformation coefficient is inversely proportional to the product of the spectral emission width and the natural width of the two-photon transition, and not the Doppler width. From this follows, on the one hand, the possibility for the effective rotation of the wave front in nonmonochromatic fields and, on the other hand, super-high-resolution spectroscopy using available lasers whose output is not highly monochromatic.

2. We shall discuss a three-level system (see Fig. 1). The eigen frequencies of transitions are denoted by ω_{ij}. The pumping field consists of two colliding nonmonochromatic waves E. Each wave is a set of independent modes with central frequency

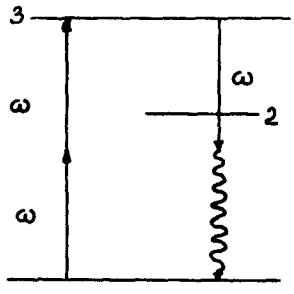

FIG. 1

that $2\omega \approx \omega_{31}$. The difference of the adjacent modes is Δ, and the spectral width is Γ_f. The mode amplitude distribution is assumed to be Gaussian

$$|E_m|^2 = |E_0|^2 \exp\{-4m^2\Delta^2/\Gamma_f^2\}. \tag{1}$$

The mode index m may assume both positive as well as negative values. We shall study the degenerate parametric process in which the third wave $\tilde{E}$ at frequency ω, which is propagated at an angle to the excitation, generates emission E_d at the difference frequency $\omega_d = 2\omega - \omega = \omega$ in a direction opposite to $\tilde{E}$. If only the ground state 1 is populated, then for calculating the nonlinear polarization in the stationary case we obtain the following system of equations for the nondiagonal elements of the density matrix

$$P_{12}^m r_{12}^m = -iG_{12}^m; \quad P_{13}^n r_{13}^n = -i\sum_m r_{12}^m G_{23}^{n-m};$$

$$P_d^l r_d^l = -i\sum_n r_{13}^n \tilde{G}_{32}^{n-l}. \tag{2}$$

Here $G_{12}^k = -d_{12}E_k/2\hbar$; $G_{23}^k = -d_{23}E_k/2\hbar$; $\tilde{G}_{32}^k = -d_{32}\tilde{E}_{\tilde{k}}/2\hbar$; d_{ij} are the matrix elements of dipole transition moment; m,n,l,k are mode indices; r_d^l and r_{ij}^k are the amplitudes of the corresponding space-time Fourier components of the nondiagonal density matrix elements in the interaction representation (for example, $\rho_{12}^m = r_{12}^m \exp\{i[(\omega + m\Delta - \omega_{21})t - \mathbf{kr}]\}$)

$$P_{12}^m = \Gamma_{12} + i(\omega + m\Delta - \omega_{21} - \mathbf{kv}) = \Gamma_{12} + i(\Omega_{12}^m - \mathbf{kv});$$

$$p_{13}^n = \Gamma_{13} + i(2\omega + n\Delta - \omega_{31}) = \Gamma_{13} + i\Omega_{13}^n; \tag{3}$$

$$P_d^l = \Gamma_{12} + i(\omega + l\Delta - \omega_{21} - \tilde{\mathbf{k}}\mathbf{v}) = \Gamma_{12} + i(\Omega^l{}_{12} - \tilde{\mathbf{k}}\mathbf{v})$$

Γ_{ij} are of the same kind (the natural or collision-broadened widths of the corresponding transitions), and $\mathbf{v}$ is speed of the atom. It is assumed in Eq. (2) that the interacting fields do not perturb the atomic system. It should be noted that the Doppler shift is absent in the equation for P_{13}.

If the detuning from the one-photon and three-photon resonance is much longer than the spectral widths of the emission and the Doppler widths of the corresponding transitions $|\omega - \omega_{21}| = |\Omega_{12}| \gg \Gamma_f \gg k\bar{v}$, then for a gas with atomic density N we obtain from Eq. (2) the following expression for the complex nonlinear polarization P_d at the frequency ω_d:

$$P_d = \frac{iN|d_{12}d_{23}|^2}{(2\hbar)^3\Omega_{12}^2}\sum_p \tilde{E}_p^* e^{-i[(\omega + p\Delta)t - \tilde{\mathbf{k}}\mathbf{r}]}\sum_{m,q}\frac{E_m E_q}{\Gamma_{13} + i\Omega_{13}^{m+2}} e^{i[2\omega + (m+q)\Delta]t}. \tag{4}$$

Here $q = n - m$ and $p = n - l$.

Therefore, the nonlinear polarization wave front and, consequently, the emission generated turns out to be rotated with respect to the wave $\tilde{E}$.

Solving Maxwell's equation in the approximation of the given excitation fields and assuming the phases of the various modes to be independent, we obtain the following results for the square of the modulus of the generated emission averaged over the mode phases.

$$\overline{|E_d|^2} \sim \frac{|d_{12}\, d_{23}|^4}{(\Omega_{12})^4} \sum_{m,q,p} \frac{|E_m|^2 |E_q|^2 |\tilde{E}_p|^2}{\Gamma_{13}^2 + (\Omega_{13}^{m+q})^2} \,. \tag{5}$$

Substituting Eq. (1) into Eq. (5), provided the value Δ is sufficiently small, and changing from the summation in terms of m, p, and q to integration of the emission over the spectrum, for $|2\omega - \omega_{31}| = |\Omega_{13}| \gg \Gamma_f$, $\Gamma_{13} \gg \Gamma_f$ we obtain

$$I_d \sim \frac{|d_{12}\, d_{23}|^4 \sqrt{2\pi}}{(\Omega_{12})^4 \Gamma_f\, \Gamma_{13}} I^2 \tilde{I} \exp\{-2\,(\Omega_{13}/\Gamma_f)^2\}, \tag{6}$$

Here, I and $\tilde{I}$ are the integrals for the corresponding waves over the intensity spectrum, for $|\Omega_{13}| \gg \Gamma_f$, we find that

$$I_d \sim \frac{|d_{12}\, d_{23}|^4 I^2 \tilde{I}}{(\Omega_{14})^2 (\Omega_{13})^2} \,. \tag{7}$$

3. Therefore, because of the compensation of the Doppler shift for the two-photon transition in the colliding waves, it follows from Eq. (6) that the intensity of the emission generated turns out to be inversely proportional to the natural and not to the Doppler width of the two-photon transition, as it would be for unidirectional waves. As a result the radiation transformation coefficient increases by 2–3 orders.

It is easily shown that the probability of the two-photon absorption of the colliding nonmonochromatic waves, which is determined by averaging the expression $\Gamma_{13}[\Gamma_{13}^2 + (\Omega_{13}^{m+q})^2]^{-1}$ over the spectrum, is independent of Γ_{13} and information about this quantity is lost.

The ratio of the experimental values (Eqs. (6) and (7)) provides a measurement of the natural widths and collisionally-broadened two-photon transitions even when the nonmonochromaticity of the emission and the Doppler broadening substantially exceed these values.

$$\frac{I_d(\Omega_{13} = 0)}{I_d(|\Omega_{13}| >> \Gamma_f)} = \sqrt{2\pi}\, \frac{\Omega_{13}^2}{\Gamma_f\, \Gamma_{13}} \,.$$

While this work was being prepared for publication, a paper[4] appeared in which a similar possibility of Doppler-free spectroscopy was discussed for monochromatic waves.

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