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STRUCTURE OF CHEMICAL COMPOUNDS. SPECTROSCOPY

Nonlinear Comb Spectroscopy

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Abstract—A numerical solution for the polarization of two-level atoms that interact with a polyharmonic field was obtained. An analytical solution for the particular case of a symmetrical position of carrier frequency relative to transition frequency is possible. The results showed that nonlinear effects occur in a polarization spectrum at the small amplitudes of comb components and small frequency distances between them. Consequently, it is necessary to take into account nonlinear effects in comb spectroscopy.

Keywords: nonlinear comb spectroscopy

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The aim of this work was to study the polarization spectrum of a two-level uniformly broadened atomic system in a weak polyharmonic light field. At present, comb spectroscopy is a vigorously developed area of spectroscopy, which makes it possible to detect atomic and molecular spectra with high sensitivity in a wide spectral range with the resolving power that depends on Doppler broadening for one-photon transitions. The emission spectrum is a set of narrow equidistant peaks (a comb spectrum). The simultaneous detection of all spectral field components is the major advantage of comb spectroscopy. Nonlinear multiphoton processes are not considered in ordinary linear comb spectroscopy. Nonlinear effects play an important role in the case that the Rabi frequencies of each of the components are lower than or commensurable with the values of the relaxation constants of levels (lines) and a frequency interval between the components. Previously [1], we found that uniformly broadened components are formed against the background of a Doppler profile in the absorption spectrum in counter comb fields due to multiphoton processes.

Let us consider a two-level atomic system under the action of a quasi-resonance polychromatic field, which consists of $2K+1$ monochromatic components:

$$E(t)=\frac{1}{2}\left(\left[E_{s0}+\sum_{m=1}^{K}E_{sm}(e^{im\Delta_s t}+e^{-im\Delta_s t})\right]e^{i\omega_{s0}t}+\text{c.c.}\right),$$

where $\omega_{s0}=(\omega_{2K+1}+\omega_1)/2$ is the center frequency of a polychromatic field, $\Delta_s=\omega_{j+1}-\omega_j$ is the frequency distance between the field components, and E_{sm} is the amplitude of the mth field component.

Equations for the density matrix of the two-level system in a quasi-resonance and rotary wave approximation take the form

$$\frac{dN_{12}}{dt}=\lambda_{12}-\gamma N_{12}-4\,\mathrm{Im}(V_{21}\rho_{12}),$$
$$\frac{d\rho_{12}}{dt}=-(\Gamma+i\delta)\rho_{12}+iV_{21}^{*}N_{12},$$

where $\lambda_{12}=\lambda_1-\lambda_2$ is the optical pumping difference to the levels; $V_{21}=-d_{21}E/\hbar$ is the matrix element of interaction energy in a dipole approximation; d_{21} is the dipole moment of the transition; $N_{12}=\rho_{11}-\rho_{22}$ is the difference of the population densities of the lower and higher states; $\delta=\omega_{s0}-\omega_{21}$ is the detuning of the medium frequency of a laser field from the transition frequency ω_{21}; and $\gamma=\gamma_1=\gamma_2$ and Γ are the longitudinal and transverse relaxation constants, which correspond to the level and line widths, respectively.

For a symmetrical case, the analytical solution of the system of differential equations is possible [2]. Polarization is determined by the nondiagonal elements of the density matrix:

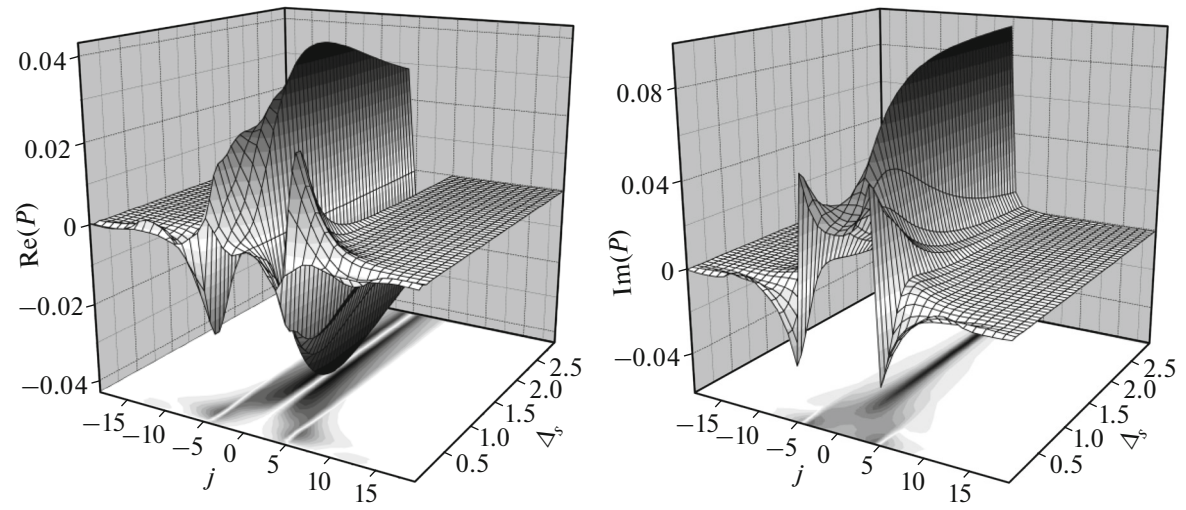
$$P(t)=d_{21}\rho_{12}(t)+\text{c.c.}$$

The polarization components P_j, oscillating at the frequencies $\omega_{sj}=\omega_{s0}\pm j\Delta_s$, are

$$P_j=-\frac{d_{21}}{2}\left\langle i\,\mathrm{Im}\,\rho_{12}e^{ij\Delta_s t}\right\rangle_t,$$

where $\langle\ldots\rangle_t$ denotes averaging over time.

After the averaging over time, the imaginary and real parts of polarization will take the form



Graphs of the dependences of the real Re(P) and imaginary Im(P) parts of polarization on the component number j and the intermode distance Δ_s. The parameters had the following values: $K = 4$, $\Omega_{s0} = \Omega_{sm} = 0.2\Gamma$, $\lambda_{12} = 1$, and $\delta = 0$.

$$\mathrm{Re}(P_j) = \frac{\gamma\lambda_{12}d_{21}}{4} \times \sum_{n_1=-\infty}^{+\infty}\cdots\sum_{n_K=-\infty}^{+\infty}\sum_{l_1=-\infty}^{+\infty}\cdots\sum_{l_K=-\infty}^{+\infty}\left(\frac{B_{n_1\ldots n_K l_1\ldots l_K}\delta_{j,f_{n_1\ldots n_K l_1\ldots l_K}}}{\gamma^2 + f_{n_1\ldots n_K}^2} - \frac{B_{n_1\ldots n_K l_1\ldots l_K}\delta_{j,-f_{n_1\ldots n_K l_1\ldots l_K}}}{\gamma^2 + f_{n_1\ldots n_K}^2}\right),$$

$$\mathrm{Im}(P_j) = -\frac{\lambda_{12}d_{21}}{4} \times \sum_{n_1=-\infty}^{+\infty}\cdots\sum_{n_K=-\infty}^{+\infty}\sum_{l_1=-\infty}^{+\infty}\cdots\sum_{l_K=-\infty}^{+\infty}\left(\frac{B_{n_1\ldots n_K l_1\ldots l_K} f_{n_1\ldots n_K}\delta_{j,f_{n_1\ldots n_K l_1\ldots l_K}}}{\gamma^2 + f_{n_1\ldots n_K}^2} + \frac{B_{n_1\ldots n_K l_1\ldots l_K} f_{n_1\ldots n_K}\delta_{j,-f_{n_1\ldots n_K l_1\ldots l_K}}}{\gamma^2 + f_{n_1\ldots n_K}^2}\right),$$

where $B_{n_1\ldots n_K l_1\ldots l_K} = \prod_{m=1}^{K} J_{n_m}(-Z_{sm})J_{l_m}(Z_{sm})$, $Z_{sm} = 2\Omega_{sm}/m\Delta_s$, $\Omega_{sm} = -\frac{d_{21}E_{sm}}{\hbar}$, $f_{n_1\ldots n_K} = \Omega_{s0} - \sum_{m=1}^{K} m n_m \Delta_s$, $f_{n_1\ldots n_K l_1\ldots l_K} = \sum_{m=1}^{K} m(n_m + l_m)$, and $\delta_{j,\pm f_{n_1\ldots n_K l_1\ldots l_K}}$ is the Kronecker symbol.

The real part of polarization determines the refractive index (dispersion), and the imaginary part, which is related to the real part by the Kramers–Kronig relation, determines the absorption coefficient. With weak fields and small intermode distances, when Δ_s is less than or in the vicinity of Ω_{sm}, the imaginary part of polarization has positive values for some harmonics in the occurrence of a laser field; consequently, these components are absorbed. However, there are components for which the imaginary part of polarization has negative values; that is, the components are strengthened. There is a strengthening at frequencies where there were no harmonics; that is, new components appear. Nonlinear processes occur with the appearance of new harmonics although weak fields are acting.

At large intermode distances, the dependence of the imaginary part of polarization on the harmonic number has an ordinary Lorentz profile for a weak field and a homogeneously broadened transition (see the figure). The real part also takes a usual anomalous form near the transition line of passage and a normal form far from it. Theoretical results are consistent with numerical calculations [3].

CONCLUSIONS

The probability of nonlinear coherent processes in the case of a polychromatic field is determined by not only the polychromatic field amplitude but also the distance between comb-field components. The phenomenon is related to the nonlinear mutual influence of weak field components located closely to each other. Multiphoton processes appear. Nonlinear effects should be taken into account in comb spectroscopy.

At certain parameters of the atom + field system, the splitting of levels into the infinite system of quasienergy sublevels and the appearance of multiphoton transitions between them would be expected.

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