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Doppler – free comb-spectroscopy in counter – propagating fields

Sergey A. Pulkin[1, 2], GuangHoonKim[2], Uk Kang[2], VasiliyArnautov[1], Svetlana V. Uvarova[1]

[1]Saint-Petersburg State University,Dept.of General Physics 1 of Physical Faculty, Saint-Petersburg, Russia
[2]Russia-Seoul Science, Seoul, Republic of Korea

Email address:
spulkin@mail.ru(S. A. Pulkin)

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Abstract: The method of Doppler – free comb – spectroscopy for dipole transitions was proposed. The numerical calculations for susceptibility spectrum for two-level system driving by strong counter propagating combs were made. The narrow peaks with homogeneous width arise on the background of Doppler counter. The contrast of these peaks is large for largest amplitudes of comb-components. Power broadening is increasing with increasing of field amplitudes. The amplitudes of peaks depend on the phase difference between carrier frequencies of combs. The spectral range of absorption spectrum is determined by the spectral range of comb generator and all homogeneous lines arise simultaneously. The spectral resolution is determined by the width of homogeneously –broadening lines. The physical nature of narrow peaks is in the existing of multi-photon transitions between manifolds of quasi-energy levels arising for different groups of atoms moving with velocities that satisfy to the resonant conditions 2kv= (n+l)Ω/j, where n, l, j-are integers and Ω - frequency difference between comb teeth.

Keywords: Doppler – Free Comb Nonlinear Spectroscopy in Counter – Propagating Comb Laser Fields

1. Introduction

The goal of present work is researching of susceptibility spectra in the strong counter-propagating polyharmonic light fields with phasing equidistant components. The frequency spectrum of that field is a spectrum of comb – generator of femtosecond laser. Generator radiates a time subsequence (train) of femtosecond pulses with repetition period of τ_p. The radiation spectrum consists of narrow equidistant peaks (comb – spectrum). Nowadays comb – spectroscopy is a fast developing area of spectroscopy [1,2] allowing to detect with high sensitivity atomic and molecular linesin the wide spectral range with resolution limited by Doppler broadening for one –photon transitions. The main advantage of comb – spectroscopy method is in the possibility to detect simultaneously all spectral lines. The method of two – photon Doppler – free comb – spectroscopy has been developed in [3,4]. The all advantages of usual two- photon spectroscopy with advantages of comb – spectroscopy method are used in this method, but the spectral resolution is determined by homogeneous broadening. In the present work we study of the mechanism of arising of narrow peaks in the absorption spectrum at the background of Doppler shape of dipole transition in one photon excitation.

The interaction of counter – propagating waves with medium of moving atoms is very important thing for our description. In the strong counter – propagating monochromatic fields multi – photon transitions already existas narrow additional peaks (dopplerons) on the background of Lamb dip [5]. Each molecule moving with velocity v “sees” not one field, but two fields, which detuned one from other on the frequency 2 kv. If one field is in resonance with transition frequency, then on the frequency detuning ω_{21} + 2 kv in the frame of moving atom coordinate the second field acts. If both fields are strong the system converts from two-level to multi – levelone consisting of manifold quasi –energy levels. The transitions between quasienergy levels are possible [6]. Two manifolds shifted on frequency 2kv arise for the case of moving atoms with velocity v. The narrow coherent peaks in the polarization and susceptibility spectrum arise additionally to known dopplerons when all fields are phased (comb – spectrum and phase control). This work is a prolongation of our previous work [7], where we considered the interaction of two counter propagating waves with two – level system – bi-harmonic (strong modulated) and weak counter – propagated monochromatic field. It was shown that in the polarization spectrum the additional narrow resonances arise on the frequencies that satisfy of the conditions of multi – photon resonances. We can explain the result of present researches using results of papers [6, 7]. The possibility of Doppler shift compensation is common feature for previous work [7] and present researches. The mechanism of Doppler shift compensation in two-photon comb – spectroscopy and present Doppler – free comb – spectroscopy is different. In the case of two-photon comb – spectroscopy one photon comes from low frequency side of comb – spectrum with frequency ω_0 - Ω and second photon with frequency $\omega_0 + \Omega$ comes from high frequency side.

The effective two – photon transition on transition frequency $\omega_{21} = 2\omega_0$ will arise for the group of atoms when the resonant condition is satisfied: kv = Ω. (Ω - is the frequency interval between comb – components). The other cause is in the case of counter – propagated combs acting near one – photon transitions. The effective interaction for multi – photon transitions between quasi – energy levels on the frequency of one – photon transition takes place for groups of atoms with velocity v, if interaction is with the frequencies ω_0 - pΩ and ω_0+nΩ. (n and p –are integers and zero) for n = - p and 2kv = nΩ.

There is partial case when the strong modulated field and counter – propagating weak probe field act on Doppler broadened medium. This case is realized in the method of modulated transfer spectroscopy (MTS) [8, 9]. The four-wave mixing in medium leads to arising of modulation of probe field on exit of cell with atoms. The order of nonlinearity is determined in this case by nonlinear susceptibility $\chi(3)$. The condition of phase synchronism is satisfied in this case. The condition of phase control and synchronism is necessary to satisfy for multi-photon interactions in our case too.

2. Theory and Results of Numerical Calculations

Criteria of strong field

The probability of coherent nonlinear processes is determined by the amplitude of acting laser field.

For pulse excitation for arising of multi – photon processes is necessary that pulse square will be of order of π. Let's estimate the intensity (density of power) for pulse duration τ_p = 100 fs and for energy ξ = 10 nJ for diameter of laser beam d = 1 mm:

$$I = 4\pi\xi/\tau_p d^2 \approx 10^{11}\,(W/_{M^2})$$

For dipole transitions in molecules the transition dipole momentum has the value $d_{12} \sim 10^{-28}$ (C * m). For that value the Rabi – frequency is equal to $\Omega_R = 10^{13}$ rad/s, and pulse square is equal:

$$Q = \int_0^{\tau} V(t) h\, dt \approx \Omega_R \tau_p \sim 1\,.$$

So, for these parameters of laser pulse the laser field is strong and multi – photon transitions in the structure of quasi –energy levels are possible.

Equations for field and density matrix equations

Let's consider ensemble of two-level atoms under the acting of two counter – propagating fields, each of them is the chain of pulses. The density matrix formalism was used for obtaining response of system. The semi classical approach and rotation wave approximation were used. The each molecule of gas is two – level system with relaxation processes. The main task of Doppler – free spectroscopy is resolution of homogeneously broadened lines on the background of wide overlapping Doppler shapes. In the known method of saturated absorption in the field of two counter propagating monochromatic waves the narrow Bennet distribution on the velocity is imposed on Maxwell distribution, that leads to arising of dip with homogeneous width in absorption on the Doppler shape. For the case of two counter – propagating comb fields there are narrow peaks at the background of Maxwell distribution. The frequency distant between these peaks is equal to frequency between comb – components.

Density matrix equations for two – level system in atom system of coordinates in the rotation wave approximation are:

$$\frac{d}{dt}\rho_{11} = 2(V_r(t)\rho_{12}^{''} - V_i(t)\rho_{12}^{'}) + \gamma\rho_{22}\,,$$

$$\frac{d}{dt}\rho_{22} = -2(V_r(t)\rho_{12}^{''} - V_i(t)\rho_{12}^{'}) - \gamma\rho_{22}\,,$$

$$\frac{d}{dt}\rho_{12}^{''} = -V_r(t)(\rho_{11} - \rho_{22}) - (\Delta + \delta)\rho_{12}^{'} - \Gamma\rho_{12}^{''}$$

$$\frac{d}{dt}\rho_{12}^{'} = V_i(t)(\rho_{11} - \rho_{22}) + (\Delta + \delta)\rho_{12}^{''} - \Gamma\rho_{12}^{'}\,,$$

whereδ – is a frequency detuning between carrier laser frequency ω and transition frequency ω_{21}; Δ = kv – shift of field frequency (in the frame of moving atom) because Doppler shift; $\gamma = \gamma_{21}$ –the transition probability on the transition 2 – 1; $\Gamma = \Gamma_{21}$ – line width.

The operator of interaction energy (perturbation operator) is equal to the sum $V = V_r + iV_i$ – real and imaginary parts of interaction energy

For two counter –propagating fields has the form for low – frequency part:

$$V = V_{0s}\sum_{m=-n}^{n}\exp(i\Omega mt) * \exp[-i2\Delta t] + V_{0p}\sum_{m=-n}^{n}\exp(i\Omega mt)$$

where Ω - frequency of repetition of pulses, m – integer , that determines the number of pulses (number of components in comb – spectrum), equal to (2n + 1);

V_{0s} and V_{0p} – are amplitudes of dipole interaction energy. The polarization of medium is equal to:

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

The polarization spectrum one can found using Fourier transform of time dependence of slow time changing part of non-diagonal element of density matrix ρ_{12}. For isotropic medium the direction of polarization vector coincides with the field direction and for susceptibility we write:

$$\chi = \frac{\mathrm{P}(w)}{E(w)} = -\frac{\mathrm{P}(w)d_{21}}{V\hbar}$$

The absorption coefficient for the central component of probe (not weak) field is equal:

$$\frac{K}{K_\Lambda} = -\frac{2\omega_{21}d_{21}}{K_\Lambda}\mathrm{Im}(\chi)$$

where

$$K_\Lambda = \frac{\omega_{21}d_{21}^2}{\hbar\gamma\Gamma_{12}}$$

is a linear absorption on the line center for absence o strong field case.

The absorption coefficient and refractive index as imaginary and real parts of susceptibility were obtained by numerical solution of density matrix equations. The one – step Runge –Cutta method of 4-rth and 5-th orders in MatLabprogram[10] was used.

Numerical simulations of interaction of atomic system with comb- spectra

Let's consider the case when carrier frequency of reference comb is in resonance with transition frequency and carrier of comb is scanning on the frequency detuning δ. The calculations are making for different groups of atoms with different projections on axis Z. The absorption coefficient for the central component of carrier frequency is shown on Fig. 1. The absorption dip with homogeneous width~ 2Γ (Γ = 0.5) on the wide background of Doppler shape takes place.

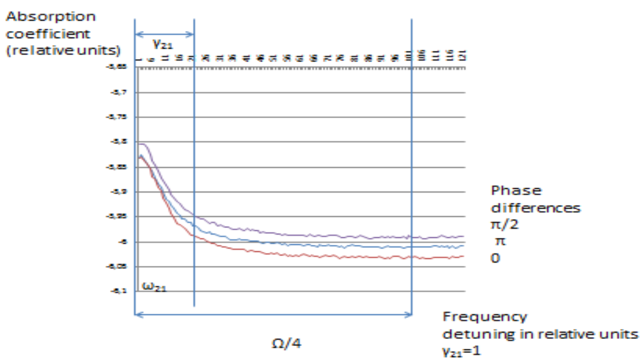

Fig. 1. *Absorption coefficient for the component of comb on carrier frequency for probe comb.(V1=V2=0.1; n=20; Ω=10).*

For obtaining of absorption coefficient moving atoms were separated into groups of atoms with different velocitiesand polarization spectra in system of the coordinates connected with each separate group were calculated. Fourier transform from that sum give us the dependence of imaginary part of susceptibility from detuning of carrier frequency – or spectrum for absorption coefficient.Dependence of absorption coefficient on the component of comb with frequency coinciding with frequency of Doppler line center$\omega_{21}$was obtained(Fig.1). The weak dependence on phase difference between counter propagating waves was found. It is because the nonlinear interaction is coherent interaction for this case. For the strong fields the field broadening of homogeneous line was found.In an absorption spectrum the dips with homogeneous width of the line widened by a strong field areshown on Fig.2. The contrast of homogeneous dip for strong fields is large than for weak fields (13% and 3% consequently for Fig.2 and Fig.1)

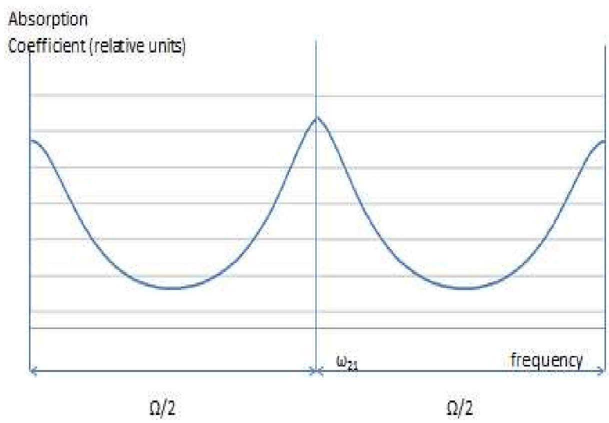

Fig.2. *Absorption coefficient for the case of strong field broadening. The same parameters as on Fig.1 except $V_1 = V_2 = 1$*

It is shown that the spectral range is defined by a spectral range of radiation of the comb-generator.

The interpretation of the results

The obtained results are possible to explain by the following way. The susceptibility and polarization spectra of two – level medium in the field of two counter – propagating waves from comb – generator of femtosecond pulses are nonlinear response of medium because multi – photon transitions between quasi – energy levels. According [6], the 2-level system is split into the on two manifolds quasi – energy components with frequency differences between them **Ω** and 2Δ.

For moving atoms and counter – propagating comb – fieldsΔ ≡ kvand the resonance condition is:

$$2kv = {}^{(n+l)\Omega}/_{j}$$

For harmonic and sub-harmonic of frequency **Ω** for these groups of atoms the resonances exist. The closest analogue of Doppler – free comb spectroscopy is a modulation transfer spectroscopy with a counter – propagating modulated pump beam and the probe beam. Due to the nonlinear medium under the influence of a strong pump field are four-wave mixing processes, in which the modulation is transferred to a probe beam. In this case, the condition of phase matching is satisfied. Doppler compensation also takes place, as in the method of saturated absorption. In the case of the gas medium driving by on opposite combs with sufficient intensity of the laser field in each pulse are also multi - wave processes because of multiphoton transitions between the two manifolds of quasi-energy levels.

The condition of compensation of Doppler shift has the form:

$$(m\Omega + 2k_m v) + (m'\Omega + 2k_{m'} v) = 0$$

Then for the transitions between quasienergy levels near resonant transition with frequency ω_{21} $k_m = -k_{m'}$; m = - m';(for counter – propagating waves) and finally, the compensation condition for groups of atoms with velocity v is:

$$2kv = l\Omega;$$

Where l=-m (0,±1, ±2, …).

More common condition one can find from [6]:

$$2kv = {(n + l)\Omega}/{j}$$

It is shown that the spectral range is defined by a spectral range of radiation of the comb-generator. The spectral range of the comb-generator can be made wide enough for simultaneous registration of a considerable quantity homogeneously - widened spectral lines. It is the big advantage of an offered method comb –spectroscopy on-comparison with existing methods Doppler - free spectroscopy.

All the components of the spectrum of a mixture of gases appear in the spectrum of polarization simultaneously. Doppler shifts which satisfy the conditions of resonance - that is, the Doppler shift must be a multiple of the intermode frequency (difference between adjacent frequency comb - spectrum) or its subharmonics. Thus, the proposed study will have all the advantages of Doppler comb - spectroscopy - high brightness, simultaneous recording in a wide spectral range of a large number of lines of various gases –the new thing is- the resolution within the Doppler contour.These resonances “drawing” the susceptibility spectrum when the carrier frequency of probe comb is scanned along the Doppler shape.

3. Conclusions

The susceptibility spectrum of medium of moving atoms driving by counter – propagating waves of strong laser field from generator of chain of femtosecond pulses were researched. It was shown that because multi – photon transitions between quasi – energy levels the waves with frequencies that satisfy of the condition of multi – photon resonance are effective absorbed. The groups of atoms with velocity and wave number that satisfy to the condition of multi-photon resonances are:

$$2kv = {(n + l)\Omega}/{j}.$$

As the result the narrow dip on the background of Doppler shape arise in the absorption spectrum for carrier frequency of probe comb.

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