Coulomb Broadening of Resonance Induced by Standing Wave

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Abstract.

Coherent preparation of quantum states of atoms and ions by laser light can lead to electromagnetically induced transparency and related effects. In particular, the standing wave at the adjacent transition induces a new type of nonlinear resonance in the probe-field spectrum of a three-level system. The resonance is due to the effect of high-order harmonics of atomic coherence. However, the resonance is broadened and the reason of the broadening was unclear. The paper describes how the velocity changing collisions broaden the resonance. The analytic theory taking into account up to 4-th order of the perturbation theory is presented. The numerical calculations are described. The analytical formulas are shown to describe the experiment qualitatively while the numerical computation demonstrates quantitative agreement. The Coulomb dephasing is discussed as the physical mechanism of the broadening.

1. Introduction

It is well-known that the light absorption in the atomic medium near a resonant frequency can be reduced substantially or even canceled with the help of strong driving field at the adjacent transition. This effect, so-called electromagnetically induced transparency (EIT) [1, 2], is a base of quite a number of atomic coherence effects, in particular, the resonant enhancement of electro-optical processes [3, 4], the lasing without inversion [5], suppression of two-photon absorption [6]. Under conditions of EIT the super-narrow resonances arise leading to the giant dispersion within the transparency window. This property is helpful for light deceleration [7–9] and makes the effect promising for application in the frequency standards [10], the precision magnetometry [11], and as a storage of quantum information [12, 13].

As a rule, in EIT experiments three-level $\Lambda$-systems interact with driving field presented by a running wave. Theoretical and experimental study of the standing wave at argon ionic lines in $\Lambda$-configuration is studied in [14,15]. Strong field was generated on laser transition $\text{ArII } mn = 4p^2S_{1/2} - 4s^2P_{1/2}$ at wavelength $\lambda = 458$ nm. Probe field was a running wave at $ml = 4p^2S_{1/2} - 3d^2P_{3/2}$, $\lambda_\mu = 648$ nm. The absorption of probe field was measured as a function of detuning $\Delta = \omega - \omega_{mn}$. At the specific probe field detuning $\Delta_\mu = \omega_\mu - \omega_{ml} = k_\mu \Delta/k$ the well known EIT resonance was observed. Along with it a new
structure at \( \Delta \mu = 0 \) was discovered and its position in the spectrum was independent of the strong field detuning \( \Delta \).

At \( \Delta = 0 \) the positions of both resonances coincide but their widths and signs differ. The new structure looks like EIT peak with a narrow dip in the centre. At higher strong field detuning \( \Delta \) the dip was transformed to a peak at the center \( \Delta \mu = 0 \). The new structure was observed for non-typical situation with a wide lower level \( n \), where the forbidden transition was wider than the allowed one \( \Gamma_{nl} \gg \Gamma_{ml} \). In the conventional physics of a three-level system the opposite limiting \( \Gamma_{nl} \ll \Gamma_{ml} \) case had been studied. For this reason the motionless structure was missed both in the perturbation theory and in numerical calculations [16]. The appearance of this structure was shown to be caused by the higher harmonics of the spatial coherence at transitions \( ml \) and \( nl \) induced by the standing wave.

At the same time the width of observed resonance occurs to be several times wider than the prediction of collisionless theory. It is naturally to assume that the broadening is a result of the velocity changing collisions. For ions in high current discharge the leading process is the Coulomb scattering [17]. In general theory of Coulomb scattering the higher harmonics of spatial coherence are neglected. The neglecting is valid for the Lamb dip in two-level system where the oscillation of populations are smoothed out after the averaging over velocities. The theory is also valid for running wave in three-level system. Calculation of the shape of known EIT resonance at \( \Delta \mu = k_\mu \Delta / k \) in the three-level system under the field of running wave demonstrates a good agreement with experiment [18]. It was shown that the resonance is caused by the Bennet dip in the population distribution over velocity. Its width becomes nearly 3 times greater than in collisionless case. For the new resonance \( \Delta \mu = 0 \) the theory taking the scattering into account was absent.

The aim of the present paper is to explain how the collision broadening occurs. The key expressions of the broadening theory of the resonance of higher spatial harmonics are presented below, while the specific details of numerical and analytical calculation can be found in papers [19,20]. The account of collisions is important not only for the quantitative interpretation of experiments in argon discharge plasma. The velocity changing collisions could broaden the resonances and decrease the slope of dispersion curve in other experiments on EIT where the standing wave is exploited.

2. Perturbation theory

Let us consider the gas of three-level systems under the driving field of standing wave. An analytical expression for the probe field absorption at high intensity of the driving field cannot be derived even for collisionless gas. Higher spatial harmonics are taken into account by the Feldman — Feld continuous fractions [16]. This expression was obtained from the known continuous fraction for populations in two-level system in bichromatic field [21,22]. In the case of equal relaxation constants and exact resonance the populations can be expressed in terms of Bessel function [23] (see also the survey by Stenholm [24]). In experiment the higher spatial harmonics were studied in cadmium spectrum. Atoms interacted with two counterpropagating waves of different frequency and amplitude [25,26] (see also [27]). Under the frequency scanning the subradiative structure was observed in the absorption spectrum that included up to 5 peaks that became more frequent in the line center (so called “1/n-resonances”). Herewith the spectrum appeared to be very sensitive to the ratio of amplitudes of the waves, especially in the case of close amplitudes.

At weak saturation the solution can be obtained as a perturbation series. The second order of the perturbation series does not include the effects of higher spatial harmonics. The second harmonics appears only in the fourth order, then we restrict ourselves by the fourth order.
2.1. Equation for density matrix

The density matrix $\rho_{ij}$ equation describes the interaction of three-level system with the parallel driving and probe waves

$$i (\Gamma_{ij} + \partial_t + v \partial_x) \rho_{ij} = [V, \rho]_{ij}. \quad (1)$$

Here $\Gamma_{ij}$ are the relaxation constants, $t$ is time, $x$ is the coordinate along the common direction of the wave vectors $k||k_\mu$ of the driving and probe waves, $v$ is the projection of the velocity vector to this common direction, $\hat{V} = -\mathbf{E} \cdot \mathbf{d}/2\hbar$ is the operator of dipole interaction, $\mathbf{d}$ is the dipole moment operator, $h$ is the Planck constant, $\mathbf{E}$ is the electric field of the light wave, indices $i, j$ possess the values $l, m, n$, square brackets denote the commutator.

We neglect the population $\rho_{mm}$ of level $m$ and polarization $\rho_{mm}$ being small under experimental conditions. The equilibrium distribution of populations are Maxwellian. Let us specify the fields of running and standing waves in the form

$$\mathbf{E}_\mu(x, t) = \frac{1}{2} \mathbf{E}_\mu^0 e^{i k_\mu x - i \omega_\mu t} + c.c., \quad \mathbf{E}(x, t) = \mathbf{E}_\mu^0 \cos k x e^{-i \omega t} + c.c., \quad (2)$$

where $\mathbf{E}_\mu^0$ and $\mathbf{E}_\mu^0$ are the amplitudes, $c.c.$ are complex conjugated terms. Denote Rabi frequencies of the driving and probe field as $\Omega \mu = \mathbf{E}_\mu^0 \cdot \mathbf{d}_{\mu ml}/2\hbar$, $\Omega = \mathbf{E}_\mu^0 \cdot \mathbf{d}_{\mu mn}/2\hbar$, where $d_{ij}$ are matrix elements of the dipole moment.

Changing the variables:

$$\rho_{ml} = \rho_\mu(x, v) \exp(i k_\mu x - i \Delta_\mu t), \quad \rho_{nl} = \rho_\nu(x, v) \exp(i k_\nu x + i \Delta - i \Delta_\mu t),$$

we get a closed steady state set for amplitudes $\rho_\mu$ and $\rho_\nu$ of polarizations:

$$(\Gamma_\mu - i \Delta_\mu + i k_\mu v + v \partial_x) \rho_\mu = -2i \Omega \cos(kx) \rho_\nu + i \Omega \nu N_l(v),$$

$$(\Gamma_\nu + i \Delta - i \Delta_\mu + i k_\mu v + v \partial_x) \rho_\nu = 2i \Omega \nu \cos(kx) \rho_\mu, \quad (3)$$

where $\Gamma_\mu = \Gamma_{ml}$, $\Gamma_\nu = \Gamma_{nl}$, $\Delta = \omega - \omega_{mn}$, $\Delta_\mu = \omega_\mu - \omega_{ml}$.

These equations take into account only interaction of ions with light, however the Coulomb ion-ion scattering with velocity change affects the probe-field spectrum strongly. The scattering is small-angle then it can be described as a diffusion process in the velocity space. The scattering is independent of the ionic internal quantum state, then it can be taken into consideration by operator $\hat{S} = -D \partial_{vv}$ in the left sides of (1) or (3), where

$$D = \frac{\nu \nu_T^2}{2}, \quad \nu = \frac{16 \sqrt{\pi} N Z^2 e^4 A}{3 M^2 v_T^6}. \quad (4)$$

Here $D$ is the diffusion coefficient in the velocity space, $\nu$ is the effective transport frequency of ionic collisions, $Ze$ is the ion charge, $\Lambda$ is the Coulomb logarithm, $N$ is the ionic density [17]. The full ionic density enters into expression (4), since the concentration of excited ions is small, and then the excited ions are scattered by the ions in the ground state.

2.2. Nonlinear absorption spectrum

Since the solutions to (1) are periodic functions of coordinate, the density matrix elements $\rho(x)$ are Fourier series over the spatial harmonics

$$\rho_{ij}(x, v) = \sum_{p=-\infty}^{\infty} \rho_{ij, p}(v) e^{i p k x}. \quad (5)$$
Nonlinear absorption spectrum is defined as the difference between probe-field absorption spectra when the driving field is turned off and turned on

$$\delta P_\mu = P_\mu(0) - P_\mu(\Omega), \quad P_\mu \propto \text{Im} \int_{-\infty}^{\infty} r_{m\ell,0}(v) \, dv.$$  \hfill (6)

Its advantage is absence of the broad Doppler contour being subtracted, as shown in Fig. 1. Scales of axis $y$ for linear and nonlinear absorption differs by an order of magnitude, then the nonlinear spectrum is better to study small nonlinear contributions of the strong driving field. Expression for $\delta P_\mu$ includes only zeroth Fourier component of the polarization $r_{m\ell,0}(v)$, since one must average the absorption over coordinate $z$ and other components give no contribution.

The dip or peak broaden compared to the collisionless case. The broadening due to velocity changing collisions could depend only on the diffusion coefficient and the wavenumber $k_{\mu} \sim k$. There is the only their combination with frequency dimension $\delta \Delta_\mu \sim D^{1/3}k^{2/3} \sim \nu^{1/3}(kv_T)^2/3$. For checking the hypothesis we plotted the series of curves $\delta P_\mu(\Delta_\mu)$, Fig. 2, at different diffusion coefficients. The curves are calculated by the formulas of perturbation theory at small Rabi frequency $\Omega$. The full width at half maximum (FWHM) as a function of $D$ is shown in the inset. The slope 0.31 is obtained by the least square fitting in agreement with estimated exponent $1/3$. This dependence has a simple physical explanation.

The broadening takes place because of the dephasing of wave in the reference frame of an ion due to the Coulomb scattering. At time $t$ the active excited ion is scattered by ions in ground state by random angle $\theta$. Its average value is zero $\langle \theta \rangle = 0$, while the mean square value is nonzero according to the diffusion law $\langle \dot{v}^2 \rangle = \nu t$. The ion started in the antinode of standing wave and having zero projection of the velocity onto the wavevector acquires velocity $\delta v \sim (\nu t)^{1/2}v_T$, then passes the distance $\delta x \sim \nu^{1/2}k^{3/2}v_T$. When the phase incursion of $n$-th harmonics reaches $\pi$ the ion leaves the interaction region, and then the effect of $n$-th harmonics vanishes. The characteristic duration of this process is of the order of $t \sim (nkv_T)^{-2/3}\nu^{-1/3}$. For estimation we take $n = 2$, then the broadening is $\delta \Delta_\mu \sim D^{1/3}k^{2/3}$. At $\delta \Delta_\mu \sim \Gamma_\mu$ the dip (in

**Figure 1.** Absorption $P_\mu(\Omega)$ (arb. units) as a function of the probe-field frequency detuning $\Delta_\mu$ (dots), the same without the strong field $P_\mu(0)$ (dashes); the ordinate axis is at the left. Their difference $\delta P_\mu$, the spectrum of nonlinear absorption (solid curve, right axis).

**Figure 2.** Central peak at $\Gamma_\nu = \Gamma_\mu = 1$ GHz and different diffusion coefficient $D$: $D/D_0 = 0.1, 0.075, 0.05, 0.025, 0.001$, $D_0 = 10^{13}$ m$^2$/s$^3$. Inset: the width of peak as a function of diffusion coefficient in logarithmic coordinates.
resonance case) is washed off and the peak (in nonresonance case) becomes wider. Notice that the considered effect of Coulomb dephasing corresponds to the density dependence $N^{1/3}$, while the known effect of Coulomb broadening of the Lamb dip gives $N^{1/2}$ [28].

3. Numerical calculation

Three level scheme is studied numerically. In the previous section we neglect the population of intermediate level $m$, and then take into account only the population of the final level $l$. This approximation answer the experimental conditions in ArII spectrum, where the initial state $n$ is relatively wide, when the intermediate and final states $m, l$ are narrow: $\Gamma_n \approx 3 \times 10^9$ s$^{-1}$, $\Gamma_m \approx 1.5 \times 10^8$ s$^{-1}$, $\Gamma_l \approx 8 \times 10^7$ s$^{-1}$. The populations of these states are nearly $N_n \approx 10^9$ cm$^{-3}$, $N_m \approx 5 \times 10^9$ cm$^{-3}$, $N_l \approx 10^{11}$ cm$^{-3}$, then $N_n, N_m \ll N_l$. In calculations all the populations are taken into account. For this purpose the full set of 9 coupled Fokker—Plank equations are solved numerically instead of simplified set (3).

To compare the computations with experiment the nonlinear spectra are fitted with the least squares. The full set of linear equations includes 13 parameters: $N_j, \Gamma_j$, the unperturbed population and the width of level $j = n, m, l$, $\Delta, \Delta_\mu$, the detunings of strong and probe fields, $k, k_\mu$, the wavenumbers of strong and probe fields, $\Omega$, the Rabi frequency of strong field, $\nu$, the effective frequency of scattering. The majority of parameters are known from independent measurements. The least-square fitting is carried out by 3 physical parameters: the effective frequency of ionic scattering $\nu$, the Rabi frequency of strong field $\Omega$, and the driving field detuning $\Delta$. Nonlinear resonances at different

![Figure 3. Nonlinear absorption $\delta P_\mu$ (arb. units) as a function of probe-field detuning $\Delta_\mu$ (GHz): experimental data (dots) and least-square fitting (curve).](image-url)
shown in Fig. 3 along with the results of fitting demonstrate a good agreement. Subfigures a–d are plotted at different detuning $\Delta$: from resonance to nonresonance case.

4. Conclusions
Thus, the shape of the standing wave resonance is calculated both numerically and analytically. The absorption is found in the 4th order of the perturbation theory in qualitative agreement with measurement. The quantitative calculations taking into consideration small population of the upper level and saturation of $mn$ transition allows fitting the experimental data with accuracy $5 \div 10\%$. The agreement occurs at experimental plasma and laser parameters.

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