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Anomalous dispersion properties of an atomic medium with a closed excitation contour

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Abstract. **This paper examines the refractive index dispersion in an atomic medium with a closed excitation contour. The refractive index as a function of the algebraic sum of the phases of excitation fields (Φ) and Rabi frequencies is shown to have a maximum for $\Phi \leqslant \pi/4$. A frequency range is found near the maximum where an increase in refractive index is accompanied by amplification of one of the optical fields.**

Keywords: *coherent population trapping, dispersion, closed excitation contour, electromagnetically induced transparency.*

1. Introduction

Recent years have seen considerable interest in optical control over the refractive index of media. The refractive index of an optical medium at its resonance frequencies may be tens of times that characteristic of the interaction of the medium with nonresonant light. The large refractive index of a medium consisting of two-level atoms is due to its considerable optical absorption. At the same time, the interference of atomic transitions in three-level systems, which leads to low-frequency coherence excitation, allows one to obtain a transparent medium with a high refractive index [1 – 5]. Physically, this is due to both coherent population trapping (CPT) [6] and a similar effect, electromagnetically induced transparency (EIT) [7 – 9], in three-level media.

It should be emphasised that, as a result of quantum coherence, there is little or no absorption in the medium (i.e. the medium is transparent to incident light) and its refractive index is comparable to the resonance index of a medium consisting of two-level atoms. Such features in the behaviour of the refractive index may be of interest for designing lasers without inversion [10 – 13]. The use of refractive index control in quantum memory systems was considered by Kalachev and Kocharovskaya [14].

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An important point is that, for an atomic medium to have high dispersion in combination with weak absorption, an additional ‘preparation’ of the medium is needed [10 – 13], which can be ensured using both additional light [12] and the propagation of polychromatic light in a medium consisting of multilevel atoms. In the latter instance, a key role is played by the scheme of interaction between optical fields and the system of quantum levels. A special place is held by atomic systems that interact with fields according to a closed scheme [15 – 18]. The dynamics of such a system depend crucially on the algebraic sum of the initial phases of fields acting on it. A closed excitation contour can be realised in a three-level Λ system when a coupling rf field is applied between its lower levels (so called Δ system) [16], or the interaction contour can be closed through an extra upper level using two optical fields (double Λ system) [15, 17, 18].

According to further research, the initial phases of fields determine whether or not systems with a closed interaction contour exhibit CPT [16 – 18] and EIT. The latter effect was studied in a closed Δ system formed by applying a microwave field between the lower levels of the system [19, 20]. Li et al. [19] and Jha et al. [21] studied absorption of light in a gas cell for systems with a closed excitation contour when four pulsed optical fields were applied. It is remarkable that such a closed excitation contour is possible for both alkali metal atoms and semiconductor quantum size effect systems [22, 23].

At the same time, the studies reported in Refs [15 – 19] focused mainly on the development and decay of CPT (EIT) effects for a closed excitation contour as a function of the relative phase Φ of excitation fields. The influence of Φ on the refractive index and absorption coefficient dispersion was not examined in those studies. Such investigation is of current interest and might yield nontrivial results. It has been the objective of this work to study the influence of the relative phase of excitation fields on the refractive index and absorption coefficient dispersion under CPT effect conditions.

2. Basic equations

Figure 1 schematically shows a three-level system with a closed interaction contour (Δ system). The transition between states $|1\rangle$ and $|2\rangle$ is a magnetic dipole transition, and $|1\rangle \to |3\rangle$ and $|2\rangle \to |3\rangle$ are strong electric dipole transitions in the optical spectral region. The upper state of the system decays to states $|1\rangle$ and $|2\rangle$ with a decay rate 2γ. The quantum system shown in Fig. 1 will be thought to interact with three resonance fields: the optical fields due to the $|1\rangle \to |3\rangle$ and $|2\rangle \to |3\rangle$ transitions have Rabi frequencies Ω_1 and Ω_2, respectively, and the microwave field applied between levels $|1\rangle$ and $|2\rangle$ has a Rabi frequency U.

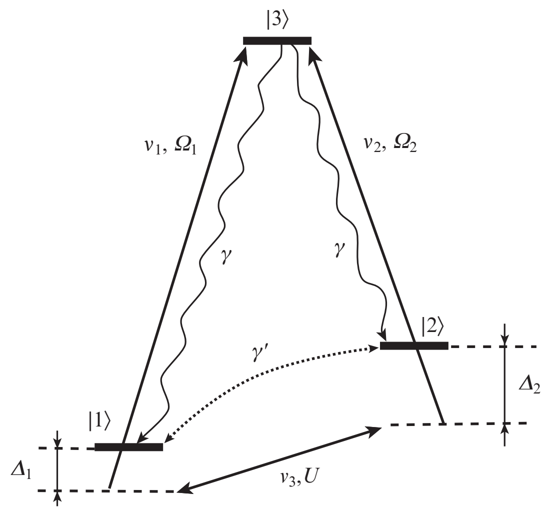

Figure 1. Energy-level diagram of a three-level system with a closed excitation contour (Δ system): ν_1 and ν_2 are optical frequencies, γ' is the microwave coherence relaxation rate, and Δ_1 and Δ_2 are one-photon detunings.

The system of coupled quantum rate equations for the density matrix elements ρ_{ij}, which determine the interaction of the closed Δ system (Fig. 1) with a three-frequency field, can be written in the form

$$\frac{\partial \rho_{ik}}{\partial t} = -\frac{\mathrm{i}}{\hbar}\sum_{l}(H_{il}\rho_{lk} - \rho_{il}H_{lk}) + \sum_{l,m}\Gamma_{iklm}\rho_{lm}, \tag{1}$$

where H is the Hamiltonian and Γ is the relaxation matrix. The Hamiltonian can be represented as $H = H_0 + H_{\mathrm{int}}$, where

$$H_0 = \sum_{i=1}^{4} E_i\,|i\rangle\langle i| \tag{2}$$

is the Hamiltonian in the absence of a laser field and H_{int} represents the interaction of the quantum system with the laser field. In the resonance approximation, we have

$$H_{\mathrm{int}} = \hbar\Omega_1\exp[-\mathrm{i}(\nu_1 t + \varphi_1)]\,|3\rangle\langle 1| + \hbar\Omega_2\exp[-\mathrm{i}(\nu_2 t + \varphi_2)]\times$$

$$\times|3\rangle\langle 2| + \hbar U\exp[-\mathrm{i}(\nu_3 t + \varphi_3)]|2\rangle\langle 1| + \text{h.c.} \tag{3}$$

Here, $\Omega_1 = \mu_{13}E_1/(2\hbar)$; $\Omega_2 = \mu_{23}E_2/(2\hbar)$; $U = \mu_{12}E_3/(2\hbar)$; E_i and φ_i are the amplitude and initial phase of the ith laser field component of frequency ν_i ($i = 1, 2, 3$); $\nu_1 = \nu_2 + \nu_3$; μ_{13} and μ_{23} are the dipole moments of the $|1\rangle\to|3\rangle$ and $|2\rangle\to|3\rangle$ transitions; and μ_{12} is the magnetic dipole moment of the $|1\rangle\to|2\rangle$ transition. The one-photon detunings of the laser fields from the $|1\rangle\to|3\rangle$ and $|2\rangle\to|3\rangle$ transitions are given by $\Delta_j = \nu_j - \omega_{3j}$ (j = 1, 2), where ω_{3j} are the frequencies of the transitions between levels 3 and j. The detuning from the two-photon resonance is $\delta = (\Delta_1 - \Delta_2)/2$.

To find the refractive index and absorption coefficient of the medium, consider its nonlinear susceptibility χ, which is in general complex-valued ($\chi = \chi' + \mathrm{i}\chi''$) and is related to the polarisability vector $\boldsymbol{P}$ by

$$\boldsymbol{P} = \varepsilon_0\chi\boldsymbol{E}, \tag{4}$$

where ε_0 is the electric constant and $\boldsymbol{E}$ is the electric field.

The real (χ') and imaginary (χ'') parts of the nonlinear susceptibility represent the dispersion and absorption at a distance equal to the optical radiation wavelength and can be expressed through off-diagonal elements of the density matrix (1) as [1]

$$\chi_{13} = \frac{N|\mu_{13}|^2}{2\hbar\varepsilon_0\Omega_1}\rho_{13},\ \chi_{23} = \frac{N|\mu_{23}|^2}{2\hbar\varepsilon_0\Omega_2}\rho_{23}, \tag{5}$$

where N is the number of active atoms in the cell and μ_{ij} is the dipole moment of the $|i\rangle\to|j\rangle$ transition. Therefore, $\chi'_{13} \sim \mathrm{Re}\rho_{13}$ and $\chi'_{23} \sim \mathrm{Re}\rho_{23}$ are dispersions (i.e. refractive indices) and $\chi''_{13} \sim \mathrm{Im}\rho_{13}$ and $\chi''_{23} \sim \mathrm{Im}\rho_{23}$ are losses (i.e. absorption coefficients) for optical fields with Rabi frequencies Ω_1 and Ω_2, respectively.

Solving Eqns (1) for a steady state, we obtain an expression for the imaginary and real parts of the optical coherences ρ_{13} and ρ_{23}. Since $\mathrm{Re}\rho_{13} = -\mathrm{Re}\rho_{23}$ and $\mathrm{Im}\rho_{13} = \mathrm{Im}\rho_{23}$, we restrict ourselves to analysis of the expressions for $\mathrm{Re}\rho_{23}$ and $\mathrm{Im}\rho_{23}$:

$$\mathrm{Re}\rho_{23} = \frac{U^2\Omega\gamma\sin\Phi\cos\Phi}{U^2\gamma^2 + 4(\Omega^2 - U^2)^2 + 12\Omega^2U^2\sin^2\Phi}, \tag{6}$$

$$\mathrm{Im}\rho_{23} = \frac{U\Omega\sin\Phi[U\gamma\sin\Phi + 2(\Omega^2 - U^2)]}{U^2\gamma^2 + 4(\Omega^2 - U^2)^2 + 12\Omega^2U^2\sin^2\Phi}. \tag{7}$$

Here, the Rabi frequencies of the optical fields are taken to be identical ($\Omega_1 = \Omega_2 = \Omega$) and the two-photon resonance condition $\delta = 0$ is thought to be satisfied. In (6) and (7), we use the total phase of the closed atomic interaction contour: $\Phi = \varphi_1 - \varphi_2 - \varphi_3$, where φ_i are the initial phases of the excitation fields.

3. Discussion

Equations (6) and (7) completely determine the refractive index and absorption coefficient as functions of Φ for a medium consisting of three-level Δ atoms provided the two-photon resonance condition is satisfied. It is known that the presence of a closed excitation contour in a three-level system leads to the decay (at $\Phi = \pi/2$) and restoration ($\Phi = 0$) of the CPT effect [14–16]. Figure 2 shows the real ($\mathrm{Re}\rho_{23}$) and imaginary ($\mathrm{Im}\rho_{23}$) parts of the optical coherence ρ_{23} as functions of Φ for identical Rabi frequencies of the optical and coupling fields. It is seen that, at $\Phi = 0$, the absorption and dispersion are also zero, which corresponds to a CPT effect in the system.

At the same time, at $\Phi = \pi/2$ there is a strong absorption associated with decay of the CPT state. As the Rabi frequencies of the excitation fields increase, the absorption coefficient saturation region shifts to lower Φ values (Figs 2b, 2c, dashed lines), which is due to the stronger excitation field effect on the CPT resonance decay. Moreover, increasing the coupling field amplitude increases the coupling between the lower levels, which has a significant effect on the optical coherences. In particular, this shows up as a reduction in absorption amplitude.

As seen in Fig. 2a, the refractive index has a maximum at $\Phi = \pi/4$ (solid line). At higher Rabi frequencies, the maximum shifts towards zero phase (Figs 2b, 2c, solid lines).

Note that the refractive index and absorption coefficient have a maximum at different Φ values. Let us find the value of Φ in the range $\Phi \in [0;\pi/2]$ at which the refractive index has the highest value:

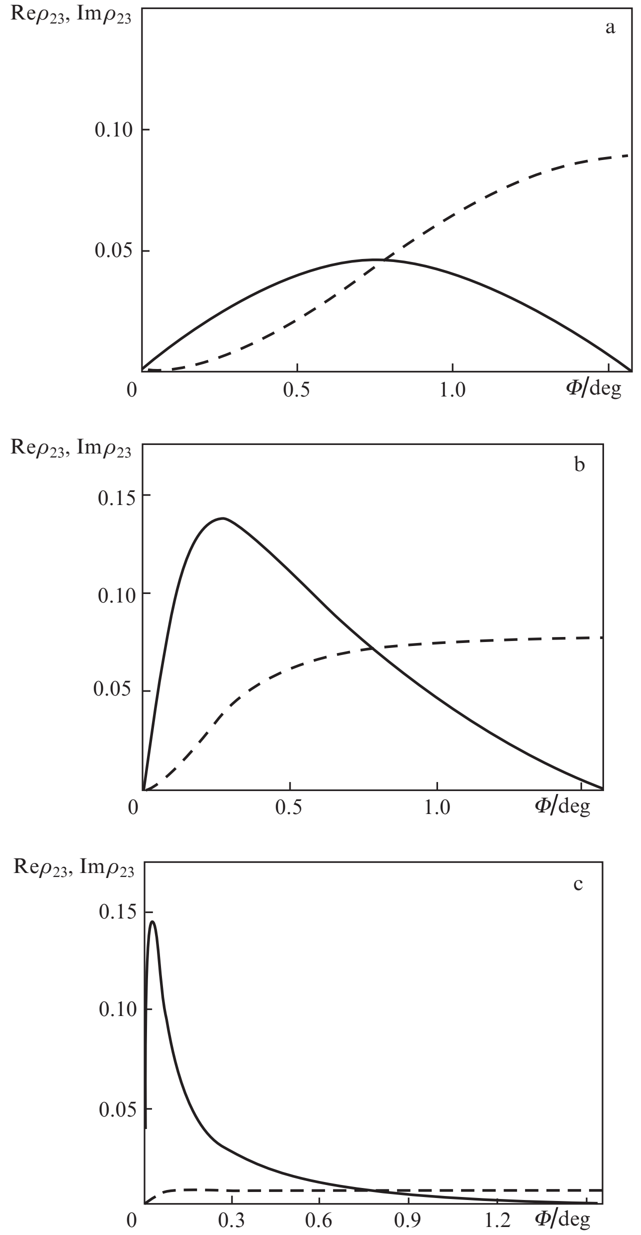

Figure 2. Dispersion $\mathrm{Re}\rho_{23}$ (solid lines) and absorption coefficient $\mathrm{Im}\rho_{23}$ (dashed lines) as functions of Φ for a Δ system under two-photon resonance conditions ($\delta = 0$) at Rabi frequencies (a) $\Omega_1 = \Omega_2 = U = 0.1\gamma$, (b) $\Omega_1 = \Omega_2 = U = \gamma$ and (c) $\Omega_1 = \Omega_2 = U = 10\gamma$.

$$\Phi_{\mathrm{ex}} = \arccos\left(\sqrt{\frac{4U^4 + 4U^2\Omega^2 + 4\Omega^4 + U^2\gamma^2}{8U^4 - 4U^2\Omega^2 + 8\Omega^4 + 2U^2\gamma^2}}\right). \qquad (8)$$

When the optical and microwave fields have identical Rabi frequencies ($\Omega_1 = \Omega_2 = U$), (8) takes the form

$$\Phi_{\mathrm{ex}} = \arccos\left(\sqrt{1 - \frac{\gamma^2}{12\Omega^2 + 2\gamma^2}}\right). \qquad (9)$$

It follows from (8) that $\Phi_{\mathrm{ex}} = \pi/4$ at Rabi frequencies $\Omega \ll \gamma$ (Fig. 2a), whereas for $\Omega \gg \gamma$ we have $\Phi_{\mathrm{ex}} = \gamma/(\Omega\sqrt{12})$, which corresponds to Fig. 2c.

Above we examined the refractive index and absorption coefficient as functions of Φ in the case of two-photon resonance ($\delta = 0$). Consider now these parameters as functions of two-photon detuning δ at different phases of the closed con-

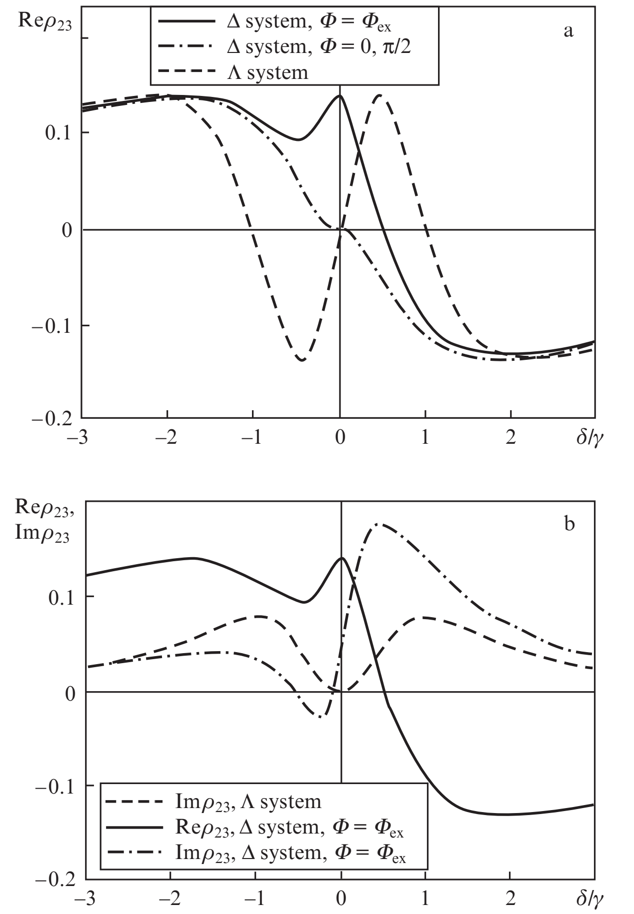

Figure 3. (a, b) Dispersion $\mathrm{Re}\rho_{23}$ and (b) absorption coefficient $\mathrm{Im}\rho_{23}$ as functions of two-photon detuning δ at different values of Φ and $\Omega_1 = \Omega_2 = U = \gamma$.

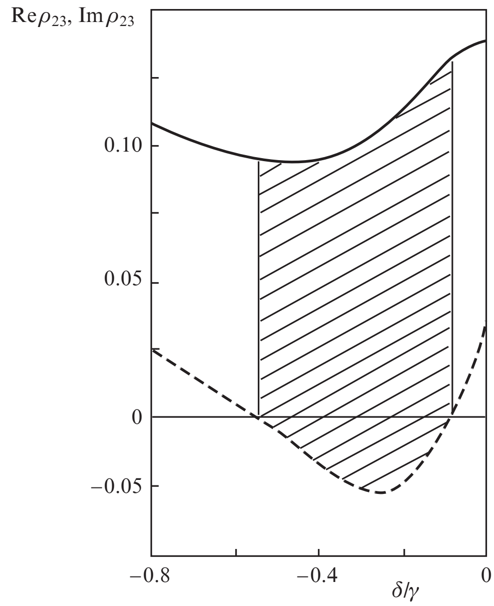

Figure 4. Dispersion $\mathrm{Re}\rho_{23}$ (solid line) and absorption coefficient $\mathrm{Im}\rho_{23}$ (dashed line) as functions of two-photon detuning δ on expanded scales for a Δ system at $\Phi = \Phi_{\mathrm{ex}}$ and $\Omega_1 = \Omega_2 = U = \gamma$.

tour. Figure 3a shows the imaginary part of the coherence ρ_{23} as a function of detuning δ at three phases in a closed Δ system and Reρ_{23} as a function of two-photon detuning for a three-level Λ system. It is seen that, in the two-photon resonance region ($\delta = 0$), the refractive index is zero at $\Phi = 0$ and $\pi/2$ in both the Δ system (dot-dashed line) and Λ system (dashed line). At $\Phi = \Phi_{ex}$, the refractive index has the highest value. Comparing it to the absorption coefficient, which is proportional to imaginary part of the coherence (Fig. 3b, dot-dashed line), we see that there is a detuning range where, in the absence of absorption, a rather high refractive index can be obtained.

Consider in greater detail the detuning range corresponding to Fig. 3b: $-0.8 \leqslant \delta/\gamma \leqslant 0$ (Fig. 4). It is seen in Fig. 4 that there is a gain in the hatched region and that the refractive index increases across this region (solid line). These features in the behaviour of the refractive index and gain coefficient can be used in a variety of nonlinear optical applications.

4. Conclusions

We have studied a three-level system interacting with two optical fields and one microwave field that form a closed excitation contour (Δ system). The refractive index of the system as a function of the algebraic sum of the phases of excitation fields, Φ, is shown to have a maximum. The maximum in the refractive index shifts from $\Phi = \pi/4$ to $\Phi = \gamma/\sqrt{12\Omega^2}$, depending on Rabi frequency. For the Φ value corresponding to the maximum in the refractive index, a detuning range is found where the refractive index increases, with a concurrent amplification of one of the optical fields.

It should be emphasised that the refractive index has a maximum for $\Phi \leqslant \pi/4$ not only in the system considered here but also in any system with a closed excitation contour. In such a case, the refractive index is of the same order as that in a three-level Λ system [1].

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