

Coherent Control Theory of off Resonance Refractive Index of Medium with a Gaussian Pulse of Coherent Light

Rui-Hua Xie and Paul Brumer^a

Max-Planck-Institute für Strömungsforschung, Bunsenstr.10, D-37073 Göttingen

^a Chemical Physics Theory Group, University of Toronto, Toronto M5S 3H6, Canada

Reprint requests to Dr. R.-H. X., E-mail: rxie@gwdg.de

Z. Naturforsch. **54 a**, 171–176 (1999); received March 4, 1999

In this paper we develop a coherent control theory of the off resonance refractive index of a medium with a Gaussian pulse of coherent light. The refractive index of gaseous N_2 is studied by using this theory. It is found that the off resonance refractive index of gaseous N_2 can be enhanced through the coherent control approach by changing the coherent parameters or the pulse duration of the coherent light.

I. Introduction

Quantum coherence and interference in atomic systems can lead to interesting optical phenomena such as nonabsorbing resonances [1] and lasing without inversion [2]. Recently, Scully [3] and Scully and Fleischhauer [4] have shown that the refractive index of a gaseous medium prepared in a phase-coherent state (phaseonium) can be resonantly enhanced while the absorption is vanishingly small. For this effect, several coherence-establishing schemes have been investigated [5, 6]. Furthermore, Quang and Freedhoff [7] have reported that this novel effect can also be obtained from the study of a coherently driven two-level atom damped by a normal vacuum. An extension of the work of Quang and Freedhoff to include Lorentzian and Doppler broadenings was given by Ling and Barbay [8], and Szymanowski and Keitel [9]. The generation of coherence via incoherent pumping to produce a high refractive index accompanied by vanishing absorption has also been reported [11], and the nonlinear effect, which takes place in the vicinity of an atomic resonance, has been taken into account for potential applications of high-index materials [12]. Very recently, Zibrov et al. [10] have experimentally demonstrated that a resonant enhancement of the refractive index is accompanied by vanishing absorption in a cell containing a coherently prepared Rb vapour. All of these theoretical and experimental studies focus mainly on the resonant case. In this paper, we turn our attention to the off resonance case.

It is known that modern lasers can generate bursts as short as 10^{-14} second. Unlike continuous-wave radiation, a light pulse is made up of a collection of distinct frequencies and, hence, of a collection of photons with different energies. Such light has a perhaps counterintuitive property. The briefer the pulse, the broader the range of energies within it. Rice et al. [14] have allowed for the coherent control of the yield of a chemical reaction with pulses. The idea is that the first pulse brings the molecule into a superposition state, and the second pulse breaks up the molecule into different products. In this paper, following such a coherent control scenario, we develop a coherent control theory of an off resonance refractive index of a medium with a Gaussian pulse of coherent light. As an example, we have shown, by using this theory, that the off resonance refractive index of gaseous N_2 can be enhanced by changing the coherent parameters or the pulse duration of the coherent light.

II. Theoretical Model of Molecule-Field Interactions

The radiation-free molecular Hamiltonian, its discrete or continuous set of energy eigenvalues, and the corresponding eigenfunctions are denoted by \hat{H}_0 , E_n , and $|\phi_n(\mathbf{r})\rangle$, respectively, which satisfy the time-independent Schrödinger equation

$$\hat{H}_0 |\phi_n(\mathbf{r})\rangle = E_n |\phi_n(\mathbf{r})\rangle = \hbar\omega_n |\phi_n(\mathbf{r})\rangle, \quad (1)$$

where $\langle\phi_m(\mathbf{r})|\phi_n(\mathbf{r})\rangle = \delta_{mn}$, and ω_n is the angular frequency.

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In the presence of the radiation field, the time evolution of the molecule-field coupling system is governed by the Hamiltonian

$$\hat{H}(t) = \hat{H}_0 + \hat{V}(t), \quad (2)$$

where the interaction Hamiltonian, $\hat{V}(t)$, which describes the interaction of the molecule with the radiation field, is given in the dipole approximation by

$$\hat{V}(\mathbf{r}, t) = -\hat{\mathbf{u}} \cdot \mathbf{E}(t), \quad (3)$$

where $\hat{\mathbf{u}} = -e\hat{\mathbf{r}}(t)$ is the electric dipole moment operator, $-e$ is the charge of the electron, and $\mathbf{E}(t)$ represents the radiation field. Assuming that all of the properties of the molecule-field coupling system can be described by the wave function $|\Psi(\mathbf{r}, t)\rangle$, we have

$$i\hbar \frac{\partial}{\partial t} |\Psi(\mathbf{r}, t)\rangle = \hat{H}(t) |\Psi(\mathbf{r}, t)\rangle. \quad (4)$$

In general, the above time-dependent Schrödinger equation cannot be solved exactly. So, it is often adequate to solve it through the use of perturbation theory. In order to solve (4) systematically in terms of a perturbation expansion, we replace the Hamiltonian $\hat{H}(t)$ by

$$\hat{H}(t) = \hat{H}_0 + \lambda \hat{V}(t), \quad (5)$$

where λ is a continuously varying parameter ranging from zero to unity that characterizes the strength of the interaction, and $\lambda = 1$ corresponds to the actual physical situation. Now we seek a solution to the Schrödinger equation (5) in the form of a power series in λ :

$$|\Psi(\mathbf{r}, t)\rangle = |\Psi^{(0)}(\mathbf{r}, t)\rangle + \lambda |\Psi^{(1)}(\mathbf{r}, t)\rangle + \lambda^2 |\Psi^{(2)}(\mathbf{r}, t)\rangle + \dots + \lambda^N |\Psi^{(N)}(\mathbf{r}, t)\rangle. \quad (6)$$

Thereby, introducing (6) into (4) and requiring that all terms that are proportional to λ^N satisfy the equality separately, we obtain the set of equations

$$i\hbar \frac{\partial}{\partial t} |\Psi^{(0)}(\mathbf{r}, t)\rangle = \hat{H}_0 |\Psi^{(0)}(\mathbf{r}, t)\rangle, \quad (7)$$

$$i\hbar \frac{\partial}{\partial t} |\Psi^{(N)}(\mathbf{r}, t)\rangle = \hat{H}_0 |\Psi^{(N)}(\mathbf{r}, t)\rangle + \hat{V}(\mathbf{r}, t) |\Psi^{(N-1)}(\mathbf{r}, t)\rangle, \quad N = 1, 2, 3, \dots \quad (8)$$

Moreover, we expand the full time-dependent wavefunction $|\Psi^{(N)}(\mathbf{r}, t)\rangle$ in terms of eigenfunctions $|\phi_n(\mathbf{r})\rangle$ of the radiation-free molecular Hamiltonian \hat{H}_0 , i.e.,

$$|\Psi^{(N)}(\mathbf{r}, t)\rangle = \sum_l C_l^{(N)}(t) e^{-iE_l t/\hbar} |\phi_l(\mathbf{r})\rangle, \quad (9)$$

where $C_l^{(N)}$ gives the probability amplitude that, to N th order in the perturbation, the molecule is in the energy eigenstate $|\phi_l(\mathbf{r})\rangle$ at time t . If (9) is substituted into (8), we find that the probability amplitudes are given by

$$C_m^{(N)}(t) = (i\hbar)^{-1} \sum_l \int_{-\infty}^t V_{ml}(t') C_l^{(N-1)}(t') e^{i\omega_{ml} t'} dt', \quad (10)$$

where $\omega_{ml} = (E_m - E_l)/\hbar$ is the transition frequency between eigenstates $|\phi_m(\mathbf{r})\rangle$ and $|\phi_l(\mathbf{r})\rangle$ of \hat{H}_0 , and $V_{ml}(t') = \langle \phi_m(\mathbf{r}) | \hat{V}(t') | \phi_l(\mathbf{r}) \rangle$ is a matrix element of the perturbing Hamiltonian. The form of (9) demonstrates the usefulness of the perturbation technique. Once the probability amplitudes of order $N - 1$ are determined, the amplitudes of the next higher order N can be obtained by straightforward time integration.

Following the control scenario [14], here we assume that the molecule-field coupling system is initially in the superposition of the eigenstates $|\phi_a(\mathbf{r})\rangle$ and $|\phi_b(\mathbf{r})\rangle$ of \hat{H}_0 , which can be realized by using a pulse light, i.e.,

$$|\Psi(\mathbf{r}, t=0)\rangle = C_a |\phi_a(\mathbf{r})\rangle + C_b |\phi_b(\mathbf{r})\rangle. \quad (11)$$

Then we have

$$C_l^{(0)}(t) = C_l \delta_{la} + C_l \delta_{lb}, \quad (12)$$

$$\begin{aligned} C_l^{(1)}(t) &= C_a (i\hbar)^{-1} \int_{-\infty}^t V_{la}(t') e^{i\omega_{la} t'} dt' \\ &\quad + C_b (i\hbar)^{-1} \int_{-\infty}^t V_{lb}(t') e^{i\omega_{lb} t'} dt' \\ &= C_a (-i\hbar)^{-1} U_{la} I(t, \omega_{la}) \\ &\quad + C_b (-i\hbar)^{-1} U_{lb} I(t, \omega_{lb}), \\ &\dots \end{aligned} \quad (13)$$

where $U_{ij} = \langle \phi_i(\mathbf{r}) | \hat{\mathbf{u}} | \phi_j(\mathbf{r}) \rangle$ is the transition-dipole matrix element between $|\phi_i(\mathbf{r})\rangle$ and $|\phi_j(\mathbf{r})\rangle$,

and $I(t, \Omega)$, the spectral density at the frequency Ω , is defined as

$$I(t, \Omega) = \int_{-\infty}^t \mathbf{E}(t') e^{i\Omega t'} dt'. \quad (14)$$

Once the radiation field $\mathbf{E}(t)$ and the dipole matrix elements are given, we get $|\Psi(\mathbf{r}, t)\rangle$, which describes all the properties of the molecule-field coupling system.

III. Definition of the Refractive Index of a Medium

From classical optics, the refractive index of a medium is defined as a measure of the speed v of an electromagnetic wave in the medium with respect to its speed c in the vacuum, i.e.

$$n = \frac{c}{v} = \sqrt{\frac{\mu\epsilon}{\mu_0\epsilon_0}}, \quad (15)$$

where μ is the permeability, ϵ is the permittivity of the medium, and the subscript 0 is to denote the values in the vacuum. For most materials, $\mu \approx \mu_0$. Then we have

$$n = \sqrt{\frac{\epsilon}{\epsilon_0}} = \sqrt{\epsilon_r}, \quad (16)$$

where ϵ_r is the relative permittivity of the medium.

The permittivity is a measure of the response of a medium to an applied field \mathbf{E} :

$$\mathbf{D} = \epsilon \mathbf{E} = \epsilon_0 \epsilon_r \mathbf{E}, \quad (17)$$

where \mathbf{D} is the displacement vector. In the multipolar component formula, it is expanded as

$$\mathbf{D} = \epsilon_0 \mathbf{E} + \mathbf{P} - \Delta \cdot \mathbf{Q} + \dots \quad (18)$$

where \mathbf{P} is the dipole moment, and $\Delta \cdot \mathbf{Q}$ is the quadrupole moment. Since we only consider the cases of a weak radiation field, we have

$$\mathbf{D} \approx \epsilon_0 \mathbf{E} + \mathbf{P}. \quad (19)$$

Comparing (17) with (19), we get

$$\mathbf{P} = \epsilon_0(\epsilon_r - 1)\mathbf{E}. \quad (20)$$

In addition, the polarization of a medium, \mathbf{P} , is defined by the sum of the individual dipole moments p_i per unit volume, i.e.

$$\mathbf{P} = \sum_{i=1}^N \frac{p_i}{V}. \quad (21)$$

Assuming a uniform density ρ of the medium, we have

$$\mathbf{P} = \rho p. \quad (22)$$

The polarization can be decomposed into a permanent component and a transitory response due to the electric field. The latter one is characterized by the susceptibility χ , i.e.

$$\mathbf{P}(\mathbf{r}, t) = \epsilon_0 \int_{-\infty}^{\infty} \chi(\tau) \mathbf{E}(\mathbf{r}, t - \tau) d\tau. \quad (23)$$

After taking the Fourier transformation, the susceptibility χ is defined in the frequency ω by

$$\mathbf{P}(\mathbf{r}, \omega) = \epsilon_0 \chi(\omega) \mathbf{E}(\mathbf{r}, \omega). \quad (24)$$

Moreover, from (20) and (24) we have

$$\epsilon_r = 1 + \chi(\omega). \quad (25)$$

Finally, we get

$$n(\omega) = \sqrt{1 + \chi(\omega)}. \quad (26)$$

Since the susceptibility is complex, i.e. $\chi = \chi' + i\chi''$ is complex, the refractive index n is also complex. The real part n' of the refractive index n , given by

$$n' = \sqrt{\frac{\sqrt{(1 + \chi')^2 + \chi''^2} + 1 + \chi'}{2}}, \quad (27)$$

is related to the propagation speed of the wave front in the medium. The imaginary part n'' of the refractive index n describes the modification of the amplitude of the electromagnetic wave as it propagates through the medium, which is given in detail by

$$n'' = \sqrt{\frac{\sqrt{(1 + \chi')^2 + \chi''^2} - 1 - \chi'}{2}} \text{sign}(\chi''), \quad (28)$$

where $\text{sign}(x) = 1$ for $x \geq 0$ and $\text{sign}(x) = -1$ for $x < 0$.

Therefore, if we know the dipole moment $\mathbf{P}(\omega)$ and the field $\mathbf{E}(\omega)$, we can calculate the susceptibility $\chi(\omega)$ and finally get the refractive index of a medium.

IV. Coherent Control of the Refractive Index of a Medium

The expectation value of the electric dipole moment of a molecule is exactly given by

$$\begin{aligned} p(t) &= \langle \Psi(\mathbf{r}, t) | \hat{u} | \Psi(\mathbf{r}, t) \rangle \\ &= \langle \Psi^{(0)}(\mathbf{r}, t) | \hat{u} | \Psi^{(0)}(\mathbf{r}, t) \rangle \\ &\quad + \lambda \langle \Psi^{(0)}(\mathbf{r}, t) | \hat{u} | \Psi^{(1)}(\mathbf{r}, t) \rangle \\ &\quad + \langle \Psi^{(1)}(\mathbf{r}, t) | \hat{u} | \Psi^{(0)}(\mathbf{r}, t) \rangle \\ &\quad + \lambda^2 \langle \Psi^{(0)}(\mathbf{r}, t) | \hat{u} | \Psi^{(2)}(\mathbf{r}, t) \rangle \\ &\quad + \langle \Psi^{(1)}(\mathbf{r}, t) | \hat{u} | \Psi^{(1)}(\mathbf{r}, t) \rangle \\ &\quad + \langle \Psi^{(2)}(\mathbf{r}, t) | \hat{u} | \Psi^{(0)}(\mathbf{r}, t) \rangle \\ &\quad + \dots \end{aligned} \quad (29)$$

As mentioned in Sect. II, we assume that the system is initially in the superposition state (11). Up to the first order and setting $\lambda = 1$, we have

$$\begin{aligned} p(t) &= |C_a|^2 U_{aa} + |C_b|^2 U_{bb} \\ &\quad + |C_b| |C_a| e^{i\delta} U_{ab} e^{i\omega_{ab}t} + |C_b| |C_a| e^{-i\delta} U_{ba} e^{-i\omega_{ba}t} \\ &\quad + \left\{ \frac{i}{\hbar} |C_a|^2 \sum_l U_{la} U_{al} I(t, \omega_{la}) e^{-i\omega_{la}t} + \text{c.c.} \right\} \\ &\quad + \left\{ \frac{i}{\hbar} |C_b|^2 \sum_l U_{lb} U_{bl} I(t, \omega_{lb}) e^{-i\omega_{lb}t} + \text{c.c.} \right\} \\ &\quad + \left\{ \frac{i}{\hbar} |C_b| |C_a| e^{i\delta} \sum_l U_{la} U_{bl} I(t, \omega_{la}) e^{-i\omega_{lb}t} + \text{c.c.} \right\} \\ &\quad + \left\{ \frac{i}{\hbar} |C_a| |C_b| e^{-i\delta} \sum_l U_{lb} U_{al} I(t, \omega_{lb}) e^{-i\omega_{la}t} + \text{c.c.} \right\}. \end{aligned} \quad (31)$$

where δ is the relative phase between the states $|\phi_a\rangle$ and $|\phi_b\rangle$. In the above equation, the first and second terms denote the contribution from the permanent dipole moment of the medium, the first and second sum terms are the contribution when the system is initially in a pure state, and the other terms show the interference effect between the states $|\phi_a\rangle$ and $|\phi_b\rangle$.

Following the coherent control scenario [14], we suppose that the molecule interacts with a pulse of coherent light. Classically, a general pulse can be written as an integral over all the modes:

$$\mathbf{E}(t) = \int_{-\infty}^{\infty} \mathbf{E}(\omega) e^{-i\omega t} d\omega, \quad (32)$$

where ω is the mode frequency. Then the spectral density $I(t, \Omega)$ can be written as

$$I(t, \Omega) = I_{\text{RW}}(t, \Omega) + I_{\text{CRW}}(t, \Omega), \quad (33)$$

$$I_{\text{RW}}(t, \Omega) = \int_0^{\infty} \mathbf{E}(\omega) \frac{e^{i(\Omega - \omega)t}}{i(\Omega - \omega)} d\omega, \quad (34)$$

$$I_{\text{CRW}}(t, \Omega) = \int_0^{\infty} \mathbf{E}(-\omega) \frac{e^{i(\Omega + \omega)t}}{i(\Omega + \omega)} d\omega, \quad (35)$$

where $I_{\text{RW}}(t, \Omega)$ and $I_{\text{CRW}}(t, \Omega)$ denote the rotating-wave and counter-rotating wave contributions, respectively. It is known that the spectral density comes mainly from the rotating-wave term in the resonant case where the counter-rotating wave contribution may be neglected, but in the off resonance case, both terms play an important role for the spectral density [15].

Using a Gaussian pulse, we have

$$\mathbf{E}(t) = E_p e^{-(t-t_c)^2/\tau^2} e^{-i\omega_c(t-t_c)}, \quad (36)$$

$$\mathbf{E}(\omega) = \pi^{-1/2} E_p \alpha e^{-\alpha^2(\omega - \omega_c)^2} e^{-i\omega t_c}, \quad (37)$$

where t_c and ω_c are the central time and frequency positions of the Gaussian pulse, respectively; $\tau = 1/(2\sqrt{\ln 2}\Gamma)$ is the pulse duration; $\alpha = \tau/2$; Γ^{-1} is exactly the fwhm of $\mathbf{E}(t)$, and $\Sigma^{-1} = 2\sqrt{\ln 2}/\alpha$ is the fwhm of $\mathbf{E}(\omega)$. Then we have

$$\begin{aligned} I_{\text{RW}}(t, \Omega) &= \pi^{1/2} E_p \alpha e^{-\alpha^2(\Omega - \omega_c)^2} e^{i\Omega t_c} \text{sign}(t - t_c) \\ &\quad \cdot \{2\theta(t - t_c) - e^{\beta^2} W[\text{sign}(t - t_c)\beta_+]\}, \end{aligned} \quad (38)$$

$$\begin{aligned} I_{\text{CRW}}(t, \Omega) &= \sqrt{\pi} E_p \alpha e^{-\alpha^2(\Omega + \omega_c)^2} e^{-i\Omega t_c} \text{sign}(t - t_c) \\ &\quad \cdot e^{\beta^2} W[\text{sign}(t - t_c)\beta_-], \end{aligned} \quad (39)$$

where $\theta(x)$ is the Heaviside function ($\theta(x) = [1 + \text{sign}(x)]/2$), $W[z]$ is the complex error function [16],

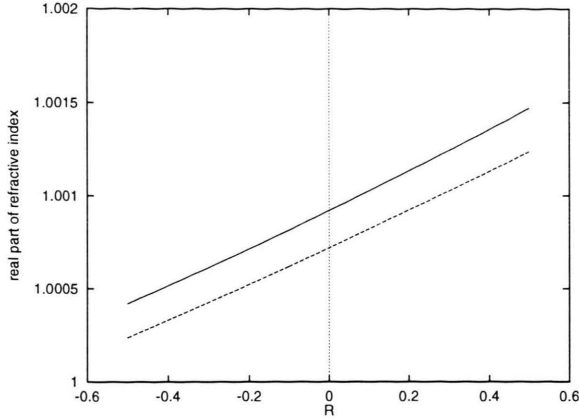


Fig. 1. The real part of the refractive index of the gaseous medium N_2 when the system is initially in the superposition of the ground state $|\nu = 0, J = 0, M = 0\rangle$ and the excited state $|\nu = 0, J = 2, M = 0\rangle$ of the ground electronic state $X\Sigma_g^+$ with $|C_a| = |C_b| = \sqrt{2}/2$ and $\delta = 0$, and the central frequency $\omega_c (= 0.072$ a.u.) of the Gaussian pulse is far away from the transition frequency between the ground and excited electronic states. The dashed line is the result when the system is initially in the ground state $|\nu = 0, J = 0, M = 0\rangle$ of the ground electronic state. Here, $R = (\omega - \omega_c)/\Sigma^{-1}$, $\rho = 2.68845 \times 10^{25}$ at STP, $E_p = 10^9$ Vm $^{-1}$, $t_c = 2.5\tau = 5\alpha$, $\Sigma^{-1} = 0.1\Delta$, and $\Delta = 0.0106209$ a.u. is the frequency width between the ground state $|\nu = 0, J = 0, M = 0\rangle$ and the excited state $|\nu = 0, J = 2, M = 0\rangle$ of the ground electronic state.

and β_{\pm} are given by

$$\beta_+ = \alpha(\Omega - \omega_c) + i\frac{t - t_c}{2\alpha}, \quad (40)$$

$$\beta_- = \alpha(\Omega + \omega_c) + i\frac{t - t_c}{2\alpha}. \quad (41)$$

Given the spectral density $I(t, \Omega)$, we are able to get the time-dependent dipole moment $p(t)$ from (31). Then, we do a Fourier transformation and get the frequency-dependent dipole moment $p(\omega)$. Finally, using (24), we get the refractive index of the medium. It is clear that the refractive index of the medium does not depend on the pulse amplitude E_p , but on the parameters $|C_a|$, $|C_b|$, δ , and τ (Γ^{-1} or Σ^{-1}). So, by changing those parameters, it is possible to control and even get an enhanced refractive index of the medium.

Taking the gaseous medium N_2 as an example, we study its refractive index. Since we focus on the molecule N_2 , the dipole transition is forbidden between $|\phi_a(r)\rangle$ and $|\phi_b(r)\rangle$ if they are in the same elec-

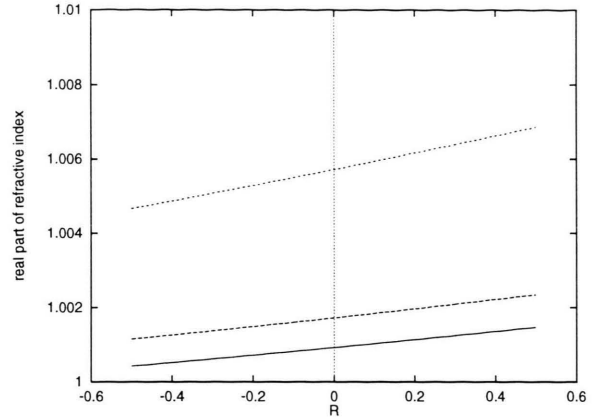


Fig. 2. The real part of the refractive index of the gaseous medium N_2 when the system is initially in the superposition of the ground state $|\nu = 0, J = 0, M = 0\rangle$ and the excited state $|\nu = 0, J = 2, M = 0\rangle$ of the ground electronic state $X\Sigma_g^+$ with $|C_a| = |C_b| = \sqrt{2}/2$ and $\delta = 0$. (a) $\Sigma^{-1} = 0.1\Delta$ (solid line); (b) $\Sigma^{-1} = \Delta$ (long dashed line); (c) $\Sigma^{-1} = 10\Delta$ (short dashed line). The others are the same as in Figure 1.

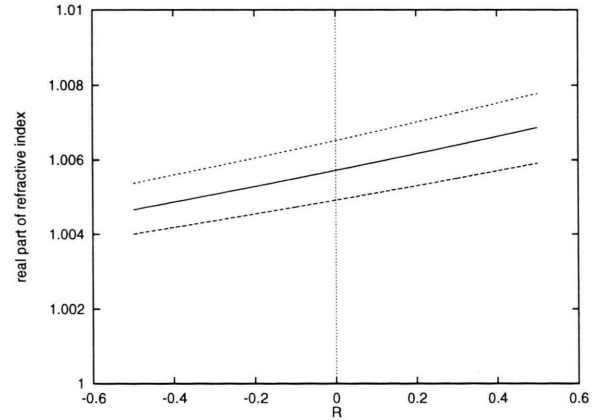


Fig. 3. The real part of refractive index of the gaseous medium N_2 when the system is initially in the superposition of the ground state $|\nu = 0, J = 0, M = 0\rangle$ and the excited state $|\nu = 0, J = 2, M = 0\rangle$ of the ground electronic state $X\Sigma_g^+$ with $|C_a| = |C_b| = \sqrt{2}/2$. (a) $\delta = 0.0$ (solid line); (b) $\delta = \pi/3$ (long dashed line); (c) $\delta = \pi/2$ (short dashed line), where $\Sigma^{-1} = 10\Delta$, and the others are the same as in Figure 1.

tronic state. In this case, $U_{ab} = U_{ba} = 0$. In addition, our medium has no permanent dipole moment, i.e., $U_{aa} = U_{bb} = 0$.

In Fig. 1, we show the results when the central frequency ω_c is 0.072 a.u., far away from the transition

frequency ($\Omega = 0.473$ a.u.) between the ground state $|\nu = 0, J = 0, M = 0\rangle$ of the excited electronic state $b'\Sigma_u^+$ and the ground state $|\nu = 0, J = 0, M = 0\rangle$ of the ground electronic state $X\Sigma_g^+$. The dashed line is the result when the system is initially in the ground state of the ground electronic state, while the solid line is the result when the system is initially in the superposition of the ground state $|\phi_a(\mathbf{r})\rangle = |\nu = 0, J = 0, M = 0\rangle$ and $|\phi_b(\mathbf{r})\rangle = |\nu = 0, J = 2, M = 0\rangle$ of the ground electronic state $X\Sigma_g^+$. It is seen that the refractive index is enhanced due to the interference effect between the states $|\phi_a\rangle$ and $|\phi_b\rangle$. In Fig. 2, we show the relation between the refractive index and the pulse width Σ^{-1} . It is seen that the off resonance refractive index is enhanced by increasing the pulse width Σ^{-1} (or say, decreasing the pulse duration τ). Finally, in Fig. 3, we show the phase-dependence of the refractive index.

Based on this numerical calculation, we find that the off resonance refractive index of gaseous N_2 can be enhanced through the coherent control approach, i.e., by changing the parameters, $|C_a|$, $|C_b|$, δ , and τ (or Σ^{-1} or Γ^{-1}).

V. Summary

In Summary, we have developed a coherent control theory of off the resonance refractive index of a medium with a Gaussian pulse of coherent light. Taking the gaseous N_2 as an example, we have shown that the off resonance refractive index can be enhanced by changing the coherent parameters or the pulse duration of the coherent light.

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