Realizing negative index of refraction in an ensemble of ground-state polar molecules with lasers

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Abstract

We propose a coherent optical method for creating negative refractive index (NRI) for a gaseous ensemble of ground-state polar molecules possessing both permanent electric and magnetic moments. Exploiting the pure rotational transition between the two lowest rotational levels of the ground vibrational state, one can generate two dressed states of mixed parity using a microwave laser. These dressed states are then used as the two lower states of a $\Lambda$-type three-level scheme using two infra-red lasers to couple them to another ro-vibrational level in the ground-state manifold. Our analytical and numerical results show that, when one of infra-red lasers used as a probe and the other as a control field with a fixed detuning, there are two distinct probe detuning regimes where the system can exhibit NRI with figure of merit reaching as high as 10. The first regime dubbed as NRI-I is the red-detuned side of the probe field and away from the two-photon or dark-state resonance. The other regime called as NRI-II is close to the dark-state resonance or electromagnetically induced transparency (EIT). While the first regime has a broad frequency span the second one has relatively narrow width. We interpret our results in terms of the proximity of EIT, quantum interference and dispersive behavior of the system.

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I. INTRODUCTION

Half-a-century ago, Russian Physicist Victor G. Veselago [1] theoretically predicted the possibility of an exotic material possessing simultaneously both negative electric permittivity $\varepsilon_r$ and magnetic permeability $\mu_r$. Based on fundamental principles of classical electrodynamics, he showed that such materials must have negative index of refraction. Materials exhibiting negative index are not found in nature. Nearly three decades after Veselago’s seminal work, Sir John Pendry first showed a practical way to design such a negative index material (NIM) using split ring resonators (SRR) and metal wires [2]. Research on NRI got a tremendous boost when in 2000 Sir Pendry showed that a slab of NRI material can amplify evanescent waves and thereby can lead to perfect lensing [3]. Note that focusing of propagating waves by such a slab had been shown by Veselago [1]. NRI was first experimentally demonstrated in 2001 by Smith et al. [4] in microwave frequency regime using an artificially created periodic array of SRRs and metal wires. Since then, there has been a flurry of research activities in negative index materials and their optical properties [5]. These designer materials are also known as left-handed material (LHM) or metamaterial, as the electric and magnetic field vectors and the propagation vector form a left-handed triplet [1, 4, 6]. These materials exhibit surprising and counterintuitive electromagnetic and optical effects like propagation of phase opposite to energy flow, reversal of both Doppler shift and Cherenkov radiation citevaselago, amplification of evanescent waves [3, 7], negative Goos-Hänchen shift [8], anomalous refraction [1], sub-wavelength focusing and superresolution [3, 9, 10], reversed circular Bragg phenomena [11], photon helicity inversion [12], $H$-field circulation and inverted $E$-field line in propagating structures [13], unusual photon tunneling effects [14]. Several other effects like switched field intensity locations in anisotropic transmission structures [15], fast and slow light [16, 17], lasing spasers [18] and optical nanocircuits [19] were also reported. There have been several other ways to realize left-handed materials based on classical electromagnetic principles, including chiral material [20–23] and photonic crystal structures [24–26].

Most of the metamaterials [5, 27] created so far using solid-state elements exhibit negative index in the long wavelength domain, namely microwave and tera-Hertz [5, 28] regime. Moreover, because of the underlying lattice structures, most of these materials are anisotropic in nature. Currently, there is tremendous research interest in designing new isotropic metamaterials at short wavelength or optical regime. In recent years coherent manipulation of the absorptive and dis-
perspective properties of a medium of atomic or molecular gases are being theoretically explored as a route to achieve isotropic negative index at optical frequency domain. In this context, Oktel and Müstecaplıoğlu [29] first proposed a three-level Λ-type scheme based on electromagnetically induced transparency (EIT) in order to achieve NRI. Since then, several coherent optical schemes using multilevel atoms or molecules have been proposed [12, 30–38]. Kästel [30] and Thommen [39] have suggested that a LHM can be obtained in an atomic four level system. Even a five-level atomic system has been studied [36] for this purpose. In recent years, there have been several suggestions [40–43] for creating LHM with atomic gases, using EIT and gain mechanism. Despite all these theoretical works, negative index in an atomic or molecular system is yet to be realized experimentally.

With the possibility of achieving negative refraction with suppressed absorption near EIT in mind, here we propose a new approach using a polar molecular gas possessing both permanent magnetic and electric dipole moments. Our system is schematically shown in Fig.1. We consider microwave-dressed states $|+\rangle$ and $|-\rangle$ which are linear superposition of two bare states $|a\rangle$ and $|b\rangle$. As the dressed states have mixed parities, both electric and magnetic dipole allowed transitions take place between either of two dressed states and a third level $|c\rangle$ with a definite parity. To be specific, we consider the molecule to be initially prepared in an absolute ground state $|a\rangle$, meaning it is electronically, vibrationally and rotationally in the lowest energy eigenstate. By exploiting the permanent dipole moment, this state is coupled to the rotational level $|b\rangle \equiv |v = 0, J = 1\rangle$ (where $v$ and $J$ are the vibrational and rotational quantum numbers, respectively) with a strong microwave laser $L_1$ leading to the two dressed states of two rotational levels. Two infrared lasers $L_2$ and $L_3$ are applied to couple these two dressed states to the level $|c\rangle \equiv |v = 2, J = 2\rangle$ forming a three-level Λ-type system as depicted in Fig.1(b). Here $v = 2$ for the the uppermost level is chosen in order to have appreciable Franck-Condon overlap integral with the vibrational ground state $|b\rangle$. We consider radiative decay of $|c\rangle$ to $|b\rangle$ with decay constant $\gamma$ due to electric dipole coupling.

Our scheme allows us to manipulate the values of $\epsilon_r$ and $\mu_r$ over a certain frequency band in infrared regime while minimizing the absorption suitably. Considering one of the infrared lasers (say, $L_3$) as the control field tuned between the upper dressed state $|-\rangle$ and $|c\rangle$ while the other laser ($L_2$) treated as a probe is tuned between the lower dressed state $|+\rangle$ and $|c\rangle$. Keeping the detuning of the control field fixed, we scan the frequency of the probe across the two-photon resonance. We find two distinct probe frequency regimes of NRI. The first regime, named as NRI-I is a dispersive
regime on the red side of the frequency of the transition $|+\rangle \leftrightarrow |c\rangle$ and has a broad width. The other regime dubbed as NRI-II lies close to the two-photon resonance or EIT regime and may lie in the blue side of the transition $|+\rangle \leftrightarrow |c\rangle$. The figure of merit, defined as the magnitude of ratio of the real to the imaginary part of the refractive index, can reach as high as 10 depending on the control field intensity and the mixing angle for dressing of the two lower ro-vibrational levels.

Another pertinent point is the proper implementation of causality via the Kramers-Kronig relations [6], since all such NRI materials are essentially lossy. A great deal of research has gone into how to choose the proper signs of the real and imaginary parts of the electric and magnetic susceptibilities and consequently the signs of both real and imaginary parts of the refractive index [44–46]. Note that absorption plays a deciding role on the feasibility of the exotic effects like Pendry lensing and superresolution [47, 48]. In our study we pay due attention to proper implementation of causality following Kinsler [46], in contrast to most of the recent studies on NRI in atomic and molecular systems.

Our work is motivated by the recent experiments [49–51] in producing cold polar molecules in absolute ground state having permanent electric as well as magnetic dipole moment. In contrast to other proposed three-level schemes, our scheme differs in several aspects. First, our scheme does not involve any electronically excited state, and so our model has relatively stronger immunity to spontaneous decays, offering greater opportunity for coherent manipulation. Second, because of microwave-dressing, in our model both electric and magnetic transitions are possible between the same two levels, unlike the Λ-type model [29] where the magnetic transition occurs between the two lower levels and the electric one between the middle and the uppermost level. Third, our model appears to be realizable with currently available technology of cold polar molecules.

The remainder of the paper is organized in the following. In section-II, we discuss our model and obtain the expressions for electric permittivity and magnetic permeability in terms of the density matrix elements. In section-III, we present our analytical and numerical results under certain parameters regime to obtain NIM in an ensemble of ground state polar molecules. Finally, we summarise the main findings of our study in conclusions.

II. THE MODEL

We consider a three-level scheme of electronically ground-state polar molecules possessing both permanent electric and magnetic dipole moments, interacting with three laser fields. The
configuration is depicted in Fig. 1. The state $|a\rangle$ is an absolute ground state meaning that it has vibrational quantum number $v = 0$ and rotational quantum number $J = 0$, while the next higher state $|b\rangle$ has $v = 0$ and $J = 1$. This means that these two states being in the same vibrationally ground-state, have opposite parity as their rotational quantum numbers differ by unity. So, owing to the permanent electric dipole moment, these two states are coupled via electric dipole transition with a strong microwave laser $L_1$ (possible in diode laser) of frequency $\omega_1$, leading to the formation...
of two dressed states

\[ |+\rangle = \cos \theta |a\rangle + \sin \theta |b\rangle \]  

(1)

\[ |-\rangle = -\sin \theta |a\rangle + \cos \theta |b\rangle \]  

(2)

where \( \theta \) is the mixing angle defined by \( \tan 2\theta = -\frac{\Omega_1}{\delta_1} \) with \( \Omega_1 \) being the Rabi frequency and \( \delta_1 = \omega_1 - \omega_{ba} \) the detuning of the laser frequency from the atomic frequency \( \omega_{ba} \) for the transition \( |b\rangle \leftrightarrow |a\rangle \). The two eigenvalues are

\[ \hbar \tilde{\omega}_+ = \frac{1}{2} \hbar (-\delta_1 - \sqrt{\delta_1^2 + \Omega_1^2}) \]  

and

\[ \hbar \tilde{\omega}_- = \frac{1}{2} \hbar (-\delta_1 + \sqrt{\delta_1^2 + \Omega_1^2}) \].

We restrict \( \theta \) to lie in the range \( 0 < \theta < \pi/2 \) implying \( \delta_1 < 0 \) and \( \tilde{\omega}_+ < \tilde{\omega}_- \). So, the two dressed states always remain to be of mixed parity. For \( \delta_1 = 0 \), \( |+\rangle \) and \( |-\rangle \) reduce to a symmetric and anti-symmetric combinations, respectively, of the two bare states \( |b\rangle \) and \( |a\rangle \).

The two dressed states are coupled to a third level \( |c\rangle \) having \( v = 2 \) and \( J = 2 \) with two infrared lasers \( L_2 \) and \( L_3 \) as schematically shown in Fig.1(b). The state \( |c\rangle \) differs in parity from \( |b\rangle \) but has the same parity as that of \( |a\rangle \). So, the components of the state \( |b\rangle \) in the two dressed states will be coupled to \( |c\rangle \) via electric dipole transition while the components of \( |a\rangle \) will be coupled to \( |c\rangle \) via magnetic dipole transition. However, since magnetic dipole Rabi coupling is smaller than the electric one by two orders of magnitude, we consider only the electric dipole coupling in calculating the atomic density matrix. Nevertheless, the important point here is that we can generate magnetic response out of purely electrical couplings as originally suggested by Pendry [2].

The Rabi couplings of the states \( |+\rangle \) and \( |-\rangle \) to \( |c\rangle \) are \( \Omega_+ = \Omega_2 \sin \theta \) and \( \Omega_- = \Omega_3 \cos \theta \), respectively; where \( \Omega_2(\Omega_3) \) is the Rabi coupling between the states \( |b\rangle \) and \( |c\rangle \) (\( |a\rangle \) and \( |c\rangle \)) due to the laser \( L_2 \) (\( L_3 \)). Since we are considering all the three levels to lie in electronic ground state manifold, the radiative damping or spontaneous decay rate \( \gamma \) from the upper level \( |c\rangle \) to the lower level \( |b\rangle \) is quite small. Since electric dipole coupling between \( |c\rangle \) and \( |a\rangle \) is forbidden by parity selection rule, there is no spontaneous emission from \( |c\rangle \) to \( |a\rangle \) due to electric dipole emission. Spontaneous emission linewidth is proportional to the square of the electric dipole matrix element. So, the linewidths for spontaneous emission from \( |c\rangle \) to \( |+\rangle \) and \( |-\rangle \) are \( \gamma_+ = \gamma \sin^2 \theta \) and \( \gamma_- = \gamma \cos^2 \theta \), respectively. In our model, we assume that the state \( |-\rangle \) can decay to \( |+\rangle \) with small damping constant \( \gamma_0 \).

At probe frequency \( \omega_2 \), the electric dipole transition (\( |b\rangle \leftrightarrow |c\rangle \)) and magnetic dipole transition (\( |a\rangle \leftrightarrow |c\rangle \)) can occur simultaneously, producing electric and magnetic polarization of the medium. The relative permittivity and permeability are defined by \( \epsilon_r = 1 + \chi_e \) and \( \mu_r = 1 + \chi_m \),
respectively; where $\chi_e$ and $\chi_m$ are the electric and magnetic susceptibility, respectively. The refractive index is $n = \pm \sqrt{\varepsilon_r \mu_r}$. Here the negative sign (-) before $\sqrt{\varepsilon_r \mu_r}$ is applicable only when both $\varepsilon_r$ and $\mu_r$ are real and negative or the phase velocity of the electromagnetic field opposes the velocity of energy flow of the field [46, 52]. For a dissipative system, both $\varepsilon_r$ and $\mu_r$ are complex. Let $\varepsilon_r = \varepsilon_r' + i\varepsilon_r''$ and $\mu_r = \mu_r' + i\mu_r''$, where primed and double-primed quantities refer to the real and imaginary parts, respectively. The real and imaginary parts of $\varepsilon_r$ or $\mu_r$ are related to each other by causality conditions expressed by celebrated Kramers-Kronig relations. According to the general criterion [46], the refractive index $n$ becomes negative when the quantity $K_E = \varepsilon_r' |\mu_r| + \mu_r' |\varepsilon_r|$ is negative.

We solve the density matrix master equation of the system including dissipative terms in Lindblad form. For our system, we have

$$\chi_e = \frac{2Nd_{cb}\rho_{bc}}{\varepsilon_0 E_2}$$

(3)

$$\chi_m = \frac{2N\mu_{ca}\rho_{ac}}{H_2}$$

(4)

where $H_2 = \sqrt{\frac{\varepsilon_r\varepsilon_0}{\mu_r\mu_0}E_2} = \sqrt{\frac{\varepsilon_r E_p}{\mu_r c \mu_0}}$ with $\mu_0$ being the permeability of the vacuum and $c$ the speed of light in free space. Here $E_2$ is electric field amplitude of the probe laser $L_2$, $N$ is the number density and $\varepsilon_0$ is vacuum permittivity. The quantity $d_{cb}$ is permanent electric dipole moment between states $|c\rangle$ and $|b\rangle$ and $\mu_{ca}$ is permanent magnetic moment between states $|c\rangle$ and $|a\rangle$, respectively. $\mu_r$ can be expressed in terms $\varepsilon_r$ and other parameters as a solution of the quadratic equation

$$\mu_r^2 - \mu_r(2 + R) + 1 = 0$$

(5)

where

$$R = \frac{4N^2 \mu_0 \mu_{ca}^2 \rho_{ac}^2}{\varepsilon_0 E_p^2}$$

(6)

The solutions Eq.5 are

$$\mu_r = \frac{(2 + R) \pm \sqrt{(2 + R)^2 - 4}}{2}$$

(7)

where $\chi_e$ or $\chi_m$ depend on bare-state density matrix elements which are related to the dressed-state density matrix elements $\rho_{+c}$ and $\rho_{-c}$ by

$$\rho_{bc} = \sin \theta \rho_{+c} + \cos \theta \rho_{-c}$$

(8)

$$\rho_{ac} = \cos \theta \rho_{+c} - \sin \theta \rho_{-c}$$

(9)
which can be readily obtained by using Eqs. 1 and 2.

The effective time-independent Hamiltonian under rotating wave approximation (RWA) and electric dipole approximation can be written as

$$H = \hbar (\tilde{\omega} + \delta_{23}) |+\rangle \langle +| + \hbar \tilde{\omega} |\rangle \langle -| + \hbar (\omega_c - \omega_3) |c\rangle \langle c| + \frac{\hbar}{2} \Omega_+ |c\rangle \langle +| + \frac{\hbar}{2} \Omega_- |c\rangle \langle -| + h.c$$

where $\delta_{23} = \omega_2 - \omega_3$. From this Hamiltonian, the density matrix equations of motion can be expressed as

$$\dot{\rho}_{--} = -\frac{i}{2} \Omega_- [\rho_{c-} - \rho_{-c}] + \gamma_- \rho_{cc} - \gamma_0 \rho_{--}$$

$$\dot{\rho}_{++} = -\frac{i}{2} \Omega_+ [\rho_{c+} - \rho_{+c}] + \gamma_+ \rho_{cc} + \gamma_0 \rho_{--}$$

$$\dot{\rho}_{cc} = \frac{i}{2} \Omega_+ [\rho_{c+} - \rho_{+c}] + \frac{i}{2} \Omega_- [\rho_{c-} - \rho_{-c}] - \gamma \rho_{cc}$$

$$\dot{\rho}_{+-} = i[\delta_+ - \delta_-] \rho_{--} - \frac{i}{2} \Omega_- \rho_{+c} + \frac{i}{2} \Omega_+ \rho_{-c} - \frac{\gamma_0}{2} \rho_{--}$$

$$\dot{\rho}_{-+} = -i[\delta_+ - \delta_-] \rho_{++} + \frac{i}{2} \Omega_- \rho_{+c} - \frac{i}{2} \Omega_+ \rho_{-c} - \frac{\gamma_0}{2} \rho_{--}$$

$$\dot{\rho}_{+-} = -i \delta_+ \rho_{+c} - \frac{i}{2} \Omega_+ (\rho_{cc} - \rho_{++}) + \frac{i}{2} \Omega_- \rho_{-c} - \frac{\gamma_0}{2} \rho_{++}$$

$$\dot{\rho}_{++} = i \delta_+ \rho_{cc} + \frac{i}{2} \Omega_+ (\rho_{cc} - \rho_{++}) - \frac{i}{2} \Omega_- \rho_{-c} - \frac{\gamma_0}{2} \rho_{cc}$$

$$\dot{\rho}_{-c} = -i \delta_- \rho_{-c} + \frac{i}{2} \Omega_+ \rho_{cc} - \frac{i}{2} \Omega_- \rho_{-c} - \frac{\gamma_0}{2} \rho_{-c}$$

$$\dot{\rho}_{-c} = i \delta_- \rho_{-c} + \frac{i}{2} \Omega_+ \rho_{cc} - \frac{i}{2} \Omega_- \rho_{-c} - \frac{\gamma_0}{2} \rho_{-c}$$

Note that $\gamma = \gamma_+ + \gamma_-$ is the total decay rate of $|c\rangle$. The above density matrix equations are constrained by $\rho_{--} + \rho_{++} + \rho_{cc} = 1$ and $\rho_{ij} = \rho_{ji}^\dagger$. Here, two detuning parameters are defined as $\delta_+ = \omega_2 - (\omega_c - \tilde{\omega}_+)$, $\delta_- = \omega_3 - (\omega_c - \tilde{\omega}_-)$. Although we solve numerically the density matrix equation in steady state, for gaining insight we derive exact analytical expressions for the steady-state density elements in the appendix-A.

The equations 3, 4, 8 and 9 show that, in order to make the real parts of both $\chi_c$ and $\chi_m$ negative, the real part of $\rho_{+c}$ must be negative satisfying the inequality $\text{Re}[\rho_{+c}] < \tan \theta \text{Re}[\rho_{-c}]$. This means that either the real parts of both $\rho_{+c}$ and $\rho_{-c}$ should be negative satisfying the inequality or the real part of $\rho_{+c}$ is negative but the real part of $\rho_{+c}$ is positive satisfying $| \text{Re}[\rho_{+c}] | > \tan \theta \text{Re}[\rho_{-c}]$. Note that here we have restricted $\theta$ within the range $0 < \theta < \pi/2$. So, the problem at hand is to search for an appropriate parameter regime where these conditions are fulfilled. In the next section, we discuss in some detail such possibility both analytically and numerically.
FIG. 2. (a) The diagonal density matrix elements $\rho_{ii}$ ($i = +, -, c$) as a function of $\delta_+/\gamma$ for $\gamma_0 = 0.1\gamma$, $\theta = \pi/\gamma$, $\Omega_2 = 0.5\gamma$, $\Omega_3 = 2.0\gamma$ and $\delta_- = 0.5\gamma$. (b) The ratio $r = w_-/w_+ = (\rho_{++} - \rho_{cc})/(\rho_{++} - \rho_{cc})$ of the $w_-$ to $w_+$ as a function $\delta_+/\gamma$ for the same parameters. The value of $r_d$ is 0.014, $r$ becomes equal to $r_d$ at $\delta_+ = 0.55\gamma$ which is shifted from the two-photon resonance $\delta_+ = \delta_- = 0.5$ due to finite decoherence decay rate $\gamma_0/2$.

III. RESULTS AND DISCUSSIONS

It is well known that, for $\gamma_0 = 0$ and $\Delta = 0$ (two-photon resonance), there is dark-state resonance at which $\rho_{cc} = 0$ or equivalently $\text{Im}[\rho_{++}] = 0$ and $\text{Im}[\rho_{--}] = 0$ meaning that all the absorption absolutely vanishes. All the population is then coherently shared by the two lower states $|+\rangle$ and $|−\rangle$ with population ratio given by $\rho_{--}/\rho_{++} = w_-/w_+ = r = \Omega_-^2/\Omega_+^2$. This happens due to the quantum interference between optical transition pathways connecting the uppermost state $|c\rangle$ to the lower states $|+\rangle$ and $|−\rangle$. It is clear from the expression (A7) that at such dark-state resonance conditions, $\rho_{+c} = 0$ implying that both the real and imaginary parts vanish. The question here is whether there exists a dispersive regime close to the dark-state resonance or quasi-dark state resonance ($\gamma_0 \ll \gamma$, but $\gamma_0 \neq 0$) where the desirable negative index of refraction is possible.

We have derived exact analytical expressions for the density matrix element $\rho_{+c}$ given by the
equation (A7) in the appendix-A. Setting $\gamma_0 = 0$, \(\text{Re}[\rho_{+c}]\) can be expressed in a compact form

$$\text{Re}[\rho_{+c}] = F_+ [A + B(r - r_d)] \quad (19)$$

where

$$F_+ = \frac{w_+ \Omega_+}{8|\kappa_+|^2 [(\Delta - \Delta_{\text{shift}})^2 + (\Gamma/2)^2]} \quad (20)$$

$$A = [4\delta_+ \Delta (\Delta - \Delta_{\text{shift}}) + \Delta \gamma_0 \Gamma] \quad (21)$$

$$B = \frac{\Omega_+^2}{|\kappa_-|^2} [(\Delta - \Delta_{\text{shift}})(\delta_+ \delta_- + \gamma_0 \gamma_+ / 4) + \Gamma / 4 (\gamma_- \gamma_- - \gamma_+ \gamma_+)] \quad (22)$$

Here $r_d = \Omega_+^2 / \Omega_-^2$ and

$$\Delta_{\text{shift}} = \frac{1}{4} \left[ \frac{\Omega_+^2 \delta_+}{|\kappa_+|^2} - \frac{\Omega_-^2 \delta_-}{|\kappa_-|^2} \right] \quad (23)$$

$$\Gamma = \gamma_0 + \frac{1}{2} \left[ \frac{\Omega_+^2 \gamma_+}{|\kappa_+|^2} + \frac{\Omega_-^2 \gamma_-}{|\kappa_-|^2} \right] \quad (24)$$

The structure of equations A5 and A6 suggest that $\rho_{-c}$ can be obtained from the expression of $\rho_{+c}$ by changing all subscripts ‘+’ $\rightarrow$ ‘-’ and vice versa. So, substituting these expressions in equations 8 and 9, we obtain exact analytical expressions for $\rho_{bc}$ and $\rho_{ac}$. As the Eq. (19) suggests and the Fig.2 illustrates, near two-photon resonance (\(|\Delta| < < \gamma\)) and $\delta_+ > 0$, there is a small frequency domain of the probe for which \(\text{Re}[\rho_{+c}]\) can be negative. Here for notational convenience, we use $\rho_{ij} \equiv \rho_{ji}$. This may happen when $r < r_d$ with $B > 0$ and $A << B$. On the other hand, for $\delta_+ < 0$ there may exist a broad rage of frequencies where \(\text{Re}[\rho_{+c}]\) becomes negative. Note that for $\Omega_2 << \Omega_3$, $\Delta_{\text{shift}}$ will be negative for small $|\delta_+|$ and $\delta_- > 0$, but for large $|\delta_+|$ it may become negative.

Having discussed the analytical results, we next turn our attention to numerical results which do corroborate our analytical findings. Making use of the numerical solutions of density matrix equations in steady state, we calculate the the complex relative permittivity $\epsilon_r$, the magnetic permeability $\mu_r$ and the index of refraction $n$. The figure of merit (FOM) of a negative index material can be defined as $\text{FOM} = |\text{Re}[n]/\text{Im}[n]|$ that characterizes the performance of a negative index material. Although, numerically one obtains two values of $\mu_r$, only one of them satisfies Kramers-Kronig relations ensuring the causality in the system, and so the other one can be discarded. In all our numerical calculations, we use the number density $N = 5 \times 10^{18}$ cm$^{-3}$, dipole moment
FIG. 3. (a) and (c): The real parts \( \epsilon'_r \) and \( \mu'_r \) (red, solid) and the imaginary parts \( \epsilon''_r \) and \( \mu''_r \) (blue, dotted) of \( \epsilon_r \) and \( \mu_r \) as a function of dimensionless detuning \( \delta_+/\gamma \). (b): The real part \( n_r \) and the imaginary part \( n_i \) of the refractive index \( n \) as a function \( \delta_+/\gamma \); (d): The ratio \( n_r/n_i \) as a function of \( \delta_+/\gamma \). The other parameters are same as in Fig. 2. NRI-I and NRI-II indicate the two frequency regimes of negative refractive index.

\[ d_{cb} = 0.4 \text{ Debye}, \mu_{ca} = 2\mu_B, \theta = \pi/7. \] The values of permanent dipole and magnetic moments correspond to polar molecule NaLi in triplet ground state. In our calculations, we scale all the frequency parameters by spontaneous linewidth \( \gamma \) for transition \( |c\rangle \rightarrow |a\rangle \). The complex refractive index is \( n = n_r + in_i \), where \( n_r \) and \( n_i \) are the real and imaginary parts of \( n \).

In the subplots (a) and (c) of Fig. 3, we show the variation of the real parts \( \epsilon'_r \) and \( \mu'_r \) (red, solid) and imaginary parts \( \epsilon''_r \) and \( \mu''_r \) (blue, dotted) as a function of \( \delta_+ \) while the subplots (b) and (d) of the same figure show the variation of \( n_r \) and \( n_i \) and their ratio as a function of \( \delta_+ \). These plots clearly demonstrate the two frequency regimes of NRI, marked as NRI-I and NRI-II. For the stated input parameters, the maximum FOM turns out to be about 9 in NRI-I, while the maximum FOM is found to be smaller in NRI-II. We have found that the FOM changes with the mixing angle \( \theta \), the maximum FOM of about 10 occurs in NRI-I for \( \theta \approx 24^\circ \). For too small or too large value of \( \theta \), FOM is degraded and the NRI regimes shrink. As equation 9 indicates, depending on the system parameters, the imaginary part of \( \rho_{ac} \) can be negative even if the imaginary parts of \( \rho_{+c} \) and \( \rho_{-c} \) are positive. This means that there can be gain in magnetic response which is reflected in the plot of Fig. 3(b).
In Fig. 4, we have plotted the same as in Fig. 3 but for higher value of the control field Rabi frequency $\Omega_3 = 5\gamma$. In comparison to Fig. 3, we notice that the maximum FOM in NRI-I reduces while that in NRI-II increases marginally, also the width of the NRI-II increases. We have found that for too small or too large values of $\Omega_3$, NRI regimes vanish. So, there are optimum values of $\Omega_3$ and $\theta$ for which one can maximize the NRI effect and the FOM.

IV. CONCLUSIONS

We have proposed a scheme to realize negative refraction in an ensemble of gaseous polar molecules having both permanent electric and magnetic dipole moment. Mixed parity dressed states are created by applying a microwave field between two rotational levels of ground vibrational states. Along with two more IR lasers coupling the dressed states to the excited state enables one to extract both electric and magnetic response from the system. The underlying EIT level scheme ($\Lambda$ system) and the associated low losses and flexibility for dispersion control are shown to lead to two distinct regimes of NRI behavior. One of them lies in the red detuned side of the probe frequency having a broad frequency band. The other regime is near the two-photon resonance, albeit with lower FOM in a narrow frequency band. We report the possibility of having a FOM as high as 10 in the first NRI regime. Our calculations are based on a density matrix approach.
with numerical and exact analytical results for the steady states. The permittivity and permeability calculations are carried out in full conformity of the Kramers-Kronig relations ensuring causality. We have made use of the permanent electric and magnetic dipole moments of ground-state polar molecules to build-up our model. Of late, cold polar molecules emerge as a novel system for applications in coherent optics. In our model, micro-wave-dressed states of the two rotational levels play an essential role in generating both electric and magnetic response at the same frequency. Field-dressing of atomic levels have been used over the years for coherent manipulation of the optical properties of the atoms. In a recent work by Rempe’s group [53], atomic dressed states are employed to build a multi-level atomic scheme to control nonlinear response of the system at a quantum level. Since atoms do not have permanent electric dipole moment, fitting our model into an atomic system seems to be unlikely. However, our model may be extended to include electronically excited manifold of cold polar molecules to obtain NRI at higher frequencies beyond infrared domain.

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Appendix A: Steady-state solution of the density matrix

In the steady-state, the Eqs. 10 and 11 result in

\[ \rho_{cc} = \frac{1}{\gamma_-} \{ \Omega_- \text{Im}[\rho_{-c}] + \gamma_0 \rho_{--} \} = \frac{1}{\gamma_+} \{ \Omega_+ \text{Im}[\rho_{+c}] - \gamma_0 \rho_{+-} \} \] (A1)

The Eqs. 15 and 17 in the steady-state yield

\[ \rho_{+c} = \frac{1}{2(\delta_+ - i\frac{\gamma}{2})} \left[ -\Omega_+ (\rho_{cc} - \rho_{++}) + \Omega_- \rho_{+-} \right] \] (A2)

and

\[ \rho_{-c} = \frac{1}{2(\delta_- - i\frac{\gamma}{2})} \left[ -\Omega_- (\rho_{cc} - \rho_{--}) + \Omega_+ \rho_{-+} \right] \] (A3)

respectively. Substituting Eqs. (A2) and (A3) in Eq. 14, we obtain the steady-state value of

\[ \rho_{+-} = \frac{\Omega_+ \Omega_- \left[ w_+/\kappa_+ - w_-/\kappa_- \right]}{4(\Delta - i\gamma_0/2) - [\Omega_+^2/\kappa_+ - \Omega_-^2/\kappa_-] / \kappa_-} \] (A4)
where $\Delta = \delta_+ - \delta_-$ is the two-photon detuning, $w_+ = \rho_{++} - \rho_{cc}$, $w_- = \rho_{--} - \rho_{cc}$, $\kappa_+ = \delta_+ - i\frac{\gamma}{2}$ and $\kappa_- = \delta_- - i\frac{\gamma_0}{2}$. Substitution of Eq. (A4) in Eq. (A2) and Eq. (A3) leads to

$$\rho_{+c} = \frac{1}{2\kappa_+} \Omega_+ \left[ w_+ + \frac{\Omega_+^2 [w_+ / \kappa_+ - w_- / \kappa_-]}{4(\Delta - i\gamma_0/2) - [\Omega_+^2 / \kappa_+ - \Omega_-^2 / \kappa_-]} \right]$$  \hspace{1cm} (A5)

$$\rho_{-c} = \frac{1}{2\kappa_-} \Omega_- \left[ w_- + \frac{\Omega_-^2 [w_+ / \kappa_+ - w_- / \kappa_-]}{4(\Delta + i\gamma_0/2) - [\Omega_+^2 / \kappa_+ - \Omega_-^2 / \kappa_-]} \right]$$  \hspace{1cm} (A6)

By taking $w_+$ and $w_-$ outside the third bracket of Eqs. (A5) and (A6) respectively, both these equations can be expressed in terms of the ratio $r = w_- / w_+$ of the two population inversions. Thus we have

$$\rho_{+c} = \frac{w_+}{2\kappa_+} \Omega_+ \left[ 1 + \frac{\Omega_+^2 [1/ \kappa_+ - r / \kappa_-]}{4(\Delta - i\gamma_0) - [\Omega_+^2 / \kappa_+ - \Omega_-^2 / \kappa_-]} \right]$$  \hspace{1cm} (A7)

$$\rho_{-c} = \frac{w_-}{2\kappa_-} \Omega_- \left[ 1 + \frac{\Omega_-^2 [1/ \kappa_+ - 1 / \kappa_-]}{4(\Delta + i\gamma_0/2) - [\Omega_+^2 / \kappa_+ - \Omega_-^2 / \kappa_-]} \right]$$  \hspace{1cm} (A8)

Equation (A1) provides

$$\frac{\Omega_- \text{Im}[\rho_{-c}] + \gamma_0 \rho_{-c}}{\Omega_+ \text{Im}[\rho_{+c}] - \gamma_0 \rho_{+c}} = \frac{\gamma_-}{\gamma_+}$$  \hspace{1cm} (A9)

Equations (A7) and (A8) show that the left hand side Eq. (A9) will be a function of $r$ and $\rho_{--}$. The population inversions $w_+$ and $w_-$ can be evaluated by using the Eq. (A9), (A1) and the normalization condition. $\rho_{cc} + \rho_{++} + \rho_{--} = 1$.

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