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Coherent control of refractive index in far-detuned lambda systems

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Enhancement and control of the index of refraction in a mixture of two three-level atomic species that form a pair of far detuned lambda schemes under two-photon resonance has been studied. We employ the density matrix approach to properly take into account population relaxation and describe the interaction of each lambda system with the electromagnetic fields. Both lambda systems are driven by a corresponding far-detuned coherent field at one atomic transition and probed by the same weak field. In the dressed state basis, it represents a superposition of effective two-level subsystems with the positions, widths and amplitudes of the resonances controlled by the driving fields and allows for efficient control of the susceptibility of the total system; leading to refractive index enhancement with vanishing absorption in the absence of amplification. We analyze the experimental implementation of such a system in a cell of Rb atoms at natural abundance of isotopes. An upper limit estimate of the refractive index enhancement is obtained.

I. INTRODUCTION

Tightly focused laser radiation allows for the selective addressing of small regions of a medium. In microscopy, it is used to image tiny objects such as biological cells, organic molecules or NV centers in diamond. In lithography, it is used for production of miniature semiconductor integral circuits. In information processing, it is used to provide multiple parallel optical channels. For all of these applications, a key issue is the spatial resolution which is defined by the minimum spot size the laser radiation can be focused to. This focal spot size is fundamentally limited by the wavelength of light in the medium, λ, which depends on the refractive index n as following: $\lambda = \lambda_{\rm vac}/n$. Thus, high refractive index (RI) is very important for achieving high spatial resolution in all of these applications. Materials with enhanced RI on demand would also be important for phase shifters, interferometers, and magnetic Faraday rotators.

Index of refraction characterizes the response of a medium to electromagnetic radiation and hence it is strongly enhanced near the atomic resonance. However, if a medium is in thermal equilibrium the enhancement of RI near the atomic resonance is accompanied by an enhancement of absorption. Such that, when the maximal contribution from the atomic resonance to the RI is reached, the contribution to the absorption is the same. As a result, a 2π phase shift and e-fold absorption take place at the same distance in a medium, which prevents usage of the obtained RI in transmission experiments. In a inverted medium, high RI in the vicinity of the atomic resonance is accompanied by high gain. However, even higher gain is present at exact resonance which makes such a system unstable and again non-suitable for high index applications.

A mixture of atomic species will provide overlapping absorption and gain if the difference in resonance frequencies is on the scale of the linewidth and one of the atomic species is inverted. A proper overlap could result in high refractive index with vanishing absorption for a weak field properly tuned between two atomic resonances. However the difficulties associated with the practical implementation of such a combined system (finding proper species, providing for an even mixture, and providing population inversion for one species while avoiding spatial fluctuations of density and population exchange, etc.) would hardly be surmountable [1].

We consider here an idea, to use coherent effects in a mixture of different species to induce strong and overlapping electromagnetic responses to provide high refractive index with vanishing absorption. The use of a coherent preparation of a medium for elimination of absorption and index enhancement was pioneered by Scully [2], which was further generalized in [3] by including the density dependent near dipole-dipole interactions. A number

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of other three and four level schemes followed. They involved resonant driving at one or two atomic transitions and probing in a way that resonant enhancement of refractive index enhancement is accompanied by vanishing absorption [1]. It was expected that $\chi \sim 1$ without absorption would be possible in high density ($N\lambda^3 \approx 50$) alkali but with a undesirable amplification region. Two schemes, however a so called double dark resonance and degenerate double Λ-scheme allowed for the elimination of the gain region though under a rather exotic and narrow range of parameters [4]. These developments led to a proof of principal experiment in Rb vapors, with a density of 10^{12} cm^{-3}, which showed refractive index enhancement with vanishing absorption. Although $N\lambda^3 \approx 1$ was achieved, the magnitude of index enhancement in this experiment was quite low, of an order of $\Delta n = 10^{-4}$ [5].

Recently, a new scheme for coherent control and index enhancement was suggested by Yavuz [6]. This scheme is based on a resonant four level system involving two Raman transitions optically pumped into the ground state and driven by two far off-resonant control fields forming two Λ-systems with the same probe field. The dispersive and absorption characteristics in such a system as functions of two-photon detuning essentially interchange so that the maximum resonant refractive index is accompanied by vanishing absorption. Similar to the previous proposals involving resonant driving the effect was attributed to interference and an index on the order of 10 for alkali vapors with densities (10^{17} cm^{-3}) was predicted. Undesirable gain in the vicinity of vanishing absorption was also present.

We study a similar but simpler system. It represents itself as a mixture of two three-level atomic species each driven by a corresponding far-detuned coherent field at one atomic transition and probed by the same weak field at a adjacent transition in the vicinity of two-photon resonance. This system was used in a proof of principal experiment which showed an enhancement of $\Delta n = 2.2{\cdot}10^{-7}$ [7]. In this work, we analyze the physical mechanisms responsible for index enhancement in the case of far off-resonant driving and its limitations. We present a simple physically intuitive picture in the decaying dressed state basis [8]. We then extend this analysis to include inhomogeneous broadening. In order to understand the limitations of refractive index enhancement in general and in this two lambda scheme, we give a proper treatment of collisional broadening to show that any given mixture has a maximum possible index enhancement. We then give a detailed analysis of the proposed system in a cell of Rb atoms with natural abundance (72% ^{85}Rb and 28% ^{87}Rb) and compare to the experimental results from [7]. An upper limit due to the dipole-dipole broadening at high atomic density of the refractive index enhancement in such a system is estimated as $\Delta n \simeq .2$.

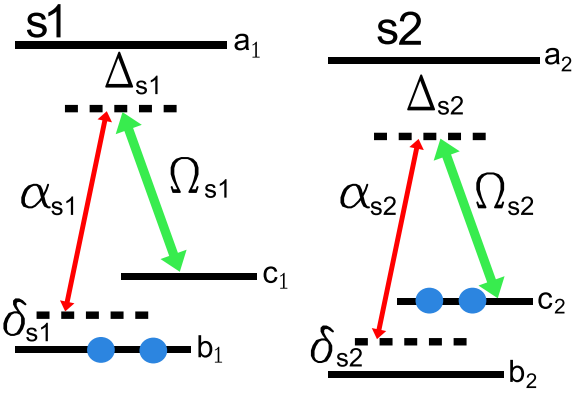

FIG. 1. (Color online) Mixture of two three-level Λ systems. The initially populated level is indicated by the dots.

II. THREE-LEVEL COHERENTLY DRIVEN SYSTEM: DENSITY MATRIX FORMALISM

We consider a mixture of two atomic species with the density of atoms for each species N_{s1} and N_{s2} being free parameters. Each of them is represented as a three-level system, labeled by si $\in$ {s1, s2}, with one excited state and two ground state sub-levels labeled $|a_i\rangle$, $|b_i\rangle$ and $|c_i\rangle$, correspondingly (see Fig. 1). The system, driven by a pair of coherent fields with frequencies ω_{si} and Rabi frequencies Ω_{si}, is probed by a weak field with frequency ω_{pr} and Rabi frequency α_{si}. The driving field Rabi frequencies $\Omega_{si} = d^{ac}_{si}E_{si}/(2\hbar)$ are defined by the applied electric field E_{si} and the dipole moment of the transition d^{ac}_{si}. The Rabi frequencies of the probe field $\alpha_{si} = d^{ab}_{si}E_{pr}/(2\hbar)$ in each system may be different because the dipole moment of the probed transition in s1-system can be different from the dipole moment of the probed transition in s2-system. All-fields are far off-resonance from the atomic transitions so that one-photon contributions are negligible, implying $\Delta_{si} \gg \Omega_{si}$. The frequencies of the fields are chosen in such a way that they result in one two-photon transition in each three-level system involving one photon from the probe field and one photon from the corresponding driving field. So each three-level system forms a Λ scheme with the same probe and corresponding driving field (see Fig. 1). Each three-level system is initially prepared in one of the two ground state sublevels via optical pumping as indicated in Fig. 1. So the first scheme exhibits two-photon absorption for the probe field while the second scheme provides two-photon gain.

The index of refraction and absorption coefficient can be found if the complex susceptibility is known. In our system the total complex susceptibility is equal to the sum of individual contributions from each of the three-level systems. The complex susceptibility itself can be calculated if the optical coherence excited by a weak probe field is known:

$$\chi_{si} = \frac{3}{8\pi^2}\gamma_r^{si}N_{si}\lambda_{pr}^3\frac{\sigma_{ab}^{si}}{\alpha_{si}}, \qquad (1)$$

where $\gamma_{\rm r}^{\rm si} = \frac{4}{3}\frac{(d_{\rm si}^{\rm ab})^2}{4\pi\epsilon_0\hbar}\frac{(2\pi)^3}{\lambda_{\rm pr}^3} = 8\pi^2(d_{\rm si}^{\rm ab})^2/(3\epsilon_0\hbar\lambda_{\rm pr}^3)$ is the radiative decay rate of the probed transition, $\lambda_{\rm pr}$ is the wavelength of the probe, and $\sigma_{\rm ab}^{\rm si}$ is the coherence of the probed transition.

We use density matrix formalism and the rotating wave approximation to calculate optical coherence induced by a weak probe field applied to a↔b transition in a three-level system driven off-resonance by a field applied to an adjacent transition a↔c. This formalism allows for including dephasing rates $\gamma_{\alpha\beta}^{\rm si}$ at $\alpha\leftrightarrow\beta$ transitions, where $\{\alpha,\beta\} = \{a,b,c\}$. Similar to [6] we assume that driving fields do not disturb an initial population distribution, which implies either sufficiently short interaction time $t_{\rm int}\Omega_{\rm si}^2/\Delta_{\rm si} \ll 1$ or sufficiently strong optical pumping through some additional levels (not indicated in the Fig. 1).

The slowly varying amplitude of the optical coherence induced by a weak probe field $\alpha_{\rm si} \ll \Omega_{\rm si}, \gamma_{\rm ab}^{\rm si}$ is found to be:

$$\sigma_{\rm ab}^{\rm s1} = \frac{\left(\delta_{\rm s1} - i\gamma_{\rm cb}^{\rm s1}\right)\alpha_{\rm s1}}{\left(\delta_{\rm s1} + \Delta_{\rm s1} - i\gamma_{\rm ab}^{\rm s1}\right)\left(\delta_{\rm s1} - i\gamma_{\rm cb}^{\rm s1}\right) - \left|\Omega_{\rm s1}\right|^2}, \qquad (2)$$

for s1-system presented on Fig. 1 (left), while for s2-system presented on Fig. 1 (right) it is found to be:

$$\sigma_{\rm ab}^{\rm s2} = -\frac{\left|\Omega_{\rm s2}\right|^2\left(\Delta_{\rm s2} + i\gamma_{\rm ac}^{\rm s2}\right)^{-1}\alpha_{\rm s2}}{\left(\delta_{\rm s2} + \Delta_{\rm s2} - i\gamma_{\rm ab}^{\rm s2}\right)\left(\delta_{\rm s2} - i\gamma_{\rm cb}^{\rm s2}\right) - \left|\Omega_{\rm s2}\right|^2}. \qquad (3)$$

In these equations, we introduced the following parameters for si-system: $\Delta_{\rm si} = \omega_{\rm ab}^{\rm si} - \omega_{\rm cb}^{\rm si} - \omega_{\rm si}$ and $\delta_{\rm si} = \omega_{\rm si} + \omega_{\rm cb}^{\rm si} - \omega_{\rm pr}$ are the one- and the two-photon detunings for si-drive field, respectively.

Finally, we can write down the expression for complex susceptibility of the system:

$$\chi\left(\omega_{\rm pr}\right) = \frac{3\lambda_{\rm pr}^3}{8\pi^2}\left(\frac{{\rm N}_{\rm s1}\gamma_{\rm r}^{\rm s1}\left(\delta_{\rm s1} - i\gamma_{\rm cb}^{\rm s1}\right)}{\left(\delta_{\rm s1} + \Delta_{\rm s1} - i\gamma_{\rm ab}^{\rm s1}\right)\left(\delta_{\rm s1} - i\gamma_{\rm cb}^{\rm s1}\right) - \left|\Omega_{\rm s1}\right|^2} - \frac{{\rm N}_{\rm s2}\gamma_{\rm r}^{\rm s2}\left|\Omega_{\rm s2}\right|^2\left(\Delta_{\rm s2} + i\gamma_{\rm ac}^{\rm s2}\right)^{-1}}{\left(\delta_{\rm s2} + \Delta_{\rm s2} - i\gamma_{\rm ab}^{\rm s2}\right)\left(\delta_{\rm s2} - i\gamma_{\rm cb}^{\rm s2}\right) - \left|\Omega_{\rm s2}\right|^2}\right). \qquad (4)$$

In the following sections, individual contributions are discussed and physical insights are given.

III. DRESSED-STATE ANALYSIS: EFFECTIVE TWO-LEVEL SYSTEMS

The slowly varying amplitude of the optical coherence induced by a weak probe field in a Λ-configuration is inversely proportional to a quadratic polynomial in terms of the two-photon detuning $\delta_{\rm si}$. Zeros of this polynomial correspond to the two main contributions to the optical coherence. Expanding the coherence into Lorentzians defined by each zero gives the decaying dressed states as has been previously discussed in [8].

For the case of a far-detuned driving field, $\Delta_{\rm si} \gg \Omega_{\rm si}, \gamma_{\rm ab}^{\rm si}, \gamma_{\rm cb}^{\rm si}$, these resonance contributions are far-detuned as well and associated with one- and two-photon resonances. This is clearly seen for s1 after expanding Eq. 2 in terms of the small parameter $\xi_{\rm si} = |\Omega_{si}|^2/\Delta_{si}^2$:

$$\sigma_{ab}^{\rm s1} = \frac{\alpha_{\rm s1}(1-\xi_{\rm s1})}{\delta_{\rm s1} - \Delta_{\rm s1}(1-\xi_{\rm s1}) - i[\gamma_{ab}^{\rm s1}(1-\xi_{\rm s1}) + \gamma_{cb}^{\rm s1}\xi_{\rm s1}]} + \frac{\alpha_{\rm s1}\xi_{\rm s1}}{\delta_{\rm s1} - \Delta_{\rm s1}\xi_{\rm s1} - i[\gamma_{cb}^{\rm s1}(1-\xi_{\rm s1}) + \gamma_{ab}^{\rm s1}\xi_{\rm s1}]}. \qquad (5)$$

If $\Delta_{\rm s2} \gg \gamma_{\rm ac}^{\rm s2}$ as well, a similar expression can be found for s2 with the exception that the two photon amplitude is now negative and therefore provides gain due to the population in level $|c_2\rangle$; while the one photon amplitude is different since the feature would no longer be present in the absence of a control field:

$$\sigma_{ab}^{\rm s2} = \frac{\alpha_{\rm s2}\xi_{\rm s2}}{\delta_{\rm s2} - \Delta_{\rm s2}(1-\xi_{\rm s2}) - i[\gamma_{ab}^{\rm s2}(1-\xi_{\rm s2}) + \gamma_{cb}^{\rm s2}\xi_{\rm s1}]} + \frac{-\alpha_{\rm s2}\xi_{\rm s2}}{\delta_{\rm s2} - \Delta_{\rm s2}\xi_{\rm s2} - i[\gamma_{cb}^{\rm s2}(1-\xi_{\rm s2}) + \gamma_{ab}^{\rm s2}\xi_{\rm s2}]}. \qquad (6)$$

Furthermore, our probe field $\omega_{\rm pr}$ is tuned to the vicinity of two-photon resonance ($\delta_{\rm si} \ll \Delta_{\rm si}$) therefore the contribution from the one photon-resonance of the second system can always be neglected, while the one photon contribution of the first system can be neglected when $\gamma_{\rm ab}^{\rm s1}/\Delta_{\rm s1} \ll \xi_{\rm s1}$.

When the low frequency coherence decays slower than the optical one $\gamma_{\rm ab}^{\rm si} \gg \gamma_{\rm cb}^{\rm si}$, the contribution from the two-photon resonance can be nearly as large as the contribution from the one-photon resonance. This would simply require $\xi_{\rm si} \geq \gamma_{\rm cb}^{\rm si}/\gamma_{\rm ab}^{\rm si}$. Hence, both of our three level schemes behave as effective two level schemes with susceptibilities on the same order as the ones for the original transition. The presence of the drive fields allow for the control of the strength, width, and position of the resonances. This, in turn, leads to the manipulation of the atomic responses of the individual systems (see Fig. 2). We will use this flexibility combined with appropriate mixing of the species to obtain enhanced refractive index without absorption.

At first, we take an approach similar to [6]. It is based on the absorption and amplification resonances having the same magnitude and width; while being separated by the full width at half maximum (FWHM). This arrangement results in the absorption being compensated by

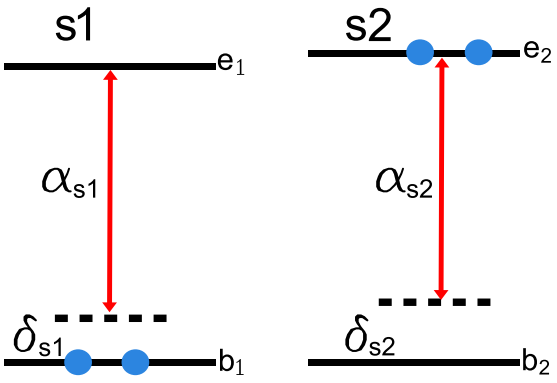

FIG. 2. (Color online) An equivalent representation of a mixture of two three-level sub-system driven by coherent off-resonant fields.

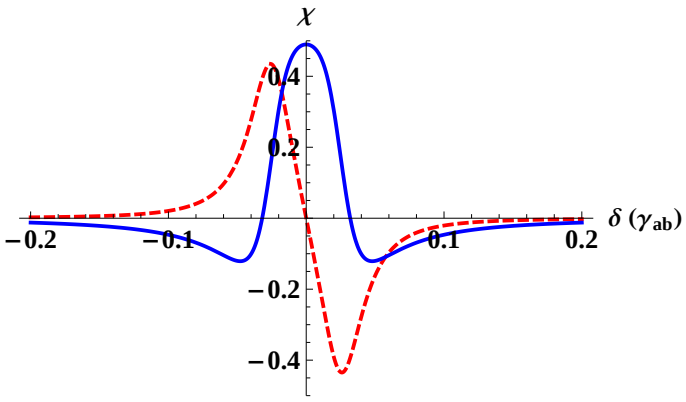

FIG. 3. (Color online) Combined real (solid) and imaginary (dashed) parts of the susceptibility from two three-level systems. Where the x-axis is normalized to $\gamma_{ab}^{s1} = \gamma_{ab}^{s2} = \gamma_{ab}$, and the y-axis is normalized to $\eta = 3N_{s1}\lambda_{pr}^3\gamma_r^{s1}/(8\pi^2) = 3N_{s2}\lambda_{pr}^3\gamma_r^{s2}/(8\pi^2) = 1$. With $\Omega_{s1} = \Omega_{s2} = 2\gamma_{ab}$, $\Delta_{s1} = \Delta_{s2} = 20\gamma_{ab}$, and $\gamma_{cb}^{s1} = \gamma_{cb}^{s2} = 0.016\gamma_{ab}$. Resonances have equal strength and width, but are shifted by FWHM. Obtained maximum at zero absorption is 0.5η.

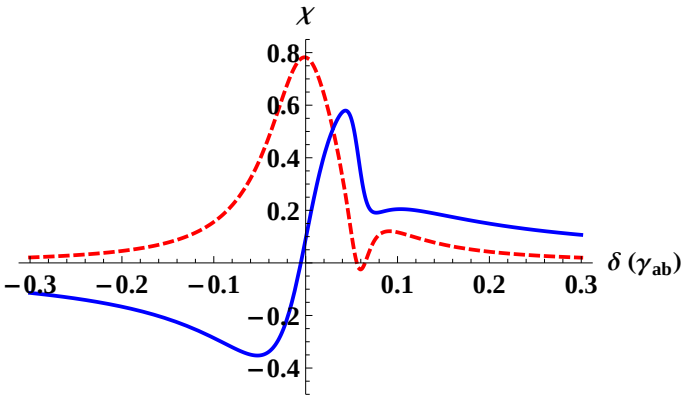

FIG. 4. (Color online) Combined real (solid) and imaginary (dashed) parts of the susceptibility from two three-level systems. With $\Omega_{s1} = 4\gamma_{ab}$ and $\Omega_{s2} = 1.5\gamma_{ab}$, and all other parameters the same as in Fig. 3. Amplification is weaker and narrower than absorption. The relative shift is adjusted to get zero absorption and no gain. Obtained maximum at zero absorption is 0.33η.

nearby gain. Furthermore, at the point of no absorption the maximum(minimum) of the real part of the complex susceptibility associated with the absorption resonance adds up with the maximum(minimum) of the real part of the complex susceptibility associated with the gain resonance. We demonstrate this (see Fig. 3) by calculating the atomic response from a mixture of two three-level sub-systems for the case of $\xi_{si} \approx \gamma_{cb}^{si}/\gamma_{ab}^{si}$. Numerical values of the parameters used are listed in the caption to the figure. Although this arrangement provides a high value of refractive index with no absorption, the disadvantage of such an approach can easily be seen in Fig. 3. Namely, non-compensated gain is present in close proximity to the point of enhanced refractive index. In order to avoid undesirable gain in the system, we suggest an alternative to the previously outlined approach. This is done at the expense of a reduced enhancement of the refractive index. In this approach a narrower amplification resonance is superimposed on top of a broader absorption resonance as previously suggested in [9]. The amplification resonance is positioned at the maximum of the real part of the complex susceptibility associated with the absorption resonance. The magnitude of the amplification resonance is chosen to compensate present absorption in the narrow region without providing gain. For such an arrangement, the amplification resonance provides no contribution to the refractive index enhancement at the point of no absorption. In order to demonstrate the approach, we present the atomic response of the mixture of two three-level systems in Fig. 4 for the parameters listed in the caption of the figure.

As an alternative for the species providing absorption, instead of a three level system we can just use a two level atom that provides absorption. This is natural since we want the absorption feature to be both stronger and wider than the gain feature, and the effective two level transition will always be weaker and narrower than the original two level transition. The only reason this is not always ideal is it eliminates one of knobs we can turn to match the transitions. Since the same probe field will address both the two level absorption transition and the far-detuned lambda scheme, it is necessary for the one photon detuning of our gain system to match the difference in transition frequencies between the two transitions i.e., $\Delta_{s2} \approx \omega_{ab}^{s2} - \omega_{ab}^{s1}$. Since we also need the one photon detuning to be much larger than the linewidth of the optical transition then for this implementation to be appropriate we need $\omega_{ab}^{s2} - \omega_{ab}^{s1} \gg \gamma_{ab}^{si}$, but since the control field Rabi frequency is proportional to Δ_{s2}, we need the difference to be small enough such that a reasonably sized control field Rabi frequency can still satisfy $\xi_{s2} > \gamma_{cb}^{s2}/\gamma_{ab}^{s2}$. If either of these conditions are not satisfied we are forced to use a far-detuned lambda scheme

for both absorption and gain.

So far, we have been discussing the atomic response of a pair of three-level systems. This discussion has demonstrated that refractive index with no absorption is possible to obtain in the presented system. The maximum value for the refractive index is limited by the value for the original two-level system with the main difference that it is achieved with no absorption or amplification in the vicinity of maximum value. Therefore in order to get the maximum value of refractive index enhancement the original two level resonant susceptibility has to be maximized by choosing an appropriately high concentration.

IV. EFFECT OF INHOMOGENEOUS BROADENING

It has been shown in the previous section that the two photon resonant feature is equivalent to an effective two level atomic system with the amplitude, frequency, and width of the transition controlled by the driving field. Furthermore, the electromagnetic response of this effective system can be as strong as the resonant response of an actual two level system. This was shown under the assumption of homogeneous broadening, when values of the Rabi frequency $\Omega_{\rm si}$ and one-photon detuning $\Delta_{\rm si}$ are well defined by intensity and frequency of the driving field. In the case of inhomogeneous broadening of the probed transition the frequency of the driving field defines only the mean value of one-photon detuning Δ_0 while variance is defined by inhomogeneous broadening. Namely, $\Delta_{\rm si} = \Delta_0 + \Delta_{\rm inh}$ with $\left\langle \Delta_{\rm inh}^2 \right\rangle - \left\langle \Delta_{\rm inh} \right\rangle^2 = \left(\gamma_{\rm ab}^{\rm inh}\right)^2$, where $\langle \cdots \rangle$ is averaging over the inhomogeneous profile. For the case of inhomogeneous broadening of the c↔b transition we only have a mean value of the two photon detuning δ_0 while the varying detuning is given by $\delta_{\rm si} = \delta_0 + \delta_{\rm inh}$ with $\left\langle \delta_{\rm inh}^2 \right\rangle - \left\langle \delta_{\rm inh} \right\rangle^2 = \left(\gamma_{\rm cb}^{\rm inh}\right)^2$. Therefore, the two-photon transition probability, frequency, and width are also not well defined.

The inhomogeneous profile in both solids and gases will be Gaussian, but in order to easily deal with the analytic expressions we will approximate with a Lorentzian profile. If we start with the coherence given by Eq. (2) or Eq. (3), we can integrate over the Lorentzian distributions of the one and two photon detunings:

$$\sigma_{\rm ab}^{\rm inh} = \int_{-\infty}^{\infty} d\Delta_{\rm inh} \frac{\gamma_{\rm ab}^{\rm inh}/\pi}{\Delta_{\rm inh}^2 + (\gamma_{\rm ab}^{\rm inh})^2} \int_{-\infty}^{\infty} d\delta_{\rm inh} \frac{\gamma_{\rm cb}^{\rm inh}/\pi}{\delta_{\rm inh}^2 + (\gamma_{\rm cb}^{\rm inh})^2} \sigma_{\rm ab}(\Delta_{\rm inh}, \delta_{\rm inh}). \tag{7}$$

In the limit examined in Section III, $\Delta_0 > \gamma_{\rm ab}^{\rm si}, \gamma_{\rm cb}^{\rm si}, \Omega_{\rm si}$, these integrals can be solved analytically which shows that the inhomogeneous profile can be taken into consideration in all the equations we derive simply by replacing the homogeneous linewidths with the total linewidth that includes the inhomogeneous broadening i.e., $\gamma_{\rm ab}^{\rm si} \to \gamma_{\rm ab}^{\rm si} + \gamma_{\rm ab}^{\rm inh} + \gamma_{\rm cb}^{\rm inh}$ and $\gamma_{\rm cb}^{\rm si} \to \gamma_{\rm cb}^{\rm si} + \gamma_{\rm cb}^{\rm inh}$. Except for system 2 where we also have to replace $\gamma_{\rm ac}^{\rm si} \to \gamma_{\rm ac}^{\rm si} - \gamma_{\rm ab}^{\rm inh}$. It is important to keep in mind this is not a change in the linewidth of the a↔c transition, just in how the decoherence of this transition comes into Eq. (3), and Eq. (6) is still valid when $\Delta_{\rm s2} \gg \gamma_{\rm ab}^{\rm inh}$.

Therefore, one can see that the quantity that matters is the total linewidth. Thus all previous discussion could be repeated here with $\gamma_{\rm ab}^{\rm si}$ being replaced by the total linewidth. This makes our final statement to sound as follows: electromagnetic response of an effective two-level system, which is fully controlled by a weak off-resonance driving field, is as strong as the resonant electromagnetic response of the actual two-level a↔b transition without a driving field being present regardless of the broadening if the total linewidth is considered. Broadening of the transition beyond the natural linewidth weakens the response of the system as well as the intensity required to reach the maximal obtainable value.

V. COLLISIONAL BROADENING

Electromagnetic response of an atomic system can be increased by improving the $\gamma_{\rm r}^{\rm si}/\gamma_{\rm ab}^{\rm si}$ ratio and by having more atoms per cubic wavelength $\mathrm{N}_{\rm si}\lambda_{\rm pr}^3$. The second approach seems to be the easiest but it leads to a decrease of the aforementioned ratio due to atomic interaction in dense media. The large estimates for achievable refractive indexes in previous works were due to not considering the effect of increasing density on the ratio $\gamma_{\rm r}^{\rm si}/\gamma_{\rm ab}^{\rm si}$. According to Lewis [10], the collisional contribution to the linewidth is proportional to concentration N, namely the HWHM is:

$$\Gamma_{\rm coll}^{\rm si} \simeq f_{\rm si} c r_e \lambda_{\rm pr} \mathrm{N} \sqrt{g_g^{\rm si}/g_e^{\rm si}}, \tag{8}$$

with $g_g^{\rm si}$ and $g_e^{\rm si}$ to be the degeneracies of the ground and excited states, respectively; r_e is the classical radius of the electron, and $f_{\rm si}$ is the oscillator strength of the transition. This broadening comes from resonant dipole-dipole interaction between induced optical dipoles. Where in Eq. (8) we have used the total population $\mathrm{N} = \mathrm{N}_{\rm s1} + \mathrm{N}_{\rm s2}$, since atoms with similar transition frequencies will be identical with regard to collisions. For example take the ^{85}Rb D1 line, Eq. (8) gives $\Gamma_{\rm coll} = .365{\cdot}10^{-13}\mathrm{MHz{\cdot}cm^3{\cdot}N}$ and [11] measured the self broadening as $\Gamma_{\rm coll} = .375(\pm.12){\cdot}10^{-13}\mathrm{MHz{\cdot}cm^3{\cdot}N}$ therefore this equation gives an accurate estimate.

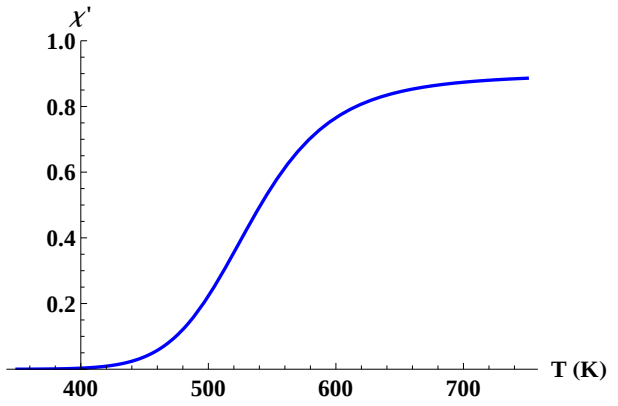

FIG. 5. (Color online) The peak real part of the susceptibility for the D2 line of ^{85}Rb as a function of absolute temperature.

For a Doppler broadened gas, the inhomogeneous broadening additively contributes to the total broadening in our scheme. The Doppler contribution is given by the half width at half maximum (HWHM) of the Maxwell distribution for each species:

$$\Gamma_{\rm D}^{\rm si} = \sqrt{\frac{2kT\ln 2}{m_{\rm si}c^2}}\omega_0. \tag{9}$$

Where k is Boltzmann's constant, T is the absolute temperature of the gas, $m_{\rm si}$ is the mass of the atomic species, c is the speed of light, and $\omega_0 = 2\pi c/\lambda_{\rm pr}$ is the transition frequency.

In a hot gas cell the density will be determined by the temperature of the cell, so both the inhomogeneous Doppler broadening and the homogeneous collisional broadening are dependent on the density. Since we want a large resonant refractive index we are interested in high gas densities. The susceptibility of the effective two level system will always be constrained by the original two level susceptibility of the a↔b transition given by:

$$\chi_{\rm max}^{\rm si} = \frac{3}{8\pi^2}\frac{{\rm N_{si}}\lambda_{\rm pr}^3\gamma_{\rm r}^{\rm si}}{.5\gamma_{\rm r}^{\rm si}+\Gamma_{\rm D}^{\rm si}+\Gamma_{\rm coll}^{\rm si}}. \tag{10}$$

Since the linewidth and total atomic response grow linearly with concentration, eventually with increased density the susceptibility will saturate. This happens when $\Gamma_{\rm coll}^{\rm si} \gg .5\gamma_{\rm r}^{\rm si} + \Gamma_{\rm D}^{\rm si}$.

Consider the D2 line of ^{85}Rb with $\Gamma_{\rm coll} = .515{\cdot}10^{-13}{\rm cm^3MHz{\cdot}N}$, when the collisional broadening becomes much larger than the other broadening terms the two level susceptibility saturates at 750K or a density of N$\approx 6{\cdot}10^{17}{\rm cm^{-3}}$ (see Fig. 5). This leads to a maximum real part of the resonant susceptibility of .885 or a refractive index of 1.37. Therefore there is no way to enhance the refractive index of Rubidium past $\Delta n = .4$.

VI. RATIO OF HYPERFINE COHERENCE TO OPTICAL COHERENCE

The main limitation for our effective 2 level transition to have as high a susceptibility as the original transition, is the need for a strong control field Rabi frequency $|\Omega_{\rm si}|^2 > \Delta_{\rm si}^2\gamma_{\rm cb}^{\rm si}/\gamma_{\rm ab}^{\rm si}$. Therefore to minimize the needed intensity, we need as small hyperfine decoherence as possible $\gamma_{\rm cb}^{\rm si} \ll \gamma_{\rm ab}^{\rm si}$.

At low densities the main contribution to the hyperfine decoherence will be due to time of flight in our control beams, since as atoms leave the interaction region the coherence decreases. This decoherence rate can be described as [12]:

$$\Gamma_{\rm TF}^{\rm si} = \frac{\sqrt{2}\ln 2}{2\pi d}\sqrt{\frac{2kT}{m}}, \tag{11}$$

where d is the 1/e diameter of the beam. While at high densities the main contribution to the hyperfine broadening is the decay of hyperfine population due to spin-exchange collisions between two atoms. This self broadening, like collisional broadening is linearly proportional to the density. For example for ^{85}Rb we can estimate the decoherence as $\Gamma_{\rm SB}^{\rm si} = 2\pi{\cdot}2.83{\cdot}10^{-16}{\rm N{\cdot}MHz}$ [13].

The time of flight decoherence can be decreased by including a neutral buffer gas. Then as our atomic species is leaving the beam area it will repeatedly collide with the buffer gas atoms leading to a longer path length in the beam. With the background gas, the time of flight decoherence rate given by Eq. (11) will be replaced by [10]:

$$\Gamma_{\rm TF}^{\rm si} = \left(\frac{4.81}{d}\right)^2 {\rm D_0}\frac{\rm P_0}{\rm P_{BG}}, \tag{12}$$

where D_0 is the diffusion coefficient measured at a reference pressure of $\rm P_0$, and $\rm P_{BG}$ is the buffer gas pressure. The buffer gas will also broaden both transitions due to collisional broadening, while even at high buffer pressures, collisions with the buffer gas will only have a negligible effect on $\gamma_{\rm cb}^{\rm si}$, but will have a noticeable addition to the optical coherence that scales linearly with buffer gas pressure. At high buffer gas pressures this effect can significantly reduce the maximum susceptibility possible.

For example consider the Rb D2 line, we can express the buffer gas contribution to the collisional decoherence is $\Gamma_{\rm BG}^{\rm si} = 2\pi{\cdot}(4.735{\rm MHz/Torr}){\cdot}{\rm P_{BG}}$ with $\rm P_{BG}$ measured in Torr [14]. $D_0 = .21{\rm cm^2/s}$ for $\rm P_0 = 760$ Torr [15], giving for $d = 1$mm, $\Gamma_{\rm TF}^{\rm si} = 2\pi{\cdot}.369{\rm MHz{\cdot}Torr/P_{BG}}$. For this case the effect of the buffer gas on the decoherence ratio is shown in Fig. 6. Since the collisional broadening increases as the buffer gas pressure is increased, for maximum refractive index enhancement it is better to use a low pressure buffer gas as can be seen in Fig. 7.

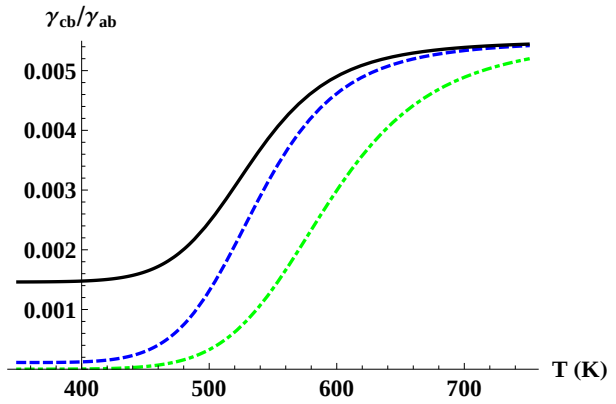

FIG. 6. (Color online) The ratio of hyperfine to optical decoherence rates for the D2 line of ^{85}Rb plotted as a function of temperature for the case where there is no buffer gas (solid), with a Neon buffer gas at 10Torr (dashed), and at 300Torr (dot-dashed).

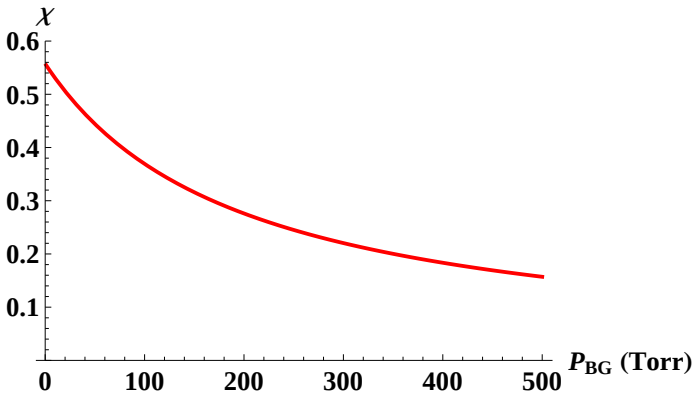

FIG. 7. (Color online) The maximum real part of the 2 level susceptibility for Rb D2 line plotted as a function of the buffer gas pressure. Plotted at a temperature of 550K.

VII. IMPLEMENTATION IN GAS

Alkali metals such as Lithium, Rubidium, Potassium have been good test systems for demonstrating many coherent effects. To demonstrate refractive index enhancement, one needs to find two transitions with frequency differences in the MHz to GHz range. This is possible if a mixture of isotopes is considered. Of the three alkali atoms with stable isotopes however, only Rubidium has a comparable ratio of naturally occurring isotopes (28% of ^{87}Rb and 72% of ^{85}Rb) and large enough hyperfine splitting. Thus as a physical example, let us consider a mixture of Rb vapors at natural abundance.

Rb atoms have two suitable transitions called D1 at 794.8 nm and D2 at 780.2 nm. The D1 and D2 transitions have common ground levels and differ in the excited state. The excited level structure for the D1 and D2 transitions has a separation less than 0.8GHz thus for one-photon detunings much larger than this separation the value of effective far-detuned dipole moment can be used. Numerical values for π-polarized light are $1.727ea_0$ and $2.44ea_0$ for D1 and D2 transitions correspondingly. A stronger dipole moment guaranties a stronger atomic response and therefore lager susceptibilities. A stronger dipole moment also implies a lower intensity requirement for the control fields to reach the needed Rabi frequency. Therefore the D2 transition seems to be the optimal choice from all the accounts. The natural linewidth of the Rb D2 absorption line is $2\pi\cdot6.067$ MHz, although the radiative decay rate is $2\pi\cdot5.12$MHz.

In order to implement refractive index enhancement with vanishing absorption while maintaining no nearby regions of gain in a Rb gas it is necessary to implement system 1 in ^{85}Rb and system 2 in ^{87}Rb. Since at natural abundance the density of ^{85}Rb is nearly three times larger than ^{87}Rb and we need the effective absorption transition to be stronger than the effective gain transition. This choice also determines the one photon detuning for system 1 since if we want the same probe field to address both transitions then the difference in transition frequencies $\omega_{ab}(^{87}\text{Rb}) - \omega_{ab}(^{85}\text{Rb}) = 2\pi\cdot.39$GHz determines the difference in one photon detunings. The ground state of Rb has 2 hyperfine levels separated by $2\pi\cdot3.036$GHz and $2\pi\cdot6.835$GHz for ^{85}Rb and ^{87}Rb isotope respectively. We assume that the probe field is applied to the lower of the two hyperfine levels and the control fields to the upper. First it tells us that we need $\Delta_{si} > 2\pi\cdot8$GHz in order to avoid one photon resonance, therefore we will take $\Delta_{s2} = 2\pi\cdot10$GHz, implying $\Delta_{s1} = 2\pi\cdot10.385$GHz.

Except for the transition frequencies all other properties of interest for ^{85}Rb and ^{87}Rb including the dipole moments and the decoherence rates are essentially the same, when the slight mass difference is neglected. The decoherence rate of the optical transition has four contributions: $\gamma_{ab}^{si} = .5\gamma_r^{si} + \Gamma_{coll}^{si} + \Gamma_D^{si} + \Gamma_{BG}^{si}$. The radiative decay rate is fairly unaffected by density and is given by $\gamma_r^{si} = 2\pi\cdot5.12$MHz. As discussed in Section V, we can take the ideal density for Rb to be $N = 6\cdot10^{17}\text{cm}^{-3}$; unfortunately, at this density $\gamma_{ab}^{si} = \gamma_{ac}^{si} = 197$GHz which violates our condition to avoid the one photon absorption of $\Delta_{si} \gg \gamma_{ab}^{si}, \gamma_{ac}^{si}$, since having such a large one photon detuning would lead to a unachievable Rabi frequency.

Say for our control fields we are limited to focusing a 100mW beam into a diameter of 1mm, this would give us Rabi frequencies of $2\pi\cdot150$MHz, so we take $\Omega_1 = 2\pi\cdot150$MHz and $\Omega_2 = 2\pi\cdot135$MHz. Implying the max control field ratio ξ_{si} we can achieve is $2.25\cdot10^{-4}$. The decoherence ratio is equal to this ξ_{si} at a temperature of T = 450K, or a density of $N = 3.3\cdot10^{14}\text{cm}^{-3}$ so at natural abundance we have $N_{s1} = .72N$ and $N_{s2} = .28N$. For a fixed ξ_{si}, a smaller buffer gas pressure is better so we will take $P_{BG} = 10$Torr. This buffer gas will add $2\pi\cdot47$MHz to the optical decoherence, and the time of flight broadening is $\Gamma_{hyp,BG}^{si} = 2\pi\cdot37$kHz. At this density the hyperfine self broadening is $\Gamma_{hf,self}^{si} = 2\pi\cdot93$kHz, so $\gamma_{cb}^{si} = 2\pi\cdot130$kHz. The collisional broaden-

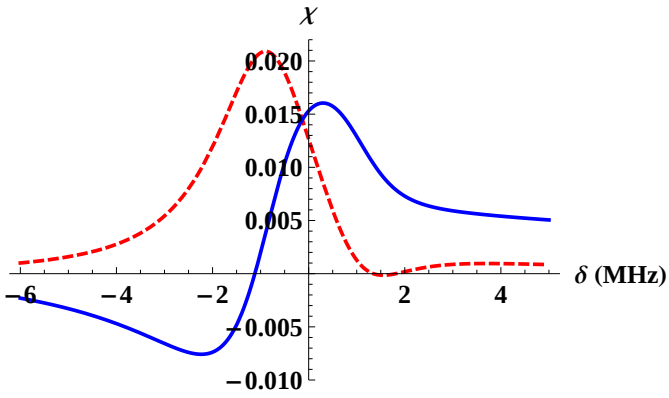

FIG. 8. (Color online) The real (solid) and imaginary (dashed) part of the susceptibility as a function of the detuning, plotted for the scheme without gain and with $\Omega_1 = 2\pi{\cdot}150$MHz described in the text.

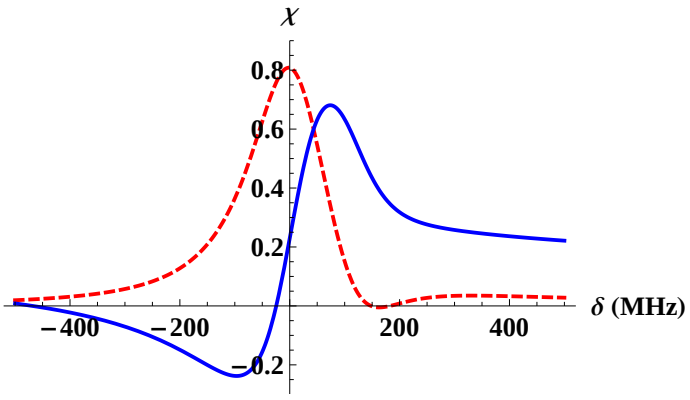

FIG. 9. (Color online) The real (solid) and imaginary (dashed) part of the susceptibility as a function of the detuning, plotted for the scheme with $\Omega_1 = \Omega_2 = 2\pi{\cdot}1.5$GHz as described in the text.

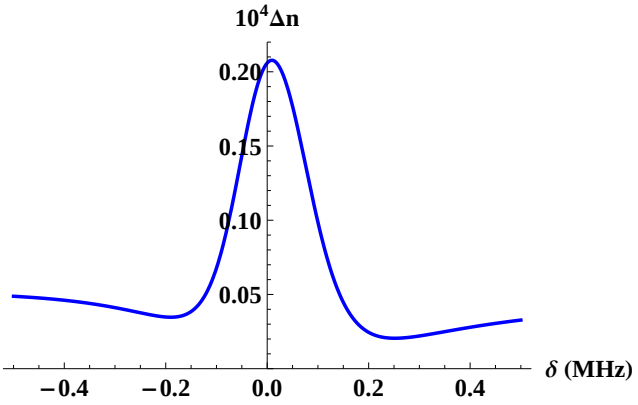

FIG. 10. (Color online) The expected refractive index enhancement for experiment described in [7] plotted as a function of two photon detuning, using the experimental numbers as reported in the text.

ing is $\Gamma^{si}_{coll} = 2\pi{\cdot}17$MHz, and the Doppler broadening is $\Gamma^{si}_{D} = 2\pi{\cdot}315$MHz, therefore $\gamma^{si}_{ab} = 2\pi{\cdot}382$MHz. Giving a ratio of $\gamma^{si}_{cb}/\gamma^{si}_{ab} = 3.4{\cdot}10^{-4}$.

With these values for the detunings and Rabi frequencies the susceptibility is plotted in Fig. 8. There is a particular frequency where we have vanishing absorption and a significant resonant susceptibility $\mathrm{Re}\chi = .0095$. At the same time we have no regions of nearby gain so that the probe field will remain stable at that frequency. Therefore in a hot Rb gas we have a maximum refractive index enhancement on the order of $\Delta n \approx 4.7{\cdot}10^{-3}$.

When it is possible to use strong control fields we can achieve the maximum possible susceptibility increase. Consider focusing a 10W beam into a 1mm diameter, then the Rabi frequencies are $\Omega_{s1} = \Omega_{s2} = 2\pi{\cdot}1.5$GHz. This allows us to use larger one photon detunings to $\Delta_{s1} = 2\pi{\cdot}20$GHz, $\Delta_{s2} = 2\pi{\cdot}20.27$GHz. Which gives $\xi_{s1} = .0055$, allowing for a higher ratio of $\gamma^{si}_{ab}/\gamma^{si}_{cb}$. Taking T = 600K, gives us $N = 4{\cdot}10^{16}\mathrm{cm}^{-3}$ which leads to $\gamma^{si}_{ab} = 2\pi{\cdot}4.667$GHz and $\gamma^{si}_{cb} = 2\pi{\cdot}71$MHz. Then the change in index is $\Delta n = .18$.

Since the isotope shift for Rubidium $\omega_{ab}(^{87}\mathrm{Rb}) - \omega_{ab}(^{85}\mathrm{Rb}) = 2\pi{\cdot}.39$GHz is smaller than our linewidths it would not be possible to use the two level transition rather than the effective two level transition for absorption as discussed at the end of Section III. To implement such a system, we would need isotope shifts on the order of 10GHz, which are only possible when the masses of the two isotopes are significantly different compared to the isotope mass. For example such a system could be implemented in Lithium.

VIII. EXPERIMENTAL REALIZATION

A proof of principal experiment to demonstrate resonant enhancement of refractive index in hot Rb gas was performed by Yavuz [7]. The same two lambda systems in ^{85}Rb and ^{87}Rb at natural abundance using the D2 line and a 10Torr Neon buffer gas was implemented. The Rb cell was kept at 363K so the Rb density is $N = 2.4{\cdot}10^{12}\mathrm{cm}^{-3}$, the optical broadening should be $\gamma^{si}_{ab} = 2\pi{\cdot}$ 334MHz, and the hyperfine broadening should be $\gamma^{si}_{cb} = 2\pi{\cdot}16$kHz. The laser power is given as a 100mW focused in a 2.4mm diameter spot size, implying that $\Omega_{si} < 2\pi{\cdot}63$MHz. Based on the experimental results of both the gain and absorption being roughly equal in height and width with a two photon HWHM of 125kHz we can assume that $\Omega_{s1} = 2\pi{\cdot}34$MHz and $\Omega_{s2} = 2\pi{\cdot}58$MHz. The control fields in the experiment are taken such that $\Delta_{s1} = 2\pi{\cdot}15.6$GHz and $\Delta_{s2} = 2\pi{\cdot}16$GHz. With an applied pump field intensity of $1.77\mathrm{W/cm}^2$, numerical simulations show that for a pump field with a bandwidth of about 500MHz the population difference for both systems would be close to 1 as needed, with the largest deviation in system 2 with $\sigma^{s2}_{cc} - \sigma^{s2}_{aa} = .93$.

The theoretical curve for the resonant refractive index

with these parameters is plotted in Fig. 9. The theory predicts a change in index of $\Delta n = 1.7{\cdot}10^{-5}$. The theory also shows that there is a background index due to the one photon feature of $\mathrm{n} = 1 + 4{\cdot}10^{-6}$, since $\xi_{\mathrm{s1}} < \gamma_{\mathrm{cb}}^{\mathrm{s1}}/\gamma_{\mathrm{ab}}^{\mathrm{s1}}$.

The reported refractive index change of $\Delta \mathrm{n} = 2{\cdot}10^{-7}$ is two orders of magnitude less than what was possible in the experiment [7] due to issues with the cross pumping of the ^{87}Rb and ^{85}Rb populations. As explained in [7], this could be due to the frequency width of the pump fields being larger than the separation between the hyperfine level of ^{87}Rb and ^{85}Rb which would reduce the population difference between levels $|c_{\mathrm{i}}\rangle$ and $|b_{\mathrm{i}}\rangle$ and thus significantly reduce the RI enhancement.

IX. CONCLUSION

We have given a simple model for how to implement refractive index enhancement without absorption in Rb gas while avoiding any nearby regions of gain. This is done by implementing a far detuned Λ system in two different atomic species evenly mixed in a hot gas. In the decaying dressed states basis the whole system can be presented as a superposition of two effective two-level schemes with positions, widths, and amplitudes of the resonances determined by the driving fields. It allows for a simple, analytic, and intuitive understanding of the susceptibility for the total system. Thus a variety of absorption, amplification, and dispersion profiles may easily be engineered. In particular, maximum refractive index with vanishing absorption results from the simple summation of susceptibilities of the effective absorbing and amplifying two-level schemes whose resonances are positively and negatively tuned with respect to the probe field frequency. Engineering a larger width for the absorptive resonance allows one to eliminate any amplification region in the vicinity of the enhanced index and vanishing absorption. Proper tuning also allows for the strong increase or decrease of refractive index under vanishing absorption.

We have shown that with reasonable beam intensities this scheme can be implemented in ^{85}Rb and ^{87}Rb at natural abundance for refractive index enhancement on the order of $\Delta n \simeq 5{\cdot}10^{-3}$. This can be done while maintaining vanishing absorption and with no nearby regions of gain. Potentially higher resonant refractive indexes with vanishing absorption could be obtained with much stronger beam intensities or in solids (in particular, in rare-earth and/or transition metal ions doped dielectric crystals or stoichiometric crystals including such ions) [9, 16]. The further analysis of the limitations of resonant index enhancement requires careful studies of optical line self-broadening with an increase of the density, and the inclusion of local field effects [17]. Along with the achievement of high refractive index, it would also be very beneficial for a number of applications to provide for its temporal or spatial modulation. For example, it would allow for the production of controllable photonic structures in a homogeneous media simply by applying optical fields [18].

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