

Pumped quantum systems: Immersion fluids of the future?

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Quantum optical techniques may yield immersion fluids with high indices of refraction without absorption. We describe one such technique in which a probe field experiences a large index of refraction with amplification rather than absorption, and examine its practicality for an immersion lithography application. Enhanced index can be observed in a three-level system with a tunable near-resonant coherent probe and incoherent pump field that inverts the population of the probe transition. This observation contradicts the common belief that large indices of refraction are impossible without absorption; however, it is well in accord with existing electromagnetic theory and practice. Calculations show that a refractive index $\gg 2$ is possible with practical experimental parameters. A scheme with an incoherent mixture of pumped and unpumped atoms is also examined, and is seen to have a lower refractive index (~ 2) accompanied neither by gain nor loss. © 2005 American Vacuum Society. [DOI: 10.1116/1.2121732]

I. INTRODUCTION

Immersion lithography is currently the technology of choice for integrated circuit fabrication at the 65-nm-gate-length node.^{1,2} Further scaling of this technology will require either a reduced source wavelength or a higher-refractive-index immersion fluid. But efforts to make economical sources and optics at wavelengths shorter than 193 nm have so far been unsuccessful. Additionally, refractive-index scaling is often denigrated based on classical arguments that index enhancement near an atomic resonance is always accompanied by absorption. Thus, it may appear that device scaling is nearing its end. However, when atomic resonances are treated quantum mechanically, the connection between absorption and index enhancement can be circumvented. In this article, we apply quantum-optical techniques to the problem of index enhancement for continued scaling of optical lithography.

We present a scheme that can yield an enhanced index of refraction ($n \gg 1$) with gain instead of absorption. In this scheme, a near-resonant probe laser experiences a high-refractive index in a pumped quantum system. The refractive index is a function of both the intrinsic properties of the quantum system, such as spontaneous decay rates, and the controllable properties, such as atomic density.³ A few possible applications of this scheme are immersion lithography,^{1,2} microscopy,⁴ all-optical switching, and optical buffering.⁵ While previous proposals for phase-coherent quantum-optics-based index-enhancement techniques promise an appreciable refractive index with no absorption,^{6–11}

only enhancements of $\Delta n \approx 10^{-4}$ have in fact been demonstrated due to practical experimental obstacles.¹² On the other hand, our calculations show that a maximum refractive index of ~ 6 is possible with our scheme for reasonable experimental parameters. In principle, the refractive index can be enhanced beyond 6 with a higher atomic density. In addition, our scheme should prove easier to implement than existing schemes based on quantum interference because the pump and probe lasers do not need to be collinear or phase locked. Thus, our proposed scheme is likely to be more practical and more effective than existing quantum-optical schemes at achieving enhanced index without absorption.

To apply the proposed scheme to immersion lithography, several issues would have to be resolved. An immersion medium consisting of a pumped quantum system would generate poor image contrast due to amplified spontaneous emission. Additionally, due to power broadening, the refractive index rapidly decays from its enhanced value to unity in a short propagation distance. These issues are discussed in Sec. III. A solution to some of the problems is suggested in Sec. IV where we describe a system composed of a mixture of quantum gases described by *absorptive* two-level quantum systems and *amplifying* (pumped) three-level quantum systems probed by a single laser.⁸ The mixture exhibits enhanced index of refraction without loss or gain.

II. PUMPED THREE-LEVEL SYSTEM: HIGH- n WITH GAIN

A simple pumped atomic resonance illustrates the concept of refractive-index enhancement with gain. In an unpumped system, a probe beam tuned near resonance experiences a high refractive index, but is absorbed in a fraction of a wave-

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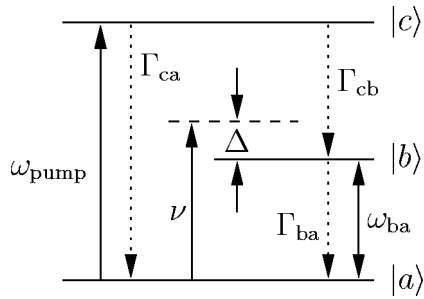


FIG. 1. Energy-level diagram for a pumped three-level system showing driving fields (probe ν and pump ω_{pump}) and incoherent decay rates Γ_{ij} corresponding to decay from level $|i\rangle$ to $|j\rangle$. The probe field is detuned by Δ from the $|b\rangle-|a\rangle$ resonance.

length. In our scheme, we use a pump laser to invert the population on the probe transition. A probe beam interacting with a population-inverted transition also experiences a high-refractive index but is amplified instead of absorbed. In the following section, we specify the parameters of our system.

A. Energy-level structure

We chose to illustrate our index-enhancement scheme in a three-level system as shown in Fig. 1, although other three-level or four-level systems will also suffice. The three-level system shown in Fig. 1 has a ground state $|a\rangle$, excited state $|b\rangle$, and upper lying level $|c\rangle$. A pump field ω_{pump} is used to invert population on the $|a\rangle-|b\rangle$ transition via an incoherent de-excitation from $|c\rangle$ to $|b\rangle$. A probe field detuned from the $|b\rangle-|a\rangle$ transition by frequency Δ will see an enhanced index. In the following section, we quantify the amount of refractive index enhancement and show that the probe field will also experience amplification rather than absorption due to population inversion.

B. Index enhancement for unbroadened thin media

To calculate the refractive index as seen by the probe laser, we formulate the density matrix equations of motion¹³ for the system. The population in level $|i\rangle$ is denoted by the on-diagonal density matrix element ρ_{ii} and decays into another level $|j\rangle$ at rate Γ_{ij} . The coherence between levels $|i\rangle$ and $|j\rangle$ is given by ρ_{ij} and decays at rate γ_{ij} . A phenomenological treatment of spontaneous decay rates and coherence decay rates leads to the following equations:

$$\dot{\rho}_{aa} = (\Gamma_{ca} - \Gamma_{ba})\rho_{cc} - (r_{ac} + \Gamma_{ba})\rho_{aa} + \frac{i\Omega_R}{2}(\rho_{ba} - \rho_{ab}) + \Gamma_{ba}, \quad (1a)$$

$$\dot{\rho}_{ab} = -\frac{i\Omega_R}{2}(\rho_{cc} + 2\rho_{aa} - 1) - (\gamma_{ab} + i\Delta)\rho_{ab}, \quad (1b)$$

$$\dot{\rho}_{ca} = -(\gamma_{ca} + i(\omega_{cb} - \Delta))\rho_{ca} - \frac{i\Omega_R}{2}\rho_{cb}, \quad (1c)$$

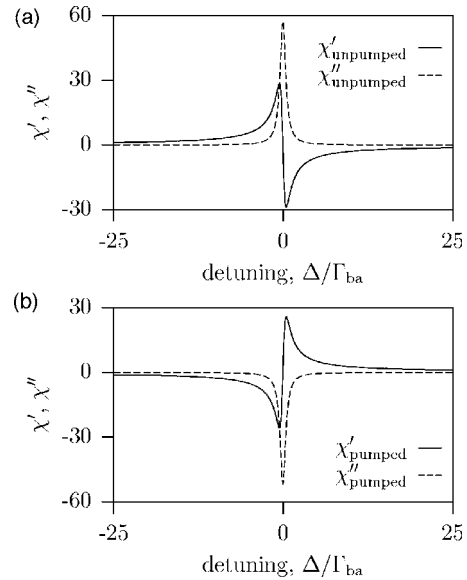


FIG. 2. Plot of the susceptibility $\chi = \chi' + i\chi''$ calculated to first order in Ω_R for (a) an unpumped system, and (b) a pumped system with population inversion. Parameters are $N=10^{16} \text{ cm}^{-3}$, $\gamma_{ab}=0.5\Gamma_{ba}$, $\Gamma_{ca}=0.1\Gamma_{ba}$, $\Gamma_{cb}=10\Gamma_{ba}$, and $r_{ac}=200\Gamma_{ba}$. We see that $\chi' < 0$ for the pumped case, resulting in amplification (rather than absorption) of the probe laser.

$$\dot{\rho}_{cb} = -\frac{i\Omega_R}{2}\rho_{ca} - (\gamma_{cb} + i\omega_{cb})\rho_{cb}, \quad (1d)$$

$$\dot{\rho}_{cc} = -(\Gamma_{ca} + \Gamma_{cb})\rho_{cc} + r_{ac}\rho_{aa}, \quad (1e)$$

where r_{ac} is the pump rate, Ω_R is the Rabi frequency of the $|b\rangle-|a\rangle$ transition, and we have used $\rho_{aa} + \rho_{bb} + \rho_{cc} = 1$.

The susceptibility χ as seen by the probe is related to the coherence between states $|b\rangle$ and $|a\rangle$ given by the off-diagonal density matrix element ρ_{ba} (see Ref. 13 for more details). Equations (1a)–(1e) were solved under the rotating-wave approximation and steady-state conditions to yield the following expression for χ :

$$\chi = N\Gamma_{ba} \frac{3}{8\pi^2} \lambda_{ba}^3 \frac{1}{\Delta + i\gamma_{ab}} (\rho_{bb} - \rho_{aa}), \quad (2)$$

where the population difference between states $|b\rangle$ and $|a\rangle$ is given by

$$\rho_{bb} - \rho_{aa} = \frac{\Gamma_{cb}r_{ac} - \Gamma_{ba}\Gamma_c}{r_{ac}\Gamma_{cb} + \Gamma_{ba}(\Gamma_c + r_{ac}) + \eta(r_{ac} + 2\Gamma_c)}. \quad (3)$$

Here $\eta = \Omega_R^2 \gamma_{ab} / 2(\Delta^2 + \gamma_{ab}^2)$ and $\Gamma_c = \Gamma_{cb} + \Gamma_{ca}$. From Eqs. (2) and (3), we see that χ depends on controllable parameters, namely N , Δ , Ω_R , and r_{ac} , and parameters that are intrinsic to the atomic species, namely decay rates and the probe transition wavelength λ_{ba} . In order to illustrate the magnitude of refractive-index enhancement, we use a three-level system with $\Gamma_{ba} = 2.2 \times 10^8 \text{ s}^{-1}$ and $\lambda_{ba} = 422.8 \text{ nm}$.¹⁴ In our calculations, the $|c\rangle-|a\rangle$ and $|c\rangle-|b\rangle$ transitions were treated phenomenologically with parameters that enabled a large population inversion.

Figure 2 shows the frequency dependence of the susceptibility χ for an unpumped and pumped system with χ taken

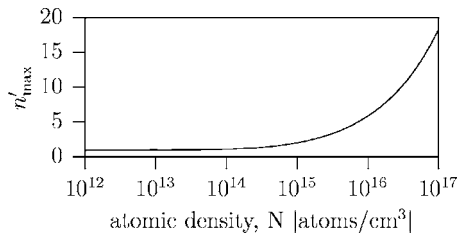


FIG. 3. Plot of maximum value of refractive index n'_{\max} (with respect to detuning) as a function of atomic density N . See Fig. 2 caption for other parameters.

to first order in Ω_R . For these plots, $\chi = \chi' + i\chi''$, where $\chi'' > 0$ implies an absorptive medium, while $\chi'' < 0$ implies an amplifying medium. Propagation effects were not taken into account, so these plots apply only for infinitesimally thin media. Figure 2(a) was obtained by expanding Eq. (3) to first order in Ω_R , setting $r_{ac} = 0$, and substituting the result into Eq. (2). From this plot, it is evident that a large susceptibility is possible, but at the cost of high absorption. Figure 2(b) was obtained via an identical analysis except that a value for r_{ac} was chosen to ensure population inversion between levels $|b\rangle$ and $|a\rangle$. χ'' shown in Fig. 2(b) has the opposite sign of the χ'' curve shown in Fig. 2(a) but χ' is changed only by a reflection in the detuning axis, signifying that a very high susceptibility can be obtained with *gain* rather than *absorption*. This is one of the key points of this article.

The refractive index can be calculated from the susceptibilities via $n = n' + in'' = \sqrt{1 + \chi}$, where n' denotes the refractive index, while n'' is the absorption coefficient of the medium. For the parameters used in Fig. 2(b), at the optimum detuning, the refractive index can be as high as ~ 6 . It is possible to enhance the refractive index still further: n' can be increased by opting for an atomic species with different intrinsic parameters, e.g., a larger dipole matrix element for the $|b\rangle - |a\rangle$ transition. Another way to increase n' is by changing the controllable atomic parameters, e.g., by using a vapor with a high atomic density. The effect of this change is shown in Fig. 3, where we have plotted the maximum achievable refractive index n'_{\max} with respect to detuning as a function of the atomic density. As the plot shows, an increase in the atomic density of one order of magnitude from 10^{16} to 10^{17} atoms/cm³ increases n'_{\max} by a factor of ~ 3 . The importance of the ability to increase the refractive index with a controllable parameter will become evident in the following section, where we will find that the refractive index drops in response to temperature-related broadening effects.

C. Index enhancement for broadened thin media

To evaluate the effect of temperature on the refractive index, we consider the effects of collisional, Doppler, and power broadening. Throughout this subsection, however, we neglect effects of the propagation of the probe beam. Thus, these results apply only for infinitesimally thin media. Propagation effects will be discussed later. We will now elaborate on the various broadening mechanisms:

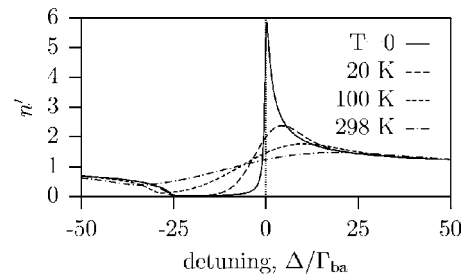


FIG. 4. Plot of refractive-index n' as a function of detuning for a broadened system for various temperatures. The numerical calculation incorporated collisional, power, and Doppler broadening mechanisms for an infinitesimally small propagation distance of the probe beam. Parameters are $d_{crrs} = 3.6$ Å, probe laser intensity $I_0 = 1$ mW/cm², $M = 40$ a.m.u. See Fig. 2 caption for other parameters.

- (1) *Nonradiative collisional broadening* is attributed to the random change in the phase of atomic wave functions that results from elastic collisions. Nonradiative collisional broadening dominates over radiative collisional broadening that occurs due to inelastic collisions.¹⁵ Collisional broadening was taken into account by adding an extra decay term in the equations of motion for the off-diagonal matrix element ρ_{ba} . The decay term is given by $4d_{crrs}^2 N(\pi k_B T/M)^{1/2}$,¹⁵ where d_{crrs} is the average distance between atoms during a collision, T is the temperature, k_B is the Boltzmann constant, and M is the mass of the colliding atoms.
- (2) *Power broadening* occurs for probe intensities so large that the Rabi frequency Ω_R is greater than the natural decay rate for the $|b\rangle$ to $|a\rangle$ transition. Power broadening was taken into account by simply using the full expression in Eq. (3) rather than taking it only to first order in Ω_R .
- (3) *Doppler broadening* occurs when a moving atom interacts with the probe field and sees a frequency shift due to the Doppler effect. Interaction of the probe field with the temperature-dependent velocity distribution of the atomic vapor then leads to broadening of the absorption line shape. Doppler broadening was taken into account by numerically performing a convolution integral between the line shape function for Doppler broadening and the line shape for susceptibility. The line shape function for Doppler broadening, when expressed in terms of Δ , is a normal distribution with variance $(2\pi/\lambda_{ba})^2 k_B T/M$.

These three mechanisms together act to broaden and reduce the peak value of the refractive index. Figure 4 shows a plot of the refractive index as a function of detuning for a pumped system at various temperatures. The maximum value of the refractive index gets reduced from ~ 6 at 0 K to ~ 1.5 at room temperature, while the entire curve broadens as the temperature is increased.¹⁶ While Doppler broadening was the dominant mechanism that lead to a decline in refractive index for the parameters chosen, power broadening contributed more significantly when propagation effects were included. This issue is discussed in the following section along with the problem of amplified spontaneous emission.

III. IMPLEMENTATION HURDLES

The engineering issues that need to be addressed in order for a pumped atomic system to be used as an immersion medium are:

- (1) The medium has to be chemically and physically compatible with conventional immersion-lithography systems and processing techniques;
- (2) The pumping scheme geometry must not inadvertently expose the photoresist. A pump field directed towards the photoresist may be a problem if the pump frequency falls within the exposure spectrum of the photoresist;
- (3) It may not be practical to have an immersion medium with refractive indices higher than ~ 2 due to the current unavailability of high-index photoresists. One solution may be to use thin resists that can be exposed using evanescent fields.¹⁷ This problem will be faced by any index-enhancement scheme used for immersion lithography.

In this article, we will not discuss these issues in detail because we believe that with appropriate chemical and optical engineering, they could in principle be resolved. Instead, we will focus for now on two potential “show stoppers,” both a consequence of high gain in the medium. In the following section, we show that power broadening reduces the initially high refractive index within a very short propagation distance. In Sec. III B, we discuss the effect of amplified spontaneous emission in an immersion lithography application.

A. Power broadening

While power broadening had a negligible effect for an infinitesimally thin medium, it becomes the dominant mechanism that lowers the refractive index when propagation of the probe field in a macroscopic immersion medium is considered. A qualitative understanding comes from the following reasoning: As the intensity of the probe beam increases (i.e., Ω_R increases) due to amplification, the population difference [see Eq. (3)], and thus the refractive index, both decrease. The effect can be calculated for a propagation distance z within a sample by numerically solving the differential equation

$$\frac{dI}{dz} = -\frac{4\pi n''I}{\lambda_{ba}}, \quad (4)$$

where I is the intensity of the probe beam. Note that n' and n'' are both functions of I and therefore z . The result of Runge–Kutta integration of Eq. (4) for various detunings is plotted in Fig. 5. The plot reveals an inherent trade-off between refractive index and propagation distance: If one is willing to settle for a lower refractive index (achieved by increasing the detuning Δ), then the maximum distance in the medium in which that index can be maintained (before n' decays rapidly to 1) is increased. However, for technologically interesting indices of refraction (greater than 2), the maximum thickness is $\sim 4 \mu\text{m}$ for an atomic density of 10^{16} atoms/cm³. A medium only $4 \mu\text{m}$ thick may present

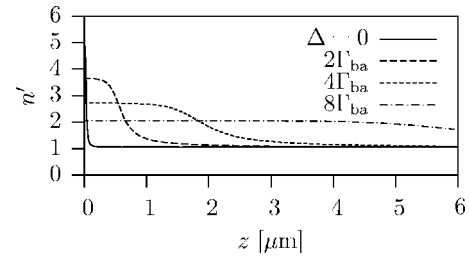


FIG. 5. Refractive index n' as a function of propagation distance z in the atomic medium for various detunings Δ . The index of refraction decreases as a function of z due to an increase in power broadening as the propagating probe beam undergoes amplification. Parameters used are $I[z=0] = 10^3 \text{ W/m}^2$ and $N = 10^{16}$ atoms/cm³. See Fig. 2 caption for other parameters.

arduous practical challenges to implement in an immersion lithography tool. The following section explores another critical challenge to implementation.

B. Amplified spontaneous emission

Amplified spontaneous emission is a problem endemic to many optical systems, including erbium-doped fiber amplifiers¹⁸ and master oscillator power amplifiers.¹⁹ In our system, the gain medium amplifies photons not only in the probe beam, but also photons that originate from spontaneous-emission events. In one simulation, we found that spontaneous emission was amplified by a factor of ~ 100 in a propagation distance of only $1 \mu\text{m}$. This magnitude of amplification is undesirable for two reasons: (1) An atom that has undergone stimulated emission from a spontaneous emission photon is unavailable for emission stimulated by a probe-beam photon, thus the population inversion is reduced, leading to a reduction in the refractive index of the medium; and (2) a large amount of noise from spontaneous emission reduces the image contrast in an immersion lithography application. In the following section, we discuss a scheme that mitigates both this effect and power-broadening issues.

IV. A SCHEME FOR HIGH- n WITH NEITHER GAIN NOR LOSS

One solution to the power broadening and amplified spontaneous emission problems is to find a scheme that eliminates gain altogether, yet still retains enhanced refractive index. Electromagnetically induced transparency would achieve this,⁶ as would a simple mixture of absorptive and amplifying high-index components in the medium.⁸ We examine such a scheme in the following section, using the results of Fig. 2, which shows that absorptive unpumped systems and amplifying pumped systems both exhibit high refractive indices near resonance.

A. Energy level structure

We examine a scheme with a homogeneously distributed, equal mixture of absorptive two-level systems and amplifying three-level systems probed concurrently by a single probe beam. Figure 6 shows the energy level structure of

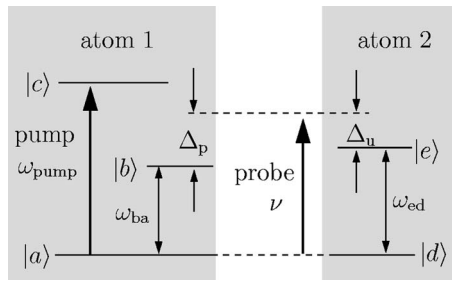


FIG. 6. Energy-level diagram of a pumped three-level (atom 1) and unpumped two-level (atom 2) quantum system. In the intended mixture, both three- and two-level systems are probed concurrently at frequency ν and are assumed to be uncoupled and homogeneously distributed within the probe interaction region.

such a scheme. The mixture consists of uncoupled two- and three-level systems with slightly different resonant frequencies on the probe transition ($\omega_{ba} \neq \omega_{ed}$). The two-level system is detuned from the probe field by Δ_u while the three-level system is detuned by Δ_p . We now describe the calculation of refractive index for this system.

B. Calculation details and result

The calculation for the refractive index for this scheme is a simple extension of the calculation for the pumped system given in Sec. II B. We first found the susceptibilities for both pumped and unpumped systems separately, denoted by χ_p and χ_u respectively. To find χ_p as a function of Δ_p , we substituted parameters specific to the pumped system (e.g., detuning Δ_p , wavelength of probe transition λ_p , atomic density N_p) in Eqs. (2) and (3). The susceptibility for the unpumped system was found by letting $r_{ac}=0$ in Eq. (3) and substituting parameters specific to the unpumped system (Δ_u, λ_u, N_u). We then found the susceptibility of the mixture $\chi_{\text{mix}} = \chi_p + \chi_u$, and expressed χ_{mix} in terms of the average detuning $\Delta = (\Delta_u + \Delta_p)/2$. Finally, we computed the refractive index as seen by the probe field from $n_{\text{mix}} = \sqrt{1 + \chi_{\text{mix}}}$.

Figure 7 shows the dependence of refractive index on detuning for a system with an equal mixture of pumped and unpumped atoms. The plot of n''_{mix} shows that there is a region of absorption where $n''_{\text{mix}} > 0$ and a region of gain where $n''_{\text{mix}} < 0$. Corresponding to the minima and maxima of the n''_{mix} curve are resonant peaks in the plot of n'_{mix} : One corresponding to the pumped species and the other to the unpumped atomic species. The most interesting and important feature of this plot is that there is a point on the n''_{mix} plot where $n''_{\text{mix}} = 0$. For that detuning, a probe field propagating in the medium would neither experience gain nor loss. In effect, the medium becomes *transparent* to the probe field but by a very different mechanism than electromagnetically induced transparency.⁸ Moreover, at the transparency point, the refractive index is greatly enhanced: for this example it is more than 2.

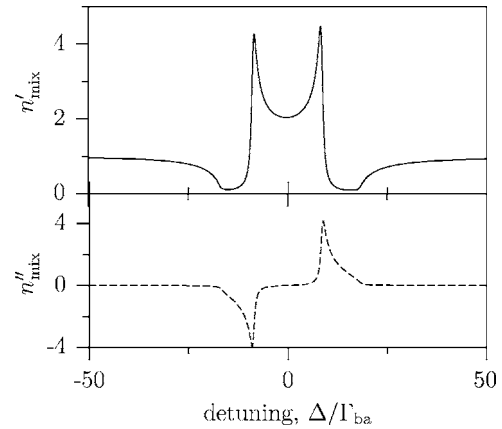


FIG. 7. Plot of refractive index n'_{mix} and absorption coefficient n''_{mix} as a function of detuning for a homogeneous equal mixture of uncoupled two- and three-level systems as pictured in Fig. 6. Note that $\Delta = (\Delta_u + \Delta_p)/2$ and that n'_{mix} and n''_{mix} were calculated to first order in Ω_R . Other parameters are $N_p = N_u = 0.5 \times 10^{16}$ atoms/cm³, $\Gamma_{ba} = \Gamma_{ed}$, and $\omega_{ed} - \omega_{ba} = 52.7$ GHz. See Fig. 2 caption for remaining parameters. $n''_{\text{mix}} = 0$ implies a medium transparent to the probe, which passes through it without gain or loss.

V. CONCLUSION AND FUTURE WORK

The central result of this article is the presentation of a scheme for refractive-index enhancement ($n' \sim 6$ for the example system) accompanied by gain rather than absorption. We showed that one can achieve extremely high refractive indices, but only for a very thin layer of the atomic medium. We identified the main implementation hurdles as power broadening and amplified spontaneous emission, both due to the high gain experienced by the probe beam. Finally, we examined a modified scheme consisting of a mixture of pumped and unpumped systems to tackle these problems and to achieve enhanced refractive index without either gain or loss.

We believe that quantum-optical techniques that take advantage of high susceptibilities near atomic resonances may not only result in a high-index medium for next-generation immersion lithography, but also represent a new direction for refractive-index engineering. Indeed, an all-optically controllable refractive index can be useful for applications in optical communications and microscopy. Future work will concentrate on finding quantum systems with which we can experimentally verify the predictions made in this article.

ACKNOWLEDGMENTS

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