Maximal Refraction and Superluminal Propagation in a Gaseous Nanolayer

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We present an experimental measurement of the refractive index of high density Rb vapor in a gaseous atomic nanolayer. We use heterodyne interferometry to measure the relative phase shift between two copropagating laser beams as a function of the laser detuning and infer a peak index $n = 1.26 \pm 0.02$, close to the theoretical maximum of 1.31. The large index has a concomitant large index gradient creating a region with steep anomalous dispersion where a subnanosecond optical pulse is advanced by >100 ps over a propagation distance of 390 nm, corresponding to a group index $n_g = -(1.0 \pm 0.1) \times 10^5$, the largest negative group index measured to date.

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Controlling the speed of light in a medium is key to applications in quantum and optical communication and computation. For optical pulses, the group velocity v_g determines the speed of the peak, and depends on the variation of the refractive index n with frequency ω. Around a resonance v_g can vary significantly, from slow light ($v_g < c$) [1], to fast light ($v_g > c$) [2], or even backwards light ($v_g < 0$) [3]. Employing techniques such as electromagnetically induced transparency [4,5], where a control laser is used to modify the atomic coherence, light has been slowed experimentally to 17 m/s, corresponding to a group index $n_g = c/v_g > 10^7$ [1]. By tuning the control field adiabatically, it is possible to store light in the form of an atomic excitation and retrieve it at a later time [6], forming a quantum memory [7]. Much effort has gone into other applications of slow light, such as tunable optical delay lines [8] and polarization control [9]. When $n_g < 0$ the peak of the pulse exits the medium earlier than at light speed, leading to the term "superluminal." This surprising phenomenon has led to a wealth of research, including topics such as how fast information can be transmitted in a fast-light medium (signal velocity) [10], and the quantum mechanical implications for noise properties in a slow- or fast-light regime [11,12].

To observe fast light, one must satisfy the inequality $-dn/d\omega < n/\omega$. One way to achieve this is to center the pulse frequency at the center of an absorption line. However, this means that a large gradient $dn/d\omega$ is accompanied by large absorption. Achieving a large group index in a medium without large absorption is a key topic, and one which has attracted considerable attention over the past decades. Most solutions to this problem have utilized atomic coherence and quantum interference in multilevel excitation schemes, proposed by Scully [13] and first experimentally demonstrated by Zibrov *et al.* [14], creating a region with vanishing absorption with nonzero dispersion on resonance. Using a similar concept, the first experiment to measure superluminal propagation used the anomalous dispersion region in between two narrow gain lines [2], where a group index $n_g = -310$ was measured. Similarly, backward propagation of pulses was first reported in a gain medium using an erbium-doped fiber amplifier [3], with a measured index of $n_g = -4 \times 10^3$. All these techniques, however, suffer from the disadvantage that the absolute variation of the refractive index Δn is small and as such are limited to low bandwidth $\Delta\omega$, which can be expressed as

$$\Delta\omega \approx \omega \frac{\Delta n}{n_g}, \tag{1}$$

assuming $|dn/d\omega| \gg n/\omega$. For a large group index with large bandwidth, a large index enhancement is required. In thermal vapors with a double resonance line, large fractional delays of up to 80 with GHz bandwidth have been demonstrated for slow light using off-resonant excitation [8]. In principle, the maximum observable fractional advance is limited to ~2 by a signal-to-noise argument [15], while experimentally fractional advances up to 0.25 have been observed [16], but with severe attenuation (2% transmission).

In this Letter, by using a dense atomic vapor with subwavelength thickness we observe an enormous index enhancement $\Delta n = 0.26 \pm 0.02$. As we are in the cooperative regime [17,18], the dipole-dipole interaction-induced broadening gives rise to a GHz-bandwidth region with very little group velocity dispersion. This allows superluminal propagation of subnanosecond pulses with little distortion, and we observe over 100 ps advance across a distance of 390 nm, corresponding to $n_g = -(1.0 \pm 0.1) \times 10^5$, the largest negative group index measured to date and close to the maximum predicted by a weak probe theoretical model.

Figure 1 shows the experimental setup. We circumvent the problem of high optical depth using a thin layer, much less than the wavelength of excitation light. The atomic

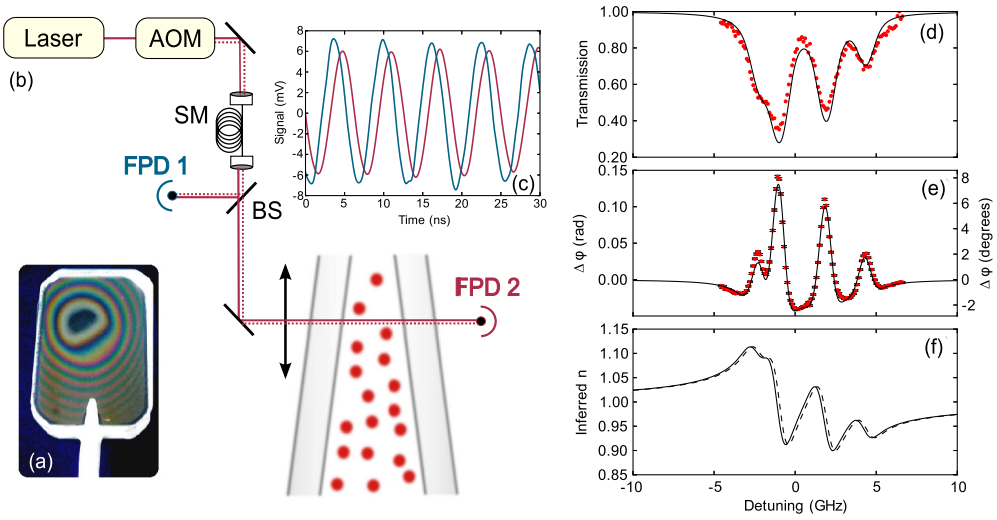

FIG. 1 (color online). Experimental setup and example data. (a) Photograph of experimental nanocell used in the experiment. The cell has a wedge profile (shown schematically) resulting in a tunable vapor thickness. (b) Schematic of experimental setup. For details see main text. AOM—acousto-optic modulator, SM—single mode fiber, BS—50:50 beam splitter, FPD1/2—fast photodiodes. (c) Beat frequency between shifted and unshifted beams for each detector, from which a phase difference is measured. As the laser frequency is scanned, we obtain transmission (d) and relative phase shift (e) information, shown here for a vapor thickness $\ell = \lambda/2$ and $T = 250\,^{\circ}\mathrm{C}$ across the $D2$ resonance, fitted to theory (solid black lines). Panel (f) shows the inferred refractive indices for the shifted (dashed) and unshifted (solid) beams—the solid line in (e) is the difference of these two curves. Zero on the detuning axis represents the weighted line center of the $D2$ line.

vapor layer is confined between two sapphire plates with a tunable separation between 30 nm and 2 μm [the cell is shown in Fig. 1(a), for more details see Ref. [17]]. Sapphire is used for the cell windows as it is chemically resistant to Rb vapor up to $\sim$1000 °C [19]. By controlling the temperature of the cell, we control the atomic number density N. We heat from room temperature to 350 °C, corresponding to $N \approx 10^{17}$ cm^{-3}. To measure the refractive index, we employ heterodyne interferometry similar to that of Pototschnig *et al.* [20]. An acousto-optic modulator (AOM) is used to generate a second beam at $\Delta_{\mathrm{AOM}}/(2\pi) =$ 160 MHz, which is power matched and then recombined with the probe beam and coupled into a single-mode optical fiber (which ensures mode matching). Half of the light is sent to a fast photodiode (FPD1) and the other half focussed to a spot size ($1/e^2$ radius) of 50 μm through the cell and onto a second fast photodiode (FPD2). Both detectors measure the beat frequency of the two light fields, and for any one measurement we observe $\sim$300 oscillations. The phase of the two signals is measured as the laser is scanned across the $D2$ resonance line of Rb ($\lambda = 780.2$ nm). Since the two beams are at nearly the same frequency, the phase difference is given by

$$\phi = \phi_0 + \Delta\phi = \phi_0 + [n(\Delta + \Delta_{\mathrm{AOM}}) - n(\Delta)]k\ell, \qquad (2)$$

where ϕ_0 is an unimportant global phase due to different optical path lengths to the two detectors. Sample data are shown in panel (c). We reconstruct the refractive index profile from fitting the measured relative phase $\Delta\phi$ using Eq. (2). Panels (d), (e), and (f) show example data for a vapor thickness equal to half the optical wavelength, $\ell = \lambda/2$. For high N where the homogeneous broadening is much greater than Δ_{AOM}, we can simultaneously measure transmitted intensity by measuring the amplitude of the oscillations on FPD2.

An interesting question that arises when considering an index enhancement is what is the maximum possible index of the medium? To predict this we use a model of atomic susceptibility that includes hyperfine structure, Doppler broadening [21], Rb-Rb self-broadening [22], and magnetic fields [23], with excellent agreement to experimental transmission spectra at the 0.5% level. We have also extended this model to include effects of Dicke narrowing, atom-wall interactions and the cooperative Lamb shift in vapor cells with nanometre thickness [17].

While other works have speculated that the near-resonance refractive index of a gaseous medium could be "as high as 10 or 100" [24], this estimate was based on an independent dipole model and neglected the dipole-dipole interactions which are invariably present at high density. Eventually, the increase in susceptibility due to adding more dipoles is exactly canceled by the higher damping rate due to resonant dipole–dipole interactions [25,26], so there is no further increase in index. A maximum index of $n \approx 1.4$ has previously been predicted for Rb [27]. Using our model, we calculate the maximum refractive index to be $n = 1.31$. This occurs at $T \approx 360\,^{\circ}\mathrm{C}$ at a detuning $\Delta \sim -7$ GHz from the weighted line center, shown in Fig. 2(a). The redshift of the resonance due to the cooperative Lamb shift can be seen clearly (calculated for

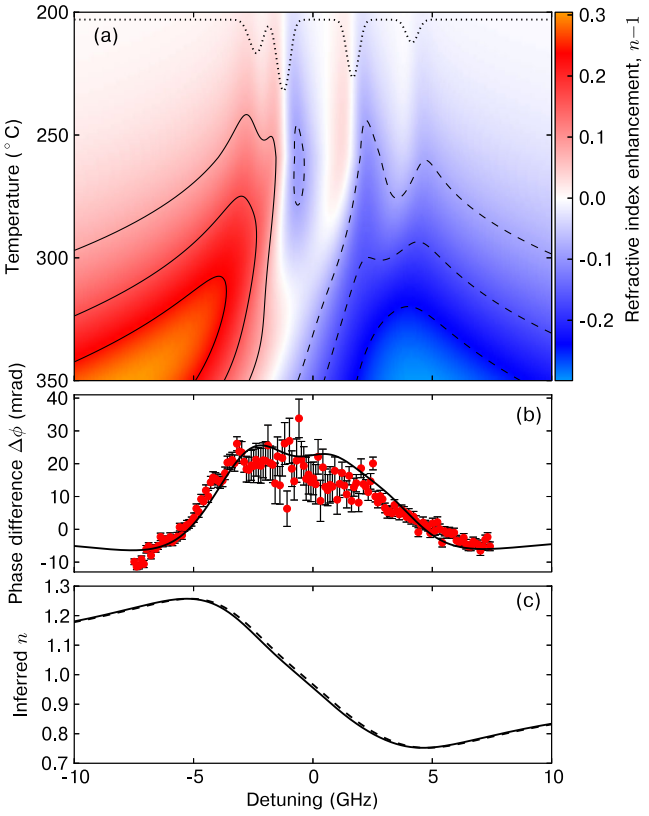

FIG. 2 (color online). (a) Calculated refractive index as a function of temperature and detuning for the Rb $D2$ resonance. Solid (dashed) contour lines denote positive (negative) values. The maximum predicted index is 1.31. The density-dependent redshift can clearly be seen, and has a significant effect on the position of the maximum index. The dotted line shows the positions of the unshifted Doppler-broadened lines. (b, c) Experimental data for a thickness $\ell = 250$ nm and $T = 330\,^{\circ}\mathrm{C}$ ($N = 5 \times 10^{16}$ cm^{-3}). Close to resonance, the large optical depth reduces signal to the point where accurate phase information is lost. Despite this, the fit is reasonable and we infer a maximum index $n = 1.26 \pm 0.02$.

$\ell \gg \lambda$, see Ref. [17]) from the figure. Beyond $T \approx 360\,^{\circ}\mathrm{C}$ the binary approximation breaks down, and one must consider a multiperturber model, which is beyond the scope of this work.

To observe the maximum index, we require $T = 360\,^{\circ}\mathrm{C}$, $N \approx 1 \times 10^{17}$ cm^{-3}. Consequently, for these parameters the resonant absorption coefficient is extremely large, and therefore the vapor thickness must be much less than λ to transmit a measurable amount of light. As one reduces the vapor thickness, however, atom-wall interactions start to have a significant effect on the linewidth, introducing further undesirable broadening and shifts which act to reduce the maximum observable index. Despite this, we can still observe a large index, as shown in Figs. 2(b) and 2(c) for experimental data obtained with vapor thickness $\ell = 250$ nm, at $T = 330\,^{\circ}\mathrm{C}$. For these conditions, the large on resonance optical depth reduces the signal to the point where phase cannot be accurately measured. Away from resonance, however, we observe good agreement with theory. From this we infer a peak refractive index of 1.26 ± 0.02 approximately 5 GHz red detuned of line center with $\sim$40% transmission. The absorption drops off with detuning much faster than the refractive index decreases. Extrapolating to larger detuning at $\Delta = -15$ GHz we can still have $n \sim 1.13$ with $>$90% transmission. We attribute the difference in theoretical and experimental refractive indices here to additional broadening from the van der Waals atom-surface interaction [included in the theory curves of Figs. 2(b) and 2(c)], which has been characterized for the Rb-Sapphire interface based on a $1/r^3$ potential from each cell wall [28].

Intuitively, one might anticipate that the largest group index coincides with the largest index enhancement, but this is not the case. The group index is given by $n_g(\omega) = n + \omega dn/d\omega$ and can thus be calculated in the same way as for Fig. 2(a). This is shown in Fig. 3(a). We see a clear maximum around the position of the strongest ground state transition (^{85}Rb, $F_g = 3 \rightarrow F_e = 2, 3, 4$, $\Delta \approx -1.2$ GHz), where we predict a very large negative group index $n_g \approx -1.2 \times 10^5$ at $T \approx 255\,^{\circ}\mathrm{C}$. By tuning the laser frequency we can move between fast- and slow-light regimes, but the maximum positive group index is smaller by a factor of $\sim$3. The spectral broadening at this density creates a region approximately 1 GHz wide with little group velocity dispersion. While increasing the density further than this increases the bandwidth available, the additional broadening smears out the resonance, reducing $dn/d\omega$ and hence n_g. For clarity, the cooperative Lamb shift has been neglected in Fig. 3(a), since it has a negligible effect on the position of the maximum group index.

We probe this region with a weak optical pulse with a full width at half-maximum (FWHM) of 800 ps, corresponding to a pulse bandwidth of 1.1 GHz, in a region with vapor thickness $\ell = \lambda/2 = 390$ nm. To measure optical pulses, we lock the laser at an arbitrary frequency electronically using a laser wavelength meter and build up a histogram of the arrival time of photons on a single photon counting module. The bandwidth of the detection equipment is much larger than that of the detected features. We record a reference pulse with the laser far detuned from resonance (pulse center detuning $\Delta_c = -13$ GHz) so there is no interaction with the medium ($n_g \approx 1$). We switch the laser onto resonance to measure the effect of the medium. Figure 3 panels (b) and (c) show experimental data. Panel (b) shows both signals normalized to the peak intensity of the reference pulse, while panel (c) shows the pulses scaled to have the same height, to highlight the advance of the resonant pulse. On resonance the peak is advanced, arriving a time $t_d = (-0.13 \pm 0.01)$ ns earlier than the vacuum pulse. Using $t_d = n_g \ell/c$, this gives a group index $n_g = -(1.0 \pm 0.1) \times 10^5$. The dashed lines in Fig. 2(b) are theory curves based on a Fourier transform method and the susceptibility model, assuming a Gaussian input pulse

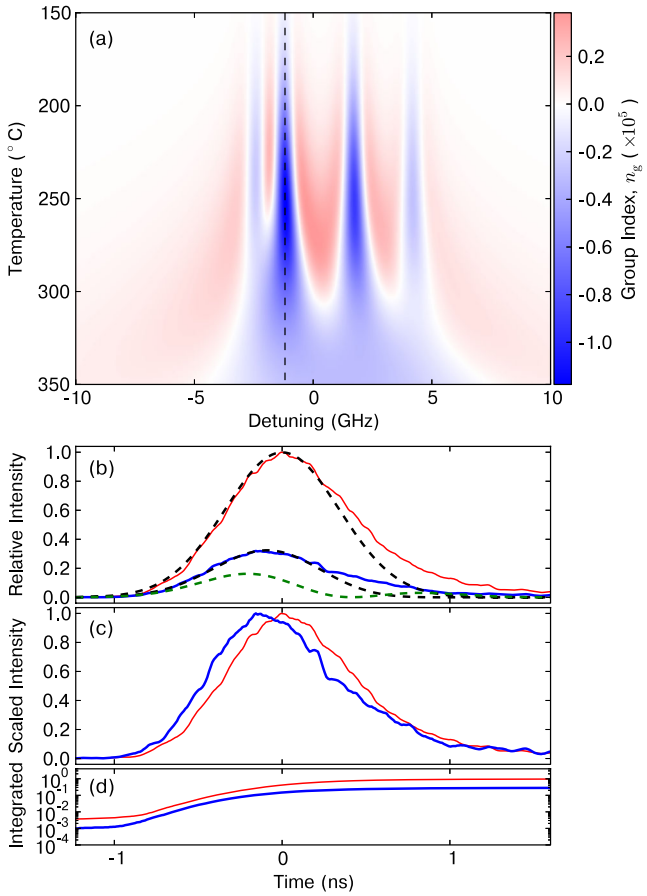

FIG. 3 (color online). (a) Calculated group index as a function of temperature and detuning. The dashed black line represents the carrier detuning for the superluminal pulse. (b,c) Superluminal propagation of a 800 ps pulse through a vapor thickness $\ell = \lambda/2$ and temperature $T = 255\,^{\circ}\mathrm{C}$. Solid lines are experiment, dashed black lines are theory with an 800 ps FWHM Gaussian input pulse. The dashed green line is the theory without dipole-dipole interactions. Off resonance ($\Delta_c = -13$ GHz, thin red line) there is no interaction at this temperature and the pulse propagates through the vapor as it would through vacuum. On resonance, ($\Delta_c = -1.2$ GHz, thick blue line), the pulse is attenuated and arrives (0.13 ± 0.01) ns earlier than the off-resonance reference pulse, corresponding to a group index $n_g = -(1.0 \pm 0.1) \times 10^5$. (d) Total integrated counts for both signals verifies preservation of causality—the probability of detecting a photon is always higher in the reference pulse.

with 800 ps FWHM, which agree well with data on the rising edge. On the falling edge, the structure is complicated by fluorescence from the decay of the excited state, which has an exponential decay with a lifetime $\tau \approx 1$ ns, corresponding to the time of flight of atoms across the cell. The large spectral bandwidth available means we see little distortion of the resonant pulse. It is important to note that without the dipole-dipole interactions, the propagation of such temporally short pulses would not be possible without heavy distortion. To illustrate this, the green dashed line in Fig. 3(b) shows the calculated pulse profile if there were no dipole-dipole interactions. In this case, the pulse has a much larger bandwidth than the transition (and spans multiple excited state hyperfine levels) and therefore distortion of the output is evident. Even though there is a superluminal component which is more advanced than for the interacting ensemble, there is also a subluminal component which can be seen at $t \sim 0.8$ ns. Panel (d) shows the total integrated counts over the detection period, which verify that the advanced pulse preserves causality [11], since there is always greater probability of detecting a photon in the reference pulse than the advanced pulse. From this we can immediately surmise that the information velocity of the advanced pulse is $< c$.

Since the advance scales linearly with the thickness of the medium, by making the vapor thicker we would increase the advance of the resonant pulse, but at the cost of more attenuation which scales exponentially with the thickness. We estimate that with the present experimental setup we could achieve a maximum fractional pulse advancement of 0.55 with 1% transmission, which is the limit set by the current signal-to-noise ratio.

In demonstrating giant refractive index and group index, we open the door to further investigation of slow- and fast-light effects in the cooperative interaction regime, where dipole-dipole interactions play an important role [17]. The GHz bandwidth available enables control of the dynamics on a faster time scale than the natural lifetime of the excited state (27 ns). Since cooperative effects are dependent on the level of excitation in the medium [29], a pump-probe setup with a strong and weak pulse and a short time delay may yield interesting physics, such as sub- or superluminal gain in a transiently inverted medium. This will form the basis of future research.

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[1] L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, Nature (London) **397**, 594 (1999).
[2] L. J. Wang, A. Kuzmich, and A. Dogariu, Nature (London) **406**, 277 (2000).
[3] G. M. Gehring, A. Schweinsberg, C. Barsi, N. Kostinski, and R. W. Boyd, Science **312**, 895 (2006).
[4] S. E. Harris, J. E. Field, and A. Imamoğlu, Phys. Rev. Lett. **64**, 1107 (1990).
[5] M. Fleischhauer, A. Imamoğlu, and J. Marangos, Rev. Mod. Phys. **77**, 633 (2005).
[6] M. Fleischhauer and M. D. Lukin, Phys. Rev. Lett. **84**, 5094 (2000).
[7] B. Julsgaard, J. Sherson, J. I. Cirac, J. Fiurášek, and E. S. Polzik, Nature (London) **432**, 482 (2004).
[8] R. M. Camacho, M. V. Pack, J. C. Howell, A. Schweinsberg, and R. W. Boyd, Phys. Rev. Lett. **98**, 153601 (2007).

[9] P. Siddons, N. C. Bell, Y. Cai, C. S. Adams, and I. G. Hughes, Nature Photon. **3**, 225 (2009).
[10] M. D. Stenner, D. J. Gauthier, and M. A. Neifeld, Nature (London) **425**, 695 (2003).
[11] A. Kuzmich, A. Dogariu, L. J. Wang, P. W. Milonni, and R. Y. Chiao, Phys. Rev. Lett. **86**, 3925 (2001).
[12] R. W. Boyd, Z. Shi, and P. W. Milonni, J. Opt. **12**, 104007 (2010).
[13] M. O. Scully, Phys. Rev. Lett. **67**, 1855 (1991).
[14] A. S. Zibrov, M. D. Lukin, L. Hollberg, D. E. Nikonov, M. O. Scully, H. G. Robinson, and V. L. Velichansky, Phys. Rev. Lett. **76**, 3935 (1996).
[15] R. W. Boyd and P. Narum, J. Mod. Opt. **54**, 2403 (2007).
[16] H. Tanaka, H. Niwa, K. Hayami, S. Furue, K. Nakayama, T. Kohmoto, M. Kunitomo, and Y. Fukuda, Phys. Rev. A **68**, 053801 (2003).
[17] J. Keaveney, A. Sargsyan, U. Krohn, I. G. Hughes, D. Sarkisyan, and C. S. Adams, Phys. Rev. Lett. **108**, 173601 (2012).
[18] J. D. Pritchard, D. Maxwell, A. Gauguet, K. J. Weatherill, M. P. A. Jones, and C. S. Adams, Phys. Rev. Lett. **105**, 193603 (2010).
[19] E. Jahier, J. Guéna, P. Jacquier, M. Lintz, A. V. Papoyan, and M. A. Bouchiat, Appl. Phys. B **71**, 561 (2000).
[20] M. Pototschnig, Y. Chassagneux, J. Hwang, G. Zumofen, A. Renn, and V. Sandoghdar, Phys. Rev. Lett. **107**, 063001 (2011).
[21] P. Siddons, C. S. Adams, C. Ge, and I. G. Hughes, J. Phys. B **41**, 155004 (2008).
[22] L. Weller, R. J. Bettles, P. Siddons, C. S. Adams, and I. G. Hughes, J. Phys. B **44**, 195006 (2011).
[23] L. Weller, T. Dalton, P. Siddons, C. S. Adams, and I. G. Hughes, J. Phys. B **45**, 055001 (2012).
[24] M. Fleischhauer, C. H. Keitel, M. O. Scully, C. Su, B. T. Ulrich, and S. Y. Zhu, Phys. Rev. A **46**, 1468 (1992).
[25] J. Keaveney, A. Sargsyan, U. Krohn, J. Gontcharov, I. G. Hughes, D. Sarkisyan, and C. S. Adams, arXiv:1109.3669v2.
[26] C. O'Brien, P. M. Anisimov, Y. Rostovtsev, and O. Kocharovskaya, Phys. Rev. A **84**, 063835 (2011).
[27] Z. J. Simmons, N. A. Proite, J. Miles, D. E. Sikes, and D. D. Yavuz, Phys. Rev. A **85**, 053810 (2012).
[28] J. Keaveney, A. Sargsyan, I. G. Hughes, D. Sarkisyan, and C. S. Adams (unpublished).
[29] R. Friedberg, S. R. Hartmann, and J. T. Manassah, Phys. Rep. **7**, 101 (1973).