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Beam Propagation near the Dispersion-less Wavelength at 790nm in Rubidium

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Between any two resonance lines for an atomic media exists a dispersion-less wavelength where the ac-Stark shift from the two resonances cancel and the real part of the resonant susceptibility is zero. We experimentally demonstrate the effects of this wavelength in a pump-probe experiment in high density atomic vapour. A strong pump field excites the media, in which we counter propagate a broadband probe field. In a long cell only at the dispersion-less wavelength will scattering and self focusing/defocusing not cause attenuation, allowing a spectrometer to read off the dispersion-less wavelength. We perform a propagation experiment that shows the passband centered around 790nm dispersion-less wavelength between the D₁ and D₂ lines of Rubidium.

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I. INTRODUCTION

In between any two resonances there is a point where the contributions to the ac-Stark effect cancel with each other, leaving a wavelength for which there is no resonant dispersion. Since when red detuned to a resonance, there is positive resonant susceptibility and blue detuned to a resonance there is negative resonant susceptibility, it is clear that there must be a point where the resonant susceptibility is zero, a dispersion-less point, as shown in Fig.1. There is also a dispersion-less point exactly on resonance, as the resonant refractive index switches sign, but this wavelength has strong absorption, making it less useful for many applications. These dispersion-less wavelengths have been calculated for all Alkali atoms [1, 2], but only a few have been experimentally verified.

Since these dispersion-less wavelengths are directly tied to the matrix elements of atomic transitions, measuring them helps check theoretical calculations of atomic structure [3]. Better knowledge of the matrix elements also helps improve uncertainties in atomic clocks [3, 4]. These wavelengths can also be used for applications that require the ability to tune-out the ac-Stark shift, such as forming species selective optical lattices [2]. It also has been suggested that the position of these dispersion-less wavelengths can be controlled by applying magnetic fields to shift the frequency of the atomic energy levels in Rubidium, in order to enable the easier generation of optical lattices [4].

But the most important application of these dispersion-less wavelengths, we believe is for the observation and control of collective effects. For example, a new technique for the generation of high frequency light from low frequency pumping using collective excitation and parametric difference frequency generation was recently suggested [5], where it is necessary to work at a point where the ac-Stark effect can be neglected. Also

many experiments involving collective effects and multiple pulses, struggle to have a long interaction region due to dispersion. Thus previous collective experiments such as [6], were typically performed at resonance, sometimes utilizing a dense but sub-wavelength, thin layer of atoms [7, 8]. Working directly at resonance is not always desirable due to strong absorption of the applied fields. Therefore the dispersion-less wavelengths are ideal for future experiments that rely on a long interaction region for collective excitation.

We develop an experimental technique to study and observe the effects of these dispersion-less wavelengths on beam propagation in atomic vapour. A strong pump field excites the atomic media, in which we counter propagate a broadband probe field. The spatial(radial) intensity profile of the pump field leaves a spatially varying population as it travels through the cell. This spatially varying refractive index then acts as a graded index (GRIN) lens, for the counter-propagating probe field. Therefore, in a long gas cell only at the dispersion-less wavelength will the probe travel freely, at all other wavelengths the probe field will be focused or defocused. If a collection lens is placed in the near field both focusing and defocusing will strongly attenuate the probe light that makes it to the detector. By propagating a broadband probe field, a spectrometer can read off the dispersion-less wavelength. We use this technique to observe the dispersion-less wavelength at 790nm in Rubidium vapour. We note that dispersion-less wavelengths in Rubidium were experimentally measured to high precision using Bose-Einstein condensates (BEC) [3, 9], here we focus on the effects of dispersion on beam propagation and window of free passage around this wavelength in hot and high density Rubidium vapour, and this technique can also be used for identifying dispersionless wavelengths in other atomic species with which BEC based interferometers are difficult to achieve.

First we explain how the pump field creates a gradient refractive index lens, which focuses or defocuses any probe field that is not at the dispersion-less wavelength. Then we explain how we perform the experiment to de-

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tect the dispersion-less wavelength at 790nm, between the D_1 and D_2 resonances of Rubidium and show the results. Finally, we discuss some applications and implications for the dispersion-less 790nm, in particular how this frequency is ideal for observing collective effects in atomic vapor.

II. GRADED INDEX LENS

Between two resonance lines is a zero in the resonant susceptibility. This is demonstrated in Fig.1. At this dispersion-less wavelength, not only is there no resonant dispersion, but the ac-Stark effects from each transition cancel.

Here we are particularly interested in the dispersion-less wavelength between the D_1 and D_2 lines of Rubidium at 790nm [2]. For a probe field of linear polarization, we can treat the Rubidium as a superposition of two two-level atomic responses, one for the D_1 line and one for the D_2 line. In absence of the pump field, the resonant susceptibility as a function of probe frequency can be expressed as [10],

$$\chi(\omega) = \frac{N}{\epsilon_0 \hbar} \left(\frac{\mu_{D1}^2}{\omega_{D1} - \omega - i\gamma_{D1}} + \frac{\mu_{D2}^2}{\omega_{D2} - \omega - i\gamma_{D2}} \right), \quad (1)$$

where N is the Rubidium density, ω is the probe frequency, μ_{D1} and μ_{D2} are the dipole matrix elements, ω_{D1} and ω_{D2} are the transition frequencies, γ_{D1} and γ_{D2} are the total transition linewidths, for the D_1 and D_2 lines. Using Racah algebra for matrix elements involving coupled angular momentum states[11], the ratio between μ_{D2}^2 and μ_{D1}^2 can be calculated to be 2, higher precision calculations can be found in [12]. There exists a frequency ω , between ω_{D1} and ω_{D2} , such that the contributions from D_1 and D_2 cancel and $\chi(\omega)$ is zero at that frequency, which for Rubidium happens at 790nm. For circularly polarized probe field, the calculation of susceptibility involves more states. Yet due to the correlations among the matrix elements of Rubidium D_1 and D_2 lines, as long as the population begins evenly distributed among the spin levels of the ground state, contributions from D_1 and D_2 transitions have the same ratio as the case of linearly polarized probe, therefore the dispersion-less wavelength calculated for linear polarization from Eq.(1) is also valid for the case of circularly polarized probe field.

In Eq.(1), we have also assumed that all the atoms are in the ground state. In the presence of pump field, fraction of population will be excited to upper states, and the susceptibility seen by the probe field will be modified to:

$$\chi(\omega) = \frac{N}{\epsilon_0 \hbar} \left[\frac{\mu_{D1}^2 (\rho_g - \rho_e^{D1})}{\omega_{D1} - \omega - i\gamma_{D1}} + \frac{\mu_{D2}^2 (\rho_g - \rho_e^{D2})}{\omega_{D2} - \omega - i\gamma_{D2}} \right], \quad (2)$$

FIG. 1. Resonant susceptibility as a function of laser wavelength for a natural abundance of Rubidium, at a temperature of 250°C. The real part of the resonant susceptibility goes through zero at the two transition wavelengths and at the dispersion-less wavelength at 790nm.

where the terms $\rho_g - \rho_e^{D1}$ and $\rho_g - \rho_e^{D2}$ represent the population difference of ground and excited states. If the probe frequency is away from the dispersion-less wavelength, the D_1 and D_2 terms in the bracket of Eq.(2) will not cancel and the susceptibility depends on the population distribution, and thus can spatially vary due to the radial variation of the pump field.

Starting with the exact time domain solution of the density matrix for a two level atom [13], we can calculate the average difference in population between the ground and the excited state in the presence of a pump field:

$$\rho_g - \rho_e = 1 - \frac{|\Omega|^2}{|\Omega|^2 + \Delta^2}, \quad (3)$$

where $\Omega = \vec{d} \cdot \vec{E}/\hbar$ is the Rabi frequency of the pump, while $\Delta = \omega_p - \omega_0$ is the detuning of the transition frequency ω_0 from the pump laser frequency ω_p . We assume a pump field strong enough to saturate the transition on resonance, thus we take $|\Omega| \gg \gamma$. The pump field propagating through the medium has a radial intensity profile, which causes a radial distribution of the Rabi frequency and thus the the atomic population. This causes a radial distribution of the resonant susceptibility. Along the length of the cell the amount of absorption of the pump field is less than 1% and for simplicity we neglect the self focusing/defocusing of the pump, so there is no significant z-dependence to the susceptibility.

When the probe field propagates through the medium the presence of the radially distributed resonant susceptibility creates a GRIN lens. Where instead of having a curved interface between two different refractive index materials, there is a gradual change of the refractive index that nevertheless still behaves similarly to a classical lens. The radial profile for probe wavelengths where the resonant susceptibility is positive, creates a defocusing lens since at the center of the beam the population is saturated and thus smaller than at the edges. The radial profile for the probe wavelengths with negative resonant susceptibility a focusing lens is created since at the

center of the beam the refractive index is higher. Except when the susceptibility is zero at the dispersion-less wavelength, then the light is allowed to travel unimpeded to the detector. If the population differences between the excited states of D_1 and D_2 transitions can be neglected (i.e. assuming $\rho_e^{D1} = \rho_e^{D2}$), the dispersion-less wavelength remains at the same position as the case of no pump (Eq.(1)). If the difference between ρ_e^{D1} and ρ_e^{D2} is taken into account, the dispersion-less wavelength itself can also shift slightly from the zero-crossing position predicted by Eq.(1) and have a spatial dependence determined by the radial intensity profile of pump. However, in cases where the pump field transfers the ground state population to states other than the states of D_1 and D_2 lines (for example, to the 5^2D level via two-photon transition), the assumption of $\rho_e^{D1} = \rho_e^{D2}$ is sufficiently accurate (they both can be considered as zero) and only ρ_g has spatial dependence on radial profile of the pump beam, therefore the D_1 and D_2 terms in Eq.(2) will cancel at the same dispersion-less wavelength as in Eq.(1).

Propagation of the probe beam can be understood by studying ray tracing of the beam through the GRIN. This is done by solving the ray equation:

$$\frac{d}{ds} \left(n(\vec{r}) \frac{d\vec{r}}{ds} \right) = \nabla n(\vec{r}), \quad (4)$$

where $\vec{r}$ is the position vector of a point along the ray, ds is the element of arc length along the ray, and $n(\vec{r})$ is the radially varying refractive index distribution. Using the method of ray tracing given in [14] we plot the effects of a defocusing lens and focusing lens in Fig. 2.

III. PROPAGATION EXPERIMENT

We performed pump-probe experiments in high density Rubidium vapor. A natural abundance of Rubidium is pre-loaded in a 2.5cm diameter and 5cm long Pyrex cell without buffer gas. The cell is heated to temperatures near 200-250°C to reach a Rb density at levels of 10^{15}cm^{-3} . Two Ti:Sapphire laser systems provide the pump and probe pulses at a repetition rate of 10Hz. The pump system consists of a regenerative Ti:Sapphire amplifier seeded by a tunable cw Ti:Sapphire oscillator. The pump beam has typical energy of 0.5 to 1 mJ, pulse duration of 18ns, linewidth of 0.1nm and can be tuned from 765nm to 805nm. The pump laser is multi-longitudinal-mode with a mode spacing of $2 \times 10^{-4} \text{nm}$, therefore, about 500 modes are contained within the linewidth of pump field. At a total pump intensity on the order of 10^7W/cm^2 , the Rabi frequency of each mode is about $6 \times 10^{10} \text{s}^{-1}$ for exciting the $5S-5P_{3/2}$ transition of Rubidium, which is significantly less than the typical detuning of pump field from the $5S-5P_{3/2}$ resonance (about $3 \times 10^{12} \text{s}^{-1}$ for each nm of detuning). Eq.(3) can be used to estimate the population excited by each mode of the pump, and the total excitation is the sum for all the modes. The probe laser can be operated either as

FIG. 2. Ray tracing plots for the probe laser of diameter $250 \mu\text{m}$ propagating through 5cm of the medium where the refractive index difference Δn (compared to the area outside the pumped region) is uniform along the propagation axis, and distributed as a Gaussian in the radial direction with a full width at half maximum of $400 \mu\text{m}$, which we use to model the distribution due to the pump laser. Here we choose refractive index changes to be similar to those in the experiments. (a) The peak refractive index difference is taken as $\Delta n_{max} = -.0001$ which creates a defocusing gradient refractive index lens. (b) The peak refractive index difference is taken as $\Delta n_{max} = +.0001$ which creates a focusing gradient refractive index lens.

a narrow-band (linewidth $< 0.1 \text{nm}$) pulse tunable from 765nm to 805nm, or as a broadband pulse having a spectrum covering the range of 770-810nm. The probe has a pulse duration of 15ns, the energy of the probe pulse is attenuated to the order of 30nJ before entering the cell. The timing of both pump and probe pulses are controlled by intra-cavity Pockel cells in each system and the two systems are synchronized by digital delay generators from Stanford Research Systems (SRS). Both beams are linearly polarized and the relative delay between the pump-probe pulses is controlled by varying the Pockel cell timing.

To observe the effects of the GRIN formed by Rubidium under a near resonance pump, we compare CCD images of the spatial profile of probe beam after it passes through the cell, as shown in Fig.3. In this data, probe has a bandwidth of less than 0.03nm. The pump laser counter-propagates with the probe. The pump and probe are focused to cell center and both beams have diameters of 0.8mm. The CCD is positioned 55cm after the cell, within the Rayleigh range of the probe. Without the pump, the probe beam profiles (left column in Fig.3)

FIG. 3. CCD images of the spatial profile of a narrow band-width probe beam after passing through the rubidium cell. In the presence of pump, the rubidium vapour behaves as an positive or negative lens seen by the probe field, leading to focusing or defocusing, except at the dispersion-less wavelength. For this data, the cell is at temperature 230°C . The pump has intensity of $6 \times 10^6 \text{W/cm}^2$ and counter-propagates with the probe. Both the pump and probe are focused to cell center and have diameters of 0.8mm .

look very similar to each other, in fact they are all similar to the patterns when the cell is cold. However, the presence of the pump leads to a radially dependent phase shift on the probe, effectively forming a positive or negative lens depending on probe wavelength, as shown in Fig.3(b) and (f), except for the case when the probe is near 790nm , i.e. Fig.3(d). In the vicinity of 790nm , the phase shift due to Rubidium is nearly zero to start with, and the pump beam will not cause any spatially dependent phase shift on the probe, the spatial profile of beam pattern is preserved.

The dispersion-less wavelength of rubidium can also be observed with a broadband probe together with a spectrometer. As shown in Fig.4(a), the probe is focused into the cell and the transmitted probe spectra is recorded by a 0.5m spectrometer positioned 90cm after the cell. The

FIG. 4. (a) Experimental setup showing forward propagating pump field, and backward propagating probe field. (b) Transmitted spectra of the probe pulse with (blue) or without (red) the pump pulse. For this data, the cell is heated to 250°C , the pump is at 775nm and the intensity is $2.5 \times 10^7 \text{W/cm}^2$. The absorption of Rubidium is shown as dips at wavelengths corresponding to $5\text{S}-5\text{P}_{1/2}$ (795nm) and $5\text{S}-5\text{P}_{3/2}$ (780nm) transitions. The narrow line at 775nm in the spectra with pump is the back reflection of pump beam from several sources including stray reflections/scattering due to cell window and mirrors.

spectral resolution of spectrometer is 0.15nm at wavelength of 800nm . At cell center, the diameter of the probe beam is $250\mu\text{m}$. The counter propagating pump beam is focused into the cell at a diameter of $400\mu\text{m}$ and peak intensity of $2-3 \times 10^7 \text{W/cm}^2$. A collection lens is used after the cell to image the probe beam onto the slit of spectrometer. Fig.4(b) shows transmitted spectra of a broadband probe at a cell temperature of 250°C , with or without the pump pulse. In absence of the pump, Rubidium absorption corresponding to the $5\text{S}-5\text{P}_{1/2}$ (795nm) and $5\text{S}-5\text{P}_{3/2}$ (780nm) transitions are shown as dips in the probe spectra. The measured spectra is a convolution of the real spectrum of transmitted probe with a window function whose width is determined by the resolution of the spectrometer, so the width and depth of the absorption dips are limited by the spectrometer resolution at lower temperatures (densities) until the width of the Voigt profile of the absorption spectra exceeds the spectrometer resolution.

In the presence of a 775nm and $2.5 \times 10^7 \text{ W/cm}^2$ pump pulse, the probe beam has good transmission only near the region of 790nm. It should be emphasized that, in our detection scheme, the recorded spectra represent the fluence (i.e. energy per unit area) instead of the total energy of each frequency component in the probe beam at the position of the spectrometer entrance slit ($30\mu\text{m}$ wide). If any frequency component has a relatively different spatial distribution profile after propagating through the Rubidium vapor, it will lead to a change in the spectra recorded by the spectrometer, even if the total energy of this frequency component over the whole beam area remains unchanged. Therefore, the spectra is not only influenced by the absorption or amplification which is related to the imaginary part of the susceptibility of the media, but also by the alteration of the phase-front which is related to the refractive index or real part of the susceptibility. The extra (wavelength-dependent) focusing or defocusing caused by the Rubidium on the probe will alter the imaging geometry of the collection lens after the cell, therefore reducing the amount of light entering into the slit of spectrometer for the corresponding wavelength component in the broadband probe.

Fig.5 shows transmitted probe spectra under different cell and pump conditions. The evolution of the spectra at different pump-probe delay time in Fig.5(a) shows that the effects of the pump on the probe spectra can last at least of a few μs . The reason this is much longer than the spontaneous decay time of the $5P_{1/2}$ or $5P_{3/2}$ levels is, we believe, due to the radiation trapping effects in the high density Rubidium vapor. In high density atomic vapors, each emitted photon from an excited atom can be re-absorbed and re-emitted repeatedly by other atoms, as a result of this “radiation trapping” effect, the effective radiative decay rate of a resonance line can be significantly decreased. The effective radiative rate is often described as the natural decay rate multiplied by an escape factor “g”, which is the inverse of the mean number of absorption events. For simplicity, we adopt the approximate formulation on escape factor in [15] to have an order of magnitude estimate. Assuming conditions of Doppler broadened line, high opacity and uniformly distributed excited-state atoms in a geometry of infinite cylinder, the escape factor can be approximated as[15]:

$$g = \frac{2}{\sqrt{\pi}} \frac{\sqrt{\ln(\alpha_0 R)}}{\alpha_0 R} \quad (5)$$

where α_0 is the absorption coefficient at line center and R is the radius of the cylinder containing the excited atoms, here in our case is the radius of pump beam ($R=200\mu\text{m}$). Under conditions of our experiment, the Doppler width is 0.7GHz for the $5S-5P_{3/2}$ transition at $T=250^\circ\text{C}$, and $\alpha_0 R$ is on the order of 10^3 , the corresponding escape factor is about 10^{-3} . That is, the estimated effective decay time is 3 orders of magnitude longer than the natural decay time (26ns). The diffusion of atoms is not included in the above simplified estimate. At cell temperature of 250°C , the thermal transit time of atoms across the

FIG. 5. Transmitted probe spectra (a) at different delays after the pump pulse (b) at different pump intensities (c) at different cell temperatures. For these data, the Rubidium cell is 5cm long.

pumped region (defined by pump beam diameter $400\mu\text{m}$) is also at μs level. Therefore, the dominating time constant here observed experimentally may be combination of the thermal transit time through the active region and the delayed population decay under radiation trapping.

Fig.5(b) and (c) display the probe spectra at different pump intensities and cell temperatures. At lower pump intensities or cell temperatures (which means lower Rubidium vapor density), the “passband” near 790nm widens and approaches to the case when there is no pump. This is in agreement with the fact that, the GRIN lens is weaker with either lower pump intensity or lower vapor density. The width and shape of the passband is dependent on the imaging setup for the probe beam from the cell center to the spectrometer entrance slit and also on the optical alignment between probe and pump beam, which could lead to variations under different runs. For example, in Fig.5(c) the plot for $T=253^\circ\text{C}$ shows similar main feature of a passband near 790nm as the plot in Fig.5(b) with pump intensity $2.5 \times 10^7\text{W}/\text{cm}^2$, but the actual width and shape is slightly different, even though the cell temperature and pump intensity are similar, this is due to the drift of the spatial mode profile of the probe beam and its alignment with the pump beam.

IV. DISCUSSION

Both the spatial profile and the spectroscopy measurements on the probe have demonstrated the effects of the GRIN induced by the strong pump field. The probe field travels freely through the media only near the dispersion-less wavelength since only there does the GRIN effect not happen. The same would happen at resonance, but there the absorption is too strong for the probe to travel through the media. Similar effects could be realized from having a coherent grating in the medium, but for a coherent grating to exist, it has been shown that the coherence time of the medium needs to be longer than the pulse length [6]. For Rubidium gas at 250°C with a density of $4 \times 10^{15}\text{cm}^{-3}$, the collisional broadening for the D_1 or D_2 lines can be estimated to be around 0.2GHz [10], which means the collisional effects alone would limit the coherence time to around 2ns or less, almost an order of magnitude shorter than the pump pulse length (18ns). This was also demonstrated by changing the delay time between the pump and probe field which showed that the effect remains until delays of μs , which are much longer than the coherence time and similar to the time it takes the population distribution to diffuse, as expected with the GRIN effect. It is also possible for the signal to be reflecting off of a spatially varying refractive index, or “population grating.” Such a population grating would form along the axis from the back reflection of the pump field, but that can be ruled out since the glass at the cell exit is angled at 5° , which when combined with the low 4% reflection off uncoated glass, does not give a strong enough reflection to cause a population grating to form

FIG. 6. The transmitted probe spectrum has a FWHM width of 0.8nm with a 10cm long rubidium cell, heated to 252°C (Rubidium density of $5 \times 10^{15}\text{cm}^{-3}$). The pump laser has intensity of $2 \times 10^7\text{W}/\text{cm}^2$ and tuned to 777.9nm (in the vicinity of the two-photon resonance for the $5\text{S}-5^2\text{D}$ transition). The resolution of the spectrometer is 0.15nm at wavelength of 800nm .

in the cell.

This experiment can be used to find the dispersion-less wavelength between two transitions, which can be useful for measuring the matrix elements of transitions, but as can be seen in Fig.4 the spectrum of the transmitted pulse is not narrow enough for precise measurement. Longer cell and optimized detection geometry can improve the precision to certain extent, as shown in Fig.6, the transmitted probe spectrum has a FWHM width of 0.8nm with a cell 10cm long heated to 252°C and a 0.5m spectrometer with a line CCD detector array. The pump laser is also tuned to vicinity of the two-photon resonance for the $5\text{S}-5^2\text{D}$ transition. Our observations have found that when the pump laser is tuned closer to the two-photon resonance of the $5\text{S}-5^2\text{D}$ transition, the passband also gets narrower, which indicates stronger lens effect as more portion of population is transferred from ground state to the 5^2D level. On the other hand, if the pump is tuned to one-photon resonance of the D_1 or D_2 transitions, i.e. $5\text{S}-5\text{P}_{1/2}(795\text{nm})$ or $5\text{S}-5\text{P}_{3/2}(780\text{nm})$, the shape of passband may get distorted and the peak wavelength (dispersion-less wavelength) will also shift since the population difference between $5\text{P}_{1/2}$ and $5\text{P}_{3/2}$ levels resulting from the population transfer by the pump is significant enough to shift the dispersion curve.

The dispersion-less wavelength has useful implications for the study of coherence and collective effects in field propagation. Since at these wavelengths self focusing is negligible and dispersion is uniform, these points could be useful for collective experiments. For this reason, studying the propagation of intense pulses near the dispersion-less wavelength is important.

V. CONCLUSION

We have demonstrated that strong pumping of a long vapor cell, creates a differential in the refractive index that will act as a gradient refractive index lens. Then when counter propagating a broadband probe field through the cell, only the portion of the probe near the dispersion-less wavelength will freely propagate, while the rest of the frequencies will be refracted away from the detector. This technique could be used in other atomic vapors to experimentally verify the frequency of zeros in the resonant refractive index.

These dispersion-less wavelengths are important for observing collective effects in long cells. Therefore the dispersion-less wavelength may be useful for coherent col-

lective control experiments in the future to avoid issues with dispersion.

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- [1] B. Arora, M. S. Safronova, and C. W. Clark, *Physical Review A* **84**, 043401 (2011).
 - [2] L. J. LeBlanc and J. H. Thywissen, *Physical Review A* **75**, 053612 (2007).
 - [3] C. D. Herold, V. D. Vaidya, X. Li, S. L. Rolston, J. V. Porto, and M. S. Safronova, *Physical Review Letters* **109**, 243003 (2012).
 - [4] N. Lundblad, M. Schlosser, and J. V. Porto, *Physical Review A* **81**, 031611 (2010).
 - [5] A. A. Svidzinsky, L. Q. Yuan, and M. O. Scully, *Physical Review X* **3**, 041001 (2013).
 - [6] P. R. Hemmer, N. P. Bigelow, D. P. Katz, M. S. Shahriar, L. DeSalvo, and R. Bonifacio, *Physical Review Letters* **77**, 1468 (1996).
 - [7] J. Keaveney, A. Sargsyan, U. Krohn, I. G. Hughes, D. Sarkisyan, and C. S. Adams, *Physical Review Letters* **108**, 173601 (2012).
 - [8] J. Keaveney, I. G. Hughes, A. Sargsyan, D. Sarkisyan, and C. S. Adams, *Physical Review Letters* **109**, 233001 (2012).
 - [9] R. H. Leonard, A. J. Fallon, C. A. Sackett, and M. S. Safronova, *Physical Review A* **92**, 052501 (2015).
 - [10] C. O'Brien, P. M. Anisimov, Y. Rostovtsev, and O. Kocharovskaya, *Physical Review A* **84**, 063835 (2011).
 - [11] B. W. Shore, *The theory of coherent atomic excitation* (Wiley, New York, 1990).
 - [12] M. S. Safronova and U. I. Safronova, *Physical Review A* **83**, 052508 (2011).
 - [13] C. O'Brien and M. O. Scully, *Journal of Modern Optics* **63**, 27 (2016).
 - [14] A. Sharma, D. V. Kumar, and A. K. Ghatak, *Applied Optics* **21**, 984 (1982).
 - [15] A. F. Molisch and B. P. Oehry, *Radiation trapping in atomic vapours* (Clarendon Press, New York, 1998).