Suppression of inhomogeneous broadening using the ac Stark shift

D. D. Yavuz, N. R. Brewer, J. A. Miles, and Z. J. Simmons

Department of Physics, University of Wisconsin at Madison, 1150 University Avenue, Madison, Wisconsin 53706, USA

(Received 30 August 2013; published 20 December 2013)

We analyze a general approach for suppressing inhomogeneous broadening of atomic transitions and thereby increasing the strength of the interaction between the atomic system and near-resonant light. The key idea is to compensate for the frequency shift due to the broadening process by using an intense laser to produce an equal and opposite Stark shift.

DOI: 10.1103/PhysRevA.88.063836 PACS number(s): 42.50.Gy, 32.60.+i, 32.70.Jz

Inhomogeneous broadening is ubiquitous in light-matter interactions [1]. When a laser beam interacts with a gas, the dominant broadening is typically due to the Doppler effect, which causes atoms to experience different laser frequencies depending on their velocities. For atoms embedded in crystals, the inhomogeneous broadening is a result of the shifts in the energy levels due to local variations in the crystal field [2]. Inhomogeneous broadening is not necessarily an undesired effect; many physical processes require an inhomogeneously broadened atomic ensemble. For example, by using an appropriate sequence of excitation pulses, photon echoes utilize the rephasing of different oscillation frequencies in an inhomogeneously broadened ensemble. Optical memories that use spectral hole burning rely on choosing a particular subset of a broadened ensemble using various frequency-selective spectroscopic techniques. The ratio of the inhomogeneous linewidth to the homogeneous linewidth determines the capacity of such memory, and this ratio is typically referred to as the figure of merit for such devices [3].

There are also many physical effects where inhomogeneous broadening is quite detrimental. For example, the magnitudes of the linear and nonlinear susceptibilities of an atomic medium typically scale as the inverse of the total linewidth of a given transition. Thus, if the inhomogeneous linewidth is larger, then the ensemble-averaged optical response is weaker. In the simplest case of a laser beam interacting with a two-level atomic system, the inhomogeneous broadening reduces the optical depth and the maximum refractive index that can be achieved. In experiments that rely on quantum interference, such as electromagnetically induced transparency (EIT), inhomogeneous broadening puts stringent constraints on the collinearity of the interacting lasers, severely limiting the nonlinearities that can be obtained at single-photon energies [4,5]. In quantum computing, Doppler broadening is a key limitation on the speed and fidelity of single- and two-qubit gates [6,7].

About three decades ago, Cohen-Tannoudji and colleagues discussed and experimentally demonstrated how Doppler broadening can be suppressed by the appropriate use of the light (Stark) shift, in the emission spectrum of an excited level [8–10]. The key idea is to compensate the frequency shifts due to Doppler effect by using an intense laser to provide an equal and opposite Stark shift. In this paper, we extend this idea to the excitation processes from the ground level. We investigate the feasibility of this approach by performing numerical simulations of the density matrix under realistic experimental conditions. Our results show almost complete suppression of Doppler broadening and, as a result, this approach may provide a powerful tool in a wide range of experiments ranging from nonlinear optics to quantum computing.

Before proceeding, we cite other related prior work. Agarwal and colleagues discussed sub-Doppler line shapes in inhomogeneously broadened media under the conditions of EIT [11]. Popov *et al.* investigated suppression of inhomogeneous broadening using coherent fields for enhanced four-wave mixing [12]. Kaplan and co-workers demonstrated the suppression of inhomogeneous broadening in rf spectroscopy of optically trapped atoms using a compensating laser beam [13]. We also would like to clearly differentiate our approach from sublinewidth spectroscopic techniques such as spectral hole burning and saturated absorption spectroscopy. These techniques rely on selecting a particular class of atoms in an inhomogeneously broadened ensemble. For example, in saturated absorption spectroscopy, an intense saturating beam is used to select a particular velocity class of atoms and saturate the transition of this smaller group. Only the atoms in the specific velocity class contribute to the optical response. In contrast, the approach that we discuss below eliminates inhomogeneous broadening and forces all the atoms in the ensemble to respond in a similar way. This is achieved with the use of an appropriately tuned Stark-shift laser so that Doppler shift is canceled by the Stark shift. Because the Doppler shift is effectively absent, all the atoms in the ensemble respond to the probe laser as if they are at zero velocity. Thus, in terms of their response to the probe laser beam, the atoms in a hot vapor cell act as though they were an ultracold ensemble.

For concreteness, we will focus on the specific example of Doppler broadening, although this idea can be extended to other inhomogeneous processes. Following Ref. [8] and as shown in Fig. 1, we consider the interaction of a four-level atomic system with two laser beams, a weak probe laser (E_P) and an intense beam that is used to Stark shift the ground level (E_S). The probe laser is tuned close to the $|1\rangle \to |2\rangle$ transition. Due to atomic motion, this transition is Doppler broadened, and we will be interested in the limit where the Doppler width is much larger than the homogeneous linewidth of the transition. To suppress this broadening, we will utilize the Stark-shift laser, which couples the ground level to two other excited levels, $|3\rangle$ and $|4\rangle$ [14]. We define the Rabi frequencies of the Stark-shift beam for the $|1\rangle \to |3\rangle$ and $|1\rangle \to |4\rangle$ transitions to be $\Omega_{13} \equiv E_S\mu_{13}/\hbar$ and $\Omega_{14} \equiv E_S\mu_{14}/\hbar$, respectively. Here the quantities μ_{13} and μ_{14} are the dipole matrix elements of the respective transitions. With these definitions, the Stark shift of

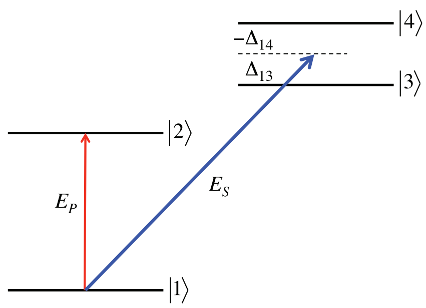

FIG. 1. (Color online) The energy level scheme for suppressing Doppler broadening of the $|1\rangle \rightarrow |2\rangle$ transition. For an atom at rest, the Stark-shift laser is tuned exactly to the middle of the two transitions, $\Delta_{13} = -\Delta_{14} \equiv (\omega_4 - \omega_3)/2$. For an atom moving at velocity v in the direction of laser propagation, the frequencies of the probe (E_P) and the Stark-shift (E_S) lasers are Doppler shifted by $k_P v$ and $k_S v$, respectively. Due to the Doppler shift of E_S, the two detunings, Δ_{13} and Δ_{14}, are modified, which in turn changes the Stark shift of the ground level $|1\rangle$. If the change in the Stark shift of the ground level exactly compensates for the Doppler shift of the probe laser beam, then Doppler broadening is suppressed.

the ground level $|1\rangle$ is

$$\delta_{\text{Stark shift}} = \frac{|\Omega_{13}|^2}{4\Delta_{13}} + \frac{|\Omega_{14}|^2}{4\Delta_{14}}, \tag{1}$$

where the detunings from the respective transitions are defined as $\Delta_{13} \equiv \omega_S - (\omega_3 - \omega_1)$ and $\Delta_{14} \equiv \omega_S - (\omega_4 - \omega_1)$. Equation (1) is valid in the perturbative limit in which most of the population remains in the ground level $|1\rangle$. For simplicity, in the remainder of this paper, we will take the dipole matrix elements of the $|1\rangle \rightarrow |3\rangle$ and $|1\rangle \rightarrow |4\rangle$ transitions to be the same, $\mu_{13} = \mu_{14}$, and therefore take the Rabi frequencies to be equal, $\Omega_{13} = \Omega_{14} \equiv \Omega_S$. We will choose the frequency of the Stark-shift laser such that for an atom at rest, this laser is tuned exactly to the middle of the two transitions, $\Delta_{13} = -\Delta_{14} \equiv \Delta_S$. As a result, for an atom at rest, the Stark shifts of the ground level due to the $|1\rangle \rightarrow |3\rangle$ and $|1\rangle \rightarrow |4\rangle$ transitions are equal in magnitude and opposite in direction, which results in $\delta_{\text{Stark shift}} = 0$. For an atom moving at a velocity v along the propagation direction of the probe laser, the frequency of the probe beam will be Doppler shifted by $k_P v$, where $k_P = \omega_P/c$. The goal is to compensate for this frequency shift by using the Stark shift of the ground level. This eliminates inhomogeneous broadening by ensuring that the probe laser detuning from the $|1\rangle \rightarrow |2\rangle$ transition, $\delta_{12} \equiv \omega_P - (\omega_2 - \omega_1)$, does not depend on the velocity of the atom. We take the propagation direction of the Stark-shift laser beam to be the same as the probe beam (copropagating). For an atom with velocity v, the Doppler shift of E_S is $k_S v$, which modifies the detunings $\Delta_{13} \rightarrow \Delta_S + k_S v$ and $\Delta_{14} \rightarrow -\Delta_S + k_S v$. As a result of the change in the detunings, the Stark shift of the ground level is

$$\begin{aligned}\delta_{\text{Stark shift}} &= \frac{|\Omega_S|^2}{4(\Delta_S + k_S v)} + \frac{|\Omega_S|^2}{4(-\Delta_S + k_S v)} \\ &\approx -\frac{|\Omega_S|^2}{2\Delta_S^2} k_S v,\end{aligned} \tag{2}$$

where in the second line we have assumed $|k_S v| \ll \Delta_S$. If the Stark shift exactly compensates for the Doppler shift $\delta_{\text{Stark shift}} = -k_P v$, then the detuning of the probe laser beam from the $|1\rangle \rightarrow |2\rangle$ transition δ_{12} remains unchanged. As a result, regardless of the velocity of the atom, the interaction of the atom with the probe laser would remain the same. Appropriate compensation requires that

$$\frac{k_P}{k_S} = \frac{|\Omega_S|^2}{2\Delta_S^2}. \tag{3}$$

Equation (3) shows that for an appropriately chosen Stark-shift beam, the Doppler shift of the probe laser can be exactly compensated for by the Stark shift of the ground level. As mentioned above, Eq. (3) is valid in the ideal perturbative limit in which most of the population is assumed to remain in the ground level $|1\rangle$. A realistic evaluation of this approach requires numerical calculations due to two key reasons: (i) From Eq. (3), for $k_P \sim k_S$, we would require $\Omega_S \sim \Delta_S$. As a result, there would be substantial population transfer to levels $|3\rangle$ and $|4\rangle$ and the perturbative approximation would break down. (ii) The analytical treatment above neglects many effects such as the power broadening of the $|1\rangle \rightarrow |2\rangle$ transition due to the intense Stark-shift laser.

We next proceed with numerical calculations in which we solve the density matrix for the four-level atomic system of Fig. 1 interacting with E_P and E_S. Within the rotating-wave approximation and using the interaction picture, the Hamiltonian that describes the atom-light interaction is

$$\hat{H} = -\frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_P & \Omega_{13} & \Omega_{14} \\ \Omega_P^* & 2\delta_{12} & 0 & 0 \\ \Omega_{13}^* & 0 & 2\Delta_{13} & 0 \\ \Omega_{14}^* & 0 & 0 & 2\Delta_{14} \end{bmatrix}. \tag{4}$$

Here Ω_P is the Rabi frequency of the probe laser beam and all the detunings are defined to be positive when the laser is tuned above the respective transition frequency. Using the Hamiltonian of Eq. (4), we form the evolution equations for the 4×4 density matrix using $i\hbar(d\hat{\rho}/dt + \frac{1}{2}\{\hat{\Gamma},\hat{\rho}\}) = [\hat{H},\hat{\rho}]$. In our numerical simulations, we consider parameters similar to D line excitations in alkali-metal atoms. For simplicity, we take the decay rates of the three excited levels to be equal, $\Gamma_2 = \Gamma_3 = \Gamma_4 \equiv \Gamma$, and we take $\Gamma = 2\pi \times 5$ MHz. We take the Rabi frequency of the probe laser beam to be $\Omega_P = \Gamma/100$. With these parameters, we numerically integrate the density matrix equations with the initial condition that all atoms are in the ground state, $\rho_{11}(t=0) = 1$. The inset in Fig. 2 shows the coherence of the $|1\rangle \rightarrow |2\rangle$ transition $|\rho_{12}|$ as the frequency of the probe laser is scanned across the resonance, in the absence of any inhomogeneous broadening and with the Stark-shift beam turned off ($\Omega_S = 0$). This coherence would result in the linear susceptibility of the medium and would cause absorption and phase shift (refractive index) of the probe laser. As expected, a Lorentzian line shape with a width of Γ and a peak (on-resonance) value of $\Omega_P/\Gamma = 0.01$ is formed.

We next introduce an inhomogeneous broadening to the system by assuming Doppler broadening with a width of $\Delta\omega_{\text{Doppler}} = 2\pi \times 250$ MHz, which is much larger than the homogeneous linewidth of $\Gamma = 2\pi \times 5$ MHz. The dashed blue

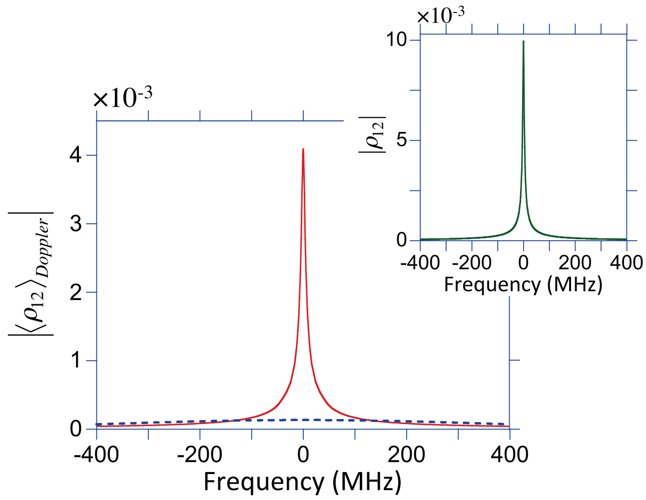

FIG. 2. (Color online) The Doppler-averaged coherence $|\langle\rho_{12}\rangle_{\rm Doppler}|$ as the frequency of the probe laser is scanned across the resonance, with (solid red line) and without (dashed blue line) the Stark-shift laser. Without the Stark-shift laser, the line is Doppler broadened with a peak (on-resonance) coherence of 1.25×10^{-4}. With the Stark-shift laser, the line shape is almost restored to its original homogeneous width with a peak coherence of 4.1×10^{-3}. For comparison, the inset shows the homogeneous Lorentzian line shape with the Stark-shift laser turned off ($\Omega_S = 0$).

line in Fig. 2 shows the calculated Doppler-averaged line shape $|\langle\rho_{12}\rangle_{\rm Doppler}|$, again with the Stark-shift beam turned off. As expected, the line shape is about two orders of magnitude broader and the on-resonance value of the coherence is reduced to 1.25×10^{-4}.

We next investigate the suppression of the Doppler broadening using the Stark-shift laser. We take $k_S = 4k_P$, which corresponds to the wavelength of the Stark-shift laser being four times smaller than the wavelength of the probe beam. We take the detuning of the Stark-shift laser from levels $|3\rangle$ and $|4\rangle$ to be $\Delta_S = 1000\Gamma$. From Eq. (3), ideal suppression requires $\Omega_S = \Delta_S/\sqrt{2} = 707\Gamma$. However, we numerically find that we get the best suppression for $\Omega_S = 810\Gamma$. This discrepancy ($\Omega_S = 810\Gamma$ vs $\Omega_S = 707\Gamma$) is likely due to the breakdown of the perturbative approximation. With these parameters, we numerically integrate the density matrix equations for atoms in different velocity classes and calculate the Doppler-averaged coherence. The solid red line in Fig. 2 shows the line shape in the presence of the Stark-shift beam as the frequency of the probe laser is scanned across the resonance. The inhomogeneous broadening is mostly suppressed with the coherence reaching an on-resonance value of 4.1×10^{-3}, only a factor of 2.5 lower than the peak value of the homogeneous line.

Figure 3 shows the numerically calculated peak (on-resonance) value of the Doppler-averaged coherence, as the Rabi frequency of the Stark-shift beam is increased. The largest suppression, and therefore the highest coherence, is obtained for $\Omega_S = 810\Gamma$, which was the Rabi frequency used in the numerical simulation of Fig. 2. For $\Omega_S < 810\Gamma$, the Stark shift of the ground level only partially compensates for the Doppler shift of the probe laser, resulting in less effective suppression. For $\Omega_S > 810\Gamma$, the Stark shift overcompensates

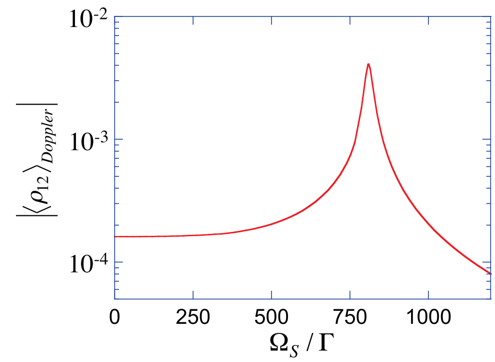

FIG. 3. (Color online) The peak (on-resonance) value of the Doppler-averaged coherence as the Rabi frequency of the Stark-shift laser is increased. The largest suppression and therefore the highest coherence is obtained for $\Omega_S = 810\Gamma$. For comparison, the peak coherence of the homogeneous line (the inset in Fig. 2) is 10^{-2}.

for the Doppler shift, which again results in reduced values of the established coherence. As shown in the inset of Fig. 2, the on-resonance value of the coherence for the homogeneous line is 10^{-2}.

As mentioned above, the numerical simulations of Figs. 2 and 3 assume $k_S = 4k_P$. In Fig. 4, we investigate the effectiveness of our scheme as the ratio k_S/k_P is varied. Here, for each value of k_S/k_P, we numerically find the optimal value for the Stark-shift laser Rabi frequency Ω_S that results in the most effective suppression. We then numerically calculate the Doppler-broadened line shape and record the peak (on-resonance) value of the Doppler-averaged coherence. For small values of k_S/k_P, satisfying Eq. (3) requires a very high laser intensity which causes large population transfer from the ground level. As a result, the suppression of Doppler broadening is not very effective, resulting in a low value of Doppler-averaged coherence. However, for $k_S/k_P > 3$, the scheme works effectively, resulting in an on-resonance value of $\approx 4 \times 10^{-3}$.

For experimental demonstration of our approach, a number of technical challenges will have to be met: (i) The atomic system must have appropriate transitions with sufficiently strong dipole matrix elements. Strong dipole matrix elements are required in order to keep the power requirements of the

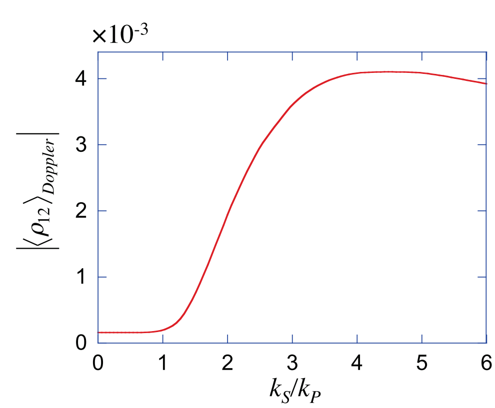

FIG. 4. (Color online) The peak (on-resonance) value of the Doppler-averaged coherence as the ratio k_S/k_P is varied.

Stark-shift laser reasonable. (ii) As shown in Fig. 4, the transitions must be such that $k_S/k_P > 3$ for effective suppression, i.e., the Stark-shift laser frequency must be at least about three times larger than the probe laser frequency. We have found that these requirements can be satisfied in a number of experimental systems. For example, in metastable helium, we have identified the following level structure: $1s2s\,^3S_1$ (ground level $|1\rangle$), $1s2p\,^3P_0$ (level $|2\rangle$), $1s3p\,^3P_2$ (level $|3\rangle$), and $1s3p\,^3P_0$ (level $|4\rangle$). For this scheme, the probe and the Stark-shift lasers are at wavelengths of 1082.9 and 388.9 nm, respectively. The frequency spacing between levels $|3\rangle$ and $4\rangle$ is 8.8 GHz, which is very similar to what we have assumed in the numerical simulations of Figs. 2–4. A detailed numerical study of our approach in various experimental systems, including atomic gases and rare-earth doped crystals, will be among our future investigations.

We next discuss a number of technical requirements on the laser beams. (i) *Collinearity:* For the scheme to work effectively, the angular spread of the k vectors must be small compared to the ratio of the homogeneous linewidth to the Doppler linewidth. For the numerical calculations of Figs. 2 and 3, this translates into a beam divergence angle of less than 10 mrad, which would correspond to beam sizes larger than 100 μm if the probe and Stark-shift lasers are in the visible to near-infrared regions of the spectrum. (ii) *Stark-shift laser power:* If we assume a matrix element of 1 atomic unit, the numerical simulation of Fig. 2 would require a Stark-shift laser intensity of 15 kW/cm^2. Since the beam size must be larger than about 100 μm, these intensities would require a laser with a power exceeding 1.5 W. This number depends critically on the frequency separation of levels $|3\rangle$ and $|4\rangle$. For the numerical calculations of Figs. 2–4, this separation is assumed to be $2\Delta_S = 2000\Gamma = 2\pi \times 10$ GHz. Larger separations would translate into a higher-power requirement for the Stark-shift laser. (iii) *Intensity stability of the Stark-shift laser:* In our scheme, the fluctuations in the Stark shift of the ground level must be small compared to the homogeneous linewidth. For the numerical calculations of Figs. 2–4, this translates into a fractional intensity stability of better than 2% for the Stark-shift beam.

We note that there are many experiments that would benefit from the suppression of broadening. For the numerical calculation of Fig. 2, the peak value of the coherence is increased by a factor of 33 using the suppression of the Stark-shift beam. As a result, the optical depth of the atomic medium and the corresponding refractive index would increase by a factor of 33. Recently, there has been substantial interest in achieving a high or negative refractive index in atomic systems, both in vapors and in rare-earth-doped crystals [15,16]. Suppressing the inhomogeneous broadening in these systems would considerably relax atomic density requirements. Furthermore, any nonlinear optical process that utilizes the probe beam would also benefit from the suppression of the inhomogeneous broadening. As a result, we expect possible applications in achieving high nonlinearities at the single-photon level and in photonic quantum computation [17,18].

We would like to thank Josh Weber for many helpful discussions. This work has been partially supported by the Air Force Office of Scientific Research (AFOSR).

[1] A. E. Siegman, *Lasers* (University Science Books, Sausalito, CA, 1986).
[2] R. M. Macfarlane and R. M. Shelby, J. Lumin. **36**, 179 (1987).
[3] R. M. Macfarlane, J. Lumin. **100**, 1 (2002).
[4] S. E. Harris, Phys. Today **50** (7), 36 (1997).
[5] M. O. Scully and M. S. Zubairy, *Quantum Optics* (Cambridge University Press, Cambridge, UK, 1997).
[6] M. Saffman and T. G. Walker, Phys. Rev. A **72**, 022347 (2005).
[7] I. Roos and K. Molmer, Phys. Rev. A **69**, 022321 (2004).
[8] C. Cohen-Tannoudji, F. Hoffbeck, and S. Reynaud, Opt. Commun. **27**, 71 (1978).
[9] S. Reynaud, M. Himbert, J. Dupont-Roc, H. H. Stroke, and C. Cohen-Tannoudji, Phys. Rev. Lett. **42**, 756 (1979).
[10] S. Reynaud, M. Himbert, J. Dalibard, J. Dupont-Roc, and C. Cohen-Tannoudji, Opt. Commun. **42**, 39 (1982).
[11] G. Vemuri, G. S. Agarwal, and B. D. Nageswara Rao, Phys. Rev. A **53**, 2842 (1996).
[12] A. K. Popov and A. S. Bayev, Phys. Rev. A **62**, 025801 (2000).
[13] A. Kaplan, M. F. Andersen, and N. Davidson, Phys. Rev. A **66**, 045401 (2002).
[14] There are two main reasons why the Stark-shift laser couples the ground level to two excited levels (levels $|3\rangle$ and $|4\rangle$) instead of a single excited level: (i) The use of two levels increases the effective velocity-dependent Stark shift by a factor of 2. As a result, the requirement on the ratio of the laser frequencies k_S/k_P is relaxed by the same factor. (ii) Because the Stark-shift beam is tuned to the middle of the two resonances, the Stark shift vanishes for zero velocity atoms. This avoids high sensitivity of the technique to the intensity of the Stark-shift laser. Furthermore, the suppression process becomes symmetric for negative and positive velocity classes.
[15] D. E. Sikes and D. D. Yavuz, Phys. Rev. A **84**, 053836 (2011).
[16] P. P. Orth, R. Hennig, C. H. Keitel, and J. Evers, New J. Phys. **15**, 013027 (2013).
[17] S. E. Harris and Y. Yamamoto, Phys. Rev. Lett. **81**, 3611 (1998).
[18] M. D. Lukin and A. Imamoglu, Phys. Rev. Lett. **84**, 1419 (2000).