ISSN 0030-400X, Optics and Spectroscopy, 2011, Vol. 111, No. 4, pp. 583–588. © Pleiades Publishing, Ltd., 2011.

COHERENT EFFECTS IN OPTICAL AND ATOMIC–MOLECULAR PROCESSES

Enhancement of Electromagnetically Induced Transparency in Room Temperature Alkali Metal Vapor[1]

O. S. Mishina[a], M. Scherman[a], P. Lombardi[b], J. Ortalo[a], D. Felinto[c], A. S. Sheremet[d], D. V. Kupriyanov[d], J. Laurat[a], and E. Giacobino[a]

[a] *Laboratoire Kastler Brossel, École Normale Supérieure et CNRS, Université Pierre et Marie Curie-Paris 6, 75005 Paris, France*

[b] *European Laboratory for Non-Linear Spectroscopy, University of Florence Sesto-Fiorentino (Firenze), I 50019 Italia*

[c] *Departamento de Fisica, Universidade Federal de Pernambuco, Cidade Universitria, Recife, PE, 50670-901 Brazil*

[d] *Department of Theoretical Physics, State Polytechnic University, St. Petersburg, 195251 Russia*

Received April 18, 2011

Abstract—Electromagnetically induced transparency (EIT) has led to several quantum optics effects such as lasing without inversion or squeezed light generation. More recently quantum memories based on EIT have been experimentally implemented in different systems such as alkali metal atoms. In this system the excited state of the optical transition splits into several sublevels due to the hyperfine interaction. However, most of the theoretical models used to describe the experimental results are based on a Λ-system with only one excited state. In this article, we present a theoretical model for the Λ-type interaction of two light, fields and an atomic system with multiple excited state. In particular we show that if the control and probe fields are orthogonally circularly polarized the EIT effect in an alkali-metal vapor can almost disappears. We also identify the reasons of this reduction and propose a method to recover the transparency via velocity selective optical pumping.

DOI: 10.1134/S0030400X1111021X

1. INTRODUCTION

EIT has been observed in various materials, such as alkali-metal vapors and ultracold atomic ensembles, rare–earth doped crystals, NV centers in diamonds or quantum dots [1]. In these systems, spin coupling between an optically excited electron and the nucleus gives rise to a MHz range hyperfine splitting of the excited state [2–6]. In the case of alkali-metal vapors used on the D_2-line inhomogeneous broadening at room temperature is comparable with the hyperfine splitting so the multiple excited levels can not be resolved. In this case all the levels simultaneously participate in the interaction process which makes the dynamic very different from the case of only one excited state as widely discussed in theory [1, 7].

In this work, we calculated the linear medium response to a probe field when the weak probe and strong control fields are applied to the different sides of a Λ-system with multiple excited states. In Section 2 we will demonstrate how the hyperfine splitting modifies the dressed states of the system in the presence of the strong control field. In Section 3 we will show that it leads to the vanishing of the EIT in an alkali metal vapor where inhomogeneous broadening is comparable with the hyperfine splitting. In Section 4 we propose a method to recover the EIT in an alkali metal vapor using a velocity dependent optical pumping. Section 5 concludes the paper.

[1] The article is published in the original.

2. SUSCEPTIBILITY OF A Λ-SYSTEM WITH SEVERAL EXCITED STATES

To study the Raman–type interaction of the two fields with a multilevel system we will use a standard semiclassical approach. A strong "control" field drives the transitions between one of the two ground states called $|s\rangle$ and three excited states $|1\rangle$, $|2\rangle$, $|3\rangle$ as presented on the level scheme in the inset (a) of Fig. 1. The weak "probe" field drives the transitions between the second ground state $|g\rangle$ and the excited states. The interaction of the probe field with a multilevel system is treated with the use of a density matrix approach. The evolution of the system is described by the following Hamiltonian:

$$\begin{gathered} H = H_0 + V = H_{\text{field}} + H_{\text{atom}} + V, \\ V = V_p + V_c, \\ V_p = -\left(d_{1g}|1\rangle\langle g| + d_{2g}|2\rangle\langle g| + d_{3g}|3\rangle\langle g|\right) E_p^{(+)} + \text{h.c.}, \\ V_c = -\left(d_{1s}|1\rangle\langle s| + d_{2s}|2\rangle\langle s| + d_{3s}|3\rangle\langle s|\right) E_c^{(+)} + \text{h.c.} \end{gathered} \tag{1}$$

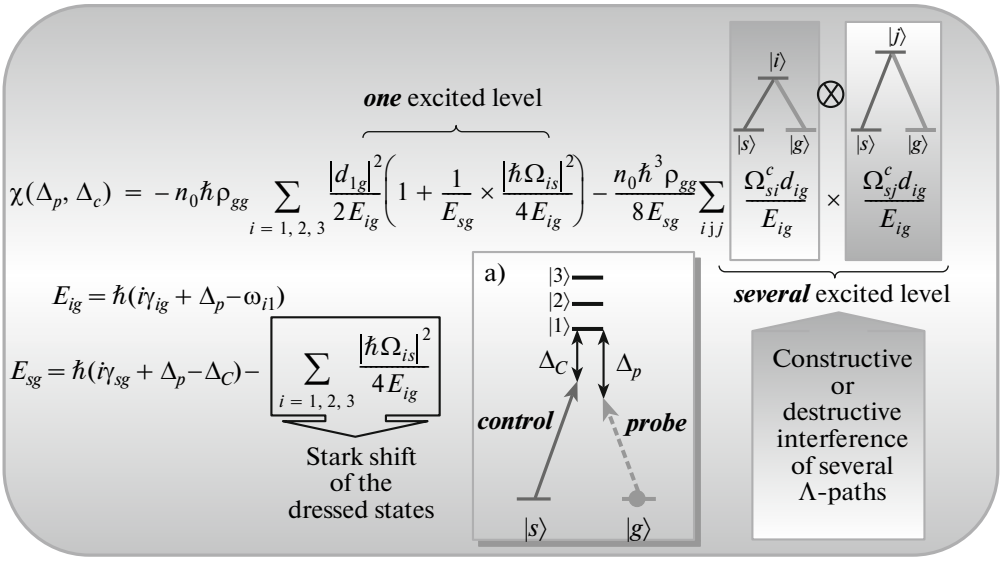

Fig. 1. (Color online) Linear susceptibility $\chi(\Delta_p, \Delta_c)$ of a Λ -system with multiple excited state (3). The level–scheme is presented in the inset (a). The first sum in the susceptibility formula shows the independent impacts of each Λ-path $|s\rangle \leftrightarrow |i\rangle \leftrightarrow |g\rangle$ ($i = 1,2,3$) taken separately. The second sum reveals the interference between the different Λ -paths (gradient color boxes indicate Λ-paths involving the different excited states $|i\rangle \neq |j\rangle$). The signs of the dipole moment matrix elements products $d_{si} \times d_{ig}$ together with the energy denominators E_{ig} (4) containing the one–photon detuning Δ_p define whether the interference is constructive or destructive. The energy denominator E_{sg} (4) presents in both terms of the susceptibility and defines the poles of the susceptibility function. The sum subtracted from the two-photon detuning $\Delta_p - \Delta_c$ in the E_{sg} expression represents the Stark shifts of a multilevel atom coupled to the strong control field. This will define the distance of the induced Raman absorption resonance in a multilevel system from the two-photon resonance.

The Hamiltonian H_0 is given by the sum of the free Hamiltonian operators of the electromagnetic field H_{field} and of the atom H_{atom}. The interaction Hamiltonian V is written in the rotating wave approximation and consists in the dipole interactions between the atom and the control and probe fields. In the interaction representation, the positive frequency components of the electromagnetic field for the control and probe modes are $E_c^{(+)} = \varepsilon_c e^{-i\omega_c t}$ and $E_p^{(+)} = \varepsilon_p e^{-i\omega_p t}$ respectively. d_{ij} are the matrix elements of the electric dipole moment of the atom.

The linear response of the atom to the weak probe field is described by the polarizability $\alpha(\Delta_c, \Delta_p)$ defined as

$$\alpha\varepsilon_p = d_{g1}\sigma_{1g}^{(1)} + d_{g2}\sigma_{2g}^{(1)} + d_{g3}\sigma_{3g}^{(1)}, \qquad (2)$$

where $\sigma_{ig}^{(1)}$ are the slowly varying amplitudes of the density matrix elements $\rho_{ig}^{(1)}$ found in the first order with respect to the probe field. They represent the optical coherences between the ground state $|g\rangle$ and the excited states $|i\rangle$ ($i = 1,2,3$). In the case of a dilute medium, when the number of particles in a volume of a cubic wavelength is small related to one, the complex susceptibility is directly expressed by the single atom polarizability $\chi(\Delta_c, \Delta_p) = n_0\alpha(\Delta_c, \Delta_p)$, with n_0 the atomic density, $\Delta_c = \omega_c - \omega_{1s}$ the control field detuning and $\Delta_p = \omega_p - \omega_{1g}$ the probe field detuning.

We find a first order solution for the slow varying optical coherences $\sigma_{ig}^{(1)}$ in the steady state regime from the master equations driven by the Hamiltonian (1) which leads to the following expression for a linear susceptibility:

$$\chi(\Delta_c, \Delta_p) = -n_0\hbar\rho_{gg}^{(0)}\left[\sum_i \frac{|d_{ig}|^2}{2E_{ig}}\left(1 + \frac{|\hbar\Omega_{is}^c|^2}{4E_{sg}E_{ig}}\right) + \frac{\hbar^2}{8E_{sg}}\sum_{i\neq j}\frac{d_{gi}\Omega_{is}^c \times d_{jg}\Omega_{sj}^c}{E_{ig}E_{jg}}\right]. \qquad (3)$$

This expression contains the Rabi frequencies of the control field $\Omega_{is}^c = 2d_{is}\varepsilon_c/\hbar$. The energy denominators E_{ig} and E_{sg} have the following form

$$E_{ig} = \hbar(i\gamma_{ig} + \Delta_p - \omega_{i1}),$$
$$E_{sg} = \hbar(i\gamma_{sg} + \Delta_p - \Delta_c) - \sum_i \frac{|\hbar\Omega_{si}^c|^2}{4E_{ig}}, \qquad (4)$$

where $\omega_{i1} = (E_i - E_1)/\hbar$ with $i = 1, 2, 3$ are the frequency separations between the excited states. The optical coherence relaxation rates are taken to be half the natural relaxation rate of an excited atom $\gamma_{eg} \approx \gamma/2$. Assuming that the ground state relaxation mechanism is the atom loss due to the finite size of the beam the decay rate of the ground state coherence is taken to be equal to $\gamma_{sg} = \gamma 10^{-4}$. It will be thus much slower than the excited state decay in all the following calculations.

Figure 1 presents expressions (3), (4) and comments the physical meaning of the different terms. The multiplicity of the excited state induces two important modifications in the susceptibility expression as compared to the case of a three-level Λ-type system. First, a new term describing the interference between the several possible Λ-transitions appears. We mark it by the two boxes containing Λ-schemes with different excited states. This interference can be destructive or constructive depending on the ratio between the dipole moments products $d_{gi}d_{is}$ and $d_{gj}d_{js}$ for $i \neq j$ and the detunings of the probe and control fields. For example in case of an alkali metal with right-handed circular polarized control and left-handed circular polarized probe fields, the interference is constructive when the fields are tuned between the excited states and it is destructive when the fields are blue or red detuned from the D_1 transition. This was shown in the reference [8] on the example of an ultracold ensemble of cesium atoms.

The second main difference is in the energy denominator term E_{sg}. It gives the poles of the susceptibility function corresponding to the positions of the absorption maxima, i.e. the positions of the dressed states created by interaction with the strong control field. In case of only one excited state the Stark shift of the dressed states is given by the first term in the sum of the energy denominator E_{sg} (see Fig. 1). In the case of more excited sublevels more Stark shifts are added into the sum. This complicates the problem of finding the exact position of the induced Raman absorption resonance as well as the EIT maximums. In reference [10] we derived analytically the position for both absorption and EIT resonances and analyzed it in details. Here we will skip this derivation and turn directly to an understanding of the EIT formation in an inhomogeneous broadened multilevel medium.

3. EIT IN AN ALKALI METAL VAPOR: JOINT EFFECT OF HYPERFINE SPLITTING AND DOPPLER BROADENING

Motivated by the fact that Doppler broadening at room temperature often has the same order of magnitude as the hyperfine splitting, we are interested in addressing the effect of both at the same time. To demonstrate this effect we chose for example an alkali-metal vapor (here ^{133}Cs). We suppose that atoms are optically pumped to the extreme Zeeman sublevel $|g\rangle \equiv |F = 3, m_F = 3\rangle$, of the ground state $6^2S_{1/2}$. The second ground Zeeman sublevel participating in the Λ-interaction is $|s\rangle \equiv |F = 3, m_F = 1\rangle$. The strong right-handed circularly polarized (σ^+) control field couples the latter with the excited states of the D_2 optical transition $6^2S_{1/2} \leftrightarrow 6^2P_{3/2}$. They are $|1\rangle \equiv |F' = 2, m_{F'} = 2\rangle$, $|2\rangle \equiv |F' = 3, m_{F'} = 2\rangle$ and $|3\rangle \equiv |F' = 4, m_{F'} = 2\rangle$. The left-handed circularly polarized (σ^-) probe field will be scanned along the transitions $|g\rangle \leftrightarrow |1\rangle$, $|g\rangle \leftrightarrow |2\rangle$ and $|g\rangle \leftrightarrow |3\rangle$.

Figure 2 shows the probe field transmission as a function of the probe field detuning Δ_p for different temperatures T of the atomic vapor. We assume here $\Delta_c = 0$, i.e. the control field is resonant to the $|s\rangle \leftrightarrow |1\rangle$ transition. To perform this calculation, we integrate the susceptibility (3) over a gaussian Doppler distribution $f(\Delta_D) = (2\pi\Gamma_D^2)^{-1/2} \exp(-\Delta_D^2/2\Gamma_D^2)$ where Δ_D is the Doppler shift and $\Gamma_D = \sqrt{k_B T/m\lambda^2}$ the half-linewidth:

$$\overline{\chi}(\Delta_c, \Delta_p) = \int \chi(\Delta_c + \Delta_D, \Delta_p + \Delta_D) f(\Delta_D) d\Delta_D. \quad (5)$$

The probe transmission through the medium of the length L is calculated from the Beer's law

$$|\varepsilon_p(L)|^2/|\varepsilon_p(0)|^2 = \exp\left[-4\pi k_p L \mathrm{Im}[\overline{\chi}(\Delta_c, \Delta_p)]\right]. \quad (6)$$

In Fig. 2, the probe transmission is given for a cesium vapor at different temperature and the same atomic density $n_0 = 2.1 \times 10^{12}$ cm^{-3}. For comparison we show the calculations for the three-level (dashed, red line) and the five-level (solid, blue line) models. When the temperature is $T = 1$ K (see Fig. 2a), which results in a Doppler broadening $\Gamma_D = 10$ MHz much smaller than the splitting between the nearest hyperfine excited states $\omega_{|1\rangle \leftrightarrow |2\rangle} = 150$ MHz, it can be clearly seen, that the EIT resonance appears in both three-level and five-level systems. This is in a good agrement with the experimental observations of EIT in a ensemble of ultracold cesium atoms despite the complicated multilevel structure [11]. However, there is already a significant, difference between the predictions of the three-level and five–level models. First, the transmittance is reduced in the multilevel configuration. This reduction can be explained by the decay of the ground state coherence σ_{sg} introduced by the control field exciting off-resonantly the levels $|2\rangle$ and $|3\rangle$ Second, the EIT resonance is shifted from the two photon resonance $\Delta_p = 0$. This light shift is also caused by the presence

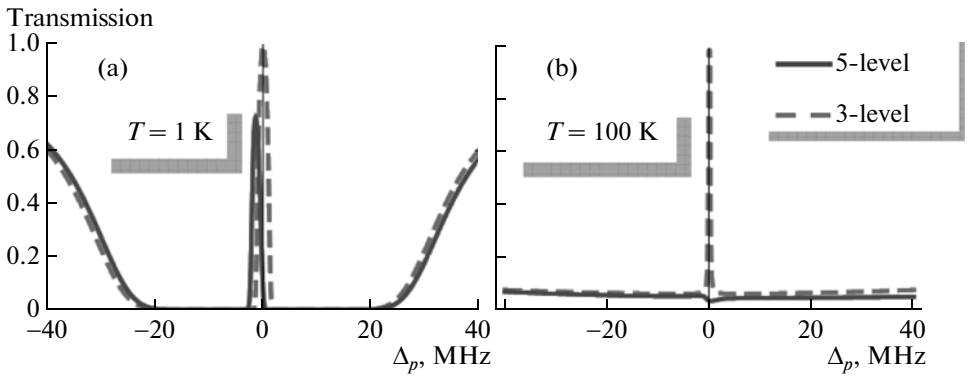

Fig. 2. (Color online) Probe transmission.in an alkali-metal vapor at different temperature. In a Doppler broadened D_2-line of ^{133}Cs vapor with the density $n_0 = 2.1\times10^{12}$ cm^{-3} we can see a high-contrast EIT resonance for a sample temperature $T = 1$ K. This is due to the fact that the Doppler broadening $\Gamma_D = 10$ MHz is smaller than the hyperfine splitting $\omega_{21} = 150$ MHz (*blue smooth curve* of plot (a)). If the Doppler broadening $\Gamma_D = 100$ MHz is close to the hyperfine splitting, the EIT peak disappears as shown on the *blue smooth* curve of plot (b) for the temperature $T = 100$ K. In the three-level Λ-system the EIT peak would remain and only get narrower at higher temperature as presented on the *red dashed* curves. The reduction of the EIT in the five-level system is caused by the atoms whose Raman induced absorbtion peaks are placed in the middle of the EIT window of the atoms from other velocity classes. In the presented calculations the control field detuning is $\Delta_c = 0$ and the Rabi frequency is $\Omega_{1s}^c = 2.3\gamma = 24\pi$ MHz.

of the excited states $|2\rangle$ and $|3\rangle$. Both effects were observed in [3] and studied in details in the [8, 12]. The analytical expression for the exact position of the EIT resonance in the multilevel system is derived in reference [10].

When the temperature of the medium is $T = 100$ K (see Fig. 2b), the Doppler broadening $\Gamma_D = 100$ MHz is getting close to the hyperfine splitting $\omega_{|1\rangle\leftrightarrow|2\rangle} = 150$ MHz. In this case the EIT is strongly reduced. As follows from the comparison between Figs. 2a and 2b, this is caused by the presence of the atoms with doppler shift $\Delta_D > 10$ MHz. Indeed the induced Raman absorbtion resonances of these atoms are shifted such as they are placed right in the EIT window of other atoms and the probe light is consequently absorbed.

The major result of this section is the demonstration that the EIT is strongly reduced in an inhomogeneously broadened ensembles of five-level atoms as compared to three-level atoms. The main reason has been clearly identified: the presence of atoms with a particular Doppler shift that absorb the probe light while the others are almost transparent. Based on this knowledge, we will discuss in the following section the possibility for the improvement of the EIT in the alkali-metal vapor.

4. ENHANCEMENT OF THE EIT VIA VELOCITY SELECTIVE OPTICAL PUMPING

In this section we propose a method to increase the EIT in a medium where inhomogeneous broadening is comparable with the hyperfine splitting of the excited state. The general idea is to remove from the interaction process the atoms which add absorbtion inside the EIT window presented on Fig. 2. To do this we propose to make these atoms with a specific Doppler shift insensitive to the probe and control fields. In the alkali metal vapor this possibility is given by the hyperfine splitting of few GHz in the ground state. We will use the second ground state, $F = 4$, as a "reservoir" for these atoms. To transfer atoms to this level we propose to use a standard optical pumping technique. By applying an additional pumping field detuned by Δ_{pump} from the transition $|F = 3\rangle \leftrightarrow |F' = 4\rangle$ we will depopulate from the level $|F = 3\rangle$ atoms whose Doppler shifts are distributed around $\Delta_D = -\Delta_{\text{pump}}$.

In Fig. 3a we present the velocity distribution calculated for atoms in the state $|g\rangle$ in the presence of an additional pumping field. We used three different values of the pump field detuning to depopulate different velocity classes. From the probe transmission on Fig. 3b we can see that EIT is enhanced when a hole is burned in a precise position of the velocity distribution: atoms from this particular velocity class absorb the probe light while the others are transparent. It is in agrement with our previous study showing that due to the multiple Stark shift in the presence of several excited states, the dressed states on these atoms will be located at the same probe frequency ω_p as the EIT of other atoms.

One more remark is needed with respect to the optical pumping process presented in this section. As well as the pump field is depopulating the sublevel

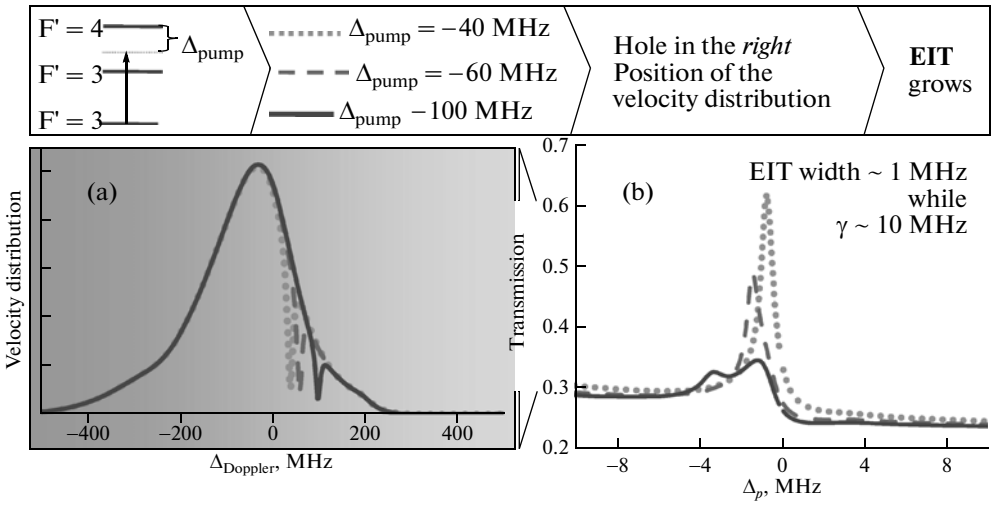

Fig. 3. (Color online) EIT enhancement in the alkali metal vapor via velocity dependent optical pumping. With an additional pumping field the "hole" can be created in the desired place of the velocity distribution of atoms in the state $|g\rangle$ as presented on (a). By depopulating the right velocity class of atoms we remove their induced Raman absorption resonances from the EIT window presented on Fig. 2a. This configuration leads to a recovery of the EIT resonance. Calculations are done with the same parameters as on Fig. 2: $\Omega_{els}^{c} = 2.3\gamma = 24\pi$ MHz and $\Delta_c = 0$.

$|F = 3\rangle$ the strong control field will do the same. So if only the control, probe and pump fields are applied to the alkali-metal vapor all the atoms will be eventually transferred into the sublevel $|F = 4\rangle$. To recover atoms in the state $|g\rangle$ we use an additional repumping field resonant to the $|F = 4\rangle \leftrightarrow |F' = 4\rangle$ transition as it was previously proposed and experimentally implemented in references [13, 14]. The full treatment of the optical pumping process in an alkali-metal vapor can be found in reference [10].

We would like to stress that the enhanced EIT peak has subnatural linewidth, showing that transparency peak presented on Fig. 2b is not due to the hole burning in the velocity distribution but indeed of the coherent action of the control and the probe fields.

5. CONCLUSIONS

In conclusion, we provide a complete theoretical, description of the stimulated Raman interaction in a Λ-system with more than one excited state. The result for the medium susceptibility makes theoretical description of the light-atom interaction closer to the experimental situation as demonstrated on the example of an alkali metal vapor. Significantly, the result can also be generalized to other systems used for quantum optics experiments where the hyperfine splitting of the excited state is comparable with inhomogeneous broadening, i.e., rare-earth doped crystals, NV-centers and quantum dots. Our work provides explanation of the EIT reduction in alkali metal vapor. The reduction of the EIT in such a system was experimentally observed [9] but did not have a proper explanation and a theoretical description heretofore. Finally, we propose a method to recover the EIT via velocity dependent optical pumping. An enhancement by a factor 5 of the EIT contrast seems possible.

ACKNOWLEDGMENTS

We thank Igor Sokolov and Alberto Amo for fruitful discussions. This work is supported by the EC under the ICT/FET project COMPAS, by the RFBR (Grants no. 10-02-00103), and by the CAPES/COFECUB project QE-COMET. O. Mishina acknowledges the financial support from the Ile-de-France programme IFRAF and A. Sheremet from the Foundation "Dynasty."

REFERENCES

1. M. Fleischhauer and J. P. Marangos, Rev. Mod. Phys. **77**, 633 (2005).
2. S. Harris, J. Field, and A. Imamoglu, Phys. Rev. Lett. **64**, 1107 (1990).
3. L. V. Hau, S. E. Harris, Z. Dutton, and C. H. Behroozi, Nature **397**, 594 (1999).
4. F. A. Beil, M. Buschbeck, G. Heinze, and T. Halfmann, Phys. Rev. A **81**, 053801 (2010).
5. R.-B. Liu, W. Yao, and L. J. Sham, arXiv:1006.5544v1 (2010).

6. A. Batalov, V. Jacques, F. Kaiser, P. Siyushev, P. Neumann, L. Rogers, R. McMurtrie, N. Manson, F. Jelezko, and J. Wrachtrup, Phys. Rev. Lett. **102**, 195506 (2009).
7. K. Hammerer, A. S. Sørensen, and E. S. Polzik, Rev. Mod. Phys. **82**, 1041 (2010).
8. A. S. Sheremet, L. V. Gerasimov, I. M. Sokolov, D. V. Kupriyanov, O. S. Mishina, E. Giacobino, and J. Laurat, Phys. Rev. A **82**, 033838 (2010).
9. K. Li, L. Deng, and M. G. Payne, Appl. Phys. Lett. **95**, 221103 (2009).
10. O. S. Mishina et al., in preparation.
11. K. S. Choi, H. Deng, J. Laurat, and H. J. Kimble, Nature **452**, 67 (2008).
12. Y. Chen, X. G. Wei, and B. S. Ham, Opt. Express **17**, 1781 (2009).
13. J. Cviklinski, J. Ortalo, J. Laurat, A. Bramati, M. Pinard, and E. Giacobino, Phys. Rev. Lett. **101**, 133601 (2008).
14. J. Ortalo, J. Cviklinski, P. Lombardi, J. Laurat, A. Bramati, M. Pinard, and E. Giacobino, J. Phys. B: At. Mol. Opt. Phys. **42**, 114010 (2009).