

Probing light forces on cold atoms by noise correlation spectroscopy

K. Theophilo, A. Kumar, H. M. Florez, C. González-Arciniegas, P. Nussenzveig, and M. Martinelli*

Instituto de Física, Universidade de São Paulo, 05315-970 São Paulo, SP-Brazil



(Received 8 August 2018; published 19 November 2018)

Enhanced sensitivity in electromagnetically induced transparency (EIT) can be obtained by the use of noise correlation spectroscopy between the fields involved in the process. Here, we investigate EIT in a cold (<1 mK) rubidium vapor and demonstrate sensitivity to detect weak light-induced forces on the atoms. A theoretical model is developed and shows good agreement with our measurements, enabling the attribution of the observed effects to the coupling of the atomic states to their motion. The effects remain unnoticed on the measurement of the mean fields but are clearly manifest in their correlations.

DOI: [10.1103/PhysRevA.98.053832](https://doi.org/10.1103/PhysRevA.98.053832)

I. INTRODUCTION

Electromagnetically induced transparency (EIT) [1] is a coherent process that has received growing attention in previous decades not only for its applications to quantum networks based on quantum memories [2,3], but also in metrology. Recent proposals have reported its use for precision measurements in atomic clocks at room temperature, using the EIT narrow linewidth to provide sensitive detection of frequency fluctuations [4]. It has also been shown that EIT is well suited for magnetometry applications [5] in particular, showing a vector magnetometer operation with angular sensitivity up to 10^{-3} deg/ \sqrt{Hz} [6]. Moreover, the use of EIT in diamonds [7] and Rydberg [8] atoms at room temperature for electrometry applications was recently demonstrated.

In spite of these metrological applications, the sensitivity of EIT to the external degrees of freedom of the atoms due to light forces has remained somewhat unexplored. Nevertheless, the subtle radiation pressure exerted by the pump and probe beams close to resonance modifies the atomic dynamics. Although this effect is negligible in the spectroscopy of hot atomic vapors, it becomes relevant for measurements in cold gases. In this paper we show that we can rely on the sensitivity of the noise correlation spectroscopy [9–11] to observe the role of the radiation pressure on the atomic dynamics, otherwise unnoticed in the average intensity signals. This extends the application of this technique from the study of internal degrees of freedom of the atom to the external ones.

EIT in a Λ transition involves two lower energy states of an atom that are coupled by a pair of fields to an excited state (Fig. 1). As a consequence of this combined interaction, the atoms are pumped into a dark state, a superposition of the ground states that decouples the atoms from the incident fields, therefore leading to a reduction of the light scattering [1]. The transmission of each field presents an increase in the intensity when the Λ resonance is satisfied, i.e., the energy difference of the incident photons matches the energy difference of the two levels with lower energy.

Atomic dynamics, as well as field fluctuations, disturb the transmitted intensities, which develop fluctuations $\delta I_i(t)$. The analysis of these fluctuations provides a useful tool for the study of atomic dynamics [12], but the measured intensity can be affected by other noise sources. Their effect can be filtered out by the observation of the Fourier component of the photocurrent $\delta I_i(\omega_a)$, and the sensitivity is increased by the use of the correlation function,

$$C(\omega_a) = \frac{S_{12}(\omega_a)}{\sqrt{S_{11}(\omega_a)S_{22}(\omega_a)}}, \text{ where} \quad (1)$$

$$S_{ij}(\omega_a) = \frac{1}{2}[\langle \delta I_i(\omega_a)\delta I_j^*(\omega_a) \rangle + \langle \delta I_i^*(\omega_a)\delta I_j(\omega_a) \rangle], \quad (2)$$

in which $S_{ii}(\omega_a)$ is the noise power of each transmitted beam and $S_{ij}(\omega_a)$ is the cross-correlation of the noise, at the analysis frequency ω_a [10]. This normalization of the correlation makes the result more robust against atomic losses in the system, in contrast to the typical measurement of the intensities of the transmitted beam or the measurement of fluorescence.

We studied an ensemble of cold ^{87}Rb atoms, investigating the coupling between Zeeman levels in the $5S_{1/2}(F=1) \rightarrow 5P_{3/2}(F'=1)$ transition (energy gap $E = \hbar\omega$), selectively addressed by cross circularly polarized laser fields with controllable detunings (Fig. 1). Field 1 (Rabi frequency Ω_1) is kept on resonance, while the frequency of the probe field (Rabi frequency Ω_2) is scanned. The one-photon detuning of each field is given by $\delta_i = (E_i - E)/\hbar$, where $E_i = \hbar\omega_i$ is the photon energy of the laser. Two-photon detuning for the Λ transition is defined as $\delta = \delta_2 - \delta_1$. To compensate for the loss of atoms by spontaneous emission into the $F=2$ level, a resonant repump field couples the transition $5S_{1/2}(F=2) \rightarrow 5P_{3/2}(F'=2)$. Any residual atomic population at the Zeeman levels with magnetic quantum number $m=0$ at the $5P_{3/2}(F'=1)$ level are rapidly pumped into levels $m=\pm 1$, $5P_{3/2}(F'=1)$ and spontaneously decay into the ground states involved in the Λ transition. Since the transition between the fields with $m=0$ is forbidden, we have an effectively pure Λ system, with a certain rate for the loss and reload of atoms that affects the coherence of the two-photon transition.

*mmartine@if.usp.br

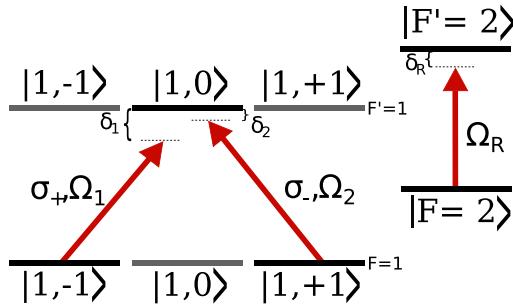


FIG. 1. Relevant levels of the ^{87}Rb atoms under study (not in scale).

II. EXPERIMENTAL SETUP

The experimental setup is presented in Fig. 2 (top) and it has been previously described in Ref. [13]. It consists of an ensemble of cold ^{87}Rb atoms in a magneto-optical trap (MOT) with nearly 10^7 atoms at $800 \mu\text{K}$. The two cross circularly polarized beams addressing the EIT transition are nearly copropagating (with an angle $\theta \sim 2^\circ$), and their frequencies and powers can be controlled by acousto-optic modulators (AOM). The repump laser is antiparallel with respect to the spectroscopy beams. To perform the correlation spectroscopy, we keep the detuning of field 1 fixed while scanning the two-photon frequency detuning $\delta' = \delta/2\pi$ of the other EIT field with respect to the atomic resonance. The power of the beams was $115 \mu\text{W}$, with spot sizes of $w \simeq 1.7 \text{ mm}$ ($I \simeq 0.2 I_{\text{sat}}$). The repump beam was kept on resonance with a power of $\sim 750 \mu\text{W}$.

The transmitted EIT beams are sent to amplified photodetectors, with a signal proportional to the mean intensity (dc) and a high gain transimpedance amplifier for high frequency components (HF). The HF signals are demodulated by an electronic oscillator at 2 MHz , and the low-pass filtered outputs ($f_c < 300 \text{ kHz}$) are acquired by an analog-to-digital converter, together with the dc signals.

The measurement cycle is shown in Fig. 2 (bottom). After a MOT loading cycle of 900 ms , the current of the trapping coils was turned off 3 ms before the measurement to avoid spurious

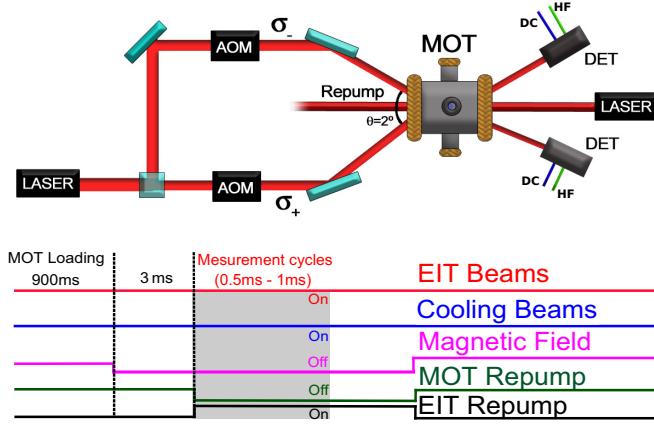


FIG. 2. (Top) Experimental setup. (Bottom) Timing sequence of our experimental cycle.

magnetic fields that could lead to decoherence or energy shifts between the Zeeman ground-state levels. The cooling beams, exciting the D2 line, could be either turned on or off during the experiment. In order to pump the atoms to the $5S_{1/2}(F=1)$ level, the repump field of the MOT was turned off and the EIT repump turned on at the beginning of the acquisition. During the measurements, the frequency detuning of one of the spectroscopy beams (δ'_2) could be left at a fixed value, or could be scanned in a 15-MHz range (2.5 times the atomic linewidth of 6 MHz) centered in the two-photon resonance, during 0.5 ms which corresponds to $d\delta'/dt = 30 \text{ MHz/ms}$. The scanning rate has an upper bound given by the ratio between the atomic linewidth and the typical time of evolution of the atomic states, of the order of $30 \mu\text{s}$ (evaluated by dynamical Bloch equations). This gives an upper bound of $\approx 200 \text{ MHz/ms}$, and our scan rate is small enough to satisfy an adiabatic evolution. On the other hand, the atomic loss due to ballistic expansion gives an upper limit for the scanning time. In the current case, the 10% loss for 1 ms can be considered as a reasonable value. A longer observation time can be obtained with lower temperatures, but the correlation technique, due to its normalization, is pretty insensitive to changes in the temperature of the atomic cloud for copropagating beams. The situation can change, though, in the counterpropagating case, reducing the quality of the signal.

III. EXPERIMENTAL RESULTS

Typical results for the noise correlation spectroscopy are shown in Fig. 3, for a fixed frequency detuning of field 1 ($\delta'_1 = (0.0 \pm 0.5) \text{ MHz}$), comparing the transmitted intensity of the probe laser and the correlation evaluated according to

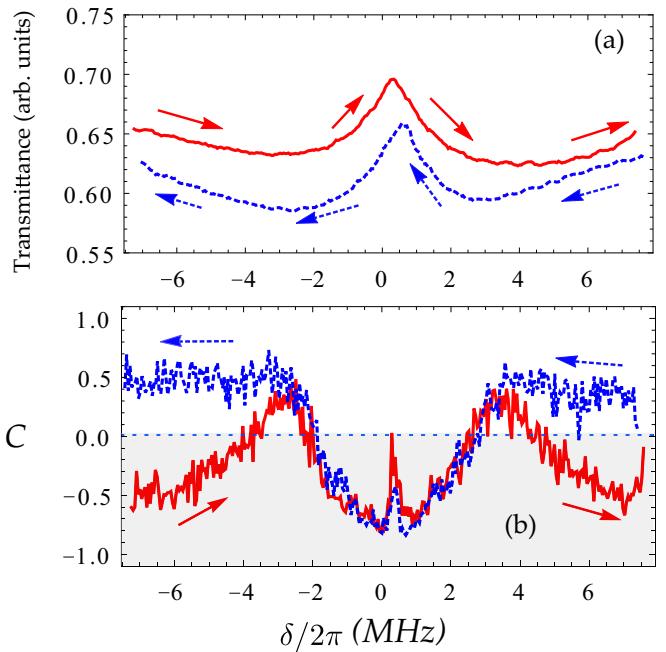


FIG. 3. Intensity (a) and correlation spectra (b) for increasing (red, continuous) and decreasing (blue, dashed) scanning in two-photon detuning $\delta/2\pi$. The arrows indicate the time evolution of the scanning. $\delta'_1 = (0.0 \pm 0.5) \text{ MHz}$.

Eq. (1). All the presented results are an average of 100 runs of the experiment. There is a relevant difference in the behavior of the correlation [Fig. 3(b)] depending on the sign of the scanning rate $d\delta'/dt$. For a positive scan in frequency (red line, continuous), the curve shows anticorrelation out of the EIT region, while for negative scan (blue line, dashed) there is correlation. On the other hand, no significant difference is observed for the mean transmitted power [Fig. 3(a)]. This asymmetry associated with the temporal evolution of the measurement is unexpected, since the sign of the scanning rate $d\delta'/dt$ should not be relevant for an adiabatic scan.

It should be noticed that the resulting spectrum is characterized by two particular regions. In the first one, close to the EIT resonance ($|\delta'| < 3$ MHz), atoms are pumped and trapped in the dark state and no asymmetry is observed, in accordance with our previous observations [13]. The situation is different beyond the transparency window ($|\delta'| > 3$ MHz), where the mismatch in correlation spectra for increasing and decreasing scanning is very distinctive. Therefore, the asymmetry is observed whenever the atoms are not in the dark state and can be excited by the laser beams.

In order to understand this asymmetry, we investigated the evolution of the system for different values of laser detuning frequency δ' . For a fixed frequency of the probe, the correlation and transmission of the fields are measured for 1 ms, while the cooling beams and magnetic fields are turned off. The time scale of this measurement was found to be sufficient to observe the evolution of these atomic responsa while the change of optical density due to ballistic expansion was smaller than 10%. The results are presented in Fig. 4.

For the exact EIT condition ($\delta' = 0$ MHz) the results of intensity and noise reach a steady value after about 40 μ s, consistent with the fast evolution of the atomic populations. Beyond that point, transmitted intensity has a very slow evolution, with a small increase in the transmittance (<0.5 %), consistent with changes in the atomic density.

As the detuning grows, the transmittance presents a fast reduction, associated with the absorption of the field. For $\delta' = \pm 1$ MHz this drop is still small, consistent with the fact that the beam frequency is within the EIT resonance. The correlations show a fast drop to negative values, consistent with the observed values in Fig. 3. Any further evolution is slow under the measured time. Nevertheless, a small asymmetry in transmittance is observed. Similar results are observed for $\delta' = \pm 2$ MHz, with the correlation evolving to positive values.

Out of the EIT peak, but within the atomic linewidth, for $\delta' = \pm 5$ MHz, absorption reaches its peak value. It is noticeable that the correlation signal presents a drop for positive detuning, at 0.3 ms. This difference in the evolution of the correlation signal according to the detuning becomes increasingly relevant as the detuning grows ($\delta' = \pm 9$ MHz), changing back and forth to negative values. For $\delta' = \pm 12$ MHz, the discrepancy in the correlation signal is large enough to prevent the anticorrelation from recovering to positive values within the measurement time.

The slow temporal dependence is, therefore, present only when the atoms are out of the exact two-photon resonance. It is consistent with the idea that light scattering from the atoms plays a role in the evolution of the signal. We propose a model where the resulting force from the absorption accelerates the

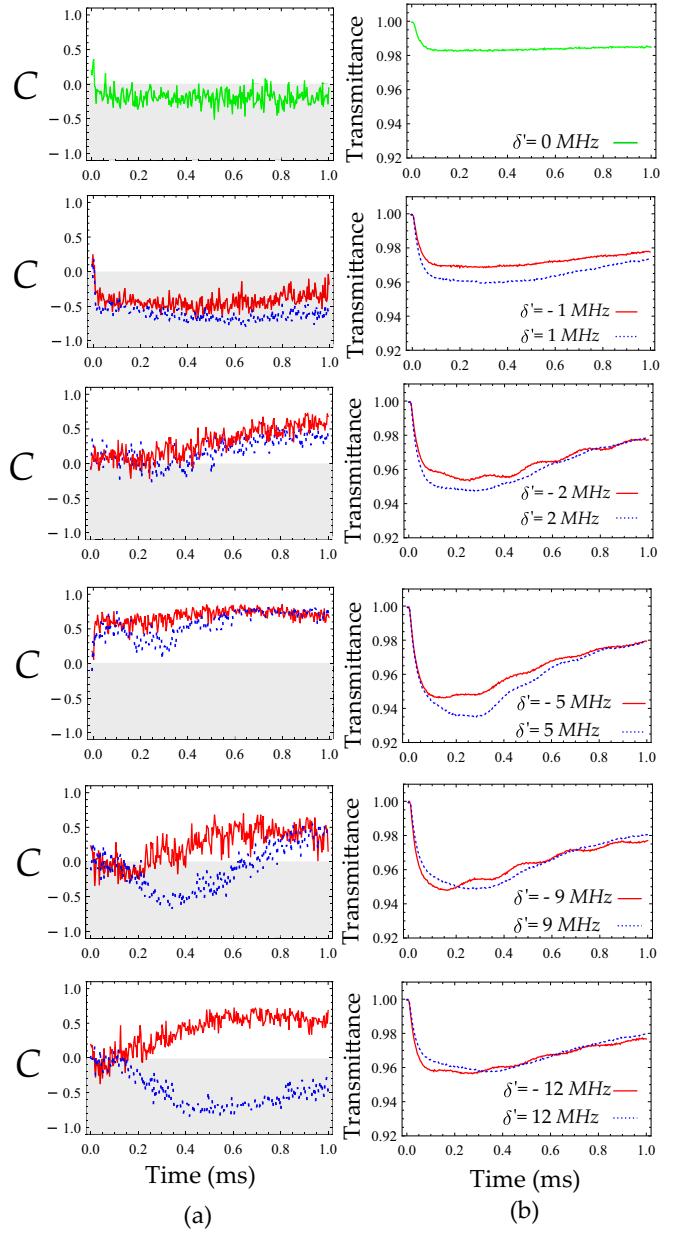


FIG. 4. Measurements of the time evolution of correlation function (first column) and transmitted power (second column) for different two-photon detuning $\delta' = \delta/2\pi$. $\delta'_1 = (0.0 \pm 0.5)$ MHz.

atomic ensemble, leading to a time-dependent Doppler shift of the atomic transitions that can be verified in the evolution of the measured fields. We describe the atom-light interaction in terms of the two dynamical processes in our system, one regarding the external degrees of freedom (atomic velocities) and the other one involving the internal degrees of freedom (atomic states). For the internal degrees of freedom along with the noise correlation we follow the calculation presented in Ref. [13]. In what follows we briefly review the main aspects.

IV. THEORETICAL MODEL

Consider a three-level system interacting with two light fields \mathbf{E}_1 and \mathbf{E}_2 with phase noise in a Λ -EIT configuration.

The phases of each field [$\phi_1(t)$ and $\phi_2(t)$] present random fluctuations, following the statistics of a Wiener process, i.e., $\langle d\phi_i(t) \rangle = 0$ and $\langle d\phi_i(t) d\phi_j(t) \rangle = 2\gamma dt$ with $i, j = \{1, 2\}$, where $\langle \dots \rangle$ stands for the statistical average and γ corresponds to the spectral linewidth of the laser. One important point in the current experiment comes from the fact that both fields come from the same laser, therefore $\phi_1(t) = \phi_2(t) = \phi(t)$.

The dynamics of the two fields coupling the three-level system is given by the Bloch equations in which the atomic density matrix is defined by the slow varying vector $\mathbf{x} = (\rho_{11}, \rho_{22}, \rho_{13}, \rho_{31}, \rho_{23}, \rho_{32}, \rho_{12}, \rho_{21})$ describing the atomic state. According to [13] the phase sensitive Bloch equation is

$$d\mathbf{x}(t) = \mathbf{M}[dt, d\phi^2, \mathbf{v}]\mathbf{x}(t) + \mathbf{B}[\mathbf{x}]d\phi(t) + \mathbf{x}_0 dt. \quad (3)$$

Here vector \mathbf{B} , proportional to \mathbf{x} , does not contribute to the calculations given that $\langle d\phi \rangle = 0$. The matrix \mathbf{M} depends linearly on $dt, d\phi^2$ and contains the Rabi frequencies, the detunings, and the spontaneous emission rate. In the present case, it also takes into account the contribution of the Doppler shift on the field detuning, given by $\mathbf{k} \cdot \mathbf{v}(t) = \mathbf{k} \cdot d\mathbf{r}(t)/dt$. Finally, the constant vector \mathbf{x}_0 is associated with the conservation of the atomic population.

In order to describe the dynamical equation for the velocity of the atoms, we assumed the dipole and long wavelength approximations, i.e., $\lambda \gg a$ (where λ is the optical wavelength and a the Bohr radius). Each field exerts scattering forces. For a traveling wave, under the rotating wave approximation [14], $\tilde{\mathbf{F}}_i = 2\hbar\mathbf{k}_i \Omega_i(\mathbf{r}_0) \text{Im } \tilde{\rho}_{i3}$ where \mathbf{r}_0 represents the position of the atomic center of mass and $\text{Im } \tilde{\rho}_{i3}$ gives the absorption of each beam. Under the effect of those forces, the atoms are accelerated, gaining a Doppler shift affecting the evolution of the internal atomic variables through Eq. (3).

We can determine the time evolution of the mean value of the atomic states $\langle \mathbf{x}(t) \rangle$ under the effect of the optical forces by numerically solving the coupled averaged equations of atomic motion (thus evaluating the atomic velocity) and Eq. (3). The values of $\langle \mathbf{x}(t) \rangle$ give the absorption and the dispersion of the fields associated with $\text{Im } \rho_{i3}$ and $\text{Re } \rho_{i3}$, respectively, for $i = 1, 2$. The transmission for each beam is given by $T_i(t) = \exp(-\alpha \text{Im}[\mathbf{x}(t)]_{2i+1})$ for $i = 1, 2$ where α corresponds to the optical depth.

We use the stochastic treatment in [15] to calculate the evolution of the density matrix elements in Eq. (3). We determine the spectral densities $S_{ij}(\omega)$ in Eq. (2) and then the correlation given by Eq. (1). It is worth noting that the approach in [15] for calculating the correlation in Eq. (1) considers a steady-state condition of the atomic states. Therefore, the evaluation of $C(\omega)$ for each $\mathbf{x}(t)$ in Eq. (3) is only valid for an adiabatic situation, where the evolution of the internal degrees of freedom is fast compared to the changes in atomic velocity. In this case the atomic state given by $\mathbf{x}(t)$ follows the slowly varying steady-state value of $\mathbf{M}[dt, d\phi^2, \mathbf{v}]$.

Resulting calculations for the evolution of correlation and transmittance are shown in Fig. 5 for different detunings. The differences in the time evolution of these signals for the blue and red detunings are evident. On the other hand, the resonant case of EIT presents no slow transient evolution, as expected.

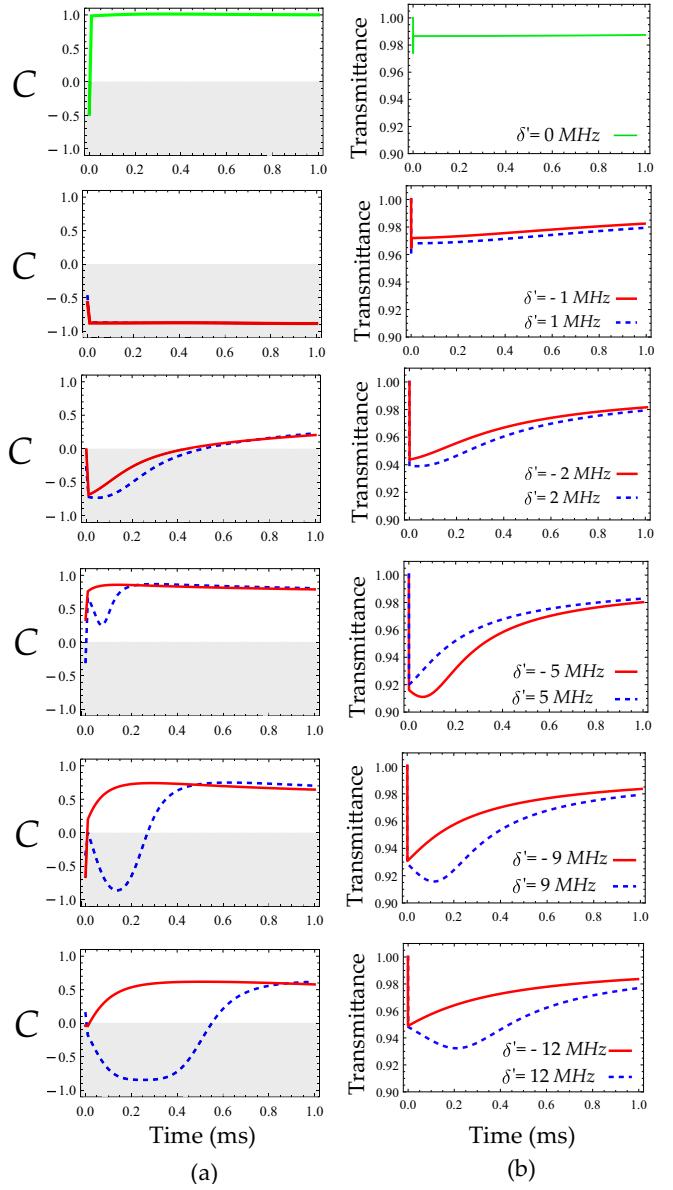


FIG. 5. Calculations for the time evolution of correlation function (first column) and transmitted power (second column) for different detuning frequencies $\delta' = \delta/2\pi$.

The correlation value at exact resonance is positive and nearly unitary.

Out of the EIT condition ($|\delta'| > 2$ MHz), we observe that, for a red detuned probe field, the accelerating atoms lead to a growing detuning, and therefore to an increase in the transmittance. On the other hand, if the probe field is initially blue detuned, absorption increases as the atoms are brought into resonance, and transmittance is increased after they pass through this condition. As for the evolution of the noise correlation signal, it can be understood by the phase-to-amplitude noise conversion observed with diode lasers [16]. Close to the atomic resonance, phase fluctuations are converted into intensity fluctuations by the atomic resonance, but the sign of the conversion term depends on the laser detuning. In the present two-field spectroscopy, for the red detuned probe, the

resulting push increases the absolute value of detuning for both fields, which become red detuned as well. Since these fields are derived from the same laser, noise conversion results in correlated intensities [13]. On the other hand, if field 2 is blue detuned, the increasing velocity of the atoms turns field 1, initially resonant, red detuned. As a consequence, the intensity correlations become anticorrelated in a first moment. This situation does not last, because field 2 eventually becomes red detuned as well, due to the acceleration of the atoms, and their intensities become correlated at the end, as observed experimentally. Typical evolution times of the transmittance and correlation will depend on laser intensities and detunings, affecting the effective Rabi frequency. Although there is no explicit direct equation for this evolution, it is evident that in the low power regime we are operating the maximum force applied to the atoms will be in the region of detunings between the EIT linewidth (of the order of 2 MHz) and the atomic linewidth of 6 MHz.

V. CONCLUSIONS

A direct comparison of the experimental results (Fig. 4) with the calculations presented in Fig. 5 shows a good agreement for the correlation signal, and a limited agreement for the intensity measurements. The measured transmittance is affected by the statistical noise introduced by fluctuations in atomic density, and the small change of transmittance for opposing detunings, of the order of 2% (Fig. 5) is within the intensity fluctuations observed for different runs. On the other hand, the wide change in the correlation makes this signal more robust against perturbations, leading to a better agreement of the observed values and the presented model, and an enhanced sensitivity to the effect of optical forces.

With the knowledge of the evolution of the system under the effect of the radiation pressure over the atom out of the EIT region, we can understand the asymmetry observed in Fig. 3. For the scanning with an increasing frequency (red line, continuous), the system may begin with an anticorrelated signal. As the lasers push the atoms, leading to a red shift for field 1, the probe laser (field 2) is scanned, ending with a blue detuning, and therefore the resulting situation leads back to anticorrelation for the fields. On the other hand, for the scan with a negative variation of the probe frequency, we may begin with correlations for a blue detuned probe. As the atoms are pushed by both fields, at the end of the scan, both fields are red detuned and the correlation shows up again after crossing the EIT region.

While the two-photon detuning is within the EIT bandwidth, the acceleration is negligible, and the detunings of the fields involved remains unchanged, even if the atoms were previously pushed by the fields involved. In this situation, evolution of the observed transmittance will depend mostly on the expansion of the atomic cloud, while correlations will be unaffected in the lowest order. We may conclude, therefore, that the observed asymmetry is associated with

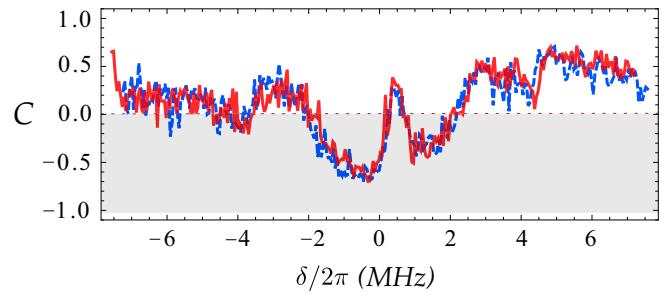


FIG. 6. Correlation spectra for increasing (red, continuous) and decreasing (blue, dashed) scanning in frequency for the counterpropagating configuration of the EIT beams.

the acceleration given by the absorption process around the two-photon resonance, out of the EIT window.

From the calculations in Fig. 5 we estimate that the acceleration due to light forces is of the order of 10^3 m/s^2 resulting in Doppler shifts of the order of the atomic linewidth ($\sim 6 \text{ MHz}$), or atomic velocities of 1 m/s (on the order of magnitude of the thermal velocity of our cold atom cloud). These accelerations for the copropagating case can be quite strong, and immediately estimated by the change of the correlation in Fig. 4. The forces over the atoms will be on the order of 10^{-22} N .

One way to reduce the effect of the radiative pressure is to perform a spectroscopy in a counterpropagating configuration for the EIT beams. Therefore, the forces are nearly balanced ($\bar{F}_1 - \bar{F}_2 \sim 0$) and atomic acceleration is negligible. As a consequence, the correlation spectra (Fig. 6) present no asymmetry with respect to the scanning direction.

In conclusion, the developed model gives a good agreement with the observed results, successfully coupling internal and external degrees of freedom of the atoms. We can observe that the sensitivity of the noise spectroscopy allows the observation of the pushing of the atoms in the vicinity of the EIT condition. Although the intensity spectrum presents a distinctive feature at different detunings, as observed in the theory (Fig. 5), its observation is limited by atomic density fluctuations, while this effect is much more evident in the correlation spectroscopy, with a swing from anticorrelation to correlation of the relevant signal (Fig. 4). Therefore, correlation can become a measurement tool for the evaluation of atomic acceleration under optical forces.

Moreover, under counterpropagating geometry, the contribution of the optical forces are compensated, thus the noise correlation could be employed to detect additional external forces (electric, magnetic, or gravitational), as well as to evaluate their slow drift in time, which would translate into optical detuning shifts.

ACKNOWLEDGMENTS

The authors acknowledge support from Grant No. 2010/08448-2, Fundação de Amparo à Pesquisa do Estado de São Paulo (FAPESP).

[1] K.-J. Boller, A. Imamoglu, and S. E. Harris, *Phys. Rev. Lett.* **66**, 2593 (1991).

[2] M. Fleischhauer, A. Imamoglu, and J. P. Marangos, *Rev. Mod. Phys.* **77**, 633 (2005).

[3] N. Sangouard, C. Simon, H. De Riedmatten, and N. Gisin, *Rev. Mod. Phys.* **83**, 33 (2011).

[4] M. A. Guidry, E. Kuchina, I. Novikova, and E. E. Mikhailov, *JOSA B* **34**, 2244 (2017).

[5] K. Cox, V. I. Yudin, A. V. Taichenachev, I. Novikova, and E. E. Mikhailov, *Phys. Rev. A* **83**, 015801 (2011).

[6] V. I. Yudin, A. V. Taichenachev, Y. O. Dudin, V. L. Velichansky, A. S. Zibrov, and S. A. Zibrov, *Phys. Rev. A* **82**, 033807 (2010).

[7] V. M. Acosta, K. Jensen, C. Santori, D. Budker, and R. G. Beausoleil, *Phys. Rev. Lett.* **110**, 213605 (2013).

[8] D. A. Anderson and G. Raithel, *Appl. Phys. Lett.* **111**, 053504 (2017).

[9] V. A. Sautenkov, Y. V. Rostovtsev, and M. O. Scully, *Phys. Rev. A* **72**, 065801 (2005).

[10] L. S. Cruz, D. Felinto, J. G. Aguirre Gómez, M. Martinelli, P. Valente, A. Lezama, and P. Nussenzveig, *Eur. Phys. J. D* **41**, 531 (2007).

[11] Y. Xiao, T. Wang, M. Baryakhtar, M. Van Camp, M. Crescimanno, M. Hohensee, L. Jiang, D. F. Phillips, M. D. Lukin, S. F. Yelin, and R. L. Walsworth, *Phys. Rev. A* **80**, 041805 (2009).

[12] D. Felinto, L. S. Cruz, R. A. de Oliveira, H. M. Florez, M. H. G. de Miranda, P. Nussenzveig, M. Martinelli, and J. W. R. Tabosa, *Opt. Express* **21**, 1512 (2013).

[13] H. M. Florez, A. Kumar, K. Theophilo, P. Nussenzveig, and M. Martinelli, *Phys. Rev. A* **94**, 012504 (2016).

[14] H. J. Metcalf and P. van der Straten, *Laser Cooling and Trapping of Atoms* (Springer Science & Business Media, Berlin, 2001).

[15] H. M. Florez, L. S. Cruz, M. H. G. de Miranda, R. A. de Oliveira, J. W. R. Tabosa, M. Martinelli, and D. Felinto, *Phys. Rev. A* **88**, 033812 (2013).

[16] T. Yabuzaki, T. Mitsui, and U. Tanaka, *Phys. Rev. Lett.* **67**, 2453 (1991).