Sensitive measurement of radiation trapping in cold-atom clouds by intensity correlation detection

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We present experimental evidence that the intensity correlations of light scattered from a cold-atom cloud are sensitive to the presence of small amounts of radiation trapping in an atomic sample of density $6 \times 10^8/\text{cm}^3$, with an optical depth (for a resonant light beam) of 0.4. This density and optical depth are approximately an order of magnitude less than the density and on-resonance optical depth at which effects of multiple scattering in cold-atom clouds have been previously observed [Phys. Rev. Lett. **64,** 408 (1990)].

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Radiation trapping in atomic vapors refers to the reabsorption of spontaneously emitted photons.[1] The decoherence introduced by this reabsorption significantly affects a variety of important experiments that rely on the preparation of coherent atomic media.[2,3] Within the context of cold atoms, radiation trapping[4] is the principal factor preventing optically trapped atomic samples from becoming colder and denser.[5] Recently an effort was made to reduce radiation trapping by introducing strong anisotropy into the trap.[6]

Clearly it is of interest to devise experimental techniques that detect extremely small amounts of radiation trapping. In Ref. 3 a laser field was used to create a coherent superposition of ground-state Zeeman sublevels, the decay rate of which was sensitive to radiation trapping at densities above $5 \times 10^{10}/\text{cm}^3$. In Ref. 4 the presence of radiation trapping induced abrupt changes in the trapped atom-cloud shape that was observable at number densities of $10^{10}-10^{11}/\text{cm}^3$ and an on-resonance optical depth (OD) of 3. Interestingly, a recent calculation suggested that a measurement of the photon statistics of the light scattered from an atomic sample may reveal the loss of coherence introduced by radiation trapping at an on-resonance OD as low as 0.1.[7]

In this Letter we measure the two-time intensity correlation function of the light scattered from atoms in optical molasses and demonstrate the sensitivity of the photon statistics to the presence of radiation trapping at a number density of $6 \times 10^8/\text{cm}^3$ and an on-resonance OD of 0.4. This density is a factor of 20 less than that of previous measurements[4] of radiation trapping in cold atoms, and the OD is a factor of 8 less.

The delayed two-time intensity correlation for a polarized light wave of intensity I emitted by a chaotic source is expressed by the degree of second-order temporal coherence $g^{(2)}(\tau)$, defined as[8] $g^{(2)}(\tau) \equiv \langle I(t)I(t+\tau)\rangle / \langle I(t)\rangle^2 = 1 + S|g^{(1)}(\tau)|^2$, where $g^{(1)}(\tau) \equiv \langle E(t)E^*(t+\tau)\rangle / \langle E(t)\rangle^2$ is the degree of first-order temporal coherence and S is the spatial coherence of the imaged portion of the source. The Fourier transform of $g^{(1)}(\tau)$ is simply the frequency spectrum.[8] In a trapped atomic sample, besides the coherent excitation by the laser, the atoms also experience incoherent pumping by spontaneous emission from other trapped atoms,[3] thus broadening the spectrum and affecting $g^{(2)}(\tau)$.

The emission spectrum of a near-resonantly excited two-level atom is the well-known Mollow triplet. For typical values of the trap laser detuning and intensity the blue side peak is close to exact resonance and is hence the dominant source of photons likely to be reabsorbed.[9] The fraction of spontaneous photons that are reabsorbed may be estimated from the on-resonance OD, which is approximately given by $n\sigma l$, where n is the number density of atoms in the cloud, σ is the resonant absorption cross section, and l is the length traversed by a resonant photon through the cloud before exiting.[7] A reasonable estimate of l for a roughly spherical cloud is the cloud diameter. Multiplying the OD by the excited-state fraction $\rho_{ee}(\Delta) = (I/2I_s)/[1 + (I/I_s) + (4\Delta^2/\gamma^2)]$ yields the probability that a photon is first emitted and then reabsorbed into the sample. This probability $n\sigma l\rho_{ee}$ may be equated to n_{th}, the thermal average photon number per mode in the incoherent radiation reservoir formed by reabsorbed photons,[3,7] within the limit $n_{\text{th}} \ll 1$. The parameter n_{th} is appropriate for the quantitative characterization of radiation trapping. For example, a value of $n_{\text{th}} = 0.01$ implies that there is a 1% chance that a photon emitted by a trapped atom will be rescattered in the cloud.

In this work we measure $g^2(\tau)$ for different values of n_{th}. We use a vapor-loaded $\sigma^+-\sigma^-$ ^{85}Rb magneto-optic trap. The sum of all six trapping beams (15 mm in diameter) at the position of the cold atoms is 3.6 mW/cm^2. A much weaker repumping light beam is added to prevent the atoms from accumulating in the lower hyperfine ground state. A pair of current-carrying coils external to the vacuum chamber provide a magnetic field gradient along their axis (say, x) of $\sim$8 G/cm. For this work a maximum number density of $\sim 1.5 \times 10^9/\text{cm}^3$ is obtained when the intensity percent ratio for the x, y, z trapping beams is 20:40:40. In this case our trapped atom cloud is approximately spherical with a diameter of $\sim$2.4 mm. By altering the relative intensity of the x, y, z trapping beams, such that the total intensity stays constant, we find

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that we can vary the shape and size of the cloud while the total number of trapped atoms varies by no more than 50% about a mean value ($\sim 10^7$ in our case). This permits us[10] to vary the number density (assumed to be uniform) systematically from $1.5 \times 10^9/\text{cm}^3$ to $\sim 1.6 \times 10^8/\text{cm}^3$. At the lowest density the two-dimensional image of the cloud is an ellipse with a minor (major) diameter of approximately 1.8 (5.4) mm. It is important to note that this method of varying the relative trap beam intensities enables us to hold the temperature constant while changing the density.[10] One needs to fulfill this condition to measure the effect of radiation trapping on the intensity correlations because $g^{(2)}(\tau)$ has been shown to depend sensitively on temperature.[11] Here we maintained a constant molasses temperature of $56 \pm 5\ \mu\text{K}$, measured with a standard time-of-flight technique, for all three densities.

Before we start measurements of intensity correlations, the magnetic field is switched off (the decay time is measured to be 1 ms) to produce optical molasses. The fluorescence from the molasses is collected by a 50-mm focal-length lens (apertured to 2 mm) placed 100 mm away (Fig. 1). The correlations are reduced for unpolarized light; hence the fluorescent light is first passed through a linear polarizer and then imaged onto the 100-μm active aperture of a single-photon-counting avalanche photodiode. For this detection geometry we calculate a spatial coherence factor S of 0.2, meaning that approximately the first four Fresnel zones of the source are imaged onto the detector. The avalanche photodiode output pulses are fed into the stop input of a time digitizer that, in this experiment, digitizes the total measurement time of 1 ms into 50-ns-wide time bins. Count rates for this experiment range from 500,000/s for the lowest atom density to 2×10^6/s for the highest density. The background counting rate, chiefly from laser light scattered off the room-temperature background gas in the vacuum chamber, is $\sim$30,000/s. To initiate measurements of intensity correlations the start input of the digitizer is triggered on a pulse sent by a timing circuit, immediately after the magnetic field gradient has been allowed to decay to zero after being turned off. On receipt of this start pulse the digitizer begins a 1-ms-long time scan and records the number of pulses received in each time bin. We verify that each bin registers mostly a 0, sometimes a 1, but practically never a 2, thereby eliminating double-counting errors. Once a scan is complete, the correlation measurement is stopped and the data are exported. The magnetic gradient is turned back on to reload the trap, and the above process is repeated many times to reduce statistical error.

In Fig. 2 we plot $g^{(2)}(\tau)$ for three different values of radiation trapping. The topmost curve in Fig. 2 corresponds to a density of $1.6 \times 10^8/\text{cm}^3$ and an on-resonance OD of 0.1 ($n_{\text{th}} = 0.007$). The OD of the cloud is measured by monitoring the absorption through the cloud of a weak probe beam swept across resonance. The middle curve in Fig. 2 corresponds to a density of $5.7 \times 10^8/\text{cm}^3$ and an on-resonance OD of 0.4 ($n_{\text{th}} = 0.02$). The lowest curve corresponds to a density of $1.5 \times 10^9/\text{cm}^3$ and an on-resonance OD of 0.9 ($n_{\text{th}} = 0.06$). It is clear that there is a substantial systematic decrease in $g^{(2)}(0)$ as the amount of radiation trapping increases.

In Ref. 7 we calculated $g^{(2)}(\tau)$ for the light scattered from a spatially coherent ($S = 1$) sample of two-level atoms at temperature T excited by a single near-resonant red-detuned laser beam propagating through the sample. If radiation trapping is neglected, one obtains the usual result, $g^{(2)}(\tau) = 1 + \exp(-\delta^2\tau^2)$, where the parameter $\delta = k(k_B T/m)^{1/2}$ is the Doppler broadening of the sample.[8] Here, k is the wave number of the laser light, k_B is the Boltzmann constant,

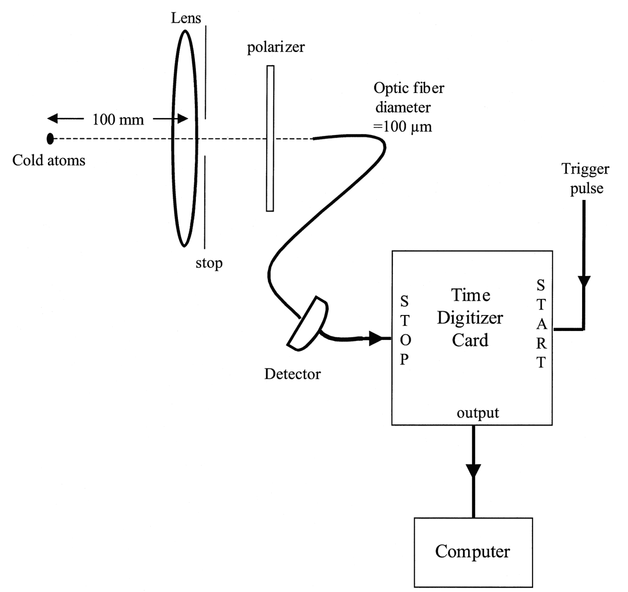

Fig. 1. Setup for measuring radiation trapping in the intensity correlations of the light scattered from cold atoms.

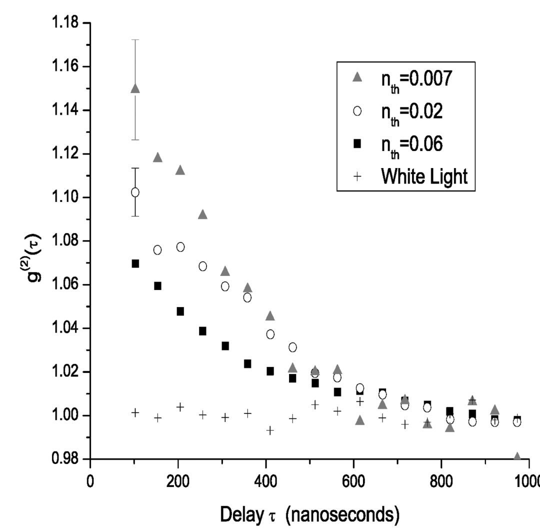

Fig. 2. Clear changes in the intensity correlation function $g^{(2)}(\tau)$ measured as radiation trapping varies. Representative error bars, where significant, are indicated for each data set. To check our counting program and electronics we verified that $g^{(2)}(\tau) = 1$ for a white-light source (shown) and for a portion of the trapping laser beam scattered directly into the detector (not shown).

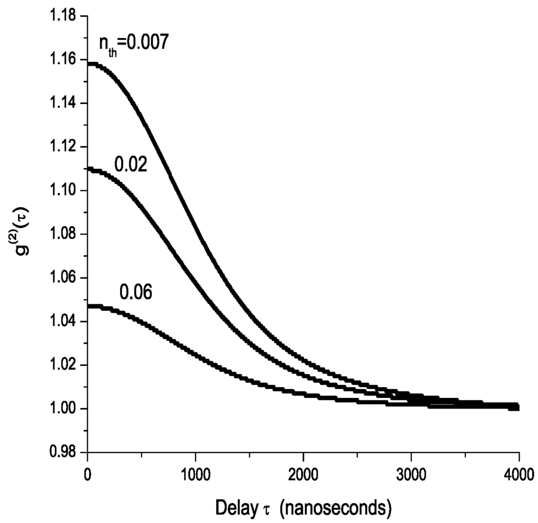

Fig. 3. Theoretical plots of the intensity correlation function $g^{(2)}(\tau)$ for the same values of radiation trapping as in Fig. 2. Note the change in time scale relative to Fig. 2.

and m is the mass of the atom. In Ref. 7 it was shown that, when one includes a small amount of radiation trapping, the prefactor for the exponential term in the expression for $g^{(2)}(\tau)$ above is no longer unity but is $\approx[\rho_{ee}/(n_{\mathrm{th}} + \rho_{ee})]^2$. This reflects the fact that besides the excited-state fraction ρ_{ee} created by coherent excitation, there is also incoherent pumping via n_{th} from the ground to the excited state by rescattered photons. The prefactor gives the probability that two photons are scattered to the detector, both of which come from a coherently excited atom. In Ref. 7 Beeler *et al.* calculated the contributions to $g^{(2)}(\tau)$ from the incoherently excited atoms as well, but to first-order when $n_{\mathrm{th}} \ll 1$ the coherent term dominates.

To model the data in Fig. 2, we extend the treatment in Ref. 7 straightforwardly to include the fact that the atoms are illuminated not by a single beam but by three pairs of counterpropagating laser beams. We obtain

$$|g^{(1)}(\tau)| \approx \left(\frac{\rho_{ee}}{n_{\mathrm{th}} + \rho_{ee}}\right) \sum_{j=1}^{6} a_j \exp(-\alpha_j \delta^2 \tau^2/2). \quad (1)$$

Here the sum is over the six laser beams traveling in the $\pm x$, $\pm y$, and $\pm z$ directions, and $\alpha_j = 2(1 - \cos\theta_j)$, where θ_j is the angle between the observation direction and the propagation direction of laser beam j. The normalized weighing factor a_j is proportional to the product of the intensity of the jth laser beam and the detected fluorescent power radiated in response to this driving beam. In our case, $\alpha_j = 45°$ for the beams in the $+x$, $+y$, $+z$ directions and 135° for the beams in the minus directions. The axis of the polarizer in front of the collection fiber is oriented along the direction $|\hat{x} - \hat{y}|$. In Fig. 2 the a_j values in the x, y, and z directions for the highest number density of $1.5 \times 10^9/\mathrm{cm}^3$ are 1/14, 2/14, and 4/14, respectively. To achieve number densities of $5.7 \times 10^8/\mathrm{cm}^3$ and $1.6 \times 10^8/\mathrm{cm}^3$ (see Fig. 2), we set the intensity percent ratio between the x, y, z trapping beams at 70:15:15 and 90:5:5, respectively, yielding corresponding a_j values of 0.3, 0.07, 0.13 and 0.43, 0.02, 0.05 for the x, y, z directions, respectively. In Fig. 3 we plot theoretical curves for $g^{(2)}(\tau)$ using Eq. (1) and the experimental parameters from the data in Fig. 2.

The decrease in the value of $g^{(2)}(0)$ in Fig. 2 as the amount of radiation trapping increases is in accordance with how the square of the prefactor $[\rho_{ee}/(n_{\mathrm{th}} + \rho_{ee})]$ changes with increasing values of n_{th} in Fig. 3. However, the experimental $g^{(2)}(\tau)$ curves decay much faster in time than the theoretical curves.[12] Our simple theory is based on a two-level atom and ignores frequency broadening caused by fluctuations in the polarization and angular distribution of the radiated light.[13] These fluctuations are due to incoherent Raman-scattering processes. This extra broadening is probably significant for real multilevel atoms, more so in the presence of weak magnetic fields.[11]

In conclusion, we have shown experimentally that intensity correlations of the light scattered from laser-cooled atoms may be used as a noninvasive probe to monitor small changes in radiation trapping in the sample.

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