Universal Scaling of Collisional Spectral Narrowing in an Ensemble of Cold Atoms

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We study the spectral narrowing induced by collisions in a dense cold atomic ensemble. We report on experiments showing a prolongation of the coherence time of optically trapped ^{87}Rb atoms as the density increases, a phenomenon we call collisional narrowing in analogy to the motional narrowing effect in NMR. We derive an expression for the new dephasing time scale in terms of the collision rate and the inhomogeneous decay time. Remarkably, this time scale universally depends only on the atomic phase space density.

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Atomic ensembles have many potential applications in quantum information, from quantum memories [1,2] through the creation of nonclassical states of light [3] and long-distance quantum networks [4]. Owing to collective enhancement, working with ensembles at high densities increases the overall efficiency of quantum operations [5], but at the same time also increases the collision rate and markedly changes the time dynamics of stored coherence. In most applications it is desirable that the atoms will be held in a conservative potential [6]. Such a potential always induces some inhomogeneity in the energy difference between the internal states, which leads to dephasing of the ensemble coherence [7].

Intriguingly, fluctuations in the atomic transition frequency can prolong the coherence time—a phenomenon called motional narrowing. Historically, motional narrowing was first observed in liquid NMR, where a large reduction in the width of spectral lines was observed in comparison to solid NMR due to the thermal motion of the nuclei [8]. Later the effect was reported in other fields such as molecular physics [9], semiconductor microcavities [10], and quantum dots [11]. Collisions-induced narrowing of a Doppler-broadened spectrum in a hot atomic ensemble (Dicke narrowing) was also observed [12,13]. Here we study motional narrowing due to elastic collisions (collisional narrowing) in a dense cold atomic ensemble. In contrast to previous experiments, our apparatus enables precise and independent control over the thermodynamic parameters of the ensemble. Owing to this, we are able to quantitatively analyze the dependence of the narrowed linewidth on the fluctuations rate and strength, and demonstrate an inverse linear dependence on the former and quadratic dependence on the latter. Remarkably, the narrowed linewidth exhibits universal scaling with the atomic phase space density. Recently, Deutsch *et al.* have demonstrated a different mechanism to increase the coherence time of trapped atomic ensembles in the opposite regime in which the velocity-changing collisions rate is slower than all relevant time scales [14].

We consider an ensemble of two level atoms in a trap, with the two internal states designated $|1\rangle$ and $|2\rangle$. The effective single particle Hamiltonian is given by

$$\hat{H} = \hbar[\omega_0 + \delta(t)]|2\rangle\langle 2| + \hbar\Omega(t)|2\rangle\langle 1| + \text{H.c.}, \quad (1)$$

where ω_0 is the free space transition frequency between the states, $\delta(t)$ is the frequency detuning from resonance and $\Omega(t)$ is a classical external control field which is used for state preparation and manipulation. Without external control fields, an atom with an initial superposition $|\psi(0)\rangle = 2^{-1/2}(|1\rangle + |2\rangle)$ will evolve in the rotating frame into the state $|\psi(t)\rangle = 2^{-1/2}(|1\rangle + e^{-i\phi(t)}|2\rangle)$, where the phase difference is given by $\phi(t) = \int_0^t \delta(t)dt$. Atoms at different positions have different detunings due to differential energy shifts [15] or mean-field density dependent interaction shifts [16]. The differential shifts are proportional to the trapping potential, and therefore the detuning δ is proportional to the total energy of the atom [7]. Also, due to the confining potential the atoms carry out oscillatory motion in the trap [Fig. 1(a)]. When the oscillation period is much shorter than the mean time between collisions (as is the case in our experiment), the oscillating $\delta(t)$ can be replaced by its value averaged over several oscillation periods, $\bar{\delta}(t)$.

Without collisions, the average detuning of each atom is constant and therefore the relative phase, ϕ, increases linearly in time [Fig. 1(a)]. The inhomogeneous distribution of the detunings leads to a distribution of phases expanding ballistically, and eventually to dephasing when its width is on the order of π. Velocity-changing elastic collisions randomize the energy of each atom without affecting its internal states. The accumulated phase is then a sum of random phases, and therefore the width of the phase distribution grows only diffusively [Fig. 1(a)]. This means that at any given time the phase distribution is narrower compared to the distribution without collisions [Fig. 1(b)]. The connection between velocity-changing elastic collisions in cold atomic ensembles and coherence times longer than expected was first suggested in [16]. Inelastic collisions are not dealt with in the present work

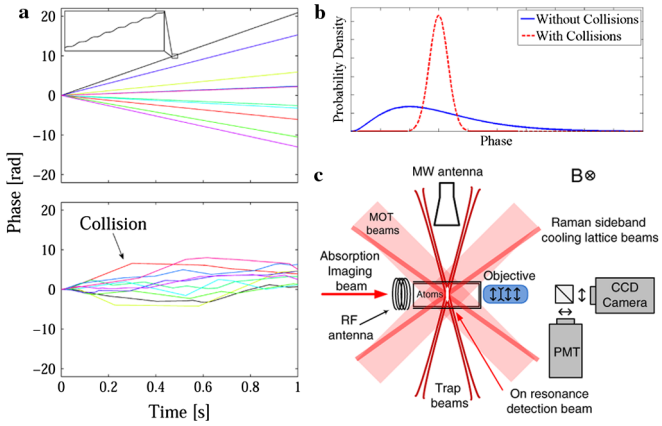

FIG. 1 (color online). (a) The phase accumulation in ten atomic realizations without collisions (upper graph) and with collisions at a rate $\Gamma_{\rm col} = 10\ {\rm s}^{-1}$ (lower graph). The inset shows small oscillations due to the fast oscillatory atomic motion in the trap. Since the oscillation period is shorter than $\Gamma_{\rm col}^{-1}$, we shall consider only the detuning averaged over several oscillations—$\bar{\delta}(t)$. (b) Schematic graph of the phase distribution with and without collisions at some arbitrary time. Both distributions have the same mean value, but with collisions the distribution is much narrower. (c) We laser cool ^{87}Rb atoms and trap them in a crossed laser beams configuration. We employ a state sensitive detection scheme using a detection beam and a photomultiplier tube (PMT), and measure the density and temperature using absorption imaging and a CCD camera.

since their rate is below $1\ {\rm s}^{-1}$, slower than the relevant time scales in our experiment [17].

We quantify the ensemble coherence with the function $R(t) = \frac{|\langle\rho_{12}(t)\rangle|}{|\langle\rho_{12}(0)\rangle|}$, where ρ_{12} is the off-diagonal element of the reduced two-level density matrix [18]. In the experiment, $R(t)$ can be measured in a Ramsey-like experiment: a short $\pi/2$ pulse produced by the external control field prepares the atoms in a superposition $|\psi\rangle = \frac{1}{\sqrt{2}}(|1\rangle + |2\rangle)$ followed by a waiting time, then a second $\pi/2$ pulse, and finally a measurement of the population at $|2\rangle$. The coherence can be written as $R(t) = \int_{-\infty}^{\infty} P_\phi(t)\cos[\phi]d\phi$, where $P_\phi(t)$ is the ensemble phase distribution at time t. To solve for $R(t)$, a specific physical model for the detuning process $\bar{\delta}(t)$ has to be assumed.

Such a model which was shown to describe well the physics of cold collisions is the discrete spectral jumps model [19], in which the detuning of each atom is constant in between jumps, which occur at times following a Poisson distribution. After each jump a new detuning is drawn from a given time-independent distribution. The coherence can be calculated exactly in this "strong collisions" model [19,20]

$$\tilde{R}(s) = \frac{\tilde{R}_0(s+\Gamma)}{1 - \Gamma\tilde{R}_0(s+\Gamma)}, \tag{2}$$

where $\tilde{R}(s) \equiv \mathcal{L}\{R(t)\}$ is the Laplace transform of the coherence, Γ^{-1} is the rate of jumps and $R_0(t)$ is the coherence without the jumps, calculated by taking the Fourier transform of the detuning distribution. Note that Γ^{-1} is the detuning autocorrelation decay time, which for cold atoms confined in a three-dimensional (3D) harmonic potential relates to the conventionally defined collision rate $\Gamma_{\rm col}$ by $\Gamma = \tilde{\Gamma}_{\rm col}/2.7$ [21]. Many times, and, in particular, for atoms in a 3D harmonic trap, the inverse Laplace transform cannot be written in terms of known functions, and it is constructive to introduce a second model which provides a good approximation to the discrete spectral jumps model but whose solution can be written simply in the time domain [19]. In this model, first solved by Kubo [22], the detuning is a *Gaussian process* and the coherence is given by $R(t) = \exp[-\sigma_{\bar{\delta}}^2 \int_0^t (t-t')\Psi(t')dt']$, where $\sigma_{\bar{\delta}}$ is the standard deviation of the detuning distribution and $\Psi(t) = \sigma_{\bar{\delta}}^{-2}\langle\bar{\delta}(t)\bar{\delta}(0)\rangle$ is the normalized detuning correlation function. Using $\Psi(t) = \exp[-\Gamma t]$ for the Poisson collision process, we obtain the following expression for the coherence:

$$R(t) = e^{-\sigma_{\bar{\delta}}^2\Gamma^{-2}(e^{-\Gamma t}+\Gamma t-1)}, \tag{3}$$

which is a generalized Gumbel function [23]. In the limit $t \ll \Gamma^{-1}$ Eq. (3) exhibits a Gaussian decay $R(t) \approx \exp[-t^2/\tau_1^2]$ with an inhomogeneous decay time constant $\tau_1 = \sqrt{2}\sigma_{\bar{\delta}}^{-1}$. In the opposite limit, $t \gg \Gamma^{-1}$, it has an exponential decay $R(t) \approx \exp[-t/\tau_2]$ with $\tau_2 = \Gamma\sigma_{\bar{\delta}}^{-2}$. The exponential decay is the hallmark of motional narrowing resulting in a Lorentzian-shaped transition line where the width is inversely proportional to the collision rate. For non-Gaussian processes Eq. (2) can be used to generalize this last relation into [19]

$$\tau_2 = \alpha^{-2}\Gamma_{\rm col}\tau_1^2, \tag{4}$$

with $\alpha = \tau_1\sigma_{\bar{\delta}}$ being a number in the order of unity which depends on the exact form of the detuning distribution, and for atoms in a 3D harmonic trap is $\alpha \approx 1.69$ (for more details, see supplementary material [24]).

We study experimentally collisional narrowing with cold ^{87}Rb atoms trapped in a far-off-resonance laser with a wavelength of 1.06 μm [see Fig. 1(c), and Ref. [24]). The two internal states used are $|1\rangle = |F = 1; m_f = -1\rangle$ and $|2\rangle = |F = 2; m_f = 1\rangle$ in the $5^2S_{1/2}$ manifold, which are, to first order, Zeeman insensitive in the applied magnetic field of 3.2 G [16]. The thermodynamic parameters of the cloud are measured using absorption imaging, and combining this with the measured oscillation frequency in the trap the collision rate is deduced. The external control of the internal levels is done by means of a two microwave-rf photons transition, and the detection is state sensitive [25]. The spontaneous scattering rate is less than $1\ {\rm s}^{-1}$ and the trap lifetime is better than 5 s, both much longer than the relevant time scales in the experiment. The typical temperature in the experiment is $T = 2\ \mu$K, low enough that we can approximate our Gaussian trap by a harmonic potential. The typical density is $\rho = 10^{13}\ {\rm cm}^{-3}$. The atomic phase space density is smaller than 0.05 which means that the motion of the atoms in the trap can be treated classically. Typical results of two Ramsey

experiments at low and high collision rates are depicted in Fig. 2. For $t < \Gamma_{\rm col}^{-1}$ the two envelopes have the same Gaussian-like decay shape, but for longer times the envelope of the Ramsey experiment with the higher collision rate deviates and changes its form to an exponential-like with a lower dephasing rate.

We fit the envelopes extracted from the Ramsey measurements with the generalized Gumbel function defined in Eq. (3) [see Fig. 2(c)]. We have carried out three sets of experiments with different temperatures, in each of which we have varied the density and extracted τ_2. The measured temperature is used to calculate τ_1 for each data set. As explained before, Eq. (3) is only an approximation to Eq. (2), and to avoid systematic errors we correct the extracted τ_2 with a factor calculated numerically beforehand (for more details, see [24]). In Fig. 3(a) we plot the extracted values of τ_2 for the three data sets, and as predicted by Eq. (4), we get that it depends linearly on the collision rate with a different slope for each temperature. An important consistency test is the comparison of the value of τ_1 calculated from the slopes of the linear fits of Fig. 3(a) to the value calculated directly from the temperature. We use the relation for a 3D harmonic trap $\sigma_{\tilde{\delta}}^{-1} = 1.69\tau_1$ and Eq. (4) and find $\tau_1 = 71 \pm 18$ ms, 47 ± 10 ms, and 30 ± 6 ms for the data sets with temperatures of 1.7, 3.1, and 4.3 μK, respectively. These values of τ_1 are in good agreement with the values $\tau_1 = 73$, 40, and 29 ms, calculated from the measured temperatures. The origin of axis is within the error margins of the three linear fits.

A striking universal scaling of τ_2 is revealed when Eq. (4) is rewritten in terms of the system thermodynamic parameters. The density of atoms can be written $\rho \sim \Phi T^{3/2}$, where Φ is the phase space density, and T is the temperature. The collision rate scaling is $\tilde{\Gamma}_{\rm col} = \rho\sigma_{\rm col}v_{\rm th}$ where $\sigma_{\rm col}$ is the collision cross section, and $v_{\rm th}$ is the average thermal velocity which is proportional to $T^{1/2}$. For low temperatures the collisions are s-wave scattering processes and $\sigma_{\rm col}$ does not depend on density or temperature. The scaling of the collision rate is therefore $\Gamma_{\rm col} \sim \rho T^{1/2}$, and when substituted into Eq. (4) we get $\tau_2 \sim \Phi T^2 \sigma_{\tilde{\delta}}^{-2}$. It can be shown that $\sigma_{\tilde{\delta}} \sim T$ for any potential of the form $U \sim x^n$, and, in particular, this is the case for a harmonic potential. The final result is that the narrowed linewidth exhibits a universal scaling with the atomic phase space density: $\tau_2 \sim \Phi$. The prefactor transforming this relation

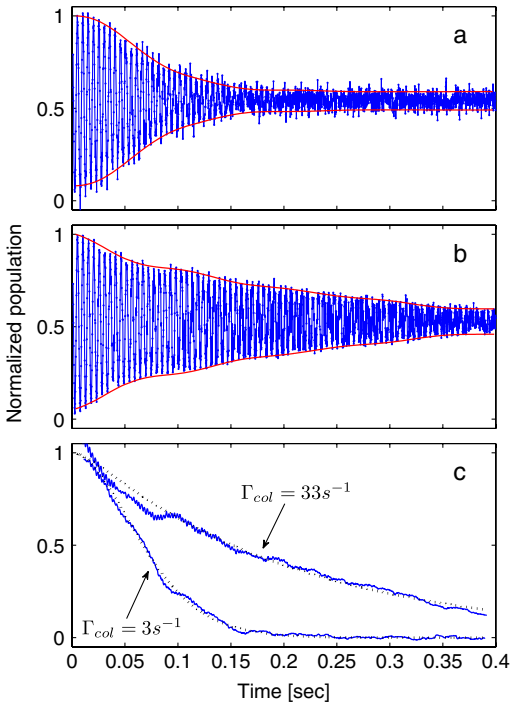

FIG. 2 (color online). Typical results of Ramsey experiments with cold ^{87}Rb atoms. Two short $\pi/2$ pulses are given, separated by a time indicated by the x axis. The y axis is the normalized population at $|2\rangle$. The data presented here were taken for atoms with a temperature of 1.7 μK, which gives a dephasing time of $\tau_1 = 73$ ms [24]. The collision rates are (a) $\Gamma_{\rm col} = 3$ s^{-1}, (b) $\Gamma_{\rm col} = 33$ s^{-1}, and graph (c) is a comparison of the envelopes of the two experiments, normalized to begin at 1 [solid (blue) line]. The dotted lines are fits to a Gumbel function, as defined by Eq. (3). The $\pi/2$ pulse duration is $\sim$300 μs. The detuning of the control field is $2\pi \times 203$ Hz, chosen such that the envelope can be easily extracted. The envelopes are extracted by calculating the standard deviation of all points in a single Ramsey oscillation and multiplying by $\sqrt{2}$.

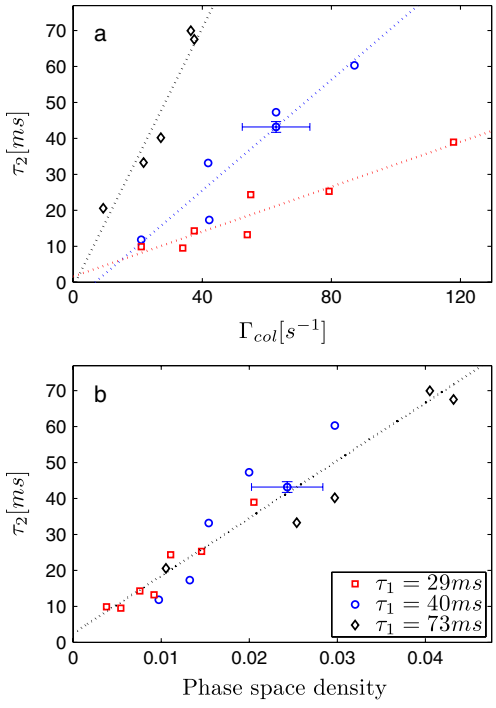

FIG. 3 (color online). The exponential decay time τ_2 as a function of the measured collision rate (a) and phase space density (b) for three data sets with different temperature and τ_1. The collision rate is the average collision rate in the cloud and it is calculated from the measured density, temperature, and oscillation frequency of the trap. The dotted lines are linear fits to the data. The measured temperature is 1.7 μK (diamonds), 3.1 μK (circles), and 4.3 μK (squares).

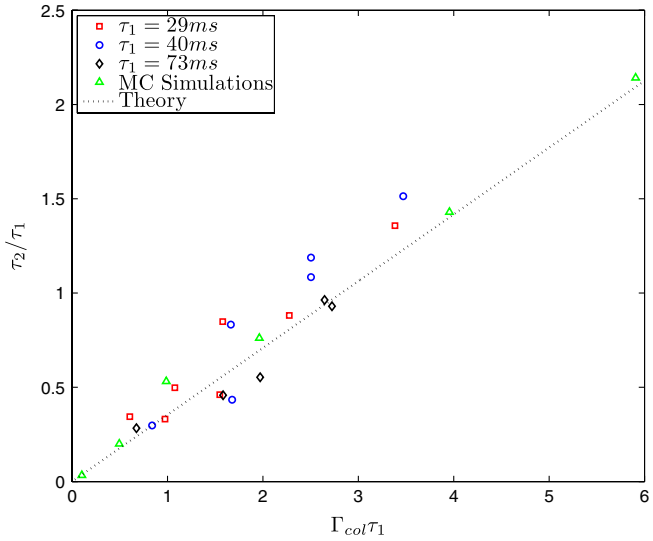

FIG. 4 (color online). A comparison of the dimensionless collisional narrowing time scale, τ_2/τ_1, versus the dimensionless collision rate $\Gamma_{\mathrm{col}}\tau_1$ for experimental data [(red) squares, (blue) circles, and (black) diamonds], Monte Carlo simulations [(green) triangles], and the theoretical prediction of Eq. (4) [(black) dotted line].

into equality is nonuniversal, however, and depends on the elastic cross section and the coupling to the trapping laser. In Fig. 3(b) we plot τ_2 of the same three experimental data sets presented before versus the measured phase space density. As predicted, all data points lie on a straight line. We fit the data with a power law function $\tau_2 = C\Phi^n$ and find $n = 0.91 \pm 0.19$, in agreement with the expected value of $n = 1$. A linear fit yields a slope of $\tau_2/\Phi = 1602 \pm 285$ ms in agreement with the calculated value of 1629 ms based on the trap parameters. All error margins are given for a 95% confidence level.

To further support our findings we perform molecular dynamics Monte Carlo simulations. We simulate separately the classical Newtonian motion of 4000 atoms in the confining potential. The atoms initial conditions are drawn from a Boltzmann distribution assuming a temperature of 4 μK. The collisions are simulated by assuming a steady state density profile to calculate each atom's local probability to undergo a collision. For any such collision, we calculate the atom's new velocity by assuming a virtual counterpart with a velocity which is drawn according to a probability distribution which depends on the velocity of the first atom. We also assume instantaneous s-wave scattering processes. We calculate the energy shift of the internal states induced by the external potential along the trajectory of each atom, and integrate this to obtain the accumulated phase difference. Finally, we use the simulated phase distribution to calculate the coherence. The use of the local density approximation in the simulation is justified since the thermodynamic conditions are not changed during the experiment. In Fig. 4 we plot the experimental results in a dimensionless form, and find that they agree well both with the theory and Monte Carlo simulations.

In summary, we have shown that velocity-changing elastic collisions lead to phase diffusion and spectral narrowing with a linewidth that universally depends only on the atomic phase space density. Though in this Letter the atomic ensemble is treated as an effective single spin system, the effect of collisional narrowing is the same also for a many-body symmetric superposition [24]. For practical applications, working with ensembles with a higher phase space density trapped in a laser light with a larger detuning can easily gain another factor of 10 in the coherence time. From a theoretical point of view, collisional narrowing is unique since the fluctuations arise from the system itself and not due to contact with a noisy environment.

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