Rydberg atom detection of the temporal coherence of cosmic microwave background radiation

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Rydberg atoms immersed in cold blackbody radiation are shown to display long-lived quantum coherence effects on timescales of tens of picoseconds. By solving non-Markovian equations of motion with no free parameters we obtain the time evolution of the density matrix, and demonstrate that the blackbody-induced temporal coherences manifest as decaying quantum beats in time-resolved fluorescence intensities of the Rydberg atoms. A measurable fluorescence signal can be obtained with a cold trapped ensemble of $10^8$ Rydberg atoms subject to suitably amplified cosmic microwave background radiation (CMB) at 2.7 K, allowing for novel insights into previously unexamined quantum coherence properties of CMB.

The interactions of atoms and molecules with incoherent light (such as blackbody radiation, BBR) play a central role in research fields as diverse as photosynthesis, photovoltaics, precision spectroscopy and measurement, and atomic and molecular cooling and trapping. Thermal BBR is a ubiquitous perturber that shifts atomic energy levels, limiting the accuracy of modern atomic clocks, and reducing the lifetime of Rydberg atoms and trapped polar molecules. Recent theoretical developments suggest, however, that quantum noise-induced coherence effects induced by BBR can be used to cool quantum systems and enhance the efficiency of solar cells.

The dynamical response of a material system to incoherent light is determined by the coherence time, a timescale over which the phase relationship between the different frequency components of the light source is maintained. A natural light source such as the Sun is well characterized as a black body radiation (BBR) emitter with temperature $T = 5.6 \times 10^3$ K, and extremely short coherence time of $\tau_c = h/kT \sim 1.3$ fs, where $k$ is the Boltzmann constant. As a consequence, incoherent excitation of atomic systems on timescales relatively long compared to $\tau_c$ produces stationary mixtures of atomic eigenstates that do not evolve in time. However, the coherence time of BBR increases with decreasing temperature and can reach values in excess of 2 ps at 2.7 K, the temperature of the cosmic microwave background radiation (CMB). This suggests the possibility of directly measuring CMB temporal coherences. However, since the CMB intensity is $(300/2.7)^4 = 1.5 \times 10^8$ weaker than that of BBR at 300 K, the absorption signal in most ground-state atoms and molecules even with suitably amplified CMB radiation, is very small. A resolution to this difficulty, shown here, is to use highly excited Rydberg atoms, whose large transition dipole moments make them extremely sensitive to external field perturbations. Previous experimental work has explored the absorption of BBR by Rydberg atoms, leading to population redistribution, photoionization, and lifetime shortening. However, these experiments were focused on measuring population dynamics with no attention to coherence effects. Similarly, no attention has been paid to coherence properties of CMB and the role it might play in enhancing cosmological information.

In this Letter we show that long-lived quantum coherence effects due to cold black body radiation (CBBR – a term that we henceforth use to denote BBR at 2.7 K) absorption can be observed with a cold, trapped ensemble of highly excited Rydberg atoms. (Note that throughout we are referring to temporal coherences.) Using a non-Markovian approach to explore the dynamics of one-photon CBBR absorption by Rydberg atoms, we show that quantum beats can be observed in time-dependent fluorescence intensities. This suggests an experiment to explore the coherence properties of CBBR using a cold trapped ensemble of Rb atoms. Our results demonstrate that non-Markovian and quantum coherence effects play a major role in short-time population dynamics induced by CBBR. A measurement of such coherence properties for the CMB could provide additional insight into cosmological models of the early Universe, and we show that such a measurement appears feasible using current technology.

Theoretically, the interaction of blackbody radiation with atoms is usually considered in the framework of Markovian quantum optical master equations, leading to Pauli-type rate equations for state populations parametrized by the Einstein coefficients. These treatments generally assume that the coherences induced by BBR are negligibly small. The non-Markovian approach adopted here allows us to examine these noise-induced coherences and memory effects arising from a finite correlation time of BBR. To our knowledge, this is the first study of blackbody radiation-driven coherent dynamics in atoms that account for the intrinsic non-Markovian character of BBR.
The time evolution of atomic populations and coherences under the influence of incoherent radiation (such as BBR), suddenly turned on at $t = 0$, is given by

$$\rho_{ij}(t) = \frac{\langle \mu_{i0}\mu_{0j}^* \rangle_p e^{-i\omega_{ij}t}}{\hbar^2} \times \int_0^t d\tau' \int_0^t d\tau'' C(\tau', \tau'') e^{i\omega_{ij}\tau'} e^{-i\omega_{ij}\tau''}$$

(1)

Here $\rho_{ij}(t)$ are the elements of the atom density matrix in the energy representation, $\rho_{i0} = \langle 0 | \hat{a}_i | 0 \rangle$ are the transition dipole moment matrix elements connecting the initial atomic eigenstate $\langle 0 | = | n_o n_0 m_0 \rangle$ and the final states $| i \rangle = | n_b n m \rangle$ with energies $E_0$ and $E_1$, $\langle \ldots \rangle_p$ denotes polarization-propagation average [29], and $\omega_{ij} = (\epsilon_i - \epsilon_j) / \hbar$. For the sake of clarity, we further assume that the atom resides in a single state $| 0 \rangle = | n_o n_0 m_0 \rangle$ before the BBR is turned on at $t = 0$. Since $\rho_{00} \approx 1$ at all times, the density matrix [Eq. (1)] describes the populations and coherences among the states populated by BBR excluding the initial state [14].

The dynamics of the atom’s response to incoherent radiation is determined by the two-time electric field correlation function $C(\tau', \tau'') = \langle E(\tau') E^*(\tau'') \rangle$ in Eq. (1). For a stationary BBR source, the correlation function depends only on $\tau = \tau' - \tau''$, and is given by

$$C(\tau) = E_0^2(0) \zeta(4, 1 + i\lambda\tau)$$

(2)

where $\zeta(4, x)$ is the generalized Riemann zeta-function [13], $\lambda = kT / \hbar$, $T$ is the temperature of the BBR, and $E_0^2(0) = [2\pi^3 / (45\hbar^3 \epsilon_0)](kT)^3$ is the mean intensity of the BBR electric field [13, 30]. Note that Eq. (2) applies when $\omega_{00} > 0$ (absorption), and $C^*(\tau)$ should be used for stimulated emission transitions ($\omega_{00} < 0$). Because $\langle E(\tau') E^*(\tau'') \rangle = 0$ for CBBR [14, 28], there is no coherence between the levels populated in absorption and the levels populated in stimulated emission from a given initial state [27]. Combining Eq. (2) with Eq. (1), and evaluating the time integrals, gives (see supplementary material [26])

$$\rho_{ii}(t) = \frac{\langle \mu_{i0}^2 \rangle_p}{\hbar^2} \left( t | K^{(+)}_0 (\omega_{i0}, t) + K^{(-)}_0 (\omega_{i0}, t) \right) - \frac{\langle \mu_{i0}^2 \rangle_p}{\hbar^2} \frac{1}{\omega_{ij}} \left( | K^{(+)}_0 (\omega_{j0}, t) + K^{(-)}_0 (\omega_{j0}, t) \right)$$

(3)

where $K^{(\pm)}(\omega, t) = \int_0^t \tau^\pm C(\pm \tau) e^{\pm i\omega \tau} d\tau$ are half-Fourier transforms of $\tau$-scaled time correlation functions. In the long-time limit ($t \to \infty$), the right-hand side of Eq. (3) grows linearly with $t$. Note that since we neglect spontaneous emission, the long-time limit is restricted to timescales short compared with the radiative lifetime of the $65\, p$ state (200 $\mu s$ [31]). Using an integral representation for the generalized Riemann zeta function, we obtain the limit [26]

$$\rho_{ii}(t) = \frac{2\pi \langle \mu_{i0}^2 \rangle_p}{\hbar^2} I(\omega_{i0}) t \quad (t \to \infty),$$

(4)

where $I(\omega) = \frac{2\mu^3}{\pi^2 c^2 \omega^3} e^{\omega c / 2\pi kT} - 1$ is proportional to Planck’s spectral density of BBR [14]. Hence, in the longtime (Markovian) limit, this approach reduces to Fermi’s Golden Rule [29], commonly used to calculate the rates of BBR-induced population transfer [8, 31].

The off-diagonal elements of the density matrix are obtained as [26]

$$\rho_{ij}(t) = \frac{\langle \mu_{i0}\mu_{0j}^* \rangle_p}{\hbar^2} \frac{1}{\omega_{ij}} \left( | K^{(+)}_0 (\omega_{j0}, t) + K^{(-)}_0 (\omega_{j0}, t) \right)$$

(5)

Note that due to the double half-Fourier transforms in Eq. (1), Eq. (5) is sensitive to frequency cross correlations in the CBBR.

We now apply this approach to examine the effects of quantum coherence in CBBR excitation of high-$n$ Rydberg atoms. In order to parametrize the equations of motion [14], we calculate the Rydberg energies and transition dipole moments for $^{85}$Rb by solving the radial Schrödinger equation for the Rydberg electron using the Numerov method [8, 27, 32]. To verify the accuracy of
our results, we calculated the spontaneous emission rates from the 30s Rydberg state to various final ns states. These results agree with those reported in [31] to < 5%.

Figure 1(a) shows the proposed setup for observing CBBR-induced coherences. A highly excited Rydberg state of an alkali-metal atom (here we focus on the 65p state of $^{85}$Rb) is created at $t = 0$ by e.g., excitation from the ground 5s state [33]. The newly prepared Rydberg state immediately starts to interact with the 2.7 K CBBR background, establishing a coherent superposition of the neighboring ns and nd Rydberg states [16]. In order to map out the time evolution of Rydberg populations and coherences, Eq. (1) is parametrized by the accurate results for the more modest gain of 10$^9$. As is typical of direct CBBR measurements, the populations in Fig. 2(a) are quite small. As such, we adopt standard CMB amplification practices [21], which can give power gains in excess of 10$^{13}$. Below we report results for the more modest gain of 10$^9$.

While clearly suggesting the existence of long-lived coherences on timescales of up to ~100 ps, the density matrix elements and the purity plotted in Fig. 2 are not experimental observables. To explore the possibility of experimentally measuring the long-lived Rydberg coherences, we evaluate the time-resolved fluorescence signal from the ns and nd states of $^{85}$Rb populated by the interaction with CBBR (see Fig. 1). These states decay
to the 5p state of Rb \(|i_f⟩\) by emitting a photon at a transition frequency of 620 nm, which can be detected with high quantum efficiency. The total power emitted on these transitions by \(N_a\) atoms is given by

\[ I(t) = I_0 \text{Tr} \{\hat{ρ}(t)\} = I_0 \sum_{i,j} \rho_{ij}(t) \]  

where \(I_0 = N_a c / (4\pi\epsilon_0 \omega^2)\), \(c\) is the speed of light, \(\epsilon_0\) is the vacuum permittivity, \(\omega\) is the transition frequency, and \(\rho_{ij}(t)\) is the density matrix element for \(|i⟩\) and \(|j⟩\) states.

Figure 3(a) shows the calculated time dependence of the fluorescence intensity for \(N_a = 10^8\) Rb atoms initially in the \(n_0 = 65p\) state interacting with CBBR amplified by a factor of \(10^9\). The final state to which fluorescence occurs is \(|f⟩ = |5p⟩\). Also shown are the incoherent and coherent contributions to the total fluorescence intensity and \(N_{ph}(t)\).

The incoherent contribution \(I_{\text{incoh}}(t)\) is expressed via the diagonal elements of the density matrix (populations) while the coherent contribution \(I_{\text{coh}}(t)\) specifically highlights the role of quantum coherences. As shown in Fig. 3(a), the coherent contribution to \(I(t)\) remains significant up until \(t < 100\) ps, suggesting the possibility of experimental observation of CBBR-induced Rydberg coherences and their subsequent decoherence.

Figure 3(b) displays the time dependence of the integrated fluorescence signal \(F(t) = \int_0^\tau I(\tau) d\tau\) with \(I(\tau)\) given by Eq. (7), which represents the experimentally measurable average number of photons emitted within the time window \([0, \tau]\): \(N_{ph}(t) = F(t)/\hbar\omega\). The calculated photon flux is \(\sim 0.2\) photons in the first 10 ps, \(\sim 2.3\) photons in the first 40 ps, and \(\sim 26.6\) photons in the first 100 ps of observation, assuming 100% photodetection quantum efficiency. These photon fluxes are well within reach of modern photodetection technology (e.g., employing cooled photocathodes capable of detecting single photons). Furthermore, increasing the amplifier gain from, e.g., the assumed \(10^9\) to \(10^{11}\) would increase the signal by two orders of magnitude.

While not showing any coherent oscillations, the integrated signal including the coherence contributions [full line in Fig. 3(b)] is smaller than its incoherent counterpart [dashed line in Fig. 3(b)] by a factor of 4 at \(t = 40\) ps and by 40% at \(t = 100\) ps. This difference represents a clear signature of time evolution of the CBBR-induced coherences.

In summary, we have computationally shown the existence and decoherence of long-lived quantum temporal coherences induced by CBBR at 2.7 K, which can be observed experimentally by measuring time-dependent fluorescence from the Rydberg states populated by the interaction with the radiation. The physical mechanism behind the coherences and their slow decay is the extremely low temperature and long coherence time of CBBR, coupled with the large transition dipole moments of the Rydberg atoms that make these coherences manifest in various physical observables. These results suggest the feasibility of directly measuring CMB coherence properties, possibly providing important insights into early Universe cosmology (22–25). Initial experimental verification of this approach could be carried out in a cold chamber with walls at 2.7 K, with subsequent experiments aimed at CMB itself.

Finally, we note that the long-lived coherences shown in Fig. 2 can, in principle, be observed not only by measuring time-resolved fluorescence, but with any other experimental technique that is sensitive to coherent superpositions of the atom’s excited states. Examples include selective field ionization (7), photoionization (7), and half-cycle pulse ionization (37). The former technique also provides a direct route to measuring non-Markovian deviations from the linear behavior of state populations at short times (Fig. 2a), which is another measure of the
coherence properties of CBBR [27].

This work suggests a number of extensions to address practical issues in the implementation of this approach. First and foremost we note the need to prepare the initial excited Rydberg state on a rapid time scale. Such fast preparation of the Rydberg state with, e.g., a 15 ps laser pulse is expected to produce a coherent superposition of five eigenstates centered around \( n = 65 \), rather than a single state, as assumed above. This superposition will then couple, via the CMB, to adjacent \( s \) and \( p \) Rydberg states. Fluorescence from this collection of levels is then expected to display a more complicated pattern of quantum beats than described above, which then decoheres in time. Hence, the principle of this experiment remains that same as that described above. Optimizing the choice of pulsed laser characteristics to enhance the quantum beat signal is a study that is now underway.

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