Experimental observation of multi-atom Dicke states in an atomic vapor using optical 2D coherent spectroscopy

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Abstract: We report the first observation of two-, three-, four-, five-, six-, and seven-atom Dicke states in an atomic vapor using optical multi-quantum 2D coherent spectroscopy. This has significant implications in the studies of many-body physics.

OCIS codes: 300.6210 Spectroscopy, atomic; 300.6530 Spectroscopy, ultrafast

Behaviors of many-body systems cannot be always understood by a simple extrapolation of the microscopic laws of a single particle [1]. In 1954, Robert Dicke introduced a coherent collection states of atoms [2], which can provide unique insights into properties of many-body systems. Experimentally, the ability to prepare cold ions, entangled photons and superconducting qubits have advanced the study of Dicke states with a scalable and deterministic number of particles. However, the studies on multi-atom Dicke states in neutral atoms/molecules have been limited to either two particles [3, 4] or a large ensemble [5] so far. Therefore, it is essential to experimentally investigate how many multi-atom Dicke states may exist and how the property scales with the number of atoms in the presence of thermal motion. Here, we report the creation and detection of multi-atom Dicke states up to seven in an atomic vapor by using optical multi-quantum two-dimensional coherent spectroscopy (2DCS). The measured 2D spectra exhibit the correlation of multi-quantum coherence and emission signal, allowing unambiguous detection of selective Dicke states. Furthermore, we found that the decoherence rate exhibits a linear dependence on the number of atoms.

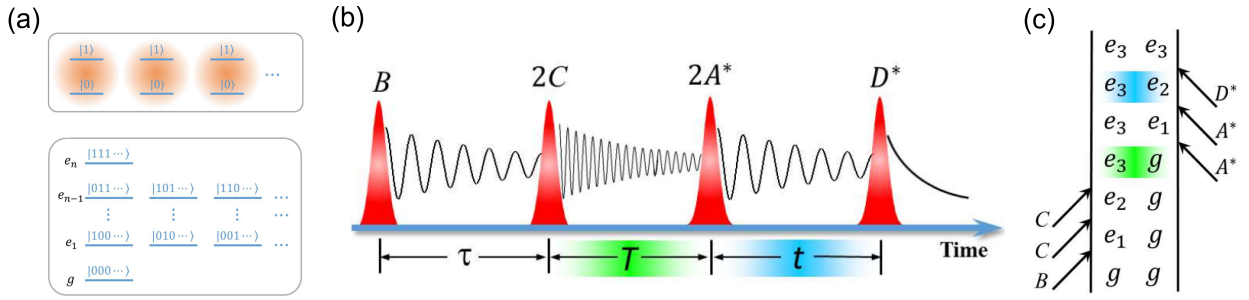

Fig. 1: (a) Energy levels of isolated atoms and multi-atom Dicke states. (b) Excitation pulse sequence of three-atom Dicke states. (c) The double-sided Feynman diagram reveals the generation of three-quantum coherence ρ_{ge_3} process.

For a two-level atom with a ground state $|0\rangle$ and a singly excited state $|1\rangle$, shown in Fig. 1(a), multiple atoms can be correlated by laser field and form Dicke states in the Hilbert space. A multi-quantum coherence $|\rho_{ge_n}\rangle$ between the ground state $|g\rangle$ and the multi-atom excited state $|e_n\rangle$ is generated through a multi-photon process. Here, we use three-quantum 2DCS as an example to describe this process. The time ordering of pulse is shown in Fig. 1(b). Four copropagating pulses B, C, A*, and D* which are phase-modulated by acousto-optic modulators (AOMs) at a specific frequency ω_i [6] are incident on the vapor. In the experiment, as shown the pathway in Fig. 1(c), pulse B generates a single quantum coherence between g and e_1; pulse C acts twice converting it to a three-quantum coherence between g and e_3, which evolves during T; applying pulse A* twice subsequently converts it into a single-quantum coherence between e_3 and e_2, which evolves during t; pulse D* is used to turn the coherence into a population e_3 to radiate a fluorescence signal. Because of this particular time ordering, the signal is modulated at the frequency $\omega_{3Q} = \omega_B - \omega_D + 2(\omega_C - \omega_A)$ and can be isolate and demodulate by lock-in detection using reference frequency ω_{3Q}. The experiment can be further extended to measure n-atom Dicke states by applying C and A* pulses $(n-1)$ times each. For this case, n-quantum coherence ρ_{ge_n} is created and evolves in the time period T, and the single quantum coherence evolves in t. The signal is then sorted out at the frequency $\omega_{nQ} = \omega_B - \omega_D + (n-1)(\omega_C - \omega_A)$ and recorded as a function of T and t. Fourier transforming the time-domain signal generates a 2D spectrum.

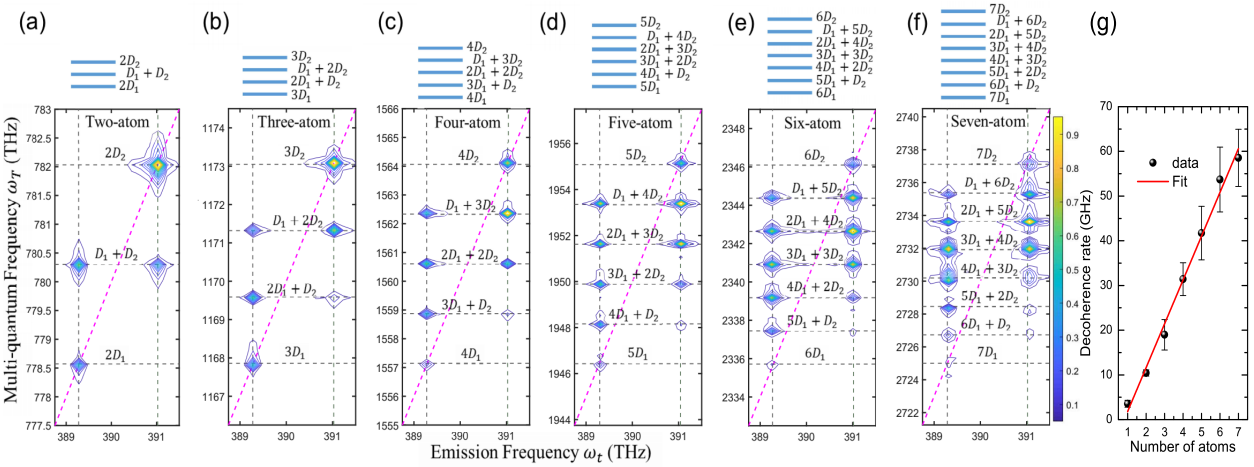

Fig. 2: (a-f) The measured 2D spectra of two atoms, three atoms, four atoms, five atoms, six atoms, and seven atoms Dicke states. (g) The extracted decoherence rate from n-atom Dicke states. For more details, see [7].

Typical multi-quantum 2D spectra are shown in Fig. 2(a)-2(f). The experiment was implemented on a potassium (K) atomic vapor and the femtosecond laser wavelength is tuned to excite the D_1 (389.29 THz) and D_2 (391.02 THz) transitions. Due to possible combinations of n atoms each being in either D_1 or D_2 states, the excited states of n-atom have $n+1$ energies. For example, in the case of three K atoms, the triply excited states can be $3D_1, 2D_1+D_2, D_1+2D_2$, and $3D_2$. Therefore, there are six peaks with three-quantum frequencies that exactly match three-atom triply excited states in the vertical direction of the measured three-quantum 2D spectrum. The peaks with a three-quantum frequency of $3D_1$ or $3D_2$ are located on the diagonal line $\omega_T = 3\omega_t$ with a single-quantum frequency of D_1 or D_2, respectively. While the three-quantum frequency has mixed contributions from D_1 and D_2, the single-quantum frequency can be both D_1 and D_2, resulting in four off-diagonal peaks. For more K atoms, the multi-quantum 2D spectra are shown in Figs. 2(c)-2(f) for four, five, six and seven atoms Dicke states, respectively. Therefore, n-quantum 2D spectra provide direct and unambiguous evidence of n-atom Dicke states.

Additionally, in order to get insights into how the property of multi-atom Dicke states scale with the number of atoms, we further measured the decoherence dynamics of n-quantum coherence at different atom numbers. The decoherence rate are obtained though scanning T. As shown in Fig. 2(g), the extracted decoherence rate linearly increases with the number of atoms, confirming the signature cooperative property of Dicke states.

In conclusion, we experimentally observed multi-atom Dicke states up to seven in a K atomic vapor by using optical multi-quantum 2DCS. In each 2D spectrum, the spectral peaks match the corresponding multi-atom Dicke excited states. The observation of n-atom Dicke states together with the technique of multi-quantum 2DCS, opens a new avenue to study the fundamental many-body physics.

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