Optical 2D Coherent Spectroscopy of Many-body Interaction and Correlation in Atoms

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Abstract: Optical 2D coherence spectroscopy was performed in dilute atomic vapors. The 2D spectra revealed long-range dipole-dipole interaction between atoms at a mean separation up to 16 micrometers and multi-atom correlation up to seven atoms.

Many body interaction and correlation in an atomic ensemble are fundamental in understanding collective and emergent phenomena that cannot be understood by a simple extrapolation of the microscopic laws of a few particles. Experimentally confirmed understanding of many body interaction and correlation in atoms is essential for many problems in cold atoms/molecules, optical atomic clock, semiconductors, and photosynthesis.

Here, we present our study of many-body interaction and correlation in potassium (K) and rubidium (Rb) atomic vapors by using optical 2D Coherent Spectroscopy (2DCS). Two-quantum 2DCS provides a sensitive and background-free detection of dipole-dipole interaction in atomic vapors [1, 2]. The long-range interaction was detected at densities of 4.81×10^8 cm^{-3} and 8.40×10^9 cm^{-3} for K and Rb, respectively, corresponding to a mean interatomic separation of 15.8 μm for K and 6.1 μm for Rb [3]. We also extended the technique to multi-quantum 2DCS and observed multi-atom Dicke states with scalable and deterministic number of atoms up to 7 atoms [4].

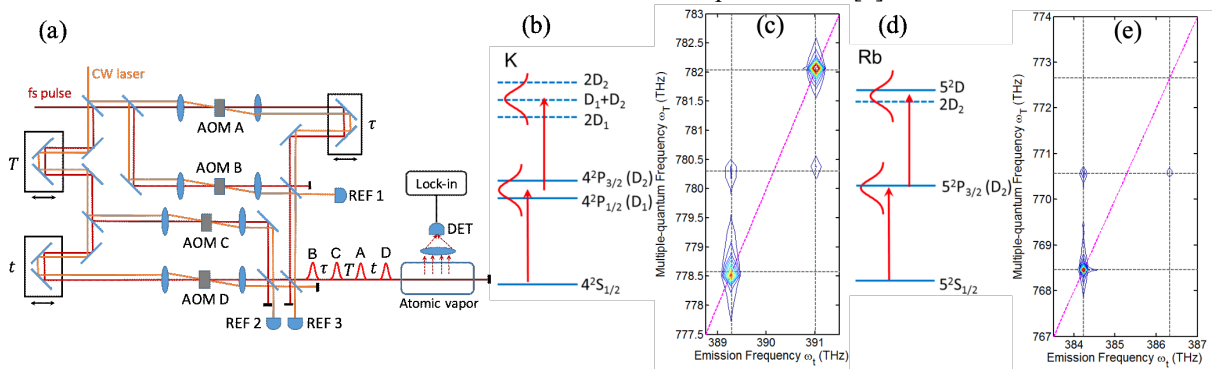

Fig. 1. (a) Schematic of the collinear 2DCS setup based on AOMs. AOM: Acousto-Optic Modulator. Relevant energy levels of single atoms (solid lines) and two atoms (dashed lines) for (b) potassium and (d) rubidium. Two-quantum 2D spectra of (c) a potassium vapor and (e) a rubidium vapor.

A collinear implementation [5] of optical 2DCS based on acousto-optic modulators (AOMs) was used in our experiment. As shown in Fig. 1(a), two nested Mach-Zehnder interferometers split a femtosecond pulse into four pulses (A, B, C and D). Each pulse is phase modulated by an AOM at a slightly different frequency ($\Omega_A, \Omega_B, \Omega_C$ and Ω_D). The pulses arrive at the sample in a sequence and the time delays are controlled by three delay stages. The generated fluorescence signal is recorded by a photodetector (DET) and a lock-in amplifier. A continuous wave (CW) laser beam goes through the same optical path and the AOMs. The beating signals of the CW laser between different arms provide reference frequencies for the lock-in detection. Different reference frequencies are used to extract the nonlinear signal for different pulse sequences and phase-matching conditions. Multi-quantum coherences can be generated by the pulse sequence and detected at the corresponding reference frequencies. The double-quantum signal can be generated and detected at the reference frequency $\Omega_{S,2} = \Omega_A + \Omega_B - \Omega_C - \Omega_D$, with each pulse acting once. This reference frequency can be obtained from REF 3. The output of REF 3 includes all 6 beating frequencies between any two beams of A, B, C, and D. The beating frequencies $\Omega_A - \Omega_C$ and $\Omega_B - \Omega_D$ can be filtered out and mixed to obtain $\Omega_{S,2}$ by a digital signal processor. Similarly, the three-quantum signal can be generated and detected at the reference frequency $\Omega_{S,3} = 2\Omega_A + \Omega_B - 2\Omega_C - \Omega_D$, with pulses C and A each acting twice. In general, the reference frequency for the n-quantum signal is $\Omega_{S,n} = (n-1)\Omega_A + \Omega_B - (n-1)\Omega_C - \Omega_D$ and pulses C and A each act multiple $(n-1)$ times. Theses reference frequencies can be obtained by digitally filtering and mixing the beating frequencies provided by REF 3.

Using this setup, we performed two-quantum 2DCS on both K and Rb atomic vapors. Two-quantum coherence can be created between the ground state and the doubly excited states, as shown in Fig. 1(b) and (d). The resulting two-

quantum 2D spectra are shown in Fig. 2(c) and (e) for K and Rb, respectively. It has been shown [1, 2] that two-quantum 2D spectra reveal two-atom correlation and interaction. We were able to acquire two-quantum 2D spectra from low-density atomic vapors in which the mean interatomic separation is 15.8 μm or $3.0 \times 10^5 a_0$ for K and 6.1 μm or $1.2 \times 10^5 a_0$ for Rb, where a_0 is the Bohr radius. This result confirms the long-range nature of dipole-dipole interaction with an effective interaction range up to tens of μm. The long-range interaction has implications in experiments with optical lattices and atom-based quantum simulators since the interaction is not just limited to the same or nearest sites but also can extend to the sites further away. The experiment in low-density atomic vapors shows that the technique has a sufficient sensitivity for the number of atoms and density in a typical magneto-optical trap, opening the possibilities of 2DCS studies in cold atoms and molecules.

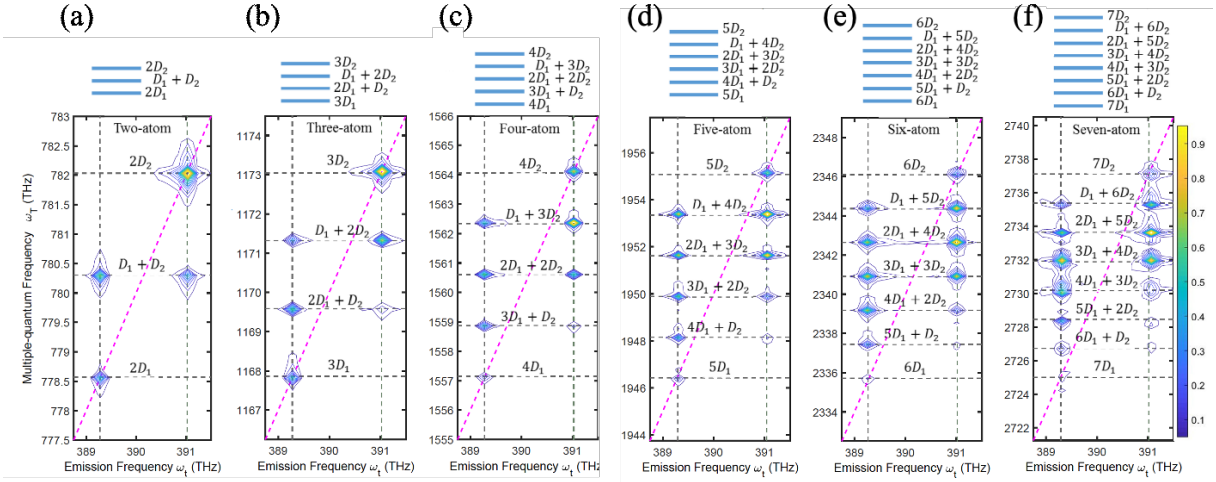

Fig. 2. (a) Experimental multi-quantum 2D spectra resulting from Dicke states of (a) two atoms, (b) three atoms, (c) four atoms, (d) five atoms, (e) six atoms, and (f) seven atoms in a K vapor. Possible energy levels of multi-atom excited states in each case are shown on the top of 2D spectrum.

Two-quantum 2D spectroscopy can be extended to multi-quantum 2D spectroscopy in which the multi-quantum coherence between the ground state and a high-lying energy state can be created and detected by a proper excitation pulse sequence. The multi-quantum coherence can be associated with a collective state of multiple particles, providing a tool to probe many-body correlations. When two-atom states are considered, the doubled excited states of two atoms have three states $2D_1$, $D_1 + D_2$, and $2D_2$ and the resulting 2D spectrum is shown in Fig. 2(a),

Similarly, multi-quantum 2DCS can create and detect higher order quantum coherence associated with multi-atom Dicke states. For three K atoms, the triply excited states have four energies, as shown in Fig. 2(b). The resulting three-quantum 2D spectrum includes six peaks with three-quantum frequencies matching three-atom triply excited states, as indicated by the horizontal dashed lines with corresponding labels. The emission frequency can be either D_1 and D_2. For the cases of more K atoms, the corresponding multi-quantum 2D spectra are shown in Fig. 2(c), (d), (e), and (f) for Dicke states of four, five, six, and seven atoms, respectively. The n-atom excited states have $n + 1$ energies due to possible combinations of n atoms each being in either D_1 or D_2 states. The spectra display a similar pattern as the ones for two- and three-atom Dicke states. The multi-quantum frequencies in the vertical direction match energies of the corresponding multi-atom excited states, while the emission frequencies are D_1 and D_2. The spectra pattern in multi-quantum 2D spectra is a direct result of n-atom Dicke states. The multi-quantum 2D spectra in Fig. 2 are the observation of Dicke states consisting of a scalable and deterministic number of atoms up to seven. The Dicke states with a specific number of atoms can be deterministically selected by using proper multi-quantum 2DCS, allowing possibilities to study the dependence of many-body properties on the number of atoms.

References:

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