Probing dipole-dipole interaction at cold-atom density range using optical two-dimensional coherent spectroscopy

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Abstract: We experimentally demonstrate that the dipole-dipole interaction in a potassium vapor at cold atom density can be observed using optical 2D coherent spectroscopy. This paves the way to implement 2D spectroscopy in cold atoms.

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

Dipole-dipole interactions play an important role in a wide range of physical and chemical systems, from simple atoms to complex molecules [1]. The interactions can lead to collective and emergent phenomena which cannot be explained within the frame of the microscopic laws of a few particles. The dipole-dipole interactions at very high atomic densities ($N > 10^{15}$ cm^{-3}) have been studied successfully. Moreover, thanks to the development of laser technology, cooling and trapping cold atoms have opened a new avenue to study atom-atom interactions at low atomic density. Recently, an extremely sensitive measurement technique named double-quantum two-dimensional coherent spectroscopy (2DCS) has been developed to probe the dipole-dipole interaction [2, 3]. It will be very interesting to study the dipole-dipole interactions at cold atoms using 2DCS. However, the dipole-dipole interaction is weaker at lower atomic densities ($N \sim 10^{10}$ cm^{-3}), posing a challenge to perform 2DCS measurement in cold atoms. Therefore, experimentally confirming the possibility of extending the 2DCS technique to cold atom density range is essential.

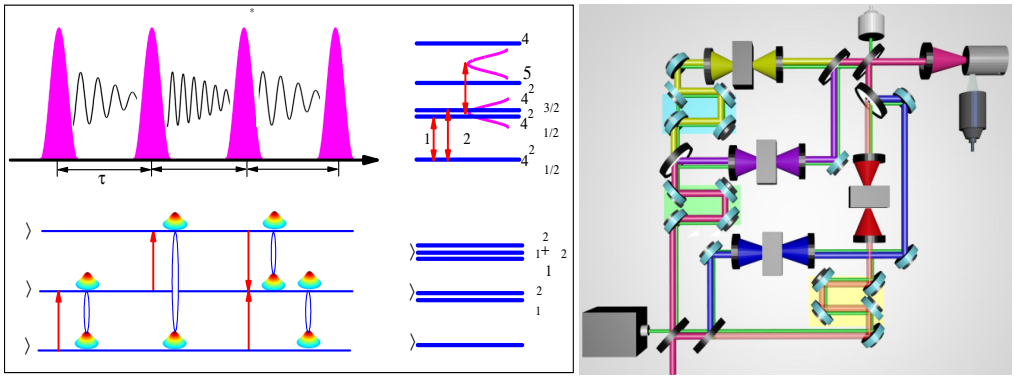

Fig. 1. (a) Pulse sequence, with corresponding time delays τ and T and t. (b) Illustration of dynamics of double-quantum 2DCS. The first pulse **B** puts the system to a single-quantum coherence between the ground state $|0\rangle$ and the first excited state $|1\rangle$. The second pulse **C** converts the single-quantum coherence into a double-quantum coherence between $|0\rangle$ and $|2\rangle$. The third pulse **A** puts the coherence back to a single-quantum coherence, $|0\rangle$ and $|1\rangle$ or $|1\rangle$ and $|2\rangle$, in which atom radiates transient four-wave mixing (TFWM) signal field. The pulse **D** is used to convert the coherence into a population to detect a fluorescence signal. (c) The energy levels of a single potassium atom. (d) The energy levels formed by two potassium atoms. (e) Scheme of the experimental setup.

As shown in Fig. 1(a) and 1(b), the double-quantum 2DCS is generated between the ground state and the doubly excited state by the excitation pulse sequence. For a single potassium atom, within the laser bandwidth tuned to excite the D1 (389.29 THz) and D2 (391.02 THz) lines, there are no atomic energy levels at twice the frequency of the laser, see Fig. 1(c). As a result, no double-quantum signal is expected in this case. However, two potassium atoms will form doubly excited states and the dipole-dipole interaction breaks the symmetry of two-atom states, resulting in the observation of the double-quantum signal [2]. In this work, we extend the double-quantum 2DCS to study the interactions in a potassium vapor at cold-atom density. The setup is shown in Fig. 1(e), more details, see Ref [4]. Instead of detecting FWM signal, we employ the fourth pulse **D** to convert the superposition into a population so that we detect a fluorescence signal. The fluorescence signal is recorded while scanning the time delays T and t. Fourier transforming the recorded data with respect to the two time variables generates a 2D spectrum.

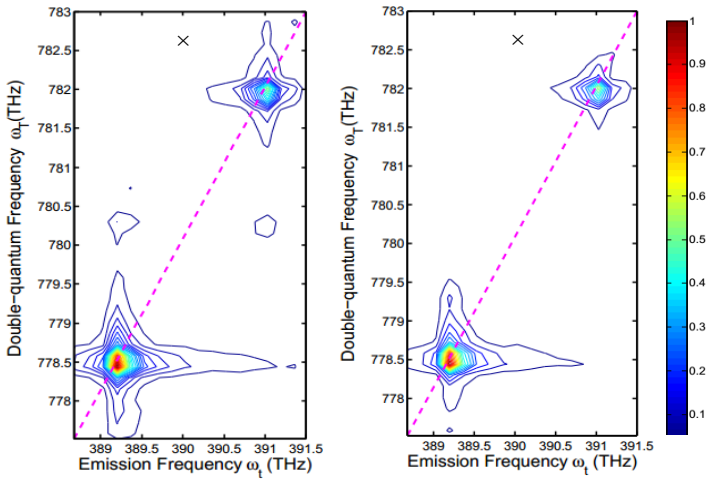

Fig. 2. (a)-(b) 2D spectra obtained from two sets of atomic densities $N = 2 \times 10^{11}$ cm^{-3} and 1×10^{10} cm^{-3}, respectively. The spectra are normalized to the most intense peak.

In Fig. 2(a) and 2(b), we show examples of the measured double-quantum 2D spectra at two sets of atomic densities 2×10^{11} cm^{-3} and 1×10^{10} cm^{-3}. For an atomic density of 2×10^{11} cm^{-3}, the spectrum exhibits four peaks marked by numbers 1, 2, 3 and 4. Peaks 1 and 4 correspond to $2D_1$ and $2D_2$ states (see Fig. 1(d)) located on the diagonal line $\omega_T = 2\omega_t$ with an emission frequency $\omega_t = 389.29$ and $\omega_t = 391.02$ THz, respectively. Peaks 2 and 3 from state D_1+D_2 show the same double-quantum frequency $\omega_T = 780.31$ THz while emitting at $\omega_t = 389.29$ and $\omega_t = 391.02$ THz, respectively. These features are in good agreement with the previous observations at an atomic density of 3.5×10^{12} cm^{-3}, which are attributed to the dipole-dipole interactions between potassium atoms [2]. With the further decrease of density to 1×10^{10} cm^{-3}, although the peaks 2 and 3 disappear, the distinct signal from peaks 1 and 4 still can be acquired. This suggests that the possibility of probing the dipole-dipole interaction at cold atom density can be implemented using 2DCS.

In conclusion, we demonstrate that it is possible to probe dipole-dipole interaction in a potassium vapor at cold-atom density range using 2DCS. This might be of particular interest for comparison study of hot atoms and cold atoms, to help us understand the effects of the thermal motion and determine the fundamental parameters of the dipole-dipole interactions, such as interaction strength, effective interaction range and the number of interacting atoms.

References

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