Collective effects in atomic and helium droplet systems revealed by phase-modulated pump-probe spectroscopy

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Synopsis Phase-modulated pump-probe spectroscopy is used to selectively detect multi-atom resonances in atomic vapors and doped helium droplet beams. A distinct phase signature is observed for the individual resonances. The observation of these effects is surprising since interparticle interactions should be insignificant in these systems.

Time-resolved nonlinear spectroscopy has opened many new directions to study ultrafast dynamics in complex quantum systems [1, 2, 3]. While most applications have been to the condensed phase, we are focusing on dilute gas phase samples, in particular, on doped helium droplet beams. To account for the small densities in our samples, we have recently significantly improved the sensitivity in our femtosecond pump-probe scheme by adapting a phase modulation technique combined with phase-synchronous lock-in detection [4].

Incorporating harmonic lock-in detection, we have further extended this technique to enable selective probing of higher-order nonlinear signals. With this, we observed for the first time the collective excitation of up to four atoms in a dilute atomic vapor ($\rho \approx 10^8$ cm^{-3}) [5]. The origin of these signals is discussed since interatomic interactions are very small in these systems [6]. Similar signatures are observed in doped helium droplet beams, where interactions between dopants sitting on different droplets should be even less significant.

The detected one- to four-atom resonances scale all linearly with the sample density which may seem counter intuitive. However, this behavior was also observed for other collective effects such as interatomic coulombic decay (ICD) [7, 8].

Most striking is the distinct phase signatures observed for the collective resonances in our experiments. High resolution measurements ($\Delta\nu \approx 1$ GHz) have revealed that this phase behavior is connected to the fundamental hyperfine sub-levels in the system. In Fig. 1 we show data obtained for a rubidium vapor. Here, the one-atom transitions $5S_{1/2} \rightarrow 5P_{3/2}$, $5S_{1/2} \rightarrow 5D_{5/2}$ exhibit the same phase signature. In contrast, the two- to four-atom response clearly reveals different phases for the individual hyperfine transitions.

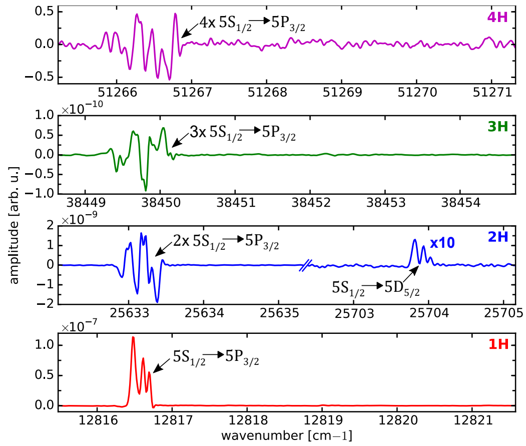

Figure 1. Spectra for one- to four-atom excitation (labeled 1H-4H) in a rubidium vapor. The 1H spectrum shows the $5S_{1/2} \rightarrow 5P_{3/2}$ transition with the onset of the hyperfine sub-structure. In the 2H spectrum the two-photon excitation $5S_{1/2} \rightarrow 5D_{5/2}$ of a single atom can be seen as well as the collective two-atom excitation $2 \times 5S_{1/2} \rightarrow 5P_{3/2}$. The 3H and 4H show the same collective excitation for three and four atoms, respectively.

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