Detecting Sodium D-line Quantum Beat in LIF via Multi-step Femtosecond Excitation

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Quantum interference is a powerful tool for manipulating atomic and molecular systems, with applications in coherent control, wave packet interferometry, and quantum beat spectroscopy (QBS)[1]. QBS involves creating and probing coherent superpositions of quantum states. Ultrashort laser pulses are particularly useful for QBS, offering broad bandwidth for exciting states with large energy separations.

In our study, we use ultrashort pulses to perform QBS on atomic sodium vapor, specifically investigating quantum beats between the $^3P_{1/2}$ and $^3P_{3/2}$ levels. Two laser pulses are used to excite the sodium atoms: the first pulse excites the atoms from the ground state ($^3S_{1/2}$) to two intermediate states ($^3P_{1/2}$ and $^3P_{3/2}$) via the D lines, while the second pulse promotes them to the $^6S_{1/2}$ state. Fluorescence at 515 nm (direct) and 330 nm (indirect) is measured.

As shown in Figure 1, adjusting the delay between the two pump pulses induces high-contrast oscillations in the 515 nm fluorescence, with a period of 1.94 ps, corresponding to the 515 GHz energy gap between the two ^3P states. Similar behavior is observed in 330 nm fluorescence, confirming quantum beating between the $^3S_{1/2}\rightarrow{}^3P_{1/2}\rightarrow{}^6S_{1/2}$ and $^3S_{1/2}\rightarrow{}^3P_{3/2}\rightarrow{}^6S_{1/2}$ pathways. A semi-classical model based on optical Bloch equations was developed to reproduces these oscillations, showing excellent agreement with experimental results, as can be seen in Figure 1.

This interference effect could enhance fluorescence detection using heterodyne methods, with potential applications in high-emissivity diagnostics, such as solid propellant analysis.

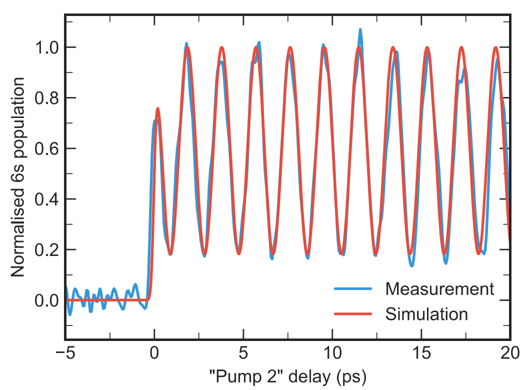

Figure 1. Comparison between normalized fluorescence intensity collected at 515 nm vs second pump delay and simulation of the 6S1/2 population

1. S. Haroche, "Quantum beats and time-resolved fluorescence spectroscopy," in Topics in Applied Physics, Vol 13, Springer (1976), pp. 253–313