# Two-dimensional Fourier-transform Spectroscopy of Potassium Vapor

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Optical two-dimensional Fourier-transformed (2DFT) spectroscopy is used to study the coherent optical response of potassium vapor in a thin transmission cell. Rephasing and non-rephasing spectra of the $D_1$ and $D_2$ transitions are obtained and compared to numerical simulations. Calculations using the optical Bloch equations gives very good agreement with the experimental peak strengths and line shapes. Non-radiative Raman-like coherences are isolated using a different 2DFT projection. Density-dependent measurements show distortion of 2DFT spectra due to pulse propagation effects.

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## I. INTRODUCTION

Vapors of alkali metal atoms are ideal for studying coherent light-matter interactions because the single outer electron results in a relatively simple optical spectrum with isolated lines. Alkali metals are the atom of choice in many areas of atomic physics such as ultracold atoms [1] and electromagnetically induced transparency [2], both of which are currently areas of extensive activity. While most studies have been carried out using continuous wave lasers and frequency domain methods, alkali atoms have also been studied using transient optical techniques. For example, tri-level [3] and two-photon [3] photon-echoes were first observed in a sodium vapor. Propagation effects [4, 5], quantum interference [4, 6, 7] and non-Markovian dynamics [8, 9] have been studied in a potassium vapor. Recently, rubidium vapor has been used as a model system for developing optical two-dimensional Fourier transform (2DFT) spectroscopy [10–12].

The technique of 2DFT spectroscopy was originally developed in nuclear magnetic resonance [13]. Implementation in the optical region of the spectrum was first proposed using a Raman excitation scheme to study molecular vibrations [14]. Currently, 2DFT techniques using infrared excitation to study molecular vibrations are widely used [15–17]. 2DFT techniques using near-IR to visible excitation is also used to study electronic excitations in molecules [18] and semiconductor nanostructures [19]. 2DFT spectroscopy explicitly measures the phase of a transient four-wave mixing (TFWM) signal during two time periods of a three-pulse excitation sequence [20]. The resulting data is Fourier transformed onto a two-dimensional frequency plane, thus separating the electronic interactions between photoexcited states.

In this paper, we present a detailed study of the coherent optical response of potassium vapor in a thin transmission cell using optical 2DFT spectroscopy. Data is acquired for the standard rephasing and non-rephasing configurations, as well as for the projection that isolates the non-radiative "Raman" coherence [21]. We observe peak shapes and amplitudes associated with two coupled $D$ lines. By increasing the number density, we can observe effects due to propagation. The rephasing and non-rephasing spectra are compared to numerical simulations based on the optical Bloch equations.

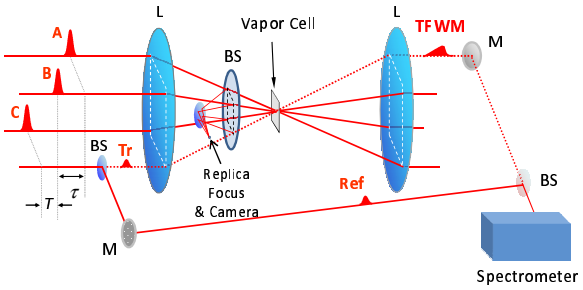


FIG. 1: (color online) Schematic diagram of the experimental setup. Notation: L:lens, BS: beam splitter, M:mirror.

## II. EXPERIMENTAL

A schematic of the experiment is given in Fig. 1. A mode-locked Ti:sapphire laser supplies $\sim 200fs$ pulses, centered at 768.2 nm, to the multidimensional optical nonlinear spectrometer [20]. The spectrometer splits the Ti:sapphire pulses into four phase-locked replicas that are aligned in the box geometry. The pulses are focused on the same spot of a home-made alkali vapor cell. Three pulses (A, B and C) generate a TFWM signal. Time delays between the pulses are denoted $\tau$, $T$ and $t$ between the first and second, the second and third and the third pulse and signal respectively. Pulse four provides the phase-stabilized tracer (Tr) and reference (Ref) separately: the tracer co-propagates with the TFWM signal and is useful for alignment, but is blocked during the 2DFT measurements; the reference beam is routed around the vapor cell and recombined with the signal, producing an interferogram that is recorded by a cooled CCD spectrometer. The phase of the signal field is determined by an all-optical method, which is realized by careful measurement of relative phases between all laser pulses and the TFWM signal [22]. Further experimental

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