2P103

Optical control of coherent phonons in bismuth

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Abstract

Observation and control of the ultrafast dynamics of physical systems under laser excitation allows a full understanding of decoherence processes. Among all the elementary excitation processes, nuclear vibrations in molecules or solids are of great interest, being covered by a variety of research fields such as photo-physics [1] and -chemistry of surfaces, solids, nanostructures[2], and molecules. Moreover a deeper knowledge of these processes may have important applications in both quantum [3] and classical [4] information.

The interaction of ultrashort laser pulses with solids often causes intense collective vibrations of the atoms of the crystal lattice. These vibrations are characterized by a high degree of spatial and temporal coherence, and are referred to as coherent optical phonons (CP). Their oscillation period generally lies in the terahertz (THz) range and is observed through reflectivity changes. These reflectivity changes are measured via a pump-probe scheme through the variation of the probe intensity at the excitation wavelength. We study bismuth (mono- and poly- crystalline) not only because it is a semimetal (zero bandgap) but also because of its relatively slow vibrational modes (330~500 fs periods) whose dynamics has been well studied [6-11]. We use a probe laser pulse with its central wavelength at 800nm and its bandwidth corresponding to about 30 fs in the Fourier transform limit.

From the viewpoint of control, several pulse shaping methods have been used in molecules [12] or alloys [13] in order to excite specific vibration modes. In our approach, we use a pair of phase-locked chirped pulse, which can serve as pump and control pulses.. The highly stabilized Michelson interferometer [14, 15](fig.1) enables us to accurately change their relative phase and as a consequence to finely manipulate the total intensity profile resulting from the beating structure of the two overlapping pulses. This rather simple manipulation allows us to control the phase and amplitude of the fully symmetric A_{1g} phonon (displacement along the trigonal axis, perild~330 fs) up to room temperatures. It is also possible at low temperatures (below 150 K) to manipulate the amplitude ratio between A_{1g} and doubly degenerated E_g (displacement perpendicular to the trigonal axis, period~500 fs) when these two coherent phonon modes are excited simultaneously.

The detailed study of the 2D images (reflectivity change vs pump-control separation and pump-probe delay) and phonon interferograms provides a clear demonstration of the phase and amplitude control.

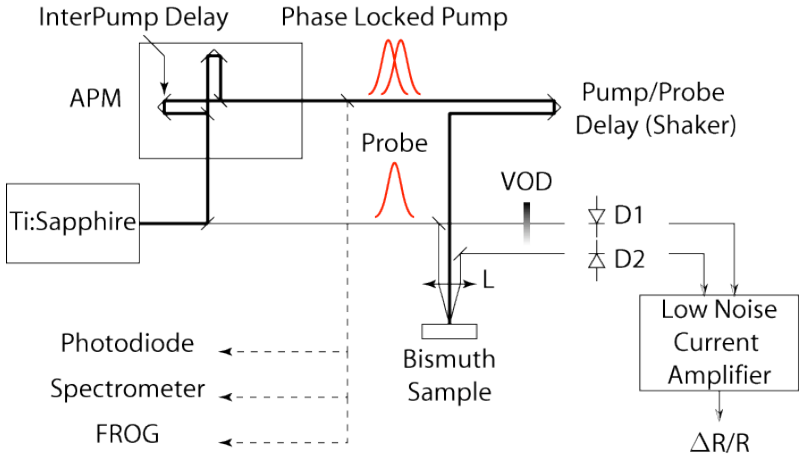

Fig. 1: Experimental setup for the optical control and detection of coherent optical phonons in solids: A femtosecond oscillator is used to generate pump, control and probe pulses at 800nm central wavelength. The phase-locked pump and control pulses are generated in an attosecond phase modulator (APM). The induced transient probe-reflectivity-change of the sample is detected with a balanced photodiode scheme.

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