Ultrafast Coherent Oscillations Of Optical Phonons In Single Layer MoS_2

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Abstract

Spontaneous Raman spectroscopy is extensively used for identifying the number of layers in transition metal dichalcogenides (TMDs) samples, according to the frequencies of the optical phonon modes. Although vibrational modes of TMDs have been largely investigated by CW Raman scattering, their behaviour in the time domain is still poorly understood.
Here we use femtosecond pump-probe spectroscopy to detect impulsively excited coherent optical phonons in single-layer MoS_2. In our experiment, a frequency tunable sub-20fs pump pulse excites electron-hole pairs in the system, while a time-delayed broadband probe pulse, covering A, B and C excitons spectral region (1.8-3.1eV), monitors the electronic relaxation. As shown in Fig.1, optical phonons coherently modulate the transient optical response predominantly across the C peak.
In the dynamics of the C exciton (Fig.2a), superimposed on the incoherent electronic relaxation, we can clearly distinguish a coherent oscillating contribution of optical phonons, with a period of 81fs, an amplitude of 2% of the signal peak, and a Fourier transform (Fig.2b) peaked at the A_1 mode (408cm^{-1} [1]). Surprisingly, the other Raman active mode E' has a negligible contribution to the time domain response. These results are supported by *ab initio* electron-phonon coupling calculations, predicting both the higher coupling of carriers with the A_1 mode, and the strong enhancement around the C exciton.

References

[1] A. Molina-Sanchez and L. Wirtz, Physical Review B, 84, 15 (2011) 1-8

Figures

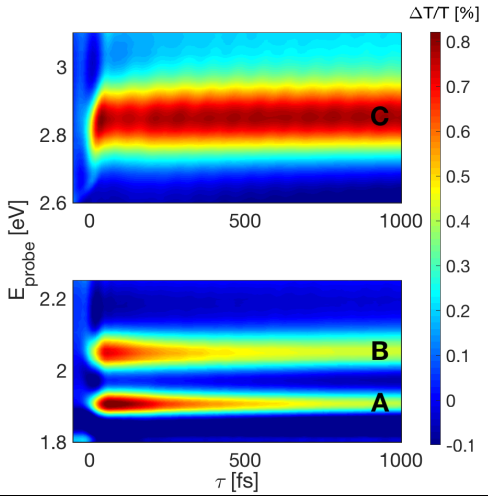

Figure 1: ΔT/T maps of single layer MoS_2 around A, B and C excitons following excitation at 2.6eV.

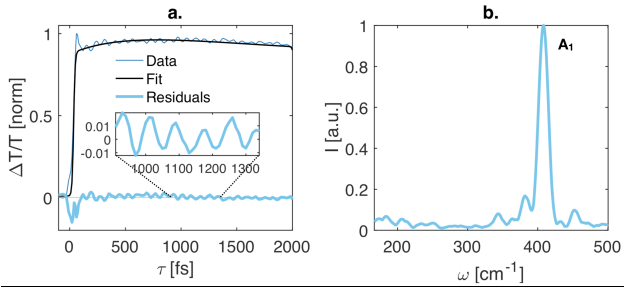

Figure 2: (a.) Dynamics around C and extracted coherent oscillations; (b.) Fourier transform