Dynamics of Long-lifetime Coherent Phonon in $MoTe_2$

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1. Introduction

Molybdenum ditelluride ($MoTe_2$), a representative transition-metal dichalcogenide (TMDC), is a promising material for optical and electrical device applications due to its unique layered two-dimensional structure and the phase change properties. $MoTe_2$ can crystallize in three different phases, i.e. 2H, 1T', and T_d phases [1]. The 2H-$MoTe_2$ is a semiconductor while the 1T'-$MoTe_2$ is metallic, and hence the control of the phase transition is of paramount importance for transistors and optical applications.

Optical excitation is believed to be one of the potential pathways for controlling ultrafast phase transitions [2]. In addition, interactions between lattice and photo-excited carrier systems are a crucial issue for TMDC-based electrical and optoelectrical device applications as the electronic properties are governed by electron-phonon interactions. However, little has been experimentally unveiled in terms of ultrafast structural dynamics and electron-phonon coupling in TMDCs. Coherent phonon spectroscopy is a powerful experimental tool to explore the lattice degrees of freedom on ultrafast time scales and enables investigation of electron-phonon coupling dynamics as well as ultrafast phase transitions. In this study, we carried out coherent phonon spectroscopy in a bulk crystal of 2H-$MoTe_2$ by employing optical pump-probe measurement. We found that the lifetime of the A_{1g} Raman-active phonon mode is unusually long even at room temperature. We also found that the frequency of the coherent phonon varies with the pump-probe time delay and its behavior strongly depends on the pump fluence (F).

2. Result & Discussion

Optical pump-probe measurements were carried out using a femtosecond Ti:sapphire laser oscillator which provided near infrared optical pulses with a pulse duration of ≤ 30 fs and a central wavelength of 800 nm. The pump beam fluence was varied from 50 to 400 μJ/cm^2. The transient reflectivity change ($\Delta R/R$) was recorded as a function of pump-probe time delay. The sample used was a small flake of 2H-$MoTe_2$ single crystal with the c-axis of the crystal corresponding to the sample normal.

Fig. 1 shows a transient $\Delta R/R$ signal for F = 50 μJ/cm^2. A sharp electronic response followed by a clear oscillatory pattern due to coherent phonons was observed at 0 ps (the arrival of the pump pulse). The oscillatory pattern persists for more than 30 ps although its amplitude decreases with time. The Fourier transformed spectrum of the time-domain signal shown in Fig. 1 indicates that the observed coherent phonon mode is the Raman-active A_{1g} mode whose

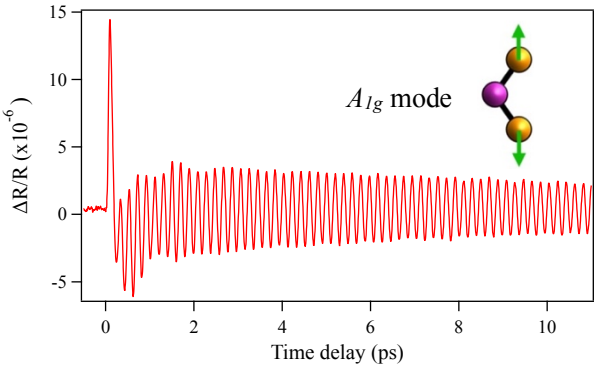

Fig. 1. (a) Time-domain $\Delta R/R$ signal of $MoTe_2$ measured with F = 50 μJ/cm^2. The schematic representation of the A_{1g} phonon mode is shown in the inset.

oscillation frequency is around 5.1 THz. With increasing F, the initial amplitude and the damping rate linearly increase.

Given the unusually long lifetime of the coherent A_{1g} phonon and the F-dependent trends, the contributions of the anharmonic phonon-phonon coupling as well as the presence of impurity and vacancy to the coherent phonon scattering are thought to be negligibly small because anharmonic phonon decay process is insensitive to the number of photo-excited carriers and the contribution of impurities and vacancies is expected to be quite small in the high-quality single crystal. On the other hand, electron-phonon coupling is thought to be the dominant factor of the coherent phonon scattering.

To obtain further insight into the coherent phonon dynamics, we performed a time-frequency-domain analysis. It was found that the transient frequency of the coherent A_{1g} mode is not constant but varies with time. In the early time stage up to ~500 fs, where the electronic temperature is much higher than the lattice temperature, the transient phonon frequency increases with increasing F. In general, phonon softening is induced by electron-phonon coupling as well as increase in lattice temperature rise. Therefore, when F is small, the coherent phono is softened through electron-phonon coupling until the photo-excited electrons relax. However, with increasing F, screening of electrons becomes significant and hence the magnitude of electron-phonon coupling is restricted. As a result, the phonon frequency is thought to increase with F.

References

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