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SELECTIVE COHERENT EXCITATION OF CHARGE DENSITY WAVES*

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Real time femtosecond pump-probe spectroscopy is used to study collective and single particle excitations in the charge density wave state of the quasi-1D metal, blue bronze. Along with the previously observed collective amplitudon excitation, the spectra show several additional coherent features. These additional resonances can be excited selectively by applying a sequence of pump pulses with intervals tuned to the period of the particular coherent excitation. A study of the pump power dependence shows a non-linear response of the amplitudon mode, in contrast to the linear power dependence of the single particle, phonon, and phason excitations, which is ascribed to the electron–amplitudon elastic scattering.

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1. Introduction

The instability of a one-dimensional Fermi surface to the formation of a charge density wave (CDW) engenders the appearance of two collective modes, related to the CDW order parameter $\Delta = |\Delta| \, \mathrm{e}^{i\phi}$. The phase mode, or phason, is associated with the sliding of CDW and has been extensively studied in transport and optical measurements [1]. The amplitude mode, or amplitudon, has received much less attention. In particular concerning the interaction between the amplitudon and single particle excitations. The

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amplitude mode is an oscillation of the single particle gap Δ. It involves an additional displacement of the underlying lattice, and its frequency ω_A is substantially smaller than the gap value, $\Delta/\hbar$, making it a well defined elementary excitation. The amplitudon has A_g symmetry and has been observed in Raman measurements [2]. Neutron experiments have shown that the phase and amplitude modes arise from a Kohn anomaly at wavevector $2k_{\mathrm{F}}$ [3]. In the vicinity of the phase transition they are coupled and substantially broadened. In the $T = 0$ limit these modes become uncoupled and well defined, with small broadening and ω_A close to its mean-field value [1].

Here we report on femtosecond pump-probe measurements of the amplitude oscillation in the quasi-1D metal $K_{0.3}MoO_3$, which undergoes the CDW transition at 183 K. IR laser pulses can couple to Raman active excitations through transient stimulated Raman scattering [4]. In comparison to previous experiments [5] we apply 10^4–10^5 more peak intensity, using the same average pump power. With this high power we reach a much higher level of population of amplitude and single particle excitations, allowing for a more detailed study of their interactions.

2. Experimental

A regenerative Ti:sapphire amplifier seeded with mode-locked Ti:sapphire laser was used to generate 130 fs pulses at 800 nm (rep. rate 1 kHz). The pump power was varied between 7 μW and 7 mW with a spot size of 100 μm, and a polarization parallel to the chains. The probe power was kept below 2 μW. The sample was placed in a He flow cryostat, which allows to vary the temperature between 10 and 300 K. In the experiment we excited first the system with the strong pump pulse or train of pulses and measured the real-time evolution of the reflectivity with the delayed probe pulse.

3. Results and discussion

Fig. 1 shows the transient reflectivity for different levels of pump power at $T = 40$ K. At each power level the response can be decomposed into six constituents. Three components have purely exponential decay with the time constants approximately 0.3 ps, 2 ps, and a long lasting process with $\tau \sim$ ns. The other three components are damped oscillations with frequencies 1.7, 2.2, and 2.5 THz (56, 74, and 85 cm^{-1}). All three resonances can be clearly seen in the Fourier spectrum $F(\nu)$ in Fig. 2. The first resonance was identified as an amplitudon mode [5]. We ascribe the last two modes to the Raman active phonons, appearing due to the CDW deformations of the lattice. The insert in Fig. 1 depicts the dependence of the amplitudon frequency ν_A and damping Γ_A upon the pump power. The absolute values

of ν_A and Γ_A agree very well with previous Raman [2, 6] and pump-probe experiments [5].

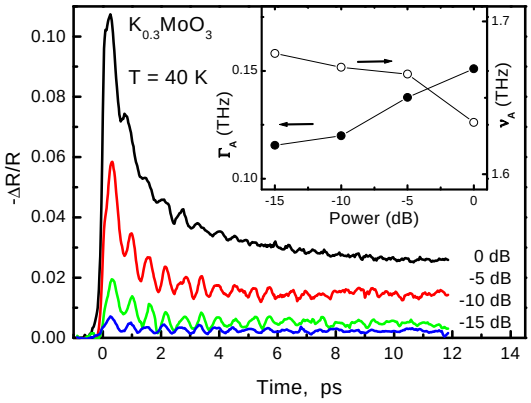

Fig. 1. Transient reflectivity for various pump powers. Insert: Amplitudon frequency ν_A and damping Γ_A as a function of pulse power.

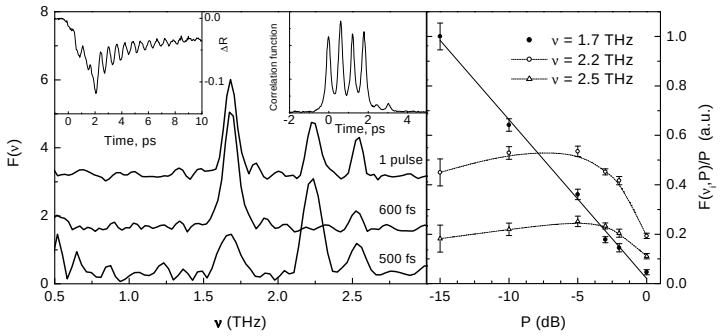

Fig. 2. Left panel: Fourier spectrum of the transient response for the single pulse pump excitation (A), and the four pulse trains for two different pulse separations. Left insert: transient reflectivity, induced by four-pulse trains. Right insert: cross-correlation of the pump pulse train with the probe. Right panel: Normalized pump-power dependence of the amplitudon (1.7 THz) and two Raman phonons (2.2 THz and 2.5 THz).

Due to the non-linear power dependence of the amplitudon, discussed below, the Raman modes become well resolved at relatively high power only. This is probably the reason why they were not reported previously [5]. Another method to increase the sensitivity to particular modes is the use of a pulse train with a spacing tuned to the period of the mode. A typical pump-probe cross-correlation function of such a pulse train is shown in the

right insert of Fig. 2. The top curve in the left panel of Fig. 2 shows a Fourier spectrum of a single pulse response at high power. When the train repetition rate is tuned to a resonance, we can suppress the other components as demonstrated by the 600 fs spectrum (*i.e.* tuned to ν_A, the transient reflectivity for this experiment is shown in the left inset).

Power dependent experiments were done with an intermediate tuning (500 fs) between the phonons and the amplitudon. From the Fig. 1 one can see that the oscillatory component, mainly determined by the amplitude mode, initially increases with the pump power, reaches its maximum at -5 dB, and falls back at higher power. The amplitude of the 1.7 THz resonance normalized to the pump power, $F(\nu_A)/P$ is plotted *vs* the pump power in the right panel of Fig. 2 together with the other two resonances. As can be seen, $F(2.2\mathrm{THz})/P$ and $F(2.5\mathrm{THz})/P$ remain fairly constant over a wide range of pump power. In contrast, $F(\nu_A)/P$ shows a strong P-dependence, which results in a highly non-linear relation $F(\nu_A) \sim P \log P_0/P$, where P_0 is a constant.

It is unlikely that the observed effects are due to heating. Because ν_A and especially Γ_A are strongly temperature dependent, they can serve as a measure of heating. From a comparison of the data in Fig. 1 to Raman and pump-probe data [2, 5] we conclude that the 4 % change in ν_A and 30 % change in Γ_A observed in the present experiments would account for no more than 20 K change in temperature. And thus, the temperature of the sample remains well below the transition temperature 183 K and well below the mean-field limit of the decoupling of the amplitude and phase ~ 100 K [3].

The most probable candidate for the observed strong decrease of the amplitudon response is the quasiparticle-amplitudon interaction. Quasiparticles (QP) excited by the pump pulse quickly relax to the single particle gap at $k \sim k_\mathrm{F}$, where they strongly scatter from the $2k_\mathrm{F}$ periodic potential. Because the scattering occurs exactly by $2k_\mathrm{F}$, the QP energy is unchanged and the scattering is inevitably elastic. This type of scattering would lead to a dephasing of the amplitudon excitations, consistent with the observed weak broadening of the amplitudon response as a function of the pump power.

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