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Disentanglement of the electronic and lattice parts of the order parameter in a 1D Charge Density Wave system probed by femtosecond spectroscopy

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Charge density waves (CDW) are metallic systems with a broken symmetry (translational) ground state, showing several unique properties like a gap in the density of states at the Fermi level, a periodic lattice modulation and a modulation of the charge density of the electronic subsystem.

One of the prototype materials of this class is blue bronze ($K_{0.3}MoO_3$) which has been extensively studies with various techniques. [1-6] A new attempt to get a deeper insight to the underlying physics has been started with the help of femtosecond spectroscopy.[7] This technique was demonstrated to be sensitive to the collective phenomena (e.g. amplitude mode) which are direct fingerprints of the charge density wave phase. It uses an intense femtosecond laser pulse to excite the electronic subsystem to a higher energy level. With a weak laser probe pulse one can track the subsequent relaxation processes which bring the system back to its ground state.

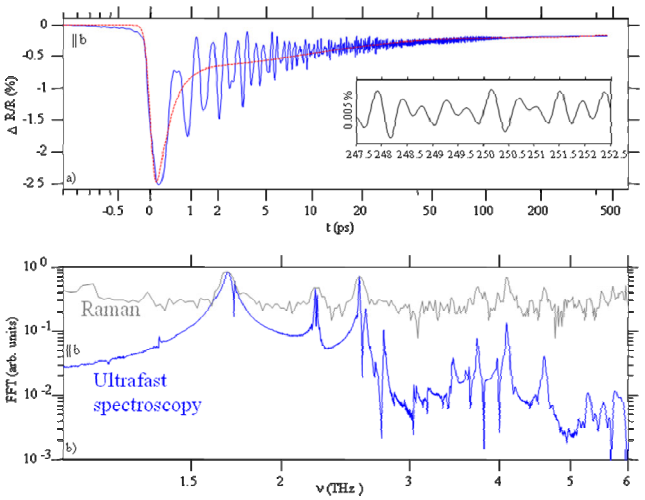

Figure 1. Panel a) shows the transient change in reflectivity after excitation with a 50 fs laser pulse of 800 nm central wavelength. The inset shows the magnified signal 250 ps after excitation which still shows strong oscillations. In b) the oscillatory signal has been Fourier transformed (blue). The data is compared to the recent Raman data (grey).[6]

In Figure 1 a) a typical Signal for the transient change in reflectivity after the excitation of blue bronze with an intense femtosecond laser pulse is shown. The signal can be decomposed into two parts: The incoherent response which shows at least two time scales with exponential behavior. Upon this signal one can observe a lot of different (phonon) modes which are excited coherently.

After a Fourier transformation of the oscillatory data, the frequencies of the oscillating modes can be obtained. A comparison to the actual Raman data set [6] shows the much higher signal sensitivity of this technique in the energy range around 1 THz (33 cm^{-1} / 4 meV).
This allowed us to study the temperature dependence of the excitation spectrum with unprecedented precision. The result is shown in Figure 2.

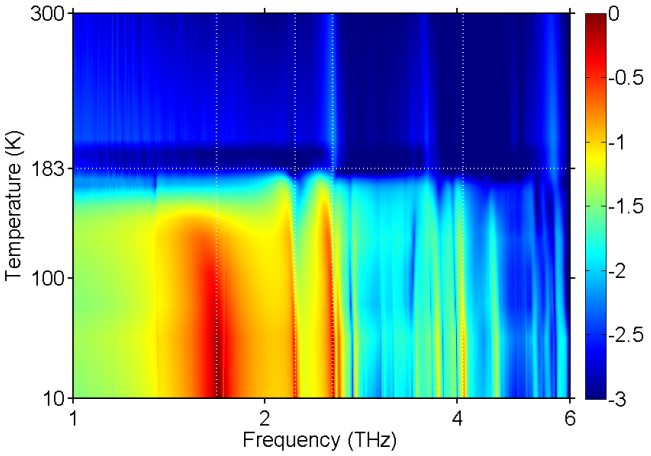

Figure 2. The temperature dependent spectrum of blue bronze. The phase transition at 183 K appears clearly in the data: Nearly all modes visible in the CDW state disappear above T_c. Additionally a softening of several modes (e.g. 1.68 THz, 2.22 THz, 2.55 THz, and 4.09 THz) is observed when T_c is approached.

The phase transition is clearly observed and the mode spectrum can therefore serve as a fingerprint of the CDW state. An interesting new feature is also observed: Several modes show softening when T_c is approached. This is in strong contrast to former interpretations, where only one mode (the so called amplitude mode) was meant to show pronounced softening.

The time dependent Ginzburg-Landau model

This observations as well as the conclusions from our previous work, [8] where we studied the photoinduced melting of the CDW state which suggested, that electronic

and lattice order are separated on ultrashort time scales, motivated us to develop a new model.[9]

The Time Dependent Ginzburg-Landau model (TDGL), which has been developed to described the observed phenomena, starts with the free energy described by

Equation 1.
$$\varphi = \varphi_0 + \frac{\alpha}{2}(T - T_{c0})\left(\Delta_{(r)}^{\ 2} + \Delta_{(i)}^{\ 2}\right) + \frac{\beta}{4}\left(\Delta_{(r)}^{\ 2} + \Delta_{(i)}^{\ 2}\right)^2 + \sum_k \frac{\Omega_k^{\ 2}}{2}\left(\xi_{(r),k}^{\ 2} + \xi_{(i),k}^{\ 2}\right) - m_k\left(\Delta_{(r)}\xi_{(r),k} + \Delta_{(i)}\xi_{(i),k}\right)$$

Where φ_0 is the free energy of the normal state and α,β are the standard Ginzburg-Landau parameters. T is temperature, Δ a complex order parameter and T_{c0} the electron phase transition temperature. The first line of the equation's right side describes the pure electronic phase transition, where the absolute value of Δ describes the amplitude of the charge density wave. When neglecting the lattice (which is represented by the second line) this already describes a 2nd order phase transition of the electronic subsystem.

The second line includes the lattice part of the energy which consists of a sum over all phonon modes k that couple linearly to the order parameter. The first term in the sum represents the energy that is needed to excite the modes with the displacement amplitude, which is given by ξ. Ω is the frequency of the corresponding phonon mode. The second term takes into account the linear coupling between lattice order and electronic order with the coupling constant m. For more details on the model see [9].

Finally this model can describe the electronic as well as the lattice part of the dynamics after the excitation. Therefore the measured data could be well fitted with the model as we show in Figure 3.

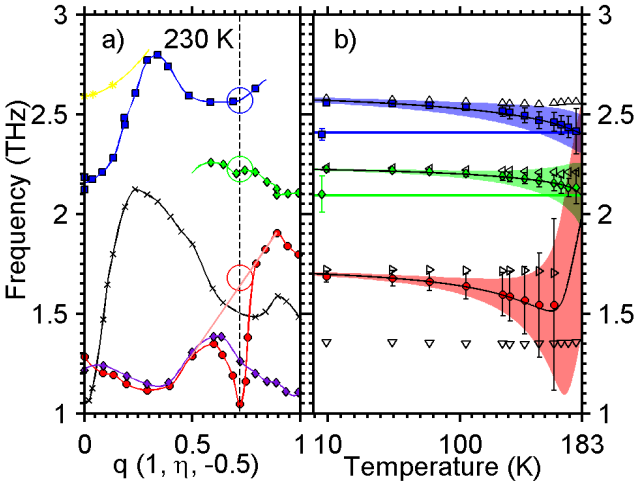

Figure 3. a) shows the neutron scattering data [4] which shows the phonon branches that cross the CDW modulation vector (dashed line) at (1, 0.748 , -0.5). In b) three of the measured 'soft' modes (red, green and blue symbols) are depicted and the model solution is superimposed as shaded areas with the same color. Four modes that do not soften are also shown. (Triangles). Straight lines (green and blue) correspond to infrared active modes expected from the model, the two data points at 6K are taken from [1].

The experimental data is shown as colored symbols in Panel b) of Figure 3. The symbols represent the frequencies of the measured modes. At low temperatures they correspond quite well to the phonon modes at the CDW modulation vector as observed with neutron scattering [4]. The measured damping of the modes is represented by the bars that cramp the symbols. The model solution is given by the black lines and the shaded areas, where the former depicts the frequency and the latter the damping.

As can be concluded from the model solution, none of the modes shows true soft mode behavior. This is because the coupling of lattice and electronic modulation is far from being adiabatic. The model solution identifies the 'electronic' amplitude mode as an overdamped mode with critical behavior as T_c is approached. [9] This is shown in Figure 4 a) where the critical behavior of the fast relaxation time is shown in comparison to the model solution (red dashed line in the inset).

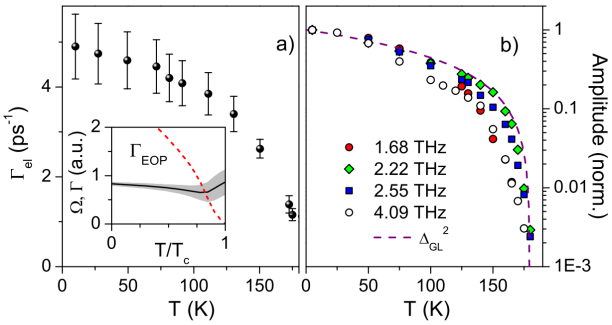

Figure 4. a) Critical behavior of the fast relaxation time scale which is attributed to the electronic order. Inset: model solution (red dahed) b) Amplitudes of the 'soft' modes extracted from the data and compared to a Δ^2 function.

Another interesting quantity that can be extracted from the data are the mode amplitudes which show all decreasing behavior with increasing temperature and are critical with respect to T_c. This can also be explained within the model solution: If the properties of the dielectric function (ε) are analyzed properly it turns out, that the mode intensities should follow Δ^2 behavior, as demonstrated.[9]

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