Ordering and low energy excitations in strongly correlated bronzes
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Chapter 6

Summary

In any solid system, whether it is superconducting, shows a charge-density-wave behavior, or any other kind of ground state, two aspects drag the attention of the scientific community. They are order and excitations in solids. The ordering may be due to electronic, lattice, spin or orbital degrees of freedom or even due to a coupling between these. The total energy of a ground state is not often a very important quantity; rather what is important is the behavior of the lower excited states or low-energy excitations relative to the ground state. Thus in most of the experiments presented in this thesis we do not address the ground state itself but rather the behavior of the excited states and in particular the low-energy excitations. These low energy states can be excited at relatively low temperatures or by external fields. There are two qualitatively different kinds of elementary excitations, the conventional and relatively well understood are quasi-particles. They closely resemble "single-electrons", however, they are relatively heavier due to their interaction with other similar excitations in solids. The other kind is, the collective excitation, which includes plasmons, phonons, excitons etc. Collective excitations can be defined as "quantized excitations due to the co-operative interaction of quasi-particles". The collective excitations in metals for instance, are plasmons, which are nothing but the quantized oscillation of electrons. The word collective implies that the excitations act as if they characterize a single mode. In the case of charge-density-wave ground state, there are two collective modes, viz., phasons and amplitudons (AM). In the present study we focused on two aspects, one: ordering in strongly correlated bronzes and, two: low energy excitations in such ordered systems. The uniqueness of the present study is that it addresses three "states" of the collective excitation; amplitudon. They are, origin, evolution and decay/dephasing of the coherent amplitudons.
In chapter two we studied the charge-density wave $K_{0.3}MoO_3$ using ultra-fast time-resolved optical spectroscopy to address the dynamical aspects of various excitations like coherent amplitudons, coherent phonons and quasi-particles and eventually their mutual interaction dynamics. At relatively large intensity of the excitation pulse (or the pump-pulse) the transient reflectivity response has contributions from the photoexcited quasi-particles, coherent amplitudons, coherent phonons and other incoherent excitations like phonons. Using intensity-dependent and wavelength-dependent ‘pump-probe’ experiments we determined the decay/dephasing time of these excitations. Our study showed that the coherent amplitudons are generated by the ultrafast-decay of the photoexcited quasi-particles. The intensity of the coherent amplitudons (peak to peak) follows the quasi-particle intensity. The coherent amplitudons are de-phased when the decay time of the photoexcited quasi-particles is larger than the period of the coherent-amplitudon. On the other hand, the attempt to achieve an optically-induced phase-transition did not give positive results. Also, our study clearly disproved the previous claim that “the long living transient reflectivity component ($\approx 1$ ns) is due to de-pinned phasons”. This may trigger researchers to conduct time-resolved pump-probe experiments on other similar CDW materials. The two coherent phonons (2.25 and 2.5 THz) are also observed.

In another case [chapter three] we carried out a comparative Raman study of the CDW materials, $K_{0.3}MoO_3$ and $Rb_{0.3}MoO_3$. The frequency of the AM in $Rb_{0.3}MoO_3$ is 3 cm$^{-1}$ smaller than in $K_{0.3}MoO_3$ which is consistent with the fact that the Rb atom is heavier than the K. A detailed temperature-dependent study revealed the interaction of the low energy excitations such as amplitudons, phonons and possibly phase-phonons. The striking feature is the anomalous temperature dependence of the intensity of the AM. We showed that this is due to the presence of strong 1D-Peierls fluctuations near $T_{CDW}$. The decay of the low energy phonons are influenced by the presence of the charge-density-wave gap. Lower energy phonons live longer due to the unavailability of the electron-phonon scattering decay channel whereas the higher energy phonons decay faster. While an incommensurate to commensurate transition is observed in $K_{0.3}MoO_3$ (as a jump in the frequency of the phonons at 100 K), in $Rb_{0.3}MoO_3$ we did not observe it.

The infrared spectroscopy on pure and doped $K_{0.3}MoO_3$ [see chapter four] showed that there are no Machida-type midgap state inside the main Peierls gap in $K_{0.3}MoO_3$, which is contrary to the previous studies. The concept of phase-phonons is revisited and the discussions ultimately leads to the conclusion that “Not all the phonons which appear along the chain-direction are phase-phonons”.

Chapter 6. Summary

The doping of W affect the CDW state since it acts as an impurity which modifies the pinning potential in the CDW state. Finally, another class of bronzes, K$_{0.3}$WO$_3$, are studied in chapter five. In the study we learnt that there are at least two local-structural-excitations (LSEs) among which the system hops between. The order-disorder transition is driven by the LSEs. A second order phase-transition is also observed, which is possibly due to the Jahn-Teller distortion. A detailed composition and temperature dependent study of K$_{0.3}$WO$_3$ may reveal the dependence of LSEs on stoichiometry at a given temperature and for a given stoichiometry it’s dependence on temperature. Thus, in the present study we have addressed ordering and low energy excitations, quasi-particles and collective excitations, in strongly correlated bronzes. Both kinds of bronzes, M$_x$WO$_3$ (M = K, Rb) and A$_{0.3}$MoO$_3$ (A = K, Rb) show metal-insulator transitions through different mechanisms. In the A$_{0.3}$MoO$_3$ case, it is through the charge-ordering driven by Peierls instability whereas in the M$_x$WO$_3$ case it is through lattice ordering driven by local-structural-excitations.