Mapping the ultrafast dynamics of the orbital-order order-parameter with transient anisotropy spectroscopy

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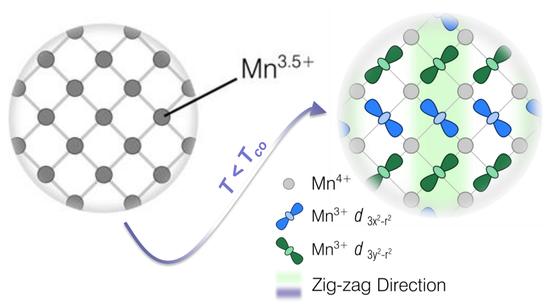
ABSTRACT

Doped manganites are an important class of correlated materials. In these systems strong coupling of spin, charge and orbital degrees of freedom leads to a complex phase diagram and exotic properties. Tracking the evolution of these complex phases after an ultrafast quench has proved a valuable approach for understanding non-equilibrium physics in correlated materials. Here we introduce a novel all-optical setup for measuring the ultrafast dynamics of photoexcited $La_{1.5}Sr_{0.5}MnO_4$ (LSMO) through changes in the in-plane optical anisotropy.

BACKGROUND

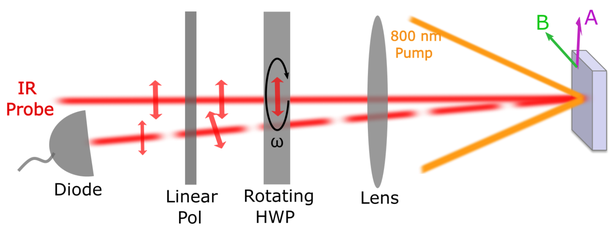


The single–layered manganite $La_{1.5}Sr_{0.5}MnO_4$(LSMO) undergoes a charge and orbital order (CO/OO) transition below T_{co} = 220 K, distortion within its MnO planes coupled to a oxygen ochtaedra distortion[1]

In the ordered state two optical axis are present, the degree of anisotropy is proportional to the order parameter of the CO/OO phase.

This opens the possibility for all-optical direct measurement of the order parameter which has previously only been studied indirectly through x-ray methods[2,3]

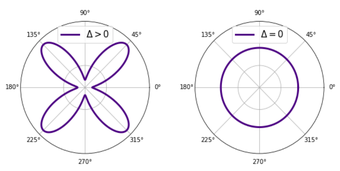
SETUP

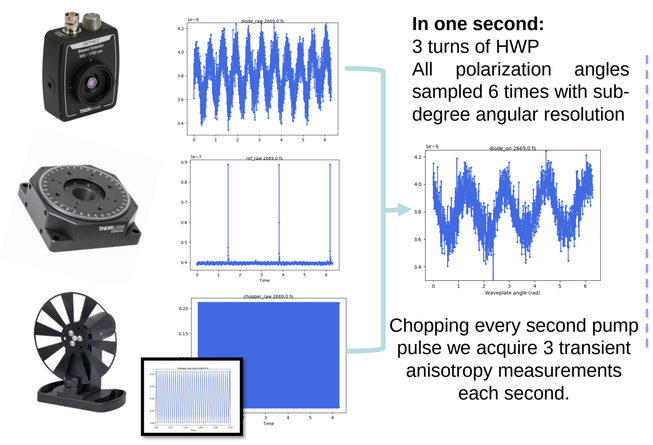


We use a pump-probe setup, modified to include a half waveplate (HWP) rotating at angular speed ω which gives us complete angular resolution at each time delay with high signal to noise on a pulse-to-pulse setting. The intensity I measured at lab time t is given by the Jones formalism:

$$I = \left[R + \frac{\Delta}{4}(1 - cos(4\omega t + \theta))\right]^2 \quad (1)$$

where the reflectivity along each optical axis is given as $r_1 = R$ and $r_2 = R + \Delta$. R and Δ are the average reflectivity and optical anisotropy of the sample, respectively. In our setup, optical anisotropy yields a characteristic 4-lobe pattern in the intensity vs. HWP angle, as shown in the figure below:

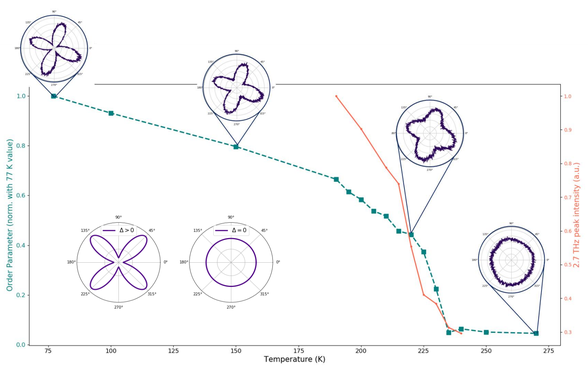




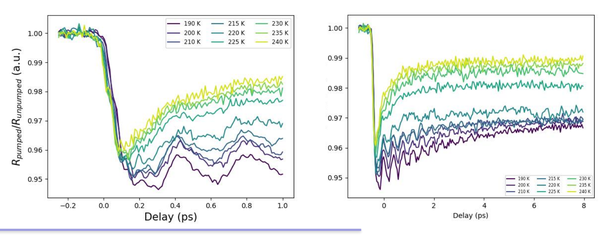
In one second:
3 turns of HWP
All polarization angles sampled 6 times with sub-degree angular resolution

Chopping every second pump pulse we acquire 3 transient anisotropy measurements each second.

STATIC MEASUREMENTS



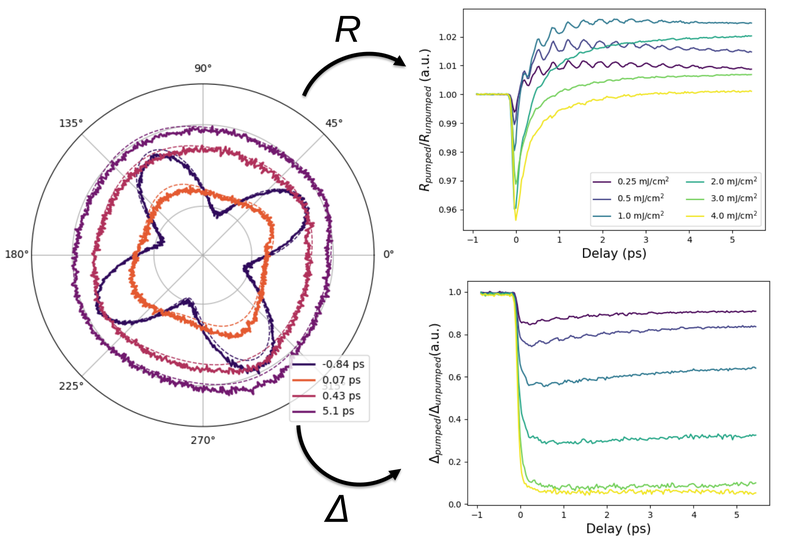
Below: Temperature dependence of LSMO reflectivity upon low-fluence photoexcitation with 15 fs duration, 1800nm pump laser. 12 fs long probe pulses at 650 nm with fixed polarization show the characteristic 2.7, 6 and 14 THz phonon, which disappear at T_{co} 225 K).



The figure above shows the evolution of the optical anisotropy in LSMO with temperature, probed with 1200 nm light. When heated above Tco = 225 K, the Δ parameter is suppressed and the 4-lobe pattern disappears.

The red line shows the temperature dependence of the intensity of the 2.7 Hz phonon mode. We can observe that it disappears together with the optical anisotropy

DYNAMICS: RESULTS



Time and polarization resolved measurement. (1200nm probe, 800 nm pump)

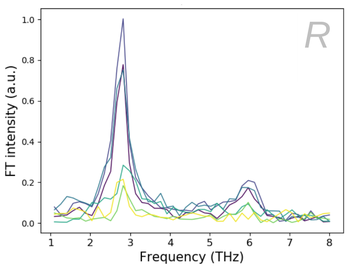
Fitting equation (1) at each delay we extract R (upper ploy) and Δ (lower plot). The dynamical response of the parameter is found to be highly different.

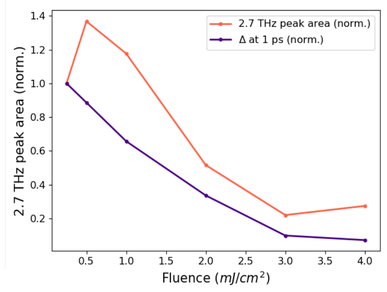
The polar plot shows anisotropy patterns corresponding to a selection of pump-probe delays for a fluence of 4 mJ/cm2, enough to erase the optical anisotropy.

Below: optical anisotropy Δ and the strength of the 2.7 THz phonon mode (extracted from R) normalized to the value at minimum fluence.

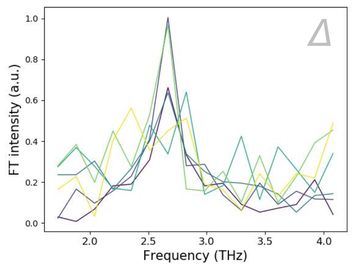
Both parameters are suppressed with photoexcitation intensity by a similar factor (80%) and at similar rate.

From the isotropic component, R, we can identify how the 2.7 and 6.0 THz phonon modes are strongly suppressed with increasing photoexcitation strength.





The 2.7 THz mode can also be observed in the anistotropic component, Δ. for lower fluences. This suggest that the phonon mode could be modulating the orbital order.



CONCLUSIONS

- Our all-optical setup can succesfully track the ultrafast response of the orbital-order order parameter.
- Full polarization resolution allows the decoupling of isotropic and anisotropic components in a charge-ordered material.
- These components show different dynamics and the characteristic ordered-pase 2.7 THz appears to modulate the charge-ordering at low fluences.

REFERENCES

[1] J. Herrero et al., Phys. Rev. B 83, 184101 (2011).

[2] P. Beaud et al., Nature Materials, 13 0923-927 (2014).

[3] M. Porer et al., Physical Review B, 101 075119 (2020).

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