Electronic Phase Control on the Femtosecond Timescale

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LETTERS

Control of the electronic phase of a manganite by mode-selective vibrational excitation

Matteo Rini[1], Ra'anan Tobey[2], Nicky Dean[2], Jiro Itatani[1,3], Yasuhide Tomioka[4], Yoshinori Tokura[4,5], Robert W. Schoenlein[1] & Andrea Cavalleri[2,6]

Paper Summary by Nathan Traynor

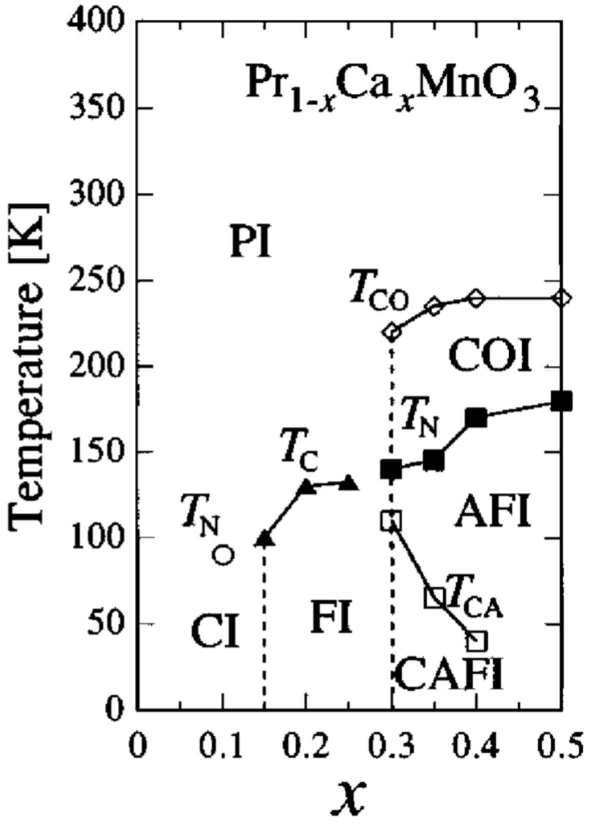
Department of Physics and Astronomy

The University of Tennessee-Knoxville

Overview

- $Pr_{1-x}Ca_xMnO_3$- General properties and fundamental physics
- Pump-Probe optical experiments
- Ultrafast (<300 fs) insulator-metal transition (IMT) in $Pr_{0.3}Ca_{0.7}MnO_3$
- Technology and fundamental science applications

$Pr_{1-x}Ca_xMnO_3$: A Prototypical Example of a Strongly Correlated Svstem



Rich phase diagram from strong correlations and *robustly insulating*

Figures from: Y. Tomikoa *et al.*, Phys. Rev. B **53** 1689

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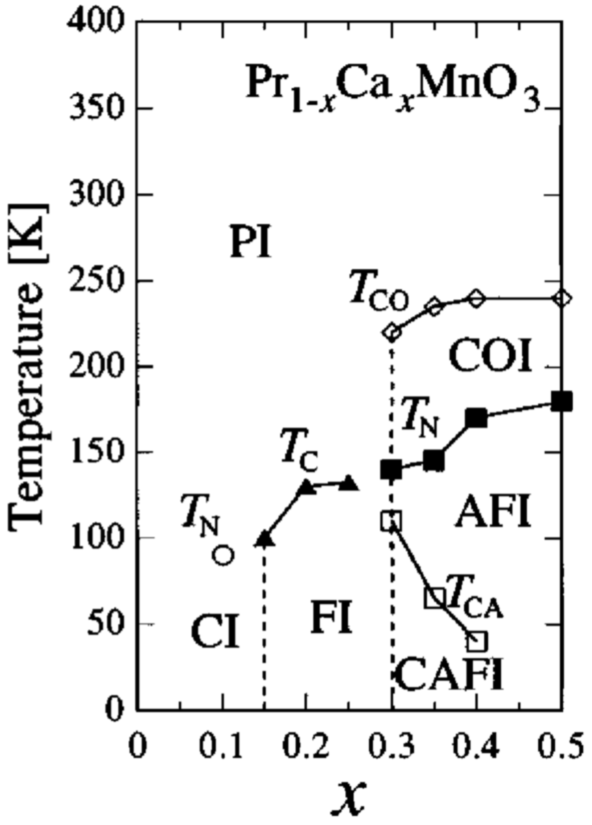

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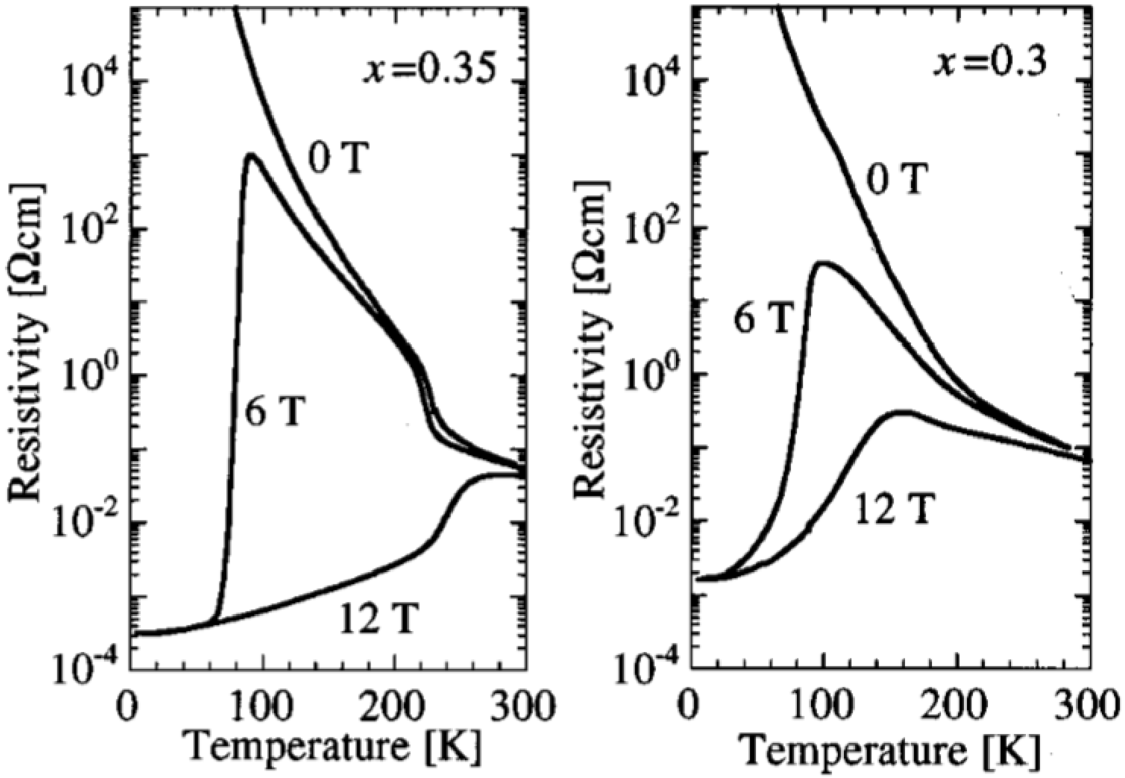

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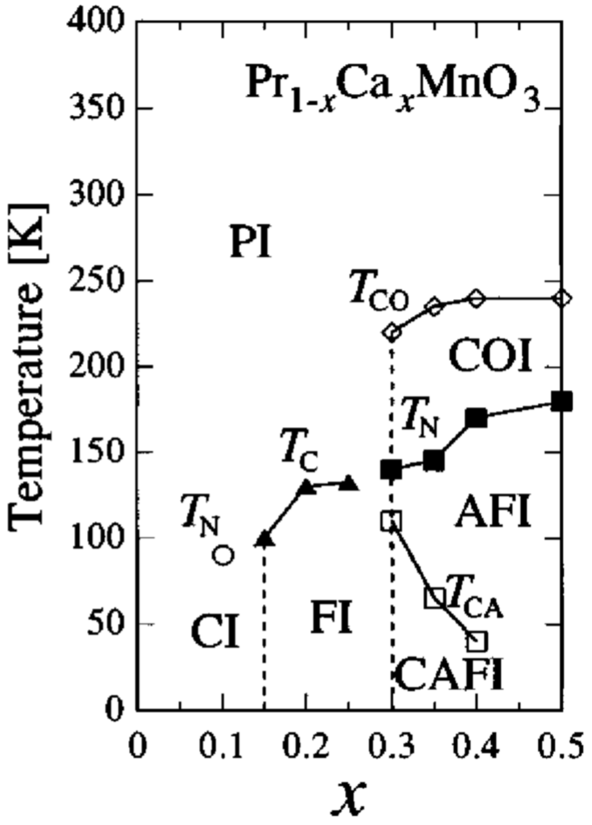
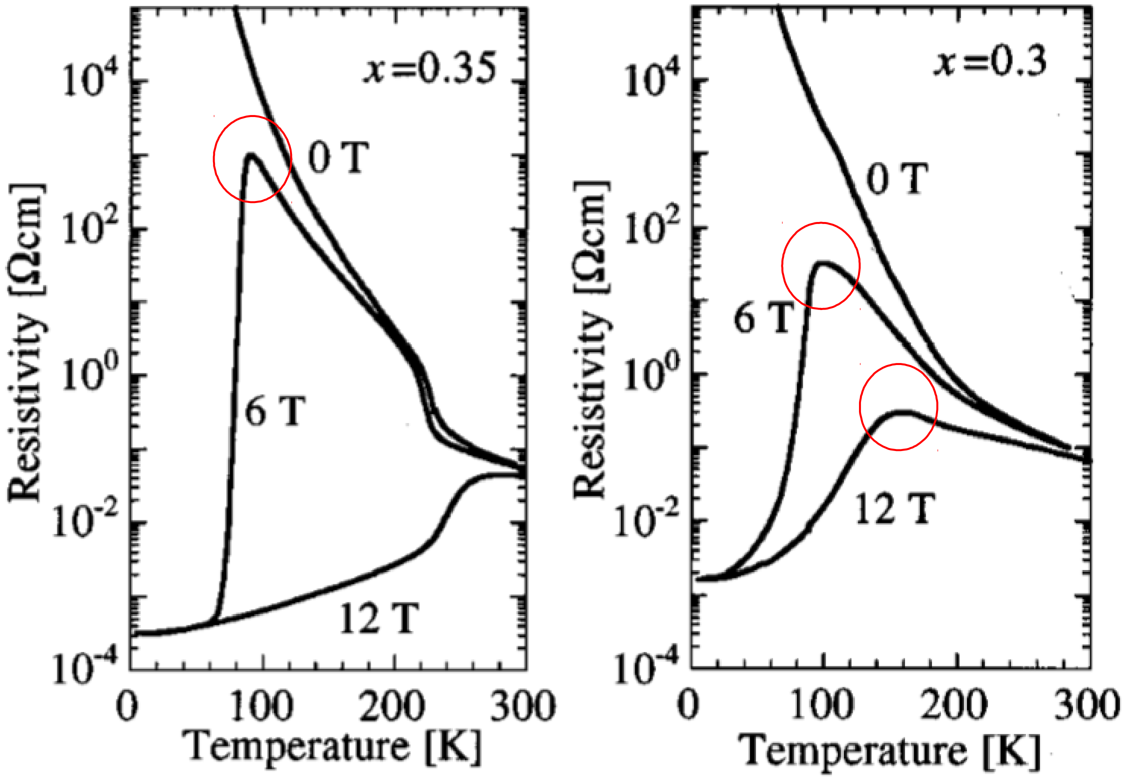


Rich phase diagram from strong correlations and robustly insulating

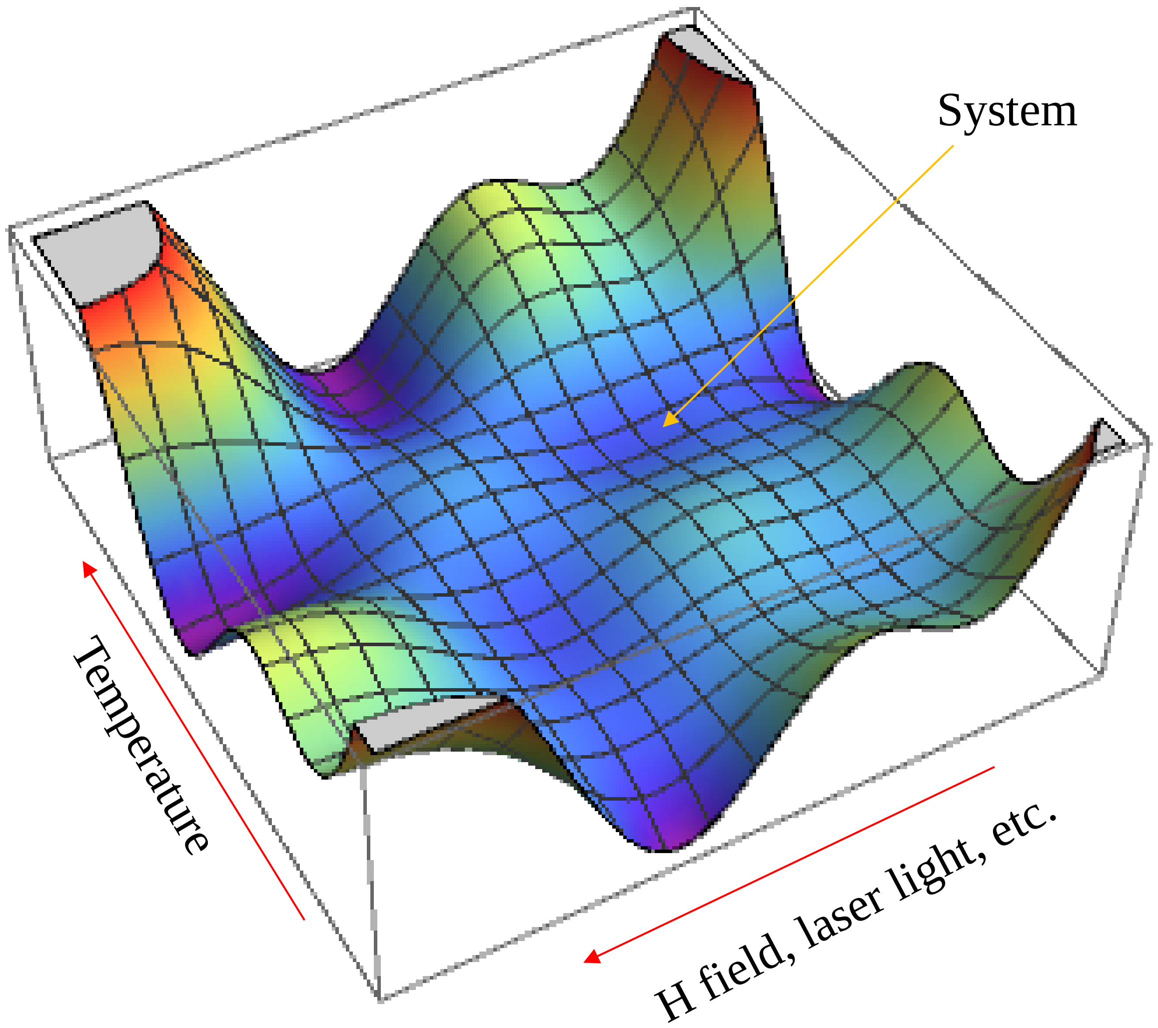


Magnetic field induced insulator-metal transitions. *Hidden phases!*

Figures from: Y. Tomikoa *et al.*, Phys. Rev. B **53** 1689

Strongly Correlated Systems- A Delicate Balancing Act

Complicated Potential Landscape



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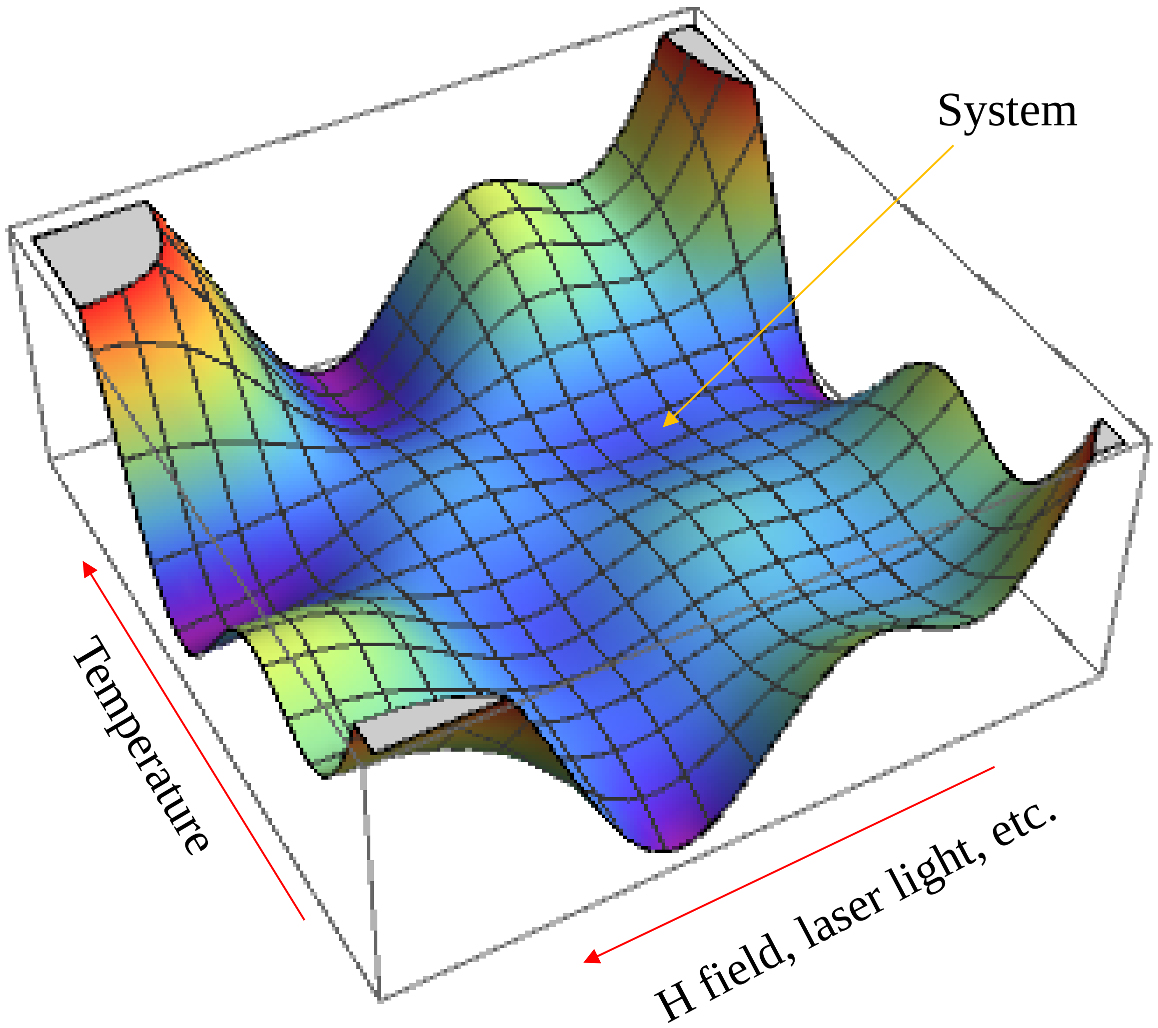
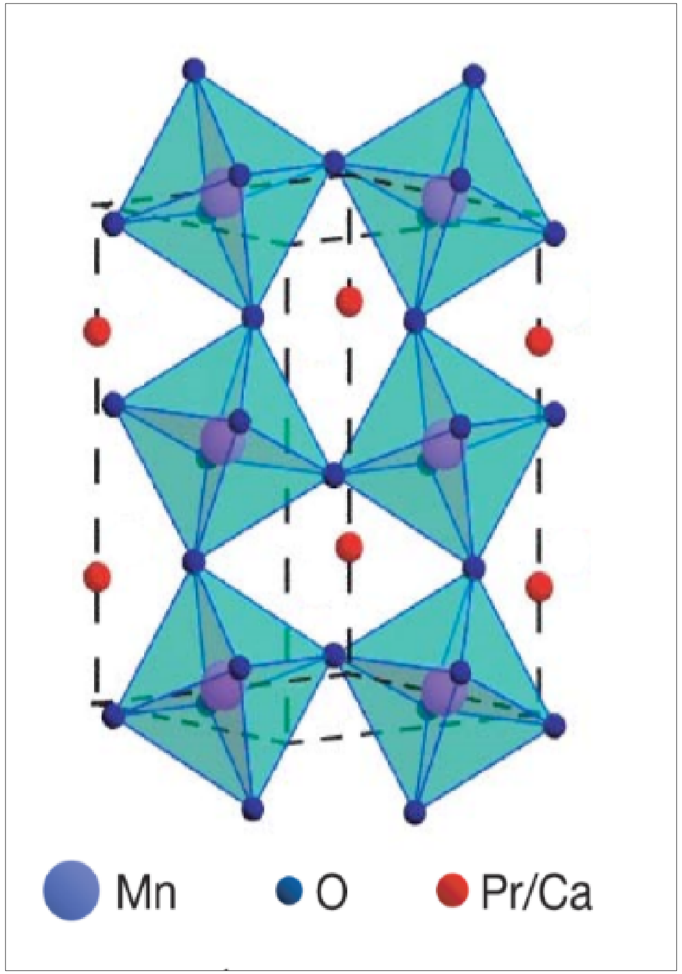


-Small perturbations=large material property changes

-Some "hidden" states are not accessible by changing only temperature

-Complex potential landscape yields rich functionalities

$Pr_{1-x}Ca_xMnO_3$: Structure and Tolerance Factor



M. Rini *et al*., Nature **449**, 72 (2007).

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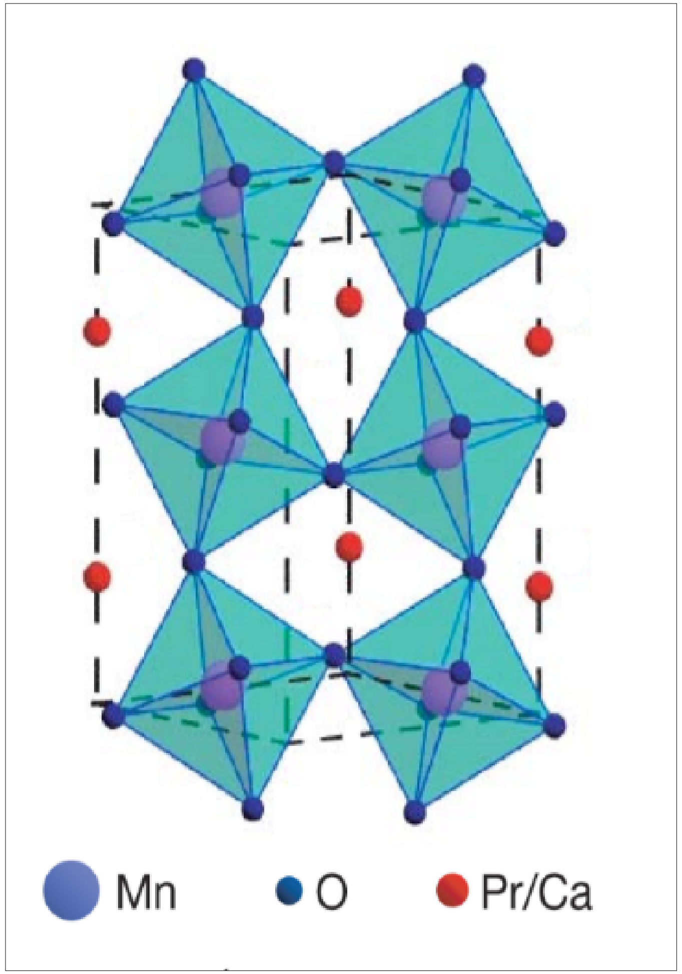

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$$\Gamma = \frac{r_{AO}}{\sqrt{2}r_{BO}}$$

r_{AO} - Average Pr/Ca-O distance

r_{BO} - Average Mn-O distance

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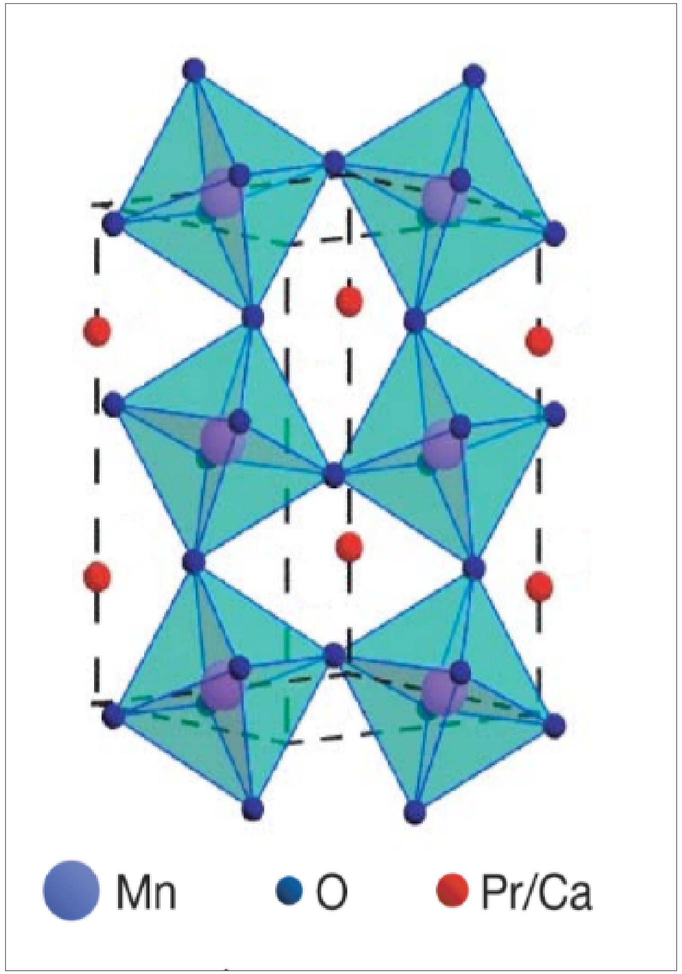

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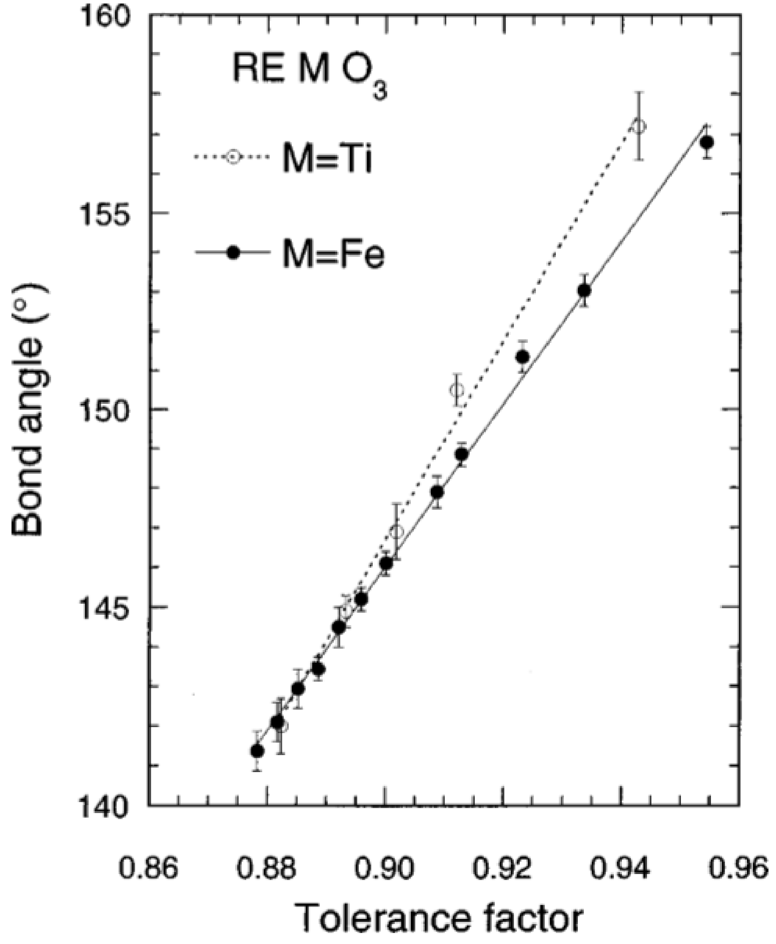
$$\Gamma = \frac{r_{AO}}{\sqrt{2} r_{BO}}$$

r_{AO} - Average Pr/Ca-O distance

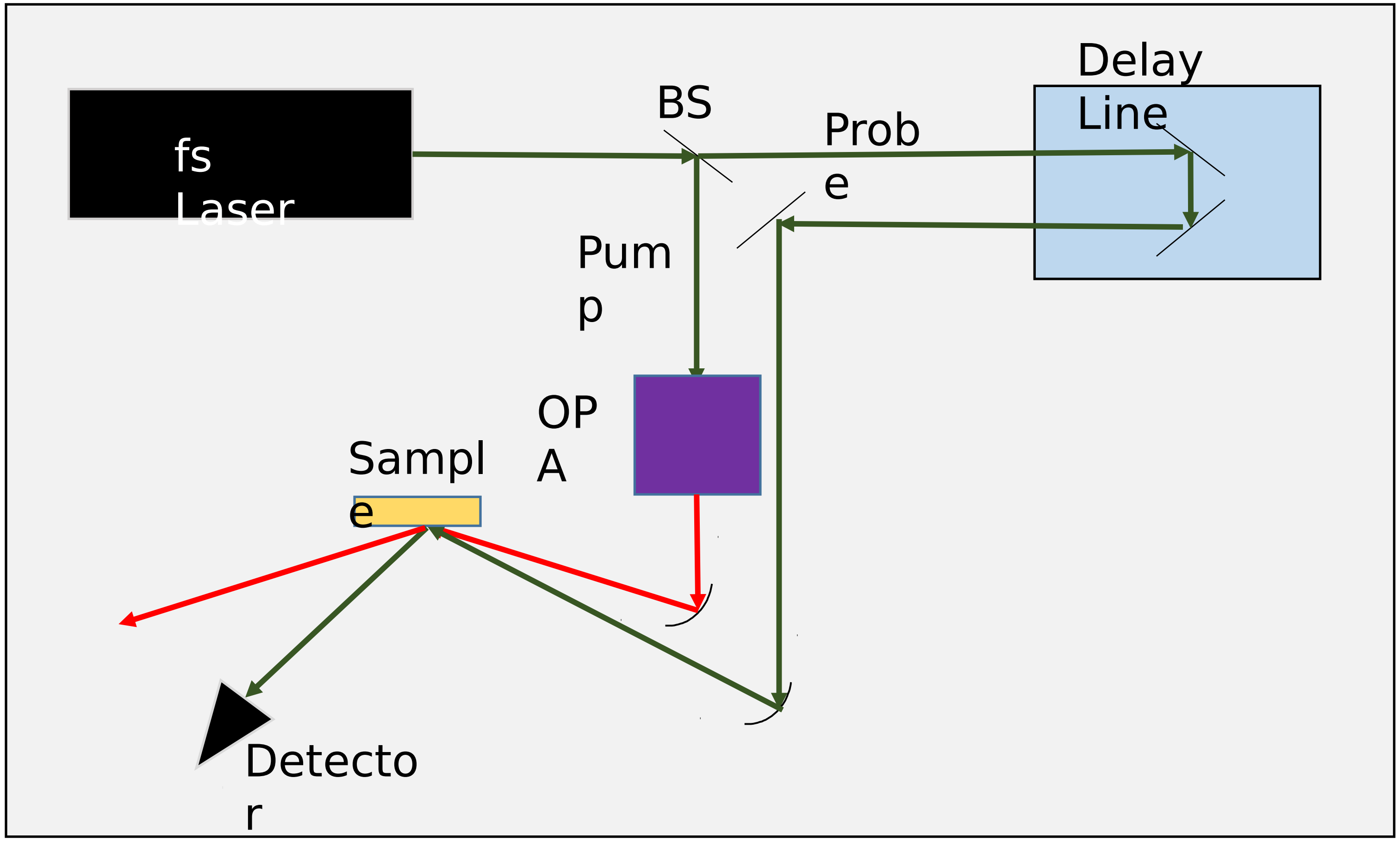
r_{BO} - Average Mn-O distance

Tolerance factor quantifies the orthorhombic distortion and Mn-O-Mn bond angle.



M. Imada *et al.*, Rev. Mod. Phys. **70** 1039 (2004).

Experiment: M. Rini *et al.*, Nature **449**, 72 (2007).



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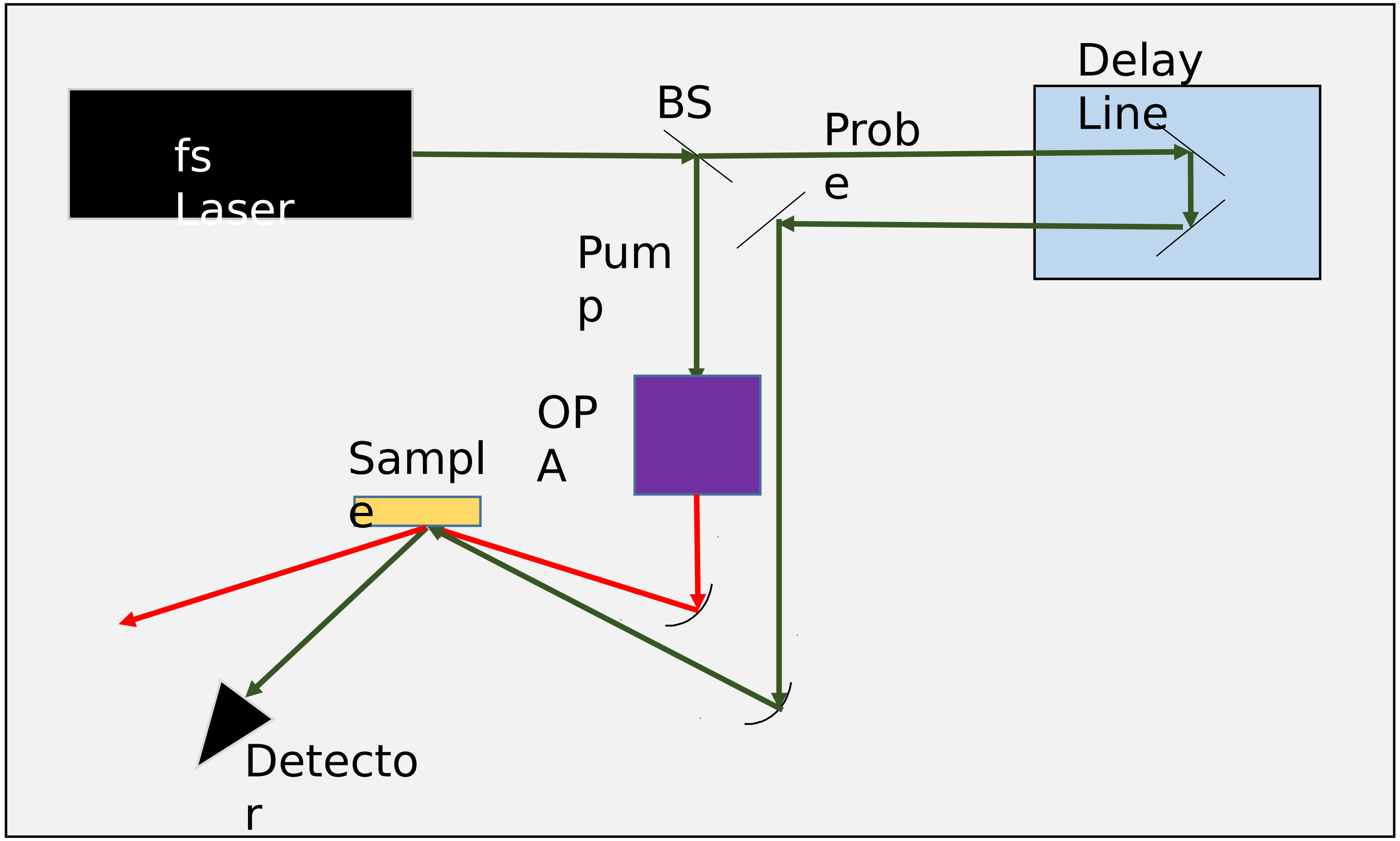


-Pump and probe pulses are ~200fs long

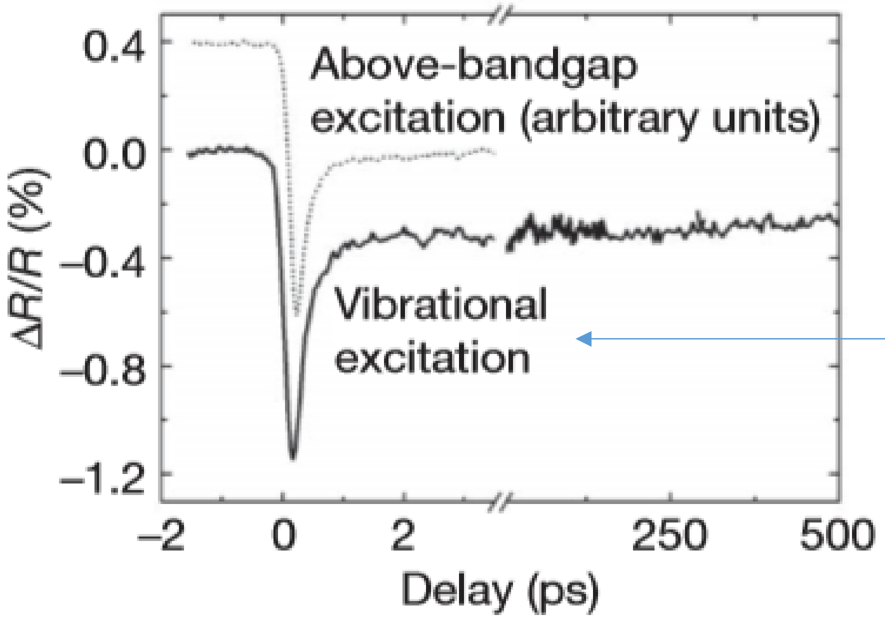
-Many electronic processes, such as the recombination of electron-hole pairs in Na_2IrO_3[1], happen within ~200fs ($1fs=10^{-15}s$)

-Measure change in reflectivity at different probe delay

-Electrodes on the sample can measure conductivity after pump pulse

[1] M Gandolfi *et al.*, Phys. Scr. **92** 1 (2017)

A insulator-metal transition in $Pr_{0.3}Ca_{0.7}MnO_3$ driven by vibrational excitation



At phonon resonance, below band gap

Figures from: M. Rini *et al*., Nature **449**, 72

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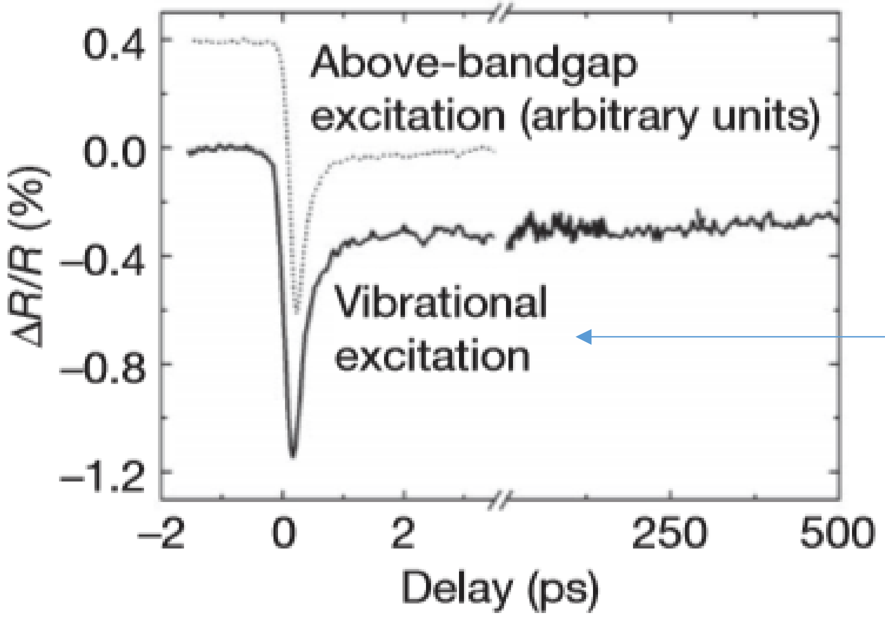

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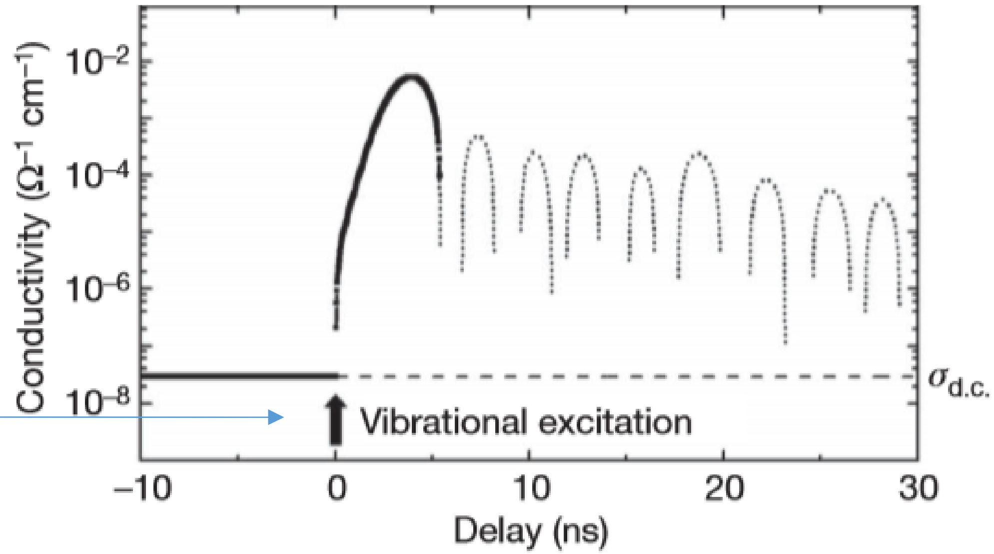

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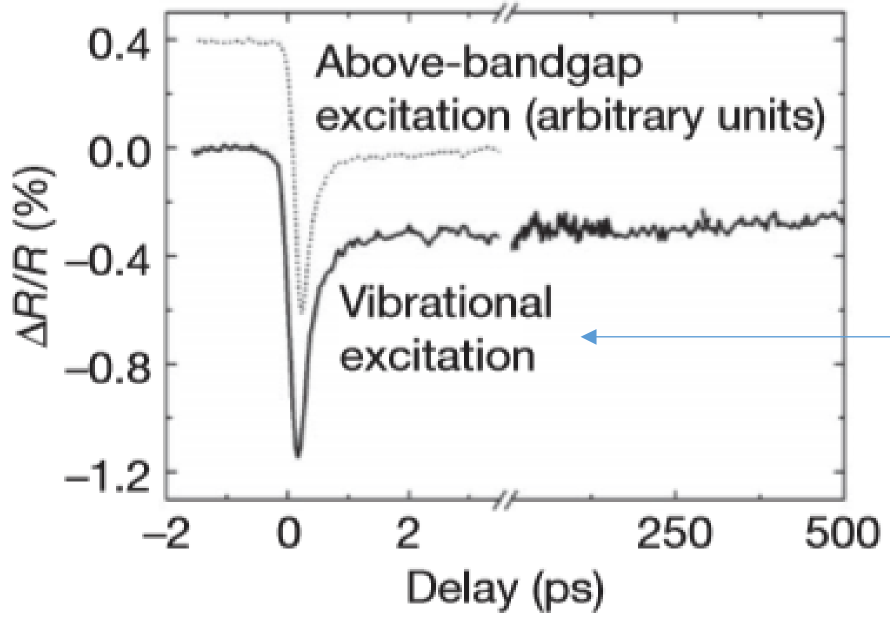
A insulator-metal transition in $Pr_{0.3}Ca_{0.7}MnO_3$ driven by vibrational excitation

Experimental artifact



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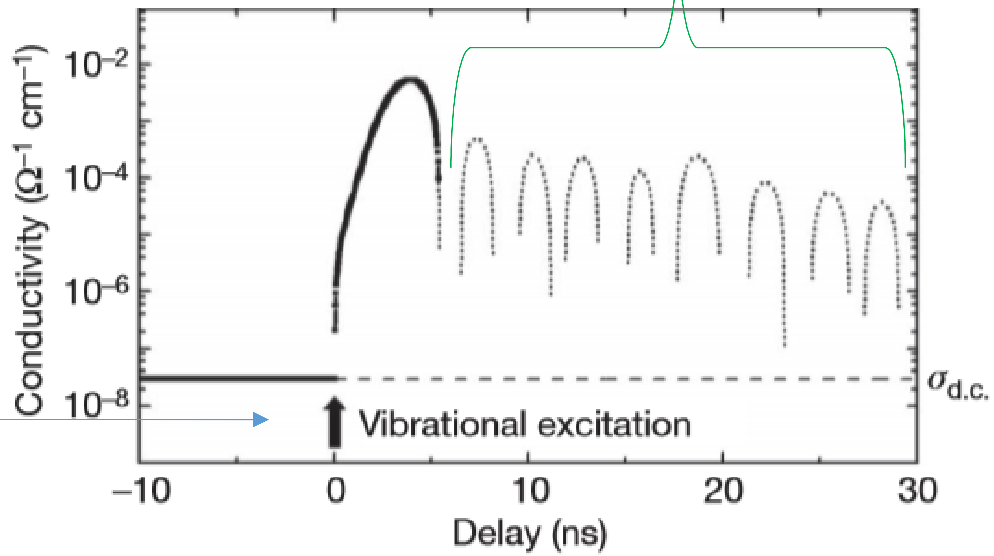
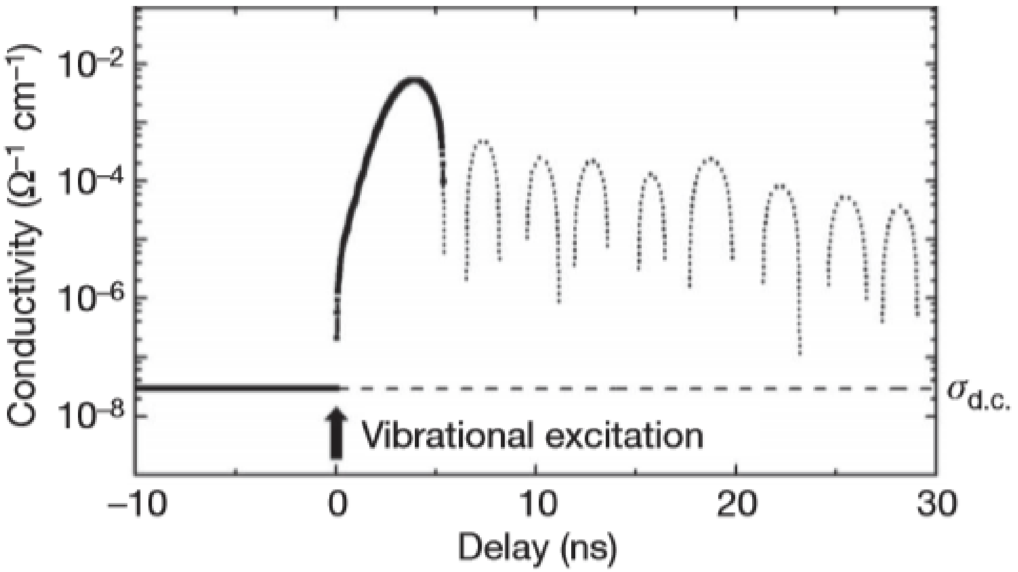


<u>Main Result:</u> ***Through selective excitation of a phonon mode an insulator-metal transition to a hidden state occurs in less than 300 fs!***

Figures from: M. Rini *et al*., Nature **449**, 72

Why is this important?



Conductivity increases by ~10^5!

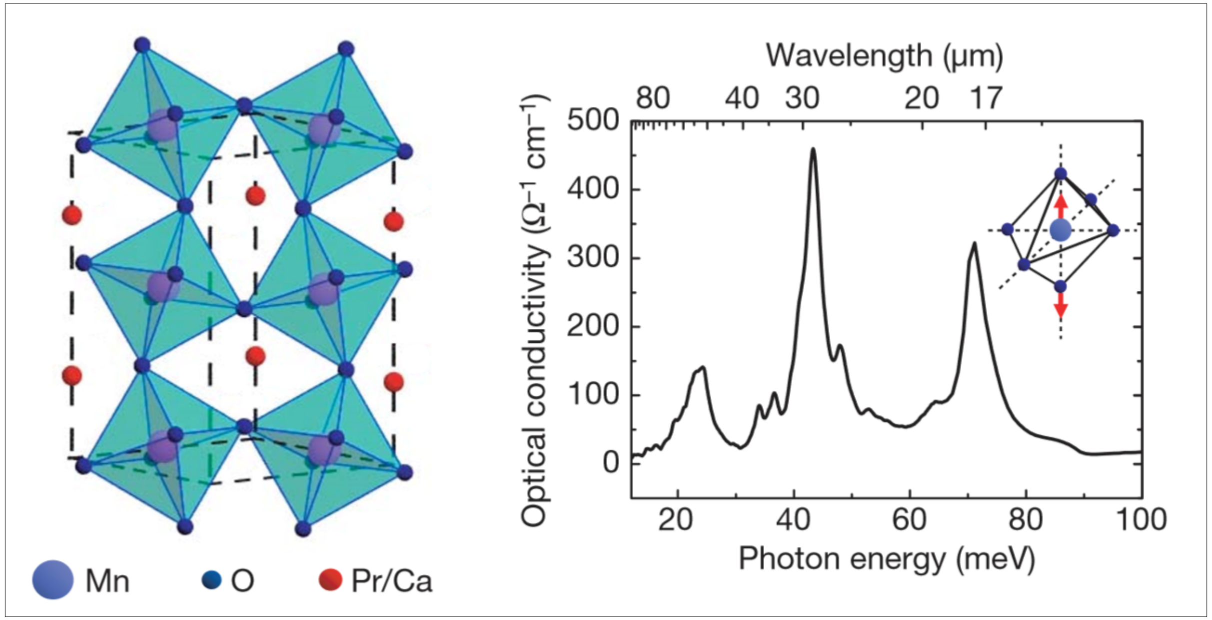
Technology Applications: electrical functionality driven by strong correlations (ultrafast switches/ sensors)

Fundamental Science Insights: novel responses and transitions to hidden phases could be used to study other strongly correlated systems

Figure from: M. Rini *et al*., Nature **449**, 72

$Pr_{0.3}Ca_{0.7}MnO_3$ Phonon Identification

M. Rini *et al*., Nature **449**, 72 (2007).



Phonon mode at 71 meV modifies the tolerance factor (Mn-O-Mn bond angle)

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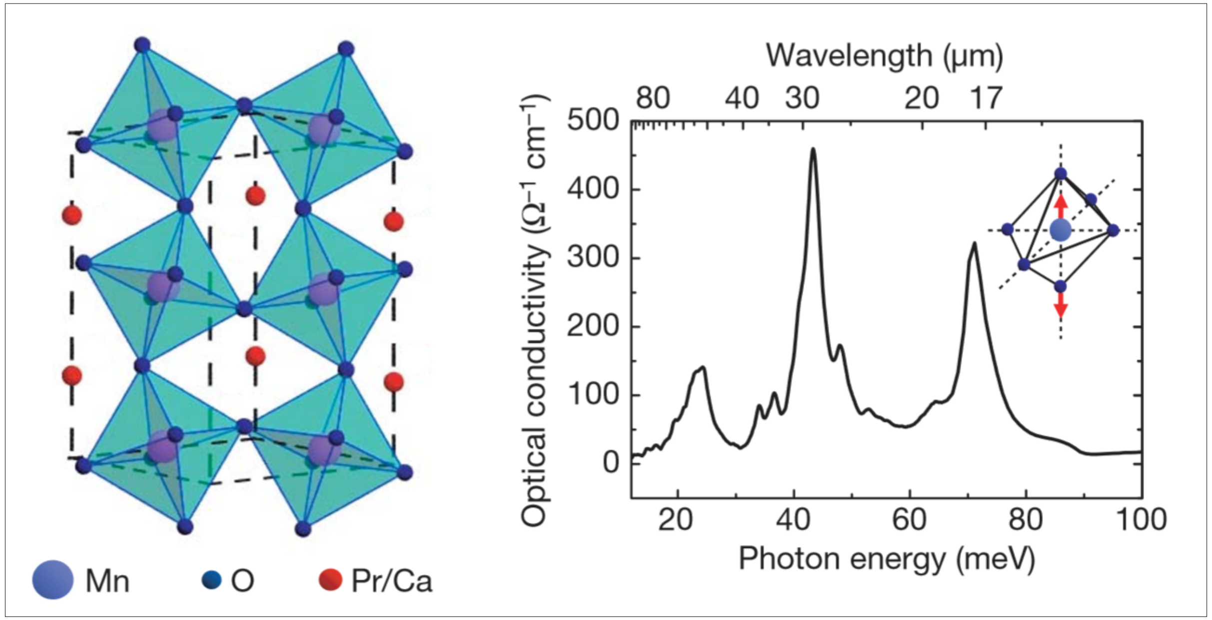

Phonon mode at 71 meV modifies the tolerance factor (Mn-O-Mn bond angle)

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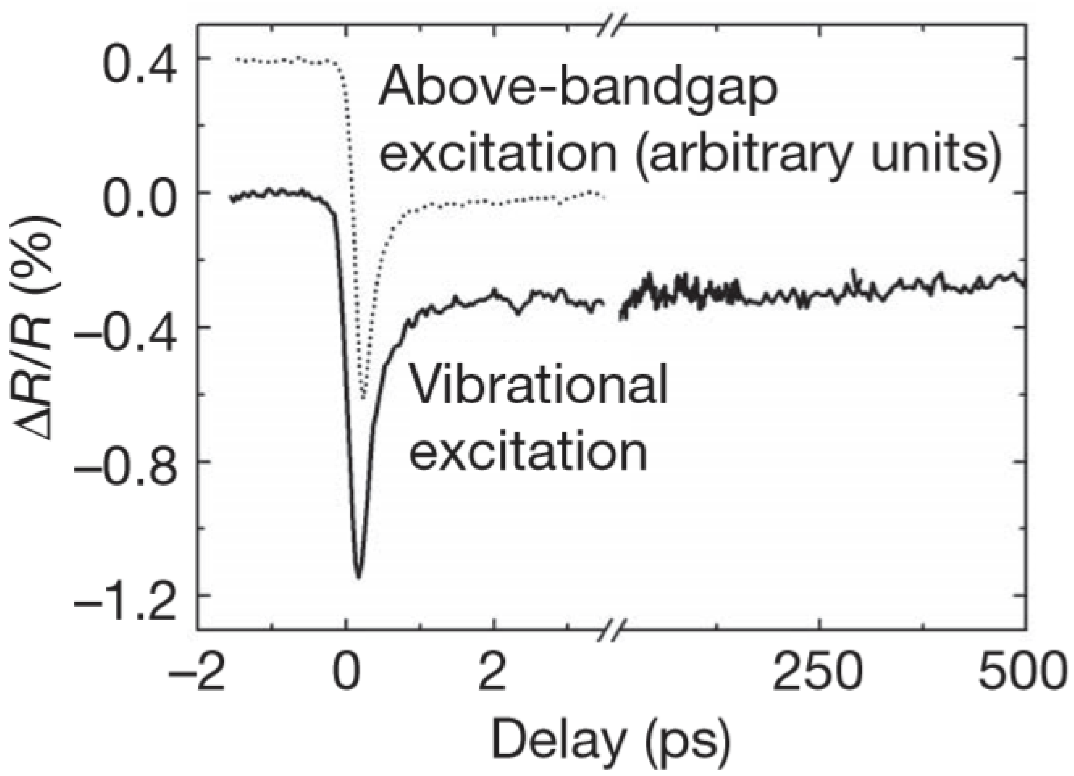
Conduction in 3d orbital systems (manganites) arises from hopping between Mn 3d states mediated by an O 2p[1]

Changes in the tolerance factor = changes in electronic properties

[1]E. Dagotto, in *Nanoscale Phase Separation and Colossal Magnetoresistance: The Physics of Manganites and Related Compounds*

$Pr_{0.3}Ca_{0.7}MnO_3$ Vibration Induced IMT



Reflectivity change develops within 300 fs of vibrational excitation

Figures from: M. Rini *et al*., Nature **449**, 72

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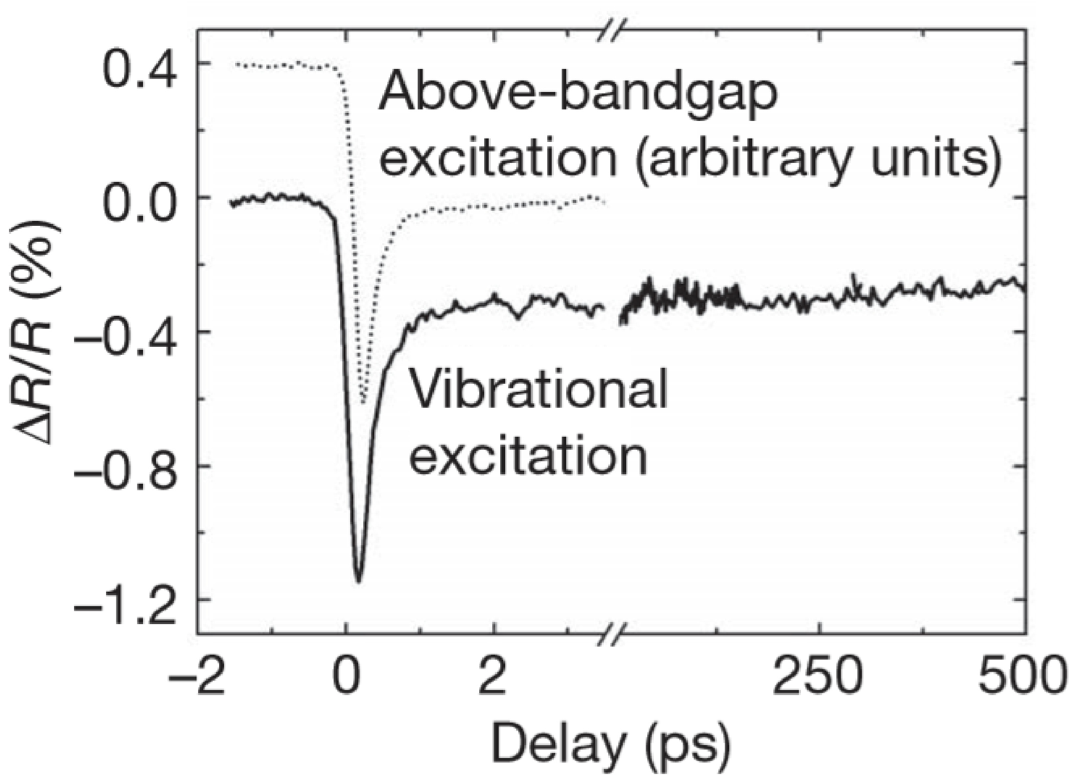

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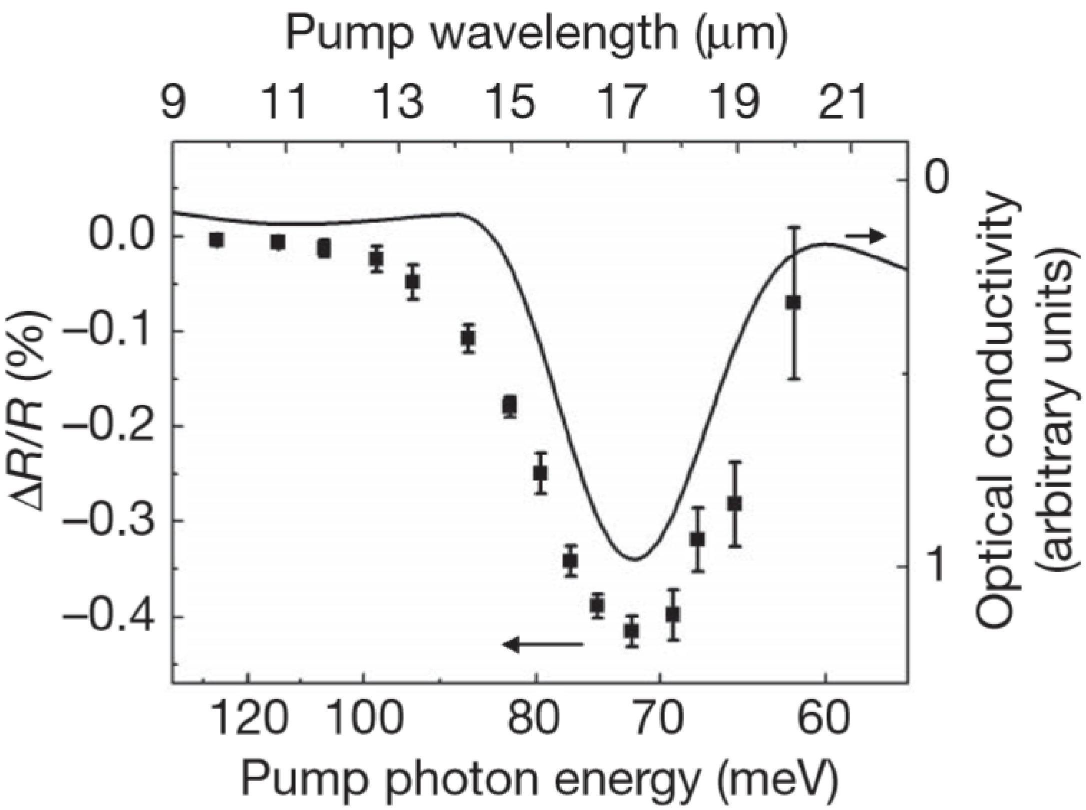
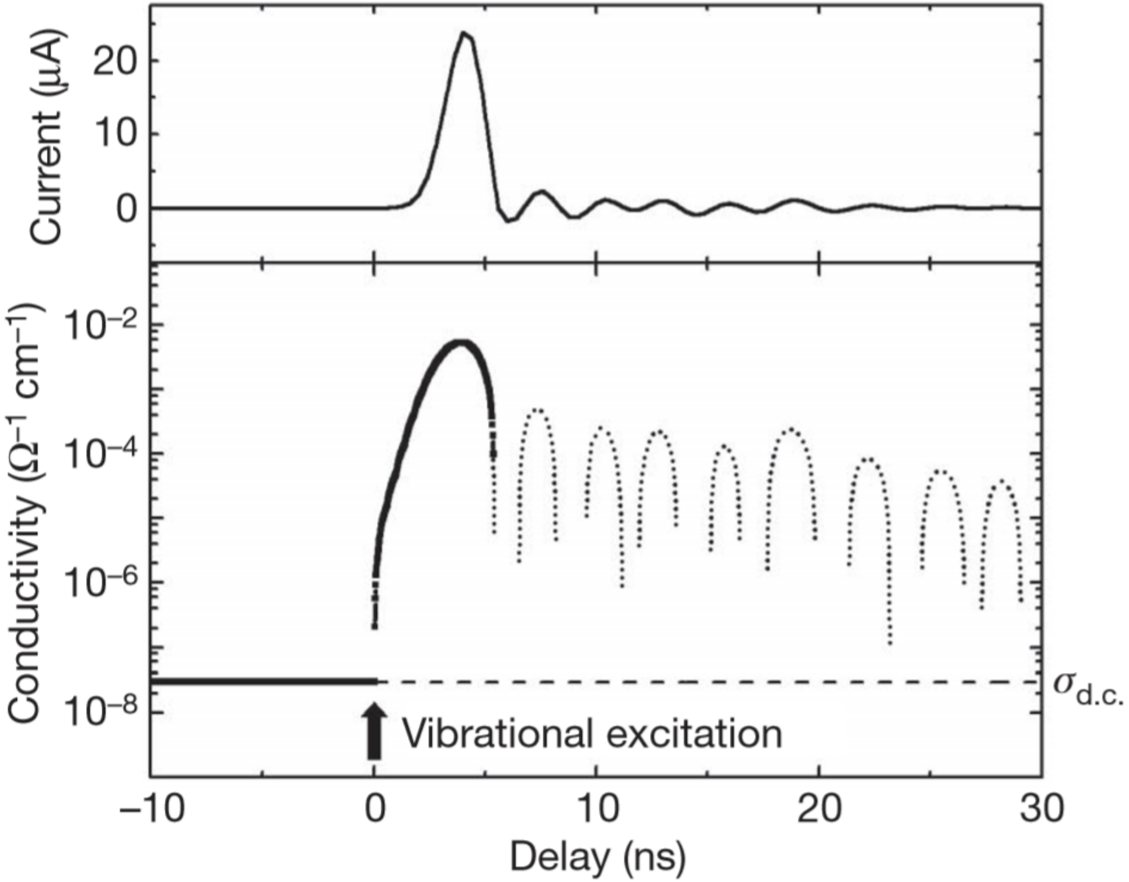


No reflectivity change at 1ps delay when pump is off phonon resonance

Figures from: M. Rini *et al*., Nature **449**, 72

$Pr_{0.3}Ca_{0.7}MnO_3$ Vibration Induced IMT (cont.)



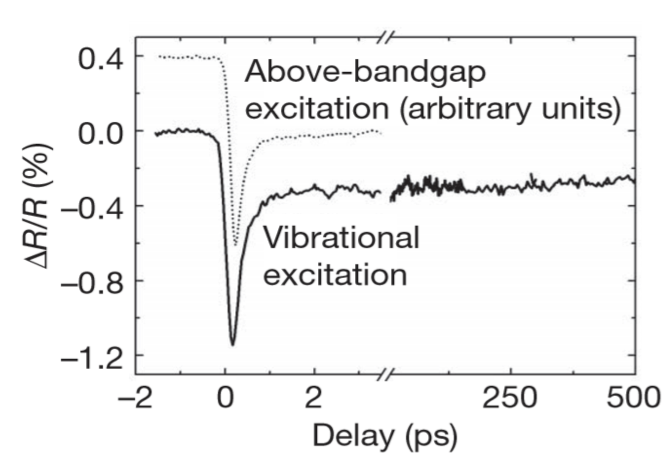
Sample biased and conductivity measured after vibrational excitation

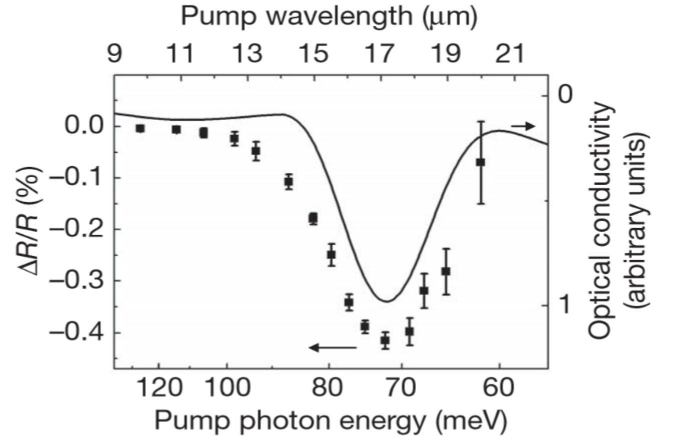
Conductivity increases by ~10^5!

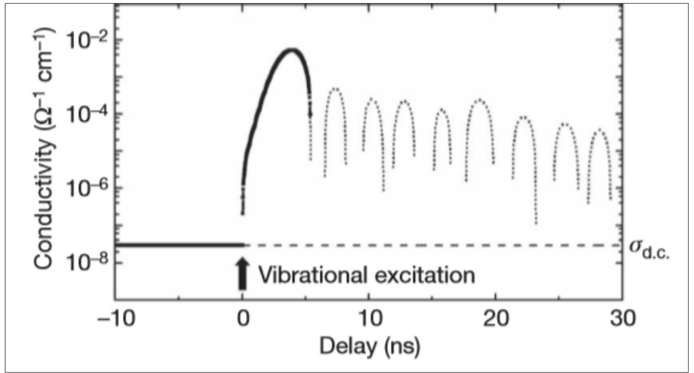
Figures from: M. Rini *et al*., Nature **449**, 72

Conclusions

Main Result: *Rini et al. demonstrate launching an insulator-metal transition to a hidden phase in less than 300 fs through selective excitation of a phonon mode*







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Conclusions

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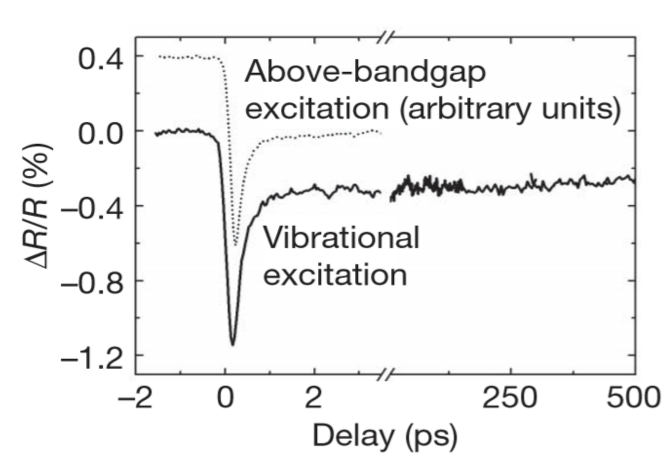
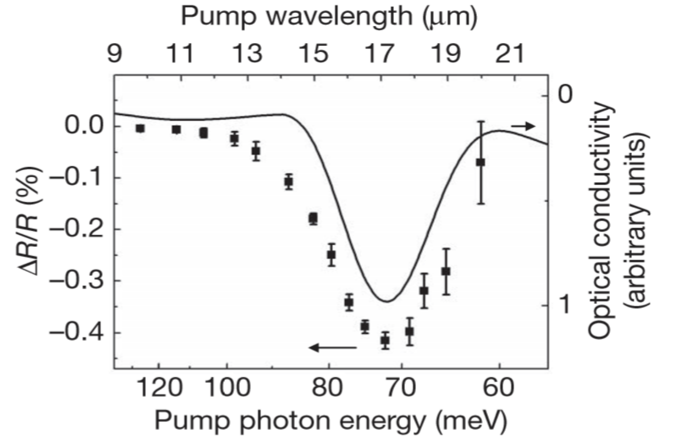
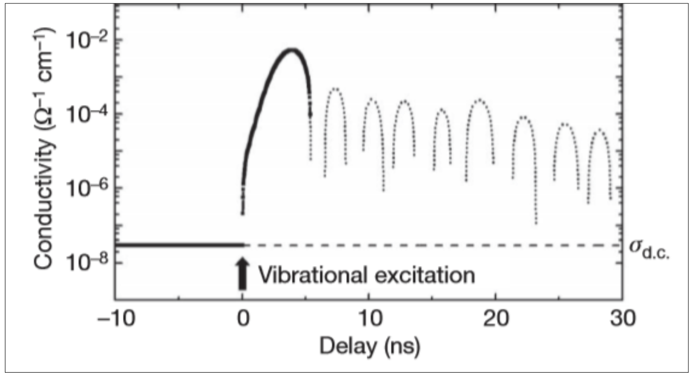


<u>Why is this important?</u>

1.) Leveraging strong correlations to drive novel material property changes holds great promise for functional materials (ultrafast switches and sensors)

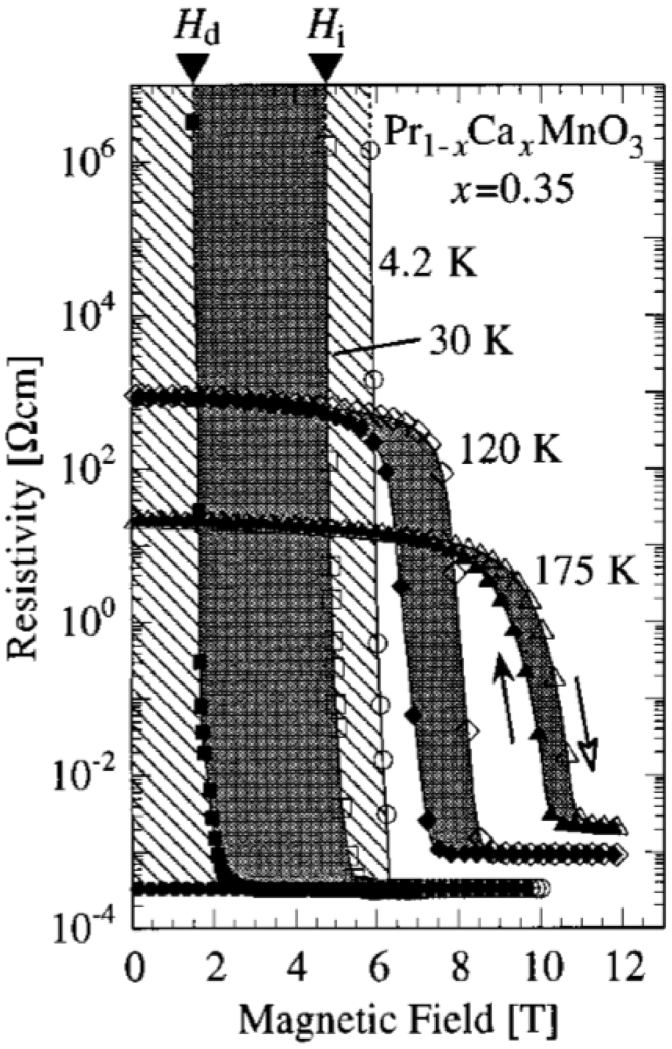


2.) Generating ultrafast responses in other strongly correlated systems could yield new ways to study what mechanisms drive the formation of interesting ground states



Figures from: M. Rini *et al.*, Nature **449**, 72

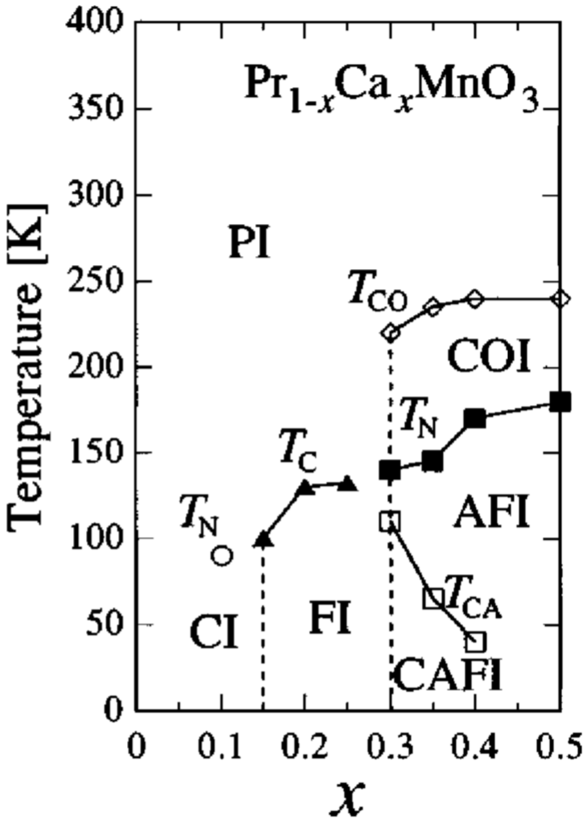
$Pr_{1-x}Ca_xMnO_3$: Resistivity vs Field



- **Critical field for transition is >2T for temperatures of 30K or larger**

Figure from: Y. Tomikoa *et al.*, Phys. Rev. B **53** 1689

$Pr_{1-x}Ca_xMnO_3$: Hidden Phase and Laser Heating



Rich phase diagram from strong correlations and robustly insulating

- Rini *et al.*, estimate laser heating at ~2K by vibrational excitation and *rule out heating as the cause of IMT*

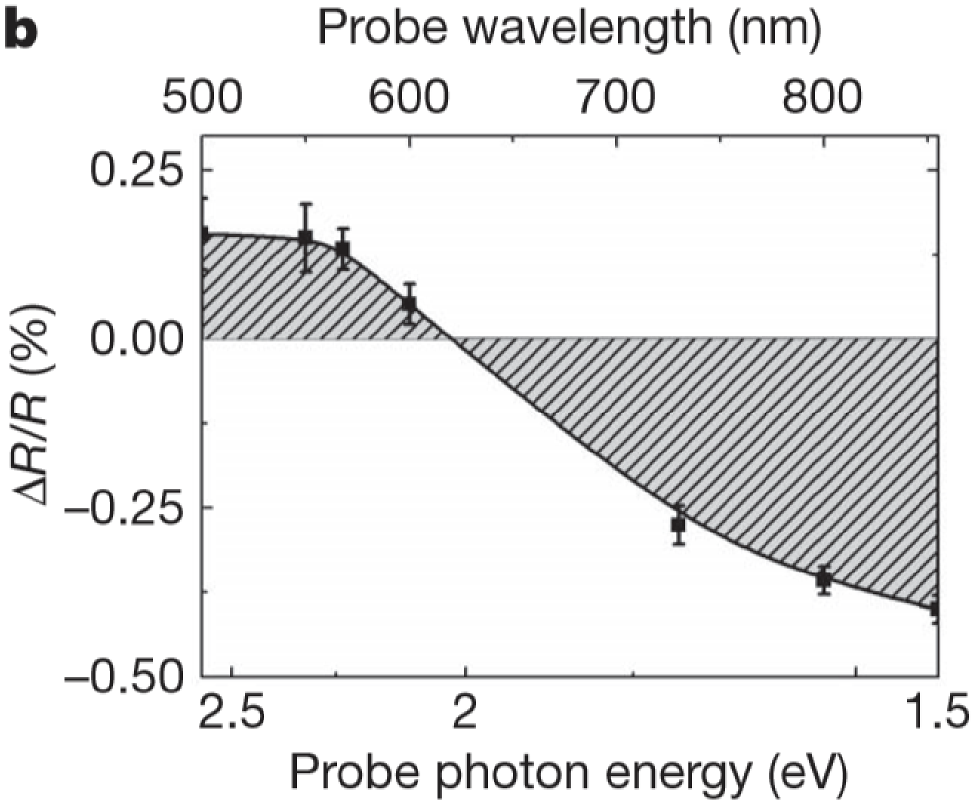
From Rini *et al.*:

“ In a magnetoresistive manganite metallicity is associated with ferromagnetism through the double-exchange mechanism[21], so the formation of a metallic state implies the possibility of generating ferromagnetic domains on ultrafast timescales by excitation of specific vibrational degrees of freedom … ”

→ *Authors only speculate on the magnetic nature of the metallic phase*

Figure from: Y. Tomikoa *et al.*, Phys. Rev. B **53** 1689 (1996).
[21] C. Zener, Phys. Rev. 82, 403 (1950)

$Pr_{1-x}Ca_xMnO_3$: Additional Evidence for Metallic state formation



Reflectivity at 1ps delay changing probe wavelength

-Decreased reflectivity at low photon energies is indicative of formation of a metallic state (seen in other studies where pump is above band gap)

-Likely due to formation of a plasma edge in the metallic state

Figures from: M. Rini *et al.*, Nature **449**, 72