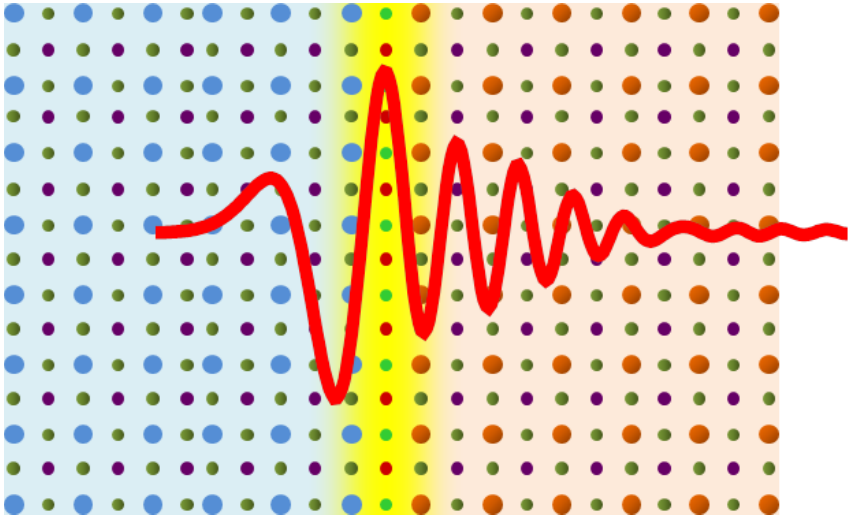
Spectroscopy of correlated electrons in nickelates and titanates

Metal-insulator transitions and novel 2DEGs



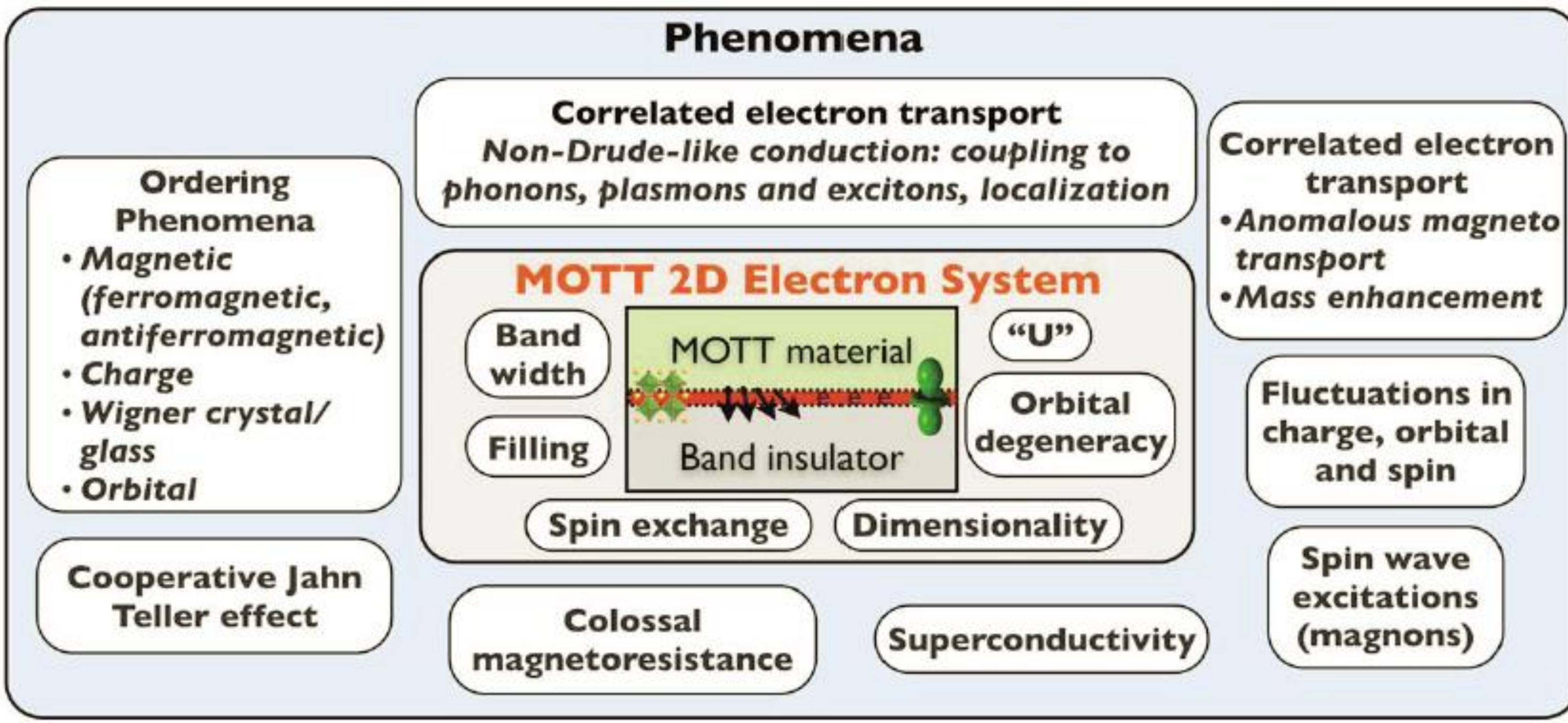
Dan Ouellette

Dept. of Physics

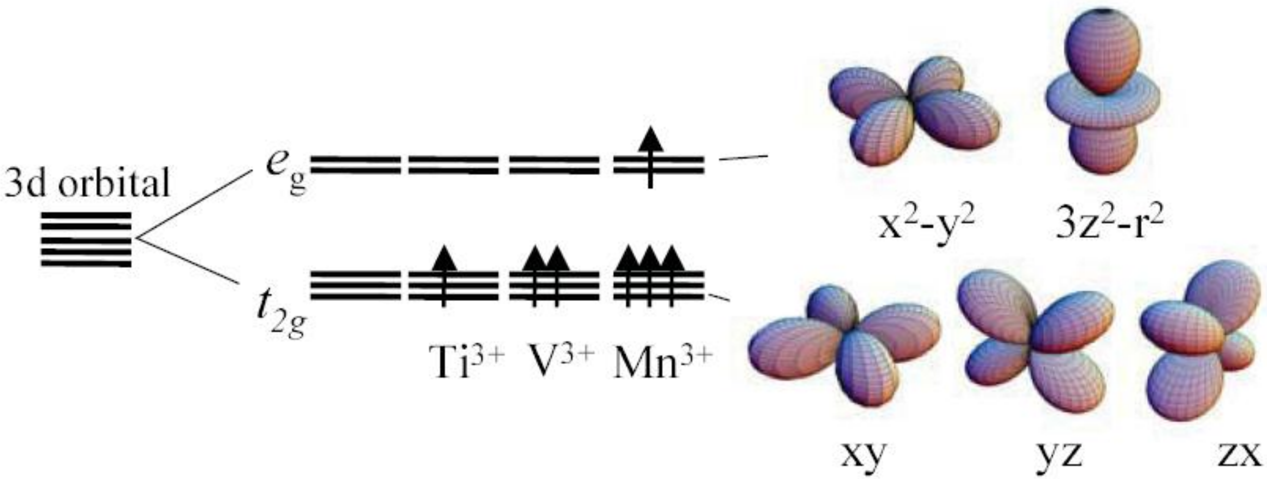
University of CA, Santa Barbara

Strong Electron Correlations

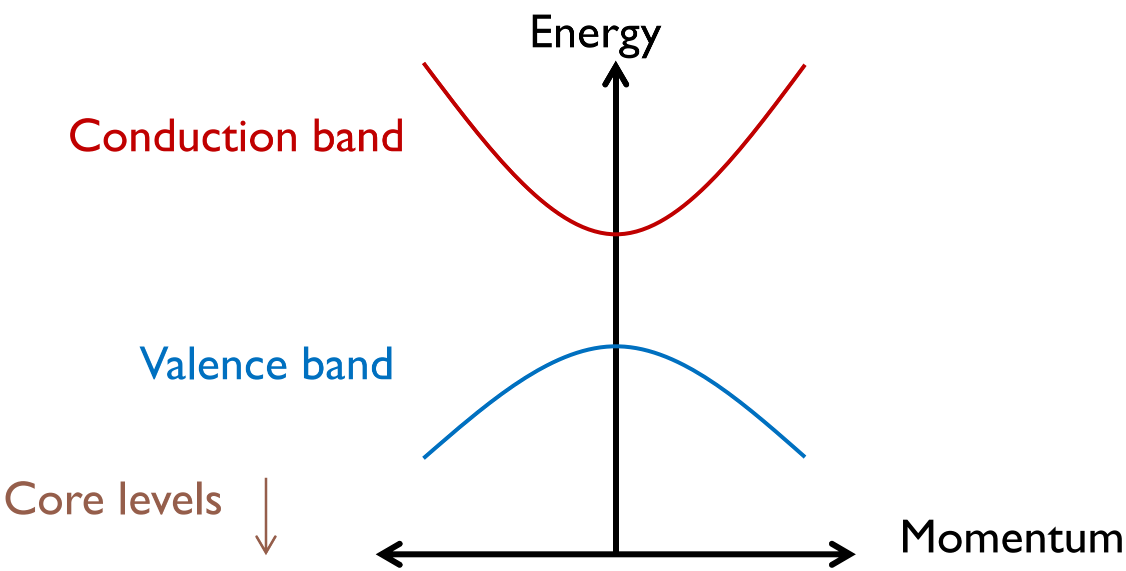
- Materials which depart from our "typical" description of solids.
- Phenomena that cannot be described by single electron physics.



Starting point of Solid State Physics



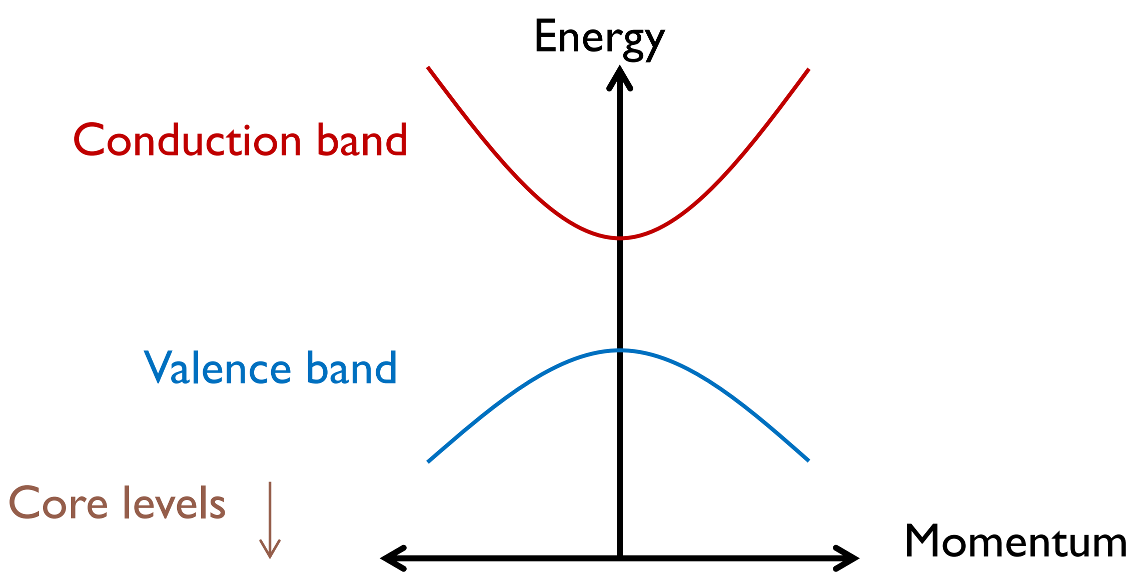
Atoms:
- Orbitals
- Discrete energies



Solids:
- Bloch states
- Continuous energy bands
- Energy gaps

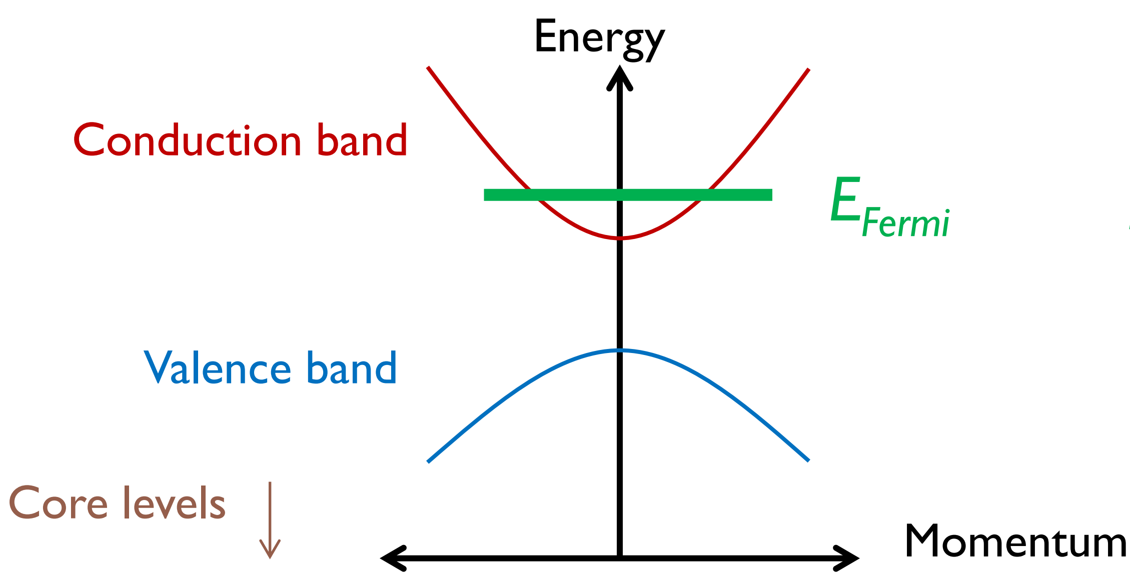
Starting point of Solid State Physics

- Calculate the energy bands
- Single particle physics within a band
  "Band" mass



Starting point of Solid State Physics

- Calculate the energy bands
- Single particle physics within a band
  "Band" mass

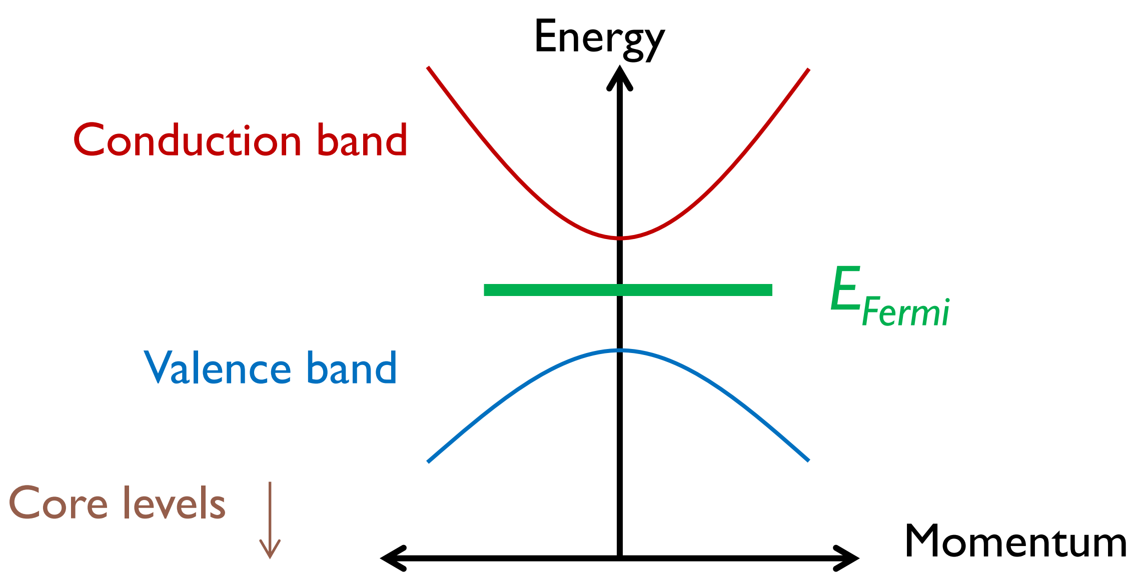


Metal

Starting point of Solid State Physics

- Calculate the energy bands
- Single particle physics within a band

  "Band" mass



Insulator

But don't the electrons interact?

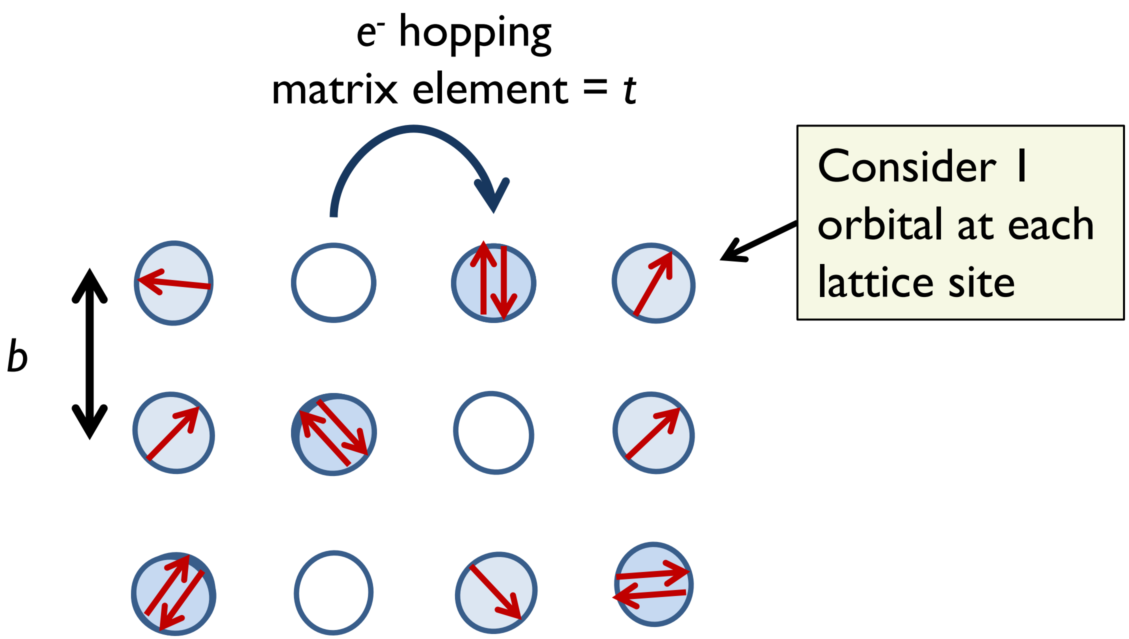
Accounting for e-e interactions

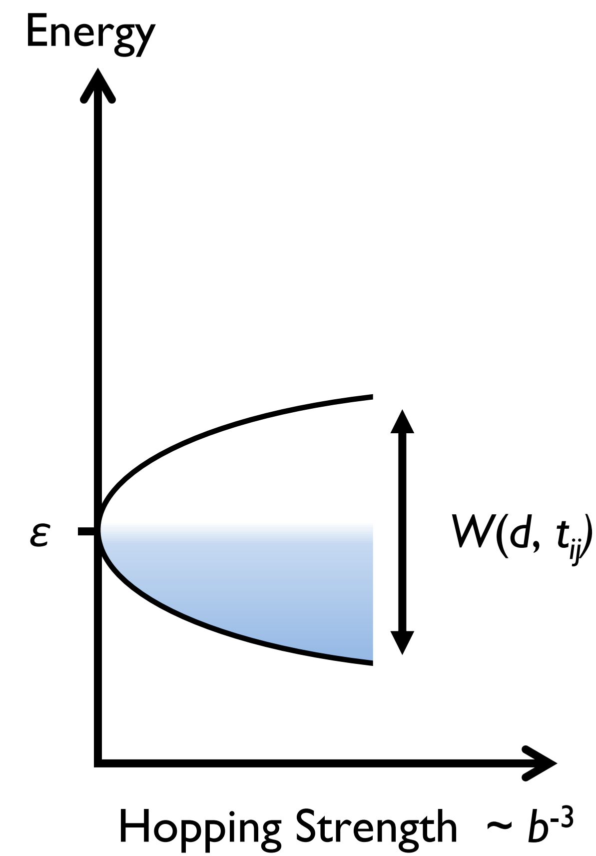
- Density functional theory
  - Replace quantum interactions with average density to obtain energy bands
  - Then consider single electron dynamics in these bands…
- Confined semiconductor structures
  - Schrodinger-Poisson

VERY SUCCESSFUL

What if the motion of the electrons is inherently collective?

Example: The Hubbard Model



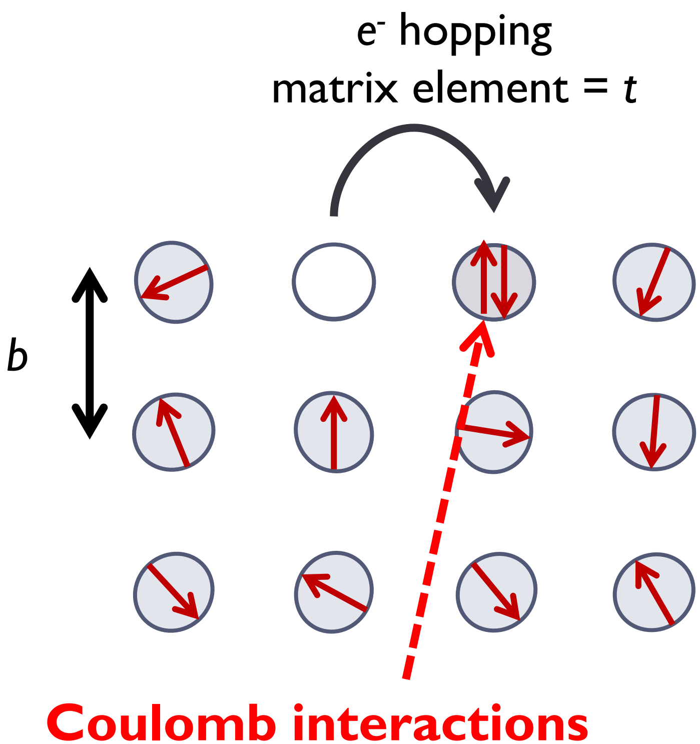


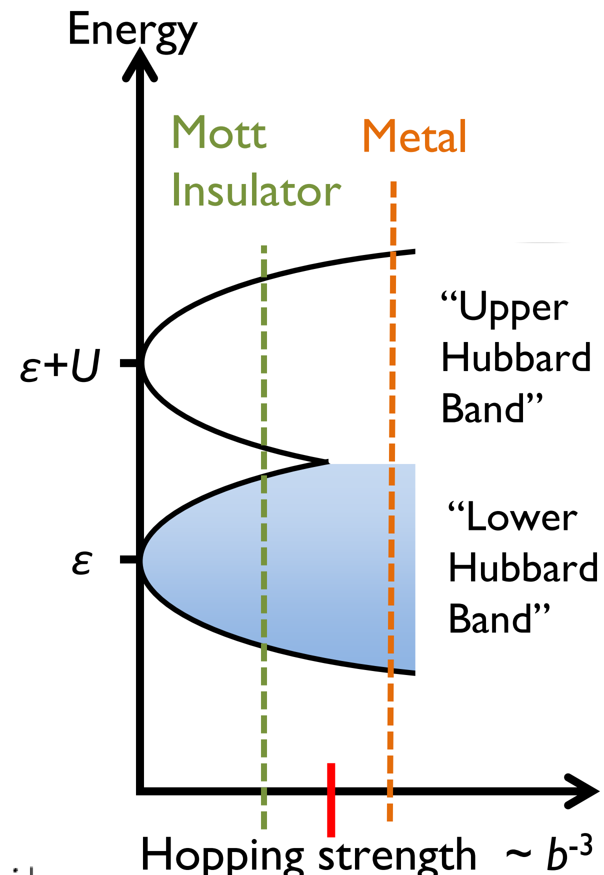
A partially filled band is metallic.

$$H = \sum_{i,\sigma} \epsilon_i n_{i\sigma} + \sum_{ij\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma}$$

N. F. Mott (1949, 1956, 1977, 1978), Hubbard (1964)
Kamimura and Aoki "The Physics of Interacting Electrons in Disordered Systems" Oxford 1989

Example: The Hubbard Model



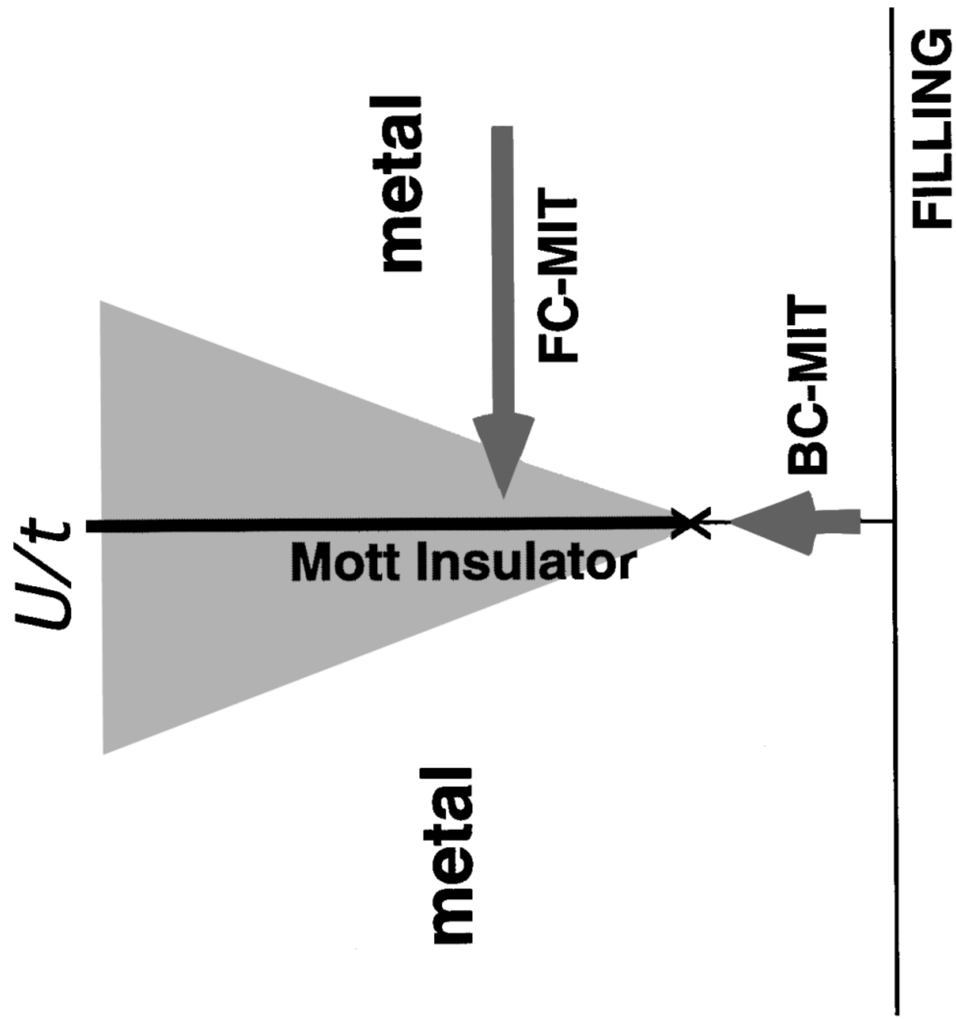


$$H = \sum_{i,\sigma} \epsilon_i n_{i\sigma} + \sum_{ij\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

N. F. Mott (1949, 1956, 1977, 1978), Hubbard (1964)

Kamimura and Aoki "The Physics of Interacting Electrons in Disordered Systems" Oxford 1989

A “Mott” insulator arises from correlations

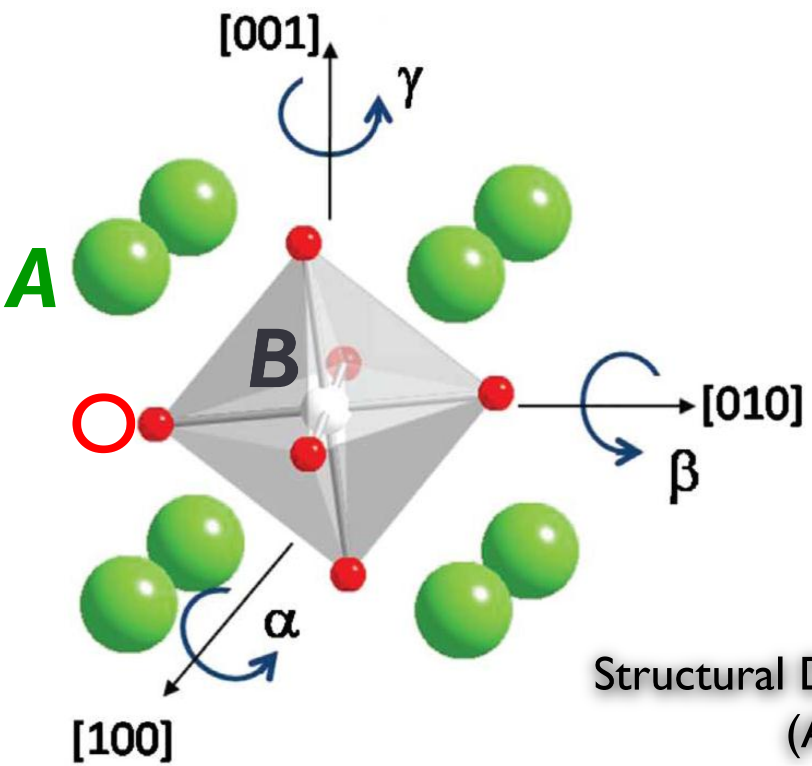


3*d* perovskite transition metal oxides

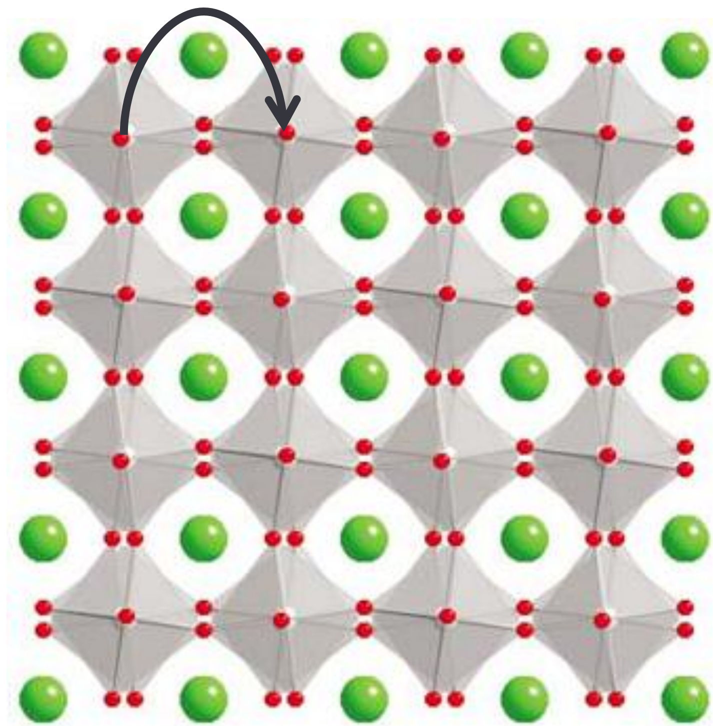
Figures adapted from S. J. May *et al.* Phys. Rev. B **82**, 014110 (2010)

Distorted Perovskites ***A**B*O_3

- Ionic*
- *A* = alkaline earth or rare-earth
- ***B*** = 3*d* transition metal ion

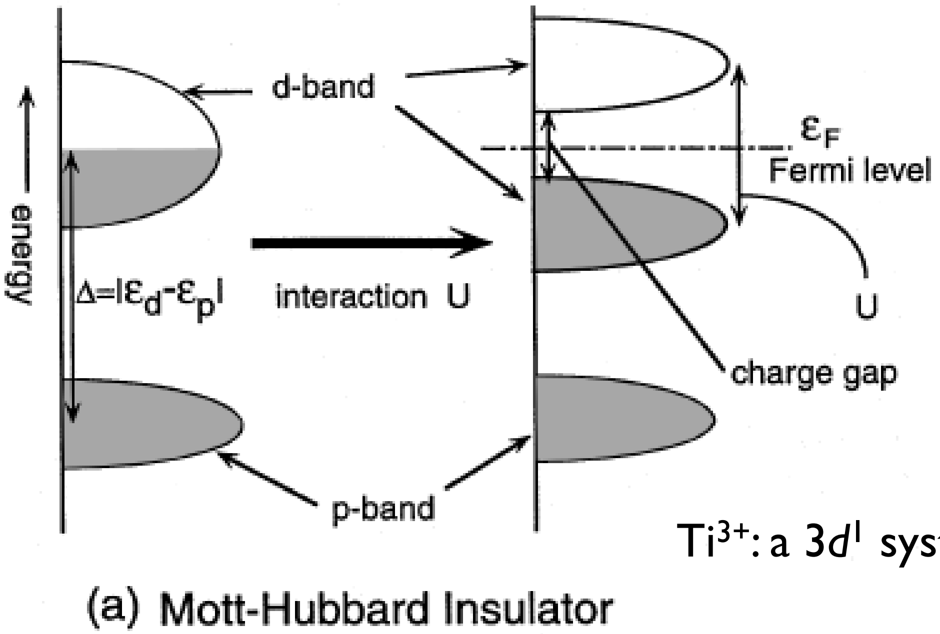


e⁻ hopping

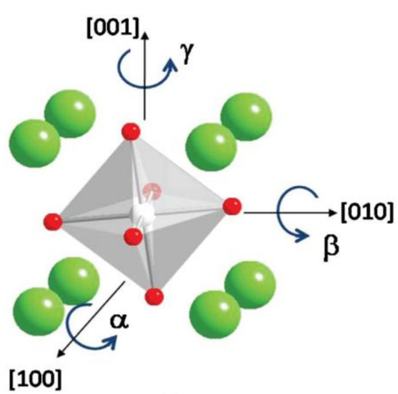


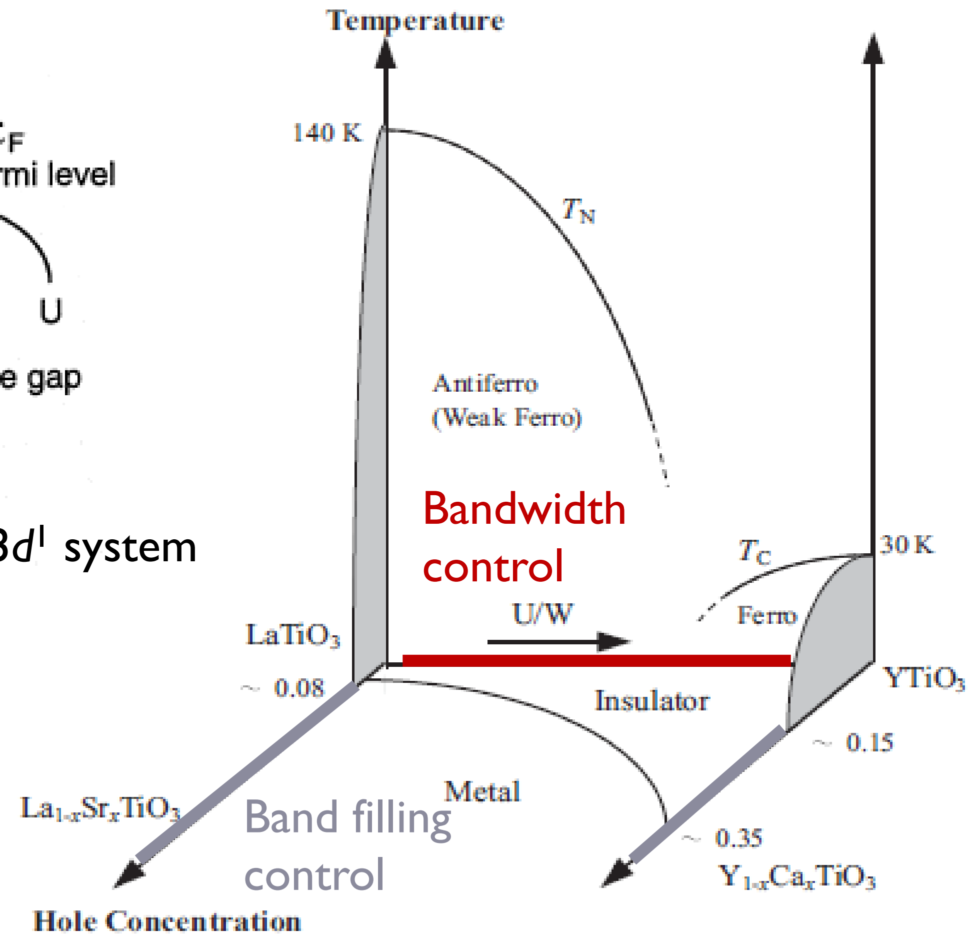
Structural Distortions: Less hopping
(Analogous to spacing in H crystal)

*R*TiO: a family of Mott insulators

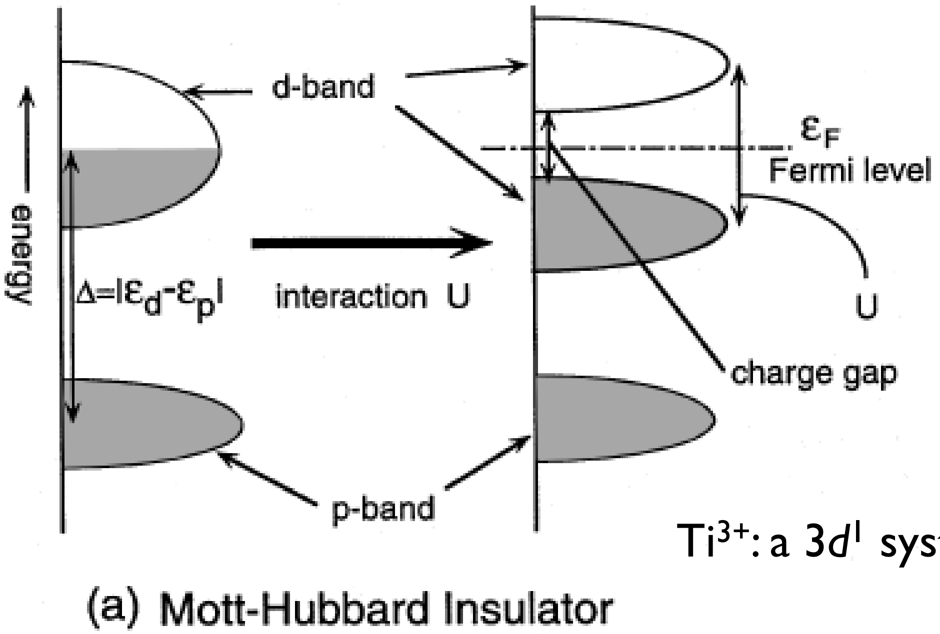


Ti^{3+}: a 3d^1 system

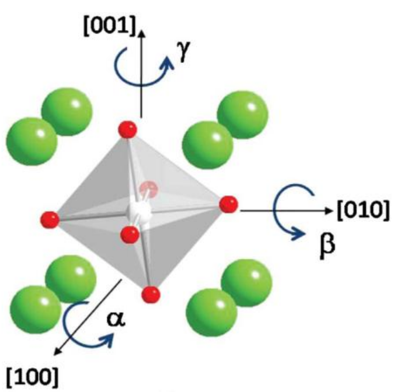


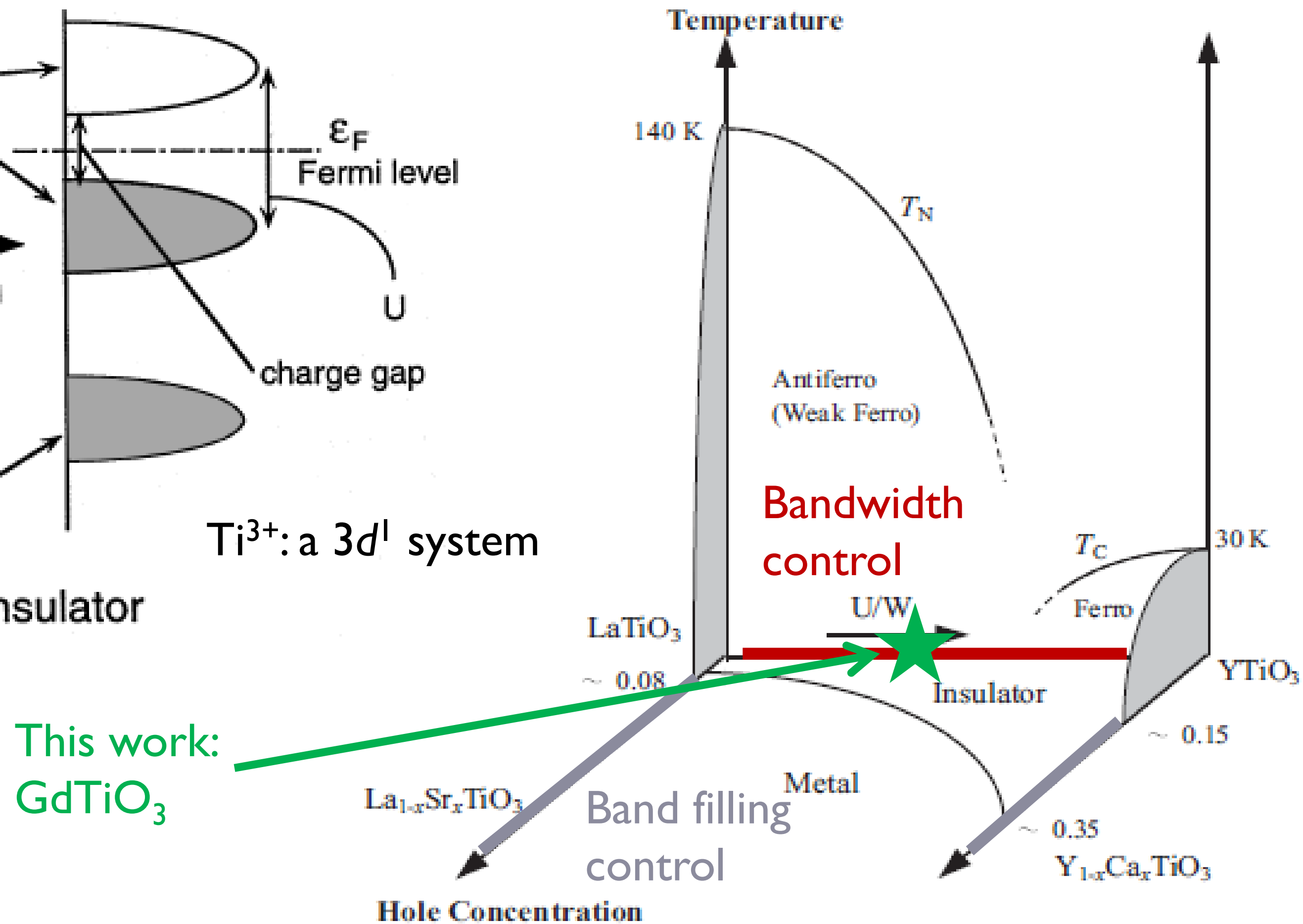


*R*TiO: a family of Mott insulators



Ti^{3+}: a 3*d*[1] system





- Related compound $SrTiO_3$:Ti^{4+}, a 3d^0 "band" insulator

Thin Films of $GdTiO_3$ and $SrTiO_3$

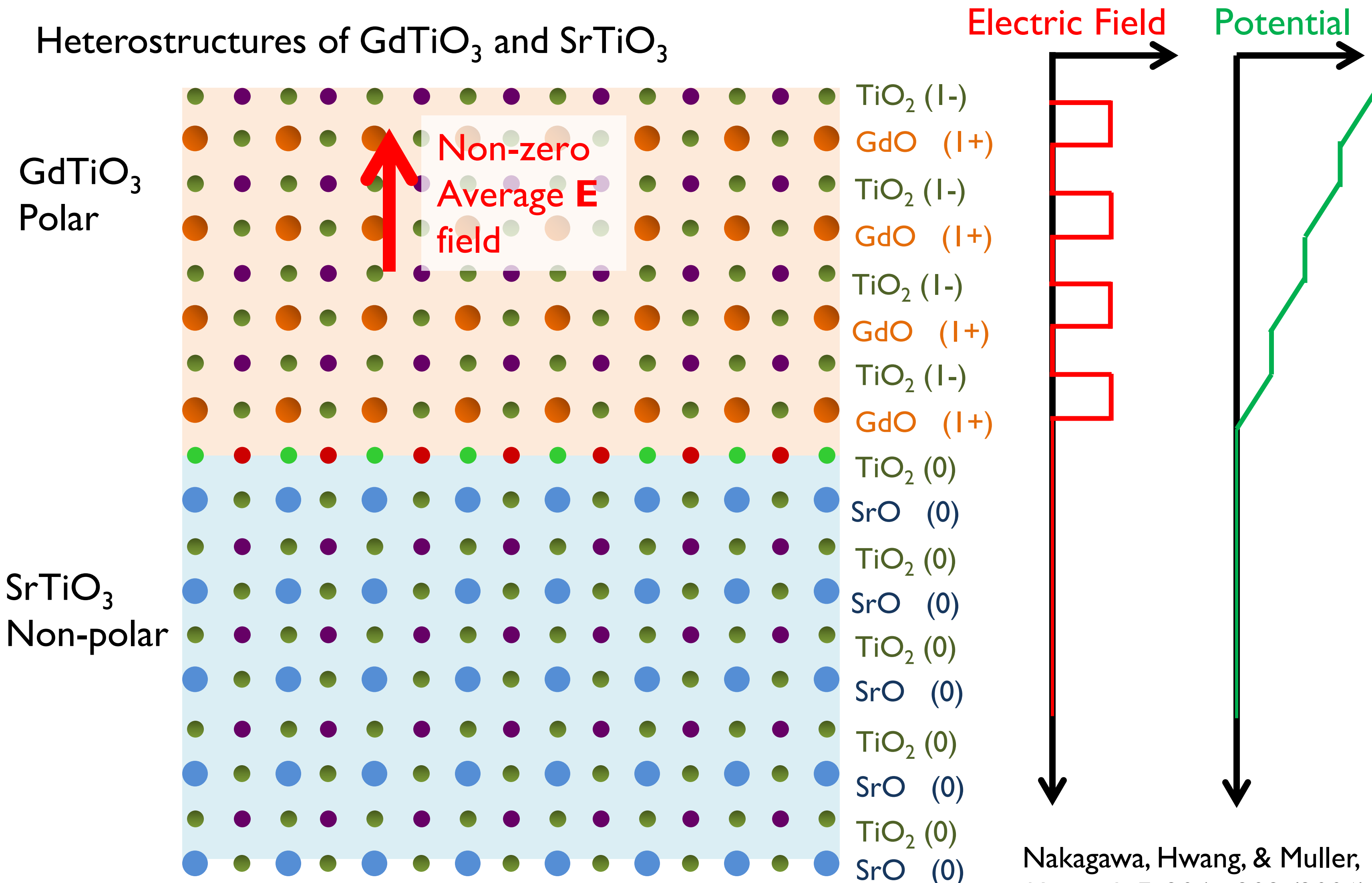
Stemmer Group, UCSB

- Hybrid MBE: metal organic precursor for Ti.
- Insulating films
- $GdTiO_3$ is ferromagnetic

Alexander Kaiser, Chuck Fadley (UC Davis)

The polar catastrophe and a novel 2DEG

Heterostructures of $GdTiO_3$ and $SrTiO_3$



Nakagawa, Hwang, & Muller, *Nature Materials* **5**, 204 - 209 (2006)

The polar catastrophe and a novel 2DEG

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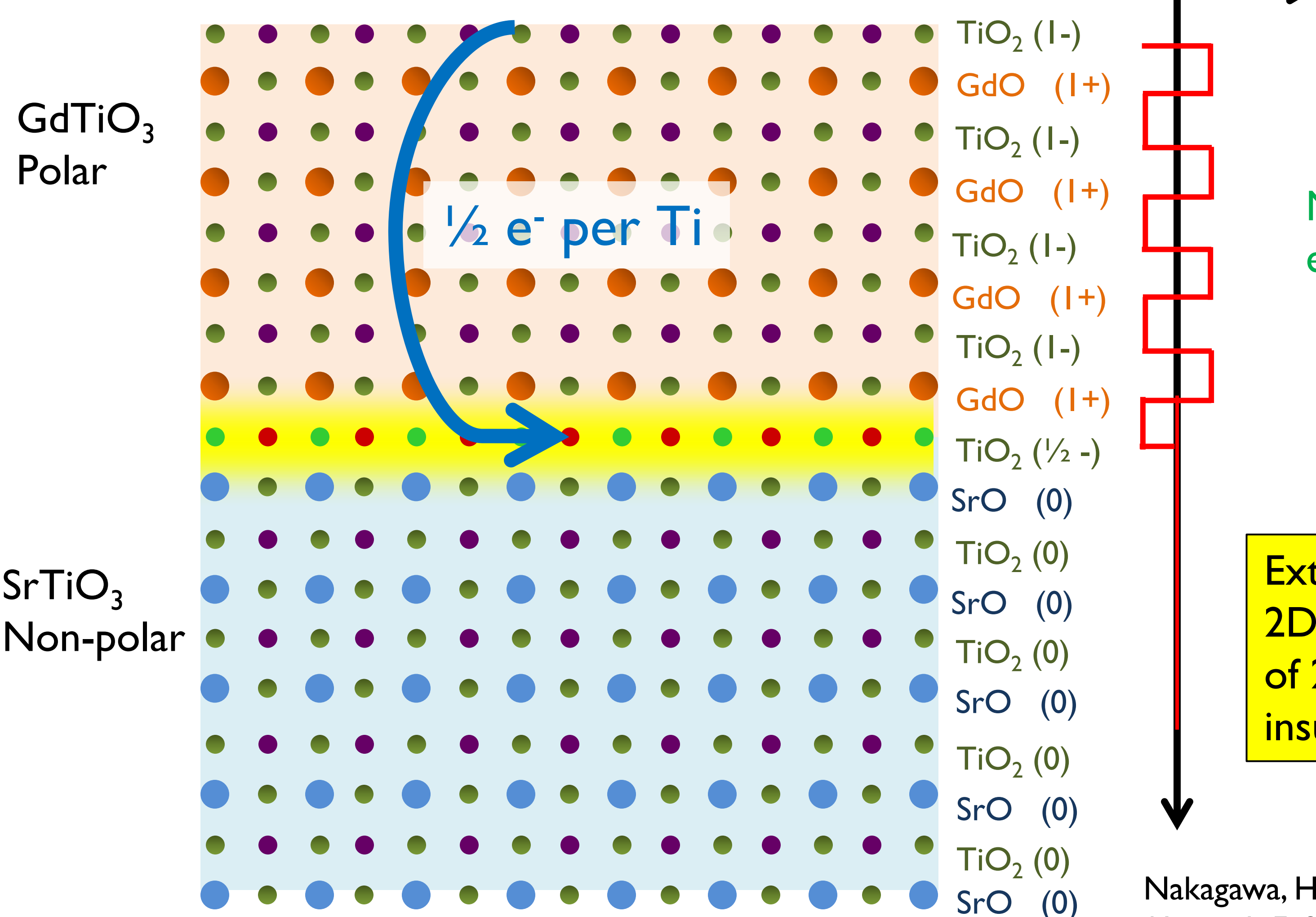
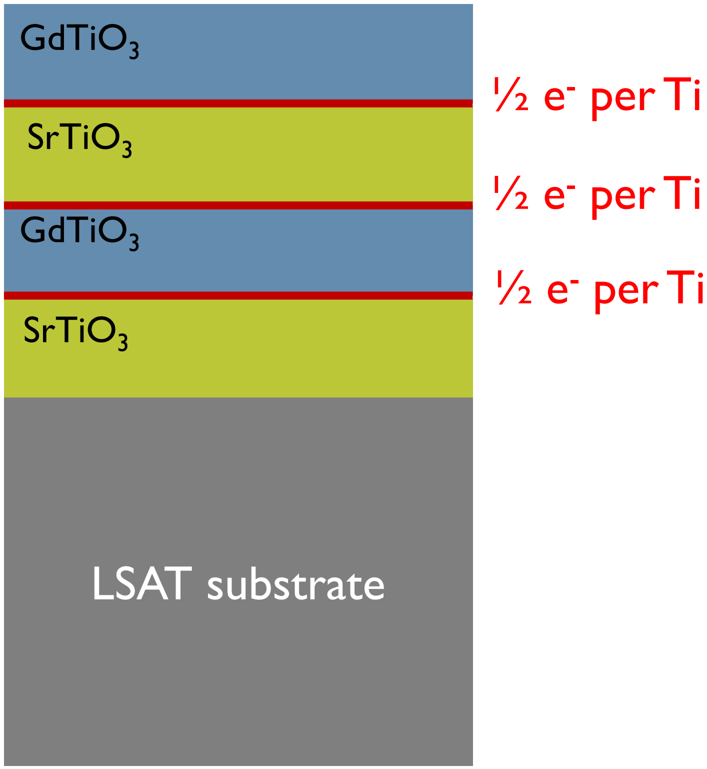


No macroscopic electric potential

Extreme density 2DEG at the interface of 2 (undoped) insulators

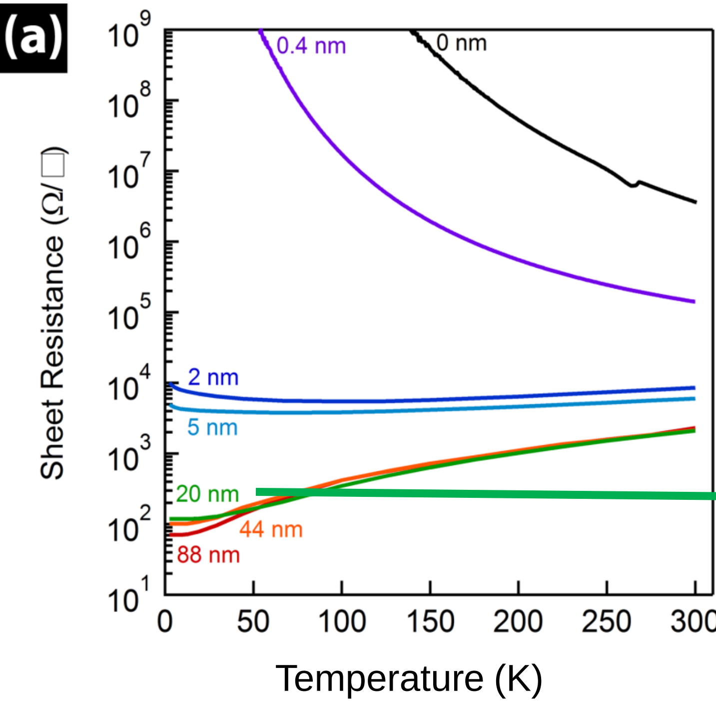
Nakagawa, Hwang, & Muller, *Nature Materials* **5**, 204 - 209 (2006)

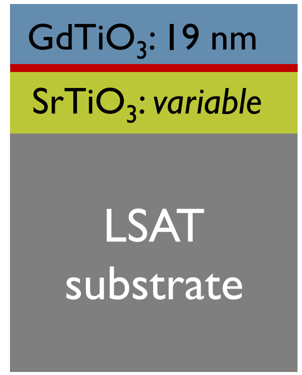
2DEG at the interface

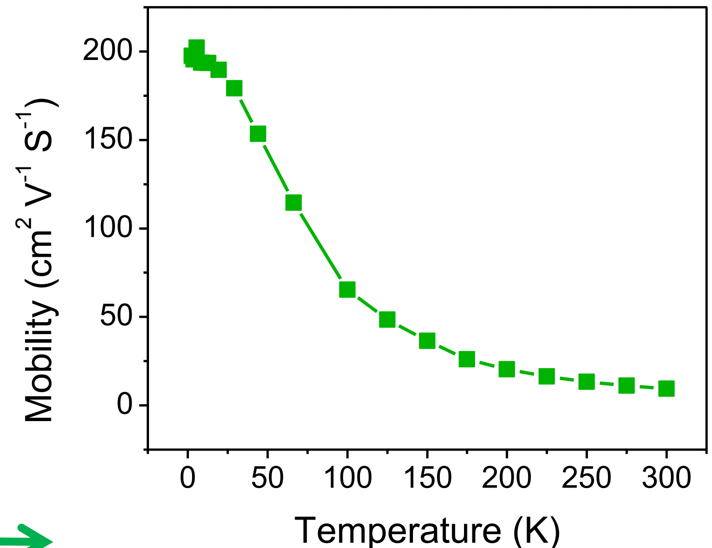


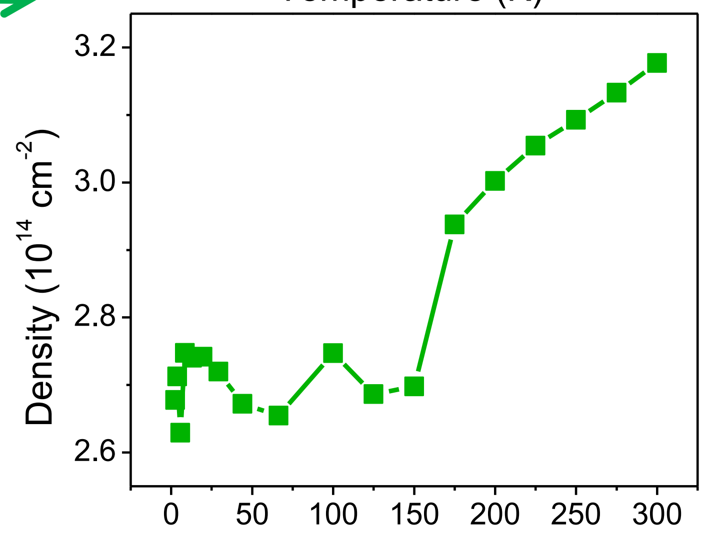
Pouya Moetakef *et. al.* APL **98** 112110 (2011)

Where is this 2DEG?









Mobility and interface scattering suggest confinement to within a few nm of interface.

Pouya Moetakef *et. al.* APL **98** 112110 (2011), Pouya Moetakef *et al. in press*

Optical response

- More information from frequency dependent transport!

Conductivity in the plane: Drude like?

$$\sigma(\omega) = \frac{ne^2\tau/m}{1 - i\omega\tau}$$

Out-of plane? Couple to intersubband transitions and plasmons?

Optical properties of solids

Drude Model

$$\sigma(\omega) = \frac{ne^2\tau/m}{1 - i\omega\tau}$$

Sum Rule

$$\frac{2}{\pi}\int_0^\infty \sigma_1(\omega)\,d\omega = \frac{ne}{m}$$

More general then Drude Model!

Frequency dependent conductivity: Drude (free carriers), interband transitions, phonons, polarons,...

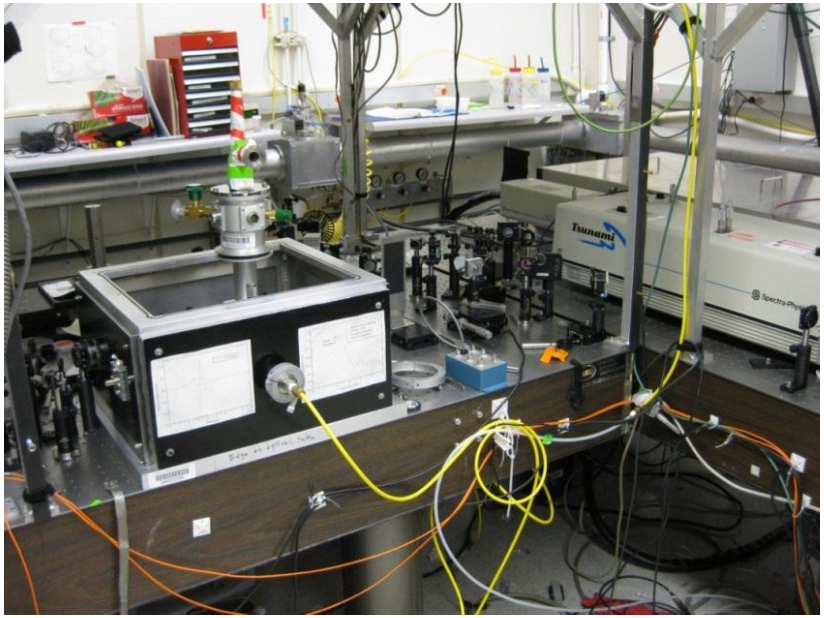
Dielectric polarizability:

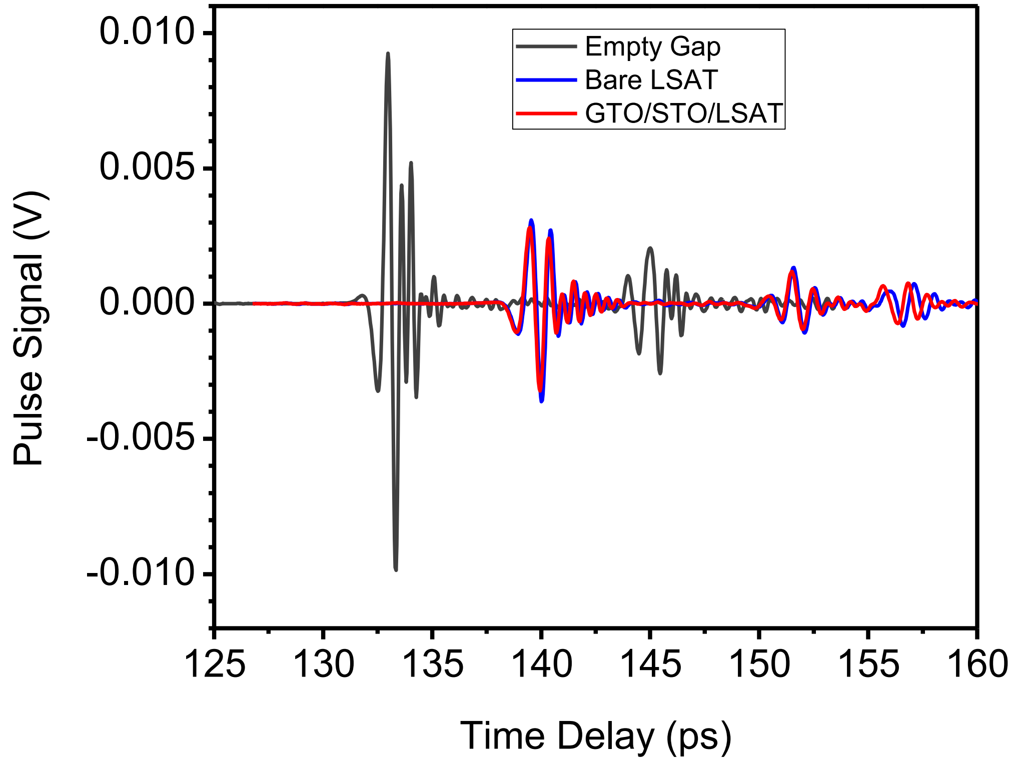
$$\epsilon(\omega) = \epsilon_\infty + \frac{i\sigma(\omega)}{\omega}$$

Index of refraction and absorption coefficient

$$\tilde{n}(\omega) = n(\omega) + i\frac{c\alpha(\omega)}{2\omega} = \sqrt{\epsilon(\omega)}$$

THz Time Domain Spectroscopy





Femtosecond NIR pulse

→Picosecond broadband THz pulse

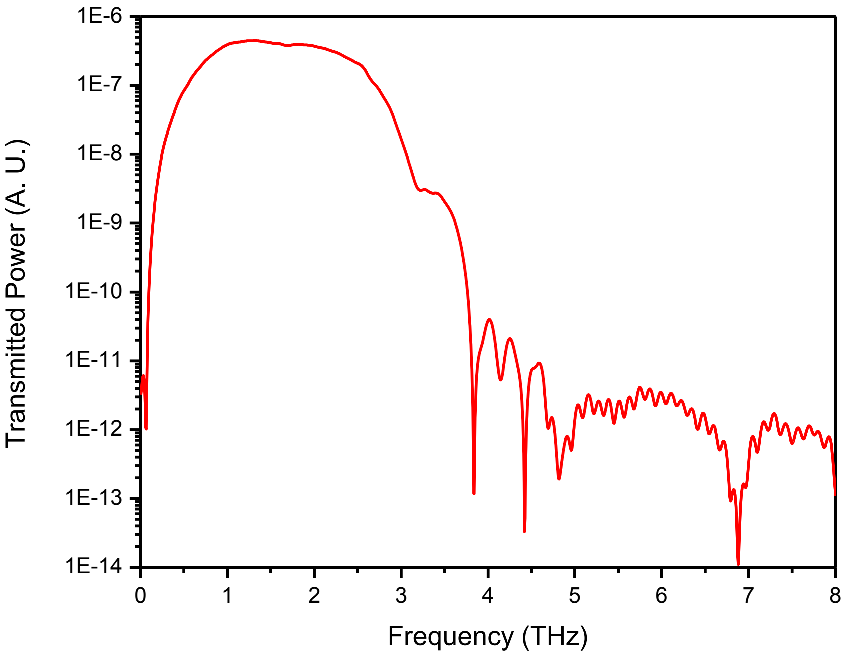
→Electric field versus time

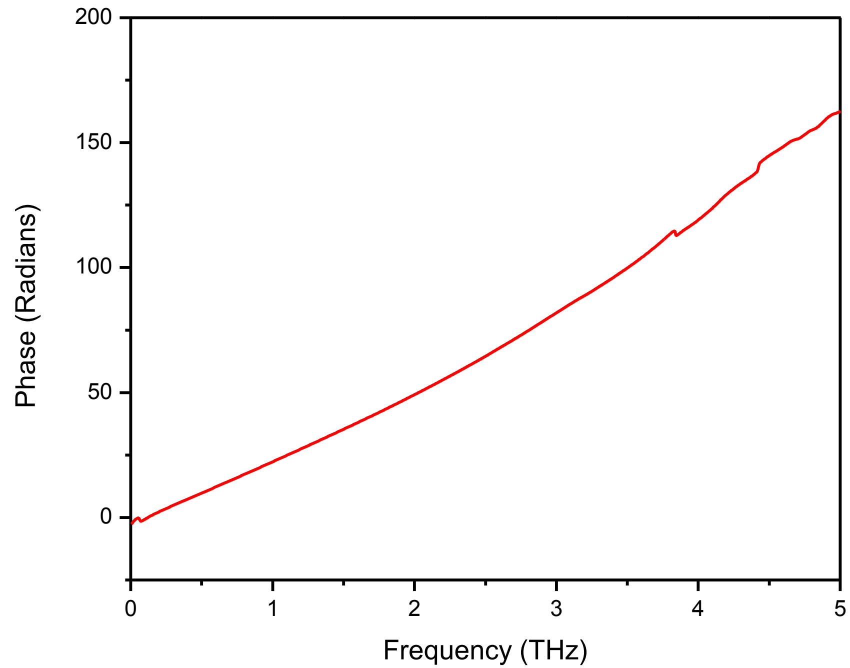
→Fourier transform for COMPLEX spectrum

With Mark Sherwin and Chris Morris

THz Time Domain Spectroscopy

Amplitude and Phase Information





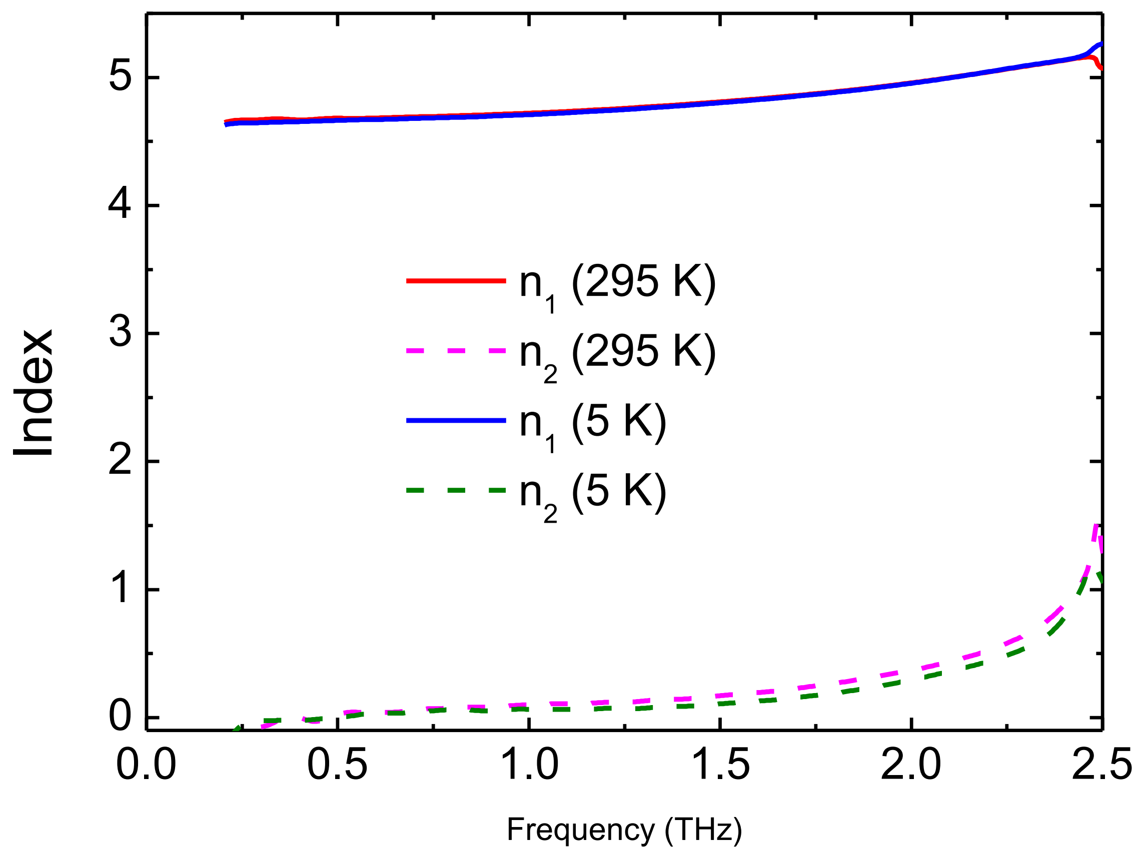
THz Time domain spectroscopy

- Preliminary step: substrate properties

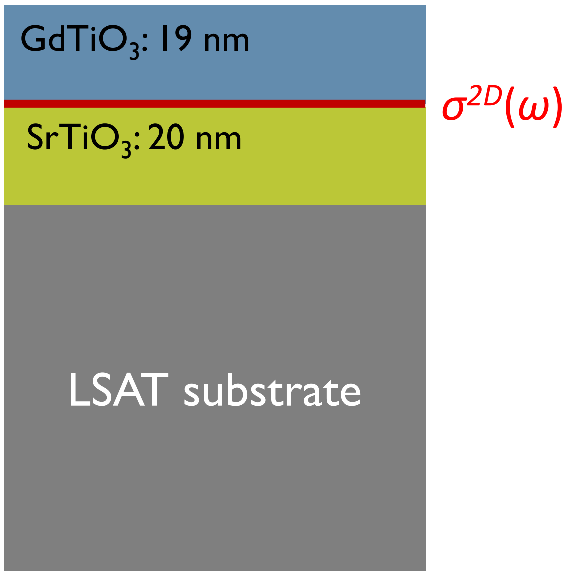
"LSAT": ($La_{0.18}Sr_{0.82})(Al_{0.59}Ta_{0.41})O_3$

Phase shift
→ real refractive index

Absorption and real index
→ absorption coefficient



THz TDS of a 2DEG



$$\frac{E_{sample}(\omega)}{E_{substrate}(\omega)} = \frac{1+\tilde{n}^{substrate}}{1+\tilde{n}^{substrate}+\sigma^{2D}\mu_0 c}$$

↓

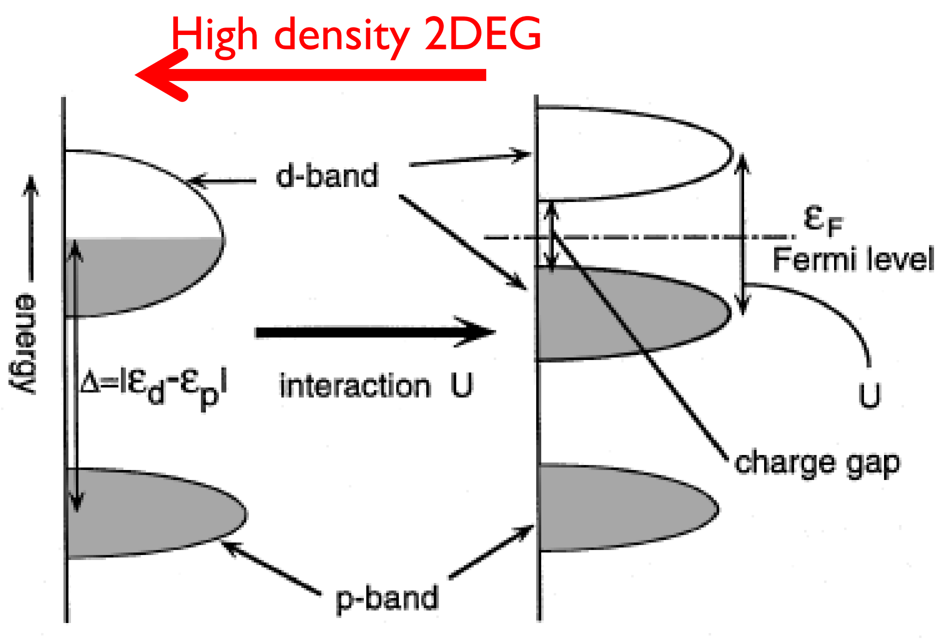
Expect phase shift < 3°

- Get real part of conductivity from power absorption

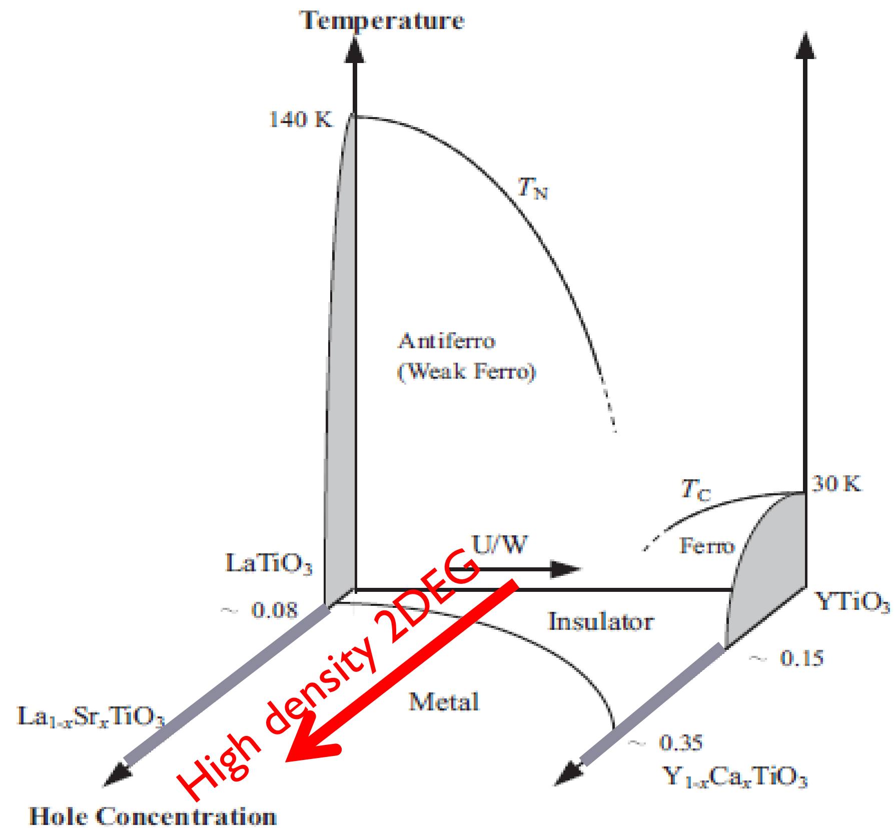
↓

$$\frac{E_{sam}(\omega)}{E_{ref}(\omega)} \approx \left|\frac{E_{sam}(\omega)}{E_{ref}(\omega)}\right| \approx \frac{1+n_1^{substrate}}{1+n_1^{substrate}+\sigma_1^{2D}\mu_0 c}$$

Alternate (complimentary?) hypothesis



(a) Mott-Hubbard Insulator

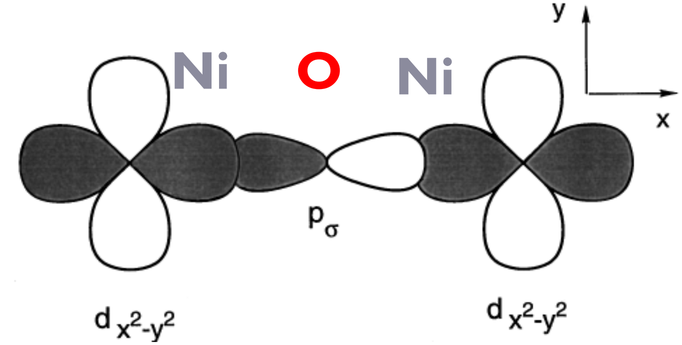


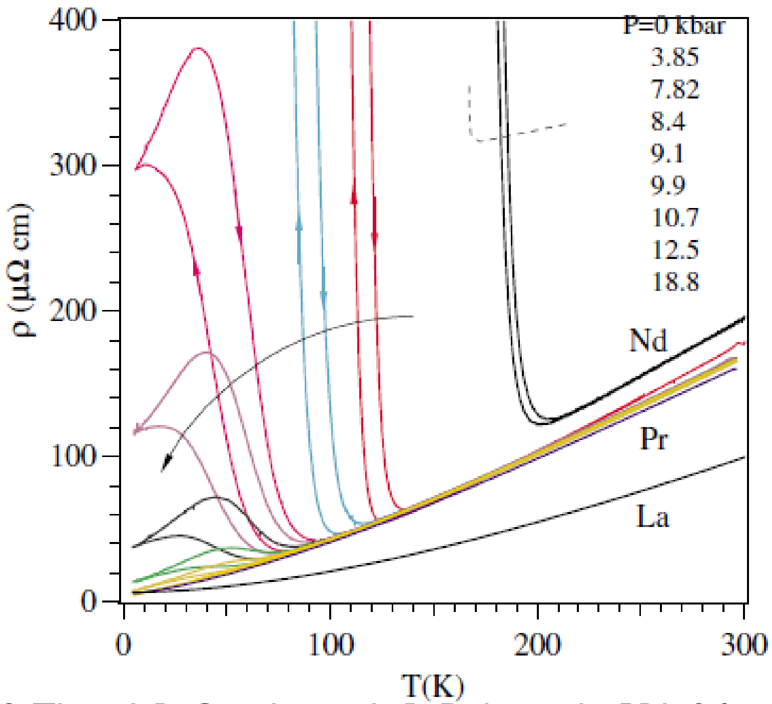
- Perhaps the Mott-Hubbard gap in the $GdTiO_3$ collapses near the interface? This was not allowed for in the self-consistent modeling!

MIT in rare-earth nickelates

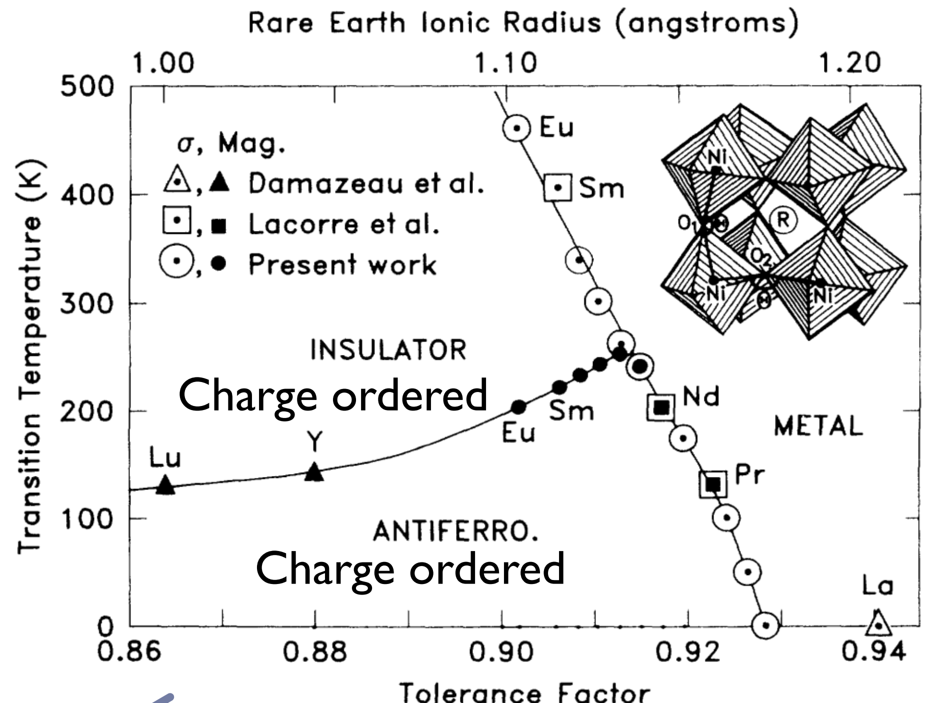
RNiO$_3$ (LaNiO$_3$, NdNiO$_3$)
- MIT vs. Temperature

Ni^{3+}, nominally d^7, e$_g^1$





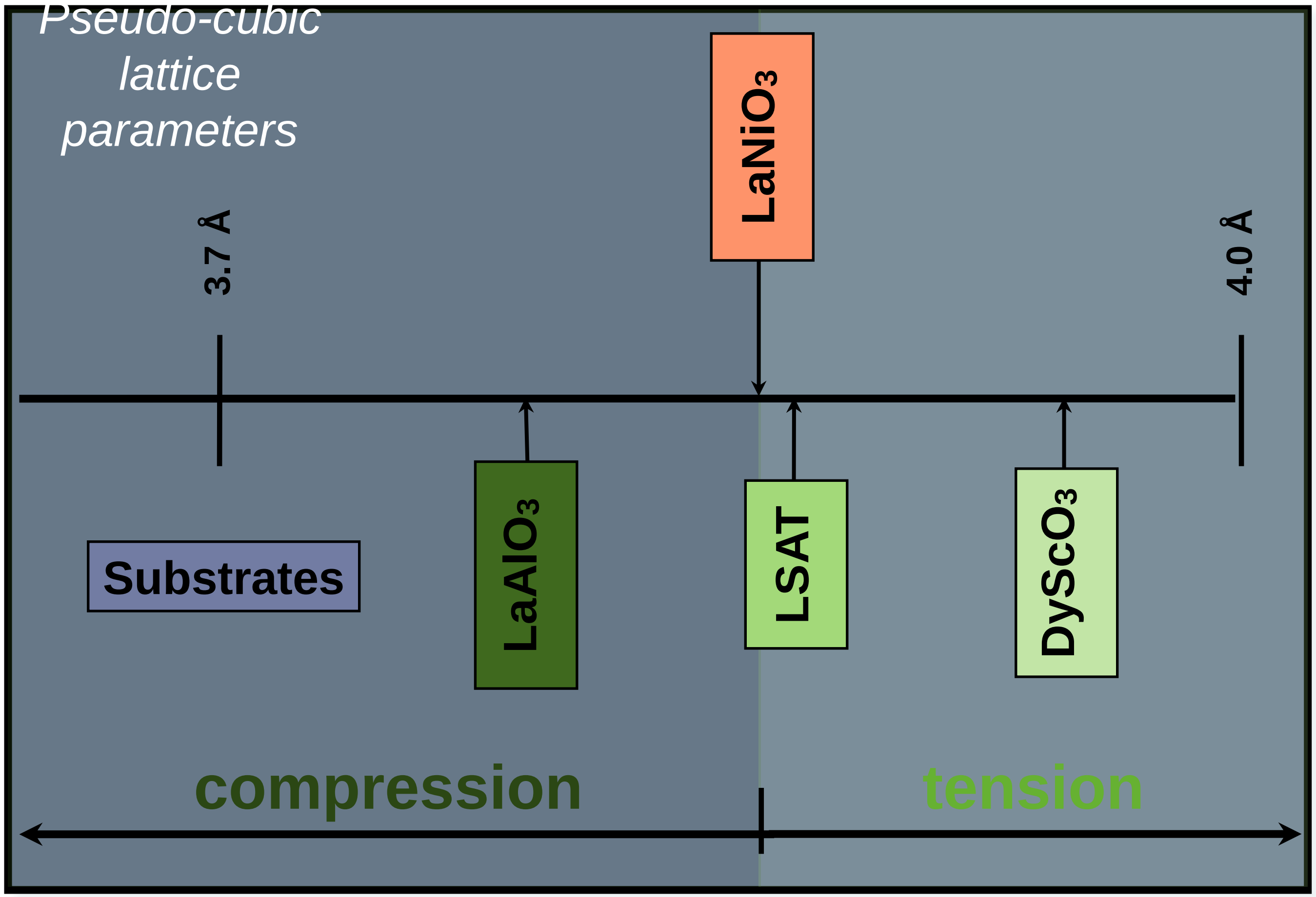
J.-S. Zhou, J. B. Goodenough, B. Dabrowski, PRL **94** 226602 (2005)



Increasing orthorhombic distortion

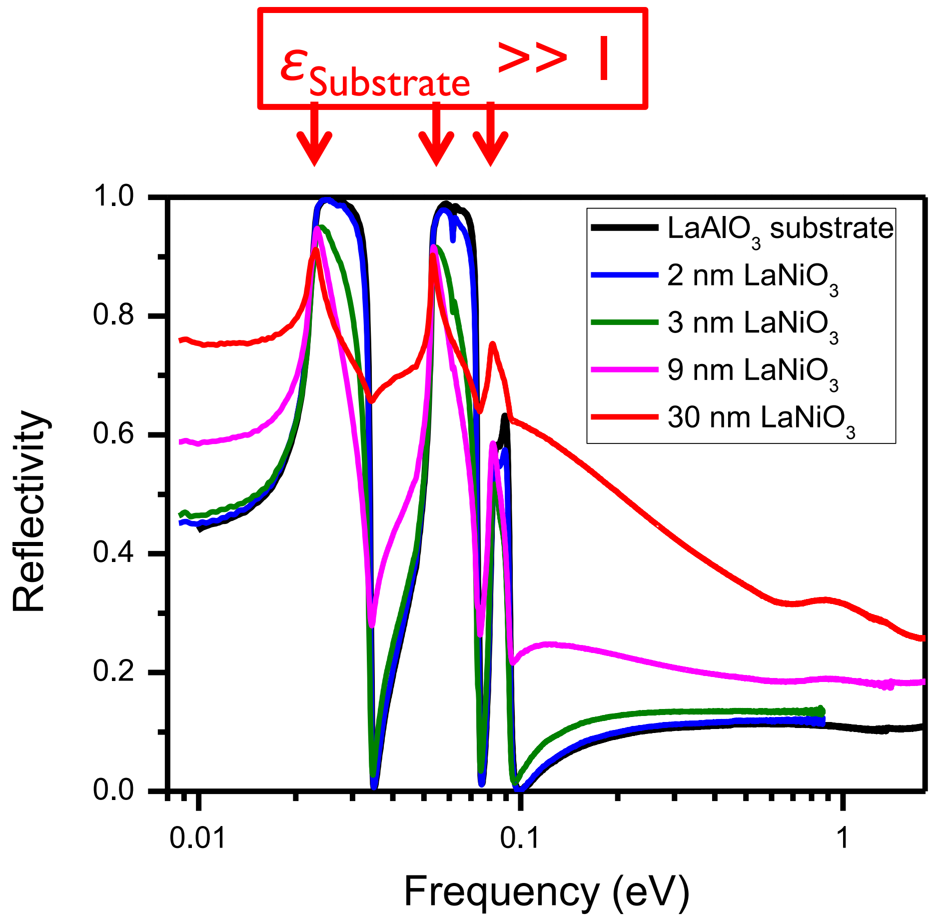
J.B. Torrence PRB **45**, 8209 (1992)

Bandwidth control using epitaxial strain



Optical reflectivity in FTIR

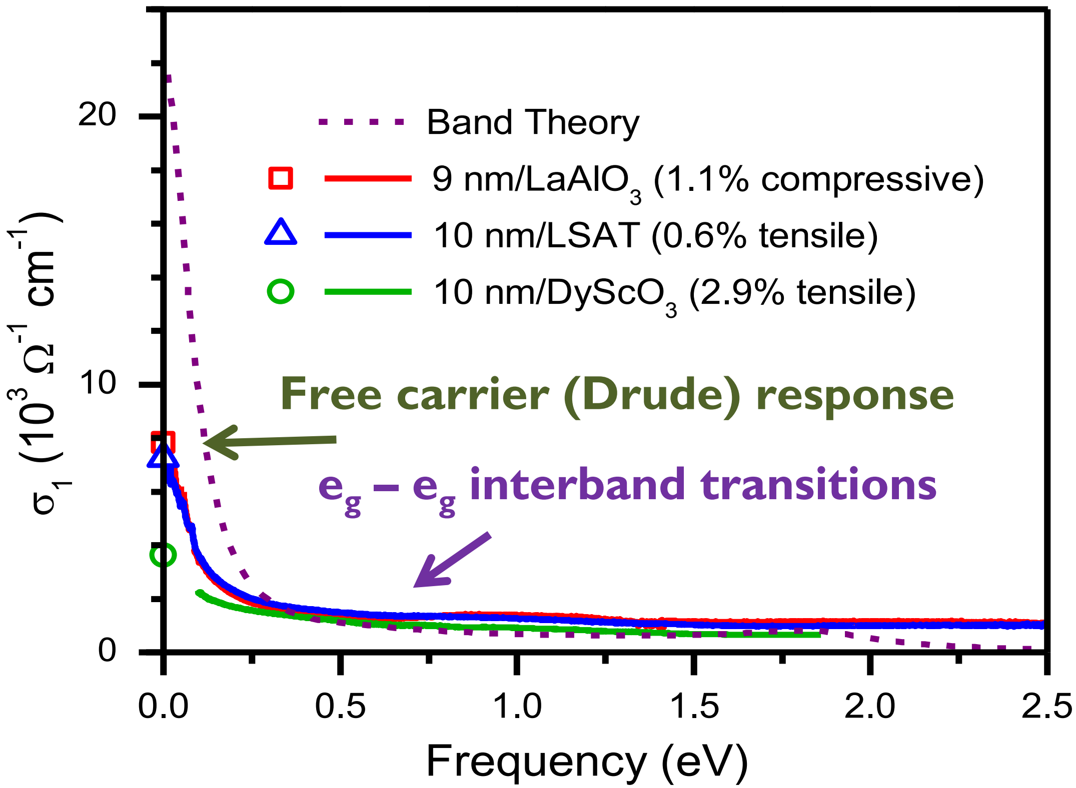
Substrate matters! Optical penetration >> film thickness



Direct inversion

- Model independent
- Impose Kramers-Kronig consistency
- Substrate artifacts remain

Optical conductivity of strained $LaNiO_3$

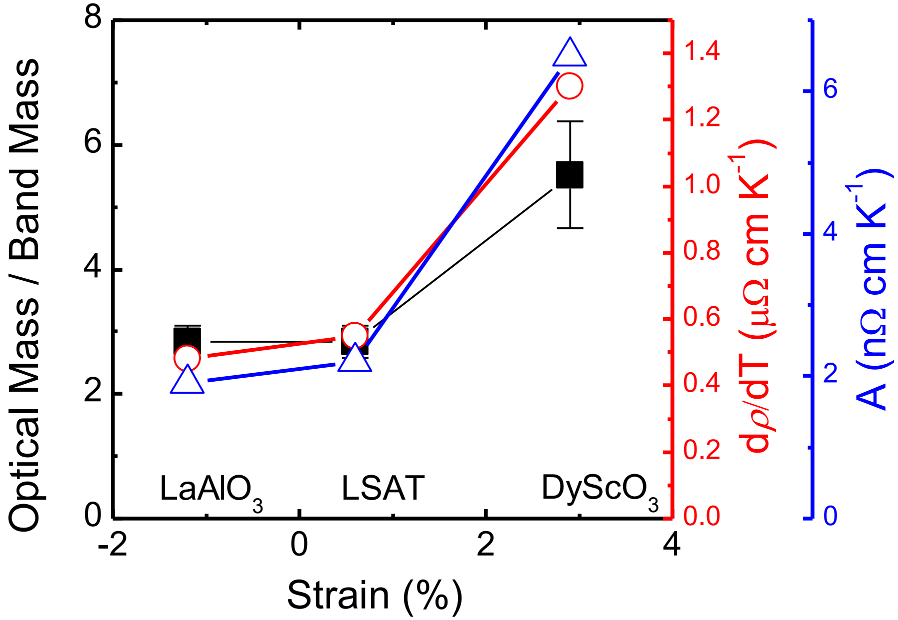


D. G. Ouellette *et al.* Phys. Rev. B **82** 165112 (2010)

In collaboration with SungBin Lee, Leon Balents, and Andy Millis (Columbia)

Carrier mass is renormalized by strong correlations not included in band theory.

Strain control of bandwidth

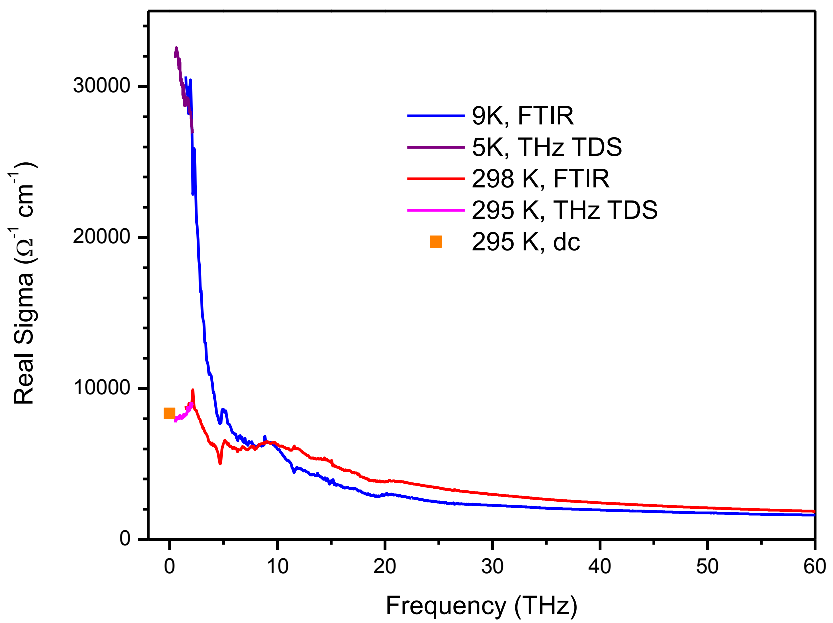


Optical mass, **quadratic** and **linear** contributions to resistivity show similar strain dependence.

Tensile strain reduces hopping, i.e. bandwidth, increasing the importance of electron correlations

D. G. Ouellette *et al.* Phys. Rev. B **82** 165112 (2010)

THz TDS: 30 nm $LaNiO_3$/LSAT



- Very good agreement between reflectivity measurements in the FTIR, THz TDS transmission, and dc transport
- At 300 K, evidently the conductivity increases with frequency below 2 THz. Could be a phonon contribution?

Summary: Rare-earth nickelates

Results

- Strong electron correlations enhance the mass ~ threefold.
- Bandwidth control using epitaxial strain
- Zero-bias tunneling anomaly: disorder or correlations?
- Band offsets from internal-photoemission

Outlook

- Modulated transport and absorption to study field effect in remotely doped $NdNiO_3$.
- Improved tunneling and internal photoemission measurements.

Thank You!

Allen Group

Prof. Jim Allen

Dan Ouellette

Alex Kozhanov

James Kally

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Vinh Nguyen

Prof. Chris Van-de-Walle
Anderson Janotti

Prof. Leon Balents
SungBin Lee

Prof. Andy Millis (Columbia)

Everyone in the MURI collaboration