Nanoscale Electrodynamics of Strongly Correlated Electron Materials

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06/10/2019  Simons Many Electron Collaboration Summer School
Why: Electronic, magnetic, and structural phase inhomogeneities are ubiquitous in strongly correlated quantum materials.

How: Optical Spectroscopy and optical near-field microscopy

What: Few examples:
- vanadium dioxide (VO$_2$),
- calcium ruthenium oxide (Ca$_2$RuO$_4$),
- strontium-doped lanthanum manganites (La$_{0.67}$Sr$_{0.33}$MnO$_3$).
Strongly correlated electron materials (SCEM)

Vanadates: (VO$_2$, $V_2O_3$); manganites (Pr$_{1-x}$Ca$_x$MnO$_3$, La$_{1-x}$Sr$_x$MnO$_3$); nickelates (NdNiO$_3$);
Iridiates, Ruthenates (Ca$_2$RuO$_4$, Ca$_3$Ru$_4$O$_7$), heavy fermion compounds (SmS etc.); High $T_C$ Superconductors (K-FeSe)
M₃ or T: a new triclinic phase intermediate between M₁ and M₂.
Optical conductivity of VO$_2$ film

Metal: $\sigma > 10^3$ (Ωcm)$^{-1}$
Insulator: $\sigma < 10^{-2}$ (Ωcm)$^{-1}$

VO$_2$ film on Al$_2$O$_3$

Spectral Weight transfer to lower frequencies

✓ Phase inhomogenity and phase separation

✓ Different electronic phases and structural phases coexistence

✓ Mesoscopic physics is important!!

Temperature dependent IR Near-field Study of highly oriented VO$_2$ films (on [110]$_R$ TiO$_2$)

Compare highly oriented VO$_2$ films to unstrained polycrystal VO$_2$ films across the insulator to metal transition.

Random electronic phase separation

Homogeneous insulator

Inhomogeneous bad metal

Rutile metal

Unidirectional phase separation

Anisotropic stripe state

Metallic islands
IR Near-field investigation of VO$_2$ films and single crystals

100nm VO$_2$ film on sapphire


300nm VO$_2$ film on [110]$_R$TiO$_2$


250nm VO$_2$ film on [100]$_R$TiO$_2$


Bulk single crystal VO$_2$

2D $\rightarrow$ 3D mapping

Cross-sectional scanning!!

M. K. Liu et al. APL 104, 121905 (2014)
Cross-sectional near-field imaging

$\text{VO}_2$ on $[100]_R \text{ TiO}_2$

M. K. Liu et al. APL 104, 121905 (2014)
Many interesting results for quantum materials (Selected results)

**La$_{0.67}$Sr$_{0.33}$MnO$_3$ on LaAlO$_3$ substrate (RT)**

Stripes only occur at a certain thickness!

**Ca$_2$RuO$_4$ single crystal**

**K$_x$Fe$_{2-y}$Se single crystal (RT)**

Percolative metal film

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**VO$_2$ films on TiO$_2$ (100) substrate**

1. T=50°C
2. T=55°C
3. T=60°C
4. T=70°C
5. T=75°C
6. T=85°C

**Percolative metal film**

**Stony Brook University**
Hierarchical control of length scales in inhomogeneous SCEMs

$\lambda / 2 \leftrightarrow \lambda \leftrightarrow I$

M. K. Liu, A. J. Sternbach, D. N. Basov, Reports on Progress in Physics (in publication, 2016)

- W. D. Wise et al., Nature Physics 4, 696 (2008). $Bi_{2-y}Pb_ySr_{2-z}La_zCuO_{6+x}$ (STM) (checkerboard-CDW)
- G. Campi et al., Nature 525, 359 (2015). $HgBa_{2}CuO_{4+y}$ (micro X-ray) (CDW-qd)
- F. Chen et al., Physical Review X 1, 021020 (2011). $K_xFe_{2-y}Se_2$ (PE) (SC-AFII)
- K. Lai et al., Science 329, 190 (2010). $Nd_{1/2}Sr_{1/2}MnO_3$ (MIM) (COO-I - FMM)
- M. M. Qazilbash et al., Science 318, 1750 (2007). $VO_2$ (SNOM) (M-I)
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Advanced IR-THz spectroscopy

- Energy gap
- Ultrasmall optics
- Conventional optics
- Temperature dependent spectral weight transfer
- Imaging Mesoscopic Physics
- Ultrafast THz switching \(\sim 1\text{ps}\)
- Disentangle e-e (\(<1\text{ps}\)) and e-\(\text{ph} (\geq 1\text{ps})\) interactions

1 THz \(\approx 10^{12}\) Hz \(\approx 1\) ps \(\approx 4\) meV \(\approx 300\) \(\mu\)m \(\approx 48\) K \(\approx 33.4\) cm\(^{-1}\)

Resolution of Near-field Optics
<table>
<thead>
<tr>
<th>Representative Microscopy Techniques</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lateral resolution</td>
</tr>
<tr>
<td>Dominant contrast</td>
</tr>
<tr>
<td>Representative Image</td>
</tr>
</tbody>
</table>
Why “Near-field”??

Far-field zone

In the strong coupling regime, the probe (tip) and the sample lose their own identities and become one “entangled” entity.
Asymmetric Michelson Interferometer

Moving mirror introduces path difference (reference)

beam splitter

fixed mirror

sample

detector

Reflected Amplitude and reflected phase!

Real and Imaginary part of the IR signal (or Reflected Amplitude and reflected phase)

Real and Imaginary part of dielectric function $\varepsilon$
Asymmetric Interferometer for Near-field Nano-Spectroscopy

Far field (background): $\Omega$

Near-field signal: $2\Omega, 3\Omega, \ldots$ (600, 900... kHz)

Moving mirror introduces path difference (reference)

Real and Imaginary part of the near-field signal (or Reflected Amplitude and reflected phase)

Real and Imaginary part of dielectric function $\varepsilon$ (with 10 nm resolution)
Fundamentally Different From
Nobel Prize in Chemistry for 2014

**Stimulated emission depletion (STED) microscopy**

Stefan Hell

**Single-molecule microscopy**

Eric Betzig and William Moerner
Extract dielectric constant below 10 nm?

\[ \varepsilon_r(\omega, r, q), \varepsilon_i(\omega, r, q) \]
Tip modeling

Increasing complexity

Dielectric function of the sample $\leftrightarrow$ scattered near-field signal

In collaboration with Alex McLeod (Columbia U) and Tat Chui (U of Delaware)


$\varepsilon_1$ and $\varepsilon_2$ dependence of the near-field signal (@ 10 $\mu$m)

(S$_3$ phase can be calculated as well)

Cone model with conformal mapping

PRB rapid communications, 97, 081406 (R) (2018)
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Real and imaginary near-field images of $\text{VO}_2/(110)_R \text{TiO}_2$

Measured with 10 μm CO$_2$ laser
\( \varepsilon(\omega = 10 \, \mu m) \) reconstruction!

- (a) \( \text{Exp Re}(S3) \)
- (b) \( \text{Exp Im}(S3) \)
- (c) \( \varepsilon_r > 0, \varepsilon_r < -1, \varepsilon_r \approx -1 \)
- (d) \( \varepsilon_i \approx 2, \varepsilon_i \approx 30, \varepsilon_i \approx 10 \)
Inhomogeneous strain versus inhomogeneous $\varepsilon$

$\varepsilon$ extracted from Near-field

$\varepsilon_r > 0$

$\varepsilon_r < -1$

$\varepsilon_r \approx -1$

Inplane micro-X-ray

Monoclinic

Rutile

High strain, intermediate state!!

Jiawei Zhang, Alex McLeod et al. under review in Nature Communications
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Insulator-metal-transition in single crystal Ca$_2$RuO$_4$
S, S’ and L phase in single crystal Ca$_2$RuO$_4$

In collaboration with Yoshi Maeno’s group
Polarity switching

Ca$_2$RuO$_4$
S, S’ and L phase in single crystal Ca$_2$RuO$_4$
S, S’ and L phase in single crystal Ca$_2$RuO$_4$
Near-field imaging of phase boundary in Ca$_2$RuO$_4$
Near-field imaging of phase boundary in $\text{Ca}_2\text{RuO}_4$
Lattice parameter change in Ca$_2$RuO$_4$

During the IMT, $a$ shrinks 1%, $b$ shrinks almost 3%
Near-field imaging of phase boundary in Ca$_2$RuO$_4$

Jiawei Zhang, Alex McLeod, Qiang Han et al. PRX 9, 011032 (2019)
Low energy properties of Ca214 is still not clear!

THz frequency is required!

(1 THz ≈ 4 meV ≈ 300 μm ≈ 33 cm⁻¹)
We didn’t observe striped phase front in VO$_2$ bulk single crystals

*Insulator-metal phase boundary formed due to thermal gradient*
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La$_{0.67}$Sr$_{0.33}$MnO$_3$

Phase diagram

Transport measurements

Construct Electronic Moiré pattern in transition metal oxides

Twin-induced electronic stripes

Miscut-induced electronic stripes

Electronic Moiré pattern in transition metal oxides

Rhombohedral domains

OC-SEM

AFM

Homogeneous

Twinning of LSMO

Miscuts in LAO

MS

Moiré pattern
Emergence of Moiré fringes

Miscut stripes

Domain stripes
Systematic evolution of the Moiré fringes
Emergence of curved Moiré pattern (on a different film)
Emergence of curved Moiré pattern (on a different film)
Looking into the future
Table Top vs Beamlines

**Broadband Near-field IR spectroscopy**

**Photon Energy [meV]**

**Wavelength [µm]**

**Brightness [Watts/cm²/mrad²/cm²]**

**Frequency [cm⁻¹]**

- **Table Top broadband (5 mW)**
- **ALS broadband nano-IR (0.5 mW)**
- **NSLS-II** Designed to have the lowest noise
Broadband Nano-IR: Motivation:
IR active phonon modes in VO$_2$

Monitor not only electronic phase transition but also structural phase transition with 10nm resolution!!!
Energy-length-time diagram

Low Temp, high magnetic field!
Group Members and collaborators

- **Broadband nano-IR & gold film**: Michael C. Martin, Hans A. Bechtel (ALS); Larry Carr (BNL);
- **VO₂**: D. N. Basov, A. McLeod (Columbia University)
  Paul C. Canfield, Sergey L. Bud’ko, Tai Kong (Iowa State University);
  Stuart A. Wolf, Jiwei Lu, Salinporn Kittiwatanakul (University of Virginia);
  Hyun-Tak Kim, Tetiana V. Slusar (Korean University of Science and Technology);
  Haidan Wen (APS, Argonne National Lab), Hu (Tiger) Tao (UT Austin)
- **Ca₂RuO₄**: Yoshiteru Maeno (Kyoto University (Japan)); Andrew Millis (Columbia University)
- **La₀.₆₇Sr₀.₃₃MnO₃**: Changgan Zeng (USTC); D. N. Basov (Columbia U.); Andrew Millis (Columbia University)