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Optical properties of nickelate heterostructures

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www.elettra.eu

Oxide heterostructures

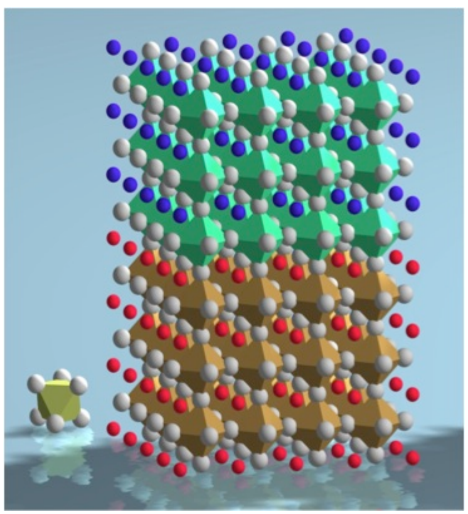
2-dimensionality

lattice polarizability

several parameters

strain

doping



Creation and control of spin and charge

metal to insulator transition

high T_c superconductivity

spintronics

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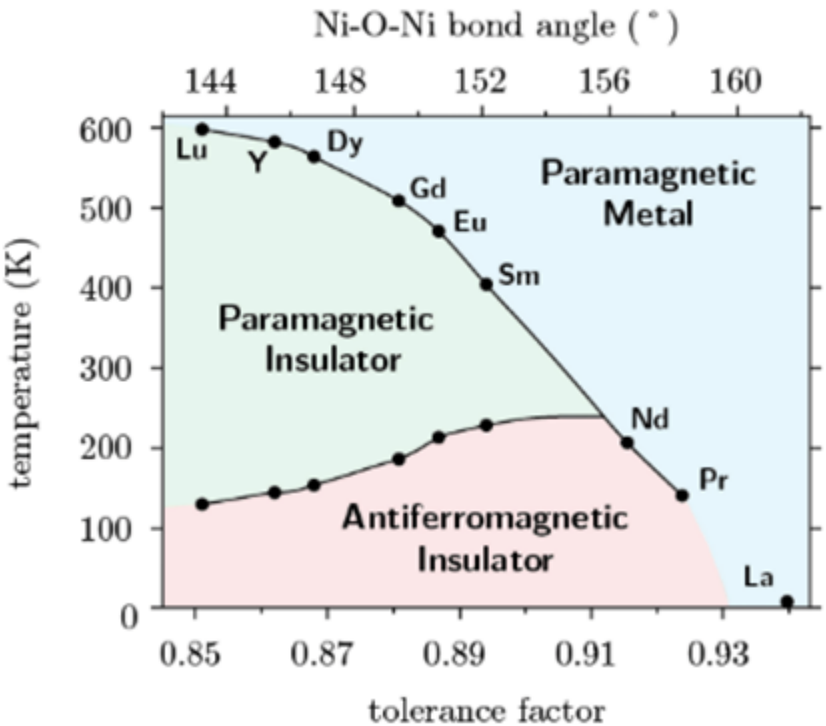
Motivation: nickelate $LaNiO_3$

$LaNiO_3$ is the only nickelate that does not undergo a metal to insulator transition

It does not exhibit a AF-insulating ground state!

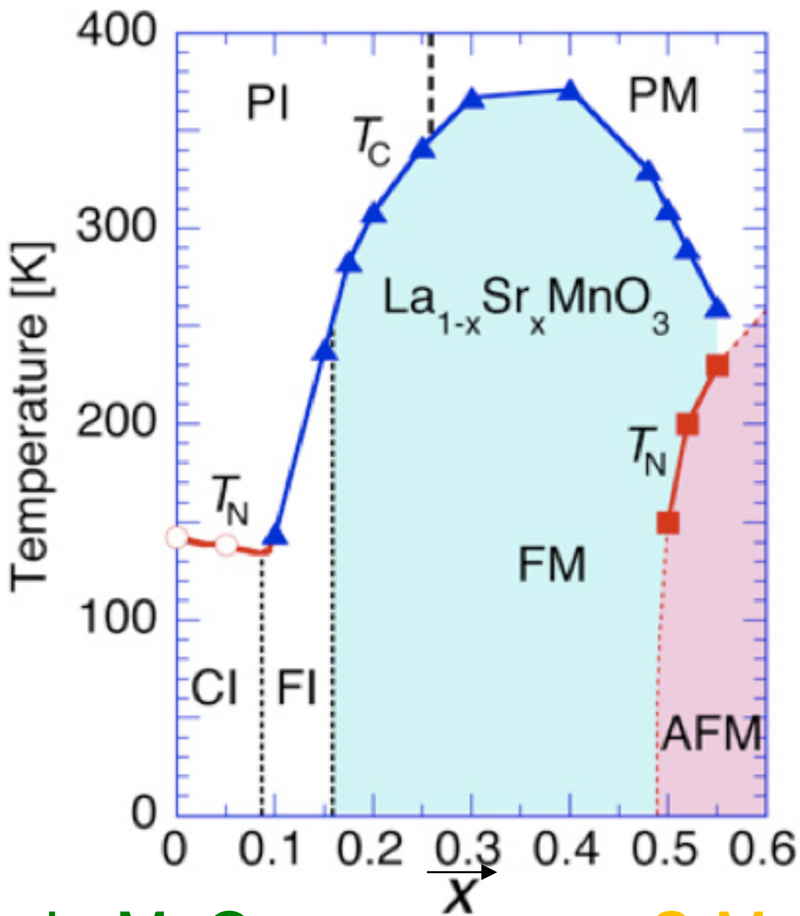


Controlling the $LaNiO_3$ ground state with

- ✓ Interfacial doping (**LNO/LMO**)
- ✓ Dimensionality (**LNO/LAO**)
- ✓ THz light (opportunities @TeraFERMI)

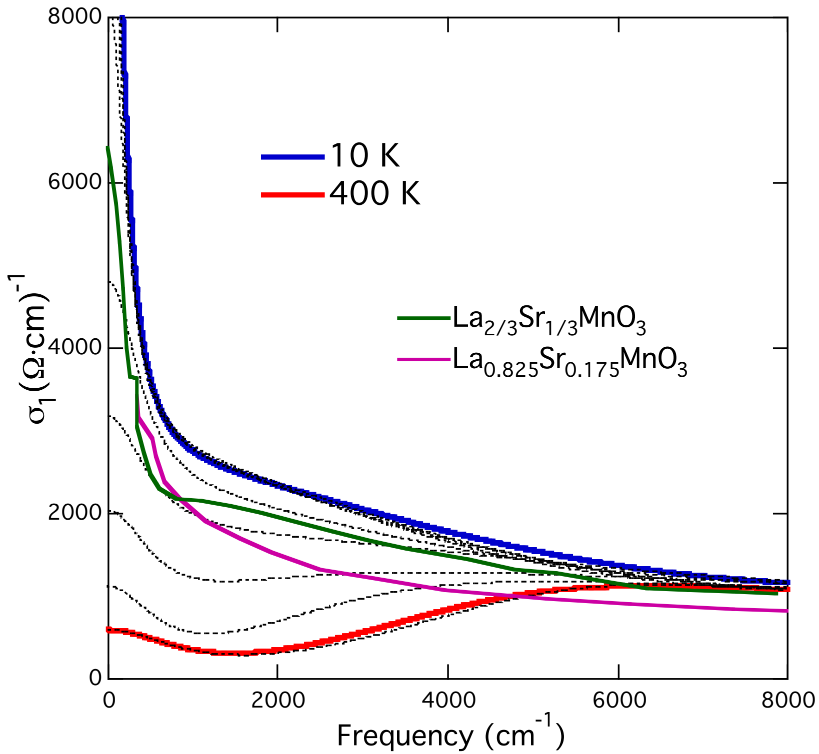
$(LaNiO_3)_n/(LaMnO_3)_2$ superlattices
n=2,3,4,5

Interfacial doping: the case of LMO/SMO



$LaMnO_3$ (Mn^{3+})

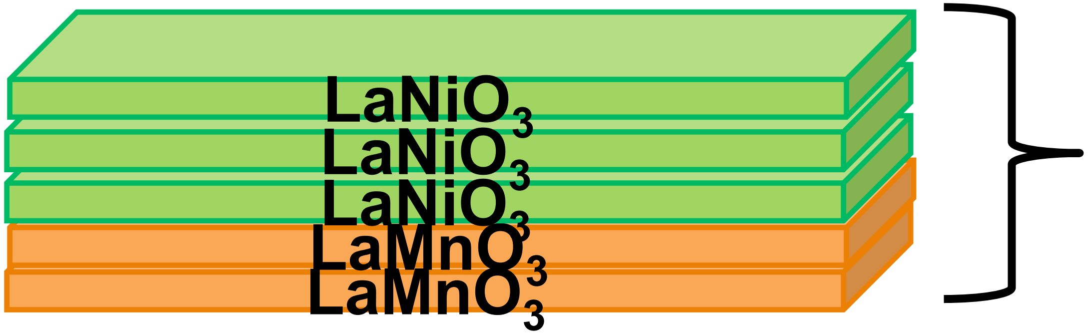
$SrMnO_3$ (Mn^{4+})



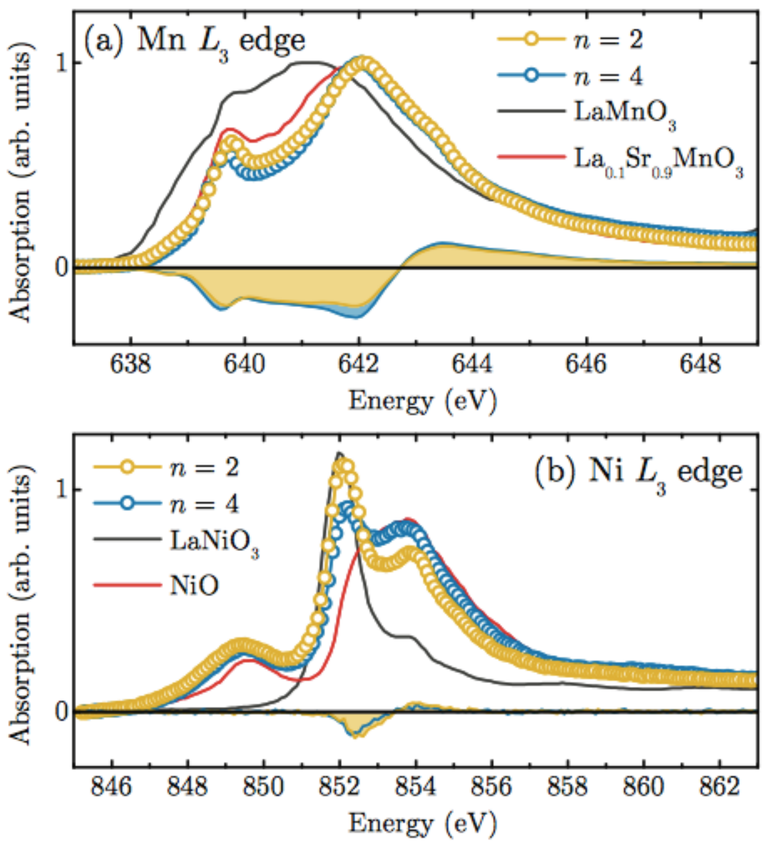
Perucchi et al., Nano Letters 2010

The electronic properties of $(LMO)_2/(SMO)_1$ SL are fully equivalent to those of the corresponding alloy $La_{2/3}Sr_{1/3}MnO_3$

LNO/LMO superlattice

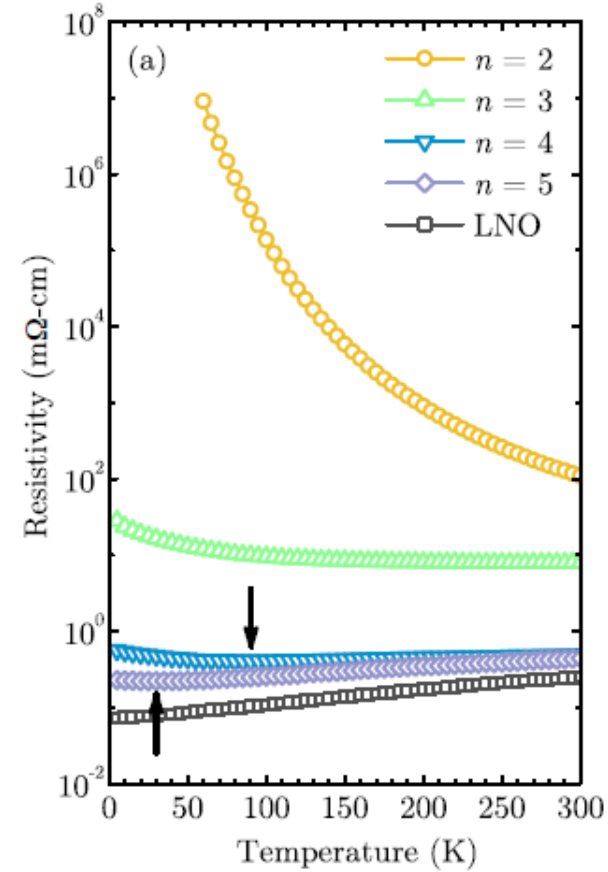


$(LaNiO_3)_n/(LaMnO_3)_2$
n=2, 3, 4, 5



With increasing $LaNiO_3$ thickness, the SLs undergo an **insulator-to metal transition**

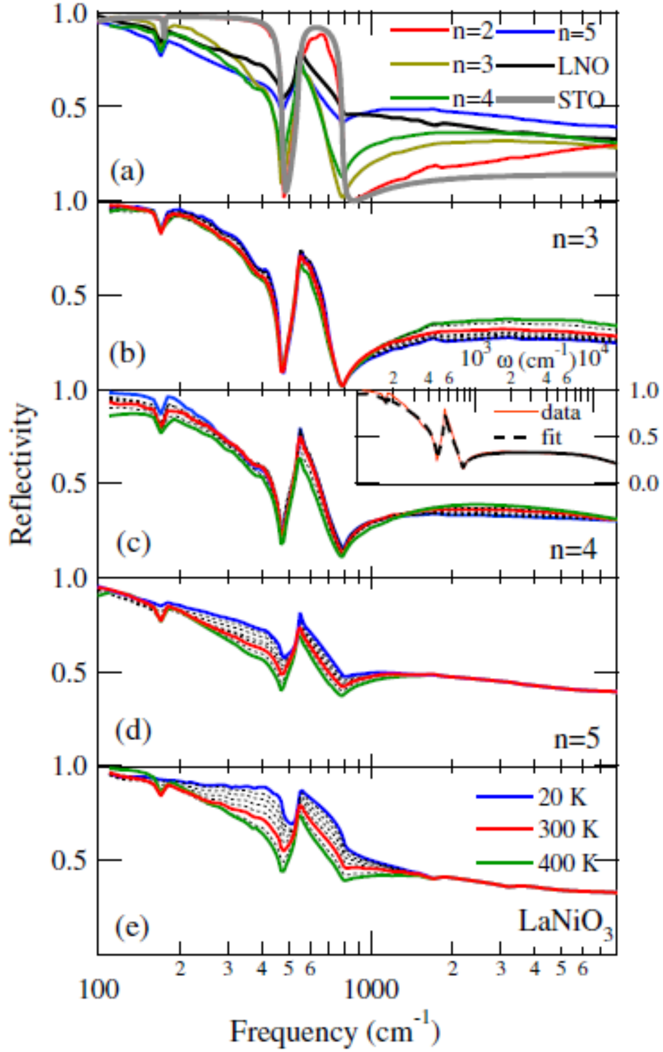
X-ray spectroscopy of this system shows that the Mn oxidation state is converted from 3+ to 4+, while Ni is intermediate between 2+ and 3+



Hoffman et al., Phys Rev B 2013

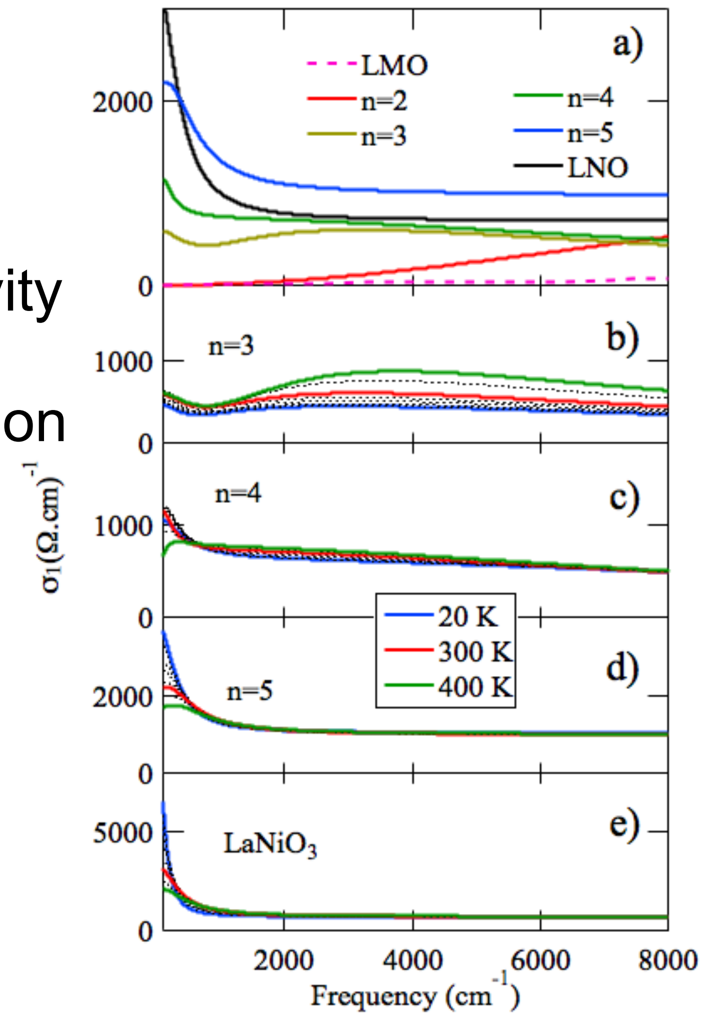
Reflectivity and Optical Conductivity

$(LaNiO_3)_n/(LaMnO_3)_2$ n=2, 3, 4, 5



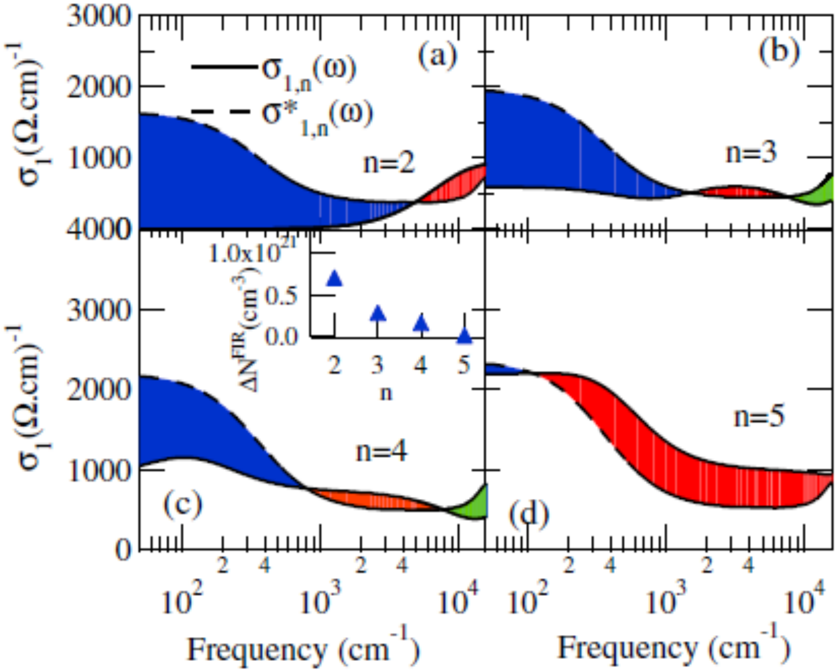
Di Pietro et al., Phys Rev Lett 2015

as n increases:
the optical conductivity increases:
onset of a metallization



Average Optical Conductivity

$(LaNiO_3)_n/(LaMnO_3)_2$ n=2, 3, 4, 5



Di Pietro et al., Phys Rev Lett 2015

$$\sigma^*_{1,n} = \frac{n\sigma_{1,\mathrm{LNO}} + 2\sigma_{1,\mathrm{LMO}}}{n+2}$$

The comparison between $\sigma_{1,n}^*$ and $\sigma_{1,n}$ allows us to single out the features directly related to the **interfaces**

The red area is always larger than the blue area

The additional MIR spectral weight is only in part due to the loss of SW at low frequencies, and a redistribution from the green area is also at play

Average Optical Conductivity

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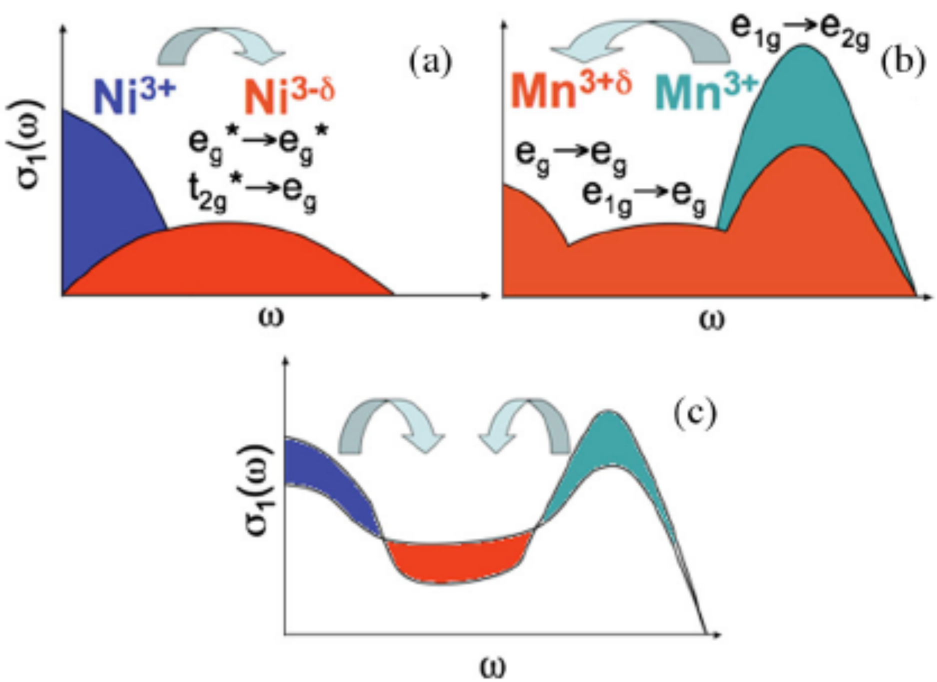
$(LaNiO_3)_n/(LaMnO_3)_2$ n=2, 3, 4, 5



In the SLs the valence of **Ni** decreases from its nominal Ni 3+ value, as n decreases

⬇

MIT at Ni valence 2.75

In the SLs **Mn** valence increases from 3+ to a higher value: polaronic states appear in the MIR, whose SW is taken from the Jahn-Teller band at 2.5 eV

introduction of LMO layers in the SL, by reducing the Ni valence, progressively depletes the low energy, coherent SW of LNO, which piles up in the infrared at expenses of FIR and VIS/UV

Di Pietro et al., Phys Rev Lett 2015

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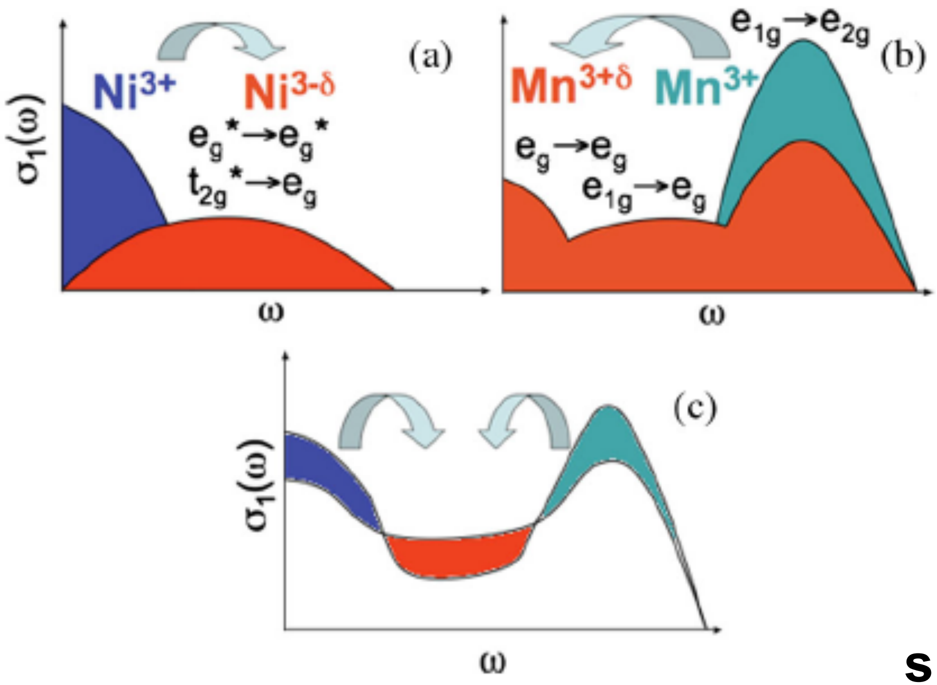
Average Optical Conductivity

$(LaNiO_3)_n/(LaMnO_3)_2$ n=2, 3, 4, 5



Di Pietro et al., Phys Rev Lett 2015

Infrared data show that the LNO/LMO SLs display the presence of significant midinfrared excitations that are not present in LNO or LMO alone.

Interfacial charge redistribution is identified as the origin of changes to the MIR spectral response.

This is in contrast to what is observed in **ultrathin LNO films** and **in LNO/LAO superlattices**, where localization is believed to occur due to dimensional confinement and enhanced correlations…

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($LaNiO_3$) thin films and ($LaNiO_3$) /($LaAlO_3$) superlattice

ARPES results on LNO thin films

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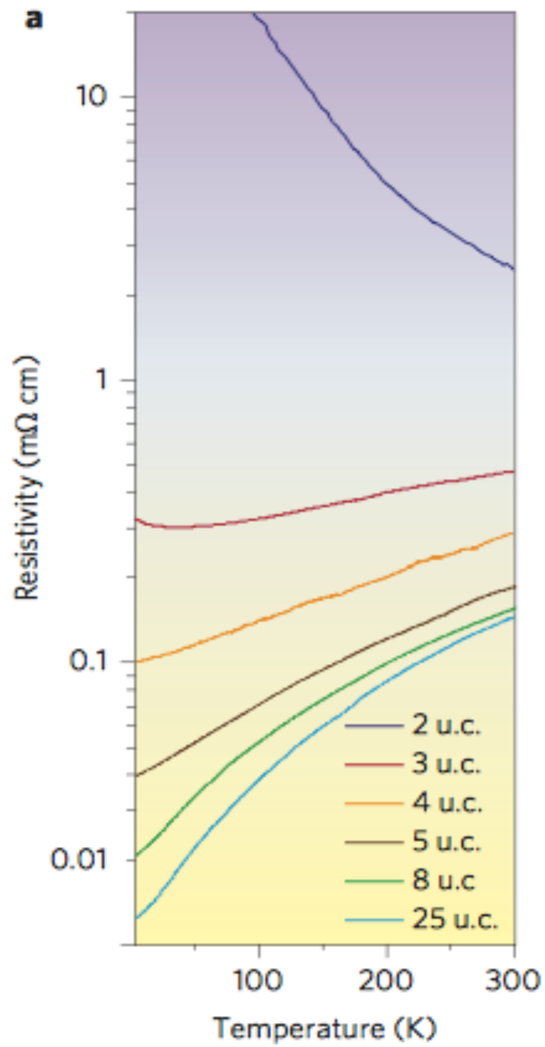
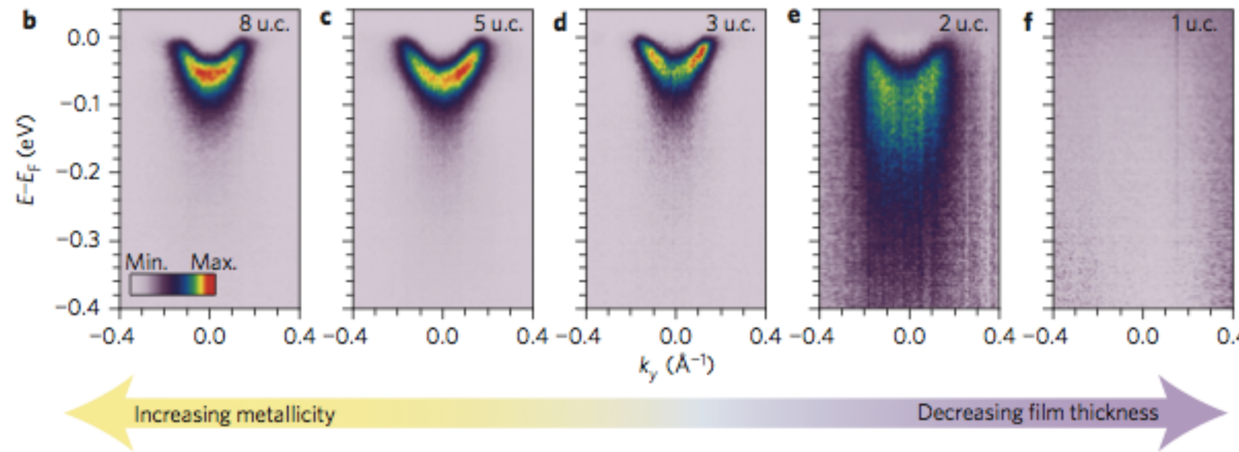
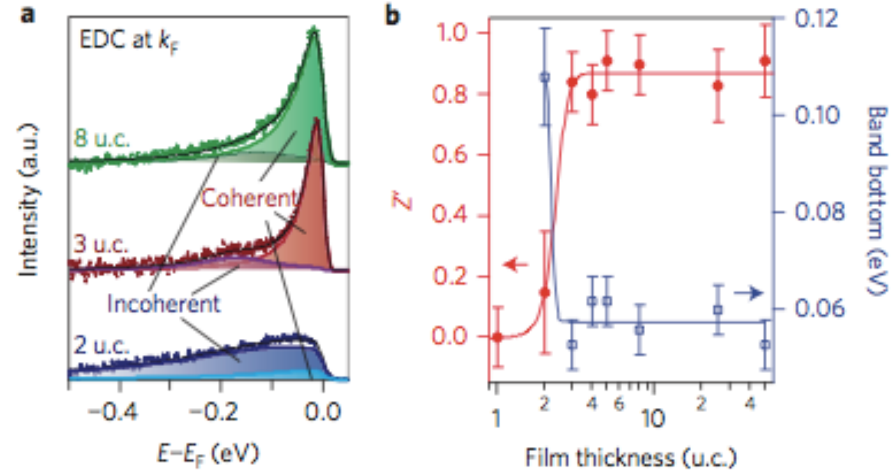


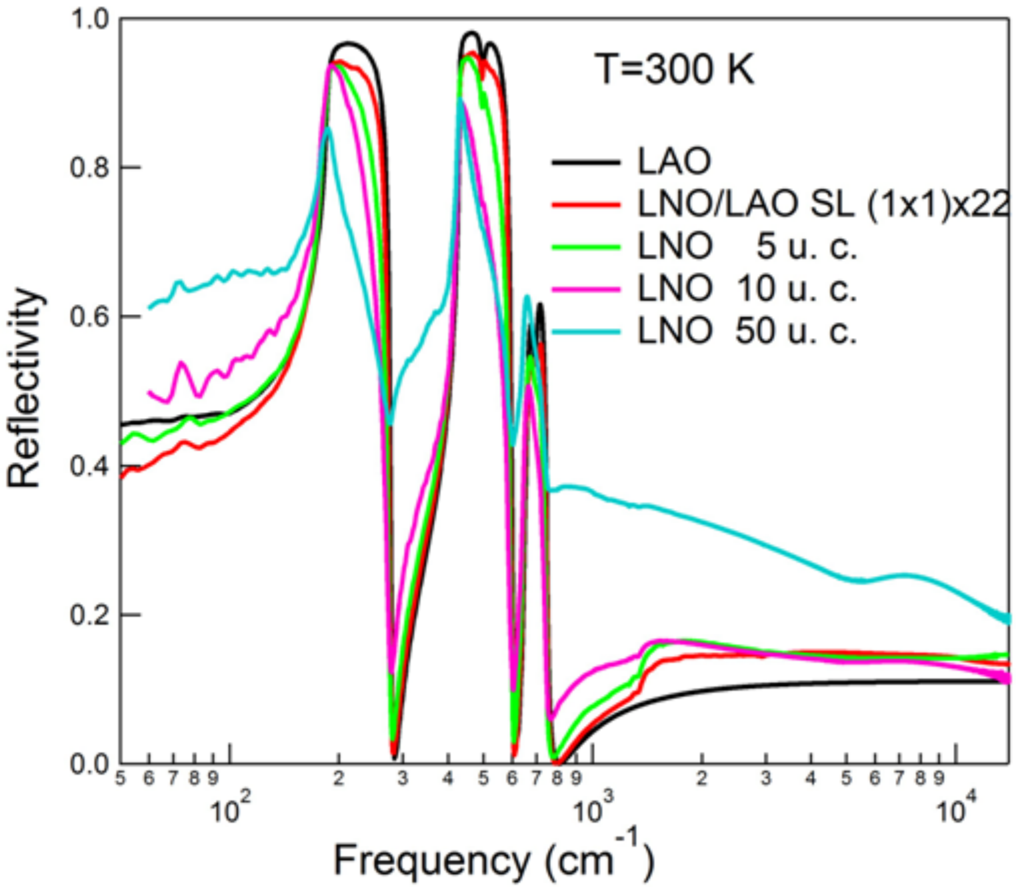
King et al., Nature Nano 2014





abrupt destruction of Fermi liquid-like quasiparticles in the correlated metal LNO when confined to a critical film thickness of 2 unit cells

LNO thin films and LNO/LAO SL: reflectivities

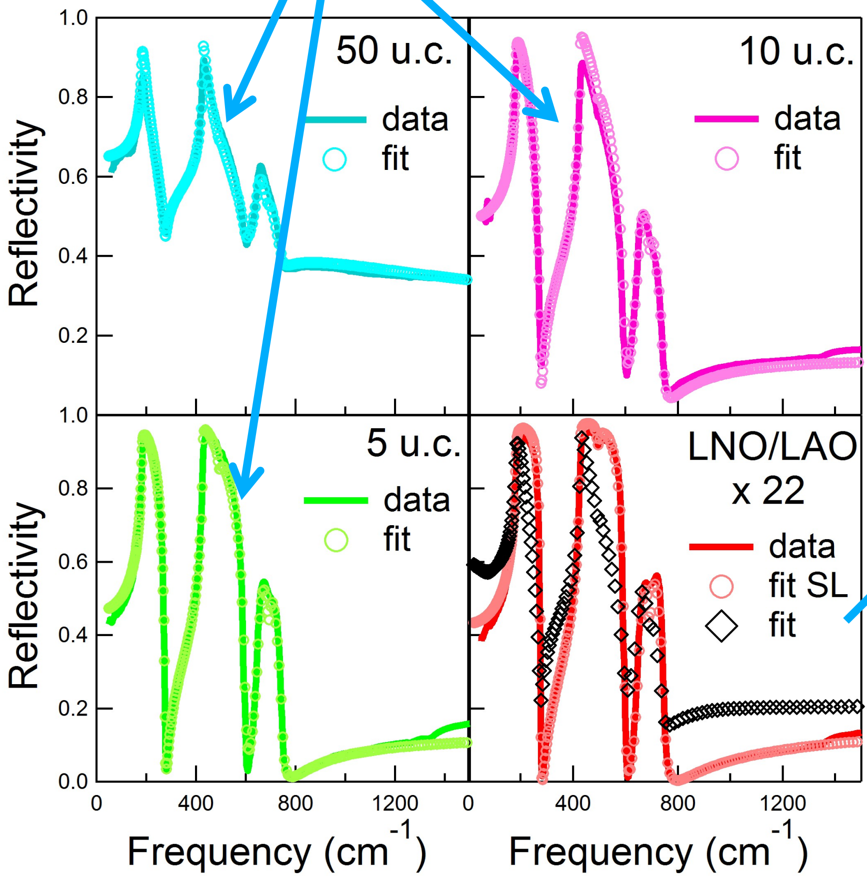


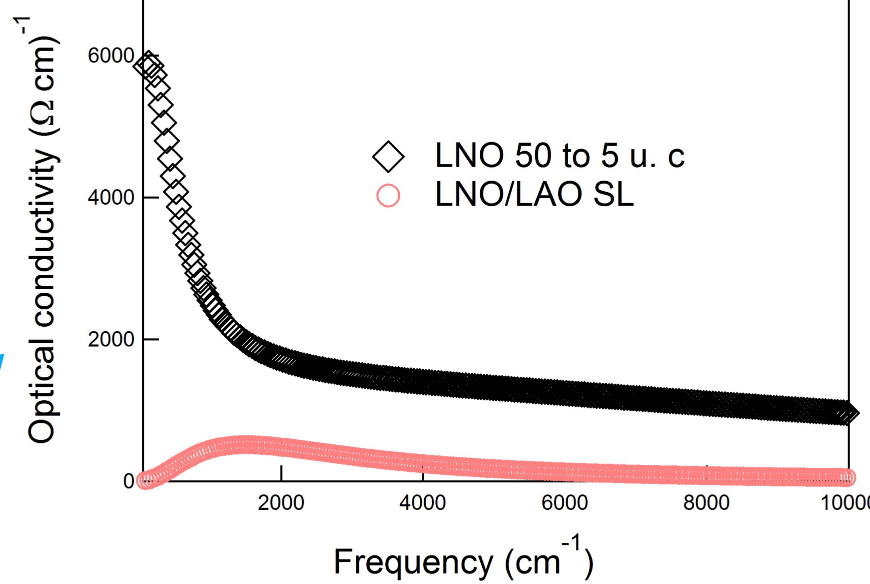
Fitting this data to a Drude-Lorentz model we can show the effect of the thickness in LNO....

Dimensionality-driven MIT

T=300 K

Same Drude-Lorentz parameters!





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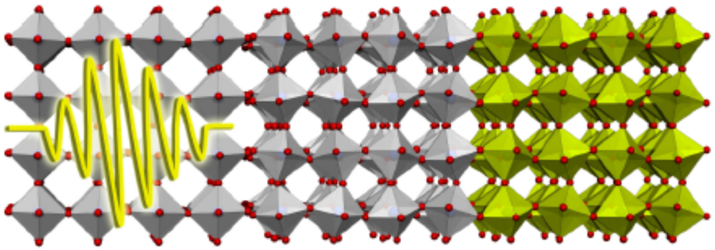
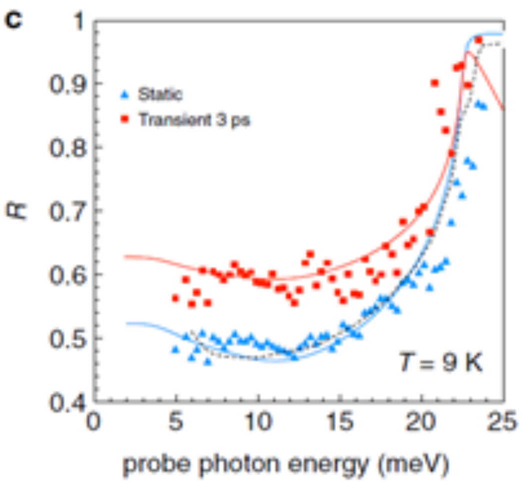
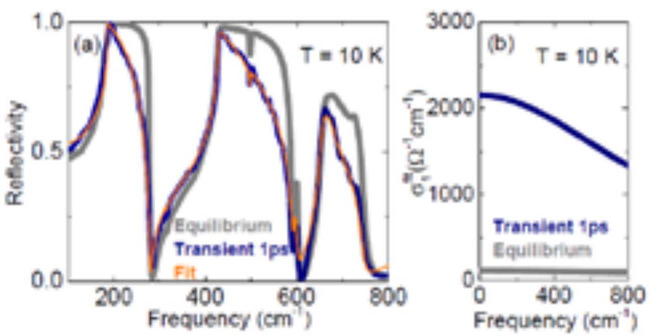
THz control in Nickelate heterostructure

THz-induced MIT





Caviglia et al., Phys Rev Lett 2012



Hu et al., arXiv 2016

Nickelates
are very sensitive to **strain**
due to different substrates

a MIT transition in **$NdNiO_3$** and **$SmNiO_3$** is induced by pumping on the lattice of the **LAO** substrate

Exploiting all available phonon modes (substrate + film) to induce the MIT!

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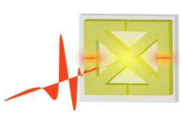
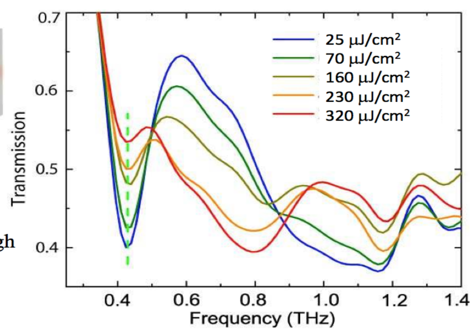
The TeraFERMI beamline

THz control of matter

THz light couples with electronic, vibrational and magnetic excitations

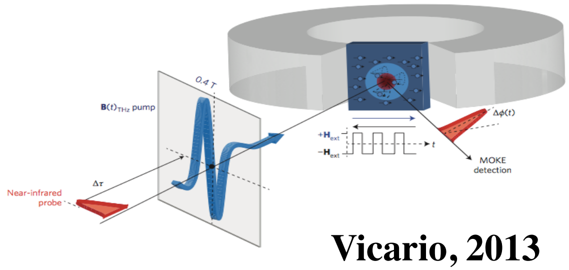
Giant quasi-static fields





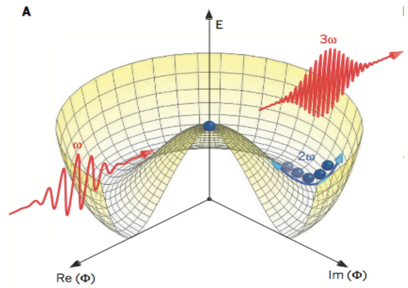
THz resonance disappears at high THz fluence

Liu, 2012

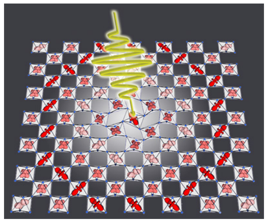


Vicario, 2013

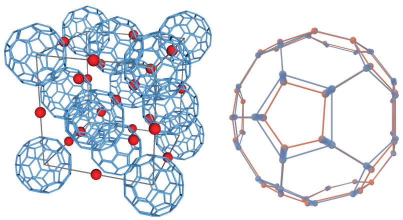
Resonant excitations



Matsunaga, 2014



Rini, 2007



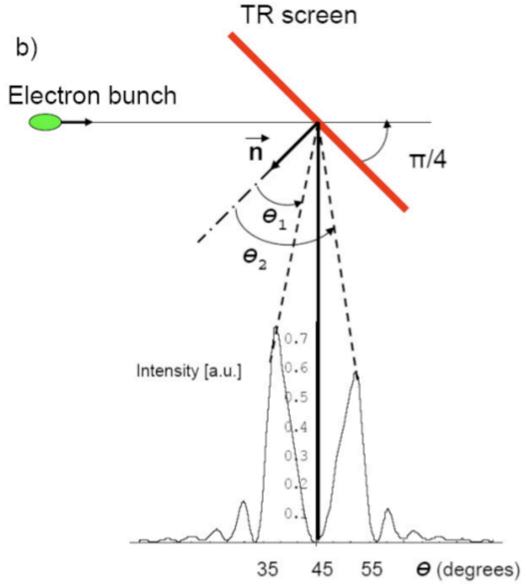
Mitrano, 2016

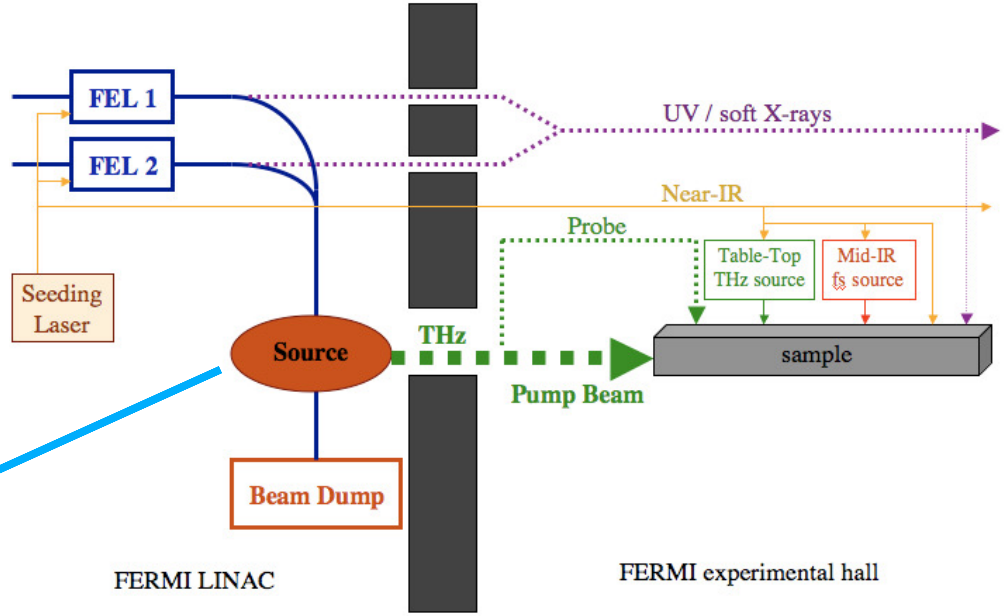
The TeraFERMI idea

Exploiting the properties of the FERMI-FEL electron beam to produce
Short (sub-ps), Powerful (>MV/cm), Broadband (0.1-15 THz) THz pulses

to be used as a **Pump** beam for ***ultrafast nonlinear spectroscopies***

Transition Radiation occurs when relativistic electrons cross the boundary between two media of different dielectric constant



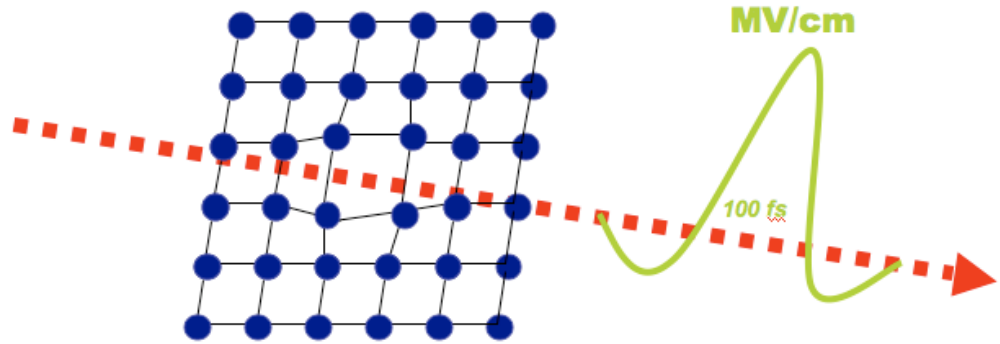


TeraFERMI will not affect overall FEL available beamtime:
THz light always available
Possibility for THz pump / FEL probe

Beamline parameters

Ultra-short, high-power THz pulses between 1 mm - 20 μm (0.1 -15 THz)
Access to the Reststrahlen-band gap!
Pumping on electronic, vibrational, magnetic excitations

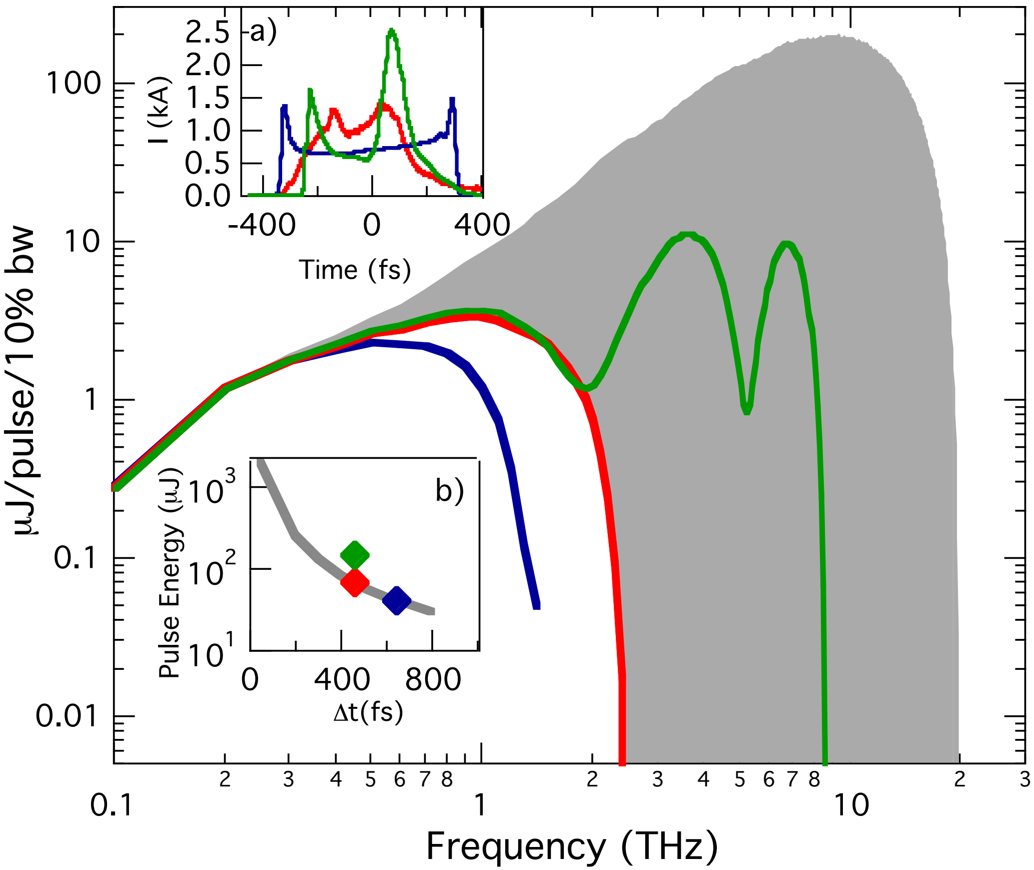
Wavelength range	3 mm - 20 μm (0.1 - 15 THz)
THz pulse energy	50 μJ - mJ
Operation Conditions	0.9-1.5 GeV FEL1/FEL2 10 - 50 Hz 700 pC



FEL1: 65-20 nm

FEL2: 20-4 nm

Seeded FEL operation



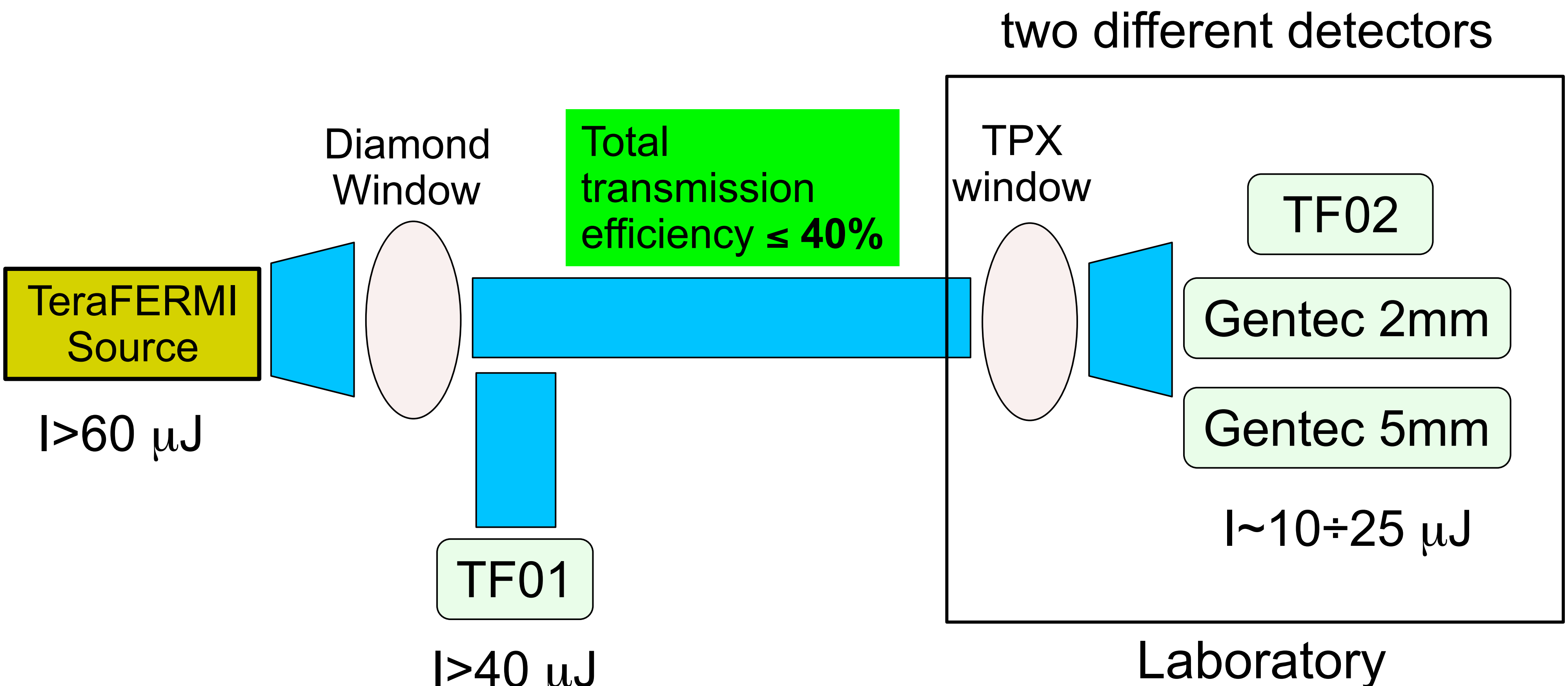
Expected performances

Blue: as expected at the entrance of FEL1

Red: predicted at TeraFERMI extraction point (FEL Off)

Green: predicted at TeraFERMI extraction point (FEL On)

Integrated power



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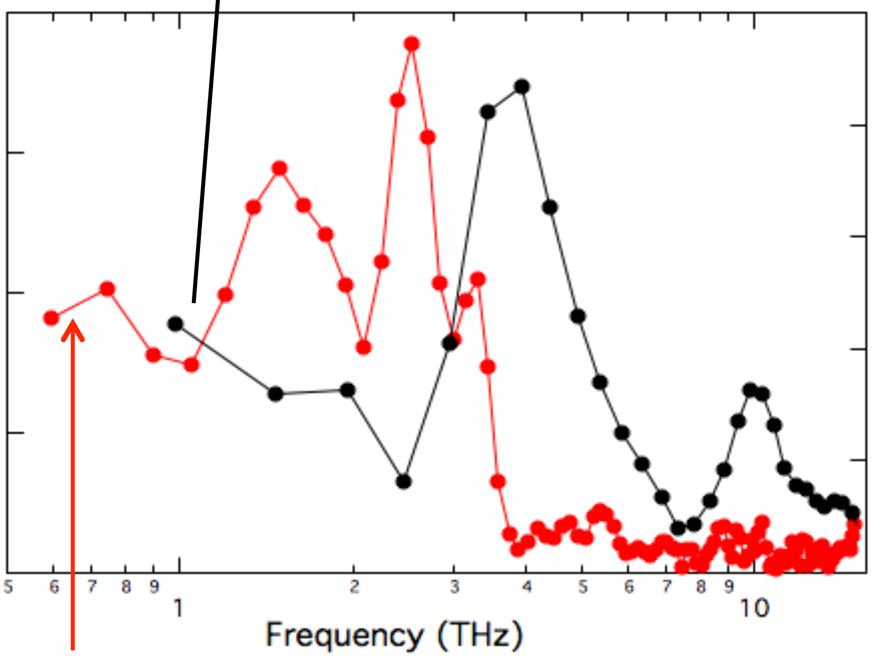
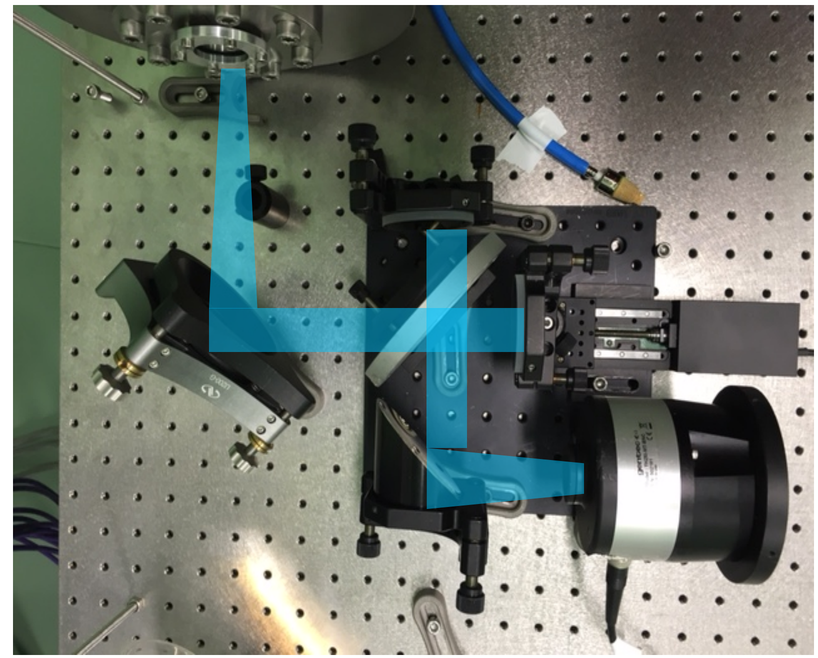
Spectral content

TeraFERMI commissioning shift (**March 2016**)



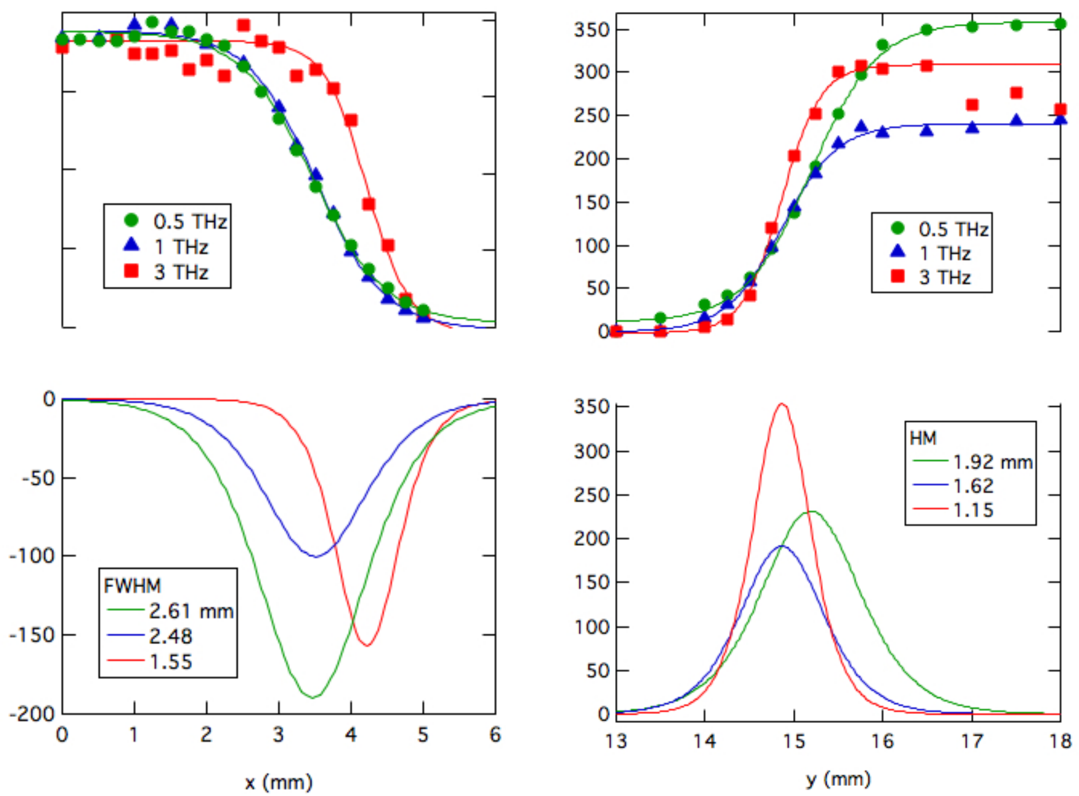
e-beam optimized for FEL users, flat current profile at the undulator (**April 2016**)

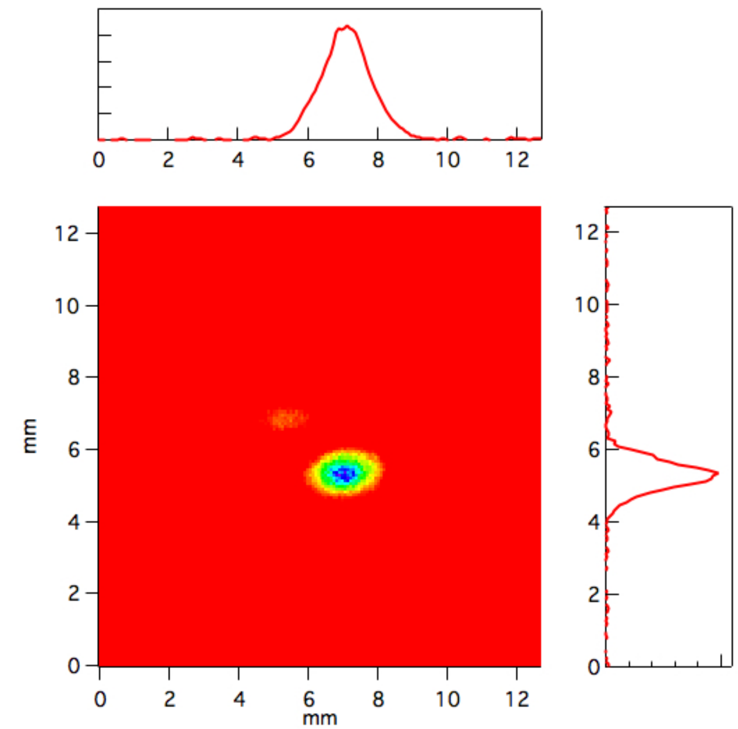


ΔL = 0.3 mm → $\Delta\omega_{res}$ = 0.5 THz
δL = 3 μm → N = 100 pts
30 averages for each point
1 Spectrum in ~ 5 minutes @ 10Hz

Focusing

For 10 μJ/pulse
~MV/cm





THz beam profile at sample position performed with the help of a **Pyrocam IIIHR camera.**

Conclusions

- ✓ Infrared data show that the **LNO/LMO SLs** display the presence of significant mid-infrared excitations that are not present in LNO or LMO alone
- ✓ Interfacial charge redistribution is identified as the origin of changes in the MIR spectral response of LNO/LMO
at variance with LNO thin films
- ✓ LNO heterostructures are a great platform to manipulate the interplay of electronic magnetic and vibrational degrees of freedom

opportunity to THz control of matter **@TeraFERMI**

Perspectives @TeraFERMI

- Synchronizing an external laser source
- Electro-optic sampling
- THz pump - NIR probe (780 and 1560 nm)

Acknowledgments

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