Modulation of manganite nano-film properties mediated by strong influence of strontium titanate excitons

Chi Sin Tang[†,Ⅱ,#], Xinmao Yin[†,‡,Ⅱ], Le Wang[⊥], Mark B. H. Breese[†,Ⅱ], Andrew T. S. Wee[Ⅱ,#,∇], Junling Wang[⊥,*], and Andrivo Rusydi[†,§,Ⅱ,#,*]

[†]Singapore Synchrotron Light Source (SSLS), National University of Singapore, 5 Research Link, 117603, Singapore
[§]NUSNNI-NanoCore, National University of Singapore, 117411, Singapore
[Ⅱ]Department of Physics, Faculty of Science, National University of Singapore, 117542, Singapore
[⊥]School of Materials Science and Engineering, Nanyang Technological University, 639798, Singapore
[#]NUS Graduate School for Integrative Sciences and Engineering, 117456, Singapore
[∇]Centre for Advanced 2D Materials and Graphene Research Centre, National University of Singapore, 117551, Singapore

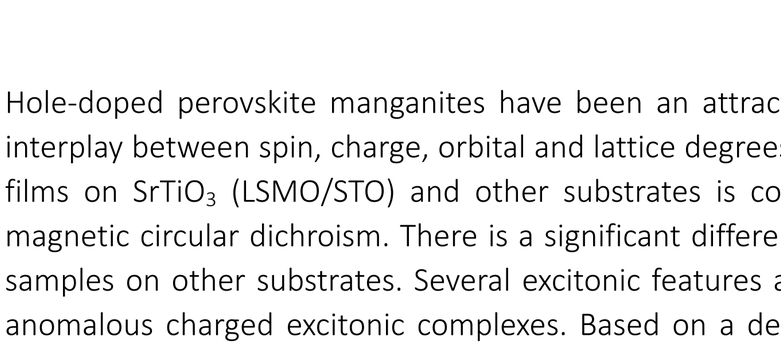
Abstract

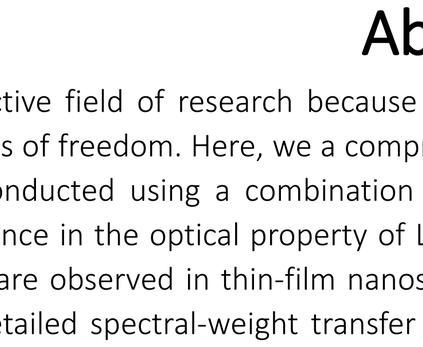
Hole-doped perovskite manganites have been an attractive field of research because of their unique manifestation of optical, electronic and magnetic properties induced by the interplay between spin, charge, orbital and lattice degrees of freedom. Here, we a comprehensive investigation of the optical, electronic and magnetic properties of $La_{0.7}Sr_{0.3}MnO_3$ thin-films on $SrTiO_3$ (LSMO/STO) and other substrates is conducted using a combination of temperature-dependent transport, ellipsometry, X-ray absorption spectroscopy and X-ray magnetic circular dichroism. There is a significant difference in the optical property of LSMO/STO that occurs even in LSMO/STO as thick as ~87nm. This is in comparison with LSMO samples on other substrates. Several excitonic features are observed in thin-film nanostructure LSMO/STO in the energy regime of ~4eV. This can be attributed to the formation of anomalous charged excitonic complexes. Based on a detailed spectral-weight transfer analysis, we deduce that the anomalous excitonic effects from STO substrate strengthen the electronic-correlation in LSMO films, thus, resulting in the occurrence of optical spectral changes related to the intrinsic Mott-Hubbard properties in manganites. Our results reveals that while lattice strain from the substrate has an effect in influencing the optical properties of the LSMO thin-films, the coexistence of strong electron-electron (e-e) and electron-hole (e-h) interactions which bring about resonant excitonic effects from the substrate play a more significant role. This onset of anomalous excitonic dynamics in manganite oxides could potentially generate new approaches and interests in manipulating exciton-based optoelectronic applications.

Sample

High-quality LSMO thin-films grown by Pulsed-Laser Deposition (PLD) on [001]-$SrTiO_3$ (STO), [110] $DyScO_3$ (DSO), [001] $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$ (LSAT)

Fully-strained & **coherent** between all films & substrates

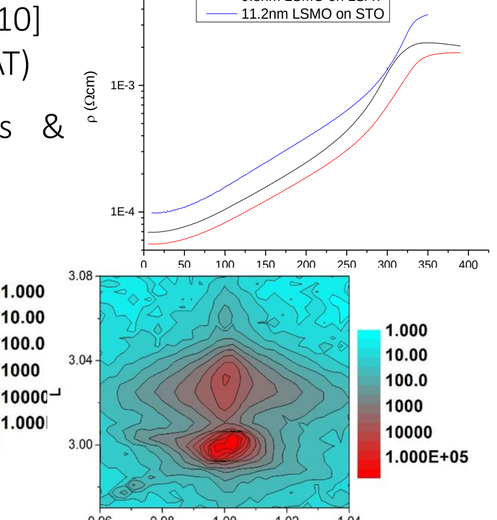




Sample	Strain (%)
(Thin) 11.2nm LSMO/STO	0.6 (tensile)
(Thick) 87.2nm LSMO/STO	0.6 (tensile)
12.6nm LSMO/DSO	1.9 (tensile)
9.8nm LSMO/LSAT	-0.3 (compressive)

Spectroscopic Ellipsometry

0.4 – 5.6 eV spectral range
3 K – 420 K (Janis Cryostat)
Measures ellipsometric parameters Ψ and Δ (magnitude ratio & phase difference between p- and s- polarized reflected light) at $70°$ incident angle
Dielectric function $\varepsilon(\omega)$, optical conductivity $\sigma(\omega)$, data extracted from Ψ and Δ using air/LSMO/Substrate multilayer model



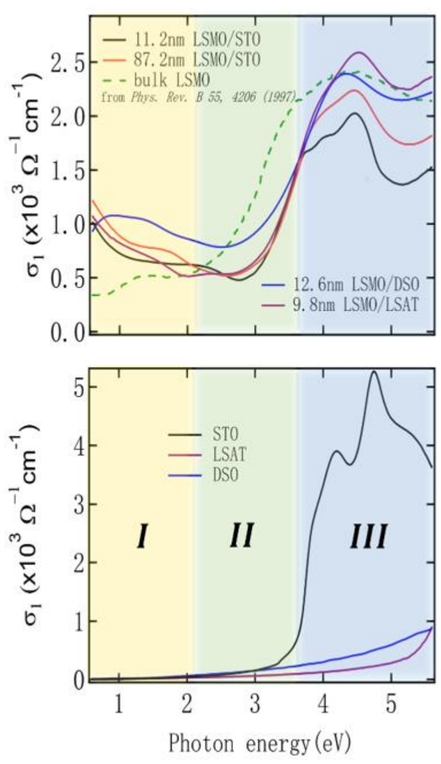
Results

Region I (<2.1eV)

- Spectra-weight (SW) mainly due to Jahn-Teller & charge-transfer excitations
- Distinct increase in SW from bulk LSMO to LSMO thin-films on various substrates
- Corresponds to respective increase in interfacial strain caused by different substrates
- Jahn-Teller & Charge-Transfer effects amplified with increasing strain.

Region III (>3.7eV)

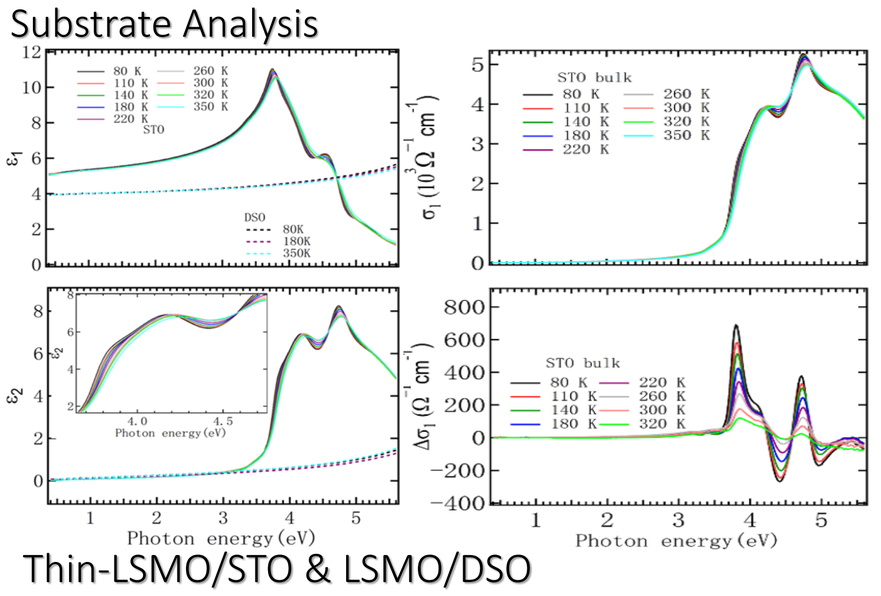
- Optical conductivity, σ_1
- Thin-LSMO/STO < LSMO/LSAT & LSMO/DSO
- Thick-LSMO/STO slightly higher
- *f*-sum Rule: steep drop in Region III due to SW-shift to significantly higher energy region
- An evidence of significance changes in the electronic-correlations of LSMO films on STO substrate



Strain not main contributing factor for optical spectra in Region III

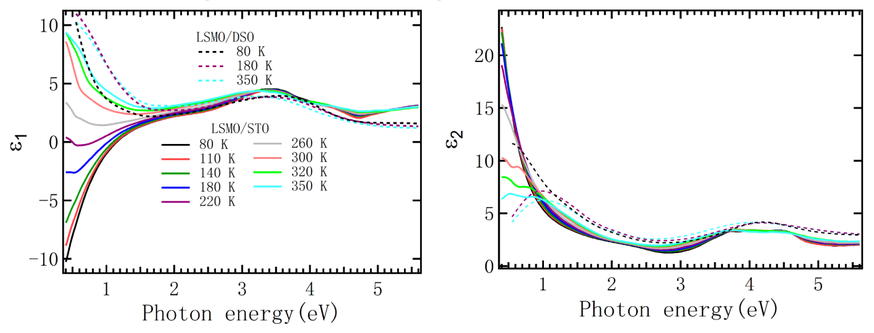
Temperature-Dependent Results

Substrate Analysis



DSO: Near-constant behaviour
STO: Increasing excitonic effects as temperature decreases

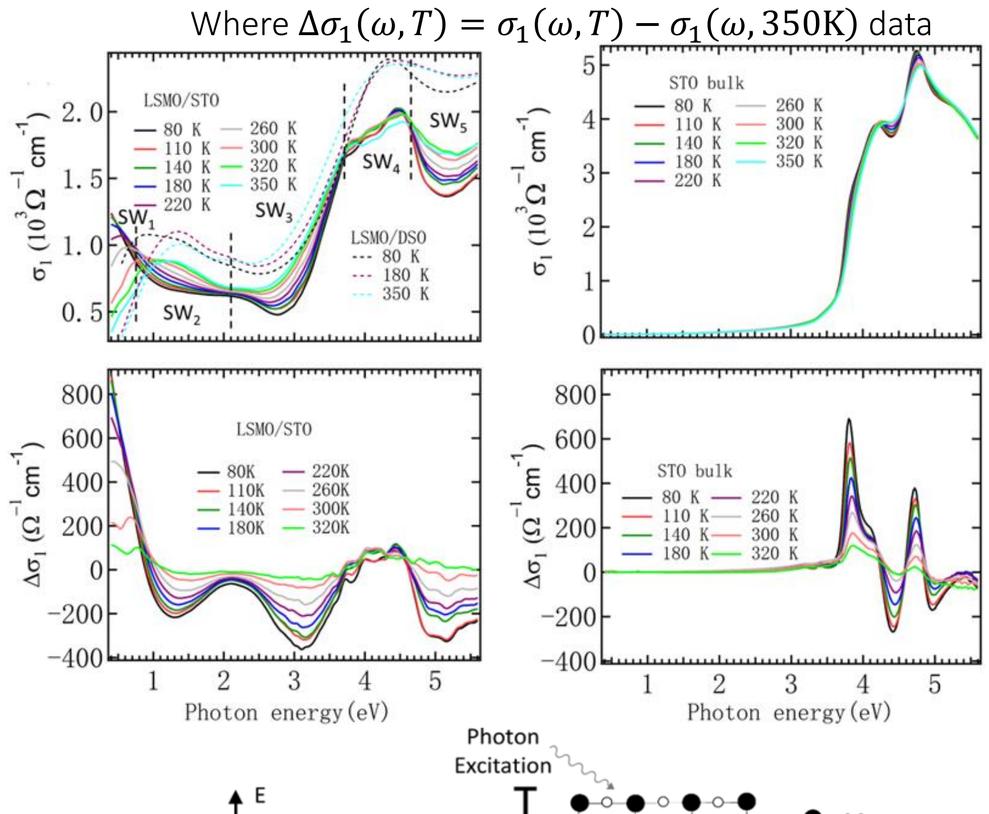
Thin-LSMO/STO & LSMO/DSO

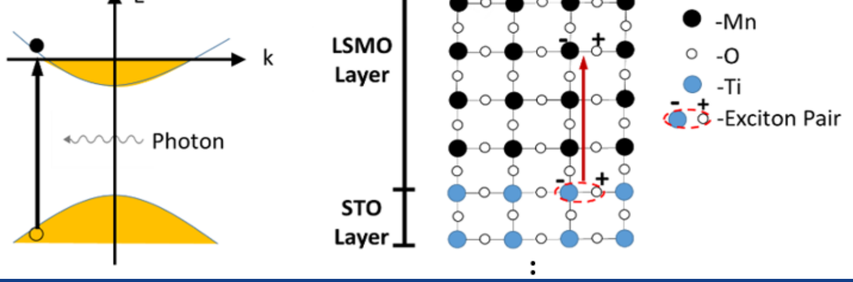


ε_2: LSMO/STO transforms into metallic state (growing Drude response with decreasing temperature)

LSMO/DSO does not

Where $\Delta\sigma_1(\omega,T) = \sigma_1(\omega,T) - \sigma_1(\omega,350\text{K})$ data





Conclusion

we discover the propagation of resonant excitons from STO substrate into LSMO films. Surprisingly, the resonant excitonic effects can propagate deep into the LSMO films (even for films as thick as 87.2 nm) and are able to change LSMO optical properties dramatically and strengthen its electronic correlation. These anomalous excitonic properties have separate effects as compared to that of interfacial strain on the optical conductivity and electronic correlations of the thin film.

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