Structural phase diagram and optical properties of superconducting bilayer nickelates under high pressure

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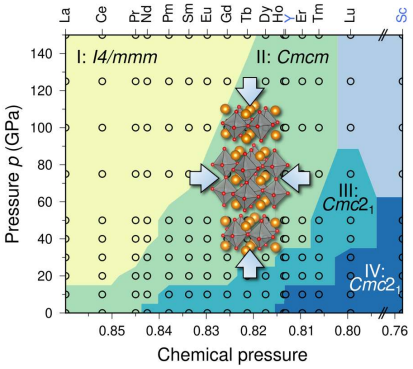
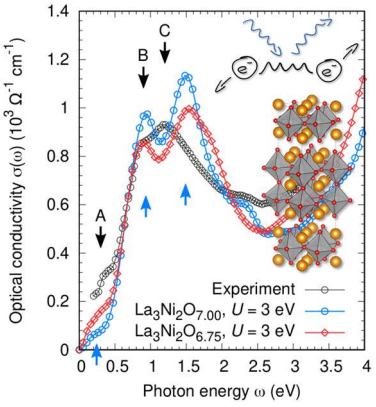


Motivated by the recent observation of superconductivity with T_C~80 K in pressurized $La_3Ni_2O_7$ [1], we explore the structural and electronic properties in $A_3Ni_2O_7$ bilayer nickelates (A=La-Lu, Y, Sc) as a function of hydrostatic pressure (0-150 GPa) from first principles including a Coulomb repulsion term [2]. At ~20 GPa, we observe an orthorhombic-to-tetragonal transition in $La_3Ni_2O_7$ at variance with recent x-ray diffraction data, which points to so-far unresolved complexities at the onset of superconductivity, e.g., charge doping by variations in the oxygen stoichiometry. We compile a structural phase diagram that establishes chemical and external pressure as two distinct and counteracting control parameters. Unexpected correlations between T_C and the in-plane Ni-O-Ni bond angles are found for $La_3Ni_2O_7$. Moreover, two novel structural phases with significant c^+ octahedral rotations and in-plane bond disproportionations are uncovered for A=Nd-Lu, Y, Sc that exhibit a surprising pressure-driven electronic reconstruction in the Ni e_g manifold. We identify $Tb_3Ni_2O_7$ as an interesting candidate for superconductivity at ambient pressure. Finally, we assess the role of electronic correlations and explicit oxygen vacancies in $La_3Ni_2O_7$ by comparing experimental and simulated optical spectra, which corroborates the proposed $s^{+/-}$ superconductivity mechanism.



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[1] H. Sun *et al.*, Nature **621**, 493 (2023)
[2] B. Geisler, J. J. Hamlin, G. R. Stewart, R. G. Hennig, P. J. Hirschfeld, arXiv:2309.15078 [cond-mat.supr-con]