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Quantum Materials and Chiral Phenomena: Fundamental Science and Applications

E-book of Abstracts

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Optical signatures of spin symmetries in unconventional magnets

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Spin symmetries have been an important ingredient for the recent classification of unconventional magnets [1,2]. In this talk, I will focus on the relation between spin symmetries and the photoresponse of unconventional magnets. Firstly, I will use a 2D variant of the Lieb lattice [3] to study the impact of spin-orbit coupling on spin symmetry predictions of the standard linear absorption of altermagnets [4]. Then, I will focus on nonlinear responses such as the shift current [5], given the close relationship of nonlinear effects to the symmetries of a material. As a concrete example, I will primarily focus on the low temperature phase of Mn_5Si_3, a material that features the two possible classes of unconventional p-wave magnetism in the form of two competing spin structures [6,7]. I will show that the dominant spin and charge photoresponse of the two structures is determined by spin symmetries rather than the conventional magnetic symmetries [8]. I will point out the main implications of this effect and propose an experimental protocol to identify the spin configuration of this promising material in photogalvanic or transport measurements.

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[7] N. Biniskos, F. J. dos Santos, K. Schmalzl, S. Raymond, M. dos Santos Dias, J. Persson, N. Marzari, S. Blügel, S. Lounis, and T. Brückel. Phys. Rev. B 105, 104404 (2022).

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Relativistic Spin-momentum locking in altermagnets

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Spin-momentum locking has been demonstrated to exist in altermagnets in the non-relativistic limit [1]. When spin-orbit coupling is taken into account, all altermagnets display antisymmetric exchange interactions. These interactions lead to spin canting, although the canting may vanish for certain orientations of the Néel vector. We demonstrate that when spin-canting occurs, the spin-momentum locking can evolve and change significantly. Focusing on the centrosymmetric altermagnets, we will show that the spin-momentum locking can be present and differs for all three components of the spin S_x, S_y and S_z. The combination of the three spin-momentum lockings is named relativistic spin-momentum locking. To discuss this effect, we consider two prototypical centrosymmetric altermagnets, namely the orthorhombic YVO_3 and the hexagonal MnTe, which have bulk d-wave and bulk g-wave spin-momentum locking in the non-relativistic limit. For the G-type magnetic ordering of YVO_3 and Néel vector along the z-axis, the relativistic spin-momentum locking is composed of s-wave, d_{xy}-wave and d_{xz}-wave for the S_x, S_y and S_z components, respectively. As in the non-relativistic case, the relativistic spin-momentum locking is protected by rotational symmetries. In MnTe, the main component Sy of MnTe inherits the polarized charge distribution and the non-relativistic spin-momentum locking bulk g-wave, but the breaking of the C_{6z} rotational symmetry by the Néel vector lowers the symmetry from g-wave to d-wave. The relativistic spin-momentum locking for MnTe is composed of d_{xz}-wave, d_{yz}-wave and s-wave for the S_x, S_y and S_z components, respectively [2,3]. There are several orders of magnitude of difference between the size of the main spin component and the components raised by the canting. Despite this, the spectral weight of the canted components on the spin-resolved band structure is significant. Indeed, the spectral weight of the canted components is smaller but of the same order of magnitude as that of the main spin component. Finally, using Ca_2RuO_4 as a testbed, we address the challenges arising in altermagnets under an electric field that breaks inversion symmetry[4].

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[4] A. Fakhredine, **C. Autieri** *et al.* Manuscript in preparation.

A-approach in ARPES and chiral topological superconductivity in $PtBi_2$

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Topological superconductivity is a key ingredient for realizing Majorana fermions, yet intrinsic material platforms remain scarce. Using angle-resolved photoemission spectroscopy (ARPES), we have recently demonstrated that the Weyl semimetal t-$PtBi_2$ hosts superconductivity confined to its topological surface states, the Fermi arcs [1]. This superconductivity is unconventional, with nodal i-wave symmetry and signatures of emergent Majorana modes [2], and can be robustly detected under both laser- and synchrotron-based ARPES conditions [3]. Moreover, t-$PtBi_2$ fulfills the canonical criteria favorable for high-T_c superconductivity, opening the door to tuning and optimizing this exotic state [4]. These findings establish t-$PtBi_2$ as a rare platform where ARPES directly reveals superconductivity of topological surface states, paving the way toward intrinsic realizations of Majorana physics. I will also briefly review a newly developed A-approach in ARPES technique [5].

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Imaging Twisted Electrons by Angle-Resolved Photoemission Spectroscopy

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Owing to their wave nature, electrons in crystalline solids are usually described in momentum space, in which each point represents an electron wave that is (in real space) spread over the entire crystal. The momentum dependence of the quantum degrees of freedom associated with these wave functions leads to a variety of properties, such as spin-momentum locking in topological surface states or chiral quasiparticles in topological semimetals. In my talk, I will shed light on the k-dependent textures, in particular of spin and orbital angular momentum, in the band structure of quantum materials. Using angle-resolved photoemission spectroscopy in combination with dichroism and spin resolution, we are able to directly image these textures. On this basis, I will show recent results in which we have observed exotic topological objects, such as chiral topological charges or momentum-space vortex lines, i.e. electrons with a 'twist'.

Electronic structure and charge-density wave modulation in monolayer $TiSe_2$

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When two periodic patterns are superimposed, interference can produce new patterns with extended periodicities termed as a moiré. In two-dimensional heterostructures, Moiré superlattices can be constructed by introducing small twist angles between homobilayers or simply by stacking heterobilayers with different lattice parameters. The spatial modulation of the Moiré lattice introduces long wavelength potentials with scales far larger than typical crystal lattices, inducing the formation of band replicas associated with the periodicity of the Moiré lattice and leading to band hybridisation with flattening of the electronic states [1-2]. Under intense band flattening regime, these localized electronic states underpin the emergence of a host of strongly correlated phenomena [3-4]. In this work, we examine the interplay between charge-density waves (CDW) and the Moiré potential arising from a lattice mismatch between 2D $TiSe_2$ and graphite in a 2D heterostructure. Using a combination of high-resolution angle resolved photoemission spectroscopy measurements and quasi-particle interference patterns obtained from scanning tunnelling microscopy and spectroscopy, I will demonstrate that this leads to a strong modulation in the electronic, both in the occupied and unoccupied states. I will show how this can be further tuned by controlling the ground state of the $TiSe_2$ layer, offering new insights into the connection between Moiré and the rich many-body phases of $TiSe_2$.

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Tunable room-temperature nonlinear Hall effect from surfaces of elementary bismuth thin films

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In the past years, there is an active research of materials displaying the nonlinear Hall effect with time-reversal symmetry [1-5]. From a fundamental point of view, this quantum transport effect provides a direct way to detect in nonmagnetic materials the Berry curvature – a quantity in which the geometry of the electronic wavefunctions is encoded. The nonlinear Hall effect is also at the basis of terahertz optoelectronic applications of interest for instance for sixth generation (6G) communication networks.

An appropriate material platform for such applications should satisfy a number of criteria: i) the nonlinear Hall effect should survive up to room temperature; ii) the effect should be tunable; iii) the material fabrication should be technologically relevant (simple chemical composition of the material and low-cost microstructure); iv) ideally the material should not contain toxic heavy rare-earth elements. So far, candidate materials address only partially these requirements.

Here, we discover the first material addressing all the requirements at the same time: polycrystalline bismuth thin films [6]. We demonstrate that in this elemental green (semi)metal, the room-temperature nonlinear Hall effect is generated by surface states that are characterized by a Berry curvature triple: a quantity governing a skew scattering effect that generates non-linear transverse currents. Furthermore, we also show that the strength of nonlinear Hall effect can be controlled on demand using an extrinsic classical shape effect: the geometric nonlinear Hall effect. We demonstrate this by fabricating arc-shaped bismuth Hall bars. This endows the nonlinear Hall effect of Bismuth with the tunability encountered only in low-dimensional materials at low temperatures.

To show the potential of polycrystalline Bi thin films for optoelectronic applications in the terahertz (THz) spectral domain, we have performed high harmonic generation experiments. Polycrystalline Bi thin films reveal a high efficiency of THz third-harmonic generation (THG) that reaches levels >1% at room temperature. Moreover, our material possesses a non saturating trend of the efficiency of the THz THG. This enables the use of Bi thin films for high- and wide- THz bandwidth electronics which works at high peak power and long pulses.

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Hallmarks of Spin Textures for High-Harmonic Generation in two-dimensional materials

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Spin-orbit coupling and quantum geometry are fundamental aspects in modern condensed matter physics, with their primary manifestations in momentum space being spin textures and Berry curvature when inversion symmetry is broken. In this talk, motivated by recent technological advances in laser and attosecond science, I will discuss the interplay of spin textures and Berry curvature with high-harmonic generation in two dimensional non-centrosymmetric materials, as we investigated in a recent paper [1], with an emphasis on even-order harmonics. Our analysis reveals that the emergence of finite even-order harmonics in such systems necessarily requires a broken twofold rotational symmetry in the spin texture, as well as a nontrivial Berry curvature in systems with time reversal invariance. This symmetry breaking can arise across various degrees of freedom and impact both spin textures and optical responses via spin-orbit interactions. These findings underscore the potential of high-harmonic generation as a powerful tool for exploring electronic phases with broken rotational symmetry, as well as the associated phase transitions in two-dimensional materials, and provide novel perspectives for designing symmetry-dependent nonlinear optical phenomena.

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Three-Dimensional Polar Topology in Ferroelectric Nanoparticles

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In the early 2000s, it was anticipated that low-dimensional systems would exhibit unique polar structures like vortices or skyrmions, depending on boundary conditions. Some variations of these structures have been seen in experiments with epitaxially strained thin film models. However, fully understanding and categorizing these structures in ferroelectric nanostructures has been challenging due to the need for atomic-scale, three-dimensional polarization mapping. We present here the discovery of such structures in $BaTiO_3$ nanoparticles using atomic electron tomography. Our findings reveal distinct topological patterns, including size dependent transitions from single to multiple vortices, aligning with theoretical expectations. This discovery, independent of strain, expands research possibilities and offers potential for contact-free switchable toroidal moments in practical applications [1].

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Low-density superconductivity in the presence of strong spin-orbit coupling

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In conventional superconductors, Cooper pairs are formed in singlet states. In some materials, spin-orbit coupling (SOC) on the scale of the Fermi energy introduces entanglement between orbital and spin degrees of freedom, changing this paradigm and leading to novel effects. I will argue that SOC is essential to explain superconductivity at anomalously low densities in incipient ferroelectrics, like strontium titanate and potassium tantalate.

Filtering Spin and Orbital Moment in Achiral Systems

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The control of spin and orbital angular momentum in nonmagnetic materials is commonly achieved by breaking inversion symmetry, enabling charge-to-spin conversion and spin-selective electron transport in chiral systems. In this work, we show that orbital moment filtering can also be realized in centrosymmetric systems, where electron states can be selectively manipulated to favor the transmission of electrons with a specific orientation of their orbital moment. We demonstrate that this effect arises from orbital couplings that simultaneously break mirror and rotational symmetries, and we identify the corresponding symmetry requirements for efficient orbital filtering. Furthermore, we show that atomic spin– orbit coupling within the centrosymmetric medium leads to the joint filtering of spin and orbital moments. These results allow one to identify optimal regimes for achieving highly efficient and simultaneous spin and orbital moment filtering even across achiral media.

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Dirac-like fermions anomalous magneto-transport in a spin-polarized oxide two-dimensional electron system

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In two-dimensional electron systems (2DESs), the breaking of the inversion, time-reversal and bulk crystal-field symmetries intertwines with the spin-orbit coupling (SOC), giving rise to exotic quantum phenomena. By engineering a spin-polarized oxide 2DES with Rashba-like SOC and hexagonal band warping, here we present the first report of an anomalous quantum correction to the magnetoconductance, originating from Dirac-like fermions experiencing competing weak anti-localization and weak localization back-scattering (Fig. 1(a) and ref. [1]). This phenomenology closely resembles that of gapped topological insulators [2]. The results were obtained on the 2DES formed at the epitaxially grown interfaces between (111) $LaAlO_3$, $EuTiO_3$, and $SrTiO_3$ single crystal, characterized by a trigonal crystal field splitting and ferromagnetism induced by Eu and Ti ions magnetic ordering [3]. Notably, the anomalous magnetoconductance disappears at the magnetic critical temperature [4], showing a direct link with the ferromagnetic order. The data are explained theoretically in a single band scenario as the combined effects of the Rashba-SOC, of the band-warping induced by the 2DES trigonal symmetry, and of the magnetic gap opening at spin-orbit induced Dirac-like point, giving rise to a non-trivial Berry phase (Fig. 1(b, c)). These findings open perspectives for the engineering of novel spin-polarized functional 2DES holding promises in spin-orbitronics and topological electronics.

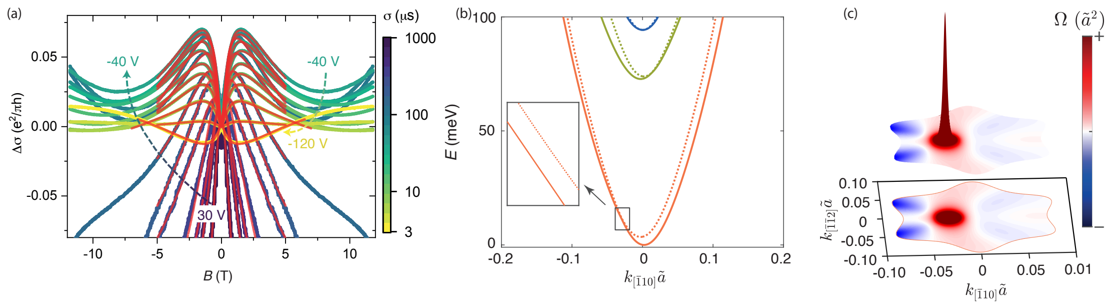

Figure 1: (a) Gate dependence of the anomalous magnetoconductance. (b) Electronic band structure in presence of in-plane magnetization. (c) The corresponding non-trivial Berry curvature with a hot-spot at the avoided crossing point.

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Skyrmions: from hadrons to magnetic textures

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Skyrmions were proposed in 1961 by Tony Skyrme as a mathematical description of hadrons. From 2010, observation of a simple O(3) version of the skyrmion started to get reported in a variety of magnetic materials, which sparked keen interests in its structure, dynamics, and potential device applications. I devote a part of my talk describing the history of the theory and experimental progress in magnetic skyrmions over the past 15 years. I also present recent attempts to extend existing theories of magnetic skyrmions, which are technically classified as CP(1) skyrmions, to CP(N) skyrmions with arbitrary N.

Spintronics and Correlation Effects in Two-Dimensional Magnetic Materials: From $\mathbf{Fe_nGeTe_2}$ to p-Wave Magnets

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The rapid discovery of two-dimensional (2D) magnetic materials has unveiled a wealth of complex magnetic orders and exciting opportunities for spintronics. In this talk, I will briefly introduce several such materials [1], focusing in particular on the Fe_nGeTe_2 (n = 3, 4, 5) family of metallic ferromagnets and on NiI_2, a recently reported p-wave magnet [2]. Using density functional theory (DFT) combined with the non-equilibrium Green's function (NEGF) formalism, I will discuss their transport properties in tunnel magnetic junction setups. I will first demonstrate that FenGeTe2 exhibits nearly half-metallic conductance along the out-of-plane direction, thus leading to a large tunnel magnetoresistance (TMR) [3,4]. Then, I will show how the concept of TMR can be extended from conventional ferromagnets to p-wave magnets. Finally, I will address the role of correlation effects in transport by introducing a framework that combines DFT, Dynamical Mean-Field Theory (DMFT), and NEGF. Within this framework, we show that devices based on Fe_nGeTe_2 under finite bias can enter a novel "hot-electron correlated" regime driven by electron scattering processes [5].

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Magnetic and Chiral skyrmion materials

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Recently, there has been considerable interest in the physics of magnetic skyrmions due to their huge potential for use in spintronic devices, such as in racetrack memories and logic devices. Magnetic skyrmions are topological magnetic spin structures originally identified in the B20 class of materials. More recently, skyrmions have been found and investigated in other non-centrosymmetric classes of materials and in centrosymmetric intermetallics. To make headway in experiments to understand the basic physics of these skyrmion materials, high quality single crystals are essential. This has motivated us to embark upon a study of several classes of skyrmion materials and to explore a wide composition range of each of the family of compounds. The materials investigated range from centrosymmetric intermetallics such as Gd_2PdSi_3, $GdRu_2Si_2$, magnetic layered van der Waals materials such as Fe_3GeTe_2, to a large family of intercalated transition metal dichalcogenides (TMDCs) and other frustrated and topological magnets.

In this talk, I will present an overview of the materials characteristics of several of the above materials including the challenges in the synthesis of these materials using a variety of techniques at Warwick. Investigations of the effects of substitution and the resulting structural order/disorder as well as magnetic frustration on the existence of the skyrmion phase in these crystals sheds light on the origin and the tuning of the skyrmion lattices. The study of their important structure–property correlations is vital to the understanding of these materials for possible future device applications.

Controlling the functionality of quantum materials by light

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The properties of complex quantum materials (QM), such as transition metal oxides, arise from the interplay of electrons, phonons, and magnons, making them highly sensitive to external parameters like pressure, doping, fields, and temperature. This susceptibility makes QM ideal for experiments where tailored electromagnetic fields can be used to induce novel properties on ultrafast timescales [1].

I will present our efforts to manipulate material properties through light, both in free space and optical cavities. After reviewing our work on cuprates, which demonstrates the feasibility of light driven phase control [2–4], I will introduce new spectroscopic methods that merge quantum optics with time-domain techniques to probe fluctuations in non equilibrium phases [5–11].

Building on our recent demonstration that a metal–insulator transition in 1T-TaS_2 can be controlled by resonant cavity coupling [12, 13], I will outline future directions aimed at controlling by cavity electrodynamics metal-insulator transitions in Calcium Rutanate and explore new light–matter coupling regimes to bypass thermodynamic limits and dynamically sustain quantum coherence in high temperature superconductors.

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The Dual Role of Killer Jahn–Teller Modes and Screw Symmetry in Ferromagnetic Two-Dimensional Hybrid Perovskites

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Chiral two-dimensional hybrid perovskites offer compact platforms where lattice handedness, light and spin intersect. We report the enantiomeric layered perovskite (R/S-3-fluoropyrrolidinium)2$CuBr_4$, in which chirality from cyclic organic cations imprints a handed distortion onto the Cu–Br framework. From an achiral tetragonal parent with undistorted $CuBr_6$, a cooperative Jahn–Teller–mode condensation lowers the symmetry to orthorhombic, enantiomorphic phases by splitting the high-symmetry octahedron into two orthogonal square units. We identify killer modes: selecting one Jahn–Teller eigen-direction "kills" (collapses) one of the two interpenetrating $CuBr_4$ squares while stabilizing the orthogonal one as a distorted, oriented square pyramid—thereby effectively transferring chirality into the framework. Choosing the perpendicular JT mode kills the other square and produces the opposite handedness, mapping directly onto the R/S domains. The chirality is quantified by a pseudo-scalar local order parameter for each $CuBr_4$ unit, which is positive for R, negative for S, and vanishes in the high-symmetry limit. In parallel, we reveal killer symmetry operations: the chiral, non-polar space group $P2_12_12_1$ arranges permanent molecular dipoles into two screw-related, non-collinear antiferroelectric sublattices, enforcing exact macroscopic cancellation while locking local dipoles to the inorganic handedness. This hybrid-improper pathway–non-chiral lattice modes coupled with JT distortion–removes all improper symmetries without producing net polarization, yielding efficient chirality transfer. The chiral phase displays strong natural circular dichroism (anisotropy $|g|$ up to $3.4{\cdot}10^{-33}$), a chirality-dependent MCD band near 330 nm, and in-plane ferromagnetism ($T_c \approx 6$ K), establishing a single-material platform where the dual role of JT-modes and screw symmetry operations co-engineer lattice handedness, dipole textures and spin order.

Towards the twisting of nickelate membranes

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Stacking and twisting of released freestanding membranes has brought to light a promising path- way towards the design of materials with novel properties. Following the discovery of supercon- ductivity in twisted graphene [1] and, more recently, in twisted transition metal dichalcogenides WSe_2 [2], the twisting approach is currently being implemented also for exfoliated membranes of transition metal oxides [3]. In particular, the appearing of a Moiré pattern, usually associated to two superposed honeycomb lattices, has been clearly demonstrated for ferroelectric $BaTiO_3$ twisted membranes [4]. Such Moiré patterns, accompanied by coherent chemical bonding at precise coincidence-site lattices, have the potential to reshape the electronic landscape at the twisted interface between two transition metal oxide layers, leading to the emergence of unexpected collective states. From the growth point of view, releasing freestanding oxide membranes poses a dual challenge: the functional oxide must first be grown on a suitable sacrificial-layer without compromising its intrinsic properties, while keeping the good structural match over the entire oxide stack. Here, I present our results on the use of epitaxial lift-off techniques to fabricate freestanding layers of $LaNiO_3$ thin films and related twisting. By using a pulsed laser deposition (PLD) technique assisted by high-energy reflection electron diffraction (RHEED), we have epitaxially grown $LaNiO_3$-based heterostructures on a sacrificial layer of water-soluble $(Ca,Sr)_3Al_2O_6$ onto $SrTiO_3$ substrates. A technique that is largely used in literature [5]. Structural analyses conducted via X-ray diffraction confirmed the good crystalline quality of the grown heterostructures as well as of the released membranes. Transport measurements performed by evaporating metallic top electrodes directly onto the membrane, showed similar temperature dependencies of the seed films, i.e. a metallic behavior with resistivity values within the μOhm cm^{-1} range, and more importantly, with no upturn at low temperatures. This largely demonstrate the high quality of the transferred membranes mainly associate to the good growth of the oxide stack and the optimal transfer processes. Finally, I will demonstrate the impact of twist angle on the transport properties, and report about preliminary topotactic reduction experiments underwent to stabilize freestanding infinite-layer $LaNiO_2$ membranes.

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