Electronic and magnetic excitations in La3Ni2O7

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[Ques] The key question of this study is to unveil the microscopic origin of the low-energy electronic structure, magnetic excitations, and ordered states in the bilayer nickelate $La_3Ni_2O_7$ at ambient pressure

[关键问题]：在常压下，揭示双层镍酸盐 $La_3Ni_2O_7$的低能电子结构、磁激发及其有序态的微观起源

[Sum] This study, using X-ray absorption spectroscopy and resonant inelastic X-ray scattering, reveals that $La_3Ni_2O_7$ at ambient pressure is a system with a small charge-transfer energy and strong electronic correlations. Its low-energy physics is dominated by the strong hybridization of the Ni $3d_{x^2-y^2}$ Ni 3d_z ² orbitals with the O 2p orbitals, and features ligand hole states analogous to those in cuprate superconductors. Most crucially, the research directly uncovers, for the first time, an interlayer antiferromagnetic superexchange coupling (J_z) that is much stronger than the intralayer interactions. This coupling originates from the unique molecular bonding between the two Ni 3d_z ²orbitals via the apical oxygen, and leads to the formation of a quasi-two-dimensional **spin density wave (SDW) order below 150 K.

[亮点 1] 文章通过 RIXS 测量磁子色散，并拟合海森堡模型，定量地揭示出层间反铁磁超交换作用（J_2）比面内相互作用（J_1, J_2）强一个数量级

[亮点 2] 研究成功地在常压母体中观测到源于强层间耦合的准二维自旋密度波（SDW）有序，其波矢为(0.25, 0.25)。

[亮点 3] 通过 O K-edge XAS 中观测到的显著前峰，明确指出 $La_3Ni_2O_7$中存在与铜基超导体“张-莱斯单态”相似的电子基态，即具有小电荷转移能和大量的氧配体空穴

[思考 1] Ni $3d_{x^2-y^2}$和 Ni 3d_z 轨道共同主导低能物理,在具体的超导配对中分别起什么作用？

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[思考 2] $La_3Ni_2O_7$存在类似“张-莱斯单态”的配体空穴态。这种态在双层镍酸盐中的具体形式是什么？它与铜基超导体中有何区别？

[拓展阅读 1] 张-莱斯单态是一个由中心 Cu^{2+}的 $3d_{x^2-y^2}$轨道和其周围四个 O^{2-}的 2p 轨道通过强杂化共同形成的、局域的自旋单态。它描述的是，那个额外的空穴并非定域在某个单一的原子上，而是以一种量子纠缠的方式，均匀地分布在四个氧原子上，并且与中心铜离子的自旋锁定在一起，形成一个总自旋为零（单态）的复合对象。在张-莱斯单态图像下，复杂的多轨道 Cu-O 模型可以简化为一个单带 Hubbard 模型或 t-J 模型，其中张-莱斯单态本身就扮演着该单带中的“空穴”角色。这个巡游的、无自旋的准粒子，被认为是实现高温超导配对的关键载体。

[拓展阅读 2] RIXS（共振非弹性 X 射线散射）是一种强大的实验技术，通过入射 X 射线在材料中引起核心能级电子的共振激发，再测量散射出的 X 射线能量和动量的变化，从而获得材料内部的能量损失信息。这种能量损失对应材料中的局域电子激发，如 dd 激发（3d 轨道电子之间的跃迁）、磁激发（自旋波或磁序激发）以及声子激发（晶格振动），使 RIXS 能够直接探测材料的电子结构、磁性以及电子-晶格相互作用等微观物理过程。

[拓展阅读 3]强色散激发指的是一种激发态（如声子、磁激发、电子激发等），其能量随动量明显变化，也就是说激发的色散关系 E(k)有很大的斜率。在晶体或固体材料中，如果某种激发的能量在布里渊区不同动量点上变化很大，它就被称为强色散。换句话说，强色散意味着该激发可以在晶体中高速传播，具有明显的动量依赖性，而非局域化或几乎动量独立的“平带”型激发。

[Introduction]

The striking discovery of high-temperature superconductivity (HTSC) at 80 K in a bilayer nickelate $La_3Ni_2O_7$ under a pressure of about 14 GPa ignited a new wave of studying HTSC in nickelates. Unlike cuprate superconductors with a Cu^{2+} $3d^9$ electron configuration, $La_3Ni_2O_7$ hosts Ni ions with mixed 2^+ (3d8) and 3^+ (3d7) valences with unpaired electrons in both $3dx^2$-y^2 and $3dz^2$ orbitals from a Ni-O bilayer structure. In particular, the molecular bonding between the two inter-layer Ni 3dz2 orbitals through the apical O pz orbital, together with Ni $3dx^2$-y^2 orbitals, are proposed by theory as a critical ingredient for the low-energy electronic structure of $La_3Ni_2O_7$. The orbital character governing the electronic

properties of the unconventional superconductors is essential for understanding the underlying pairing mechanism. In cuprates, the small charge-transfer energy and strong hybridisation between Cu $3dx^2$-$y^2$and O 2p orbitals lead to the formation of the strongly correlated Zhang-Rice singlet band, which serves as the foundation for describing the electronic properties including the superconducting pairing interaction with $3dx^2$-y^2 symmetry. On the other hand, the iron-based superconductors feature relatively weaker correlation and multiple 3d bands near the Fermi surface. The orbital-dependent correlation and the strong anisotropy in the electronic hopping result in a distinct s pairing symmetry. At first sight, $La_3Ni_2O_7$ appears to be a sibling of iron-based superconductors owing to the multi-orbital nature and the bad metallicity in the undoped parental phase. However, perovskite nickelates are also known to exhibit strong electronic correlation and small chargetransfer energy, resembling cuprates. Theories to date vary in their opinions on which orbitals are most relevant for the electronic properties, especially the superconductivity, in $La_3Ni_2O_7$. The antiferromagnetic (AFM) superexchange interaction is accepted as another important ingredient of unconventional superconductors. Upon the doping of charge carriers, the long-range AFM ordered parental phase evolves into one with short-range AFM spin fluctuations, which may mediate the superconducting pairing. In a sizable part of the phase diagram, the interplay among spin, charge, and lattice degrees of freedom often leads to exotic ordering phases such as the periodic density modulation of charge or spin. In cuprates and iron-based superconductors, charge (CDW) and spin density waves (SDW) intertwine with superconducting phase which is regarded as being closely relevant to HTSC. The bilayer structure and the multi-orbital nature of $La_3Ni_2O_7$ have profound impact on its magnetism as well, which plays a pivotal role in theories on this novel superconductor. Some theory suggest the importance of the interlayer antiferromagnetic coupling Jz between dz^2 orbitals; some others advocate that the strong interlayer coupling would cause the bilayer splitting of band structure, while in-plane magnetic exchange interactions play a dominant role in superconductivity. In the asgrown $La_3Ni_2O_7$ crystal at ambient pressure, resistivity measurements found a kink-like transition at around 153 K implying a possible CDW or SDW state. NMR studies found CDW order possibly mixed with SDW order in polycrystalline $La_3Ni_2O_7$, and most recently SDW order was revealed in single crystal $La_3Ni_2O_7$. In addition, μSR experiments suggested that a static long-range magnetic order emerges in polycrystalline $La_3Ni_2O_7$ ~ 150 K. Despite the proposals of potential density waves, NMR and μSR experiments reported that the

magnetic moment per Ni site is ~ 0.08 μB and 0.3–0.7 μB, respectively. Given the currently limited knowledge on the essential electronic and magnetic properties, such as the charge-transfer energy and the magnetic exchange interactions, experimental verification is indispensable. In this work, we employ X-ray absorption spectroscopy (XAS) and resonant inelastic X-ray scattering (RIXS) at both Ni L3-edge and O K-edge of $La_3Ni_2O_7$ single crystal at ambient pressure. These spectroscopic and scattering techniques are sensitive to low-energy electronic and magnetic structures together with elementary excitations, and thus are ideally suited for tackling the core issues in $La_3Ni_2O_7$.

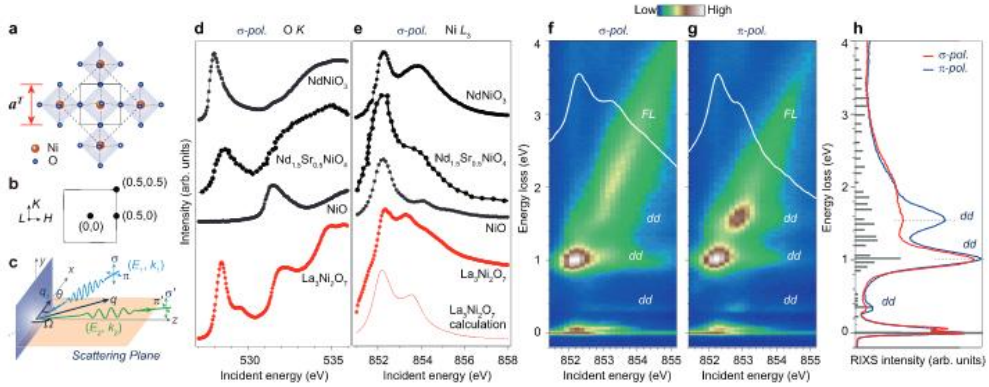

Fig. 1 | XAS spectra and the incident energy-dependent RIXS maps in $La_3Ni_2O_7$.

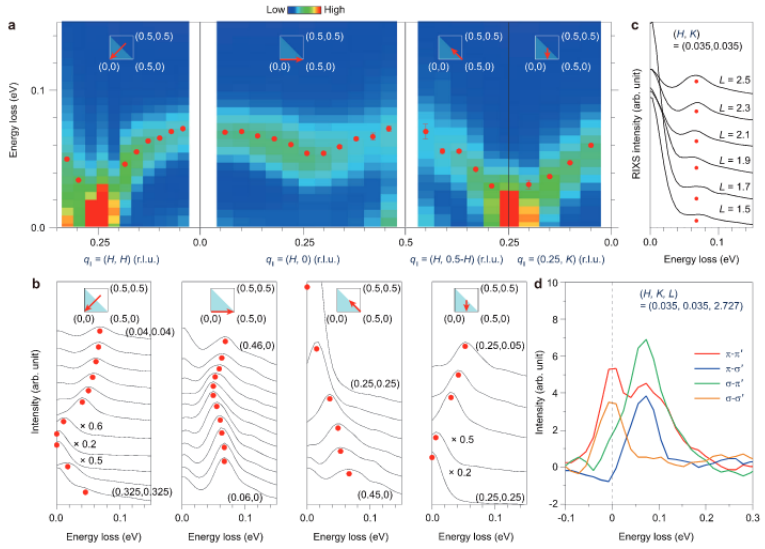

Fig. 2 | Energy-momentum dependent magnon in $La_3Ni_2O_7$

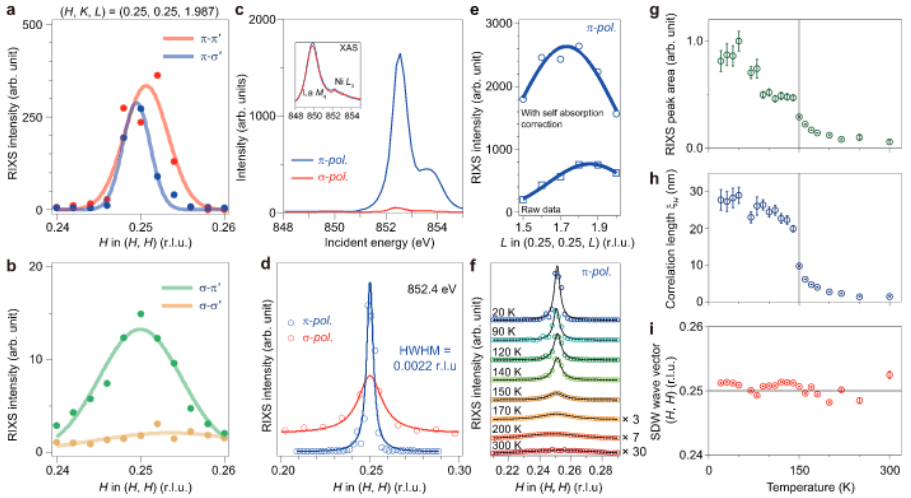

Fig. 3 |SDW order at (0.25, 0.25) of $La_3Ni_2O_7$

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