Spin density wave rather than tetragonal structure is prerequisite for superconductivity in La3Ni2O7-δ

Mengzhu shi(University of Science and Technology of China)

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[Ques] Is the tetragonal structure a necessary condition for realizing superconductivity in $La_3Ni_2O_{7-\delta}$?

[关键问题] 四方结构是否是 $La_3Ni_2O_{7-\delta}$ 中实现超导的必要条件？

[Sum] This study demonstrates that the tetragonal structure is not a prerequisite for superconductivity in $La_3Ni_2O_{7-\delta}$. Tetragonal $La_3Ni_2O_{6.92}$ single crystals synthesized at ambient pressure show metallic behavior but no SDW transition or superconductivity, even under pressures up to ~70 GPa. In contrast, superconductivity under high pressure occurs in the orthorhombic $La_3Ni_2O_{6.85}$ structure. These results highlight a strong correlation between SDW order and superconductivity, providing important constraints on the mechanism of pressure-induced superconductivity in nickelates.

[亮点 1] 首次在常压下合成出四方结构的 $La_3Ni_2O_{6.92}$单晶，并通过高氧压退火修复氧缺陷，使其具有金属性但不显示 SDW 转变。SDW 的存在与抑制是超导出现的关键，而非晶体结构的对称性变化。

[亮点 2] 在高压下，正交结构的 $La_3Ni_2O_{6.85}$中观察到了超导转变，而四方结构的样品在高达 70 GPa 的压力下仍未出现超导。

[亮点 3] 使用氦气作为传压介质，确认高压超导相仍为正交结构，而非先前认为的四方结构。

[思考 1] 使用各种光谱探针证实了斜方晶 La3Ni2O7-δ 中的 SDW 阶数，包括共振非弹性 X 射线散射（RIXS）、核磁共振（NMR）和μ子自旋旋转（μSR）实验，具体是怎样？

[思考 2] 四方 La3Ni2O7-δ 中 SDW 跃迁的缺失和超导性与 c/a_p 对底层电子结构影响之间的具体关联是？

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[拓展阅读 1] 什么是 SDW？

[拓展阅读 1] 自旋密度波（SDW）是一种在特定材料中出现的量子有序态，它本质上是材料内部电子自旋方向（即微小磁矩）呈现出的周期性空间波动图案。其产生的原因主要源于“费米面嵌套”的电子结构不稳定性，即材料中电子的能量-动量关系使得一个特定的波矢能够连接费米面上大面积的平行区域，从而驱动电子系统通过形成这种自旋调制来降低整体能量，达到更稳定的状态。它的主要用处在于为我们理解和设计新颖量子材料提供了关键理论基础，例如它是某些非常规超导体的母态，有助于探索高温超导机制。

[拓展阅读 2] 第一第二临界场和磁场涡旋

[拓展阅读 2] 在II型超导体中，第一临界场 H_{c1} 和第二临界场 H_{c2} 是描述超导—磁场相互作用的两个关键参数：当外加磁场 H 小于 H_{c1} 时，超导体能够完全排斥外部磁场，表现出完全抗磁性（Meissner 效应），内部没有磁场渗入，磁感应强度 B=0；当外加磁场 H 大于 H_{c2} 时，超导性被完全破坏，材料变成普通金属，内部磁场完全渗透，不再具有零电阻和抗磁性；在 $H_{c1}<H<H_{c2}$ 的区间，超导体允许磁场部分渗入，表现为混合态或涡旋态（vortex），即磁通以量子化的磁通管形式穿透超导体，每个涡旋中心超导电流密度为零，周围环绕闭合超导电流，将磁场束缚在涡旋核心内，每个涡旋携带固定的量子化磁通 $\Phi_0=h/2e$，随着外加磁场增加，涡旋密度增加，直到超过 H_{c2} 时涡旋填满材料，超导消失。

[Introduction]

Search for high-temperature superconductivity in nickelates has attracted significant interest among physicists and materials researchers following the discovery of cuprate superconductivity. In 2019, the breakthrough in infinite-layer nickelate $Nd_{1-x}Sr_xNiO_2$ thin films triggered a new wave of searching for novel nickel-based superconductors. Recently, the nickelate superconductor family has been successfully expanded to the Ruddlesden-Popper (RP) phases $La_{n+1}Ni_nO_{3n+1}$, with n = 2 and n = 3. The superconducting transition temperature (T_c) in $La_3Ni_2O_{7-\delta}$ exceeds the liquid-nitrogen boiling temperature, suggesting a new family of high-temperature superconductors. However, unlike the infinite-layer nickelate thin films, the realization of superconductivity in these RP nickelates requires extremely high-pressure conditions, which hinders most spectroscopic measurements of the superconducting state and complicates the study of the underlying mechanism. Exploration of superconductivity at ambient pressure in the RP-phase $La_{n+1}Ni_nO_{3n+1}$ not only offers a promising strategy to overcome the above challenges but also has significant importance for the application of nickelate superconductors.

In $La_3Ni_2O_{7-\delta}$, the NiO_6 octahedra are tilted away from the longest axis at ambient pressure, resulting in an orthorhombic structure with space group Amam (Fig. 1a) rather than a tetragonal structure with space group I4/mmm (Fig. 1b). In the double-layer $La_3Ni_2O_{7-\delta}$ with Amam structure, the bond angle of Ni–O–Ni between adjacent NiO_6 octahedra is 168°. Earlier transport and spectroscopic measurements have revealed a density-wave transition around 150 K at ambient pressure. A similar density-wave transition has also been observed in the trilayer nickelate $La_4Ni_3O_{10-\delta}$, in which an intertwined density wave with both charge and spin order is revealed (~136 K). As pressure increases, the unit cell volume shrinks significantly, making the tilted NiO_6 octahedra unstable. Recent X-ray diffraction (XRD) experiments under high pressure revealed that high pressure induces a structural transition from orthorhombic to tetragonal, and the pressure-induced superconductivity occurs in a tetragonal structure with I4/mmm. In the I4/mmm structure, the bond angle of Ni–O–Ni between adjacent NiO_6 octahedra becomes 180°. Earlier theory suggested that this change of the Ni–O–Ni bond angle can significantly affect the interlayer coupling between NiO planes and is thought to be important for achieving superconductivity under high pressure. Therefore, stabilizing the tetragonal structure was considered a prerequisite for exploring nickelate superconductivity at ambient pressure.

On the other hand, recent density functional theory (DFT) calculations indicate that this change of the Ni–O–Ni bond angle has no significant effect on the band structure, especially for the dz^2-orbital-dominated band. Following this line of reasoning, the tetragonal structure is not crucial for superconductivity. Instead, the suppression of the density-wave transition in orthorhombic double-layer $La_3Ni_2O_{7-\delta}$ is important for achieving superconductivity at ambient pressure.

In the present study, we successfully obtained the tetragonal structure with I4/mmm in $La_3Ni_2O_{7-\delta}$ single crystals at ambient pressure. Moreover, the superconducting structure in pressurized orthorhombic $La_3Ni_2O_{7-\delta}$ has been revisited. Our results indicate that a tetragonal structure is not a necessary condition for achieving superconductivity in

$La_3Ni_2O_{7-\delta}$. The existence of spin-density-wave (SDW) order at ambient pressure seems more vital for pressure-induced superconductivity.

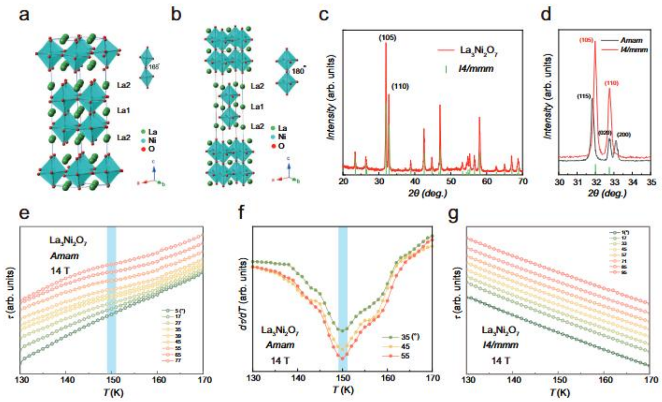

Fig. 1 | The structure and magnetic torque measurements for orthorhombic and tetragonal $La_3Ni_2O_{7-\delta}$ at ambient pressure.

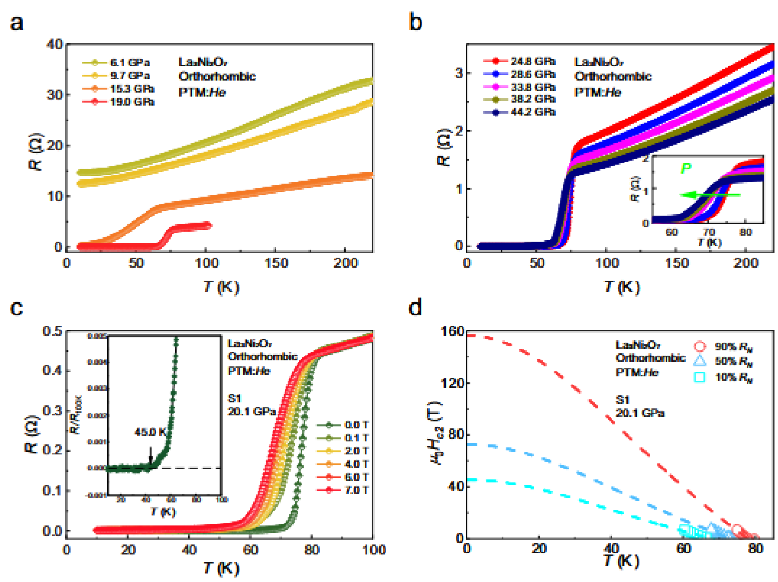

Fig. 2 | Temperature dependence of resistance (R) under various pressures for orthorhombic $La_3Ni_2O_{7-\delta}$.

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编辑：闫婷

投稿人：秦文玲

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