Robust Superconductivity against Al Substitution in $LiTi_2O_4$ Thin Films

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The origin of superconductivity in $LiTi_2O_4$ remains an important question because its mixed-valence Ti network supports a complex electronic state in which carrier filling, disorder, and charge frustration may influence pairing. Building on earlier substitution studies [1], we grow Al-substituted $Li(Ti_{1-x}Al_x)_2O_4$ thin films on $MgAl_2O_4(001)$ by pulsed-laser deposition to test how superconductivity responds when this network is chemically perturbed. Superconductivity persists up to nominal 50% Ti-site substitution even as the normal state becomes substantially more resistive (Fig. 1). This contrast indicates that normal-state conduction and superconductivity have markedly different sensitivities to Al substitution. Zero-resistance transitions and comparatively stable transition temperatures and upper-critical-field scales support the robustness of superconducting transport. The fourfold angular response previously reported in undoped films [2] also survives at the heavily substituted endpoint. Structural characterization reveals lattice contraction, while spectroscopic ellipsometry and density-functional calculations probe the accompanying electronic evolution. The calculations identify stoichiometric $LiTiAlO_4$ as a carrier-depleted limit, emphasizing that Al substitution changes nominal electron filling and Ti connectivity as well as introducing nonmagnetic disorder. The superconducting response must therefore be understood in the context of a changing electronic system, beyond a simple impurity-scattering picture. Because Al substitution is expected to disrupt the mixed-valence network that can support charge frustration, persistent superconductivity argues against charge frustration on an intact Ti network being essential to pairing. These films provide a platform for disentangling the roles of carrier filling, disorder, and charge frustration in oxide superconductivity.

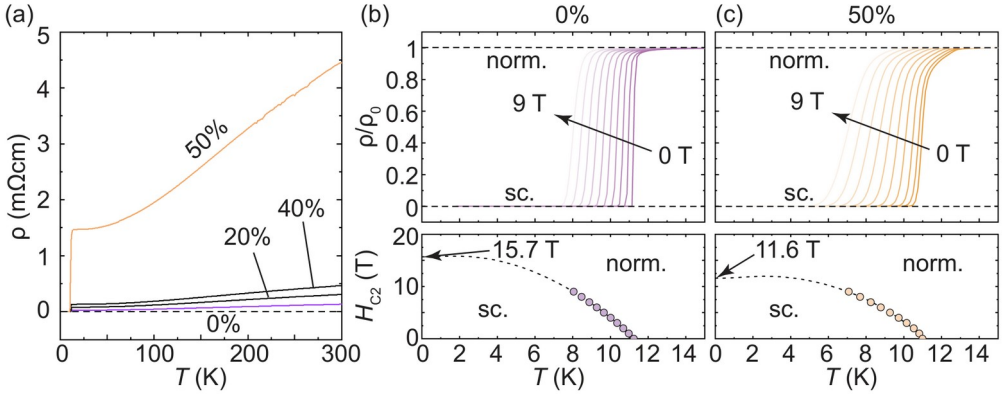

Figure 1. (a) Resistivity versus temperature. (b,c) Field-dependent transitions and critical-field boundaries for undoped and nominal 50%-Al films; curves include extrapolations.

[1] P. M. Lambert, P. P. Edwards, and M. R. Harrison, J. Solid State Chem. 89, 345–360 (1990).
[2] H. Xue et al., ACS Nano 16, 19464–19471 (2022).