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#### ACCEPTED MANUSCRIPT

# Effect of Hydrothermal Conditions on Superconductivity and Magnetism in $[Li_{1-x}Fe_xOH]FeS$

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#### Abstract

Recent reports of superconductivity and magnetism in single crystals of  $[\text{Li}_{1-x}\text{Fe}_x\text{OH}]\text{FeS}$  show unexplained variations in both superconducting and magnetic properties. We investigate the effect of hydrothermal growth conditions on these properties and find that increasing the growth temperature systematically increases the superconducting transition temperature  $(T_c)$ , sharpens the magnetic transition, and decreases the scattering rate  $(\Gamma)$ . The slow rate of  $T_c$  suppression with increasing  $\Gamma$  indicates a conventional s-wave superconducting state according to the Abrikosov-Gorkov expression. Samples with higher scattering rate show broader magnetic transitions and a stronger temperature dependence in the magnetic susceptibility. These results identify disorder, due to interstitial iron impurities, as the unique internal parameter responsible for the unexplained variations in  $T_c$  and magnetic ordering. We demonstrate the optimal hydrothermal growth conditions to minimize disorder and maximize  $T_c$  in  $[\text{Li}_{1-x}\text{Fe}_x\text{OH}]\text{FeS}$  crystals.

Keywords: Intercalation, Hydrothermal, Superconductivity, Magnetism

#### 1. Introduction

Mackinawite is a naturally occurring mineral of iron and nickel with the formula  $(\text{FeNi})_{1+x}$ S and a tetragonal unit cell in the space group P4/nmm [1]. Pure FeS crystallizes in two forms: the stable hexagonal phase (h-FeS) with a NiAs-type structure, and the metastable tetragonal phase (t-FeS) with an anti-PbO-type structure [2]. Recently, single crystals of the metastable t-FeS were grown using a hydrothermal method and found to be superconducting below 5 K [3, 4]. The search for superconductivity in tetragonal iron sulfide with Mackinawite structure was motivated by the closely related tetragonal iron selenide (t-FeSe) which is also metastable and superconducting [5]. Whereas the stable hexagonal phase is not superconducting in both FeS and FeSe, the metastable tetragonal phase is superconducting.

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