ThFeAsN: Nitrogen-containing iron-pnictide superconductor

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Iron-based superconductors (FeSC) are currently under intense study. These materials contain a building block of Fe^{2+} square lattice, with tetrahedral coordination of Pn (As and/or P) or chalcogenide ions. Compared to other high T superconductors such as cuprates, FeSCs bears the merits of resistant to impurities and high current density through grain boundary.[1,2] In FeSCs, superconductivity arises from electron pairing through attractive interaction while coulombic repulsion is avoided, instead of through phonon-mediated attractive interaction, as described in BCS Theory.[3]

Iron-pnictides are FeSCs based on elements from group 15 in the periodic table. Among the iron-pnictides, the 1111-type family, which represents a 1:1:1:1 stoichiometric composition, is well known and widely studied. In 2008, F doped LaFeAsO, was first reported to exhibit superconductivity at T_c = 26K, which was comparable to that of cuprate.[4] Sm doped SrFeAsF, with T_c = 56K[5], is one of the iron-pnictide superconductors with the highest T_c to date. Fig. 1 shows the schematic phase diagram of 1111-type FeSCs. For the majority of iron pnictides, doping of electrons or holes into the stoichiometric, antiferromagnetic (AFM) parent material is necessary to modify their Fermi-surface so as to suppress its magnetic order and induce superconductivity.[1] Dopant-free iron pnictides superconductors with stoichiometric composition are scarce, and are known to conduct with zero resistance at very low temperature. In the 1111-type family, LaFePO was previously reported to be a dopant-free bulk superconductor, with T_c = 4K.[6]

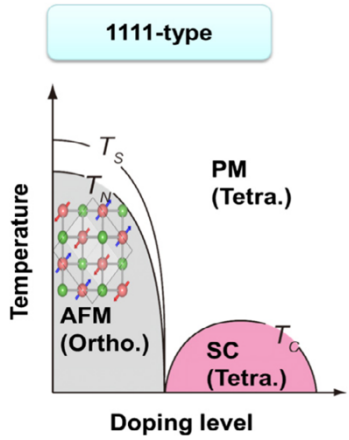

Figure 1. Schematic phase diagrams of 1111-type FeSCs.

ThFeAsN, the first nitrogen-containing iron pnictide superconductor, has recently been discovered. Its unique composition, stoichiometric bulk superconductivity at a relatively high T_c, and absence of magnetic order and structural distortion has aroused intensive research on this material.[7,8,9]

In 2016, Wang et al. first reported on the superconductivity of ThFeAsN. The material was synthesized using Th metal, Th_3N_4 and FeAs as precursors. The reactions were carried out in an oxygen-free environment. Powder XRD and Rietveld refinement profile using the 1111-type structure model support a tetragonal unit cell, with fluorite-like $[Th_2N_2]^{2+}$ layers at the top and bottom of the unit cell, separated by antifluorite-like $[Fe_2As_2]^{2-}$ layers. No impurity was observed from the XRD pattern. Resistivity and magnetic susceptibility χ measurements both support the onset of a bulk superconductivity at 30K (Fig. 2). Suppression of T_c was observed with doping, indicating that the bulk ThFeAsN already possesses its maximum T_c.[7]

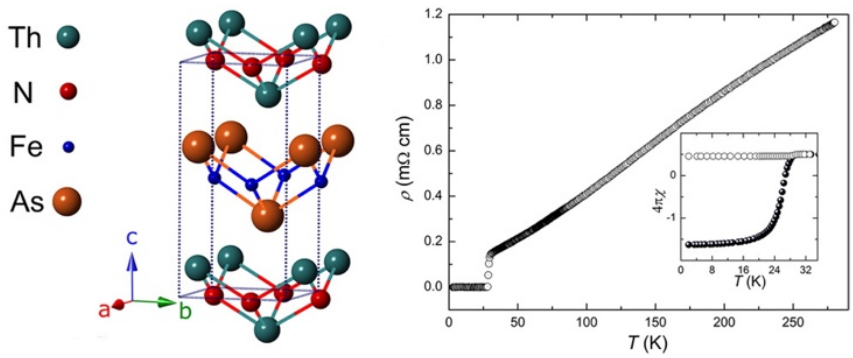

Figure 2. Crystal structure (Left) and temperature dependence of resistivity at zero magnetic field of polycrystalline ThFeAsN (Right).

Following the first report on ThFeAsN, Shiroka et al. used muon-spin rotation/relaxation (μSR) to study its magnetic and electronic properties. Zero field (ZF)-μSR measurements (Fig. 3) showed no muon-spin precession above and below T_c, which indicates that no long-range magnetic order was present preceding the onset of superconductivity, which is different from previously studied $LaFeAsO_{1-x}F_x$, for which a doping of $x \geq 0.04$ is necessary to suppress its magnetic order.[8,10]

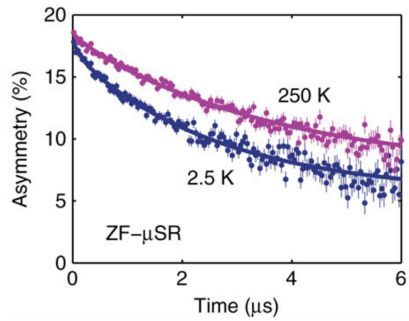

Figure 3. ZF-μSR time spectra at 250K and 2.5K.

Neutron powder diffraction was also carried out on the polycrystalline ThFeAsN from 6 K to 300 K. The matching of patterns at 6 K and 40 K once again proves the lack of magnetic order or structural distortion in the material. This contradicts previous studies on LaFeAsO, where new peaks were observed in the stoichiometrically composed parent material through the cooling process due to emerging magnetic order and structural transition from tetragonal to orthorhombic.[9,11]

All of the measurements above contradict the first-principle calculations of the material, where its lowest magnetic ground state is expected to be a stripe-type antiferromagnetic state.[12,13] Attempt to induce the magnetic order by doping Y and O into ThFeAsN, has turned out to be unsuccessful. A possible explanation is the self-doping effect, or intrinsic interlayer charge transfer, which leads to a suppression of the magnetic order in the stoichiometrically composed material. This is supported by a shorter distance between N^{3-} in the crystal structure, indicating covalent bonding between them. This lowers effective nitrogen valence, facilitating interlayer charge tranfer.[6,14]

Since its recent discovery, ThFeAsN has been widely studied in the past two years. The absence of magnetic order in this stoichiometric bulk superconductor and a relatively high T_c has been a heated topic in the field of iron pnictides and FeSCs. Future studies include development of single crystal synthesis and ARPES study. Better understanding of its phase diagram, further investigation on its Fermi-surface, and identifying the origin of its superconductivity might be crucial to unraveling the mystery of FeSCs.

References

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