Fe-based superconducting thin films and their potential for high field applications

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Iron based superconductors (IBSs)

**FeAs or FeSe or FeTe tetrahedron**

- Common structure
- Important role for superconductivity

\[ T_{c}^{\max} \approx \begin{align*} 15 \text{ K} & \quad \text{FeAs} \\ 18 \text{ K} & \quad \text{FeSe} \\ 56 \text{ K} & \quad \text{FeTe} \\ 43 \text{ K} & \quad (\text{Li, Fe})\text{OHFeSe} \\ 38 \text{ K} & \quad A\text{Fe}_{2}\text{As}_{2} \end{align*} \]

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Progress of $J_c - H$ for NdFeAs(O,F) (Nd-1111)

$J_c \sim 8$ MA/cm$^2$ for NdFeAs(O,F)

4.2 K $H \parallel c$

2019 Kauffmann-Weiss et al., Nanoscale Adv. 1, 3036

2016 Tarantini et al., Sci. Rep. 6, 36047
Measured $J_c$ is around 10~20% of $J_d$

$J_c \approx 8 \text{ MA/cm}^2$ for NdFeAs(O,F)

Higher the $J_d$, higher the $J_c$

Depairing current density $J_d$

(theroretical upper limit)

$$J_d(T) = \frac{\phi_0}{3\sqrt{3}\pi\mu_0\lambda^2(T)\xi(T)}$$
Over-doping increases the condensation energy

- $J_c \sim 8$ MA/cm² for NdFeAs(O,F)
- Higher the $J_d$, higher the $J_c$
- Depairing current density $J_d$ (theoretical upper limit)

$$J_d(T) = \frac{\phi_0}{3\sqrt{3}\pi\mu_0\lambda^2(T)\xi(T)}$$

- Over-doping increase $J_d$, and hence $J_c$

Combining over-doping and microstructural modification

- Depairing current density $J_d$ (theoretical upper limit)
- Higher the $J_d$, higher the $J_c$
- Over-doping increase $J_d$, and hence $J_c$

$J_d(T) = \frac{\phi_0}{3\sqrt{3}\pi\mu_0\lambda^2(T)\xi(T)}$

- $J_c \sim 8 \text{ MA/cm}^2$ for NdFeAs(O,F)
- $\sim 150 \text{ MA/cm}^2$ @ 4.2 K

** M. Miura et al., NPG Asia Mater. 14, 85 (2022).
Solubility limit depends on F and H

\[
\text{O}^{2-} \rightarrow \text{F}^- \text{ or H}^- + e^- \quad \text{(electron doping)}
\]

- Substitution level is limited up to \(~0.2\) (For \(\text{SmFeAsO}_{1-x}\text{F}_x\))
- For H, the substitution level is increased up to \(~0.8\)
- Heavily electron doped film can be obtained
- \(T_c\) keeps constant around 50 K up to \(x=0.4\)

\(\text{LnFeAsO} \ (\text{Ln}: \text{lanthanoide})\)


IEEE-CSC, ESAS, and CSSJ SUPERCONDUCTIVITY NEWS FORUM (global edition), October 2023. Invited presentation given at EUCAS 2023, Sept. 5, 2023, Bologna, Italy
**LnFeAsO: penetration depth and coherence length**

- $\lambda$ of $LnFeAsO$ may be decreased with electron doping
- No clear relation between $\xi$ and electron doping

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Growth of F- and H-doped NdFeAsO thin films

NdFeAsO$_{1-x}$F$_x$

parent NdFeAsO

NdFeAsO$_{1-x}$H$_x$

Nd(O,F) overlayer

NdFeAs(O,F) $\rightarrow$ F$^-$/O$^2-$

MgO(001) sub.

NdFeAsO

vacuum

CaH$_2$ powder

heat treatment

400~500°C up to 72 h

Phase pure & c-axis oriented
d~24 nm


K. Kondo, K. Iida et al., SuST 33, 09LT01 (2020).
Comparison between H- and F-doping: $T_c$ and carrier density

- A constant $T_c$ of ~ 50 K up to $c$~8.54 Å, corresponding to carrier density $n$~$2 \times 10^{21}$ /cm$^3$

- $c$ varied in a wide range, 8.44 Å and 8.55 Å with $T_c$~50 K [NdFeAs(O,H)]

- The maximum $n$ was around $6 \times 10^{21}$ /cm$^3$ [almost 3 times higher than NdFeAs(O,F)]

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K. Kondo, K. Iida et al., Sust 33, 09LT01 (2020).

Invited presentation given at EUCAS 2023, Sept. 5, 2023, Bologna, Italy
\( J_c \) for NdFeAs(O,H) is higher than that for NdFeAs(O,F)

- \( J_c(4\,\text{K},0\,\text{T}) > 17\,\text{MA/cm}^2 \), which is twice larger than NdFeAs(O,F)
  - c.f.) 20 MA/cm\(^2\) for the irradiated SmFeAs(O,F) single crystal \[1\]
- \( F_p \) of NdFeAs(O,H) is 1.5 times higher than that of NdFeAs(O,F)

NdFeAs(O,H) shows higher $H_{\text{irr}}$ than NdFeAs(O,F)

- Upper critical field $H_{c2}$ of NdFeAs(O,H) is almost comparable to that of NdFeAs(O,F)
- Irreversibility field $H_{\text{irr}}$ of NdFeAs(O,H) is higher than that of NdFeAs(O,F)

$H_{\text{irr}} \propto H_{c2}^{\gamma_2}$ for $H \parallel c$

Hanzawa et al., PRM 6, L1118 (2022).
Band calculation

Fully substituted $H$ and $F$ of 1111

\begin{align*}
\text{CaFe}_{0.9}\text{Co}_{0.1}\text{AsH} \\
\text{CaFe}_{0.9}\text{Co}_{0.1}\text{AsF}
\end{align*}

Evolution of band structure with $H$

\begin{align*}
\text{NdFeAsO} & \quad \text{NdFeAs(O,H)} \\
\text{LaFeAsO} & \quad \text{CaFe}_{0.9}\text{Co}_{0.1}\text{AsH}
\end{align*}

V. Vildosola et al., PRB 78, 064518 (2008).

Y. Muraba et al., PRB 89, 094501 (2014).
(Ba,K)Fe$_2$As$_2$
K-doped $\text{AElFe}_2\text{As}_2$: Only epitaxial thin film has not been realised

- High $T_c \sim 38$ K
- $J_d \sim 170$ MA/cm$^2$
- $\gamma \sim 1 - 2$

- High $T_c \sim 38$ K, high $J_d \sim 170$ MA/cm$^2$ & low electromagnetic anisotropy
- The most promising material for applications (e.g. PIT wires and bulk magnets)
- All material forms except for epitaxial thin films have been available
K-doped Ba122 epitaxial thin film on CaF$_2$ sub.

Fluoride sub. & low growth temperature (~400$^\circ$ C) are the key to epitaxial growth

Toward bi-crystal K-doped Ba122 experiments

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$^{1}$ La$_{0.3}$Sr$_{0.7}$Al$_{0.65}$Ta$_{0.35}$O$_3$
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1) La$_{0.3}$Sr$_{0.7}$Al$_{0.65}$Ta$_{0.35}$O$_3$

- Oxygen is released during deposition (SrTiO$_3$)
- Reaction layer is present at the interface (LSAT)
- Ba122 grows epitaxially on MgO
(Ba,K)Fe$_2$As$_2$ was not epitaxially grown -> due to the low growth temperature (cf. 800°C for Co-$^{[1]}$ and P-doped$^{[2]}$ Ba122)

30° rotated grains were present

Strategy for K-doped Ba122 on MgO

K-doped Ba122

Ba122

CaF₂ cover layer

K-doped Ba122

Ba122 buffer

MgO sub.

20 nm (720°C)

80 nm (400°C)

~30 nm

Interface

K-doped Ba122

Ba122 buffer layer

Sharp interface between Ba122 and K-doped Ba122!

D. Qin et al., Sust 35, 09LT01 (2022).

Invited presentation given at EUCAS 2023, Sept. 5, 2023, Bologna, Italy
The 00l peaks arising from K-doped Ba122 and Ba122 were observed. K-doped Ba122 was grown epitaxially on Ba122-buffered MgO. $T_c$ of K-doped Ba122 was 37.5 K.
K-doped Ba122 bicrystals are realised

- Structurally fine K-doped Ba122 films are grown on MgO bi-crystal substrates
- Microbridge was fabricated by all dry processes (Ar ion etching or laser cutting)

\[ \theta_{GB} = 13^\circ \]

103 $\phi$-scans

Grain Boundary (GB)

intra-grain

inter-grain

GB

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Transport properties of K-doped Ba122 bicrystals

- Even at 12 K and 20 K, inter-grain $J_c$ of K-doped Ba122 is higher than those of other IBS [Co-doped Ba122 (12 K), Fe(Se,Te) (4 K), NdFeAs(O,F) (4 K)].

- Critical grain boundary angle $\theta_c$ of (Ba,K)122 is ~9°, similarly to other

T. Hatano et al., in preparation
Transport properties of K-doped Ba122 bicrystals

- Even at 12 K and 20 K, inter-grain $J_c$ of K-doped Ba122 is higher than those of other IBS [Co-doped Ba122 (12 K), Fe(Se,Te) (4 K), NdFeAs(O,F) (4 K)]
- Critical grain boundary angle $\theta_c$ of (Ba,K)122 is $\sim 9^\circ$, similarly to other IBS
- The $\theta_c$ of (Ba,K)122 is unchanged by applied magnetic fields (c.f. Co-doped Ba122)
1. Over-doped NdFeAsO showed a high $J_c$ and low anisotropy

   -> The strategy for over-doping method can be applicable

2. Low angle GBs and their networks work as strong pinning centers in K-doped Ba122

3. Inter-grain $J_c$ exceed 1 MA/cm$^2$ even at $\theta_{GB}=24^\circ$ and 12 K

4. The critical angle $\theta_c$ for all IBSs seem to be $\sim 9^\circ$

5. The $\theta_c$ for K-doped Ba122 is unchanged by magnetic field