Thin-film Growth of 1111-type Iron-based Superconductors

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After our report on heteroepitaxial growth of LaFeAsO thin films by pulsed laser deposition (PLD) using 2ω of a Nd:YAG laser [1], molecular beam epitaxy (MBE) is the most effective technique to fabricate superconducting 1111-type F-doped ReFeAsO (Re = rare earth) epitaxial films [2]. However, technical difficulties in the 1111-type film-growth have not been completely resolved both for PLD and MBE because only a few research groups can grow the films and the number of papers on the 1111 films is quite limited, compared with other iron-based films such as 122- and 11-types. Particularly, there is still no report on *in-situ* PLD growth of a superconducting 1111-type thin film.

Under such circumstances, we focused on H-doping of parent SmFeAsO (Sm1111) epitaxial films. We initially attempted H-doping by *in-situ* PLD under H_2 gas and rf-generated H radical atmospheres as well as by post-deposition thermal annealing treatment under H_2 gas atmosphere. Then, we adopted an atomic hydrogen source into the PLD growth chamber because thermally cracked hydrogen atoms should be more active. However, no Sm1111:H film could be obtained. Therefore, we selected a topochemical reaction between undoped Sm1111 epitaxial films and CaH_2 powders, succeeding in fabricating high-T_c Sm1111:H films [3]. Binary hydrides other than CaH_2, such as MgH_2, SrH_2, and BaH_2, were also effective to dope hydrogen [4]. Magnetization measurements of the films potentially revealed high critical current densities (J_c) of ~1 MA/cm^2 both under a self-field and 9 T [4]. Iida *et al*. clarified that the H-doping method using CaH_2 is applicable also to 1111-type MBE-grown NdFeAsO films, which exhibit a high self-field J_c >10 MA/cm^2 [5].

Although a high-pressure synthesis technique at 2–5 GPa is useful to fabricate polycrystalline bulks of 1111-type ReFeAsO:H, it is still difficult to grow enough large-size single crystals of Sm1111:H [6]. Thus, more detailed investigations on intrinsic physical properties, particularly depending on crystallographic orientation, were difficult. However, the Sm1111:H epitaxial films enable us to examine them. By employing high pulsed magnetic fields up to 130 T, we experimentally clarified that the upper critical field ($H_{c2}||ab$) and its anisotropy parameter (γ) of the Sm1111:H are 120 T and 2, respectively [7]. Especially, it should be noted that the small γ value is comparable to that of 122-type $BaFe_2As_2$ with lower T_c and much smaller than that of F-doped 1111. Therefore, the small γ is the distinct feature of Sm1111:H. Furthermore, recently Miura *et al*. precisely examined transport J_c and H_{c2} of our Sm1111:H epitaxial films, and found that the depairing current density (J_d) of the Sm1111:H is extremely high (maximum 415 MA/cm^2), comparable to that of cuprates. Additionally, they also performed proton irradiation to introduce high-density effective pinning centers in the Sm1111:H films, leading to demonstration of a quite high in-field $J_c||c$ of ~3 MA/cm^2 at 25 T [8].

References

[1] H. Hiramatsu *et al.*, *Appl. Phys. Lett.* **93**, 162504 (2008). DOI: 10.1063/1.2996591
[2] *For a review*, J. Hänisch *et al.*, *Supercond. Sci. Technol.* **32**, 093001 (2019). DOI: 10.1088/1361-6668/ab1c00
[3] J. Matsumoto *et al.*, *Phys. Rev. Mater.* **3**, 103401 (2019). DOI: 10.1103/PhysRevMaterials.3.103401
[4] H. Hiramatsu *et al.*, *Appl. Phys. Express* **13**, 073002 (2020). DOI: 10.35848/1882-0786/ab998f
[5] *For a review*, K. Iida *et al.*, *Supercond. Sci. Technol.* **36**, 063001 (2023). DOI: 10.1088/1361-6668/acccb2
[6] *For a review*, S. Iimura and H. Hosono, *J. Phys. Soc. Jpn.* **89**, 051006 (2020). DOI: 10.7566/JPSJ.89.051006
[7] K. Hanzawa *et al.*, *Phys. Rev. Mater.* **6**, L111801 (2022). DOI: 10.1103/PhysRevMaterials.6.L111801
[8] M. Miura *et al.*, *Nat. Mater.* **23**, 1370 (2024). DOI: 10.1038/s41563-024-01952-7