In-situ Growth of Superconducting *Ln*FeAs(O,F) (*Ln*=Nd, La) Thin Films by Molecular Beam Epitaxy

Abstract :

High-quality epitaxial thin films are indispensable for exploring the intrinsic properties and for electronic device applications of a superconductor. Many research groups have reported on thin film preparation of Fe-based superconductors, but reports on *Ln*FeAsO (*Ln*=lanthanide) thin films, the so-called 1111 family, which possesses the highest T_c among the Fe-based superconductors known to date, are very limited. In our previous work, we had succeeded in growing Nd-1111 thin films on GaAs substrates by molecular beam epitaxy (MBE) [1,2]. The films grown with a growth time of 1 h were single phased but did not show a superconducting transition. On the other hand, the films showed a well defined superconducting transition when the growth time was increased, although the superconducting films contained some impurities such as NdOF. The highest onset temperature T_c^{onset} of these films was 48 K with a zero resistance temperature T_c^{0} of 42 K. This was achieved without an ex-situ heat treatment, and the observed T_c is the highest among the so-far reported thin films of Fe-based superconductors.

The reason why a superconducting thin film was obtained with increasing the growth time is probably related to the formation of a NdOF layer on top of the NdFeAs(O,F) layer, as was found by a depth profile analysis using Auger electron spectroscopy [2]. The formation of the NdOF layer can be attributed to an excessive supply of fluorine due to the use of NdF₃ as the source of Nd and F. This means that an independent control of fluorine and Nd is necessary to improve the film quality. On the other hand, SEM observations revealed that the GaAs substrate was etched during the early stage of the growth. We think that the etching was caused by fluorine as it can react with GaAs and forms GaF₃, while the formed GaF₃ immediately sublimate because the substrate temperature was higher than the sublimation temperature of GaF₃. This model suggests that Ga may work as a getter and can be used to regulate the amount of fluorine. Indeed, the substrate was not etched by fluorine when a Ga flux with an adequate density was supplied during the growth of the Nd-1111 film. We also found that the etching of the GaAs substrate was prevented when a F-free layer of NdFeAsO was grown on the substrate prior to the F-containing NdFeAs(O,F) layer. These methods enable a more controlled growth of Ln-1111 films, which is important for further improving the film quality.

We have also grown *Ln*FeAsO thin films on MgO substrates. The use of Ga as a F-getter was crucially important for the growth of Nd-1111 on MgO substrates, because MgO does not react with fluorine unlike GaAs. The Nd-1111 phase was not formed on MgO substrate when no Ga flux was supplied, but a single phase film of F-free NdFeAsO was obtained by adjusting the flux density of Ga. We also succeeded in the growth of LaFeAsO films by changing the deposition parameters only slightly. An attempt of growing F-doped *Ln*FeAsO thin films on MgO substrates is underway by further optimizing the Ga flux.

[1] T. Kawaguchi et al., Appl. Phys. Express 2, 093002 (2009).

[2] T. Kawaguchi et al., Appl. Phys. Lett. (in press).