Morphological variability of CeIn₃ thin films grown by molecular beam epitaxy

Foyevtsov O and Huth M
Physikalisches Institut, Goethe University, Frankfurt am Main, Germany

Abstract. We report results concerning the thin film growth of CeIn₃ using the molecular beam epitaxy technique. A series of films on α-Al₂O₃ (a- and c-plane), and MgO(100) substrates was grown by co-evaporation of the constituent elements. We observed strong islanding, and, as a consequence, discontinuous films with a Ce:In ratio of 1:3. We suggest that the islanding is driven by de-wetting processes induced by the In component, since In is well known for its de-wetting characteristics when grown on insulating surfaces. We furthermore observed a pronounced morphological variability for films grown under nominally identical conditions on MgO (100) and α-Al₂O₃ (c-plane) substrates. For films on α-Al₂O₃ (a-plane) the morphological structures proved to be more reproducible. By optimizing the growth process it is possible to stabilize a surface morphology consisting of well-isolated micron-sized CeIn₃ islands. Electronic transport measurements were performed on these micro crystals and on thin films.

1. Introduction
CeIn₃ is a heavy-fermion antiferromagnet that becomes superconducting under pressure [1]. Structurally, it is related to another heavy fermion superconductor CeCoIn₅, which is highly interesting due to its non-Fermi liquid behavior and its highest transition temperature among all heavy fermion superconductors [2]. A commonly used method for synthesizing these compounds is growing from In flux. However, samples obtained by this method are not suitable for surface-sensitive measurements, such as tunneling spectroscopy, which is a valuable method for probing the superconducting state. Such measurements are possible on thin films [3, 4].

2. Results and discussion
CeIn₃ films were grown using the molecular beam epitaxy (MBE) by evaporation of constituent components of purity no less than 99.9 % from individually controlled effusion cells. Tantalum and pyrolitic boron nitride crucibles were used for Ce and In, respectively. Pressure during the growth was lower than 6 × 10⁻⁹ mbar. The films were grown on a-, c-plane α-Al₂O₃ and MgO (100) epitaxially polished substrates, which were chemically cleaned before every growth. The temperature of the substrates was varied between 500 C and 600 C. For structure characterization, we used x-ray diffraction taking ω/2θ scans and rocking curves. For high-resolution morphology investigations we used a scanning electron microscopy (SEM). Analysis of the sample composition was performed by energy-dispersive x-ray spectroscopy (EDX).

The series of films that we obtained demonstrate a rich variability of the surface morphology at nominally identical growing conditions. We observe a strong tendency for island growth with occasionally rather well isolated micron-sized CeIn₃ crystallites. This morphology was
most reproducible on $\alpha$-Al$_2$O$_3$ (a-plane) substrates at a substrate temperature of 600 °C. X-ray diffraction shows that the crystallites of CeIn$_3$ are mostly of (111) orientation.

We assume that this discontinuous structure of the films is due to the well-known dewetting properties of the In component, which renders the growth process extremely sensitive to small variations in growth parameters. Such uncontrollable factors as small fluctuations in the component fluxes from growth to growth or small variations of the substrate temperatures may have a great influence on the resulting surface morphology.

In an attempt to improve growth control a set of films was simultaneously grown during a single growth process so that only the substrate temperature varied between the films in one set. A number of substrates were glued on a holder constructed in such a way that its temperature varied from substrate to substrate. We estimated the temperature gradient by numerically solving the heat conduction and radiation coupling equation for this holder. The thus obtained films demonstrate a systematic change in their surface morphology with substrate temperature (see Fig. 1). The film grown at the highest temperature (with the nominal value of 550 °C) has the lowest density and largest average diameter of the CeIn$_3$ islands. The films grown at lower temperatures demonstrate a logical sequence of increasing density of islands with decreasing substrate temperature. This morphology evolution can simply be explained by the increasing mobility of the adatoms as the substrate temperature increases. The compositions of all films in this set are equal within the accuracy of the EDX method and correspond to the Ce:In ratio of 1:3. The stability of the films’ composition indicates that, except of the variations in substrate temperature, all other growth conditions were identical.

![Figure 1](image_url)

**Figure 1.** Morphological variability of the surface for different substrate temperatures (A: highest substrate temperature, D: lowest substrate temperature). The area shown for all pictures is approximately 11×10 µm$^2$.

Since we achieved good reproducibility for films with well-isolated micron-sized CeIn$_3$ crystallites, it was possible to perform electronic transport measurements on an individual micro-crystal. Contact preparation was done in several steps. First, four 20 nm thick Cr contact pads were prepared on the substrate surface with an area of $1 \times 2$ mm$^2$ and an inter-contact distance of 50 µm. Next, using a stencil mask with an open, round window matched to the point in between the four Cr contact pads, we grew a film of CeIn$_3$ composition with the previously optimized growth parameters. In the SEM we selected a CeIn$_3$ micro-crystal and checked its composition via EDX. The micro-crystal was then isolated using focused ion beam etching. Finally, ion beam induced deposition of Pt was employed to wire the micro-crystal to the Cr contact pads. In Fig. 2, we present preliminary results of the resistance measurements of the micron-sized CeIn$_3$ crystal (right) and of the whole film (left) as a function of temperature. The measurements were performed by the four-probe method.

Evidently, we observe the same qualitative behavior in the resistivity of the film and of the micro-crystal. This behavior is completely analogous to that observed for bulk crystals [5] of CeIn$_3$ obtained by the most commonly used method of growing from In flux. The residual
resistance ratio (RRR) of our micro-crystal is about 1.1 and about 2 for the film. These small values for RRR are indicative of an inferior crystal quality of our film and micro-crystal if compared to that of good single crystals. Work to optimize the MBE process is currently in progress.

3. Conclusion
A series of (111)-textured CeIn₃ films on α-Al₂O₃ (a- and c-plane) and MgO(100) substrates were obtained using the MBE technique. The films exhibit a strong morphological variability at nominally identical growth conditions. The α-Al₂O₃ (a-plane) substrate surfaces provides the best template for obtaining a sufficient reproducibility of the surface morphology of the films. We were able to investigate the influence of small variations of the substrate temperatures on the morphology and observed systematic changes which we could reconcile with the influence of the substrate temperature on the mobility of the adatoms of the growing layers. We conducted resistance measurements on an individual micro-crystal of CeIn₃, as well as on the films. The temperature dependence of the resistance was in good qualitative agreement with that known from bulk crystals obtained by growth from In flux.

4. Acknowledgment
This work was supported by the Deutsche Forschungsgemeinschaft (DFG) through grant No. HU 752/3-3

References
[1] Mathur N D, Grosche F M, Julian S R, Walker I R, Freye D M, Haselwimmer R K W and Lonzarich G G 1998 Nature 394 39
[2] Petrovich C, Pagliuso P G, Hundley M F, Movshovich R, Sarrao J L, Thompson J D, Fisk Z and Monthoux P 2001 J. Phys.: Cond. Mat. 13 L337-42
[3] Jourdan M, Huth M and Adrian H 1997 Physica 230 335
[4] Jourdan M, Huth M, Hessert J and Adrian H 1998 Nature 398 47
[5] Walker I R, Grosche F M, Freye D M and Lonzarich G G 1997 Physica C 282-287 303-6