Formation of Single-Crystal Copper Oxide by Laser-induced Crystallization: Chevron Beam-Profiles Work While Gaussian Beam-Profiles Fail

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Laser-induced-crystallization (LIC) frequently adopted in attempts of crystallizing non-single-crystal thin films offers attractive features advantageous for functional devices that need to be built on non-single-crystal substrates such as glasses for which epitaxial growth–a conventional technique to obtain single-crystal thin films–does not serve. LIC has a long history, dating back to the late 1970s with a significant emphasis on elementary semiconductors such as Si. Conventional LIC frequently adopts laser with a Gaussian beam-profile; however, it merely produces polycrystal (i.e., conventional LIC fails to yield single-crystal). In this paper, selective area crystallization of non-single-crystal copper(II) oxide (CuO) is described. The crystallization is achieved by unconventional LIC with a beam-profile in the shape of chevron–a marked contrast to Gaussian beam-profiles. The crystallization is verified by observing a transition from an non-single-crystal phase consisting of small crystalline (~100 nm × 100 nm) grains of CuO to an single-crystal phase of copper(I) oxide (Cu_2O). Provided the experimental demonstration, a theoretical assessment based on a cellular automaton model, with the behaviors of localized recrystallization and stochastic nucleation, is developed. The theoretical assessment qualitatively predicts the dependence of vital observable features (e.g., size and gross geometry of crystalline domains) obtained in the experiment on the key LIC conditions. The theoretical assessment further predicts that differences in resulting crystallinity, either single-crystal or polycrystal, primarily depend on the geometric details with which non-single-crystal regions exposed to laser melt in relation to the scan direction of the laser. Concave-trailing profiles yield larger crystalline domains, which lead to single-crystal, while convex-trailing profiles result in smaller crystalline grains, which lead to polycrystal, casting light on the fundamental question: Why do chevron-beam profiles succeed in producing single-crystal, while Gaussian-beam profiles fail? Fig. 1 compares the two cases.

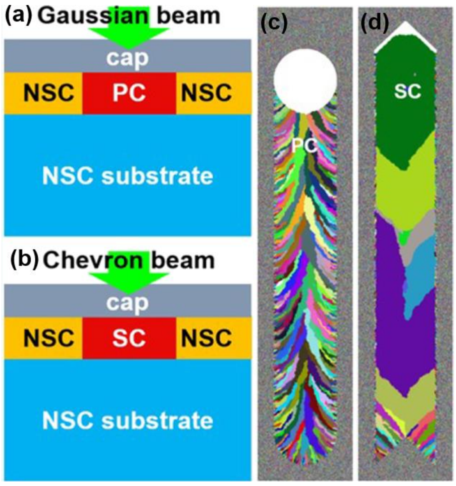

Fig. 1: (a) Conventional LIC with a Gaussian beam-profile produces polycrystal (PC) while (b) LIC with a chevron beam-profile yields single-crystal. (c)(d) the two cases used in the modeling: Gaussian beam-profile in (c) and chevron beam-profile in (d) are compared, qualitatively suggesting that the Gaussian beam-profile is likely to fail in producing large domains while the chevron beam-profile offers a better chance of forming a single-crystal, which is consistent with the experiment.