

Cryogenic deposition and crystallization of ultrathin refractory superconductors

T. A. J. van Schijndel^{1,+}, Y. Wu¹, J. J. Choi², M. Coombs³, J. T. Dong³, Y. Gul⁴, G. P. Mazur³,
C. J. Palmström^{1,3}

¹ Electrical & Computer Engineering Department, University of California Santa Barbara, Santa Barbara, CA 93106, USA

² Physics Department, University of California Santa Barbara, Santa Barbara, CA 93106, USA

³ Materials Department, University of California Santa Barbara, Santa Barbara, CA 93106, USA

⁴ London Centre for Nanotechnology, University College London, London WC1H 0AH, UK

Refractory metals are important materials for superconducting electronics and quantum circuits. Many of these applications require thin films with controlled morphology and crystal structure, but achieving this control becomes particularly challenging at thicknesses of only a few nanometers. In conventional physical-vapor deposition, morphology and crystal structure depend strongly on adatom mobility, which is influenced by the substrate temperature relative to the melting temperature. Therefore, the high melting temperatures of refractory metals limit adatom mobility at typical substrate temperatures. However, increasing substrate temperatures can promote unwanted interfacial reactions, particularly with underlying III-V semiconductor heterostructures.

Here, we use cryogenic deposition to intentionally suppress atomic mobility and separate deposition from crystallization in ultrathin Nb, Ta, Re, and V films on III-V heterostructures. *In situ* reflection high-energy electron diffraction (RHEED) patterns show that Ta (Fig. 1(a)), Nb, and V films are initially highly disordered or amorphous and become polycrystalline upon heating to 250 °C (Fig. 1(b)). *Ex situ* Grazing-incidence X-ray diffraction (GIXRD) reveals that thinner Ta and Re films remain amorphous, whereas thicker films crystallize into α -Ta and hexagonal close-packed (hcp) Re (Fig. 1(c) and 1(d)). This thickness dependent behavior is confirmed for Re using scanning transmission electron microscopy (STEM) (Fig. 1(e) and 1(f)). In Ta/Nb bilayers, Ta crystallizes at a thickness of only 2 nm, indicating that adjacent metal layers can modify crystallization. Transport measurements reveal relationships between crystal structure and superconductivity. The α -Ta film shows a higher superconducting transition temperature (T_c) compared to amorphous Ta. In contrast, a 4 nm amorphous Re film has an enhanced T_c of 6.1 K, whereas 6 nm hcp Re has a T_c of 2.6 K. These results show how material choice, film thickness, and adjacent layers influence crystallization following cryogenic deposition, providing a pathway to integrate ultrathin refractory superconductors on III-V semiconductor heterostructures.

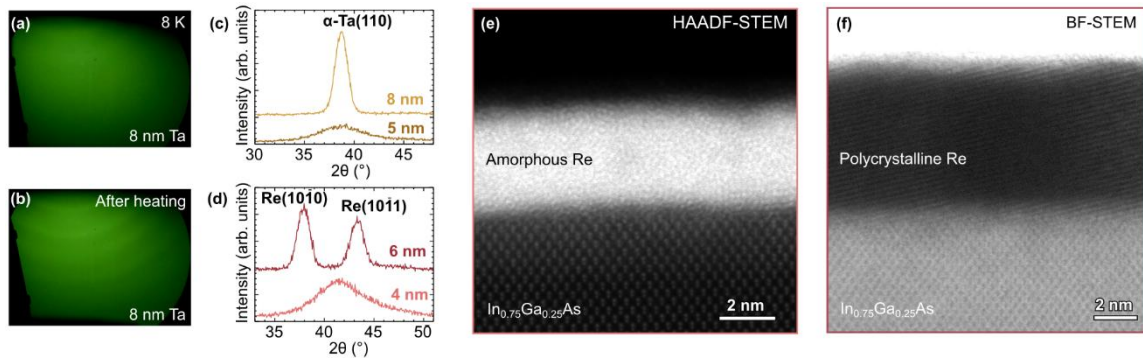


Fig. 1: Structural characterization of cryogenically deposited and crystallized refractories. (a) Reflection high-energy electron diffraction (RHEED) patterns of an 8 nm Ta film immediately after growth at 8 K and (b) after heating, showing polycrystalline diffraction rings. (c) Grazing-incidence X-ray diffraction (GIXRD) of 5 and 8 nm Ta and (d) 4 and 6 nm Re. The thinner films reveal broad diffraction feature consistent with amorphous structure. The thicker films show relative sharp diffraction peaks corresponding to α -Ta and hcp Re. (e) Cross-sectional scanning transmission electron microscopy (STEM) image of 3 nm Re showing an amorphous structure. (f) STEM image of 6 nm Re revealing a crystalline grain extending over the entire layer.

⁺ Author for correspondence: teunvanschijndel@ucsb.edu

Supplementary information:

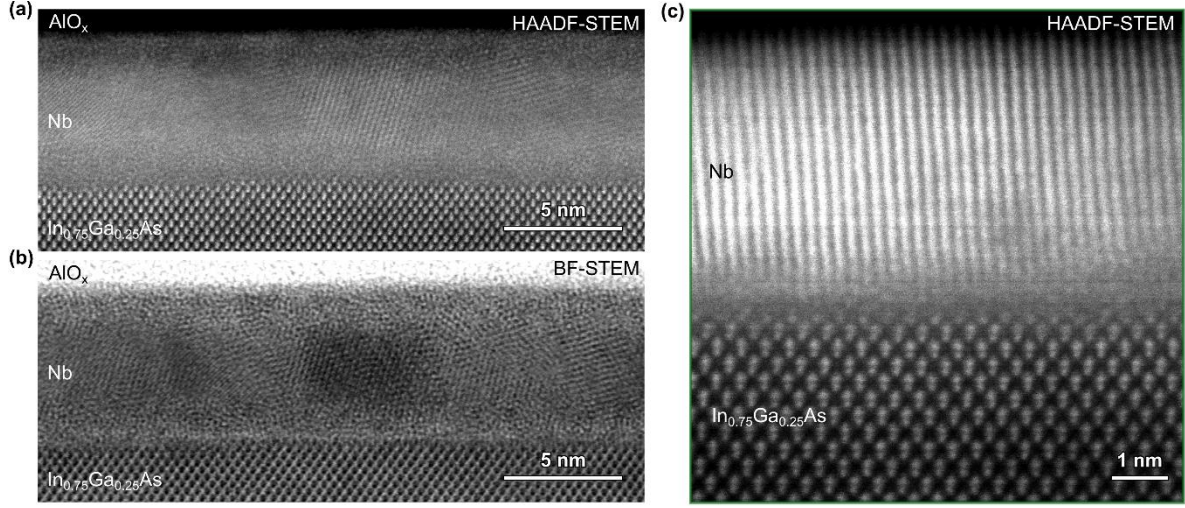


Fig. 2: Cross-sectional STEM of Nb films on III-V heterostructures after heating. (a) High-angle annular dark-field (HAADF) STEM of a 5 nm Nb film on an InGaAs/InAs quantum well heterostructure showing multiple crystalline grains. (b) Corresponding bright-field (BF) STEM image providing enhanced orientation contrast between the Nb grains. (c) High-resolution HAADF-STEM of the Nb/InGaAs interface, showing a sub-nanometer region without clearly resolved lattice fringes.

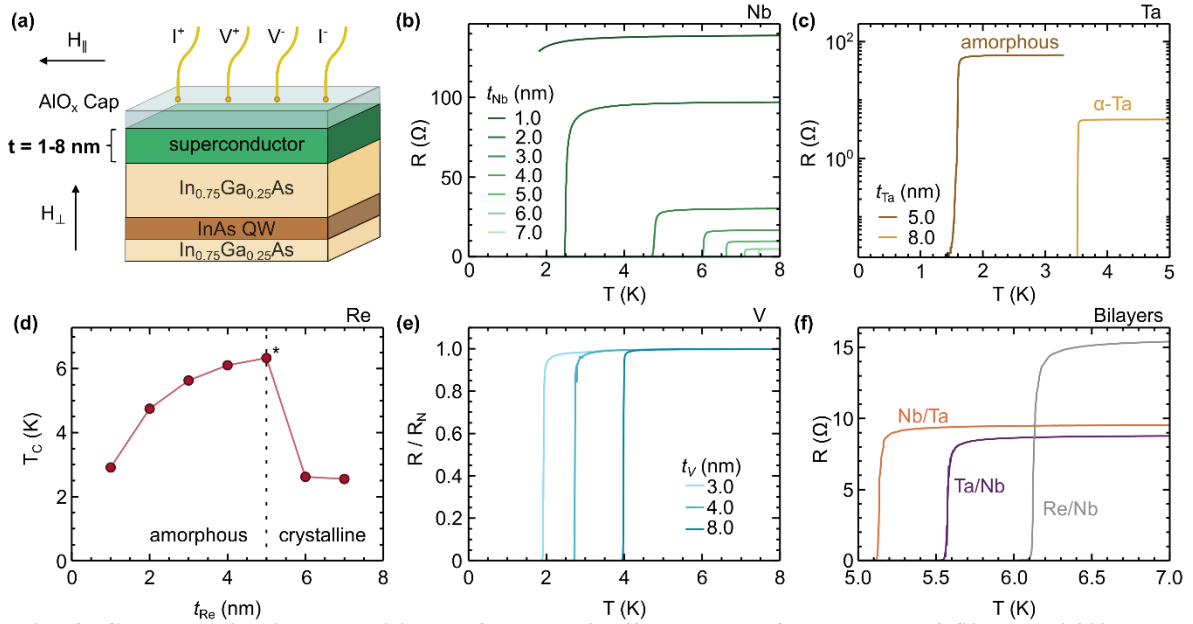


Fig. 3: Superconducting transitions of cryogenically grown refractory metal films and bilayers. (a) Schematic of the superconductor/ $\text{In}_{0.75}\text{Ga}_{0.25}\text{As}$ /InAs/ $\text{In}_{0.75}\text{Ga}_{0.25}\text{As}$ heterostructure and four-terminal transport geometry. (b) Temperature-dependent resistance, $R(T)$, of Nb films with thicknesses from 1 to 7 nm. (c) $R(T)$ of 5 and 8 nm Ta films plotted on a logarithmic scale. (d) Superconducting transition temperature as a function of Re thickness from 1 to 7 nm. The double transition of the 5 nm film is indicated by *. (e) Normalized $R(T)$ of 3, 4, and 8 nm V films. (f) $R(T)$ of 3 nm Nb/2 nm Ta, 2 nm Ta/3 nm Nb, and 2 nm Re/3 nm Nb bilayers.