



JOURNAL OF
APPLIED
CRYSTALLOGRAPHY

Volume 52 (2019)

Supporting information for article:

Significant texture improvement in single-crystalline-like materials on low-cost, flexible metal foils through growth of silver thin films

Yongkuan Li, Sicong Sun, Ying Gao, Yao Yao, Eduard Galstyan, Pratap Rudra, Monika Rathi, Pavel Dutta, Sara Pouladi, Jae Hyun Ryou and Venkat Selvamanickam

S1. Growth parameters of the Ge films

Samples *e* and *f* were fabricated by growing Ge films on the IBAD TiN substrate and on sample *a*, respectively, in the same run. Conductive buffers of NiSi₂/TiN were grown before the growth of Ge for purpose of epitaxy. The thicknesses of the NiSi₂ and TiN buffers are estimated to be ~15 and ~15 nm, respectively. The IBAD TiN substrate and sample *a* were ultrasonically cleaned in an isopropyl alcohol (IPA) bath and then blown dry with nitrogen (N₂). Films of TiN, NiSi₂, and Ge were grown consecutively on the substrates using the co-sputter tool. The targets were sputtered using ultra-high-purity gases, including Ar (99.999%), forming gas (Ar mixed with 4 vol% H₂, 99.999%), and N₂ (99.999%). The target-substrate distance was set as 10 cm, and the substrate holder was rotated at ~20 rpm to obtain uniform films. All the other process parameters are listed in Table S1.

Table S1 Process parameters of the Ge films and the buffers sequentially grown on the IBAD TiN substrate and sample *a*.

Film grown	Target, purity	Substrate temperature (°C)	Sputtering power (W)	Gas, flow (sccm)	Working pressure (mTorr)	Growth time (minutes)
TiN	Ti, 99.995%	700	250	Ar, 20 N ₂ , 2	1.75	3
NiSi ₂	NiSi ₂ , 99.9%	550	300	Ar+H ₂ , 34.5	4.20	1.5
LT ⁱ Ge	n-type Ge, 99.999%	525	250	Ar+H ₂ , 34.5	4.20	7.5
HT ⁱⁱ Ge	n-type Ge, 99.999%	700	250	Ar+H ₂ , 34.5	4.20	60

^{i, ii} The Ge films grown at 525°C and 700°C are denoted as low-temperature (LT) Ge and high-temperature (HT) Ge, respectively.

S2. Growth parameters of the Ge films

Cross-sectional Transmission Electron Microscope (TEM) images were obtained with a JEOL 2000FX TEM. Cross section TEM samples were prepared with an FEI 235 focused ion beam (FIB) milling system. The microstructure of sample *f* is shown by the cross-sectional TEM images in Figure S1. The film thickness of the Ge film is ~1.5 μm. Epitaxial growth of the Ag and Ge films in sample *f* is confirmed by the single-crystalline SAED patterns shown in Figure S1 (c) and inset of (a), respectively. In Figure S1 (c), the similar SAED patterns of the Ag and TiN films confirm the axis-by-axis epitaxy relationship of Ag on TiN, i.e. (001)⟨100⟩Ag || (001)⟨100⟩TiN. Distinct groups of spots are identified for Ag and TiN due to the ~3.7% lattice mismatch. A clear and linear interface between Ag and the materials on top is identified in Figure S1 (b), which confirms the flat and smooth surface shown in Figure 5 (e). Besides, the Ag film is found to be dense and uniform in Figure S1 (b), indicating that the pin-holes identified in Figure 5 (c) are filled in the ~330-nm-thick Ag film.

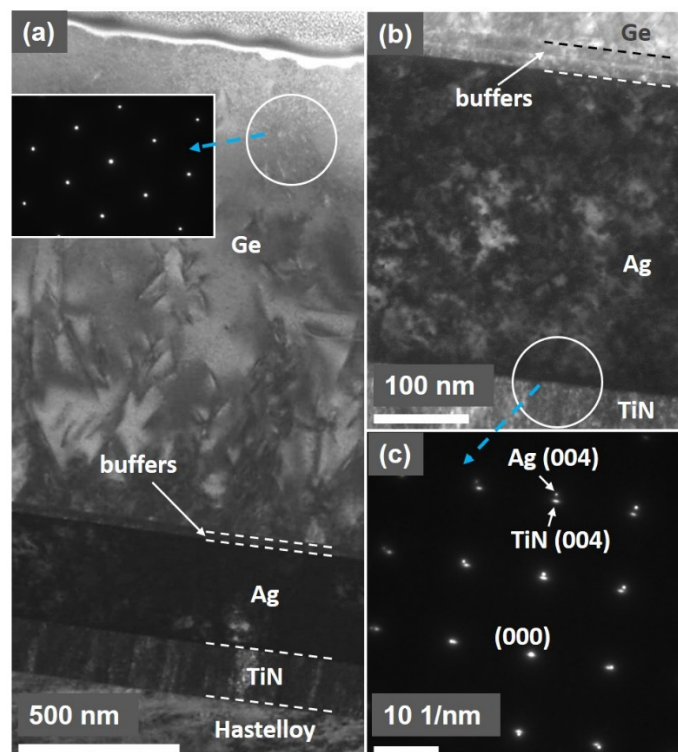


Figure S1 Cross-sectional TEM images of (a) sample *f*, (b) the Ag film in sample *f*. Inset of (a) shows the SAED pattern of the marked area of Ge. (c) SAED patterns of TiN and Ag in the marked area in (b). Sample *f* is the Ge film grown on the ~330-nm-thick Ag film on the IBAD TiN substrate.

S3. Growth parameters and texture data of the TiN film grown on sample *a*

The TiN film grown on sample *a* is used to show benefit of texture improvement by Ag. The process parameters of the TiN film are shown in Table S2. The thickness of the TiN film on the Ag film is estimated to be ~160 nm.

Table S2 Process parameters of the TiN/Ag/TiN sample on the IBAD TiN substrate.

Film grown	Target, purity	Temperature (°C)	Sputtering power (W)	Gas, flow (sccm)	Working pressure (mTorr)	Growth time (minutes)
TiN	Ti, 99.995%	700	250	Ar, 20 N ₂ , 2	1.75	30

Epitaxial growth of the TiN film on sample *a* is demonstrated by the sharp TiN (002) peak in the θ -2 θ scan in Figure S2 (a) and the four-fold symmetry in the TiN {220} pole figure in Figure S2 (b). The $\Delta\omega$ and $\Delta\phi$ of the TiN film grown on sample *a* are accordingly calculated to be $\sim 0.98 \pm 0.02^\circ$ and $\sim 2.08 \pm 0.09^\circ$, respectively, using the methods described in the Experimental Section. These texture spreads are significantly reduced compared to those of the IBAD TiN substrate.

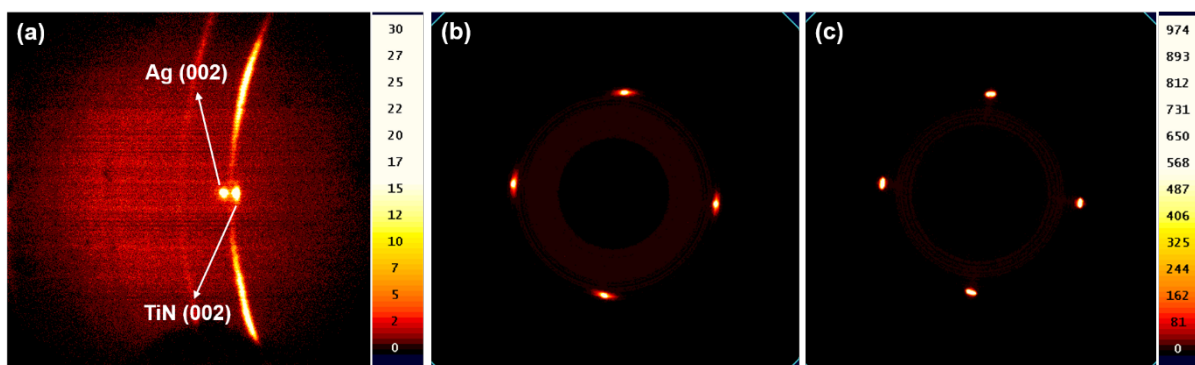


Figure S2 (a) XRD θ - 2θ scan of the TiN film grown on sample *a*. (b) TiN{220} pole figure of the TiN film grown on sample *a*. (c) Ag {220} pole figure of sample *a* displayed as a reference.

IUCr body text

S4. Epitaxial growth of all the Ag films in samples *b*, *c* and *d*

Samples *a*, *b*, *c* and *d* with a varying thickness of the Ag film are used to study how the texture improvement is achieved by Ag. The growth time was controlled to achieve Ag films with thicknesses of around 330, 220, 110, and 22 nm for samples *a*, *b*, *c* and *d*, respectively. The XRD θ - 2θ scan and pole figures conducted on sample *a* shown in Figure 2 (a) and Figure 3 (a) and (b) suggest the epitaxial growth of the ~330-nm-thick Ag film on the IBAD TiN substrate. Similar results of samples *b*, *c* and *d* are presented in Figure S3 which show the epitaxial growth of all the Ag films on the IBAD TiN substrates, including the Ag layer in sample *d* which is composed of discrete Ag grains.

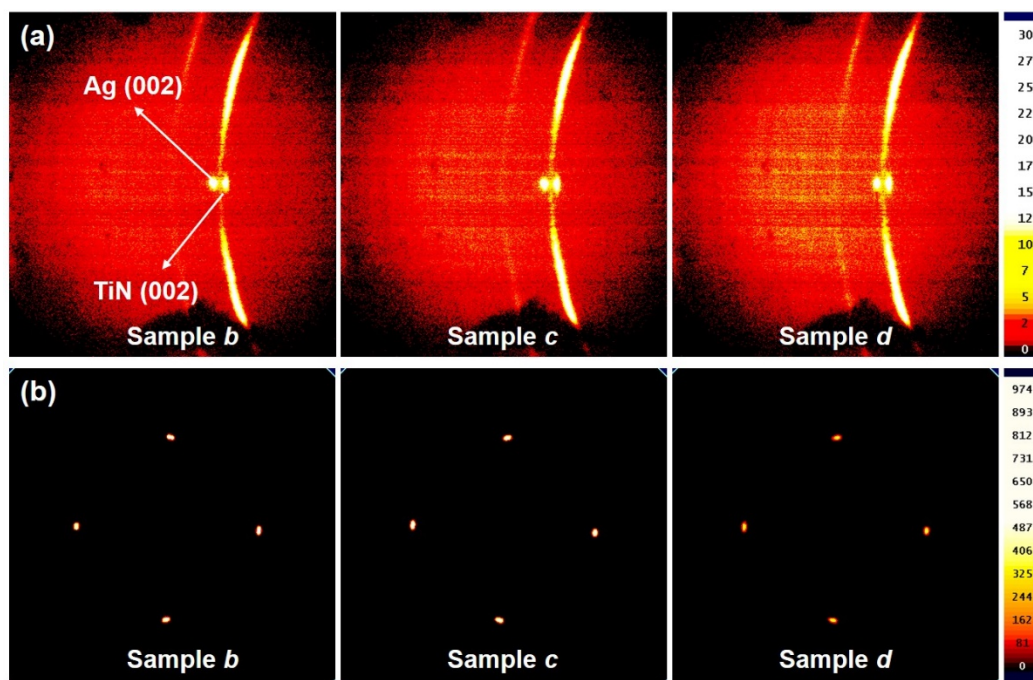


Figure S3 (a) XRD θ - 2θ scan of samples *b*, *c* and *d*. (b) Ag {220} pole figures of samples *b*, *c* and *d*. Samples *b*, *c* and *d* are Ag films with thicknesses of 220, 110, and 22 nm, respectively, grown on the IBAD TiN substrates.