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Fabrication of wafer-scale grain boundary free Cu single crystal film and copper oxide film by sputtering method and its application

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Copper thin films have been widely used as electrodes and interconnection wires in integrated electronic circuits, and more recently as substrates for the synthesis of graphene. However, the ultra-high vacuum processes required for high-quality Cu film fabrication, such as molecular beam epitaxy (MBE), restricts mass production with low cost. In this work, we demonstrated high-quality Cu thin films using a single-crystal Cu target and sputtering technique; the resulting film quality was comparable to that produced using MBE, even under unfavorable conditions for pure Cu film growth. The Cu thin film was epitaxially grown on an Al_2O_3 (0001) substrate, and had high crystalline orientation along the (111) direction. Despite the 10^{-3} Pa vacuum conditions, the resulting thin film was oxygen free due to the high chemical stability of the sputtered specimen from a single-crystal target; moreover, the deposited film had > 5 X higher adhesion force than that produced using a polycrystalline target. We applied the technique fabricating the single crystal thin film to the flexible transparent conducting electrodes, where a micromesh/nanomesh structure was fabricated on a polyimide substrate using UV lithography and wet etching. We also succeeded to fabricate a wafer-scale graphene in the formation of artificial single crystalline AB-BLG via aligned transfer of two single-crystalline monolayers. Such single crystal copper film was realized not only on the sapphire substrate, but also on the PI, PET, and PC, which enabled to apply in the region of flexible devices, metamaterial and of surface plasmonics. The important advantage of this single crystal copper film is due to the natural oxide layer with a thickness of around 2nm, which protects further oxidation, so that the copper layer maintains clean even for 2-3 years without any capping layer. Well-defined conditions enabled the copper film to convert to Cu_2O or CuO partially or totally, which are transparent and p-type semiconductor. In the partially oxidized case, the copper layer left beneath Cu_2O or CuO could be used as electrode. The color of the film varies between transparent gold and opaque metallic scarlet depending on the ratio of metal and oxides. Thanks to the grain free copper thin film, the copper oxides also maintain high crystallinity.

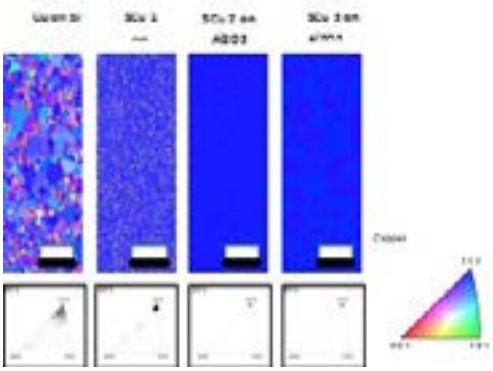

Figure 1: The EBSD images of commercial copper film (Cu on Si), single crystal copper film under unoptimized conditions SCu_1 and SCu_2) and single crystal copper film under optimized condition (SCu_3)

Recent Publications

- Nguyen VL, Perello DJ, Lee S, Nai CT, Shin BG, Kim JG, Park HY, Jeong HY, Zhao J, Vu QA, Lee SH, Loh KP, Jeong SY and Lee YH (2016) Wafer-scale single-crystalline AB-stacked bilayer graphtene. Advanced Materials, 28: 8177-8183.
- Lee S, Wi HS, Jo W, Cho YC, Lee HH, Jeong SY, Kim YI, Lee GW (2016) Multiple pathways of crystal nucleation in an extremely supersaturated aqueous potassium dihydrogen phosphate (KDP) solution droplet, PNAS 113: 13618-13623.
- Kim WK, Lee S, Lee DH, Park IH, Bae JS, Lee TW, Kim JY, Park JH, Cho YC, Cho CR, Jeong SY (2015) Cu Mesh for Flexible Transparent Conductive Electrodes. Sci. Rep. 5: 10715-10722
- Kim JY, Oh MW, Lee S, Cho YC, Yoon JH, Lee GW, Cho CR, Park CH and Jeong SY (2014) Abnormal Drop in Electrical Resistivity after Impurity Doping of Single Crystal Ag. Sci. Rep. 4: 05450-05454
- Phamcong D, Choi JH, Yun J. Bandarenka A. Kim J. Braun P. Jeong SY, Cho CR (2017) Synergistically Enhanced Electrochemical Performance of Hierarchical MoS2/TiNb2O7 Hetero-Nanostructures as Anode Materials for Li-Ion Batteries. ACS Nano 11: 1026-103.

Biography

Se-Young Jeong has his expertise in crystal growth and the investigation of physical properties such as structure, electricity and magnetism. He has been in charge of the crystal bank of a research institute for 20 years, which grow single crystals and supply the samples to the researchers. He has grown more than 100 kinds of single crystals including MgB2, GaN single crystals, and also contributed to develop ZnCoO DMS having room temperature ferromagnetism. Recently, he is studying on metal single crystal thin film. Especially, his recent study attracts lot of attention from the community, because recently developed copper films in his group by the modified sputtering system shows almost perfect crystallinity without grain boundaries even in wafer-scale, which has been required as the substrate for graphene growth and has lots of application possibilities.

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