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Effect of dopant gas pressure on the growth of magnetron sputtered CuO thin films for electrical and optical applications

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ABSTRACT

Tunability offers in the optoelectronic properties of TCOs open up the way to advanced applications in the field of photovoltaics. However, the conductivity of p-type TCOs remains an unresolved problem. In this work, an advantage of RF magnetron sputtering is employed to obtain polycrystalline CuO film at room temperature with better conductivity and optimum bandgap by controlling nitrogen gas pressure. Doping of N_2 gas pressure has a significant influence on the morphology of CuO films, resulting in improved electrical and optical properties of films at room temperature. The variation of structural characteristics with respect to N_2 gas pressure is discussed in detail using XRD, AFM, SEM, and EDX measurements, and these results confirm nitrogen's impact in improving the property of the fabricated film. The results show that the crystalline size, as well as the crystalline quality of the film, was found to increase with the increase in N_2 gas pressure at room temperature. Atomic force microscope (AFM) shows that all the deposited films are well adherent and grains are uniformly distributed over the surface of the substrate. The simultaneous effect of doping and grain boundary migration of nitrogen atom into CuO lattice increases crystallinity and the mobility of charge carriers at room temperature. This experimental strategy made CuO a good p-type material with mobility of $2.17 \times 10^3 cm^2/Vs$ and bandgap of 2.44eV at N_2 gas pressure of 2×10^{-2}mbar and resistivity is decreased due to high mobility obtained by tuning N_2 gas pressure. These results suggest that the electrical and optical properties of CuO thin films can be modulated by optimizing the carrier concentration and thus mobility by controlling the N_2 gas pressure.

1. Introduction

The research interest in the transition conducting oxides (TCOs) has continued owing to its wide variety of applications especially in the field of photovoltaics [1,2]. Researchers are very keen on exploiting the properties of oxides to enhance the efficiency of photovoltaic devices. For this fabrication of highly stable p and n-type oxide material is essential. To achieve high efficiency for PV cells, the material should possess good electrical conductivity and bandgap. But the simultaneous occurrence of these properties in the same material is incompatible. The majority of the TCOs in use and under research are n-type owing to their ease in creating lattice defects such as oxygen vacancies, proton interstitials, and their properties can be further tailored by the process of substitutional doping [3]. When compared to n-type TCOs, p-type TCOs are limited due to their lower conductivity [4,5]. In p-type TCOs their majority carriers are positively charged holes, due to their large effective mass they exhibit lower carrier density and mobility, also the difficulty associated with p-type doping limits their carrier concentration which also lowers its conductivity. So the growth of high-quality p-type material is essential for high efficient p-n junction devices. In p-type TCOs it is very difficult to obtain good mobility due to their lower carrier concentration. Carrier mobility is one of the major factors which influence the conductivity of thin films that depend upon defects formed at the crystal lattice due to doping. Scattering of charge carriers results in decreasing mobility if carrier concentration increases beyond a certain limit. Hence an optimization between carrier concentration and mobility is essential to achieve higher conductivity.

Cupric oxide (CuO) thin films have sparked a great deal of interest among transition metal oxides and several studies on copper oxides have been published in recent years, owing to their unique electronic, mechanical, magnetic, and optical properties, which makes them useful semiconductor materials for device applications including solar cells, lithium-ion batteries, gas sensing, supercapacitors, diodes applications, etc. [6–15]. The conductivity of thin films can be increased either by increasing the carrier concentration or the mobility of charge carriers. One of the efficient approaches to improve conductivity is by increasing

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.optmat.2022.112031.

References

[1] J.S. Jang, J. Kim, U. Ghorpade, H.H. Shin, M.G. Gang, S.D. Park, H.J. Kim, D.S. Lee, J.H. Kim, Comparison study of ZnO-based quaternary TCO materials for photovoltaic application, J. Alloys Compd. 793 (2019) 499–504, https://doi.org/10.1016/j.jallcom.2019.04.042.

[2] P. Aazadfar, E. Solati, D. Dorranian, Properties of Au/Copper oxide nanocomposite prepared by green laser irradiation of the mixture of individual suspensions, Opt. Mater. 78 (2018) 388–395, https://doi.org/10.1016/j.optmat.2018.02.050.

[3] S.C. Dixon, D.O. Scanlon, C.J. Carmalt, I.P. Parkin, N-Type doped transparent conducting binary oxides: an overview, J. Mater. Chem. C. 4 (2016) 6946–6961, https://doi.org/10.1039/c6tc01881e.

[4] G. Brunin, F. Ricci, V.A. Ha, G.M. Rignanese, G. Hautier, Transparent conducting materials discovery using high-throughput computing, Npj Comput. Mater. 5 (2019) 1–13, https://doi.org/10.1038/s41524-019-0200-5.

[5] K.H.L. Zhang, K. Xi, M.G. Blamire, R.G. Egdell, P-type transparent conducting oxides, J. Phys. Condens. Matter 28 (2016) 383002, https://doi.org/10.1088/0953-8984/28/38/383002.

[6] P. Chand, A. Gaur, A. Kumar, U. Kumar Gaur, Structural and optical study of Li doped CuO thin films on Si (1 0 0) substrate deposited by pulsed laser deposition, Appl. Surf. Sci. 307 (2014) 280–286, https://doi.org/10.1016/j.apsusc.2014.04.027.

[7] P. Samarasekara, N.T.R.N. Kumara, N.U.S. Yapa, Sputtered copper oxide (CuO) thin films for gas sensor devices, J. Phys. Condens. Matter 18 (2006) 2417–2420, https://doi.org/10.1088/0953-8984/18/8/007.

[8] H. Absike, Z. Essalhi, H. Labrim, B. Hartiti, N. Baaalla, M. Tahiri, B. Jaber, H. Ez-zahraouy, Synthesis of CuO thin films based on Taguchi design for solar absorber, Opt. Mater. 118 (2021) 111224, https://doi.org/10.1016/j.optmat.2021.111224.

[9] M. Dahrul, H. Alatas, Irzaman, preparation and optical properties study of CuO thin film as applied solar cell on LAPAN-IPB satellite, Procedia Environ. Sci. 33 (2016) 661–667, https://doi.org/10.1016/j.proenv.2016.03.121.

[10] T. Ishihara, M. Higuchi, T. Takagi, M. Ito, H. Nishiguchi, Y. Takita, Preparation of CuO thin films on porous BaTiO3 by self-assembled multibilayer film formation and application as a CO2 sensor, J. Mater. Chem. 8 (1998) 2037–2042, https://doi.org/10.1039/a801595c.

[11] J. Morales, L. Sánchez, F. Martín, J.R. Ramos-Barrado, M. Sánchez, Use of low-temperature nanostructured CuO thin films deposited by spray-pyrolysis in lithium cells, Thin Solid Films 474 (2005) 133–140, https://doi.org/10.1016/j.tsf.2004.08.071.

[12] S.S. Kumar Jacob, I. Kulandaisamy, I.L. Poul Raj, A.A. Abdeltawab, S. Z. Mohammady, M. Ubaidullah, Improved optoelectronic properties of spray pyrolysis coated Zn doped Cu2O thin films for photodetector applications, Opt. Mater. 116 (2021) 111086, https://doi.org/10.1016/j.optmat.2021.111086.

[13] A.S. Patil, M.D. Patil, G.M. Lohar, S.T. Jadhav, V.J. Fulari, Supercapacitive properties of CuO thin films using modified SILAR method, Ionics 23 (2017) 1259–1266, https://doi.org/10.1007/s11581-016-1921-9.

[14] P. Venkateswari, P. Thirunavukkarasu, M. Ramamurthy, M. Balaji, J. Chandrasekaran, Optimization and characterization of CuO thin films for P–N junction diode application by JNSP technique, Optik 140 (2017) 476–484, https://doi.org/10.1016/j.ijleo.2017.04.039.

[15] A. Tombak, M. Benhaliliba, Y.S. Ocak, T. Kiliçoglu, The novel transparent sputtered p-type CuO thin films and Ag/p-CuO/n-Si Schottky diode applications, Results Phys. 5 (2015) 314–321, https://doi.org/10.1016/j.rinp.2015.11.001.

[16] A.A. Al-Ghamdi, M.H. Khedr, M. Shahnawaze Ansari, P.M.Z. Hasan, M.S. Abdel-Wahab, A.A. Farghali, RF sputtered CuO thin films: structural, optical and photo-catalytic behavior, Phys. E Low-dimens. Syst. Nanostruct. 81 (2016) 83–90, https://doi.org/10.1016/j.physe.2016.03.004.

[17] A. Saldaña-Ramírez, M.R.A. Cruz, I. Juárez-Ramírez, L.M. Torres-Martínez, Influence of the power density and working pressure in the magnetron co-sputtering deposition of ZnO–SnO2 thin films and their effect in photocatalytic hydrogen production, Opt. Mater. 110 (2020), https://doi.org/10.1016/j.optmat.2020.110501.

[18] H. Mahdhi, Z. Ben Ayadi, J.L. Gauffier, K. Djessas, S. Alaya, Effect of sputtering power on the electrical and optical properties of Ca-doped ZnO thin films sputtered from nanopowders compacted target, Opt. Mater. 45 (2015) 97–103, https://doi.org/10.1016/j.optmat.2015.03.015.

[19] M.K. Shabbir, S. Bashir, Q.S. Ahmed, N. Yaseen, S.A. Jalil, M. Akram, K. Mahmood, A. Khalid, Effect of substrate temperature on the growth of copper oxide thin films deposited by a pulsed laser deposition technique, Surf. Rev. Lett. 25 (2018) 1–14, https://doi.org/10.1142/S0218625x18500531.

[20] S. Dolai, R. Dey, S. Das, S. Hussain, R. Bhar, A.K. Pal, Cupric oxide (CuO) thin films prepared by reactive d.c. magnetron sputtering technique for photovoltaic application, J. Alloys Compd. 724 (2017) 456–464, https://doi.org/10.1016/j.jallcom.2017.07.061.

[21] S. Cho, Optical and electrical properties of CuO thin films deposited at several growth temperatures by reactive RF magnetron sputtering, Met. Mater. Int. 19 (2013) 1327–1331, https://doi.org/10.1007/s12540-013-6030-y.

[22] G. Shugar, J. Ballinger, Chemical Technicians' Ready Reference Handbook, Tata McGraw Hill, New York, 1996.

[23] V.K. Arepalli, Y. Shin, J. Kim, Photovoltaic behavior of the room temperature grown RF-Sputtered SnS thin films, Opt. Mater. 88 (2019) 594–600, https://doi.org/10.1016/j.optmat.2018.12.016.

[24] R.A. Nachiar, S. Muthukumaran, Structural, photoluminescence and magnetic properties of Cu-doped SnO2 nanoparticles co-doped with Co, Opt Laser. Technol. 112 (2019) 458–466, https://doi.org/10.1016/j.optlastec.2018.11.055.

[25] M. Shaheera, K.G. Girija, M. Kaur, V. Geetha, A.K. Debnath, R.K. Vatsa, K.P. Muthe, S.C. Gadkari, Characterization and device application of indium doped ZnO homojunction prepared by RF magnetron sputtering, Opt. Mater. 101 (2020) 109723, https://doi.org/10.1016/j.optmat.2020.109723.

[26] K. Matsushima, K. Koga, I. Suhariadi, D. Yamashita, N. Itagaki, K. Kuwahara, G. Uchida, K. Oshikawa, M. Shiratani, Effects of nitrogen on crystal growth of sputter-deposited ZnO films for transparent conducting oxide, Jpn. J. Appl. Phys. 52 (2013) 11NB03, https://doi.org/10.7567/jjap.52.11nb03.

[27] G. Marinov, V. Strijkova, M. Vasileva, V. Madjarova, N. Malinowski, T. Babeva, Effect of substrate temperature on the microstructural, morphological, and optical properties of electrosprayed ZnO thin films, Adv. Condens. Matter Phys. 2018 (2018), 8957507, https://doi.org/10.1155/2018/8957507.

[28] A.A. Khadayeir, E.S. Hassan, T.H. Mubarak, S.S. Chiad, N.F. Habubi, M.O. Dawood, I.A. Al-Baidhany, The effect of substrate temperature on the physical properties of copper oxide films, J. Phys. Conf. Ser. 1294 (2019), https://doi.org/10.1088/1742-6596/1294/2/022009.

[29] R. Kumari, A. Sahai, N. Goswami, Effect of nitrogen doping on structural and optical properties of ZnO nanoparticles, Prog. Nat. Sci. Mater. Int. 25 (2015) 300–309, https://doi.org/10.1016/j.pnsc.2015.08.003.

[30] G.C.A.M. Janssen, Stress and strain in polycrystalline thin films, Thin Solid Films 515 (2007) 6654–6664, https://doi.org/10.1016/j.tsf.2007.03.007.

[31] N.H. Hashim, S. Subramani, M. Devarajan, A.R. Ibrahim, Structural and surface characterization of undoped ZnO and Cu doped ZnO using sol–gel spin coating method, J. Mater. Sci. Mater. Electron. 27 (2016) 3520–3530, https://doi.org/10.1007/s10854-015-4187-5.

[32] Ş. Baturay, A. Tombak, D. Batibay, Y.S. Ocak, n-Type conductivity of CuO thin films by metal doping, Appl. Surf. Sci. 477 (2019) 91–95, https://doi.org/10.1016/j.apsusc.2017.12.004.

[33] H.Z. Asl, S.M. Rozati, Spray deposited nanostructured CuO thin films: influence of substrate temperature and annealing process, Mater. Res. 21 (2018) 1–8, https://doi.org/10.1590/1980-5373-MR-2017-0754.

[34] M. Dhaouadi, Physical properties of copper oxide thin films prepared by sol–gel spin–coating method, Am. J. Phys. Appl. 6 (2018) 43, https://doi.org/10.11648/j.ajpa.20180602.13.

[35] M. Aftab, M.Z. Butt, D. Ali, F. Bashir, T.M. Khan, Optical and electrical properties of NiO and Cu-doped NiO thin films synthesized by spray pyrolysis, Opt. Mater. 119 (2021) 111369, https://doi.org/10.1016/j.optmat.2021.111369.

[36] D. Sudha, S. Dhanapandian, C. Manoharan, A. Arunachalam, Structural, morphological and electrical properties of pulsed electrodeposited CdIn2Se4 thin films, Results Phys. 6 (2016) 599–605, https://doi.org/10.1016/j.rinp.2016.09.004.

[37] H. Sun, S.C. Chen, W.C. Peng, C.K. Wen, X. Wang, T.H. Chuang, The influence of oxygen flow ratio on the optoelectronic properties of p-type $Ni_{1-x}O$ films deposited by ion beam assisted sputtering, Coatings 8 (2018) 1–10, https://doi.org/10.3390/coatings8050168.

[38] S.H. Lee, S.J. Yun, J.W. Lim, The characteristics of Cu_2O thin films deposited using rf-magnetron sputtering method with nitrogen-ambient, ETRI J. 35 (2013) 1156–1159, https://doi.org/10.4218/etrij.13.0213.0216.

[39] A.A. Talukder, J. Pokharel, M. Shrestha, Q.H. Fan, Improving electrical properties of sol-gel derived zinc oxide thin films by plasma treatment, J. Appl. Phys. 120 (2016), https://doi.org/10.1063/1.4964941.

[40] P. Singh, A. Kumar, Deepak, D. Kaur, Growth and characterization of ZnO nanocrystalline thin films and nanopowder via low-cost ultrasonic spray pyrolysis, J. Cryst. Growth 306 (2007) 303–310, https://doi.org/10.1016/j.jcrysgro.2007.05.023.

[41] G. Kavei, A.M. Gheidari, The effects of surface roughness and nanostructure on the properties of indium tin oxide (ITO) designated for novel optoelectronic devices fabrication, J. Mater. Process. Technol. 208 (2008) 514–519, https://doi.org/10.1016/j.jmatprotec.2008.01.024.

[42] L.R. Nivedita, A. Haubert, A.K. Battu, C.V. Ramana, Correlation between crystal structure, surface/interface microstructure, and electrical properties of nanocrystalline niobium thin films, Nanomaterials 10 (2020) 1–24, https://doi.org/10.3390/nano10071287.

[43] S. Ishizuka, S. Kato, T. Maruyama, K. Akimoto, Nitrogen doping into Cu2O thin films deposited by reactive radio-frequency magnetron sputtering, Jpn. J. Appl. Physics 40 (2001) 2765–2768, https://doi.org/10.1143/jjap.40.2765. Part 1 Regul. Pap. Short Notes Rev. Pap.