ISSN 1996-3343

Asian Journal of Applied Sciences

Knowledgia
SCIENTIFIC
A Place to Publish Outstanding Research

http://knowledgiascientific.com

Asian Journal of Applied Sciences 7 (8): 804-808, 2014
ISSN 1996-3343 / DOI: 10.3923/ajaps.2014.804.808

Deposition and Optical Characterization of Superconducting Niobium Nitride Thin Films

[1]A.V. Thanikai Arasu, [1]G. Chinnamma, [1]R. Baskaran, [2]S. Tripura Sundari and [2]A.K. Tyagi
[1]Condensed Matter Physics Division,
[2]Surface and Nanoscience Division, Materials Science Group, IGCAR, Kalpakkam, 603102, India

Corresponding Author: A.V. Thanikai Arasu, Condensed Matter Physics Division, Materials Science Group, IGCAR, Kalpakkam, 603102, India

ABSTRACT

Niobium nitride (NbN) thin films deposited by DC reactive magnetron sputtering on oxidized silicon and glass substrates at room temperature were found to be superconducting in the range of ~11.4-12.3 K depending on the sputtering conditions. All the NbN thin films showed a negative temperature co-efficient of resistance above 25 K indicating the presence of disorder. The superconducting transition temperature (T_c), onset temperature (T_o) at which temperature dependence of resistance becomes positive and width of the superconducting transition (ΔT_c) did not show direct correlation to the resistivity measured at 300 K. The optical properties of the films were measured using a rotating polarizer type spectroscopic ellipsometer in the energy range of 1.5-5 eV under ambient conditions. Assuming the low frequency dependence of real part of the dielectric function is indicative of a metallic behavior, we have found that this metallicity is not correlated to all the three superconducting properties measured.

Key words: Niobium nitride, thin films, temperature

INTRODUCTION

Niobium nitride (NbN) is a superconductor with a cubic B1 structure and superconducting at a maximum superconducting transition temperature (Tc) of 17 K (Keskar *et al.*, 1971). NbN has a potential for several applications like Josephson junctions, superconducting single photon detectors, superconducting hot electron bolometers etc. The important properties of NbN thin films for which it is anticipated to be used are its chemically inert nature and its weak dependence of T_c on radiation damage. Most of these applications require deposition and characterization of NbN thin films. The high transition temperature NbN thin films are generally obtained by reactive sputtering in argon and nitrogen plasma by DC/RF sputtering on heated sapphire substrates. Thin films deposited at ambient temperature generally become superconducting below 16 K (Wang *et al.*, 1996). Deposition of NbN at room temperature is required for processing during sensor fabrication like SQIUDs etc. Similarly NbN deposited on oxidized silicon substrates or glass show reduced T_c due to disorder and interface strain. Disorder in NbN thin films are due to the non-stoichiometric formation of NbN_x with $x<1$. Disorder in NbN a thin film increases resistivity at room temperature and reduces the T_c of NbN thin films. The correlation between disorder and T_c has been published in literature (Chand *et al.*, 2012). The temperature dependence of resistance with temperature changes from positive for small disorder to negative for samples with medium disorder. The residual resistivity ratio measured between room temperature and the temperature at which resistance starts dropping due to superconducting fluctuations is a measure of the disorder in NbN thin films. In general, the superconducting transition width (ΔT_c) increases for thin films deposited under sub-optimum conditions. The increase in width (ΔT_c) could be attributed to both disorder (Gantmakher and Golubkov, 2001) and inhomogeneties. Optical measurement on superconducting thin films can be a tool to identify the formation of superconducting phase which can be done non-invasively and could also be used during the formation of thin film. Optical measurements have earlier been done on NbN thin films to extract various parameters of Drude model and plasma frequency and hence the electron carrier density (Semenov *et al.*, 2009). In this study we have tried to relate the effect of disorder on NbN thin films deposited under specific conditions, as measured by electrical resistivity of the thin filmsand the dielectric constants measured from ellipsometry.

DEPOSITION OF NIOBIUM NITRIDE THIN FILMS

Thin films of superconducting niobium nitride (NbN) have been deposited by reactive DC magnetron sputtering on oxidized silicon substrates and glass substrates in a high vacuum deposition chamber. Base pressures of 8×10^{-6} Pa was achieved using a 1000 L $\sec^{-1}$ Elettrorava turbomolecular pump. The flow of 99.9995% pure argon and nitrogen were controlled by mass flow controllers. During the sputtering process, the deposition pressure was controlled by a 1000 position gate valve fixed above the turbomolecular pump and measured accurately by a capacitance manometer. Four films labeled A, B, C and D were deposited using this process for characterization. Samples A, B and C were deposited on glass substrates at a total pressure of 0.266, 0.333 and 0.399 Pa, respectively. The flow rates of high purity argon and nitrogen were maintained at 33 SCCM and 7 SCCM, respectively, for all the three samples. The power of the DC source was maintained at 500 W during deposition. The rate of deposition marginally increased from 0.15-0.16 nm $\sec^{-1}$ as the pressure during deposition was changed from 0.266-0.399 Pa. Sample D was deposited on oxidized silicon substrate. In this deposition process, the flow rate of argon and nitrogen were maintained at 33 SCCM and 6 SCCM, respectively. The deposition pressure was 0.266 Pa and the power of the DC source was maintained at 600 W during deposition. The rate of deposition was 0.2 nm $\sec^{-1}$. In all the deposition, pre-sputtering of niobium was carried out in pure argon atmosphere with shutters closed for 30 min to remove surface impurities. The samples are rotated to get a uniform deposition over the substrate area. The samples are cooled by flowing water onto substrate holder at 20°C. All the samples were patterned lithographically by a lift-off process into linear resistivity pattern with separate contact pads for current injunction and voltage measurement. Quartz crystal thickness monitor was used to measure the deposited thickness of thin films. A total thickness of approximately 160 nm were deposited and the actual thickness was subsequently measured accurately using a surface profiler. The NbN thin films deposited by similar process earlier was characterized by Glancing Incident X-ray Diffraction measurements (GIXRD) for the formation of NbN thin films (Baskaran *et al.*, 2013).

ELECTRICAL CHARACTERIZATION

The samples were electrically characterized using the standard four probe geometry. The electrical resistance of the sample was measured by passing a constant current of 30 μA and measuring the developed voltage across the sample using a nano-voltmeter. The resistivity at the measuring temperature can be calculated based on the thickness of the thin film, as the length and width of the measuring sample is a known constant due to photolithographic pattern. The temperature dependence of resistivity of the sample is measured by attaching the sample to the copper sample holder in a dipstick and inserting the dipstick in a partially filled liquid helium dewar. The temperature fluctuations are reduced by surrounding the sample holder by a radiation shield. The temperature of the sample is measured using a calibrated silicon diode thermometer fixed close to the sample in the sample holder. Different temperatures were obtained by varying the level of the sample holder in the dipstick inside the liquid helium dewar. The voltage developed in the sample and diode voltage is stored in a computer using lab-view software for subsequent analysis.

The room temperature resistivity of the four samples A, B, C and D are 350.3, 1264.3, 735.3 and 242.9 μΩ-cm, respectively. All the four films were found to be superconducting with a superconducting transition temperature (T_c) ranging from 11.4-12.3 K. The variation of T_c with room temperature resistivity is shown in Fig. 1. All the films exhibited a negative temperature coefficient of resistance above 25 K indicating the presence of disorder in these films. The variation of the onset temperature (T_o) at which the temperature dependence of resistance becomes positive due to superconducting fluctuations below a certain temperature (T_o) with room temperature

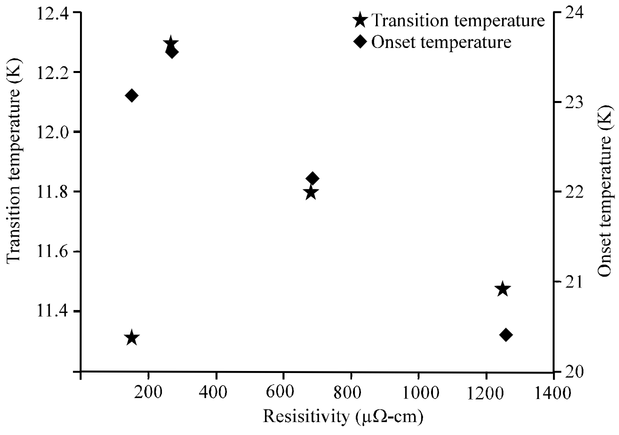

Fig. 1: Plot of transition temperature and the onset temperature variation with room temperature resistivity

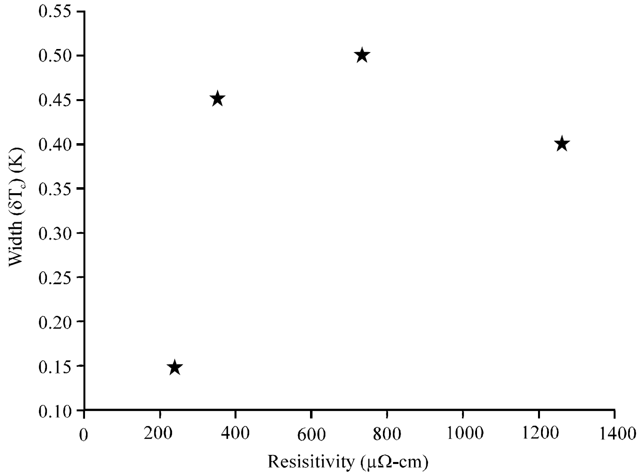

Fig. 2: Plot of variation of transition width with room temperature resistivity

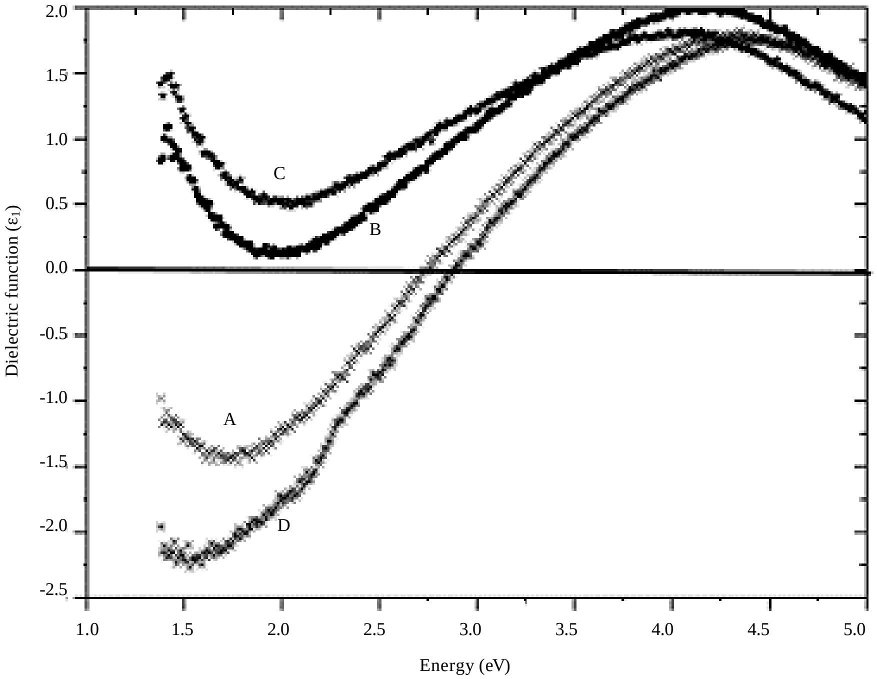

Fig. 3: Dependence of real part of the dielectric function (ε_1) on incident photon energy

resistivity is also shown in Fig. 1. The disorder present in these thin films changes the superconducting transition width and the variation of transition width ($\triangle T_c$) with room temperature resistivity is shown in Fig. 2. It may be noted that all the three parameters T_c, T_o and $\triangle T_c$ vary non-monotonically with room temperature resistivity.

OPTICAL CHARACTERIZATION

The optical characterization of the films was carried out using a rotating polarizer type spectroscopic ellipsometer (SOPRA ESVG) in the energy range 1.5-5 eV. The measurements were performed at an angle of incidence of 75° under ambient conditions. The real and imaginary parts of the dielectric functions (ε_1 and ε_2) were computed from the ellipsometric parameters using the relation (Sundari *et al.*, 2013):

$$\varepsilon(E) = N_o^2\left[\sin^2\varphi + \left[\frac{1-\rho}{1+\rho}\right]^2 \sin^2\varphi\tan^2\varphi\right] \tag{1}$$

where, N_o is the refractive index of the ambient, φ is the angle of incidence and ρ is the ratio of parallel to perpendicular components of the reflected light. Figure 3 shows the real part (ε_1) of the dielectric function calculated from the Eq. 1. It is observed that the magnitude of ε_1 is positive and negative in samples D and A, while it is only positive for samples B and C. The screened plasma frequencies are determined from the locations of frequencies where real part of the dielectric function goes to zero. The screened plasma frequency for samples D and A are 2.88 and 2.75 eV, respectively. As larger screened plasma frequency is indicative of a higher electron density, the electrical resistivity increases from D to A. Even though the samples B and C are superconducting and the resistivity is still in the range of 1 mΩ cm and below, the low frequency response on $\varepsilon_1(\omega)$ is still non-Drude like indicating presence of insulating phase. The intriguing part of the present analysis is that sample C though has lower resistivity, higher T_c and T_0 compared to sample B, the $\varepsilon_1(\omega)$ is shifted even higher than sample B. This shift is reflected in sharper transition ΔT_c for sample B compared to sample C. This implies that optical measurements alone cannot be an isolated tool to infer properties of mixed phase samples (e.g. B and C).

CONCLUSION

NbN thin films were deposited on oxidized silicon and glass substrates. The superconducting properties like transition temperature (T_c), onset temperature (T_o) and of transition width (ΔT_c) were measured using electrical characterization. The real and imaginary part of dielectric constants ε_1 and ε_2 were computed in the energy range 1.5-5 eV from the ellipsometric parameters, measured by spectroscopic ellipsometry for all the NbN thin films at room temperature. The dependence of superconducting parameters like T_c, T_o and ΔT_c show a non-monotonic behavior with room temperature resistance. The metallicity of these samples as depicted by the variation of ε_1 with energy at low frequency also show a non-monotonic variation with room temperature resistance.

REFERENCES

Baskaran, R., A.V. ThanikaiArasu, G. Chinnamma and M.P. Janawadkar, 2013. Pseudogap effects in disordered niobium nitride superconducting thin films. J. Applied Sci., 13: 330-332.

Chand, M., G. Saraswat, A. Kamlapure, M. Mondal and S. Kumar *et al.*, 2012. Phase diagram of the strongly disordered s-wave superconductor NbN close to the metal-insulator transition. Phys. Rev. B, Vol. 85.

Gantmakher, V.F. and M.V. Golubkov, 2001. Width of the zero-field superconducting resistive transition in the vicinity of the localization threshold. J. Exp. Theor. Phys. Lett., 73: 131-134.

Keskar, K.S., T. Yamashita and Y. Onodera, 1971. Superconducting transition temperatures of RF sputtered NbN films. Jap. J. Applied Phys., 10: 370-374.

Semenov, A., B. Gunther, U. Bottger, H.W. Hubers and H. Bartolf *et al.*, 2009. Optical and transport properties of ultrathin NbN films and nanostructures. Phys. Rev. B, Vol. 80. 10.1103/PhysRevB.80.054510

Sundari, S.T., K. Srinivasu, S. Dash and A.K. Tyagi, 2013. Temperature evolution of optical constants and their tuning in silver. Solid State Commun., 167: 36-39.

Wang, Z., A. Kawakami, Y. Uzawa and B. Komiyama, 1996. Superconducting properties and crystal structures of single-crystal niobium nitride thin films deposited at ambient substrate temperature. J. Applied Phys., 79: 7837-7842.