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RESEARCH ARTICLE

INVESTIGATION OF THE SPUTTERING CONDITIONS ON THE DEPOSITION OF THE NbN THIN FILMS ONTO GLASS AND Si/SiO SUBSTRATES

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ABSTRACT

NbN thin films are very useful superconducting materials for the electronic devices. Using sputter deposition technique is a cheap alternative to produce of NbN thin films on glass or SiO for the technology applications. The NbN films was coated on glass and SiO/Si substrates by DC magnetron sputtering technique. Pure Nb target was used the production of NbN thin films in different compositions of Argon and nitrogen gases. The effects of different substrate temperatures, sputtering power and, various ratios of Nitrogen/Argon gases were investigated on Superconducting Critical Temperature (Tc). It is seen that increasing the substrate temperature or sputtering power resulted in increased on the Tc. Thereby optimum condition values are deduced by the Residual Resistance Ratio (RRR) calculation and resistance-temperature (R-T) curves. It is observed that the substrate temperature and the flux of nitrogen gas effect (111) preferential orientation during the deposition, whereas the critical temperature of the thin films is affected by the of deposition chamber vacuum. Tc is also highly dependent on the Nitrogen and Argon pressures on sputtering deposited NbN films.

Keywords: *Transition Temperature, Niobium Nitride (NbN), Thin Films, Residual-Resistance Ratio, Superconductivity.*

Investigation of The Sputtering Conditions On The Deposition of The NbN Thin Films Onto Glass and Si/SiO Substrates

CAM VE Si/SiO ALTTAŞ ÜZERİNE KAPLANAN NbN İNCE FİLMLERİN KAPLAMA KOŞULLARININ ARAŞTIRILMASI

ÖZ

NbN ince filmler elektronik cihazlar için çok kullanışlı süper iletken malzemelerdir. Püskürtme biriktirme tekniğini kullanması teknoloji uygulamaları için cam veya SiO üzerinde NbN ince filmlerin üretilmesi için ucuz bir alternatiftir. . Bu çalışmada, NbN ince filmler, reaktif magnetron püskürtme yöntemi ile cam, Si ve SiO alt tabakaları üzerine kaplanmıştır. Saf Nb metali, Argon ve azot gazlarının farklı karşım oranlarında NbN ince filmlerinin üretimi için kullanılmıştır. Çeşitli alttaş sıcaklıkları, püskürtme gücü ve Argon / Azot gazı oranının süperiletken geçiş sıcaklığı (T_c) üzerine etkileri araştırılmıştır. Alttaş sıcaklığının veya püskürtme gücünün arttırılmasının T_c'yi de arttığı görülmüştür. Bunun sonunda optimum sıcaklık değerleri, direnç-sıcaklık (R-T) eğrileri ve Artık Direnç Oranı (RRR) hesaplaması yardımı ile bulunmuştur. Kaplama sırasındaki alttaş sıcaklığının ve azot gazı akış değerlerinin tercih edilen yönlendirmeyi güçlü bir şekilde etkilediği gözlenirken, ince filmlerin kritik sıcaklığı orijinal vakumun kalitesinden etkilenmektedir. Ayrıca T_c üretilen NbN filmlerinin kalitesi kaplama işlemi süresindeki Azot ve Argon basınçlarına da oldukça bağımlıdır.

Anahtar Kelimeler: *Kritik Sıcaklık, NbN, İnce Film, Kalıntı Direnç Oranı, Süperiletkenlik.*

1. INTRODUCTION

The nitrite thin film coating is used in the field of different technologies. However, NbN thin films produced by sputtering on Si or SiO2 substrate are more attractive among other studies due to their low cost and easy accessibility for the production of high-density integrated circuits. NbN thin films can produced by reactive DC magnetron sputtering [1-4], ion beam deposition [5, 6], pulse laser deposition [7, 8] metal organic chemical deposition (MOCVD) [9]. In order to obtain NbN thin films with good superconducting properties, it is necessary to define the appropriate value production conditions well, which is possible with detailed studies on the effects of conditions in the deposition process.

Previous works [10], on the development of superconductivity properties of NbN thin films, proved that the partial nitrogen pressure increase in the Argon-Nitrogen (Ar / N_2) gas mixture provides an increase in the N/Nb ratio in NbN thin films. Competition among researchers is still ongoing to find the effective mechanism that primarily suppresses superconducting critical temperature (T_c) and to increase in the T_c of NbN films. The effects of parameters such as Ar and N_2 pressures, film deposition time, and substrate selection on the NbN thin films should be investigated in more detail.

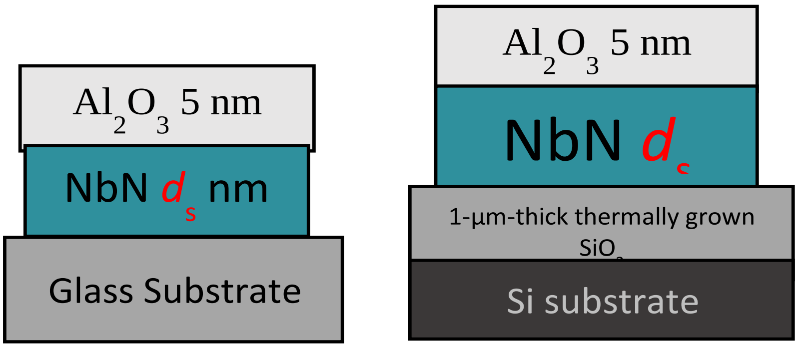
In the past, studies were carried out on the improvement of T_c at optimized production conditions. For example, the influence of the N_2 partial pressure ratio on the film produced at room temperature was studied by Z. Wang et al. [11], at the same time the effect of the temperature of the substrate and sputtering power were investigated by S. Chockalingam et al. [12]. In addition, the effects of film thickness on T_c of NbN is summarized in [13] and [14].

In this study, some basic measurements were made about the production of NbN thin films and the change in superconductivity transition temperature. NbN films were produced in ultra high vacuum system with DC reactive magnetron sputtering technique. The effect of different rates of Ar and N_2 gases on glass and Si / SiO_2 on two different substrate of T_c were investigated. T_c was found in the range of 4.5-8.5 K and 10.1-14.7 K for Nb and NbN thin films, respectively depending on the sputtering conditions.

The highest T_c of the NbN is 14.7 K on the Si/ SiO_2 substrate at 300 W sputtering power. According to our results, Si/ SiO_2 substrate improves superconducting properties of the superconducting thin films. Residual-resistivity ratio (RRR) is calculated for the thin films and the flux of N_2 have a strong effect on RRR of the thin films.

2. MATERIALS AND METHOD

NbN films were produced by reactive DC magnetron sputtering method in cryo and turbomolecular pumped chamber where base pressure prior to sputtering was below $2x10^{-6}$ Pa. The NbN films were deposited using a 99.95% pure Nb target from Kurt Lesker Co. with a diameter of 3 inch in an argon and nitrogen mixture atmosphere onto glass and thermally oxidized Si (Si/SiO_2) substrates. The pure Argon (99.999%) partial pressure was kept at 0.3 Pa and the nitrogen (99.99%) various nitrogen pressures. The nitrogen concentration in the mixed gases, which is defined as the nitrogen partial pressure divided by the total pressure of 1.0 Pa, which can be changed from 0% to 100%. In this study, partial pressure of nitrogen was varied from %15 to %20. Argon flow rate was optimized to produce stress free NbN thin films and the Nitrogen flow rate to reach maximum critical temperature. Gas pressures were precisely measured by digital mass flow meter (Sierra Mass Flow Meter-Smarttrek100). During deposition the flow rate (Ar: N_2) for glass substrate was set to 40: 9.5 sccm and Si / SiO_2 substrate (Ar: N_2) 40: 7.06 sccm. To prevent surface impurities in NbN films to be prepared, pre-sputtering was applied to the Nb target for 20 minutes by closing the cover in front of the substrate prior to the deposition process. The power of DC power supply was kept at 190 and 300 W, respectively. The distance between the target and the substrate in the chamber is 80 cm to achieve the highest deposition rate and the deposition rate was 6.8 nm/s for every sample. Thin film thickness was measured calibrated with a computer-controlled quartz crystal thickness monitor. Then, these thicknesses were confirmed by measuring with a profilometer (Bruker DXT stylus).



(a) **(b)**

Figure 1. Schematic diagram of the NbN films onto a) glass b) Si/SiO2 substrates.

Figure 1. shows the schematic diagram of the thin films which their surfaces were covered with a buffer layer Al_2O_3 of 5 nm thick to prevent the samples from being exposed to external influences and being degraded. The heater current is held constant for all thin film samples to heat the substrate and the substrate temperature was T_s = 450 ^oC.

The thin films electrically characterized by the standard 4-point method. Here, four connections made as two for voltage and two for current terminals. The voltage and current terminals were connected using indium with 0.5 mm diameter copper wire. The temperature of the samples was recorded by a calibrated RuO thermometer.

3. RESULTS AND DISCUSSION

Table 1 shows the variation critical transition temperature (T_c) of the NbN thin film deposited on the glass substrate at 450 ^oC for different thicknesses

Table 1. Variation of the critical temperature by thickness.

NbN Film Thickness (nm)	**T_c (K)**	**RRR ρ_{300}/ρ_{Tc}**	**Argon Flux (sscm)**	**N_2 Flux (sscm)**	**Partial pressure ratio % N_2**
20	10.1	0.90	40	9.5	19.2
50	10.3	0.93	40	9.5	19.2
100	12.0	0.95	40	9.5	19.2
150	13.0	0.94	40	9.5	19.2

As seen in the table 1, the N_2 partial pressure has a value of 19.2% in total pressure. The reactive DC sputtering power is 190, used for the deposition of the NbN films onto glass substrates.

Here, the calculated RRR value is the ratio of the resistivity at 300 K to resistivity at 10 K. RRR value shows the level of the impurity. High RRR value implies high material purity for re-crystallized material. Niobium nitrate thin films can be characterized by RRR as the first criterion of the level of purity. This is easier and faster way when it is compared to thermal conductivity. The results in the table 1 show that increasing the thickness values improves the superconductivity properties of NbN thin films at optimum N_2 and Ar pressure on the glass substrate.

Figure 2 shows the variation of the thickness of the films produced on the glass substrate. Although the film, has thickness of 150 nm, gives the highest superconductivity temperature (T_c = 13 K), the RRR value slightly decreases.

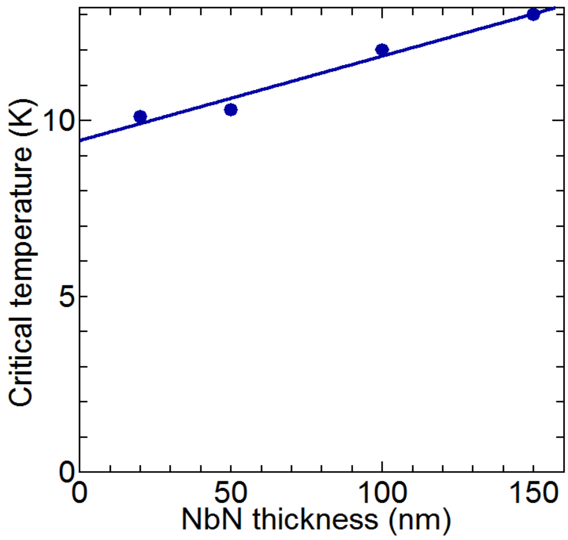


Figure 2. The thickness dependence of critical temperature (T_c).

This decrease in the RRR implies that when the superconductivity is improved on the films, the film quality may be poor. Changes in the grain structure of the films occurred by the deposition of the thicker thin films. In the study [15], the change in film quality is attributed to related the dependence of the T_c and RRR is connected the grain boundaries and scattering in point defects. For example, changes in the homogeneity of the thin film may cause this behavior. It has also been reported [1] that small particle size causes lower T_c values.

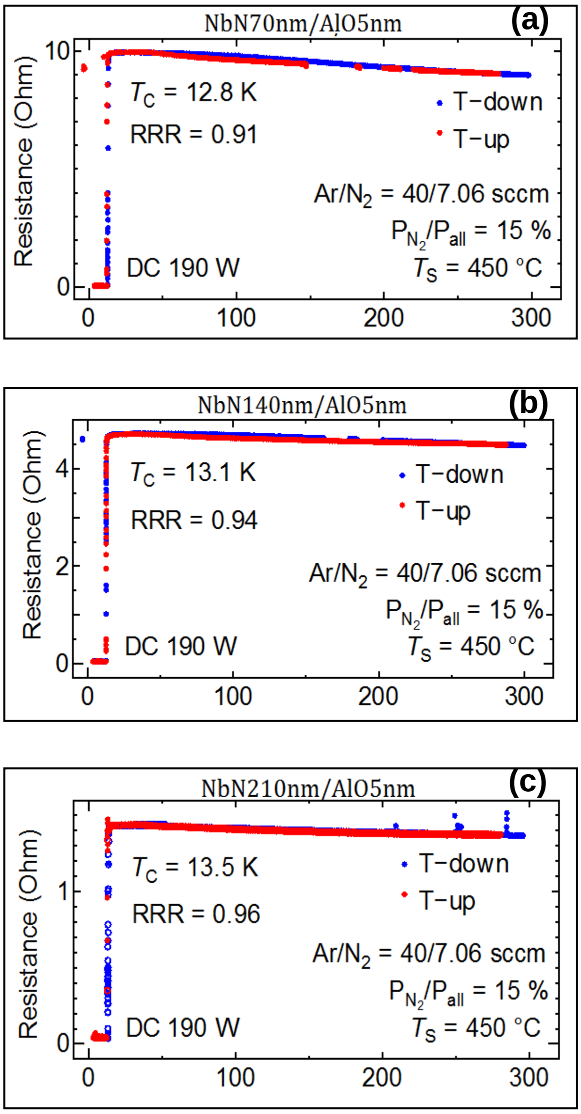


Figure 3. The resistance of NbN films as a function of temperature; a) 70 nm, b) 140nm, c) 210nm thickness.

Deposition of the NbN thin films on Si / SiO_2 substrates significantly changes the RRR and T_c values associated with film quality, despite the reduction of partial pressure N_2 gas and DC sputtering power. It is shown that how changes critical temperature and RRR values in detail for 190 W DC sputtering power, which observed highest T_c is 13.5 K, in the Figure 3.

The N_2 and Ar gas flux ratios, T_c and RRR variations for the films produced onto Si / SiO_2 substrate are given in table 2.

Table 2. Variation of the critical temperature by thickness and sputtering power.

NbN Film Thickness (nm)	**T_c (K)**	**RRR**	**Argon Flux (sscm)**	**N_2 Flux (sscm)**	**Partial pressure ratio % N_2**	**Sputter Power (W)**
70	12.8	0.91	40	7.06	15	190
140	13.1	0.94	40	7.06	15	190
210	13.5	0.96	40	7.06	15	190
210	12.7	0.94	40	7.06	18	300
210	14.7	0.95	40	7.06	19	300

As shown in Fig. 4. the highest T_c is measured as 14.7 K in our work, whereas an argon flow of 40 sccm, a partial N_2 pressure of 19% and a power of sputter of 300 W. Thus we conclude that critical temperature improved with increasing N_2 partial pressure and reaches a highest value. Especially, the effect of N_2 partial pressure on T_c at 300 W is clearly visible for this study.

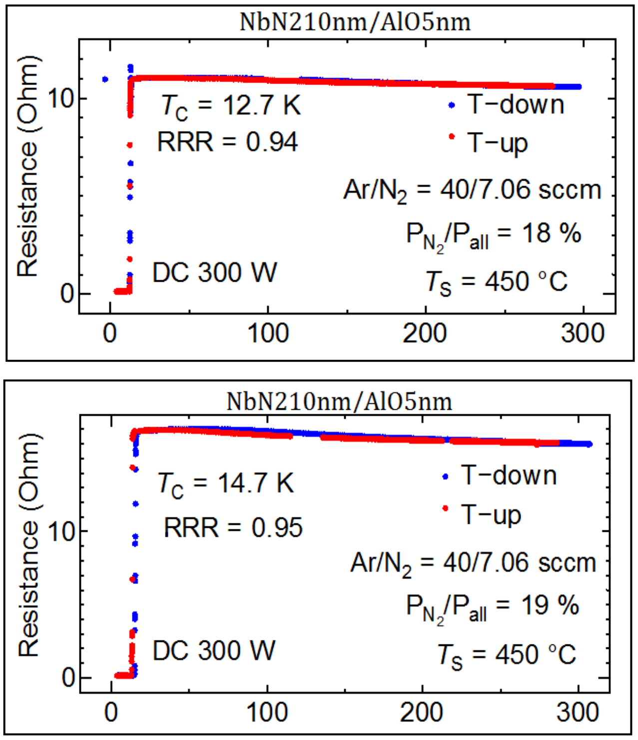


Figure 4. The resistance of NbN films as a function of temperature.

4. CONCLUSION

In this study, it is observed that with the increase in the substrate temperature and sputtering power and occasioned increase on the transition temperature (T_c) of the superconducting NbN thin films. A maximum value of T_c (14.7 K) is measured at 40/7.06 Ar/N_2 pressure ratio, 15% partial pressure of N_2 and 450 ^oC substrate temperature while the minimum is 12.7 K. These results are also routinely reproducible. The best thin films production parameters are calculated via Residual-Resistance Ratio and R-T curves. Then, it is conclude that the increase in temperature of substrate results in favor of critical superconducting temperature. It is also shown that T_c of the NbN films highly depends on the stream of N_2 and Ar gases.

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REFERENCES

[1] M.S. Wong, W.D. Sproul, X. Chu, S.A. Barnett, (1993). Reactivemagnetron sputter deposition of niobium nitride films, J. Vac. Sci. Technol. A: Vacuum, Surfaces, and Films, Vol. 11, No. 4, pp. 1528-1533.

[2] S. K. Kim, B. C. Cha, and J. S. Yoo, (2004). Deposition of NbN thin films by DC magnetron sputtering process, Surface and Coatings echnology,Vol. 177-178, pp. 434–440.

[3] J. J. Olaya, S. E. Rodil, and S. Muhl, (2008). Comparative studyof niobium nitride coatings deposited by unbalanced and balanced magnetron sputtering, Thin Solid Films, Vol. 516, No. 23, pp. 8319–8326.

[4] Kulwant Singh, A. C. Bidaye, and A. K. Suri C. S. Sandu, (2011). Magnetron Sputtered NbN Films with Nb Interlayer on Mild Steel, International Journal of Corrosion, Vol. 2011, pp. 1-11.

[5] Hoshi Y., Terada N., Naoe M., Yamanaka S., (1984). Fabrication of High T_c NbN Films by Ion Beam Deposition Technique, In: Clark A.F., Reed R.P. (Eds.) Advances in Cryogenic Engineering Materials. Advances in Cryogenic Engineering, Vol. 30. Springer, Boston, MA.

[6] M Kidszun, R Hühne, B Holzapfel and L Schultz, (2011). Ion-beam-assisted deposition of textured NbN thin films, Supercond. Sci. Technol. 23, pp. 025010-025018.

[7] G. Cappuccio, U. Gambardella, A. Morone, S. Orlando, and O. P. Parisi, (1997). Pulsed laser ablation of NbN/MgO/NbN multilayers, Applied Surface Science, Vol. 109, pp. 399–402.

[8] Y. Ufuktepe, A.H. Farha, S.I. Kimura, T. Hajiri, K. Imura, M.A.Mamun, F. Karadag, A.A. Elmustafa, H.E. Elsayed-Ali, (2013). Superconducting niobium nitride thin films by reactive pulsed laser deposition, Thin Solid Films, Vol. 545, pp. 601-607.

[9] M. Benkahoul, E. Martinez, A. Karimi, R. Sanjinés, F. Lévy,(2004). Structural and mechanical properties of sputtered cubic and hexagonal NbNx thin films, Surf. Coat. Technol., 180-181, pp. 178-183.

[10] Z. Wang, A. Kawakami, Y. Uzawa, B. Komiyama, (1996). Superconducting properties and crystal structures of single-crystal niobium nitride thin films deposited at ambient substrate temperature, J. Appl. Phys., 79, pp. 7837-7842.

[11] S. Chockalingam, M. Chand, J. Jesudasan, V. Tripathi, P. Raychaudhuri, (2008). Superconducting properties and Hall effect of epitaxial NbN thin films, Phys. Rev. B., 77, pp. 214503.

[12] L. Kang, B. B. Jin, X. Y. Liu, X. Q. Jia, J. Chen, Z. M. Ji, B. G. Wang, (2011). Suppression of superconductivity in epitaxial NbN ultrathin films, Journal of Applied Physics, 109(3), pp. 033908.

[13] S. Ezaki, K. Makise, B. Shinozaki, T. Odo, T. Asano, H. Terai, Z. Wang, (2012). Localization and interaction effects in ultrathin epitaxial NbN superconducting films, Journal of Physics. Condensed Matter: An Institute of Physics Journal, 24(47), pp. 475702.

[14] C. S. Sandu, M. Benkahoul, M. Parlinska-Wojtan, R. Sanjin´es, and F. L´evy, (2006). Morphological, structural and mechanicalproperties of NbN thin films deposited by reactive magnetronsputtering, Surface and Coatings Technology, Vol. 200, No. 22- 23, pp. 6544–6548.

[15] R. Sanjinés, M. Benkahoul, C.S. Sandu, P.E. Schmid, F. Lévy, (2006). Electronic states and physical properties of hexagonal β-Nb2N and δ′-NbN nitrides, Thin Solid Films, Vol. 494, 1-2, pp. 190-195.