Reactive magnetron sputter-deposition of NbN and (Nb, Ti)N films related to sputtering source characterization and optimization

N. N. Iosad[a)]
Delft University of Technology, Department of Applied Physics (DIMES),
Lorentzweg 1,2628 CJ Delft, The Netherlands

B. D. Jackson
Space Research Organization of the Netherlands, P.O. Box 800, 9700 AV Groningen, The Netherlands

S. N. Polyakov
Institute of Nuclear Physics, Moscow State University, 119899, GSP, Moscow, Russia

P. N. Dmitriev
Institute of Radioelectronics Russian Academy of Sciences, Mokhovaya 11,103907, GSP-3, Moscow, Russia

T. M. Klapwijk
Delft University of Technology, Department of Applied Physics (DIMES),
Lorentzweg 1,2628 CJ Delft, The Netherlands

We compare the properties of planar single track balanced and unbalanced sputtering sources by comparing the film properties of NbN and (Nb, Ti)N films sputtered from each source. Our experiments show that reducing the effectiveness of the magnetic trap by changing the magnet configuration is equivalent to reducing the sputtering pressure. We also examine the behavior of the sputtering sources throughout the target lifetime. The balanced sputtering source shows an increase in the magnetic trap effectiveness and a reduction in the heat flux towards the substrate surface as the target becomes grooved (for the fixed applied power and gas pressure), while the unbalanced design shows the opposite behavior. We also show that it is possible to optimize the configuration of the magnetron magnets to produce stable and reproducible (Nb, Ti)N films under the same gas pressure and applied power throughout the target lifetime. © *2001 American Vacuum Society.*
[DOI: 10.1116/1.1349189]

I. INTRODUCTION

Over the last 30 years, magnetron sputtering has become a well-studied subject. In particular a wide range of important aspects have been identified and independently studied including the current–voltage relations in magnetrons,[1] the pressure dependence of the stress in the sputtered films,[2] effects related to the flux of fast neutrals,[3] gas rarefaction in front of the target due to the directed ''wind'' of sputtered particles,[1] plastic flow produced by stress,[4] the formation of the compound layers on the target surface during reactive sputtering,[5] and different configurations of magnetic traps.[6] In this manuscript, we follow the opposite approach. We analyze the properties of deposited films to evaluate the performance of the sputtering source.

The original idea of this approach was suggested by the publication of Imamura *et al.*[7] Figure 1 illustrates the classical dependence of the intrinsic stress of Nb films versus sputtering pressure for different discharge currents. The position of a process on this curve determines the Nb film properties, in agreement with Thornton classification of structural zones.[8] All these intrinsic stress points fit the same curve if plotted versus the discharge voltage (Fig. 2). Imamura *et al.* suggest the following explanation: ''The Nb target's surface is sputtered by the accelerated Ar$^+$ ions. If the energy ex-change between Ar$^+$ ion and Nb occurs without any energy loss, the kinetic energy of the Nb particles ejected from the target equals that of the incident Ar$^+$ ions. Since the bombardment of deposited Nb particles causes compressive stress, we assume that the Nb film stress is strongly related to the cathode voltage.''[7] This explanation is in clear contradiction with the well-known fact that the energy of sputtered particles is approximately equal the binding energy (several electron volts),[9] while the energy of the bombarding Ar$^+$ ions is several hundred electron volts. We suggest a different explanation of the curve illustrated in the Fig. 2. Rossnagel and Kaufman have shown that considerable gas rarefaction occurs in front of the target during sputtering.[1] This effective gas density controls two processes: (a) the thermalization conditions for the sputtering yield and fast neutrals, and (b) the electron temperature in the plasma bulk layer of the glow discharge. Since the discharge voltage, V, is determined by the following equation $V = \alpha(kT_e)^{0.5}$ (where α is a constant for a given target-gas combination, k is the Boltzmann constant, and T_e is the electron temperature), we conclude that this value is indicative of the effective pressure between the cathode and the substrate. On the basis of these considerations, we formulate the concept of our approach: (a) thermalization is the main technological factor determining the properties of films deposited at ambient substrate temperature (in the other words, if the wafers are heat sunk to the

---

[a)]Electronic mail: iosad@dimes.tudelft.nl
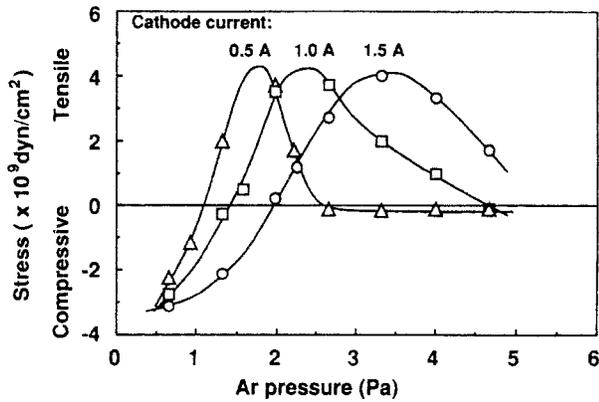
FIG. 1. Film stress vs Ar pressure (modified from Ref. 7).

FIG. 3. Cross-sectional view of the homemade unbalanced magnetron and magnetic field for new and eroded targets. The eroded target profile is indicated by shading (modified from Ref. 14).

chucks maintained at room temperature, we have a classical case of quenched growth),[10] (b) monitoring the dependence of the intrinsic stress on sputtering pressure provides sufficient information for general sputtering source characterization.

Our particular interest in the sputtering of NbN and $(Nb, Ti)N$ is motivated by the fact that these materials are candidates for use in terahertz superconductor-insulator-superconductor (SIS) mixers.[11–13] For this application we need reproducible $(Nb, Ti)N$ films with the lowest possible resistivity, the highest possible transition temperature (T_c), and an acceptable value of intrinsic stress (one that does not cause peeling of the films or photoresist). Because these materials are very sensitive to the sputtering conditions, obtaining films with parameters that are stable and reproducible from run-to-run is very important. In general a reduction in the sputtering pressure results in a change in texture from [111] to [100] and an increase in electrical conductivity and compressive stress.[14,15] Similar changes in the film properties are caused by an increase in nitrogen injection—this causes minor changes in the film chemical composition, but considerably reduces the sputtering rate, thereby resulting in an increase of the effective gas pressure.[16–19] Because these film parameters are important for the device production and also sufficient for sputtering source characterization we compare

the performance of the sputtering sources on the basics of monitoring of the intrinsic stress, texture, resistivity, and the critical temperature (T_c) of the films.

Since the growth of $(Nb, Ti)N$ films is suffering from a lack of surface diffusion,[15] we need a technique to stimulate it. Using an unbalanced sputtering source is one possible solution. The main characteristic feature of such a source is the location of the wafer in the plasma bulk layer.[20] The interaction of the plasma with the substrate surface results in an additional heating compared to the conventional sputtering source and therefore a stimulation of the surface diffusion. This plasma heating is different for the grounded and ''floating'' wafers. If the substrate is floating then the ions are accelerated towards the substrate and electrons are repelled by the ''self-bias'' potential.[21] If the wafer is grounded then the very low plasma potential is not able to repel the electrons nor to accelerate the ions significantly.[21] Therefore, in this case, only the electrons mainly cause the substrate heating.

II. EXPERIMENTAL PROCEDURE

Films of $(Nb, Ti)N$ are deposited by direct current (dc)-magnetron sputtering in a Nordiko-2000 sputtering system with a base pressure of 4×10^{-5} Pa. This machine is equipped with a cryopump and a throttling valve, which together determine the process pressure, while the injection of Ar and N_2 gases is controlled by flow meters. In order to avoid the hysteretic sputtering regime, the pumping rate is fixed at a high value of 750 l/s for all experiments.[22] An alloy target with 30 at. % Ti and 70 at. % Nb is used. In order to maximize film uniformity, the substrate-target distance is set to the maximum for our sputtering system, 8 cm.

A homemade unbalanced 75 mm magnetron sputtering source is used for the deposition of $(Nb, Ti)N$ (Fig. 3). The outer ring of a Leybold PK-75 magnet set is doubled to obtain an unbalanced magnetic trap and a magnetic lens is used to partially loop the magnetic field lines. The purpose of

FIG. 2. Nb film stress as a function of cathode voltage during sputtering (modified from Ref. 7).

the magnetic lens will be discussed in more detail later. A balanced version of this sputtering source is obtained by replacing the magnetic lens with an additional central magnet from the PK-75 magnet set. The rectangular component of the magnetic field above the center of the outer magnet ring is 351 G for both configurations. The rectangular component of the magnetic field in the center of central magnet is 577 G for the balanced design and 254 G for the unbalanced one.

NbN films are deposited in a Leybold high vacuum system L-560 UV using a RPK-125 dc sputtering source at a base pressure of 5×10^{-5} Pa. This sputtering system is not equipped with a throttling valve, so the partial pressure of the process gases is controlled by flow meters. The pumping speed of 1000 l/s, provided by the turbomolecular pump, results in the nonhysteretic sputtering regime. The substrate-target distance is 7 cm.

All films (260 ± 10 nm thick) are sputtered on glass wafers. The substrates are fixed with diffusion pump oil to a copper chuck, which is maintained at $10\,°C$ to stabilize the thermodynamics of growth. The stress in the (Nb, Ti)N films is evaluated by measuring the deflection of the wafer before and after film deposition with a profilometer. The stress value was calculated with the help of the Stoney's equation,[23] using a Young's modulus and Poisson's ratio taken from Ref. 24. T_c is evaluated from the dependence of film resistivity on temperature. The dc film resistivity is measured at 20 K, since this value is related to the radio frequency losses in a microwave tuning circuit. Measuring the resistivity at 20 K instead of 300 K is also more informative from the perspective of the quenched-in crystal point defects concentration. X-ray diffraction (XRD) $\Theta - 2\Theta$ scans in a 2Θ range from 34° to 43° are performed using a Rigaku D/max-Rc diffractometer equipped with a 12 kW x-ray generator of Cu $K\alpha$ radiation and a graphite crystal monochromator.

III. COMPARISON OF BALANCED AND UNBALANCED SPUTTERING SOURCES

In order to evaluate the effectiveness of the balanced and unbalanced magnetic traps of our homemade sputtering source, we have compared the properties of the glow discharge for the two configurations at 6 mTorr Ar pressure. The self-bias of the floating substrate table is -19 V for the unbalanced configuration and -5 V for the balanced case. Furthermore, the ionic current measured with a -30 V biased substrate table is 60 mA for the unbalanced configuration and 7 mA for the balanced case. Finally, the grounding current is equal to the discharge current (≈ -1 A) for the unbalanced configuration, while it is equal to -10 mA for the balanced configuration. These results show that, as expected, the magnetic trap in the balanced configuration is much more effective than in the unbalanced configuration.

The properties of (Nb, Ti)N films obtained using the balanced and unbalanced magnetron configurations are shown in Fig. 4. All films are sputtered under 300 W applied power on a ''floating'' substrate table, and the nitrogen flow rate is optimized to obtain the highest T_c for each total pressure.
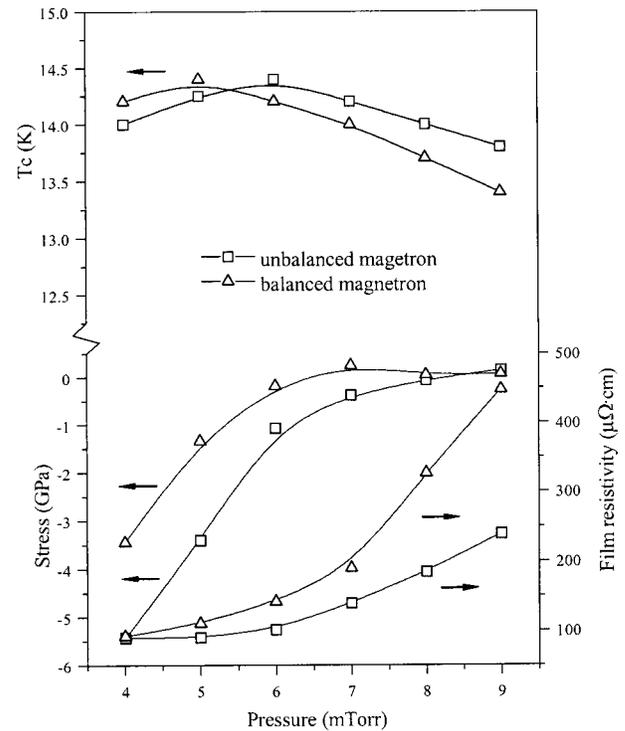
FIG. 4. T_c, intrinsic stress, and film resistivty of (Nb, Ti)N films obtained from the balanced and unbalanced sputtering sources.

This optimization of the nitrogen injection results in the same deposition rate for each data point (80 ± 10 nm), because the poisoning of the target determines both the chemical composition of the films (stochiometric films have the maximum T_c)[15] and the sputtering rate. For both sources, reducing the total sputtering pressure leads to a slight increase in T_c and a decrease in resistivity. This is attributed to a reduced thermalization of the sputtering yield and increased film bombardment by fast neutrals. At the lowest pressures used (4–5 mTorr), this bombardment becomes strong enough to damage the growing film, resulting in a saturation of the resistivity at $\approx 90\,\mu\Omega$ cm and a degradation of T_c. As is seen in Fig. 4, the properties of (Nb, Ti)N films obtained with the unbalanced configuration are similar to those of films obtained with the balanced configuration at lower total pressures. This indicates that the effect of the ion bombardment generated by the unbalanced source is similar to the effect of the increased fast neutral bombardment obtained by reducing the sputtering pressure. Grounding of the wafer during sputtering with the unbalanced source (hereby suppressing the ion bombardment and enhancing the secondary electron heating of the wafer), is also equivalent to increasing of the sputtering pressure, in terms of it's effect on film properties. This indicates that electron heating of the substrate has significantly less effect on the properties of the growing film than ion bombardment.

IV. PERFORMANCE OF THE BALANCED SPUTTERING SOURCE VERSUS TARGET EROSION

We have investigated the behavior of the Leybold dc-sputtering source RPK-125 (seen in Fig. 5) versus target ero-
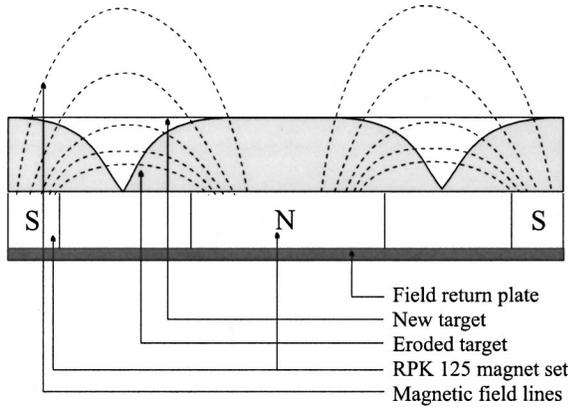
FIG. 5. Cross-sectional view of the RPK-125 magnetron and magnetic field for new and eroded targets. The eroded target profile is indicated by shading (modified from Ref. 18).

sion. NbN films are sputtered with 400 W applied power from 10% and 90% eroded 8-mm-thick Nb target. Independent of target erosion, the self-bias of the floating table is less than 1 V, indicating that the RPK-125 sputtering source has a balanced and very effective magnetic trap. All films are sputtered under a total pressure of ≈ 10 mTorr. The properties of the films sputtered with different nitrogen injection and fixed Ar flow are illustrated in Fig. 6. Increasing of nitrogen injection leads to an increase of film resistivity, a reduction of the deposition rate, and a more pronounced [111] texture for both fresh and eroded targets. We find that as the target becomes grooved, the nitrogen injection must be reduced by 30% to avoid T_c degradation. This is due to a reduction in the sputtering rate caused by a reduction in the effective power density in the erosion track of the grooved target (see Fig. 5). This reduction of the sputtering rate causes an increase in the effective gas pressure between the sputtering source and the substrate. Furthermore, because sputtered particles have an initial velocity perpendicular to the cathode surface, the particles of the sputtering yield will undergo more collisions before reaching the substrate in the case of grooved target. From this perspective the process of target erosion is also equivalent to an increase in substrate-target distance or gas pressure. This leads to a reduction of the heat flux towards the substrate surface, resulting in an increase in the resistivity of the NbN films and a change in the relative XRD intensities throughout the target lifetime. For this reason, we conclude that the process of target erosion in a balanced sputtering source is equivalent to an increase of the sputtering pressure.

V. PERFORMANCE OF THE UNBALANCED SPUTTERING SOURCE VERSUS TARGET EROSION

We have also examined the behavior of the unbalanced sputtering source versus target lifetime. Taking into account the results of the previous section, we have used the magnetic lens to stretch the erosion track, thereby keeping the effective power density as stable as possible throughout the target lifetime. As a result, it is not necessary to correct the
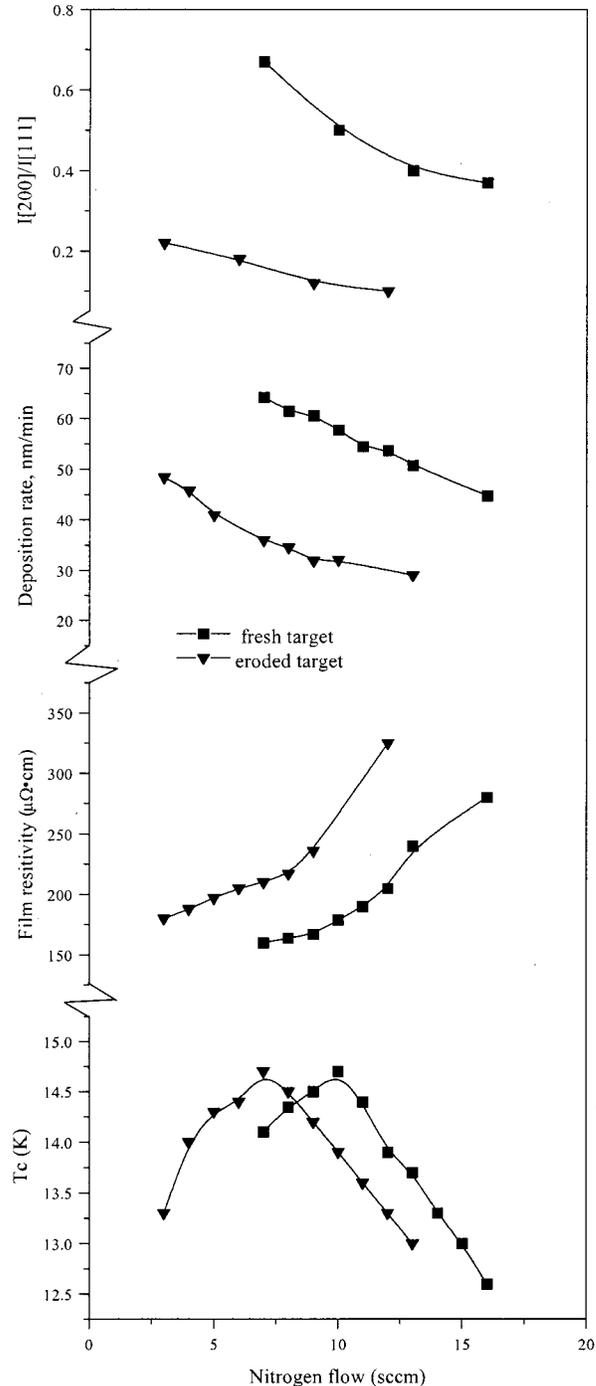
FIG. 6. Relative XRD peak intensities, deposition rate, film resistivities, and T_c of NbN films vs nitrogen injection for fresh and eroded target.

nitrogen injection for sputtering from 10%- and 90%-eroded 5-mm-thick targets. An examination of the properties of the glow discharge at 6 mTorr Ar pressure shows that the self-biasing of the substrate table is -19 V for the fresh target and -22 V for the grooved one. Additionally, the ionic current measured with a -30 V biased substrate table is 60 mA for the new target and 82 mA for the grooved one. These changes can be attributed to a decrease in the effectiveness of the magnetic trap through the target lifetime, due to the magnetic field produced by the outer ring becoming stronger as
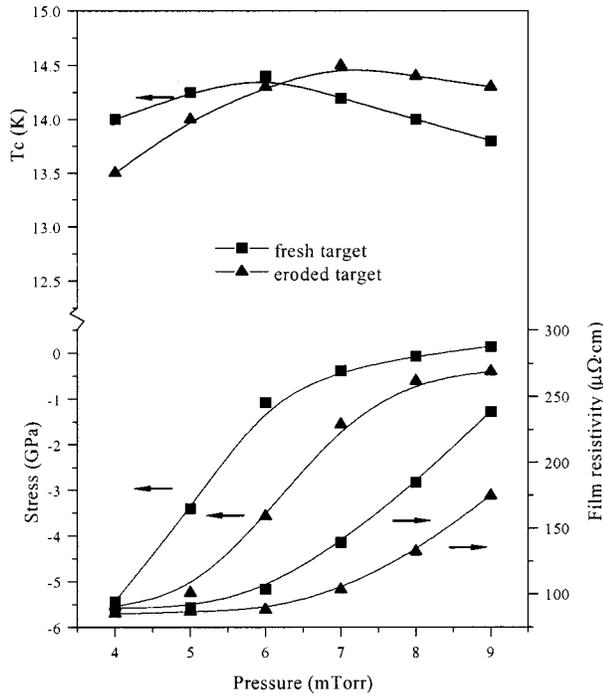
FIG. 7. T_c, intrinsic stress, and film resistivty of (Nb, Ti)N films obtained from the fresh and eroded targets using the unbalanced sputtering source and a floating substrate.

FIG. 8. Relative XRD peak intensities, T_c, intrinsic stress, and film resistivty of (Nb, Ti)N films obtained from the fresh and eroded targets, using the unbalanced sputtering source and a grounded substrate.

the target becomes grooved (see Fig. 3), making the magnetic trap more unbalanced. This explanation is supported by the fact that there is no considerable difference in the discharge voltage with and without the magnetic lens for the case of the eroded target. In contrast, using the magnetic lens in the case of the new target gives a 50 V reduction in the discharge voltage for a constant applied power.

The properties of the (Nb, Ti)N films obtained from the fresh and eroded targets are illustrated in Fig. 7. From this figure, it is seen that there is a significant increase in the conductivity and intrinsic stress of (Nb, Ti)N films deposited from the grooved target, relative to those deposited from a fresh target. Furthermore, at the lowest pressures (4–5 mTorr) the substrate bombardment actually becomes strong enough to cause a plastic flow of films sputtered from the grooved target (seen from the flattening of the stress curve in Fig. 7). These results are in direct contrast to the decrease in conductivity and intrinsic stress observed previously for NbN deposition using a balanced sputtering source. This can be explained by a significant increase in the ion bombardment of the substrate due to the previously discussed reduction in the effectiveness of unbalanced magnetic trap. This effect is insufficiently compensated by the increased thermalization of the sputtering yield and reduced fast neutral bombardment expected to result from the target becoming grooved. Thus, from Fig. 7, we can conclude that the process of target erosion for the unbalanced sputtering source is equivalent to a sputtering pressure decrease.

In order to suppress the contribution of ionic bombardment, we have repeated this experiment with a grounded substrate table. As is seen from a comparison of Figs. 7 and 8, this results in less-stressed films at low pressures and a smaller deviation of film properties throughout the target lifetime. XRD does not show any considerable differences in the film texture for the films sputtered under the same pressure from the fresh and grooved targets. Furthermore, it is seen that it is possible to select an operating point at 5 mTorr, for which the film properties are roughly independent of the target erosion state and satisfy the conditions for device production. At this point, the films have stress ≈ -2 GPa, $T_c \approx 14.3$ K, and resistivity $\approx 100\,\mu\Omega$ cm.
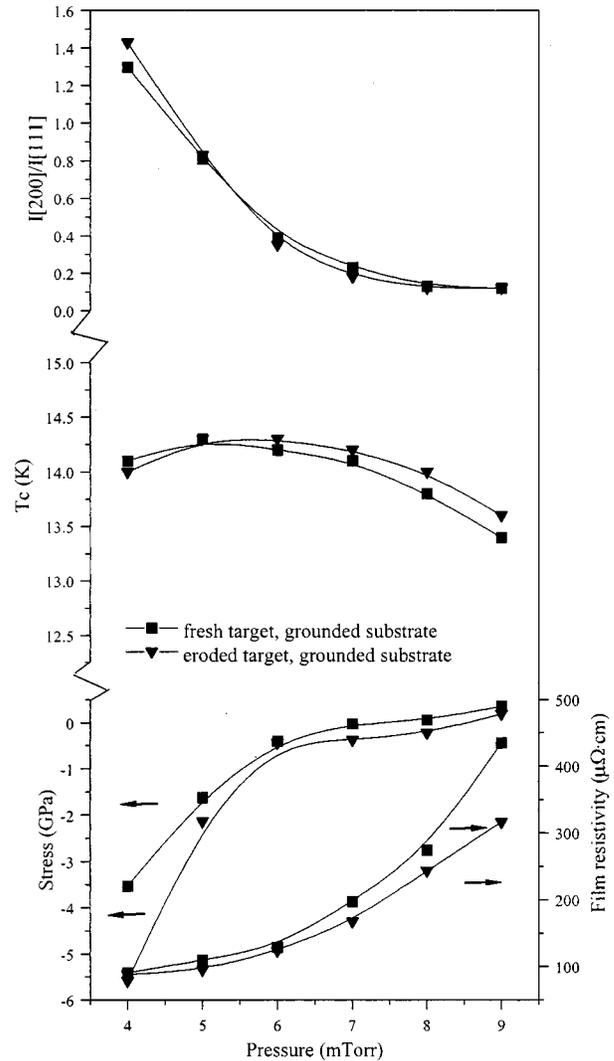
VI. CONCLUSIONS

We have characterized and compared the balanced and unbalanced sputtering sources by monitoring the dependence of intrinsic stress on sputtering pressure. This curve is used

because it can be directly related to the other film properties using the Thornton classification. Following this approach, we find no significant difference between the balanced and unbalanced sputtering sources, since the properties of the (Nb, Ti)N obtained from unbalanced sputtering source are equivalent to the properties of the (Nb, Ti)N films obtained from the balanced one under lower sputtering pressures.

An evaluation of the NbN and (Nb, Ti)N film properties leads us to a conclusion that the heat flux towards the substrate is not stable throughout the target lifetime. The unbalanced sputtering source shows a change in (Nb, Ti)N film properties equivalent to a decrease in the sputtering pressure, while the balanced sputtering source shows the opposite behavior. Furthermore, using an unbalanced source together with a grounded substrate table, it is possible to choose deposition conditions which yield stable and reproducible (Nb, Ti)N film properties throughout the target lifetime.

This approach of monitoring the film intrinsic stress-pressure curve for sputtering source characterization should be verified for a much broader spectrum of materials to establish a possible wider applicability.

ACKNOWLEDGMENTS

[1] S. M. Rossangel and H. R. Kaufman, J. Vac. Sci. Technol. A **6**, 223 (1988).
[2] D. L. Smith, *Thin Film Deposition* (McGraw-Hill, New York, 1995), p. 197.
[3] S. M. Rossnagel, J. Vac. Sci. Technol. A **7**, 1025 (1989).
[4] B. Window, J. Vac. Sci. Technol. A **7**, 3036 (1989).
[5] B. Window and G. L. Harding, J. Vac. Sci. Technol. A **8**, 1277 (1990).
[6] L. B. Jonsson *et al.*, J. Vac. Sci. Technol. A **17**, 1827 (1999).
[7] T. Imamura *et al.*, IEEE Trans. Appl. Supercond. **2**, 1 (1992).
[8] D. L. Smith, *Thin Film Deposition* (McGraw-Hill, New Work, 1995), p. 160.
[9] D. L. Smith, *Thin Film Deposition* (McGraw-Hill, New York, 1995), p. 444.
[10] K.-H. Muller, J. Appl. Phys. **62**, 1796 (1987).
[11] J. Kawamura *et al.*, Appl. Phys. Lett. **75**, 4013 (1999).
[12] Y. Uzawa *et al.*, Appl. Phys. Lett. **69**, 2435 (1996).
[13] B. D. Jackson *et al.*, Appl. Phys. Lett. (submitted).
[14] N. N. Iosad *et al.*, IEEE Trans. Appl. Supercond. **9**, 1716 (1999).
[15] N. N. Iosad *et al.*, Appl. Phys. Lett. **88**, 5756 (2000).
[16] D. D. Bacon *et al.*, J. Appl. Phys. **54**, 6509 (1983).
[17] N. N. Iosad *et al.*, IEEE Trans. Appl. Supercond. **7**, 2805 (1997).
[18] N. N. Iosad *et al.*, IEEE Trans. Appl. Supercond. **9**, 1720 (1999).
[19] M. S. Wong *et al.*, J. Vac. Sci. Technol. A **11**, 1528 (1993).
[20] D. L. Smith, *Thin Film Deposition* (McGraw-Hill, New York, 1995), p. 487.
[21] D. L. Smith, *Thin Film Deposition* (McGraw-Hill, New York, 1995), p. 465.
[22] K. L. Westra *et al.*, J. Vac. Sci. Technol. A **8**, 1288 (1990).
[23] G. C. Stoney, Proc. R. Soc. London, Ser. A **32**, 172 (1909).
[24] J. Hlavac, *The Glass Technology of Glass and Ceramics* (Elsevier Scientific, Amsterdam, 1983), p. 232.