Stress control of reactively sputtered thick NbN film on Si wafer changing the location of the substrate Si wafer against the Nb target on a magnetron cathode

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Abstract. We have been developing a superconducting NbN thin film coil in a spiral trench on a Si-wafer using MEMS technology. Connecting the coils on the different wafers using wafer-bonding process, a cylindrical wafer stack is to be formed as a unit of a compact SMES. The critical current density of our NbN film was measured to be around 1100 A/mm². We measured critical current $I_c$ of 47 mA for the previously fabricated coil of the film thickness $t_f = 0.5$ μm. $I_c$ in the spiral coil increases with $t_f$. However, if we make the NbN film thicker, the film is apt to have higher lateral stress caused by tensile or compressive stress which can cause peeling of the film from the Si substrate. It is well known that the stress of the sputtered thin films can be controlled from tensile to compressive stress by controlling the bombardment of high energy particles including argon atoms backscattered from the target surface. Based on this knowledge, a specially designed sputter-deposition apparatus was fabricated in which the substrate can be located not only at the different target-to-substrate distances but also at several different lateral distances from the central axis of the target (off-axis lateral shift). Using this apparatus, various stress conditions could be realized which contributed to fabrication of thick NbN film spiral coil in the trench. The film stress was calculated from bending analysis of the substrate Si wafer by stylus method using Stoney’s formula. The maximum compressive stress of 2.5 GPa was measured. By an off-axis lateral shift, $t_f$ could be increased from 0.5 to 1 μm. By increasing sputtering gas pressure from 0.7 to 2 Pa, the compressive stress could be mitigated and $t_f$ could be further increased from 1.0 to 3 μm. Up to now, we measured $I_c$ of 220 mA for a NbN spiral coil at $t_f$ around 3 μm. More detailed adjustment of the deposition condition will bring further increase in $t_f$, and hence $I_c$ into sight.
1. Introduction

We have been developing a superconducting NbN thin film coil in a spiral trench on a Si-wafer using MEMS technology as shown in figures 1(a) and (b) [1]. Connecting the coils on the different wafers in series using wafer-bonding process, a cylindrical wafer stack is to be formed as shown in figure 1(c). The superconductive contacts between the neighboring wafers via through-holes are to be attained, following the established conventional multi-wafer interconnection technology as described in detail in our previous paper [1].

A compact SMES system is to be composed of required numbers of the stacks and cryogenic refrigerators as well as other indispensable appendage systems such as thermal and magnetic shields as schematically shown in figure 1(d), for example. The critical current density of our NbN film was measured to be around 1100 A/mm² [2]. We measured critical current $I_c$ of 47 mA for the previously fabricated coil of the film thickness $t_f = 0.5$ μm. Figures 2(a) and (b) shows schematics of the cross-sectional structure of the reactively sputter-deposited NbN thin film spiral coil lined with Cu plated layer in the trench. $I_c$ in the spiral coil increases with $t_f$, and hence the amount of energy storage. Therefore, we would like to increase $t_f$ from the present status as shown in figure 2(a) to figure 2(b) in future.

Figure 2(c) and (d) are cross-sectional SEM images of the trench after NbN deposition in the present status. However, if we make the NbN film thicker, the film is apt to have higher lateral force caused by tensile or compressive stress which can cause peeling of the film from the Si substrate as shown in figures 3 (a)-(c). It is well known that the stress of the sputtered thin films can be controlled from tensile to compressive stress by controlling the bombardment of high energy particles including argon atoms backscattered from the target surface [3]-[13]. In this paper, conditions of reactive sputter-deposition of NbN superconductive film are studied to mitigate film stress to realize thick NbN films showing high $I_c$. Here, it should be noted that elongation of lattice parameters from that of the bulk value of NbN has been known to cause increase in $T_c$ in reactively sputtered thin NbN films for the studies on Josephson tunnel junctions [14]. Therefore, a certain amount of film stress may improve superconducting properties of NbN thin films. However, what we experienced was that thicker NbN films deposited at the corresponding high $T_c$ condition easily break up and peel off from the substrate. Based on this experience, stress free is the best condition as a priority target in this paper. Utilization of any possible effects of the film stress on the superconducting properties can be targets on the next stage.

**Figure 1.** A compact SMES to be formed. (a) a design of a spiral trench on a Si wafer, (b) a SEM image of the central part of the spiral trench formed by MEMS process, (c) a schematics of a stack of Si wafers with spiral superconducting coils connected in series between the wafers, (d) a schematic of a typical example of a compact SMES system.

**Figure 2.** Schematics of the cross-sectional structure of the reactively sputter-deposited NbN thin film spiral coil lined with Cu plated layer in the trench: (a) and (b). Cross-sectional SEM image of the trench after NbN deposition: (c) and (d). The trench geometries are different between (a)(b) and (c)(d).
2. Experimental

2.1. Instrumentation

Based on the knowledge extensively studied on the effect of bombardment of high energy particles in sputter deposition[3]-[13], a specially designed sputter-deposition apparatus was fabricated in which the substrate can be located not only at the different target-to-substrate distances but also at several different lateral distances from the central axis of the target (off-axis lateral shift)[15]. Figure 4(a) shows a top view of the sputtering chamber showing a ceiling circular flange on which a rotatable substrate holder is mounted. Since the circular flange is fixed to the chamber with 24 sets of bolts and nuts, the position of the rotating axis of the substrate holder can be laterally shifted from the central axis of the target mounted on the bottom flange of the chamber by rotating and fixing the ceiling flange by every 15 degree of angle around the center of the ceiling flange. Figures 4(c) and (d) show

(a) Detachment of NbN film from a Si wafer with the spiral trench
(b) NbN film with cracks and wrinkles outside of the spiral trench (magnified view of (a))
(c) Detachment of NbN film from a plane Si wafer

Figure 3. Wrinkles and cracks in the NbN film in the outer side of Si wafer with a spiral trench (a), (b), Detachment of NbN films from a plane Si wafer without a trench(c).

Figure 4. Lateral shift of the rotating axis of the substrate holder from the central axis of the target.
schematics to show this lateral shift of the rotating substrate holder in the case of 0 and 45 degrees of angle, respectively. Figure 4(b) shows a typical example of NbN film thickness distribution as a function of the distance from the central axis of the target when the ceiling flange was rotated and fixed to the chamber by 45 degrees of angle. The target used was of metallic Nb of 3N grade and 76.2 mm in diameter. The perpendicular distance between the target surface and substrate surface could be changed between 50 and 100 mm, but was fixed to 50 mm in this study. The temperature of the substrate holder could be increased and held up to 873 K by radiation heating. The chamber can be pumped down to 1 x 10⁻⁴ Pa using turbo-molecular pump. The sputtering gas flow rate was 44 sccm for Ar and 6 sccm for N₂, respectively. The substrate holder was rotated at 10 rpm.

2.2. Measurement of the stress in the NbN film
Reactive sputter-deposition of NbN film on Si wafers of 101.6 mm in diameter was performed in various conditions. The substrate Si wafer had not gone through the process for removal of the surface oxide layer before the deposition. In general, metallic thin film sputter-deposited at lower sputtering gas pressure than a certain critical value shows compressive stress which would likely extend its area after deposition resulting in formation of wrinkles by local detachment from the substrate as shown in figure 3(b) or bending of the substrate Si wafer convexly as shown in figure 5(a) if the adhesive strength is too high to cause the detachment [8]. At higher sputtering gas pressure than the critical value, the film is to show tensile stress and would likely dwindle its surface area resulting in bending of Si wafer in a concave shape as shown in figure 5(c). Since the thermal expansion coefficient is not so different between Si and NbN, the stress causing the substrate bending can be considered as the intrinsic stress of the film. By measuring this concavity or convexity using stylus method as shown in figures 5(a)-(d), the intrinsic film stress was obtained by the following Stoney’s formula [16].

$$\sigma_f = \frac{E_s t_s^2}{(1 - \nu_s)6R t_f}$$

where $\sigma_f$ stands for the intrinsic stress, $E_s$ for Young’s modulus of Si substrate, $t_s$ for thickness of Si substrate, $\nu_s$ for Poisson’s ratio of Si substrate, $R$ for curvature radius of bending, $t_f$ for thickness of NbN film, respectively.

![Figure 5](image-url)
3. Results

3.1. Effect of off-axis lateral shift of location of rotating substrate holder

Figure 6 (a) shows a photograph of a Si wafer of 76.2 mm in diameter with a spiral groove after deposition of NbN as thick as 0.3 μm at the rotating substrate location as shown in figure 4(c). Although the NbN film in the area of the spiral groove showed no detachment, some detachment could be found in the peripheral region outside of the spiral groove. Therefore, $t_f = 0.3 \mu m$ was considered to be the upper limit of NbN film thickness without remarkable detachment. Figure 6 (b) shows the similar photograph to figure 6(a) but at the rotating substrate location as shown in figure 4(d). The upper limit of NbN film thickness without remarkable detachment increased to 1 μm. Figure 6 (c) summarizes the increase of the upper limit of $t_f$ with increase in the rotation angle of the ceiling flange around the center of the flange.

(a) Rotation angle : 0°
with film detachment, $t_f = 0.3 \mu m$
(b) Rotation angle : 45°
without film detachment, $t_f = 1 \mu m$

Figure 6. Increase in the upper limit of NbN thickness without detachment from the substrate with increase in the rotation angle of the ceiling flange

3.2. Effect of sputtering gas pressure on the NbN film stress

Figure 7 (a) shows $dh$ shown in figure 5 (b) as a function of lateral distance for different deposition times. The deposition time was proportional to film thickness measured by SEM observation. The

(a) $dh$ as a function of the lateral distance from the edge of the Si substrate
(b) $dh$ at the center of the Si substrate as a function of the film thickness

Figure 7. Increase of bending of the sample by compressive stress of the NbN film with increase in film thickness and obtained value for the compressive stress using Stoney’s equation.
lateral distance 0 mm and 50 mm correspond to the edge and the center of the Si wafer, respectively. The substrate temperature was 150 degrees of centigrade and sputtering gas pressure was 0.7 Pa. In this condition, deposition rate was 0.6 μm/h and the film stress was compressive. Detachment of the film from Si substrate took place for the deposition time longer than 2.0 h. The value of $dh$ was almost proportional to the NbN film thickness as shown in figure 7 (b). From the proportional constant, the film stress $\sigma_f$ was obtained to be 2.5 GPa using the Stoney’s formula. Figures 8(a) and (b) show variations of $dh$ with the lateral distance from the edge of the substrate for different sputtering gas pressure. It was shown that intrinsic compressive stress in NbN films were considerably mitigated and even changed to the tensile stress by increasing sputtering gas pressure as in the case of metallic films studied by D. W. Hoffman [8]. By the adjustment of sputtering gas pressure, the compressive stress could be mitigated and NbN film as thick as 3 μm could be deposited without detachment.

4. Discussion

The intrinsic compressive stress in the reactively sputtered NbN films was found to be changed to tensile stress by increasing sputtering gas pressure. This phenomenon is apparently similar to the well-known phenomena in metallic films [8]. In these metallic films, it was considered that energetic particles coming from the target bombarded growing film on the substrate and caused compressive stress. This phenomenon was called as atomic peening effect. By increasing the sputtering gas pressure, those energetic particles were considered to be effectively scattered by the sputtering gas atoms and the atomic peening effect was mitigated. Reduction of the atomic peening effect was considered to cause growth of thin film of lower density than the bulk materials, and cause tensile stress during the densification procedure. As for the energetic particles, neutralized sputtering gas ions once accelerated toward the target and backscattered at the target surface were considered to be one of the candidates. It was shown experimentally that target of elements heavier than sputtering gas atom tended to cause intense atomic peening effect and hence remarkable compressive stress of the film. Since Nb has heavier atomic mass than argon and nitrogen, it is likely for the same phenomenon to take place. On the other hand, there found an unusual dip in film thickness distribution as shown in figure 4(b). The position of the dip seems to correspond the erosion ring of the target which is specific to the planar magnetron type cathode. The similar phenomena were reported by D. J. Kester and R. Messier in the case of four perovskite targets: PbTiO$_3$, CaTiO$_3$ and SrTiO$_3$ and BaTiO$_3$ [12]. In these
cases, the dips were more remarkably formed in the target materials which yield oxygen negative ions more easily. Therefore, the energetic particles in this case was considered to be oxygen negative ions accelerated in the cathode fall region towards the substrate. K. Tominaga et al. reported the similar dips of film thickness distribution in the case of ZnO target [13]. These oxygen negative ions accelerated towards the substrate can be also effectively scattered by sputtering gas atoms, and hence the increase in the scattering gas pressure may mitigate the atomic peening effect. In the present case, it is the point of discussion whether nitrogen negative ions may take the similar role as oxygen or not. However, phenomenologically, the similar phenomenon seems to be going on apparently.

5. Conclusion

By adjusting the sputtering gas pressure and mitigating the compressive film stress at the rotation angle of the ceiling flange of 45 degree of angle, the NbN films of the thickness more than 3 μm could be formed after continuous 12 hours’ deposition without detachment as shown in figure 9. After the deposition of NbN, Cu plating and CMP were performed to complete the spiral coil as having been reported in our previous report [1]. The input forward RF power was 200 W while the reflected power was almost zero W. The substrate temperature was 300 degree of centigrade. Figure 10 shows temperature dependence of resistivity of a spiral coil in the trench as shown in figure 2(a) on the Si wafer of 76.2 cm in diameter measured by two-terminal method in the cryostat. Clear change of resistivity from that of the Cu coil to that of the superconducting NbN coil only with the contact resistance around 2 ohm was observed at the transition temperature around 15.5 K. The maximum critical current was 220 mA at 4 K. More detailed adjustment of the deposition condition will bring further increase in the film thickness to fill the trench as shown in figure 2 (b), and hence further increase in the critical current into sight.

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