

Superconductivity and electrical properties in single-crystalline ultrathin Nb films grown by molecular-beam epitaxy

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High-quality superconducting ultrathin Nb films were grown on sapphire single-crystal substrates by molecular-beam epitaxy, of which thicknesses ranged from 1.2 to 9.0 nm. They were found to be single crystalline by reflection high-energy electron diffraction. Size effects appeared remarkably in the normal conduction range. Superconducting transition temperatures (T_c 's) of these films were higher than in the past reports for the same thicknesses. The thickness (d) dependence of T_c was $\ln[T_c(d)] \propto -1/d$, which could be explained in terms of the proximity effect. A theory of localization effects in two-dimensional superconductors by Maekawa and Fukuyama, on the other hand, could not be applied to the present experiments, since the prepared ultrathin films had better crystallinity than those discussed in the theory.

I. INTRODUCTION

Single-crystalline superconducting thin films have been studied by many researchers because of their importance in application to Josephson junctions, Josephson LSI circuits, etc.¹⁻¹³ As for the constituent material, its transition temperature (T_c) is desirable to be as high as possible for reliable operations at 4.2 K. In addition its crystal structure and composition should be simple from the viewpoint of film preparations. For the repeated cycles between room temperature and 4.2 K, thermal-expansion coefficients of the substrate and the grown film should be similar to each other.

The system of an epitaxial Nb film grown on a single crystal of sapphire, therefore, has been selected and studied extensively, so far. Although superconductivity was reported for the 0.9 nm thick Nb film having texture structure,¹⁴ the minimum thickness was 2.5 nm for the epitaxial Nb films in the past reports.⁵ Thus preparation of the epitaxial films thinner than 2.5 nm and characterization of their crystallinity and superconducting properties seem quite important.

From the viewpoint of basic research, a superconductivity of thin films has been intensely discussed. A decrease in T_c has been explained theoretically by the proximity effect¹⁵ and the electron localization effect in two-dimensional

systems.¹⁶ Some experimental results of Nb thin films have been interpreted in terms of the proximity effect,^{5,17-20} where the system is considered to consist of both a superconducting layer and a normal-metal layer. The normal conduction region has been considered to be an interface region between the Nb film and the sapphire substrate or between the Nb film and the surface region.

Under the condition that the proximity effect is negligible, the electron localization effect can be responsible for the T_c depression, as reported in the case of amorphous films of Nb (Refs. 19 and 20) and Mo-Ge.²¹ These experiments have been made intentionally by use of the amorphous or the textured films in the past reports. Thus the experiments with epitaxial films in the very thin range are expected to provide interesting results.

In this context, the purposes of the present investigation are (1) To prepare very thin (thinner than 2.5 nm) single-crystalline Nb films, and to characterize their crystallinity and superconducting properties, (2) to discuss superconducting properties in terms of the proximity effect and the localization effect.

It is already known that single-crystalline Nb films can be grown on α -Al₂O₃ substrate surfaces with different crystal orientations. The epitaxial relations are as follows:

$$\begin{aligned} \text{sapphire}(11\bar{2}0) \text{ (sapphire-A), } [0001] \parallel \text{Nb}(110), [\bar{1}11] \text{ (Refs. 1, 3-6, 9-11, and 13),} \\ \text{sapphire}(0001) \text{ (sapphire-C), } [2\bar{1}10] \parallel \text{Nb}(110), [\bar{1}10] \text{ (Refs. 3 and 8),} \\ \text{and } [2\bar{1}\bar{1}0] \parallel \text{Nb}(111), [\bar{1}10] \text{ (Refs. 1, 3, 5, 6, 9, and 10),} \\ \text{sapphire}(1\bar{1}02) \text{ (sapphire-R), } [11\bar{2}0] \parallel \text{Nb}(100), [011] \text{ (Refs. 5, 8, and 10).} \end{aligned}$$

In the present work, these three different substrates were used.

In the following, we will start with the fabrication of epitaxial films and the characterization of the prepared films, being followed by the discussions on their electrical properties.

II. EXPERIMENTAL

Growth of epitaxial Nb films was examined in an ultra-high vacuum chamber with a base pressure of 1×10^{-8} Pa. The pressure under the film growth was as low as 2×10^{-7} Pa with the aid of a liquid N₂ shroud.

Single crystals of sapphire (α -Al₂O₃) were used as the substrate. The substrates were cleaned in boiled concentrated HNO₃ for 5 min, etched in a solution of HCl and H₂O₂ for 10 min, and rinsed in distilled water, then loaded into the molecular-beam epitaxy chamber and heated at 800 °C for 60 min. After these processes, sharp streak patterns were observed in reflection high-energy electron diffraction (RHEED). *In situ* Auger electron spectrum measurement indicated that the substrate surfaces were free of carbon and other contaminants.

A Nb pellet (99.95%, Furuuchi Chemical Co.) was used for the Nb source. Niobium beam was generated with an electron-beam gun (−10 kV, 110 mA) and irradiated the substrates with a rate of about 0.0025 nm/s. The substrate temperature was 700 °C for all the substrates. Thickness of the film was monitored with a quartz crystal thickness monitor, calibrated *ex situ* with Sloan Dektak II. The Nb beam was so stable through the whole experiment that the film thickness could be evaluated precisely. Crystallographic orientations and crystal surface morphology were checked with RHEED. Before being taken out into the atmosphere, some films were covered with amorphous Se thin layers (about 3.0 nm thick) at room temperature, in order to prevent Nb surfaces from oxidation. These Se layers hardly affect the electrical properties since the resistivity of amorphous Se layer is much larger than that of Nb.

The transport properties were measured *ex situ* by a four-point dc technique in a cryostat which could be cooled down to 1.7 K. The temperatures of the samples were measured by a calibrated Si diode thermometer (Cryo Cal CD301). Sample current was changed from 1 to 20 μ A for each sample, of which value was determined to keep Ohmic conductance and to avoid sample temperature rise.

III. RESULTS AND DISCUSSION

A. RHEED patterns of the prepared films

Figures 1(a) and 1(b) show the RHEED patterns of the Nb films grown on sapphire-A and -R substrates, respectively. RHEED patterns show sharp streaks, indicating the grown films have good crystallinities. For all A and R substrates, the optimum growth temperature was 700 °C, at which RHEED patterns presented the sharpest streaks. In the case of the Nb films grown on sapphire-C, however, RHEED patterns consisted of somewhat broad streaks at any growth temperature. In addition, x-ray-diffraction analysis indicated that these films consisted of two domains of Nb(110) and Nb(111) orientations.

The present RHEED observations revealed that the epitaxial relations between the substrates and the grown Nb films were the same as the past reports described in Sec. I for all sapphire substrates.

In the previous reports on the films grown on sapphire-A and -R, some streaks originating from surface superlattice were observed,^{6,9,13} while they did not appear reproducibly in the present results. This phenomenon did not depend on substrate temperature and we think it was caused by pretreatment procedures and other uncertain factors.

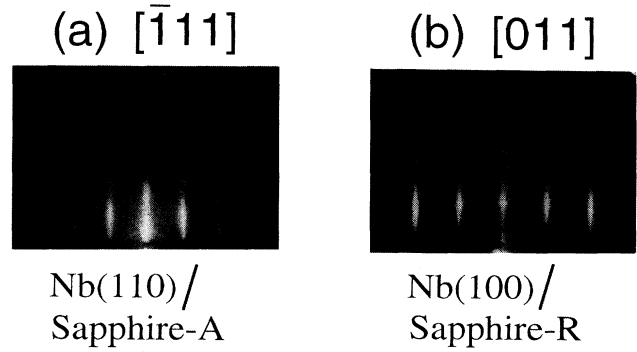


FIG. 1. RHEED patterns of 5 nm thick Nb films grown on (a) sapphire-A and (b) -R substrates, respectively. The incident electron beam was parallel to the (a) $[\bar{1}11]$ axis of Nb (\parallel sapphire-A $[0001]$) and (b) $[011]$ axis of Nb (\parallel sapphire-R $[11\bar{2}0]$), respectively.

B. Electrical properties in normal conduction range

Several parameters of the samples with various thicknesses are shown in Table I. First we will discuss the electrical properties of the films in the normal conduction range. As for the notation, d means the film thickness, ρ means the resistivity, and R_{\square} means the sheet resistance. The relation between ρ and R_{\square} is $\rho = R_{\square}d$. A residual resistivity ratio (RRR) is defined as the ratio of $\rho(295 \text{ K})$ to $\rho(10 \text{ K})$ here, since ρ -temperature curves are almost flat around 10 K for all samples and therefore ρ at 10 K [$\rho(10 \text{ K})$] can be considered as the residual resistivity in normal conduction range.

Figure 2 shows a plot of sheet resistance (R_{\square}) versus thickness (d). The size effect theories of Fuchs²² and Sondheimer²³ predict that R_{\square} is nearly proportional to d^{-2} by the boundary scattering on the condition that $d \ll l_{\infty}$, where l_{∞} is the mean free path when $d \rightarrow \infty$. To estimate the value l_{∞} , we used the relation between ρ and l (mean free path) for bulk Nb; $\rho_0 l_0 = 3.27 \times 10^{-6} \mu\Omega \text{ cm}^2$ (the subscript 0 means the values are those for bulk).²⁴ From the bulk ρ_0 (10 K) value,²⁵ l_0 is estimated to be more than 8000 nm. Although the crystallinity of the prepared films is a little inferior to that of bulk; $l_{\infty} < l_0$, the condition $d \ll l_{\infty}$ is considered to be still valid.

As is seen in Fig. 2, the theory fits well for the samples thicker than 2.0 nm (solid line for the samples with $d \geq 2.0$ nm has the slope of about -2), indicating that the prepared films have uniform thicknesses over the wide area of the entire surface. In addition, the remarkable appearance of the size effect indicates that the films have high crystalline quality, since the scattering caused by defects and impurities would play a determining part in the resistivity at low temperature in poor-crystalline-quality films.

As is seen in Table I, RRR values of thin films (thicker than 1.5 nm) were larger than unity as well as thicker films. This shows that even thin films were metallic and continuous ones. As for extremely thin films (1.2–1.5 nm), however, their values vary from sample to sample even if their thicknesses are the same. Furthermore, resistivities of some of such thin samples increased as the temperature goes down until T_c (i.e., $\text{RRR} < 1$). This indicates that these films are considered to consist of isolated islands.

Here, it should be noted that the observed temperature change in resistivity for some films is greater than the bulk

TABLE I. Electrical and superconducting parameters of the samples grown on (a) sapphire-A, (b) -C, and (c) -R substrates, respectively. The notations in this table are d : film thickness, ρ : resistivity, R_{\square} : sheet resistance, RRR: ρ (295 K)/ ρ (10 K), T_c : superconducting transition temperature, ΔT_c : transition width. Samples 3–8 in (a), 5, 6 in (b), and 5–13 in (c) were those with amorphous Se protective layers.

(a)									
Sample No.	d (nm)	ρ (295 K) ($\mu\Omega$ cm)	ρ (10 K) ($\mu\Omega$ cm)	R_{\square} (295 K) (Ω)	R_{\square} (10 K) (Ω)	RRR (ρ (295 K)/ ρ (10 K))	T_c (K)	ΔT_c (mK)	
1	7.0	31.5	10.2	45.0	14.6	3.08	8.13	146	
2	3.0	25.1	17.9	83.5	59.8	1.40	5.84	621	
3	3.0	32.1	17.9	107	59.6	1.79	6.99	331	
4	2.5	40.5	31.5	162	126	1.29	5.00	286	
5	2.0	69.0	52.0	345	260	1.33	6.30	718	
6	2.0	34.6	29.1	173	146	1.18	5.40	573	
7	1.5	322	301	2150	2004	1.07	5.83	3364	
8	1.2	84.8	91.1	707	760	0.93	3.90 ^a	> 2150	
(b)									
1	9.0	33.4	8.29	37.1	9.21	4.03	8.73	78	
2	8.0	27.7	8.24	34.6	10.3	3.35	8.66	60	
3	7.0	40.5	17.9	57.9	25.6	2.26	7.88	151	
4	5.0	39.5	17.5	78.9	34.9	2.26	7.69	286	
5	5.0	29.7	12.7	59.4	25.3	2.35	7.51	401	
6	2.0	96.8	80.6	484	403	1.20	4.06	907	
(c)									
1	7.0	24.1	8.75	34.4	12.5	2.75	8.10	321	
2	7.0	41.9	14.0	59.8	20.0	2.99	8.21	82	
3	5.0	38.1	16.1	76.1	32.2	2.36	7.61	455	
4	3.0	31.8	17.5	106	58.2	1.82	7.24	368	
5	4.0	46.0	22.5	115	56.3	2.05	6.97	453	
6	3.0	37.8	18.0	126	60.0	2.10	7.47	262	
7	2.5	24.3	12.3	97.0	49.3	1.97	7.05	335	
8	2.5	50.0	38.3	200	153	1.31	6.72	290	
9	2.0	27.6	18.8	138	93.9	1.47	4.88	483	
10	1.5	61.5	48.5	410	323	1.27	2.91	981	
11	1.5	279	275	1861	1831	1.02	5.40	670	
12	1.5	634	767	4227	5110	0.83	4.60	1835	
13	1.5	1439	2377	9596	15 844	0.61	5.79	> 4000	

^aThis value is that of onset.

value of about $15 \mu\Omega$ cm. This might be impossible provided that only the phonon part in resistivity is taken into account. Although we could not interpret this phenomenon at the present stage, it may be explained on the assumption that the boundary scattering part in resistivity is temperature dependent as well as the phonon part.

The scattering in parameters in extremely thin films (1.2–1.5 nm) is considered to arise from an interface layer formed between the sapphire substrate and the Nb film during the epitaxial growth. The effect of this layer may become prominent as film thicknesses decrease. There are two possible candidates for this interface layer. One is niobium oxide, and the other is Nb-Al intermetallic compound. Ohuchi and Kohyama pointed out that one atomic layer of niobium oxide was formed at the interface in addition to the segregation of metallic Al species into the Nb overlayer when a Nb film was grown on an α -Al₂O₃ (0001) (sapphire-C) substrate at a substrate temperature of 1000 °C.^{26,27} In the present work, the growth temperature (700 °C) was much lower and different

surfaces (sapphire-A, -R) were used as the substrates. Although the interface layer is not always the same as that in the above-mentioned report, some kinds of Nb-Al intermetallic compound as well as niobium oxide are considered to be formed. The existence of a metallic layer in the interface region could explain the behavior of T_c , which will be discussed in Sec. III D.

The resistivities of the samples thinner than 2.5 nm without amorphous Se protective layer increased with a decrease in temperature. This is considered to be caused by the surface oxidation, and the Se layer actually prevents the Nb film from surface oxidation by atmospheric oxygen.

C. General properties in superconductivity

Several parameters concerning superconducting properties of the samples were also shown in Table I. A superconducting transition temperature (T_c) was defined as the temperature at the half of the residual resistivity, and a width of

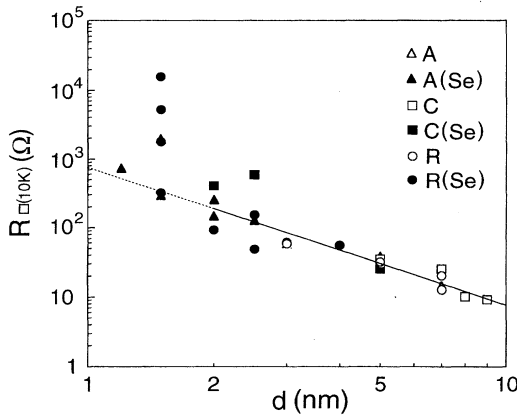


FIG. 2. Sheet resistance plotted vs film thickness. Symbols A, C, and R indicate the kinds of the sapphire substrate. The symbol of Se attached to solid triangles, squares, and circles represents the sample with a Se protective layer. Solid line for the samples with $d \geq 2.0$ nm has the slope of about -2 , in agreement with the value predicted by the boundary theories of Fuchs and Sondheimer (Refs. 22 and 23, see text).

transition temperature (ΔT_c) was defined as the temperature width between the residual resistivity of 10 and 90 %.

The transition widths of the relatively thick films were quite sharp as shown in Table I. Transition temperatures are somewhat higher than those in previous reports with the same thickness from 1.5 to 9.0 nm,^{5,14,17,20} In addition, the minimum thickness showing superconductivity was 1.2 nm, which is thinner than any results in previous reports on epitaxial Nb films. These results proved that the prepared films in the present work had very high crystalline quality.

D. Thickness and T_c : An approach from the proximity effect

The depression of T_c in thin films has already been reported in plenty of materials,^{5,14,17-21,28} and two possible explanations have been proposed for this phenomenon. One is the proximity effect,¹⁵ and the other is the localization effect in two-dimensional systems.¹⁶

First we apply the proximity effect to our experimental results. We will consider the system of a superconducting layer (thickness d) contacting with a normal-metal layer (thickness a).

Figure 3 shows a plot of T_c versus inverse of thickness ($1/d$). As seen in this graph, $\ln T_c$ decreases almost linearly with $1/d$. This relation between T_c and d (i.e., $\ln T_c \propto -1/d$) was obtained also in the study of Wolf and co-workers^{5,17} for somewhat thicker films. Although they used a model based on the proximity effect proposed¹⁵ by Cooper to explain that dependence of T_c on d , a modified form in a McMillan's report²⁹ seems preferable. According to the latter, the decrease in T_c with a decrease in d is written as follows at "Cooper limit:"²⁹

$$T_c = T_{c0} \left(\frac{2\gamma\omega_c}{T_{c0}\pi} \right)^{-[N_N(0)/N_S(0)](a/d)}, \quad (1)$$

where T_{c0} is the transition temperature of bulk, $\gamma = 1.781$, ω_c corresponds to the Debye temperature (Θ_D) of the super-

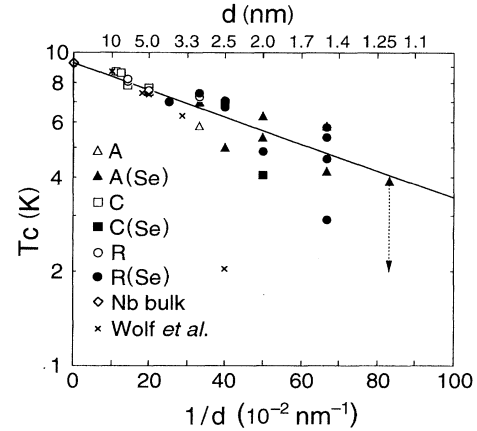


FIG. 3. Plot of T_c vs inverse thickness (d^{-1}). Solid line corresponds to Eq. (1) in text. The points indicated with \times are plotted using the data by Wolf *et al.* (Ref. 5). Symbols are the same as those in Fig. 2.

conducting layer, and $N_N(0)$ and $N_S(0)$ are the density of states for normal and superconducting layers, respectively.³⁰ Then, T_c is expected to decrease as $\ln[T_c(d)] \propto -1/d$, assuming that a is constant.

Substituting the Debye temperature of Nb $\Theta_{D_{\text{Nb}}} = 275$ K for ω_c in Eq. (1), the slope of the line in Fig. 3 gives a thickness of the normal metal layer $a = 0.3\alpha$ nm under the presumption $a \ll d$, where $\alpha = N_S(0)/N_N(0)$. This is of the order of the lattice parameter if α is of the order of unity. Then this value seems reasonable.^{5,17}

There should be the normal-metal layer adjacent to the superconducting layer, as long as the above explanation is valid. This layer is expected to be an interface compound layer between the sapphire substrate and the epitaxial Nb film, as was discussed in Sec. III B. The interface layer probably affects also the scattering in parameters in very thin films ($d \leq \sim 2.0$ nm).

In this way, T_c depression can be explained by the proximity effect. But the localization effect can also influence T_c , as will be discussed in the next subsection.

E. An approach from the localization effect: Maekawa-Fukuyama theory (Ref. 16)

The other possible explanation for T_c depression is the localization effect in two-dimensional systems.^{16,19-21} We apply the theory of Maekawa and Fukuyama,¹⁶ predicting the T_c depression of the two-dimensional superconductor as follows:

$$\ln \left[\frac{T_c}{T_{c0}} \right] = - \frac{1}{2} \frac{g_1 N(0) e^2}{2\pi^2 \hbar} R_{\square} \left[\ln \left[5.5 \frac{\xi_0}{l} \frac{T_{c0}}{T_c} \right] \right]^2 - \frac{1}{3} \frac{g_1 N(0) e^2}{2\pi^2 \hbar} R_{\square} \left[\ln \left[5.5 \frac{\xi_0}{l} \frac{T_{c0}}{T_c} \right] \right]^3, \quad (2)$$

where T_{c0} is the transition temperature of bulk, g_1 is the electron-electron interaction, $N(0)$ is the density of states, R_{\square} is the residual sheet resistance, ξ_0 is the coherence length, and l is the mean free path. This theory predicts that T_c decreases roughly in proportion to the sheet resistance.

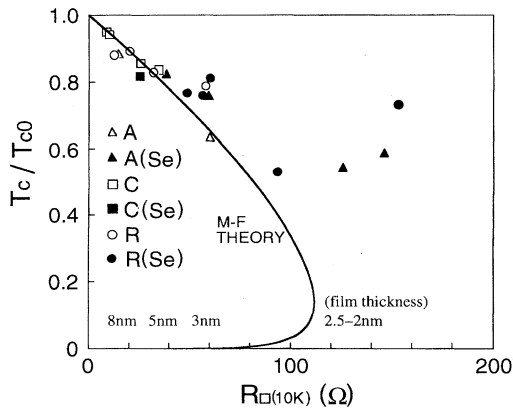


FIG. 4. Plot of T_c vs sheet resistance less than 200Ω , with fit to Maekawa-Fukuyama (MF) theory (Ref. 16), where $g_1 N(0) e^2 / 2 \pi^2 \hbar = 2.4 \times 10^{-4} \Omega^{-1}$; i.e., $g_1 N(0) \approx 20$. The value of $g_1 N(0)$ is too large and MF theory cannot be applied to the present experimental result. Symbols are the same as those in Fig. 2.

In order to apply the theory to the present results, the value of (ξ_0/l) should be estimated. We estimated it in the following way. $\xi(0)$ can be determined from a temperature dependence of an upper critical magnetic field; H_{c2} . We measured the H_{c2} -temperature curve for relatively thick samples (5.0–9.0 nm), which provided the average value of $\xi(0)$ as 16.8 nm. This value was adopted as ξ_0 . To estimate the value l , we used an already quoted relationship between ρ and l for bulk Nb (see Sec. III B). Using the average value of ρ of relatively thick samples, l was estimated to be 2.8 nm at 10 K.

Figure 4 is a plot of T_c versus R_\square at 10 K. A theoretical curve from Eq. (2) is also shown in Fig. 4. As is readily seen in this figure, the experimental result seems to agree to the theoretical prediction in appearance, for R_\square less than about 100Ω (roughly corresponding to the samples thicker than 2.0 nm). The parameters in Eq. (2) have been calculated from the experimental data with the above-estimated values of ξ_0 and l , however, resulting $g_1 N(0) e^2 / 2 \pi^2 \cdot \hbar = 2.4 \times 10^{-4} \Omega^{-1}$; i.e., $g_1 N(0) \approx 20$. This value is too large since $g_1 N(0)$ should be of the order of unity.¹⁶ Then T_c depression in the present work cannot be explained with Maekawa-Fukuyama (MF) theory. This is because the prepared films in the present work have better crystallinity than those assumed in MF theory. Actually, in the case of amorphous films, experimental results fit well to this theory.²¹

In Fig. 4 some of very thin films show relatively high T_c for their R_\square at 10 K. This may be ascribed to the fact that a little worse crystallinity in such very thin films affects the

resistivity more than the T_c . The relation between R_\square and T_c tends to approach the MF theory for such films, although significant deviation still exists.

Nevertheless seeing T_c come up in the Se coated films in Fig. 4, one may suspect that NbSe_2 ($T_c = 7.2 \text{ K}$) could be formed at an interface between a Nb film and a Se protective layer. The reaction of Nb and Se to NbSe_2 , however, is possible for the substrate temperature higher than 500°C .³¹ In the present experiment Se was deposited onto Nb film at room temperature. On the other hand, T_c of NbSe_2 is very sensitive to stoichiometry between Nb and Se.³² It is rather difficult to obtain a superconducting NbSe_2 film, even if the substrate temperature and other conditions could be optimized. Thus we can conclude that NbSe_2 formation did not occur, although we could not deny other (unknown) compound formation completely.

Although an influence of the localization effect on T_c depression still cannot be excluded from the consideration, there is no theory for such high-crystalline-quality films as those prepared in the present work.

IV. CONCLUSION

Superconducting ultrathin Nb films were prepared by molecular-beam epitaxy, of which thicknesses ranged from 1.2 to 9.0 nm. The size effect appeared remarkably in the normal conduction range. Even a sample with thickness of 1.2 nm has shown superconductivity in the present experiment, which is thinner than those in any previous reports on epitaxial Nb films. In addition, the transition temperatures were higher than those in the previous reports with same thicknesses. These results indicate that films with very high-crystalline quality were prepared in the present study.

The decrease in T_c can be explained by the proximity effect on the assumption that a very thin nonsuperconducting metallic layer exists between a Nb film and a sapphire substrate. The localization effect theory, cannot be applied to the present work, which also indicates the high crystallinity of the grown films.

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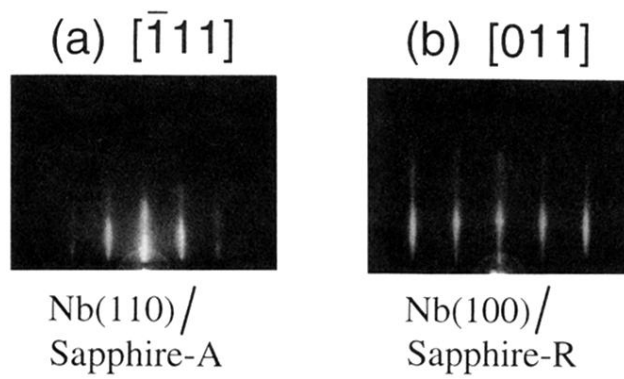


FIG. 1. RHEED patterns of 5 nm thick Nb films grown on (a) sapphire-A and (b) -R substrates, respectively. The incident electron beam was parallel to the (a) $[\bar{1}11]$ axis of Nb (\parallel sapphire-A $[0001]$) and (b) $[011]$ axis of Nb (\parallel sapphire-R $[11\bar{2}0]$), respectively.