Effects of Ion Milling on the Microwave Properties of MgB$_2$ Films

Sang Young Lee, J. H. Lee, J. Lim, H. N. Lee, S. H. Moon, B. Oh and M. A. Hein

Abstract—The new superconductor MgB$_2$ may prove useful for microwave applications at intermediate temperatures. MgB$_2$ films with the thickness of 300 – 400 nm have surface resistance $R_S$ less than 10 $\mu\Omega$ at a frequency ($f$) of 8.5 GHz and 7 K, and ~1.5 m$\Omega$ at 87 GHz and 4.2 K. The critical temperature ($T_C$) of these films is ~39 K when they are prepared under optimum conditions. $R_S$ appears to scale as $f^2$ up to 30 K. After surface ion-milling, a reduction of the $T_C$ and an enhanced resistivity $\rho(T_C)$ are observed consistently at 8.5 GHz and 87 GHz along with a reduced $R_S$ at low temperatures. The observed $\rho(T_C)$ - $T_C$ relation and the uncorrelated behavior between $\rho(T_C)$ and $R_S$ values measured at low temperatures are well explained in terms of the two-gap model, with the observed $\rho(T_C)$ - $T_C$ relation attributed to the properties of the large gap, and the $R_S$ at lower temperatures reflecting the properties of the small gap, with an enhanced small gap energy due to increased interband scattering. This study suggests that the interband scattering should be enhanced to improve the low temperature microwave properties of MgB$_2$ films and that the ion-milling process must be performed with great care to preserve the high quality of MgB$_2$ films.

Index Terms—Ion-milling, MgB$_2$ film, Microwave properties, Two gap.

I. INTRODUCTION

The boride superconductor MgB$_2$ discovered in early 2001 [1] appears attractive for device applications in an intermediate temperature range [2]; the critical temperature ($T_C$) is the highest among intermetallic compounds and the superconductors, MgB$_2$ is thought to have s-wave gap symmetry [4], which is expected to allow an exponential dependence of the surface impedance on temperature at low temperatures [5]-[7]. Furthermore, the fact that high-quality MgB$_2$ films can be prepared on sapphire substrate [8, 9] makes it easier to produce them at a relatively low cost for practical microwave applications.

Several factors must be understood and improved, however, before MgB$_2$ can become a attractive superconductor. These include the observed rapid drop of the critical current density under increased magnetic field [10] and the surface-sensitive character of MgB$_2$ films due possibly to the existence of a Mg-rich surface layer for films prepared by the two step process [11]. The possibility of two gaps in MgB$_2$, with the small gap apparently responsible for the $R_S$ of MgB$_2$ at low temperatures, also creates difficulty in optimizing the microwave properties of MgB$_2$ [12].

In this paper, we report effects of surface ion milling on the microwave properties of high-quality MgB$_2$ films, both in the normal state and in the superconducting state. The intrinsic surface resistance ($R_S$) of MgB$_2$ films prepared under optimized conditions was less than 10 $\mu\Omega$ at 8.5 GHz and 7 K, and ~1.5 m$\Omega$ at 87 GHz and 4.2 K, which was comparable to the corresponding typical $R_S$ values of epitaxially grown YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) films [13]. The changes in the surface resistance due to ion-milling are well explained by the two-gap model for MgB$_2$.

II. EXPERIMENTAL

The MgB$_2$ films were prepared on c-cut sapphire, MgO and LaAlO$_3$ by the two step process where boron films deposited on the substrates are annealed in a Mg vapor environment inside quartz tubes [9]. The films used for this experiment fall into three different groups. Films in group I (MB-1 to 4) were prepared in an earlier stage of this study and appeared to have relatively high $R_S$. The films in group II (MB-12 to 14) were surface ion-milled immediately after being prepared, and those in group III (MB-15 to 20) were prepared using an improved growth technique. In preparing the ion-milled films, the surface of the MgB$_2$ film was etched by argon ion-milling under an angle of 70 degree with respect to the film plane. The etching rate was ~10 nm/min with a beam voltage of 500 V and a current density of 0.28 mA/cm$^2$.

Properties of the MgB$_2$ films including the film thickness ($t$),...
the normal state. The normal-state resistivity $R_{S}^{n}$ in intrinsic surface resistance

effective values ($R_{S}^{eff}$) was reproducible within 5% with errors in

assumed normal skin effect for MgB$_2$ in

taken into account and assuming normal skin effect for MgB$_2$ in

appeared to depend on the measurement frequency.

Assuming a penetration depth of ~160 nm, the corresponding

where $T_c$ is the critical temperature determined at high frequency

Microwave properties were measured at ~8.5 GHz and ~87 GHz using a TE$_{01}$

Loaded quality factor measured in a weak-coupling scheme.

Figure 1 shows the temperature dependence of the effective

$N_{eff}$, $\rho(T_c)$ was used for this purpose. The normal-state intrinsic surface resistance

and 45 K. The temperature was stable within ±0.15 K. The effective surface resistance $R_{S}^{eff}$ was calculated using both the impedance transformation method [14] and a method based on

unloaded quality factor measured in a weak-coupling scheme.

Changes in the microwave properties were significant for all

Figure 2 and its inset shows the $R_S$ vs. frequency data for MB

Changes in the microwave properties were significant for all

$\rho(T_c)(HF)$ was accompanied by an increase in $\rho(T_c)(HF)$ after ion-
milling. For most films, the ratio of $\Delta T_C$(HF) to $\rho(T_C)$(HF) is ~ 1 K/µΩ-cm. The increase of $\rho(T_C)$(HF) after ion milling is very likely due to the increased defect density created during the ion milling process and the resulting increase of the scattering rate of electrons. We note that a similar correlation between $T_C$ and $\rho(T_C)$ has been observed for A15 compounds and bcc transition metals, which is attributed to electron lifetime effects by Testardi and Mattheiss [16]. Although $\rho(T_C)$(HF) and $\rho_0$ cannot be directly compared with each other, it is very likely that increased defect density in MgB$_2$ films would result in an increase in $\rho_0$ as well. For reference, $\Delta T_C/\rho_0$ is 0.1 -- 0.2 K/µΩ-cm for A15 compounds and bcc transition metals with low $\rho_0$.

When the MgB$_2$ films were ion-milled twice, the correlation between $T_C$(HF) and $\rho(T_C)$(HF) was not clear. In Fig. 3(b), we show the data for MB 11A, MB 12I-14I (films ion-milled one time) and MB 12II-14II (films ion-milled twice) measured at 8.5 GHz and 87 GHz. The data at 87 GHz still show some correlation between $T_C$(HF) and $\rho(T_C)$(HF) for MB 13 and MB 14. However, at 8.5 K, both $T_C$(HF) and $\rho(T_C)$(HF) decrease after ion-milling. The difference in the $T_C$(HF) vs. $\rho(T_C)$(HF) behavior for MB 13 and MB 14 between the data at 8.5 GHz and 87 GHz is not understood at this time.

In Figs. 3(a) and 3(b), differences in $T_C$(HF) and $\rho(T_C)$(HF) are seen between the data at 8.5 GHz and 87 GHz. For the films in groups II and III, the measured $T_C$(HF) values at 87 K are consistently lower than the corresponding ones at 8.5 GHz. Within the context of the two-gap scenario for MgB$_2$, the observed difference in the $T_C$(HF) values may be related to the presence of the small energy gap, since 87 GHz is about 10 % of the gap frequency for $\Delta k_0 T_C$ ~ 1. There are two exceptions to this general observation regarding $T_C$(HF); the $T_C$(HF) values of MB13I and MB14I at 87 GHz are a little higher than the corresponding ones at 8.5 GHz, respectively. However, the observed small differences in $T_C$(HF) for MB13I and 14I may be within the measurement errors for determining $T_C$(HF).

In Figs. 3(a) and 3(b), it is seen that the $\rho(T_C)$(HF) of the films in groups II and III is in the range of 10 - 20 µΩ-cm regardless of the ion-milling process. In particular, the $\rho(T_C)$(HF) values of the as-prepared films with the lowest $R_{eff}$ values (e.g., MB 15A, 16A, 18A and 20A) are in the range of 12 -- 15 µΩ-cm. These values are still significantly higher than the values of $\Delta k_0 T_C$ ~ 1 -- 5 µΩ-cm, which have been frequently reported for single crystal MgB$_2$ and bulk polycrystalline MgB$_2$. The difference in the $\rho(T_C)$(HF) values at 8.5 GHz and 87 GHz can be understood in terms of the measurement errors. In most cases except for MB-13I, the differences in the measured $\rho(T_C)$(HF) values at 8.5 GHz and 87 GHz are within 10 %, which can be explained assuming an measurement error of 5 %.
near $T_C$ (HF), a plausible number considering that the resonator coupling becomes drastically weak near $T_C$ (HF).

Very interesting features were observed in the temperature-dependent $R_{\sigma}^{HF}$ data for the as-prepared and ion-milled MgB$_2$ films. Figure 4 shows the $R_{\sigma}^{HF}$ vs. the reduced temperature ($T/T_C$) data for MB-19A, 19I, 20A and 20I measured at 8.5 GHz and 87 GHz. In the figure, we see a crossover in the $R_{\sigma}^{HF}$ vs. $T/T_C$ (HF) curves for the as-grown and the ion-milled MgB$_2$ films with the $R_{\sigma}^{HF}$ of the ion-milled films becoming smaller than that of the as-grown films despite the degradation of $T_C$ (HF) after ion-milling. It is noted that the crossover temperature appears almost the same at 8.5 GHz and 87 GHz, occurring at ~0.85 for MB 19A and 19I, and ~0.6 for MB 20A and 20I. Such a crossover can be well understood within the context of the two-gap scenario along with the reduction in $T_C$ (HF). First, $T_C$ (HF) cannot be affected by intraband scattering according to Anderson’s theorem. In this regard, it is believed that the reduction in the $T_C$ (HF) of the as-grown MgB$_2$ after ion-milling is attributed to the increased interband scattering between the $\sigma$-band and the $\pi$-band in MgB$_2$. Since the $\rho_0$ values would be dominated by the properties of the small gap at low temperatures, the reduced $R_{\sigma}$ of the ion-milled MgB$_2$ at low temperatures would mean an increased gap energy of the small gap due to the increased interband scattering by ion-milling-induced defects. No correlation was found between $\rho(T_C)(HF)$ and the $R_{\sigma}$ measured at low temperatures. Within this model, the $\rho(T_C)(HF)$-$T_C(HF)$ relation is attributed to the properties of the large gap, i.e., the $\sigma$-band, with the $R_{\sigma}$ at lower temperatures reflecting the properties of the small gap, i.e., the $\pi$-band when the uncorrelated $\rho(T_C)(HF)$-$R_{\sigma}$ behavior is considered.

Our arguments are consistent with the recent report by Mazin et al. [17]. According to them, although the two-band model predicts lower $T_C$ for MgB$_2$ samples with higher $\rho_0$, the extremely small interband scattering in MgB$_2$ enables many MgB$_2$ samples to have similar $T_C$ despite having a distribution in $\rho_0$. If MgB$_2$ samples can be made with enhanced interband scattering, lower $T_C$ should correlate with higher $\rho_0$. We therefore attribute the reduced $T_C$ (HF) we observe due to ion-milling as due to enhanced interband scattering.

It is also noted that for MgB$_2$ films, repeated ion-milling does not always yield reduced $R_{\sigma}$ at low temperatures. In the inset of Fig. 4, the $R_{\sigma}^{HF}$ vs. $T/T_C$ data for MB 12I and 12 II show a reduction in the $T_C$ and $R_{\sigma}$ of MB 12 I after ion-milling as observed for films in group III. Similar behavior was observed for MB-4A and 4I. However, when MB-4I was ion-milled again, its $R_{\sigma}^{HF}$ appeared significantly enhanced, with a reduction in $R_{\sigma}^{HF}$ (see the inset of Fig. 4). This suggests that reduction in $R_{\sigma}$ at low temperatures only be realized when modest disorder is introduced in MgB$_2$. Similar observations have been made in proton-irradiated bulk MgB$_2$ by Bugoslavsky et al. [18], who reported enhancement of the high-magnetic field critical current density of bulk MgB$_2$ by proton irradiation. In this regard, excessive disorder might have played some roles in enhancement of $R_{\sigma}$ for ion-milled MgB$_2$ films [5] and reduction of the critical current density for electron-irradiated bulk MgB$_2$ at high magnetic field [19].

For films not prepared under optimized conditions, a Mg-rich metallic layer could affect their $\rho_0$ values significantly [11]. Such films usually show significantly lower $R_{\sigma}^{HF}$ (see e.g., Table I for the $R_{\sigma}^{HF}$ values for MB 2 - 4 in group I) compared to the values observed for films in group II and III. For these films, $T_C$ also appeared to change little after the ion-milling.

A similar crossover behavior has been reported in the $R_{\sigma}$ vs. temperature curves for YBCO films with different defects densities [20], with the crossover attributed to the behavior of $\sigma_i$, the real part of complex conductivity, due to the temperature-dependent inelastic scattering rate and the quasiparticle density. To date, however, the $\rho(T_C)(HF)$-$T_C(HF)$ relation and the $\rho(T_C)(HF)$-$R_{\sigma}$ relation are not known for YBCO films, which makes it difficult to compare the properties of YBCO with those of MgB$_2$.

Our interpretations of the changes in the properties of MgB$_2$ films are also consistent with our recent observations for the nonlinear behavior of as-prepared and ion-milled films. At relatively high temperatures, the ion-milled films showed higher nonlinear response. However, as the temperature decreases, a crossover was observed between the nonlinear response of the as-prepared and the ion-milled films, resulting in the observation of lower nonlinear response from the ion-milled films. This observation also fits the model of the small gap becoming enhanced due to the ion-milling induced interband scattering. More details on the nonlinear responses of MgB$_2$ films will be reported elsewhere [21].
IV. CONCLUSION

High-quality MgB2 films with Rs less than 10 µΩ at 8.5 GHz and 7 K, and ~ 1.5 mΩ at 87 GHz and 4.2 K, with the critical temperature (Tc) of ~ 39 K have been prepared, and effect of ion-milling on their microwave properties were studied. After ion-milling, reduction of the Tc and enhancement of the normal-state resistivity were consistently observed along with a reduction of Rs at low temperatures. The intrinsic Rs appeared to scale with F ion up to 30 K. The observed ρ(Tc)(HF) - Tc(HF) relation and the uncorrelated behavior between ρ(Tc)(HF) and Rs measured at low temperatures are well explained within the context of the two-gap model, with the observed ρ(Tc)(HF) - Tc(HF) relation attributed to the properties of the large gap, i.e., the π-band, and the Rs at lower temperatures reflecting the properties of the small gap, i.e., the π-band, with the small gap energy enhanced by increased interband scattering. In this regard, finding ways to increase the interband scattering seems to be one of the key means to improve the low temperature microwave properties of MgB2, which is dominated by the small gap.

Our results also show that ion-milling, which is frequently used for passivation or planarization for thin films, affects the superconducting properties of MgB2 films significantly and great care needs to be taken for the properties of MgB2 films to be preserved when any processes involving ion-milling are performed.

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