Microwave Properties of MgB_2 Thin Films Grown by Reactive Evaporation

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***Abstract*—We have grown MgB_2 films using the deposition technique of reactive evaporation. This method allows high-quality, large-area, double-sided films to be grown on a large variety of substrate materials. These films are therefore well suited for applications, several of which may exploit the low-loss high-frequency properties of MgB_2, including passive microwave resonators and filters, nuclear magnetic resonance coils, and RF cavities for particle accelerators. Determining the surface resistance and nonlinear properties of our films can thus give us an idea of their suitability for these applications, as well as tell us something about their fundamental superconducting nature. We have begun measuring the microwave properties of our films using both a parallel plate technique and by patterning the films into stripline resonators and lumped-element resonators and filters. The films display very low surface resistance values.**

***Index Terms*—Intermodulation distortion, magnesium diboride, microwave properties, superconducting materials growth, thin films.**

I. INTRODUCTION

THE RECENTLY discovered medium-temperature superconductor MgB_2 [1] offers promise for potential advantages over both low-temperature (LTS) and high-temperature superconductors (HTS), depending on the application. The higher T_c of MgB_2 vs. LTS materials is clearly advantageous, opening the door for the use of smaller and less expensive closed-cycle cryocoolers. The far less complex materials difficulties of MgB_2 make up its key possible advantage over HTS materials. The path to superconducting digital electronics operating at 30 K, for example, may now be possible. Moreover, MgB_2 may offer cost and manufacturability advantages over HTS materials, though these would have to offset the cost of larger and more complex coolers. In addition, it is possible that MgB_2 offers superior performance over either LTS or HTS materials for the same operating temperature.

In addition to digital electronics, detectors, and current-carrying applications, there are several technologies that rely on the very low loss of superconductors at high frequencies. Indeed, passive microwave devices presently form the largest market for HTS thin films. Though MgB_2 operates at considerably lower temperatures than HTS materials, we nonetheless would like to investigate its suitability for RF applications and determine whether it might display superior properties in terms of either surface resistance or nonlinear performance. There is also significant interest in fabricating RF cavities of MgB_2 for accelerator applications, thereby relieving the heavy burden of low-temperature operation. And since NMR coils utilizing HTS thin films already operate at temperatures below the T_c of MgB_2, the potential exists for using MgB_2 for this application, particularly if cost or performance advantages can be gained, either through inherently superior properties or the possibility of enhanced geometries or 3D structures.

Despite the initial difficulties of depositing completely in-situ, high quality MgB_2 films on large-area substrate materials suitable for applications, we have recently developed a technique for doing so, and thus several of the potential thin-film applications listed above may become realizable.

In this paper we briefly review the growth of our MgB_2 films, and we discuss the initial measurements of their microwave properties. We have measured the surface resistance of our films by different methods, including a parallel plate technique and by patterning stripline resonator structures. Because our growth technique allows the formation of large-area, double-sided films, we can also fabricate microwave resonators and filters in a manner similar to that used for the YBCO thin-film filters developed for commercial wireless applications.

II. FILM GROWTH

Our MgB_2 films were deposited by a reactive evaporation technique. Details of this method and the properties of our films are discussed elsewhere [2], but we will briefly review them here.

The growth of high-quality MgB_2 films has been relatively difficult due primarily to the oxygen sensitivity of Mg and to the high pressure of Mg needed during growth of MgB_2 at high temperatures. However, using the growth technique of reactive evaporation in which a localized source of Mg vapor is maintained near the substrate by using a rotating pocket heater, we have been able to grow MgB_2 films completely in situ. Moreover, our technique allows growth of high-quality films on virtually any single-crystal substrate for which there is not a chemical reaction with Mg, B, or MgB_2, including MgO, SiC, $SrTiO_3$, $LaAlO_3$, $NdGaO_3$, sapphire, YSZ, and $LaGaO_3$: a well lattice-matched substrate is not required. Moreover, we have also grown excellent films on Si buffered with a variety of intermediate layers, on polycrystalline alumina, and on flexible, unpolished stainless steel shim stock. Our present heater size allows growth on substrates up to 4″ in diameter, and we have deposited

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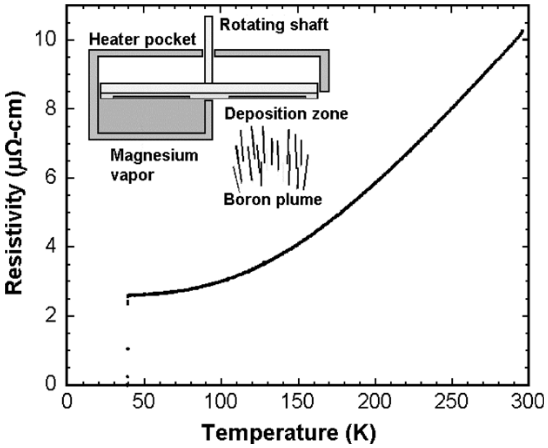

Fig. 1. $R - T$ curve of a 500-nm-thick $\mathrm{MgB_2}$ thin film deposited onto r-plane sapphire. The T_c is 39.1 K. Note the low room-temperature resistivity and residual resistivity. The pocket heater used for the reactive evaporation process is shown in the inset.

high-quality films on 4″ r-plane sapphire and on 4″ Si buffered with $\mathrm{Si_3N_4}$. Since our heater does not contact the surface of the substrates, we are able to deposit double-sided films by flipping the substrates over following deposition and growing on the back side in a subsequent run. No degradation of the front $\mathrm{MgB_2}$ surface has been observed. This is crucial for applications requiring a superconducting ground plane, such as for certain passive microwave devices.

Our technique allows $\mathrm{MgB_2}$ film growth in an intermediate growth temperature range of 400 to 600°C. This range is generally inaccessible by other methods such as MBE [3]–[5] for which the growth T must be less than about 300°C due to the high Mg vapor pressure, and HPCVD [6] for which a temperature over 700°C is desirable in order to evaporate sufficient Mg and to obtain epitaxial growth. Our films regularly display T_c values of 38 to 39 K and above, and their room-temperature resistivity values and residual resistivity values are among the lowest reported for thin films. The films are smooth, textured, and have a columnar grain structure. The films are also very clean as exhibited by their low microwave loss, low H_{c2} values [7], and a lack of oxygen or carbon contamination as determined by RBS [8]. In addition, the films are relatively stable with respect to patterning and upon exposure to water. The films described in this paper were grown at 550°C and are about 500 nm thick. Fig. 1 displays a representative $R-T$ curve for an $\mathrm{MgB_2}$ film grown on r-plane sapphire by reactive evaporation. A schematic of our heater is shown in the inset.

III. PARALLEL PLATE MEASUREMENTS

The first microwave measurements of our films were made using a parallel plate configuration. The details of this technique have been described previously [9]. The measurements were made using two 1 cm × 1 cm samples which were diced from a 2″ wafer of $\mathrm{MgB_2}$ grown on r-plane sapphire. The T_c of these samples was measured to be over 39 K. Fig. 2 shows the microwave surface resistance vs. temperature for these two $\mathrm{MgB_2}$ plates. The measurements were made at 10 GHz. The data shown in the figure *include* extrinsic coupling and radiation losses in the system; the range of these losses is indicated. Note that the microwave loss of our $\mathrm{MgB_2}$ film is significantly better than that measured for a high-density bulk $\mathrm{MgB_2}$ sample [10], which is also shown in the figure for comparison. Above a crossover temperature of 3.3 K, the loss of our $\mathrm{MgB_2}$ sample is also superior to a high-quality Nb thin film.

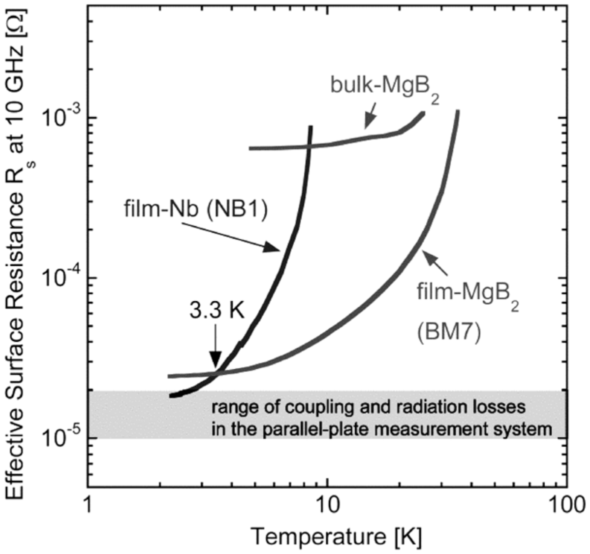

Fig. 2. R_s at 10 GHz vs. T measured by a parallel plate technique for $\mathrm{MgB_2}$ thin films deposited on r-plane sapphire. For comparison are a bulk $\mathrm{MgB_2}$ sample and a high-quality Nb film. These data include external coupling and radiation losses.

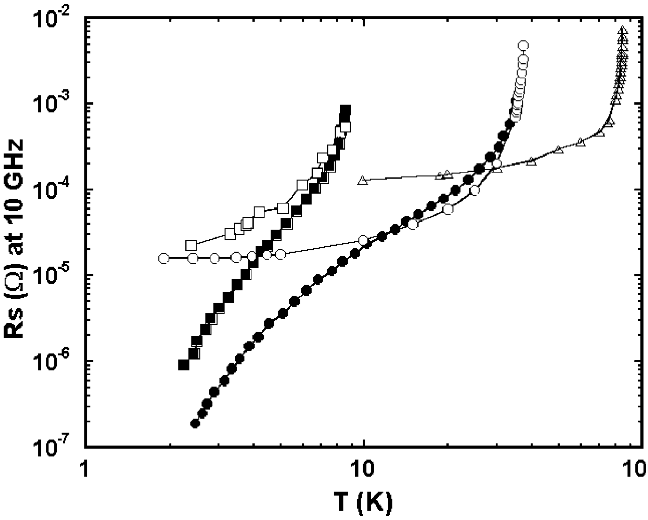

Fig. 3. R_s vs. T scaled to 10 GHz for our $\mathrm{MgB_2}$ films on sapphire (○), a YBCO film on LAO (△), and for Nb thin films (□). Filled symbols indicate parallel plate measurements with external losses subtracted, and open symbols indicate stripline resonator measurements including external losses.

We have also determined and subtracted the extrinsic losses from our measurements, and the results are shown in the plot of Fig. 3, which includes the stripline resonator measurements discussed in the next section. From this figure, it is clear that the intrinsic microwave loss of $\mathrm{MgB_2}$ is lower than Nb at all temperatures. Indeed, to our knowledge the microwave loss of our films is lower than any $\mathrm{MgB_2}$ results reported in the literature.

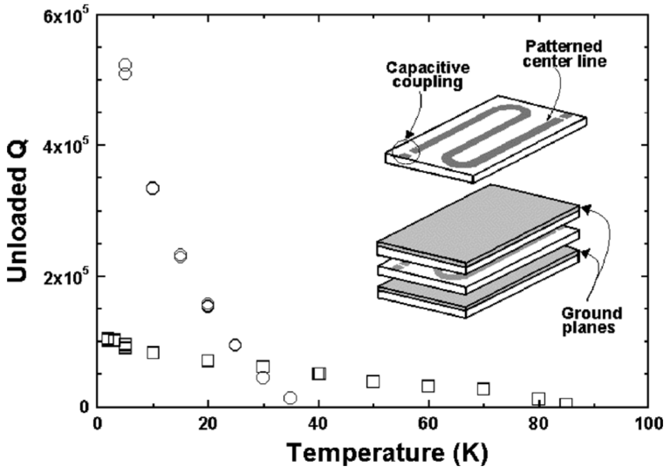

Fig. 4. Unloaded Q vs. temperature for an $\mathrm{MgB_2}$ film (○) and for a YBCO film (□). The measurements were made at 2.3 GHz using a stripline resonator structure as shown in the inset.

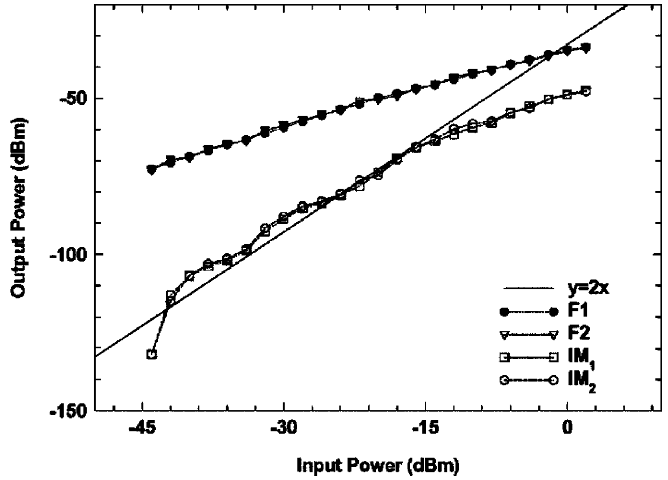

Fig. 5. Two-tone IMD measurement of an $\mathrm{MgB_2}$ film on r-plane sapphire using the stripline resonator technique described in the text.

IV. STRIPLINE RESONATOR MEASUREMENTS

We have also performed surface resistance and intermodulation distortion (IMD) measurements on our films by patterning them into stripline resonators. As for the parallel plate measurements, these were made on $\mathrm{MgB_2}$ films deposited on 2″ r-plane sapphire substrates.

A. Surface Impedance Measurements

The films were patterned using standard photolithography and ion beam etching, and the wafer was diced into 1 cm × 1 cm pieces, which provided resonator meander lines and ground planes. A meander line and two ground planes were assembled to form a stripline resonator and the nonlinear surface impedance was measured by a technique that has been described previously [11], [12], in which the Q and resonant frequency f_0 of the resonator are measured as a function of the microwave power at various temperatures between 1.7 K and T_c. The results are converted into the effective surface resistance $R_s(I_{\mathrm{rf}})$ and changes in reactance $\Delta X_S(I_{\mathrm{rf}})$, where I_{rf} is the microwave current. The measurements were done at the fundamental frequency of 2.3 GHz. After patterning, the T_c of this film was measured to be 39 K, and the extracted penetration depth was $\lambda_L(0) = 100\ \mathrm{nm}$. This value is consistent with the smaller of the two $\mathrm{MgB_2}$ gaps, as seen in other microwave measurements [13].

Fig. 4 shows the unloaded Q as a function of T for the patterned structure shown in the inset. For comparison, a high-quality YBCO film grown on 2″ $\mathrm{LaAlO_3}$ is also shown. Note the clear crossover to a higher loaded Q for $\mathrm{MgB_2}$ below about 30 K.

Surface resistance numbers were extracted from these results. These data and also an additional stripline resonator measurement of a Nb film have been scaled to a frequency 10 GHz by assuming a quadratic dependence on frequency, which has been shown to hold for $\mathrm{MgB_2}$ [14]. The results are plotted in Fig. 3 along with the parallel plate measurements discussed above. Note that the results measured by these two techniques agree rather well. The lower values of the parallel plate measurement below 10 K reflect the subtraction of extrinsic losses in that measurement. The stripline resonator measurements, on the contrary, include any additional losses in that system. Also note that the two independent measurements of different Nb films agree rather well, and that the results for our $\mathrm{MgB_2}$ films are clearly superior to these high-quality Nb films at all temperatures. Lastly, the YBCO thin film result is also included, highlighting the superior microwave loss of our $\mathrm{MgB_2}$ films below about 30 K.

We have also measured the power dependence of R_s for these stripline resonators. The R_s follows a roughly linear dependence on input power. This result is rather surprising, since it is reminiscent of poor YBCO behavior and is indicative of an extrinsic source of microwave loss. Since the grain boundaries in $\mathrm{MgB_2}$ are not known to be weak links, and our films appear to be free of oxygen incorporation, it is unclear what this additional source of loss might be, but it provides encouragement that the loss of these films may be reduced even further with additional materials development.

B. Intermodulation Distortion Measurement

The same resonator used for the Z_S measurements is used for the IMD measurements. The third-order IMD was measured in the usual way, in which two closely spaced tones of equal power at frequencies f_1 and f_2 are combined and applied to the resonator. The frequencies are centered about the resonant frequency with a tone separation of approximately 1/32 of the low-power 3-dB bandwidth. The tone separation was adjusted at each temperature and input power to maintain the same relationship to the bandwidth. The third-order mixing products at frequencies $2f_1 - f_2$ and $2f_2 - \mathrm{f1}$ are then measured in a spectrum analyzer as a function of the input power to the resonator. At each input power, the frequencies were adjusted to the peak of the resonance. This step is necessary because of the power dependence of the resonance due to the nonlinear reactance of the films.

These IMD results were measured at 20 K and are shown in Fig. 5. The figure shows the output power at the input tones and the IMD third order products as a function of the input power. The IMD's shown here are in fact rather high; we report better

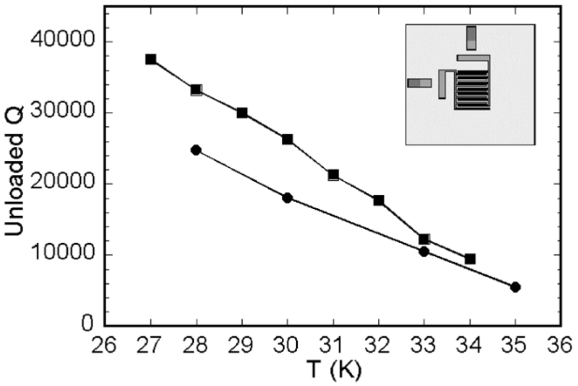

Fig. 6. Unloaded Q for two lumped-element $\mathrm{MgB_2}$ thin film resonators on sapphire with an $\mathrm{MgB_2}$ ground plane. The resonator geometry is shown in the inset. The center frequency is 1.86 GHz.

results below for our microwave filters. The slope of the IMD's vs. input power is ~2, so that the third order signal is proportional to the square of the input signal. This behavior is similar to that observed for HTS materials [15] and is also consistent with the linear $R_s(I_{rf})$ response noted above. This result may imply a nonlinear inductance which varies linearly with the magnitude of the input current, $\Delta L \sim |I|$. Whether this is a consequence of $\Delta\lambda_L \sim |J|$ or is due to extrinsic effects is unknown.

V. LUMPED ELEMENT RESONATOR AND 10-POLE FILTER MEASUREMENTS

Due to the encouraging R_s measurements discussed above and our ability to deposit large-area, double-sided $\mathrm{MgB_2}$ films, we have fabricated microwave resonators and filters in order to evaluate their suitability for use in microwave applications. These structures were formed using standard photolithographic techniques and inert Ar ion etching.

A. Lumped Element Microwave Resonator

We have patterned double-sided $\mathrm{MgB_2}$ films grown on r-plane sapphire using a standard test resonator design that we have used for several years to evaluate the Q of our YBCO films. This resonator has dimensions of about 0.5 × 0.5 cm and has the geometry shown in the inset of Fig. 6. The temperature dependence of the unloaded Q for the first two resonators we have measured is shown in Fig. 6. The temperature of 27 K is the lowest we could reach in the single-stage cryocooler used for this measurement. In future we plan to go to lower temperatures using liquid He. The resonant frequency of this resonator was 1.86 GHz at this temperature, and the frequency dependence of f_0 was about 1 MHz/K. The measured Q values increase with decreasing T as expected, and are already sufficiently high for commercial applications. For comparison, our best YBCO films that are 40% thicker than the $\mathrm{MgB_2}$ films measured here give unloaded Q values of 50 000 at 77 K for this resonator. So the value of 40 000 at about 27 K for our $\mathrm{MgB_2}$ films is quite good by comparison, particularly since it took us several years of development to optimize our YBCO films. Indeed, based on the R_s numbers presented in Fig. 3, we appear to be on the way to exceeding the best YBCO numbers at low temperatures.

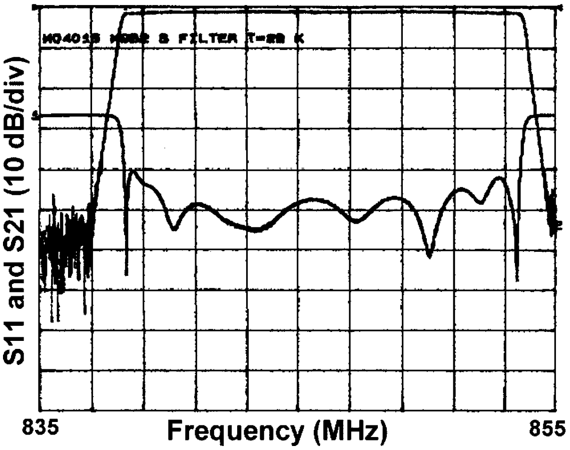

Fig. 7. Response of an untuned 10-pole $\mathrm{MgB_2}$ thin film bandpass filter on an MgO substrate with an $\mathrm{MgB_2}$ groundplane. $\mathrm{S_{21}}$ is offset for clarity.

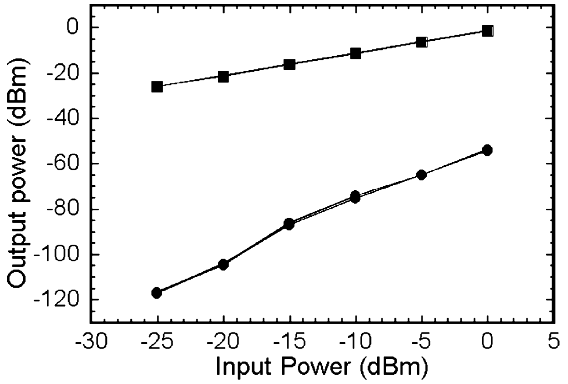

Fig. 8. Two-tone IMD measurement of a 10-pole $\mathrm{MgB_2}$ thin film filter on MgO with an $\mathrm{MgB_2}$ ground plane. The measurement was made at 30 K. The figure plots output power at the input tone frequencies f_1 and f_2 (■) and the third order IMD products (●).

B. 10-Pole Bandpass Microwave Filter

In addition to single resonators, we have also patterned complete 10-pole bandpass microwave filters with circuits similar to those used for our commercial YBCO microwave filters. These filters were patterned using 2″ $\mathrm{MgB_2}$ films deposited on MgO substrates with $\mathrm{MgB_2}$ ground planes deposited on the back side. Again, our low temperature excursion was limited by our cryocooler, but in Fig. 7 we show the filter response measured at 29 K. This filter is completely *untuned*, so the excellent response shown in this figure is quite promising.

C. Intermediation Distortion Measurements

We also made two-tone IMD measurements on this 10-pole filter. The equal-power input tones were spaced 30 kHz apart and were placed near the center of the passband. The third-order mixing products were measured as a function of the input power. The IMD measurements were made at 30 K, and the results are shown in Fig. 8. These IMD data appear to be significantly better than those shown in Fig. 5 for the stripline resonators measured at the lower temperature of 20 K. A direct comparison is difficult, however, because the input couplings and Q values of these resonators are different. The slope of the IMD's in Fig. 8 is consistent with Fig. 5, though, being equal to about 2.5. This is again similar to HTS and to $\mathrm{MgB_2}$ IMD behavior observed by others [16]. Though the model of Josephson flux motion in grain boundaries [17] has been widely used to describe this behavior in HTS materials, it is not clear whether this applies to $\mathrm{MgB_2}$, since the grain boundaries in this material are not described as weak links [18].

VI. DISCUSSION AND CONCLUSION

Our reactive evaporation growth technique allows the growth of large-area, double-sided $\mathrm{MgB_2}$ films on a multitude of substrate materials. These films are therefore ideally suited for applications. We have measured the high-frequency R_s of our films by independent techniques, both of which provide values that are among the very lowest reported for $\mathrm{MgB_2}$ thin films. This is encouraging at this early stage of development, and is likely a testament to the far less severe materials complexities of $\mathrm{MgB_2}$ in comparison to the HTS compounds. We have therefore also made fabricated microwave resonators and filters from our double-sided films on both MgO and r-plane sapphire substrates. The low R_s values are reflected in the high Q values of the resonators and excellent response of a 10-pole bandpass filter of a type useful for commercial applications.

The nonlinearities of our films as reflected in the two-tone IMD measurements of a stripline resonator and 10-pole filter are more sobering though by no means discouraging. Indeed, we are hopeful that with further materials development the nonlinearities can be improved, as has been markedly demonstrated for the HTS materials. It does appear that the small π band gap of the two-band $\mathrm{MgB_2}$ materials may be responsible for larger intrinsic nonlinearities at low temperature compared to a single-gap s-wave superconductor [19]. However, it has also been suggested that clean $\mathrm{MgB_2}$ films such as ours may be expected to have worse nonlinearities than films that are doped or contain other sufficient disorder to provide a higher scattering rate in the π band relative to the σ band [17]. There is therefore hope that such strategies can lead to optimized properties. Doping with C has, for example, already been used to achieve very high H_{c2} values in clean $\mathrm{MgB_2}$ films [7]. We also note that measurements of the nonlinear critical current density J_0 [20] of our $\mathrm{MgB_2}$ films by a mutual inductance technique indicate that very high J_0 values are possible, above $10^7\ \mathrm{A/cm^2}$ even at 35 K [21].

Given the excellent Q values and performance of our test structures along with the possibility for improvement of the nonlinear properties, we are optimistic about the use of these $\mathrm{MgB_2}$ films for applications. This is particularly true since our technique allows a relative ease of fabrication in comparison to HTS films.

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