Final technical report

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Title of the Grant: Magnesium Diboride thin Films, multilayers, and coatings for SRF cavities
Period of Performance: April 1, 2014 to March 31, 2017
PI's Name: Xiaoxing Xi

Project Scope

The objective of this project is to develop a MgB_2 superconducting RF (SRF) cavity technology. Compared to the currently-used SRF material niobium, MgB_2 has a much higher T_c of 40 K, a lower residual resistivity (< 0.1 μΩcm), and a higher thermodynamic critical field H_c. SRF cavities with MgB_2 coatings have the potentials for higher Q, higher gradient, and higher operation temperatures. A MgB_2 SRF technology can significantly reduce the operating costs of particle accelerators when these potentials are realized.

The project used the HPCVD technique to deposition MgB_2 thin films, multilayers, and coatings to investigate their applications in SRF cavities. The research activities included (1) fundamental materials physics of multilayer structures and their effects in enhancing the lower critical field H_{c1} and the vortex penetration field, and (2) the coating of SRF cavities. Coating of 3.9 GHz mock cavities has been carried out, which is the largest size we can coat in the existing deposition system.

Results from the Project

1. Enhancement of H_{c1} in thin MgB_2 films

It has been shown that H_{c1} of a type II superconductor (H_{c1} of Nb ~ 1700 Oe) plays an important role in determining the RF breakdown field.[1] Gurevich proposed[2] that a coating of thin superconductor layer less than its London penetration depth λ can impede the vortex penetration. In this project, we have investigated whether the enhancement of H_{c1} is attainable in both epitaxial and polycrystalline MgB_2 films because coatings on cavity walls are inevitably polycrystalline.

We grew the MgB_2 films by HPCVD on SiC (0001) substrate for the epitaxial films. For the polycrystalline MgB_2 films, an MgO layer was deposited first at room temperature on the SiC substrate by reactive DC magnetron sputtering prior to the deposition of the MgB_2 film. Both x-ray diffraction (XRD) and cross-sectional transmission electron microscopy (TEM) confirm that the films on the MgO layer were polycrystalline.[3]

To measure H_{c1}, a vertical magnetic field was applied parallel to the film's surface and the magnetic moment m was measured. When the applied field is below H_{c1} of a superconductor thin film, there is no vortex in the film and a linear dependence of m on H should be observed. Above H_{c1}, vortices enter the superconductor and m deviates from the linear dependence on H. Our results showed that for the epitaxial films, a 300 nm thick film had a low H_{c1} about 600 Oe at 5 K, similar to bulk MgB_2 samples reported in the literature. H_{c1} increases with decreasing film thickness, reaching 1880 Oe when the thickness is 100 nm. For the polycrystalline films grown on MgO buffer layer, the same thickness dependence trend as the epitaxial films was observed. The H_{c1} values are 520 Oe at 5 K for the 300 nm film and 1520 Oe for the 100 nm film. The thin polycrystalline MgB_2 films should have similar abilities to prevent vortex entry as in clean, epitaxial films.

In Fig. 1, H_{c1}(5 K) as a function of MgB_2 film thickness from 100 nm to 300 nm for (a) epitaxial and (b) polycrystalline films are plotted. In both cases, H_{c1} increases with decreasing film thickness to values similar to those of bulk Nb. The solid lines in the figures are theoretical fitting applicable to film thickness greater than λ, based on the free energy of a single vortex in a thin film.[4] The film to bulk ratio of H_{c1} depends exclusively on the ratio of the film thickness to the penetration depth d/λ. Taking both bulk H_{c1} and λ as fitting parameters, very good fittings are obtained with H_{c1}(bulk) = 610 Oe and λ = 51 nm for the epitaxial films and H_{c1}(bulk) = 514 Oe and λ = 54 nm for the polycrystalline films.[3] The H_{c1}(bulk) obtained are within the range of values reported in the literature. The λ values are slightly higher than the previous measurements on our clean film samples.[5]

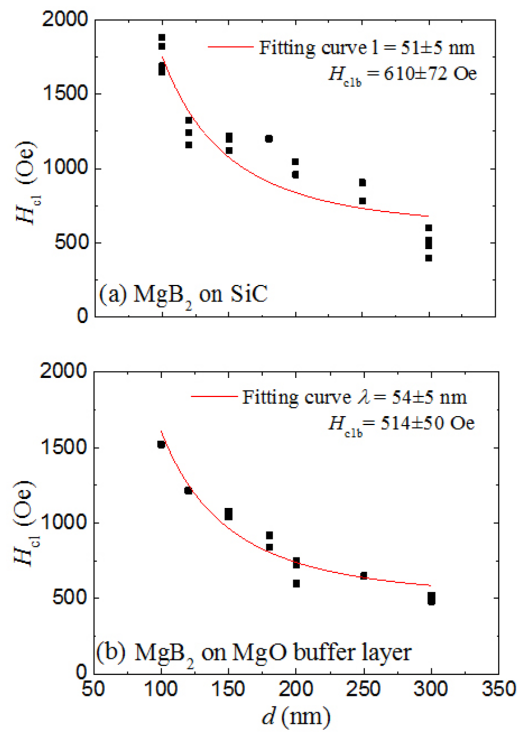

Fig. 1 Thickness dependence of H_{c1} (5 K) of (a) epitaxial and (b) polycrystalline MgB_2 films. The solid lines are theoretical fits.

The result demonstrates that the approach of preventing vortex entry, thus enhancing the acceleration gradient, by thin superconducting films can be realized using polycrystalline MgB_2 coatings on SRF cavities and in S-I-S multilayer structures.

The result is from a collaboration with scientists at Los Alamos National Laboratory and Drexel University, and has been published in *APL Mater*. 3, 041101 (2015).

2. Enhancement of H_{vp} in MgB_2-coated Nb

To test Gurevich's idea of coating the inside of Nb cavities with thin superconducting films to enhance the vortex penetration field (H_{vp}),[2] one has to consider the situation when the field is only applied on one side of the film, which is different from the case when H is applied on both sides of a film. The concept that a thin film can sustain a ΔH between both of its surfaces cannot be tested by measuring a thin film on a flat substrate because both surfaces of the film are exposed to the same ambient field. To overcome this fundamental limitation, and concomitantly also solves the resolution and alignment problems in H_{vp} measurements, in this project we fabricated macroscopic-sized ellipsoids and coated them with MgB_2 films.[6] In Fig. 2, photographs of a bare Nb ellipsoid (Nb0) and a Nb ellipsoid coated with 200 nm MgB_2 (Nb200) are shown. The Meissner effect in the film screens the entire ellipsoid volume from the applied field, so a ΔH develops between the internal and external film surfaces. Here the field is outside of the ellipsoid rather than inside as in a real SRF cavity, thus we call it the "inverse cavity" concept. We use ellipsoids because the demagnetizing effects and boundary conditions are well defined, and in the Meissner state the external field at the ellipsoid surface is parallel to the surface everywhere, the same as in real SRF cavities.

Figure 2(c) summarizes the $H_{vp}(T)$ curves for the bare Nb ellipsoid, Nb100 (coated with 100 nm MgB_2), and Nb200. The $H_{vp}(T)$ of Nb200 is about 400 Oe higher than the bare Nb ellipsoid in the temperature range from 1.8 K to the T_c of Nb. This increase is approximately constant because the properties of MgB_2 do not change significantly in this temperature range ($T < T_c/4$). The maximum field at the equator that our inverse cavity can sustain before vortex penetration is indicated on the right vertical axis. At $T \sim 2.8$ K it is ~2600 Oe, about 500 Oe above the bare Nb ellipsoid.

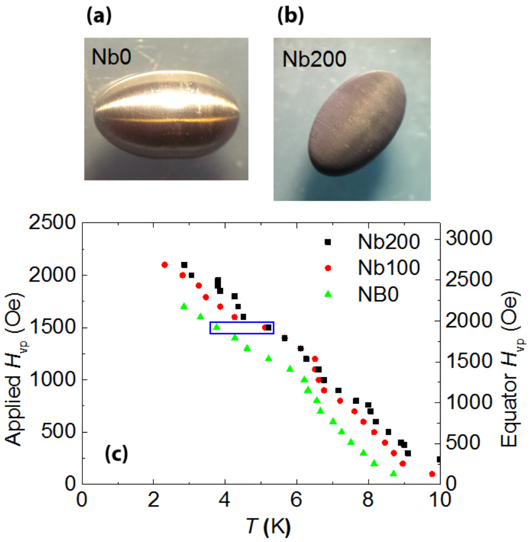

Fig. 2 Photographs of (a) bare Nb ellipsoid, Nb0 and (b) Nb ellipsoid coated with 200 nm MgB_2, Nb200. (c) Comparison of H_{vp}-T curves measured on several ellipsoids.

Our results demonstrate three important facts. First, it is indeed possible to increase the H_{vp} of the Nb material used in the state-of-the-art SRF cavities by a substantial amount (~600 Oe) by coating it with a thin superconducting film that screens vortex penetration. Second, it is technically possible to deposit a thin MgB_2 film with appropriate properties for this purpose on curved bulk Nb. Third, a single layer of superconducting thin film can reduce the magnetic field felt by the bulk material under it. These three facts are enough to demonstrate that it is possible to increase the H_{vp} of existing Nb SRF cavities by ~600 Oe. The superconducting shielding current density at the equator of Nb200 is estimated to be $J \sim \Delta H/d \sim 10^7$ A/cm^2, as compared to the depairing current density $J_d \sim 10^8$ A/cm^2 reported for MgB_2 thin films, therefore the maximum ΔH that a MgB_2 thin film can shield has not been reached. Further, a superconductor/insulating multilayer coating proposed by Gurevich would achieve even higher shielding field ΔH_{total} than with single layer films.

The result is from a collaboration with scientists at Los Alamos National Laboratory and has been published in Sci. Rep. 6, 35879 (2016).

3. MgB_2 coating of 3.9 GHz mock cavities

In the previous HEP-supported project we have established the technical feasibility of coating the inside wall of RF cavity with MgB_2 using a mock 6 GHz cavity. However, the small size of the 6 GHz cavity significantly affects the gas flow pattern inside of the cavity, leading to different qualities in the films deposited at different locations inside of the cavity. This is not easy to remedy as there is very little room to maneuver.

To solve this difficulty, we chose 3.9 GHz as the cavity frequency in this project, which has a much bigger physical sizes.[7] Because fluid dynamics plays a critical role in HPCVD, scaling up to coat the larger 3.9 GHz SRF cavities of requires significant investigation. The modified HPCVD system used in this project was similar to the 6 GHz system except that the size was larger to accommodate the larger size of the 3.9 GHz cavities. Instead of the flat sample stage heating the substrates in the standard HPCVD system, a cylindrical heater was adopted for the cavity coating system. The heater was split into two parts so that it could be open for installing the cavity for deposition. A tube-shaped molybdenum oven was used for the magnesium evaporation source. The resistive heating element wrapped around the oven and the temperature of the oven were independently controlled from the outer tube heater. The magnesium oven stood on the bottom of the vacuum chamber. A thermally insulated gas nozzle is inserted from the top of the chamber for the delivery of diborane gas. The thermal insulation is crucial because diborane gas is unstable at high temperatures. The 5% diborane in hydrogen gas mixture was introduced through this nozzle while the hydrogen carrier gas could be introduced through both this nozzle and another inlet outside the cavity. A supporting structure from the top of the vacuum chamber was used to hold and rotate the cavity while moving the cavity up and down along its vertical axis. In the meantime, the gas nozzle and the magnesium oven remained fixed during the deposition. The mock cavity was initially positioned such that the top area of the cavity was coated first. Then, it gradually moved upward so that the equator and the lower part of the cavity were coated. Small substrates of *c*-cut SiC, stainless steel foil of thickness 0.05 mm and high purity niobium pieces were mounted inside the mock cavity at different locations for deposition. Position #1 is on the cavity equator, Position #2 on the bottom part of the elliptical half-cell, and Position #3 on the bottom cavity tube.

The result of a mutual inductance measurement of an MgB_2 film on Nb placed at the equator of the 3.9 GHz mock cavity is shown in Fig. 3. A sharp superconducting transition at 38 K is observed. In Fig. 4, the real parts of the AC susceptibility versus temperature curves for several MgB_2 films placed at various positions and on various substrates are shown. Because several of the films are on metallic substrates, the imaginary parts of the signals from the films include signals from the substrates, which are difficult to separate from the film signals. Therefore only the real parts are shown here. The two films on stainless steel have the lower T_c values around 30 K regardless of their positions. The other three films, both at Position #1 and Position #2, show T_c values around 38 K. The films grown on SiC and niobium substrates have a better quality compared to films on stainless steel, which is consistent with the results from the standard HPCVD setup. Because of the small film size, the curve of an MgB_2 film grown on SiC at Position #1 does not reach full screening. Quality factor measurement at 18 GHz confirmed the results of the low-frequency measurement. An MgB_2 film on SiC substrate showed a superconducting transition at 38 K.

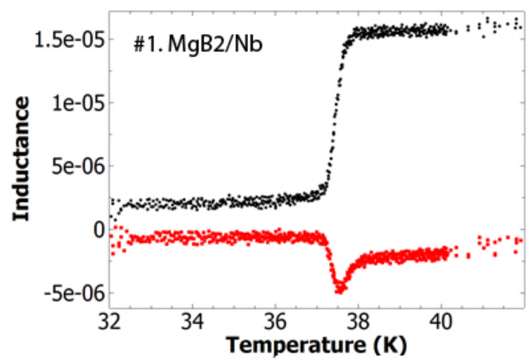

Fig. 3 Mutual inductance measurement of an MgB_2 film on Nb placed at Position #1.

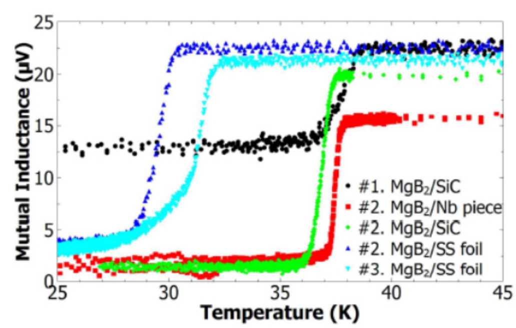

Fig. 4 Real parts of the AC susceptibility versus temperature curves for several MgB_2 films placed at various positions and on various substrates.

The results on the 3.9 GHz stainless steel mock cavity showed that a uniform film with good superconducting property can be grown across the cavity interior, demonstrating the feasibility of MgB_2 coatings with HPCVD for SRF cavities.

The result is from a collaboration with scientists at Argonne National Laboratory and has been published in IEEE Trans. Appl. Supercond. 27, 3500304 (2017).

4. MgB_2 films on 2 inch diameter copper discs

Current Nb cavities typically operate on sub-atmospheric liquid helium at 2 K, adding significant capital and operational costs for cryogenic refrigeration. Coating the inner walls of a Cu cavity with MgB_2 could bring the cavity operation temperature to 20 – 25 K. The excellent thermal conductivity of Cu is expected to improve the heat transfer from the superconducting coating and therefore provide better resistance to 'quench'. If operation at 20 - 25 K is achieved, closed-cycle refrigeration can be used, eliminating the need for liquid helium cooling and complicated cryomodules.

The main challenge in depositing MgB_2 on a Cu substrate is the reaction of Mg with the substrate. According to the Mg-Cu phase diagram (Fig. 5),[8] Mg reacts with Cu to form the alloys $MgCu_2$ and Mg_2Cu, which melt at 790 °C and 568 °C, respectively. The eutectic point for forming Mg-Cu liquid is $Mg_{0.84}Cu_{0.16}$ at 487 °C. Since the HPCVD process requires Mg vapor in the vicinity of the substrate, this effectively limits the HPCVD deposition of MgB_2 directly on Cu to around 500 °C. On the other hand, it has been found that the presence of a Mg-Cu liquid can promote the growth of MgB_2 at low temperature.[9]

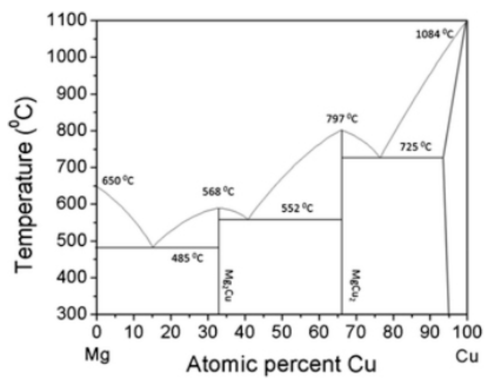

Fig. 5 Mg-Cu phase diagram.[5]

We have grown MgB_2 films directly on Cu disc at 450 °C.[10] X-ray diffraction shows that $MgCu_2$ and Mg_2Cu form on the Cu substrate before Mg vapor is supplied. Once an MgB_2 film is deposited, Mg_2Cu disappears. Evidently, the alloy Mg_2Cu acts as a source of Mg for the growth of MgB_2. Figure 6 shows (a) an optical image of an MgB_2 film on a 2 inch diameter Cu disc and (b)-(d) SEM cross section images from three different regions on the disc, marked by 1, 2, and 3, respectively. A platinum protective layer was deposited in situ prior to FIB milling. The optical image shows a purplish color characteristic for an MgB_2 thin film across the 2 inch disc, however the three different regions show different degrees of brightness. Area 1 at the edge of the disc is darker, Area 2 brighter, and Area 3 in the center is somewhere in between in brightness. The cross section images, taken at a 52° angle, show the MgB_2 film as a dark band of relatively uniform width for all the three regions. The film uniformly covers the Cu disc and is overall continuous except in Area 3, where regions of bright color interrupt the film occasionally. Results of EDS elemental mapping suggest that the bright region contains Mg and Cu and it is most likely a Mg-Cu alloy.

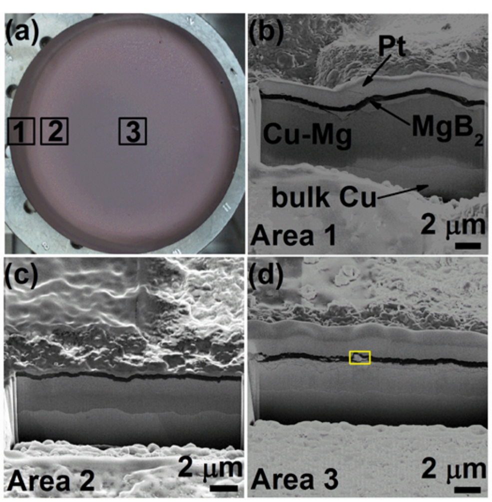

Fig. 6 (a) Optical image of an MgB_2 film on a Cu disc. (b), (c) and (d) SEM images of the FIB fabricated cross sections from three different areas.

DC magnetization measurement of a 650 nm thick MgB_2 film on a Cu disc shows that the sample has a superconducting transition at around 37 K, close to the bulk value of 39 K. Magnetic moment versus magnetic field (*m*-*H*) hysteresis loops measured in a perpendicular field shows a high J_c in the order of 10^7 A cm^{-2} at zero field, comparable to the reported values for MgB_2 films on ceramic substrates and Cu tapes. The critical current density is rapidly suppressed by the applied field, which is characteristic of clean MgB_2 films that lack strong pinning centers and a desirable property for SRF cavities.

The RF properties were measured at 11.4 GHz using two cryogenic RF cavities, one made of Cu and the other utilizing a Nb coating on Cu.[11] In both cavities, the total RF loss is a combination of the loss on the sample surface and that of the cavity body. For the measurements in the Cu cavity, the measured quality factor, Q_0, is dominated by the loss due to Cu. Therefore, it cannot be used to determine R_s of the superconducting sample, but only to determine its transition temperature. Figure 7(a) shows the Q_0 versus T curve measured from 4 to 50 K using the Cu cavity. A sharp decrease in Q_0 above 37 K signals a transition of the MgB_2 film from superconducting to normal state, confirming a T_c of 37 K from the DC measurement. Figure 7(b) shows the result for the Nb cavity from 4 to 10 K along with Cu and Nb reference samples. The transition close to 9 K is due to the superconducting transition of the Nb cavity itself. The surface resistance of both the Nb reference sample and the Nb cavity body is estimated to be 65 μΩ at 4 K and 11.4 GHz. Based on this value, Q_0 for the MgB2 sample in the Nb cavity (1.65×10^7) equates to an R_s of about 130 μΩ (R_s of the Cu reference sample is about 8.1 mΩ). The R_s crossover at 6.2 K in Fig. 7(b) is due to the high residual resistance of the MgB_2 film compared to Nb.

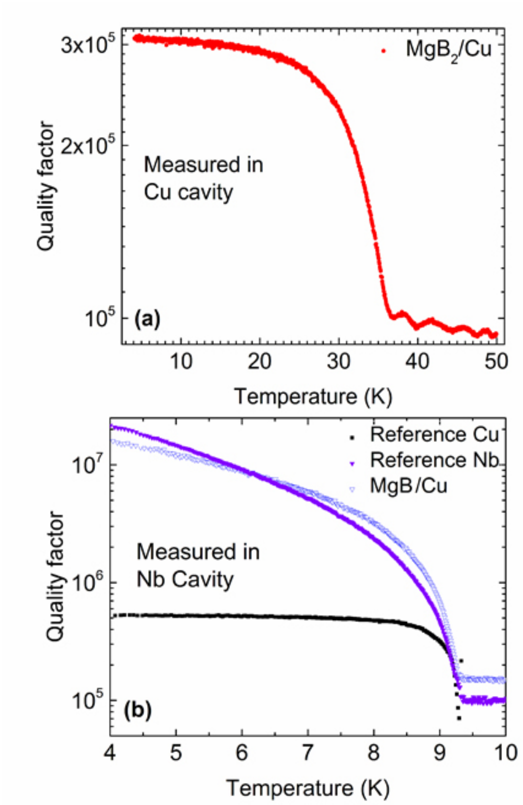

Fig. 7 Quality factor versus temperature curve of an MgB_2 coated Cu disc measured in (a) a Cu cavity and (b) a Nb cavity. The result from a Nb reference sample is included in (b) for comparison.

Combined with the results of coating mock cavities using the HPCVD process, the results demonstrate that it is possible to coat Cu cavities with high quality MgB_2 films using HPCVD. MgB_2 coated Cu could open up a possibility of using SRF cavities at 20–25 K with cryocoolers. In order to replace Nb cavities, however, investigations into the origin of high residual resistance and its reduction will be important in the future.

The result is from a collaboration with scientists at Argonne National Laboratory and SLAC National Accelerator Laboratory and has been published in Supercond. Sci. Technol. 30, 045001 (2017).

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