Superconducting MgB2 thin films by pulsed laser deposition

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Abstract
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Keywords
Physics and Astronomy, Pulsed laser deposition, Thin film growth, Thin films, Magnesium, Magnetic films

Disciplines
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Comments

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Growth of MgB$_2$ thin films by pulsed laser deposition is examined under ex situ and in situ processing conditions. For the ex situ process, boron films grown by pulsed laser deposition were annealed at 900 °C with excess Mg. For the in situ process, different approaches involving ablation from a stoichiometric target under different growth conditions, as well as multilayer deposition involving interposed Mg layers were examined and analyzed. Magnetic measurements on ex situ processed films show $T_c$ of $\sim 39$ K, while the current best in situ films show a susceptibility transition at $\sim 22$ K. © 2001 American Institute of Physics. [DOI: 10.1063/1.1385186]

The discovery of superconductivity in MgB$_2$ with $T_c$ $\sim 39$ K by Akimitsu and co-workers has caused tremendous excitement in the scientific community due to the rather simple binary intermetallic character of the material system, and the prospects for further discoveries of materials with even higher $T_c$ in this whole class of compounds.\textsuperscript{1} Quickly following the initial discovery, Bud’ko et al.,\textsuperscript{2} Finnemore et al.,\textsuperscript{3} and Canfield et al.,\textsuperscript{4} established some key facts pertaining to boron isotope effect as well as the thermodynamic and transport properties of this system, and demonstrated a method to fabricate dense wires of this new superconductor.\textsuperscript{2–4} The $T_c$, $H_c2(T)$, and $J_c$ data on these dense wires showed better performance as compared to their sintered pellet form. The development of wire is undoubtedly a key to the potential technological utilization of the superconductor. Another development which is very much needed for realization of a range of high-tech applications of MgB$_2$ superconductor involving Josephson junctions and heterostructure based devices is the reproducible preparation of epitaxial thin films of this material, preferably grown by an in situ process. There has been a report of MgB$_2$ film prepared by a possible postannealing process and pulsed laser deposition.\textsuperscript{5} In this letter, we address this issue and present the results of our experiments on the growth of MgB$_2$ films by the pulsed laser deposition (PLD) technique, either as an entirely in situ process, or as an ex situ process involving PLD of pure boron followed by annealing in Mg vapor.

For the case of pulsed laser deposition with ex situ annealing in Mg, we deposited boron films by pulsed laser deposition (Lambda-Physik excimer laser, KrF, $\lambda$=248 nm) and reacted these with Mg in a sealed Ta tube in Mg vapor at 900 °C. The boron films were deposited at 800 °C on SrTiO$_3$ (100) and (111) substrates at the rate of 0.035 Å/pulse, the thickness being $\sim 1000$ Å. The laser energy density and pulse repetition rate were 1.5 J/cm$^2$ and 10 Hz, respectively. The ambient was vacuum, with the background pressure below $10^{-7}$ Torr. We also compared these results with films deposited from an MgB$_2$ target onto SrTiO$_3$ (STO) substrates and subjected to Mg vapor reaction. For the case of in situ deposition, three different approaches were attempted: (a) PLD from a MgB$_2$ sintered target, (b) PLD of multilayers of MgB$_2$ and Mg followed by in situ annealing at high temperature, and (c) PLD of multilayers of Mg and B followed by in situ annealing at high temperature. In all cases, the same laser energy density (1.5 J/cm$^2$), pulse repetition rate (10 Hz), and ambient (vacuum) were used as stated above. Different cap layers at the bottom as well as top of the film sandwich were examined to control interface reaction and to avoid Mg escape during the high temperature anneal process. The details of these optimization studies will be reported separately in a longer paper. Here we present the conditions and results for the case, which yielded highest $T_c$ by an in situ process within the parameter framework we have examined thus far. For this case, STO (111) was used as the substrate. First, a 10 nm thin layer from MgB$_2$ target was grown at 500 °C. This was followed by the deposition of multilayers from MgB$_2$ and Mg targets at 350 °C with nominal thicknesses of a few nanometers each, with 400 bilayers in a sandwich. Finally, a cap layer of $\sim 10$ nm was deposited from MgB$_2$ target. Such a configuration was annealed in situ at 900 °C for 30 min. The heating rate was 40 °C/min and the cooling rate was 5 °C/min.

In Fig. 1 is shown the x-ray diffraction pattern for the film grown on STO (001) substrate by the ex situ process. Only one sharp line corresponding to the desired MgB$_2$ phase is seen indicating highly oriented, if not epitaxial, character of the film. The other two small peaks correspond to Mg. In fact, small Mg balls were found to be sticking on the film surface after the ex situ reaction in Mg vapor. These could not be removed by wiping with a cotton stub, but could be scrapped with a blade. These could also be removed by low temperature evaporation. We are exploring ways to remove this excess Mg by evaporation or by selective etching, without affecting the underlying film properties.

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In Fig. 2(a) we show the zero field cooled magnetization as a function of temperature for the \textit{ex situ} processed film for two magnetic field values of 25 and 200 Oe. A fairly sharp transition is seen near 39 K with a broad tail, for the field of 25 Oe. The transition shifts to lower temperature and broadens with an increase in field to 200 Oe, as expected. However, if one starts with a film deposited from a MgB$_2$ target, even after postannealing in Mg a well defined unique $T_c$ is not seen. Instead a second transition at about 20 K is seen \[\text{Fig. 2(b) and 2(c)}\] for films on STO \textit{100} and STO \textit{111} substrates, respectively.

In the case of the \textit{in situ} processed films, the transition as obtained from magnetization for the best case (conditions described earlier) is shown in Fig. 3(a). It shows a transition at 22 K with a tail. These lower values of $T_c$ could be due to departure from perfect stoichiometry in view of the possible variability of individual thicknesses in the multilayer, and also possibly loss of Mg at high temperature. We believe that careful thickness calibration followed by further optimization of the annealing sequence with an added suitably chosen cap layer would enhance the superconducting transition temperature as well as its quality. On films deposited directly from MgB$_2$ target at 800 °C, a sharp transition at $\sim$8 K has been seen \[\text{Fig. 3(b)}\]. For comparison, the ac susceptibility data of MgB$_2$ film prepared by \textit{ex situ} process is shown in Fig. 3(c).

The observation of superconducting transitions at 8, 22, and 39 K in films under different processing conditions is noteworthy. Specifically, the transition near 20–22 K was observed in the \textit{ex situ} process when implemented on films grown from MgB$_2$ target on STO(001) and STO(111) substrates and then magnesiumated, as well as in the \textit{in situ} films. These 8 and 22 K transitions may be due to stabilization of...
different phases of MgB$_y$ depending upon Mg deficiency. The sharpness of the transitions at lower temperatures raises an interesting question from the physics standpoint: do these transitions suggest occurrence of superconductivity in other stoichiometric phases of Mg–B system such as MgB$_4$ and MgB$_6$ or other, yet to be determined, MgB$_y$ compound?

In summary, *ex situ* and *in situ* processes are examined for the growth of superconducting MgB$_2$ films on SrTiO$_3$ substrate. The *ex situ* process is shown to yield a $T_c$ of $\sim$39 K, while the *in situ* process optimized thus far yields a $T_c$ of $\sim$22 K. In the *ex situ* process, the highest $T_c$ is obtained by magnesiating pure boron films, rather than films made from MgB$_2$ target. This suggests that a careful study of reaction and diffusion kinetics as a function of temperature and role of intermediate phases is warranted in this system. In addition, it demonstrates that if components can be coated with boron films, then exposure to Mg vapor can transform these films into MgB$_2$. For the *in situ* process, maintaining correct stoichiometry, especially retaining Mg in the film is a critically important issue yet to be fully resolved. Surface morphology of the growing film as influenced by the wetting properties of Mg films as a function of deposition temperature is an important issue, which deserves attention. While we have succeeded in growing films with $T_c$ close to bulk value by the *ex situ* process, an *in situ* process would be needed for realization of successful junction development. In the mean time, we are studying the systematic of the dependence of $J_c$ on temperature and field on the high quality *ex situ* films.

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