Superconductivity in Dense MgB_2 Wires

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MgB_2 becomes superconducting just below 40 K. Whereas porous polycrystalline samples of MgB_2 can be synthesized from boron powders, in this Letter we demonstrate that dense wires of MgB_2 can be prepared by exposing boron filaments to Mg vapor. The resulting wires have a diameter of 160 μm, are better than 80% dense, and manifest the full $\chi = -1/4\pi$ shielding in the superconducting state. Temperature-dependent resistivity measurements indicate that MgB_2 is a highly conducting metal in the normal state with $\rho(40\text{ K}) = 0.38\ \mu\Omega$ cm. By using this value, an electronic mean-free path, $l \approx 600$ Å can be estimated, indicating that MgB_2 wires are well within the clean limit. T_c, $H_{c2}(\mathrm{T})$, and J_c data indicate that MgB_2 manifests comparable or better superconducting properties in dense wire form than it manifests as a sintered pellet.

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I. Introduction.—The discovery of superconductivity in MgB_2 has caused a renaissance of interest in intermetallic superconductivity [1]. This, combined with the discovery of superconductivity in YPd_2B_2C and the RNi_2B_2C series several years ago [2–5], seems to indicate that the old idea of looking for high intermetallic T_c values in compounds rich in light elements is still a valid guiding principle. Measurements of the boron isotope effect in this compound [6] are consistent with the superconductivity being mediated via electron-phonon coupling, a conclusion that is also supported by recent band structural calculations [7]. Measurements of the upper critical field, $H_{c2}(\mathrm{T})$, the thermodynamic critical field, $H_c(\mathrm{T})$, and the critical current, J_c, indicate that MgB_2 is a type-II superconductor with properties that are consistent with an intermetallic superconductor that has a $T_c \approx 40$ K [8]. For example, other than the remarkably high T_c, MgB_2 appears to be quite similar to Nb_3Sn. Given this similarity, and given the far lower density of MgB_2 as well as the greater natural abundances of Mg and B, the logical questions are whether wires of MgB_2 can be easily synthesized and, if so, what are their physical properties. In this Letter we present a remarkably simple method for the synthesis of MgB_2 wires from boron filaments. In addition, we show that wires produced in this manner are of high density and have impressively low normal state resistivity and impurity scattering.

II. Experimental Methods.—MgB_2 can be synthesized in powder form by reacting stoichiometric amounts of powdered B and Mg at 950 °C for approximately two hours [6]. Given that at 950 °C the vapor pressure of Mg is approximately 200 Torr [9], it is believed that MgB_2 forms via a process of diffusion of Mg vapor into the boron grains. Based on this observation, the possibility of using this technique on other morphologies of boron appears to be promising.

MgB_2 wire was produced by sealing 100-μm-diam boron fiber [10] and Mg into a Ta tube with a nominal ratio of Mg_2B. Given that MgB_2 is the most Mg-rich binary Mg-B compound known [11], it was felt that excess Mg would aid in the formation of the proper, stoichiometric phase. The sealed Ta tube was sealed in quartz and then placed into a 950 °C box furnace for approximately two hours. The reaction ampoule was then removed from the furnace and quenched to room temperature.

Measurement of temperature- and field-dependent electrical resistivity and magnetization was performed in Quantum Design MPMS and PPMS systems. Resistivity measurements were made in a standard four probe geometry using Epotek H20E silver epoxy to make contacts. The contact resistance was approximately 1 Ω. Given the well-defined geometry of the samples, accurate measurements of resistivity were possible.

III. Results.—Upon opening the Ta tube it became clear that there had been a reaction between the boron fiber and the Mg vapor. Whereas the boron fibers were straight and moderately flexible before the reaction, the MgB_2 wires in the Ta tube after the reaction were brittle and deformed. The inset of Fig. 1 is a photograph of the resulting wires. As can be seen, there has been significant warping and bending of the fiber as a result of the reaction with the Mg vapor. Figure 2 shows scanning electron microscope images of the fiber before the reaction as well as the wire after the reaction. In both cases a tungsten core (approximately 15 μm diameter) can be clearly seen. This core is part of the original boron fiber and does not appear to be affected by the exposure of the fiber to Mg, nor, as will be seen, does it seem to affect the superconducting properties of the resulting MgB_2 wire. Whereas the boron fiber has a diameter of 100 μm and breaks with a smooth, clean surface (inset of Fig. 2), the MgB_2 wire has a diameter of approximately 160 μm and breaks with a rougher, grainier surface. The increased diameter of the wire is consistent with observations that there is an expansion associated with the formation of the MgB_2 powders during synthesis [6]. Although the MgB_2 wires are somewhat brittle, the integrity of the filament segments was preserved during the exposure to the Mg vapor, i.e., the fibers did not decompose or turn into powder.

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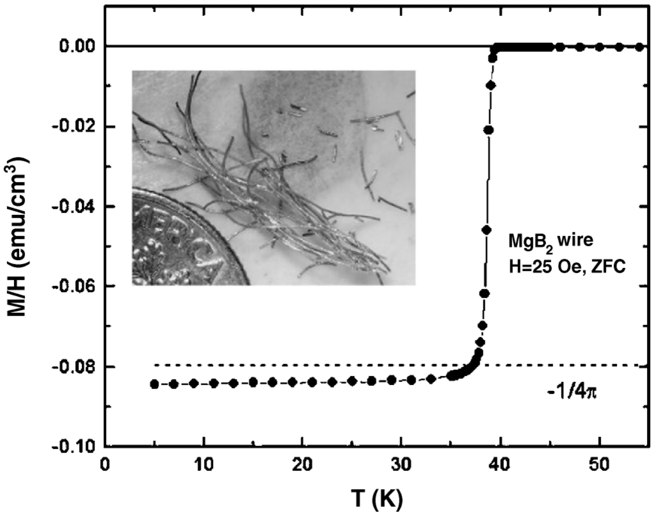

FIG. 1. Magnetization divided by applied field (25 Oe) for zero field cooled wire sample. Field was applied parallel to the wire length, leading to a demagnetization factor close to 0. Inset: photograph of wires, as they appear after removal from Ta tube, and part of U.S. dime for scale.

Using a diameter of 160 μm and measuring the length and mass of several wire segments, we determined the density of the wire to be 2.4 g/cm^3. This is to be compared with a theoretical value of 2.55 g/cm^3 for a single crystal sample using lattice parameters $a = 3.14$ Å and $c = 3.52$ Å for the hexagonal unit cell [6]. Given the rather coarse nature of our measurements, this implies that the wire samples are probably better than 80% of the theoretical density. It should be noted that the small tungsten core would come in as a roughly 10% correction, and therefore is within our level of uncertainty.

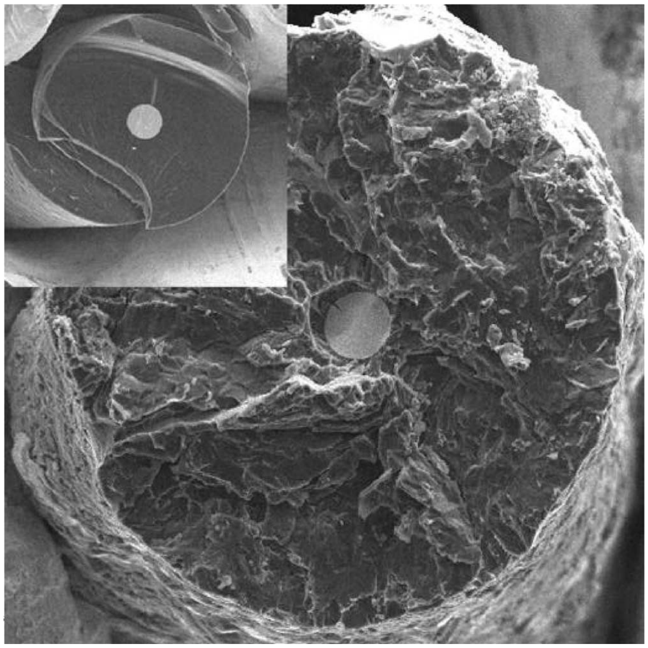

FIG. 2. Electron microscope image of cross section of grown MgB$_2$ wire. The diameter of the wire is 160 μm. Inset: electron microscope image of the unreacted boron filament. The diameter of the filament is 100 μm. For both images the wire/filament was snapped *in situ*. Note that, in both images, a central core of tungsten wire (diam $\approx$ 15 μm) can be clearly seen.

Figure 1 presents the temperature-dependent magnetization of MgB$_2$. The data were taken after cooling in zero field and then warming in a field of 25 Oe. Given the aspect ratio of the wire segments used we were able to obtain a susceptibility very close to $-1/4\pi$, the value expected for total shielding and a demagnetization factor close to 0. $T_c = 39.4$ K can be determined from these data by using an onset criterion (2% of $-1/4\pi$). The width of the transition (10%–90%) is 0.9 K.

Figure 3 presents the temperature-dependent electrical resistivity of MgB$_2$ wires. The room temperature resistivity has a value of 9.6 $\mu\Omega$ cm, whereas $\rho(77\text{ K}) = 0.6$ $\mu\Omega$ cm and $\rho(40\text{ K}) = 0.38$ $\mu\Omega$ cm. This leads to a residual resistivity ratio (RRR) of RRR = 25.3. It should be noted that the shape of the resistivity curve and the RRR values are qualitatively the same as those observed for sintered pellets of polycrystalline Mg^{10}B$_2$ [8]. The relatively low room temperature resistivity value, along with the high RRR, is not unusual for diboride samples [12]. The resistivity of the sintered pellet samples [8] is approximately 1 $\mu\Omega$ cm at 40 K. This somewhat higher value of the calculated resistivity for the pellet is consistent with the sintered sample, having an actual density substantially lower than the theoretical value.

The temperature-dependent resistivity shown in Fig. 3 can be fit by $\rho = \rho_0 + \rho_1 T^{\alpha}$ with $\alpha \approx 2.6$ between T_c and 200 K. This is comparable to the power law $R = R_1 + R_1 T^{\alpha_1}$ with $\alpha_1 \approx 2.8$ found for the sintered Mg^{10}B$_2$ sample [8] over a comparable temperature range.

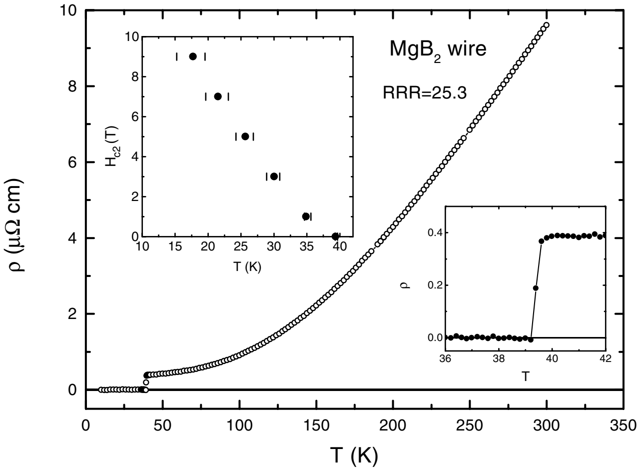

FIG. 3. Temperature-dependent electrical resistivty of MgB$_2$ wire. Lower inset: expanded view for temperatures near T_c. Upper inset: H_{c2}(T) data inferred from temperature-dependent resistivity data taken in constant applied field upon cooling. The three symbols are for onset, maximum slope, and completion temperatures.

Given the similarity of the two power laws it seems clear that the resistivity of MgB_2 will not have a linear slope for temperatures between T_c and 300 K [7]. On the other hand, using an average Fermi velocity [7] of $v_F = 4.8 \times 10^7$ cm/s and a carrier density of 6.7×10^{22} e/cm^3 (two free electrons per unit cell) we can estimate the electronic mean-free path to be approximately 600 Å at T_c. This is clearly an approximate value of the electronic mean-free path, but, given the estimated superconducting coherence length of approximately 50 Å [8], these values place MgB_2 wires well within the clean limit. Given a $\kappa \approx 26$ [8], this implies that, much like the case of the RNi_2B_2C materials [5], there may be significant nonlocal effects associated with MgB_2.

The superconducting transition temperature, $T_c = 39.4$ K, can be determined from both the magnetization and resistivity data shown in Figs. 1 and 3. This value is slightly higher than the $T_c = 39.2$ K value determined for isotopically pure $Mg^{11}B_2$, but is significantly lower than $T_c = 40.2$ K for $Mg^{10}B_2$. This is consistent with an approximate 80% natural abundance of ^{11}B. It is noteworthy that the superconducting transition is both relatively high and sharp in the wire samples. This means that either very few impurities are being incorporated into the MgB_2 or that what few impurities are being incorporated are having very little effect on either resistivity or T_c.

The temperature dependence of the upper critical field, H_{c2}(T), is shown in the inset of Fig. 3. For each field, three data points are shown: onset temperature, temperature for maximum $d\rho/dT$, and completion temperature. Qualitatively, these data are remarkably similar to the H_{c2}(T) data inferred from measurements on $Mg^{10}B_2$ sintered pellets [8] as well as recent measurements on hot-pressed powders [13]. Quantitatively, at $H = 9$ T, the width of the resistive transition for the wire sample is roughly half of the width found for the sintered sample. These data are consistent with the wire sample being of comparable or better quality as the sintered powder samples.

Figure 4 presents data on the critical current density, J_c. The open symbols are J_c values extracted from direct measurements of the current-dependent voltage across the sample at a given temperature and applied field values. The filled symbols are J_c values inferred from magnetization loops by application of the Bean model [8,14]. The direct measurement of J_c was limited to values below approximately 200 A/cm^2 due to resistive heating from the sample leads and contact resistance. As can be seen, the extrapolations of the directly measured, low J_c, data and the Bean-model-inferred, high J_c, data match up moderately well. In comparison to the J_c data presented for a sintered pellet of $Mg^{10}B_2$ [8], J_c for the wire sample is roughly a factor of 2 higher at low fields and over an order of magnitude higher at high fields.

IV. Conclusions.—We have devised a simple technique of producing low resistivity, high density, high T_c MgB_2 in wire form via the exposure of boron filaments to Mg vapor. The resulting wire has better than 80% the theoretical density of MgB_2 and measurements of the temperature-dependent resistivity reveal that MgB_2 is highly conducting in the normal state. The room temperature resistivity has a value of 9.6 $\mu\Omega$ cm, whereas the resistivity at $T = 40$ K is 0.38 $\mu\Omega$ cm. This means that, even in the normal state, wires of MgB_2 can carry significant current densities. This should be compared with the resistivity of Nb_3Sn $\rho(20\text{ K}) = 11$ $\mu\Omega$ cm and $\rho(300\text{ K}) = 80$ $\mu\Omega$ cm [15].

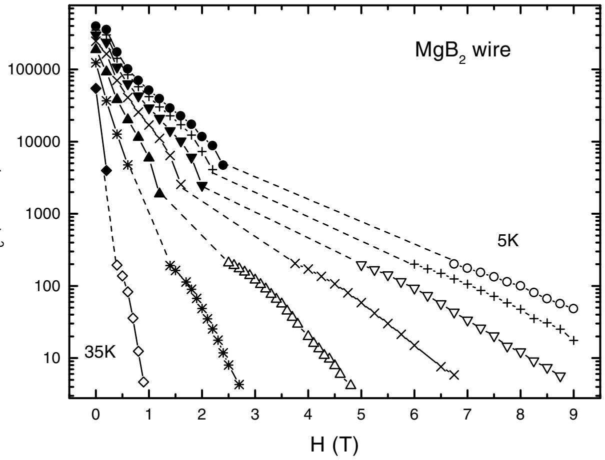

FIG. 4. Superconducting critical current density, J_c, as a function of applied field every 5 K in the 5–35 K range. Open symbols were taken via direct measurement of current-dependent voltage of the wire. Filled symbols were determined via a Bean-model analysis of magnetization data from wire samples, with the applied field parallel to the wire length. The dashed lines simply connect data sets taken at the same temperature.

Given the well-defined geometry of the wire samples we have been able to directly measure a full $-1/4\pi$ susceptibility in the superconducting state. The values of T_c for this wire sample are slightly higher than the T_c values for isotopically pure $Mg^{11}B_2$ sintered powders and the width of the resistive superconducting transition is smaller than that seen for $Mg^{10}B_2$ sintered powders. In addition, H_{c2}(T) for the wire sample is virtually the same as that found for $Mg^{10}B_2$ sintered pellets. Based on all of these observations, it appears that MgB_2 wires provide dense, high-quality samples of MgB_2. By comparing our estimate of the electronic mean-free path $l \approx 600$ Å, to the superconducting coherence length, $\xi \approx 50$ Å [8], we can see that MgB_2 wires are well within the clean limit.

All of the above, of course, present the possibilities of using such wires for both research and applied purposes. For basic research the possibilities of making weak-link Josephson junctions and other devices are currently being pursued. On the applied side, given that boron filaments are produced in a variety of sizes and of arbitrary lengths, the possibility of converting boron filament into MgB_2 wire as part of a continuous process leads to the possibility of simple manufacturing of lightweight, high T_c, wires with remarkably small normal state resistivities. In addition, this process could be used to turn boron coatings on tapes, cavities, or other devices into high-quality superconducting films. Although the low temperature J_c values are currently smaller than those for Nb_3Sn [8], as of yet very little effort has been put into optimizing J_c. A multidimensional phase space of filament purity, diameter, treatment time, and temperature has yet to be explored. Both basic and applied directions of research will have to be explored in detail over the coming months and years.

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