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Improved Critical current Density in MgB_2 by Graphene nano-platelets

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그래핀 나노플레이트에 의한 MgB_2의 임계전류밀도 향상

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Abstract

The effect of graphene inclusion in the ex-situ MgB_2 was analyzed with the help of resistivity behavior and critical current density studies. Amount of graphene was systematically varied from 0% for pristine sample to 3% by the weight of MgB_2. Graphene that is considered as a good source of carbon was found to be intact without any significant carbon doping in MgB_2 structure as reveled by XRD measurements. There was no signature of graphene inclusion as far as the superconducting transition is concerned which remained same at 39 K for all the samples. The transition width being sensitive to defect doping remained more or less about 2 K for all the samples showing no variation due to doping. Although there was no change in the superconducting transition or transition width, the graphene doped sample showed noticeable decrease in the overall resistivity behavior with respect to decrease in temperature. The graphene inclusion acted as effective pinning centers which have enhanced the upper critical field of these samples.

Keywords: MgB_2, graphene, critical current density, pinning sites

I. Introduction

The superconductivity in MgB_2 created interest amongst the scientist since it was first announced by Akmitsu et. al. [1]. Even a decade later amidst the discovery of other superconductors [2], it still remains a potential candidate towards application in superconductor science and technology. This is because it has a simple crystal structure and more significantly it is a binary compound with low cost and non toxic starting material. MgB_2 is a superconductor with multiple band gaps [3] which further manipulates it to deviate from the Werthame et.al. [4] formulation to show a higher overall upper critical field value at low temperatures with dirtier π-band [6]. This further increases the expectation from magnetic properties in MgB_2 being enhanced by a suitable dopant. Out of various dopant tried [5]

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carbon is found to be the most successful dopant which almost doubled the $H_{c2}(0)$ ranging between 32.5 T to about 60 T [6-9] in various MgB_2 samples. The critical current density is also observed to increase with carbon doping up to 10^5 A/cm^2 under self fields [10-12] for various sources of carbon. Graphene which is a few layered compound of carbon is also a good source of carbon. It shows various fascinating properties like extremely lower conductivity and reduced effect of phonon scattering from its perfect hexagonal crystal lattice structure. The relatively thin layers of single or multilayered graphene sheets are very sensitive towards its surrounding material. Hence it would be interesting to study the role of graphene sheets in MgB_2 matrix. Therefore in the present work, graphene was included in ex-situ MgB_2 bulk and its effects on superconducting properties were studied.

II. Experimental

In the present work graphene nano platelet was mixed with MgB_2 powder from Alfa Aesar through solid state synthesis. 0 %, 1 % and 3 % graphene by the weight of MgB_2 was taken and hand milled to form samples named MBG0, MBG1 and MBG2 respectively. Here MBG0 was used as a reference samples to study the effect of graphene doping. These samples were heat treated at 900 ℃ for 3 hours in 5N Ar+4%H_2 environment. The samples so synthesized were examined for their structure by X-ray diffraction measurement using Rigaku, DMAX 2200 with CuK radiation(λ=1.54 Å). The as synthesized samples were tested for its superconductivity by performing a collinear four probe resistivity measurement. Critical current density was estimated by using modified beans model from magnetization measurements performed with the help of VSM module in Quantum Design Evercool II PPMS.

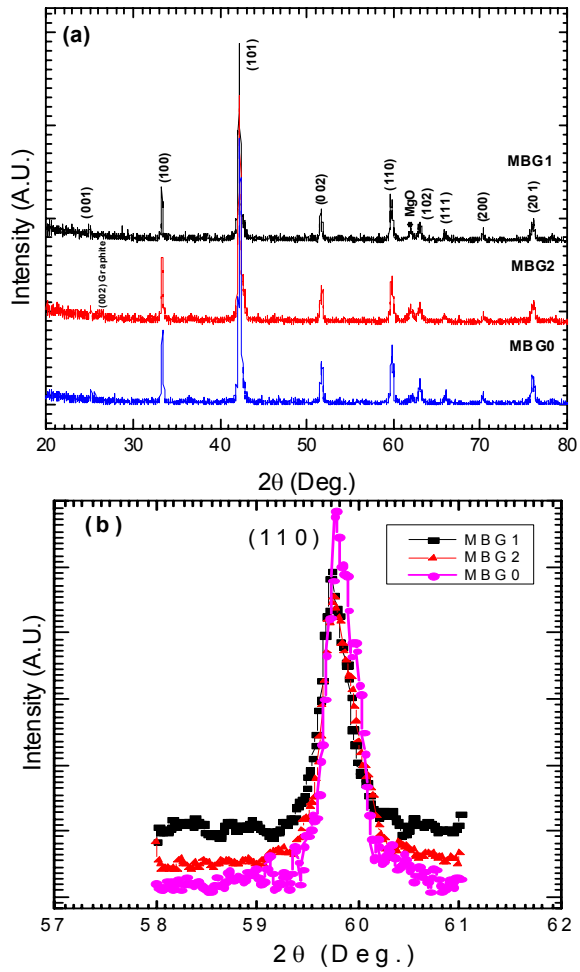

Fig. 1. X-ray diffraction pattern for (a) pure as well as graphene doped MgB_2 samples (b) enlarged view of (110) peak.

III. Results and Discussion

3.1 X-ray diffraction studies:

Graphene nano platelets were considered to form a randomly oriented two dimensional defects in MgB_2 bulk. Since graphene is a two dimensional sheet, a single or multiple layer carbon from graphite, it could also act as a probable source of carbon that will distort the MgB_2 structure by substitution at boron sites [13]. The inclusion of graphite during the synthesis of MgB_2 has already been reported to the

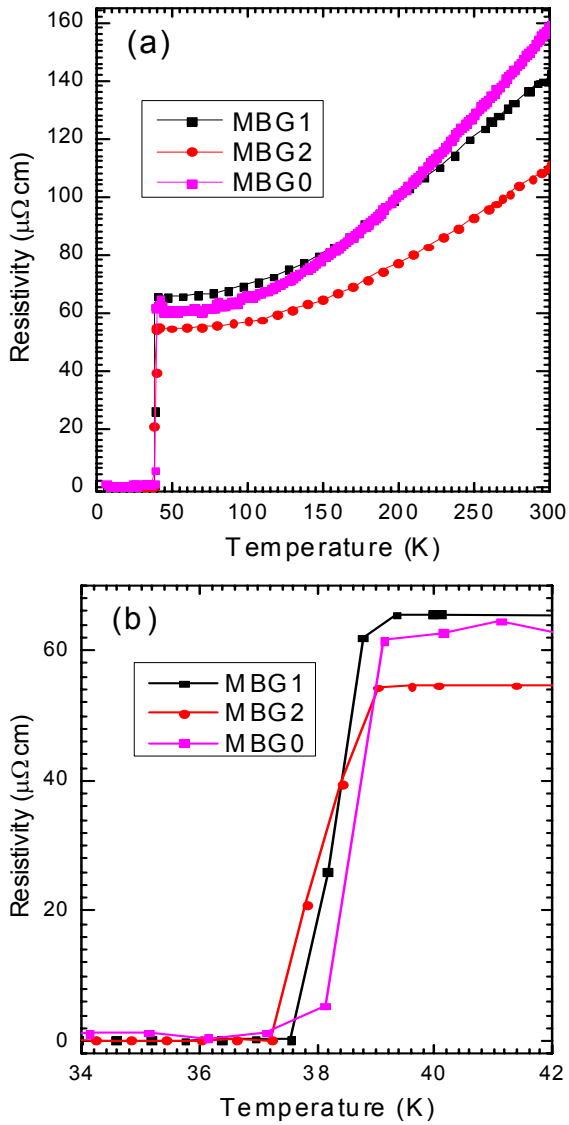

Fig. 2. (a): Resistivity response for all the samples with respect to decrease in temperature and (b) enlarged view of superconducting transition in the resistivity plot for the samples MBG0, MBG1 and MBG2.

dope carbon at boron sites [14]. Hence the samples were examined for any changes in MgB_2 crystal structure by XRD analysis. Fig. 1 shows the XRD patterns for the samples MBG0, MBG1 and MBG2 in the 2θ range of 20 to 80 degrees. The use of ex-situ MgB_2 powder as a starting material makes it obvious to exhibit all the peaks corresponding to MgB_2 hexagonal crystal structure. However, the fact to be noted is absence of shift in angle corresponding to (110) peak. It is well known that doping of carbon in MgB_2 structure leads to the decrease in the in-plane lattice parameter [13]. From fig. 1(b) it is quite clear that there is hardly any remarkable shift in peak corresponding to ab-plane of hexagonal MgB_2 structure. This indicates that even though the samples were heat treated at 900 ^oC for 3 hours, there is no carbon doping in MgB_2 from graphene. This signifies that graphene has remained more or less intact and act as a two dimensional defects in MgB_2 matrix. This can also be supported by the gentle presence of (002) graphite peak emerging at around $2\theta=26.2°$ for sample MBG2 where the concentration of graphene is more than 1%. There is a slight presence of MgO impurity which is unavoidable in case of MgB_2 due to high affinity of oxygen towards Mg [15]. The Scherrer formula, $d = 0.9\lambda/\beta\cos\theta$ was used for the measurement of crystallite size of the samples. Here d is crystallite size in nm, λ is wavelength of X-ray radiation in nm, β(rad.) is full width at half maximum after the correction for the instrumental broadening and θ(deg.) is the angle of diffraction. The crystallite size for the samples was found to be 28.9 nm, 28.3 nm and 25.2 nm respectively for MBG0, MBG1 and MBG2. It can be observed that there is very less difference in the crystallite size of MBG0 and MBG1 while, the same for MBG2 is noticeably reduced. From this we can infer that the addition of graphene results in the decrease in the MgB_2 crystallite size.

3.2 Resistivity measurements:

Fig. 2 shows the resistivity plots with respect to temperature. The resistivity behavior for all samples seems to be identical wherein the high temperature exponential behavior due to electron phonon interaction gets more or less terminated at a particular residual resistance after which it undergoes superconducting transition at critical temperature. MBG1 shows more or less similar residual resistivity as MBG0, however MBG2 has comparably lower overall resistivity. It is known that graphene is not

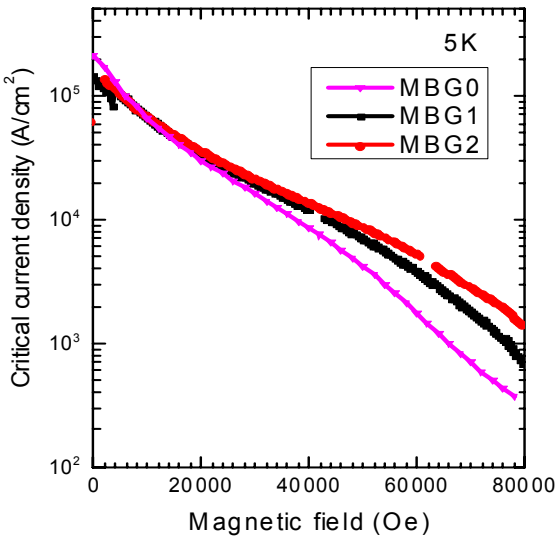

Fig. 3. Critical current density plots for samples MBG0, MBG1 and MBG2 at 5K.

only a very good conductor, but even has a low dependence on the phonons [16]. The thinness of graphene exposes the few atomic layers it has to the immediate superconducting surroundings strongly influencing its intrinsic property. This may be a reason for the reduction of overall resistivity of sample MBG2 or otherwise the resistivity should increase due to defect scattering. This is also supported by the fact that overall resistivity of MBG1 is same as that of MBG0 even with the 1% graphene. It is now widely understood that any doping in MgB_2 reduces the superconducting transition temperature. But in contrast to this, the transition temperature remains constant for all the samples. If there have been any carbon substitution from graphene, it must have reflected from the decrease in the superconducting transition. This backs the observation from XRD measurements which do not show any signs of carbon substitution in the MgB_2 structure. Moreover, the transition width is about 2 K which is also same for all the samples. Hence it is conclusive that although graphene is present in the matrix, it remains as an external phase without interfering in superconducting properties and thus keeping the optimum transition onset at 39 K.

3.3 Critical current density:

Although there is no carbon substitution, mere presence of graphene in MgB_2 matrix resulted in the enhancement of critical current density. Fig. 3 shows the critical density plots for samples MBG0, 1 and 2 at 5 K as calculated from the MH measurements using modified Bean's formula. Although the infield J_c of the samples remains the same, high field J_c shows improvement with graphene inclusion. It is remarkable that there is enhancement in the critical current density with graphene inclusion, without any drop in the transition temperature observed at about 39 K. Although the improvement in the critical current density is marginal, the fact to be stressed upon is the improvement observed without any compensation in the transition temperature, which is against the usual trend in MgB_2 superconductors.

IV. Summary

Effect of graphene on ex-situ MgB_2 is studied. Even after heat treatment at 900 ^oC for 3 hrs, graphene was found intact without any carbon doping in MgB_2 structure. The superconducting transition remained unchanged at 39 K for all the doped as well as undoped samples which otherwise decreases with any kind of doping tried in MgB_2 superconductor. The transition width which is sensitive to doping or impurity seems to be same for pristine and graphene doped samples. The inclusion of graphene in the MgB_2 matrix however has assisted in improving the critical current density of the sample. It remain in the sample as an external phase and act as an effective pinning center along with the MgB_2 grain boundaries.

Acknowledgments

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