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Effect of Mn substitution on the thermo-power of the superconductor Mg_{1-x}Mn_xB_2

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Abstract: In this communication a systematic study has been made on the effect of Mn substitution on the thermo-power of MgB_2 superconducting compounds involving 0%, 0.5%, 1.0%, 1.5% and 2% Mn content. The superconducting transition has been found to be very sharp for the pure compound of MgB_2 and it is observed that the transition width increases with Mn content. For small values of Mn content, the thermo-power S(T) increases with Mn content which suggests that the hole density increases. However, with further increase in Mn content, a small decrease in S(T) is observed. For all the compounds, we have observed the expected linear region at low temperatures. There is no systematic trend in the slopes dS/dT with Mn content.

1. Introduction:

The discovery of superconductivity in MgB_2 at 39 K [1] has resulted in intensive investigations both theoretically as well as experimentally, and the main aim of such studies is to develop some practical applications. It has the highest transition temperature, T_c, among the inter-metallic compounds. The structure of MgB_2 is a simple hexagonal with honeycomb type boron layers and the interpenetrating Mg layers [2]. There is a general agreement that MgB_2 is a two-band superconductor. It is also well established that the two bands and the respective energy gaps are associated with different parts of its Fermi surface. Compared to the cuprate based superconductors, MgB_2 shows larger coherence length, lower anisotropy and better current flow across the grain boundaries [2]. These properties, in fact, make MgB_2 a good candidate for practical applications. Chemical substitution is a powerful tool to understand the mechanism of superconductivity as well as to improve its practical applicability. There are several substitution studies on this compound and it is observed that there is a decrease in T_c in all
the substitutions [3-7]. To explain the variation in \( T_c \), several mechanisms have been proposed such as pair breaking effect, decrease in phonon frequency, reduction in density of states (DOS) and band narrowing. For instance, the reduction in \( T_c \) in Al-doped compounds has been attributed to the decrease in DOS at the Fermi level.

Among the various dopants, Fe and Mn are of great interest due to magnetic properties of these dopants. It is further seen that Mn shows the most rapid decrease in \( T_c \). There are several reports on Fe doped compounds of MgB\(_2\). Few studies seem to have been done on Mn-doped compounds. These studies are limited to properties like transition temperatures and critical fields. The studies demonstrate that there is drastic decrease in \( T_c \), which has been attributed to spin-flip scattering and pair-breaking effect [8]. Recently we had reported results on resistivity, magneto-resistance and specific heat (with and without external field) on Mn-doped compounds [9]. It was observed that the jump in specific heat decreases with Mn content.

Among the various transport probes, thermoelectric power (TEP) is one; researchers pay much attention to, as it is sensitive to band structure and scattering mechanism. TEP is also an important tool to study the nature of charge carriers. In literature, there are several reports on thermopower studies on various substitutions like Al, Fe, and Be [7, 10, 11]. In general, the nature of thermopower curves is similar; however, the trend with doping depends on the nature of dopants. For instance, Al doping induces an increase in hole concentration and hence \( S(T) \) increases with Al content [10]. A similar trend has been observed in Fe doped samples of MgB\(_2\) [7] where an increase in \( S(T) \) is seen with Fe content. However in Be doped samples of MgB\(_2\), a decrease in \( S(T) \) is observed with increasing doping content [11]. To the best of our knowledge, no studies seem to have been carried out on TEP measurements on Mn-doped compounds of MgB\(_2\). Keeping this in mind, we carried out such measurements. In this communication, we report TEP measurements on pure and Mn-doped samples of MgB\(_2\) in the temperature range of 10-300 K.

2. Experimental Details:

Bulk samples of Mg\(_{1-x}\)Mn\(_x\)B\(_2\) have been prepared using the conventional solid state reaction technique. The details of the preparation technique have been given in our previous communication [9]. Transition temperature was determined using four-probe method. In the present investigation, X-ray diffraction data were collected at room temperature from 20° to 100° using an X-ray diffractometer.

Thermo-electric power (\( S \)) measurement, in the temperature range of 10 – 300 K, was performed with a commercially available Dewar using the differential dc method, taking OFHC (oxygen-free highly conducting copper) as reference. Superconducting sample was taken in the form of a cylindrical pellet. The flat and parallel surfaces of the pellet were clamped between two cylindrical copper blocks. The upper copper block also acted as heat sink. These copper blocks hold the sample securely with the help of an SS spring-loaded axial brass screw and a teflon block attached to the metallic support. The two junctions of the Au-Fe (0.07%)-alumel thermocouple were attached thermally to the two copper blocks using GE-varnish. A Lakeshore temperature controller (Model 340) was used to control the temperature difference (\( \Delta T \)) between the two copper blocks using the thermocouple and a heater wound on the lower copper block. The data was taken during the warming cycle.
3. Results and Discussion:

The transition temperatures of the pristine and Mn-doped samples are presented elsewhere [9]. The XRD patterns are shown in figure 1. One can note that all the samples contain MgO and the corresponding peaks have been indicated in the XRD pattern. To ascertain this, we have performed Rietveld analysis for all the samples and observed that the range of MgO in these is between 4-6%. The lattice parameters are given in our earlier communication. Figure 2 shows the temperature dependence of the TEP of pristine as well as Mn-doped samples of MgB$_2$. Each curve is off-set by 2 µV for the sake of clarity. For the pure sample, one can observe a linear dependence of TEP at low temperatures which is typical behaviour of diffusive thermo-power of metals. This is shown in the inset of figure 2. The linear trend is associated with the thermal diffusion of carries. Departure from linear behaviour is seen at temperatures above 170 K. Beyond 250 K, TEP tends to saturate which is possibly due to the polarionic transport contribution at higher temperatures. This behaviour is similar in nature when compared to the reports in literature [7]. However no such saturation is observed for Mn-doped samples. It may be mentioned that the saturating behaviour of TEP at high temperatures is also observed in bismuthates and cuprates. A positive value of TEP confirms the evidence for predominantly hole-type conduction in MgB$_2$ and Hall Effect measurements have also provided evidence for the same [12]. In a polycrystalline sample of MgB$_2$, the measured value of S(T) is sum of the two bands, and . Since S(T) is positive, this means that the hole like -band dominates the
thermo-electric transport in these compounds. The transition temperatures determined from the S (T) curves, in general, match quite well with that obtained from electrical and magnetization measurements performed on these samples. For example, the transition temperature as obtained using four probe for the pure sample of MgB$_2$ is observed to be 39 K whereas that determined from S(T) measurements is determined to be 38.4 K.

We now describe the effect of Mn doping on TEP. As boron is substituted for Mg, the value of TEP increases for small concentration of Mn (up to 1%); thereby indicating that such doping introduces hole type carriers. However, with further increase in Mn concentration, a small decrease in TEP is observed. In other words, for small values of Mn content, doping enhances the role of $\sigma$-bands. However for Al and Fe-doped compounds of MgB$_2$ [7, 10] TEP is observed to increase monotonically with impurity concentrations. Figure 3 depicts the slopes of thermo-power dS/dT in the linear region and also depicts the values of S at 300K for pure and Mn-doped samples of MgB$_2$. We could not notice any systematic variations in dS/dT, however, it is seen that the room temperature thermo-power increases almost linearly for small Mn concentration. Similar variations in room temperature thermo-power have been observed in Co and Cr doped compounds and such variations have been attributed to the reduction of density of states around Fermi energy [10.13]. It may also be pointed out that no such systematic variations in room temperature S(T) have been observed in Fe-doped compounds of MgB$_2$ [9] which has been attributed to the fact that the DOS remains unaffected for small concentrations of dopants.

![Figure 2](image_url)

Figure 2: The temperature dependent thermo-power for pristine and Mn-doped MgB$_2$ compounds. For the sake of clarity each curve is off-set by 2 $\mu$V. The inset shows the linear region for pure sample.
We now use the thermal transport theory to our system. To understand in a qualitative manner, we can approximately apply the Mott formula which is given by

\[ S(T) = X_b T \] (1)

where the parameter \( X_b \) is inversely related to the Fermi energy, \( E_F \). We have already pointed out that MgB\(_2\) system is a multi-band system with \( \sigma \) and \( \pi \) bands co-existing. In addition to this, band structure calculations have shown that the Fermi surface is also not spherical and also reflect the anisotropy of the layered structure [14]. Here in our system of Mn-doped samples, we observe that the parameter \( X_b \) increases with Mn content (for low Mn content) which indicates that the hole density increases with Mn content. This is also consistent with the observation that the thermo-power values increase with increase in Mn-content. We assume that the transport in the linear region is mainly due to hole carriers, for pure sample we estimate the Fermi energy which is equal to 0.68eV which matches well with that in literature [10].

4. Conclusion:
In the present studies, we have presented thermo-power measurements of pristine and Mn-doped samples of MgB$_2$. Pristine as well as Mn-doped compounds exhibit hole-like trend. It is seen that for low Mn contents, TEP increases with Mn concentration. However, a decrease in TEP is seen for higher concentration of Mn. On contrary, no systematic trends in the slopes $dS/dT$ could be observed for pure as well as Mn-doped samples. We have also estimated Fermi energy for pure sample and it is satisfactory to note that it is in good agreement with that in literature.

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