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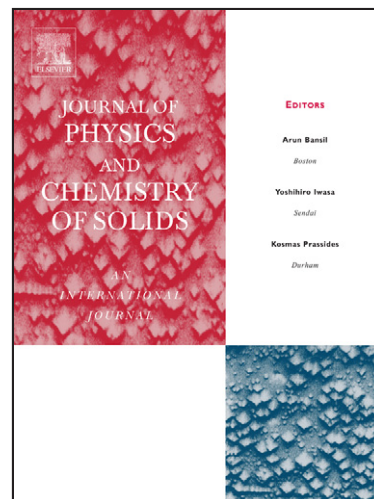
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Transport properties of MgB_2 single crystals doped with electrons and holes

K. Rogacki¹, K. Oganisian¹, C. Sulkowski¹, N. Zhigadlo², S. Katrych², and J. Karpinski²

¹*Institute of Low Temperature and Structure Research, Polish Academy of Sciences,
50-950 Wroclaw, Poland*

²*Laboratory for Solid State Physics, ETH, 8093 Zurich, Switzerland*

Abstract

The thermoelectric power of C, Mn, C:Li, and Al:Li substituted MgB_2 single crystals has been investigated in the temperature range 10-300 K. Both the in-plane (S_{ab}) and the out-of-plane (S_c) thermopowers are positive for the non-substituted crystal and both S_{ab} and S_c change a sign for crystals doped with electrons where C is substituted for B in the amount larger than 5 %at. When Li is substituted for Mg, the π band rather than the σ band is doped with holes and the doping effects are much more subtle. The anisotropy ratio of the non-substituted crystal $S_{ab}/S_c \approx 3$ and this ratio is strongly reduced by the substitution of C. Isovalent magnetic Mn ions, which substitute for Mg with a drastic reduction of T_c , do not influence the values and temperature dependences of both S_{ab} and S_c .

Substitutional chemistry can modify in the controlled way electronic structure and properties such as superconducting transition temperature, T_c , upper critical field, H_{c2} , gaps width, intra- and inter-band scattering, defect structure, etc. In order to study the influence of intra- and interband scattering and doping on the normal and superconducting properties of MgB_2 , investigation of substituted single crystals are particularly valuable.[1-6] Introducing C, Al, Li, Mn, and Fe into the melt we have grown MgB_2 crystals where Mg or B were partially substituted with iso- or heterovalent and non-magnetic or magnetic ions. Thus the scattering centers were added and/or the crystals were doped with electrons or holes. In this work we study the resistance, R , and thermoelectric power, S , of MgB_2 single crystals substituted with electron-doping C for B and isovalent magnetic Mn^{2+} for Mg. The preliminary results for crystals co-substituted with Al:Li and C:Li are also presented for comparison.

Single crystals of substituted MgB_2 were grown under high pressure using a cubic anvil press. The applied temperature and pressure conditions to obtain C, Al, and Mn substituted crystals were described in Ref. 3-5. The successful method to grow Li, Al:Li, and C:Li substituted crystals has been presented in Ref. 7. The C content in the crystals was estimated from changes in the a lattice parameter. The contents of Al, Mn and Fe were determined with EDX. The Li content was determined from the structure refinement.[7] The methods to measure R and S were described in details in Ref. 3 and 8, respectively.

Normalized R versus temperature is shown in Fig. 1 for the $\text{Mg}(\text{B}_{1-x}\text{C}_x)_2$ single crystals with $x = 0$ and 0.05. The resistance ratio RRR defined as $R(300\text{ K})/R(40\text{ K})$ decreases from about 7, observed for the non-substituted ($x = 0$) crystal, to $\text{RRR} = 2$ and 1.4 obtained for crystals with $x = 0.05$ and 0.095 (not shown in the Figure), respectively. This nonlinear decrease of RRR with the carbon content may result in a resistance saturation effect, which can be attributed to the created by substitution microscopic defects acting as electron scattering centers. Thus, the increase of R with the carbon content is not a simple effect of

electron doping but also the scattering mechanism should be considered as an important one. This conclusion is supported by the $S(T)$ results, which show a decrease of S with C content indicative of more metallic character of the substituted crystals. The $S(T)$ results for the $\text{Mg}(\text{B}_{1-x}\text{C}_x)_2$ crystals with $x = 0$ and 0.05 are shown in Fig. 2. As thermoelectric power is only moderately sensitive to the microscopic defects, the main reason for the observed changes of S with the substitution is the electron doping. This doping reduces considerably the absolute value of S and also its anisotropy, defined as a ratio S_{ab}/S_c , where S_{ab} and S_c are results obtained with a temperature gradient along the ab plane and the c axis, respectively. For the non-substituted crystals, $S_{ab}/S_c \cong 3$ at 300 K and this ratio decreases to about -1.4 for the substituted crystal with $x = 0.05$. Note, that for this crystal, S_c is negative suggestive of the significant change in the carrier distribution.

The influence of the C substitution on thermoelectric power is summarized in Fig. 3. The main effect is a rapid reduction of both S and S_{ab}/S_c , for the MgB_2 crystals with a moderate amount of substituted C ($x = 0.05$, $T_c = 34.5$ K), and then saturation of these quantities for higher substitutions. At this stage of our studies we are not able to interpret that effect. Figure 3 shows also S_{ab} for C:Li co-substituted crystals, where electrons doped with C, which is substituted for B, seem to be partially compensated with holes added with Li, which is co-substituted for Mg.[7] The results fit the $S_{ab}(T_c)$ dependence well suggestive of T_c as a parameter that characterizes the transport properties of MgB_2 adequately. Interestingly, the $S_{ab}(T_c)$ dependence is not followed by results obtained for Al:Li co-substituted crystals. This can be explained by the fact that Al substituted for Mg may dope electrons into the π band, so this substitution acts in different way. The co-substituted Li does not change T_c of the Al-substituted crystals, so the Li substitution is negligible in this case.

Finally we studied the $S(T)$ properties of MgB_2 crystals substituted with isovalent magnetic ions. Figure 4 shows $S_{ab}(T)$ and $S_c(T)$ for $\text{Mg}_{1-x}\text{Mn}_x\text{B}_2$ with $x = 0.007$ and $T_c = 29.3$ K. Surprisingly, both the values and the temperature dependences of S_{ab} and S_c are very

similar to those obtained for the non-substituted crystal, despite of the drastic reduction of T_c . This can be explained by the fact that the Mn^{2+} ions substituted for Mg are isovalent and that the very small amount of substituted Mn can create only a small amount of the electron scattering centers. The main effect that reduces T_c is magnetic pair-breaking, which does not influence the electronic structure and thus can not modify the $S(T)$ properties.

In conclusion, availability of sizeable MgB_2 crystals doped with electrons and holes enable us to perform advanced studies of their superconducting and normal-state properties including transport characteristics. Both resistance and thermoelectric power are strongly influenced by doping when C is substituted for B. This substitution, which introduces electrons into the σ band, results in a large increase of R , most likely due to the creation of additional scattering centers. On the other hand, for the C-substituted MgB_2 , S decreases with the substitution and even changes a sign to negative indicative of more metallic and electron-conducting character of substituted crystals. Another interesting observation is that the substitution of Li for Mg, which introduces holes to the π band, changes the superconducting and transport properties in a much subtle way. Thus, the electron- and hole-doping effects seem to be asymmetric. The substitution of isovalent magnetic Mn^{2+} ions, which drastically influences the superconducting properties like T_c and H_{c2} , does not appear to have any effects on both the value and the temperature dependence of S . This suggests, not surprisingly, that the pair-breaking effect, which is responsible for the rapid reduction of T_c in the Mn substituted crystals, does not modify the Fermi surface to any substantial degree.

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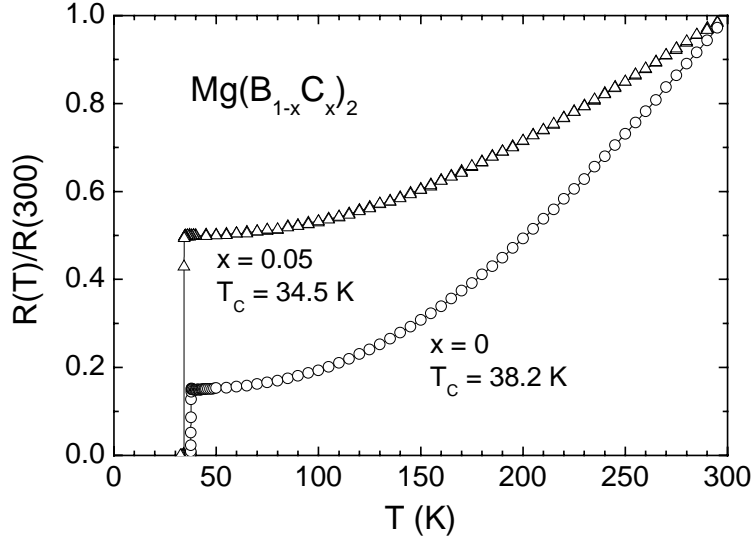


Fig. 1. Temperature dependence of the in-plane resistance ratio $R(T)/R(300)$ for $\text{Mg}(\text{B}_{1-x}\text{C}_x)_2$ crystals, non-substituted (circles) and carbon-substituted with $x = 0.05$ (triangles). The superconducting transition temperature measured with low current ($i = 2.5$ A/cm²) is $T_c = 38.2$ and 34.5 K for the samples with $x = 0$ and 0.05, respectively. Adapted from Ref. 3.

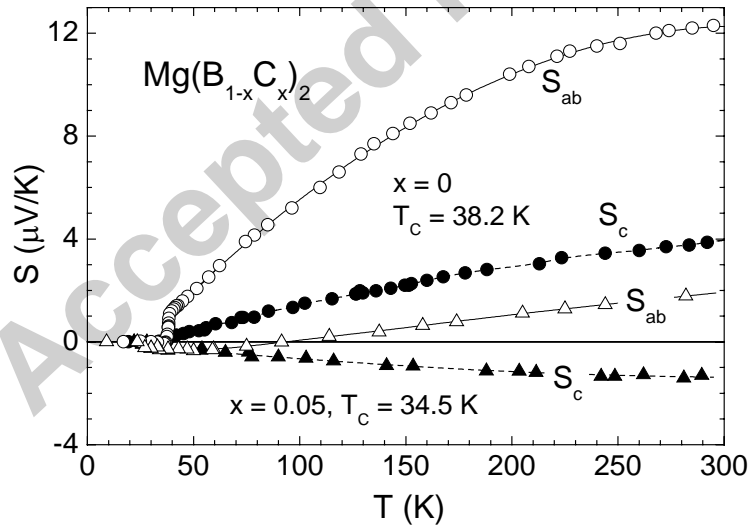


Fig. 2. Thermoelectric power of $\text{Mg}(\text{B}_{1-x}\text{C}_x)_2$ crystals, non-substituted with $T_c = 38.2$ K (circles) and carbon-substituted with $x = 0.05$ and $T_c = 34.5$ K (triangles), measured along the ab plane (S_{ab}) and the c axis (S_c).

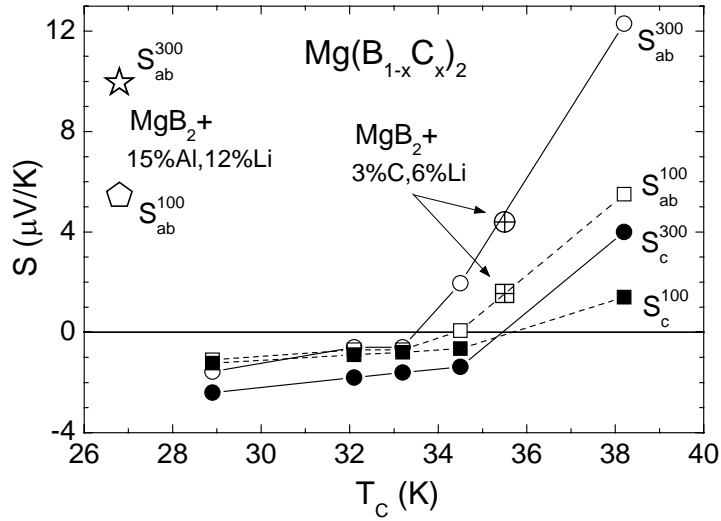


Fig. 3. Thermoelectric power along the ab plane and the c axis at 300 K (circles, star) and 100 K (squares, pentagon) versus T_c of the $\text{Mg}(\text{B}_{1-x}\text{C}_x)_2$ crystals with $0 \leq x \leq 0.1$ (circles, squares), and the MgB_2 crystals substituted with C:Li (crossed symbols) and Al:Li (star, pentagon) to compare. For $\text{Mg}(\text{B}_{1-x}\text{C}_x)_2$ with $0 \leq x \leq 0.09$, T_c decreases with the C content roughly linearly.

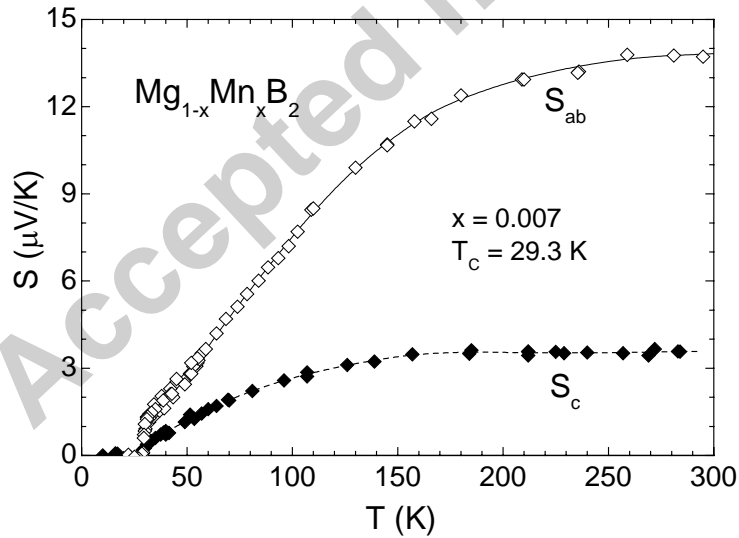


Fig. 4. Thermoelectric power of the $\text{Mg}_{1-x}\text{Mn}_x\text{B}_2$ crystals with $x = 0.007$ and $T_c = 29.3$ K, measured along the ab plane and the c axis. Comparing with the non-substituted crystal, the values of both S_{ab} and S_c are only slightly enhanced despite of the drastically reduced T_c .