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Superconductivity of various borides: The role of stretched *c*-parameter

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The superconductivity of MgB_2 , AlB_2 , NbB_{2+x} , and TaB_{2+x} is intercompared. The stretched *c*-lattice parameter ($c=3.52$ Å) of MgB_2 in comparison to $\text{NbB}_{2.4}$ ($c=3.32$ Å) and AlB_2 ($c=3.25$ Å) decides empirically the population of their π and σ bands and as a result their transition temperature T_c values, respectively, at 39 and 9.5 K for the first two and no superconductivity for the later. The nonstoichiometry induces an increase in *c* parameter with Boron excess both in NbB_{2+x} and TaB_{2+x} . Magnetization (*M-T*) and resistivity measurements (ρ -*T*) in case of niobium boride samples show the absence of superconductivity in stoichiometric NbB_2 sample ($c=3.26$ Å) while a clear diamagnetic signal and a $\rho=0$ transition for boron excess NbB_{2+x} samples. On the other hand, superconductivity is not achieved in TaB_{2+x} case. The probable reason behind is the comparatively lesser or insufficient stretching of *c* parameter. © 2009 American Institute of Physics.

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I. INTRODUCTION

The superconductivity in the family of AlB_2 type diborides like TaB_2 , NbB_2 , and ZrB_2 was boost up with the discovery of MgB_2 superconductor.¹ But in the comparison of MgB_2 , very few reports exist on other diborides; even the existence of superconductivity is suspected in some of the diborides. For example, ZrB_2 is reported to have a T_c of 5.5 K by Gasprova *et al.*,² whereas Leyarovska and Leyarovski³ report no transition. Similarly, Gasprova *et al.* and others^{2–5} have reported no observation of superconductivity in TaB_2 , while Kackzorowski *et al.*⁶ report a transition temperature of 9.5 K. The results for NbB_2 are even more diverse. Gasprova *et al.*,² and Kackzorowski *et al.* and others^{6,7} report no superconductivity, while many others^{3,8,9} report different values of transition temperature in the range of 0.62–9.2 K.

Band structure calculations in MgB_2 reveal that T_c increases with increase in *c* parameter.¹⁰ Working on the same idea, NbB_{2+x} and TaB_{2+x} samples are checked for existence of superconductivity and the systematic comparison in both NbB_{2+x} and TaB_{2+x} is carried out. The thermoelectric power of stoichiometric samples of MgB_2 , AlB_2 , and NbB_2 is also intercompared.

II. EXPERIMENTAL

The polycrystalline samples of MgB_2 , AlB_2 , NbB_{2+x} ($x=0.0$ to 0.8), and TaB_{2+x} ($x=0.0$ to 0.8) were prepared by simple solid-state reaction route. See our Refs. 11 and 12. X-ray diffraction patterns done on Rigaku-Miniflex-II and

Rietveld analysis was done by Fullprof program-2007. For details of other measurements, see Refs. 12 and 13.

III. RESULTS AND DISCUSSION

Figure 1(a) shows the x-ray diffraction patterns for MgB_2 and AlB_2 , while Fig. 1(b) shows the same for NbB_2 , $\text{NbB}_{2.4}$, TaB_2 , and $\text{TaB}_{2.4}$ samples. In order to confirm the phase purity, Rietveld refinement is done for all the samples in the space group *P6/mmm* No. 191. There is hardly any difference between the experimentally observed and theoretically Rietveld determined x-ray profiles except a small MgO peak in case of MgB_2 shown by #. We observe that the (002) peak shifts toward lower angle side with the boron excess in both NbB_2 and TaB_2 cases, which results in increase in *c* parameter. The systematic variation in the parameters can be seen from Table I. There is a slight decrease in *a* parameter with increasing boron content in both NbB_{2+x} and TaB_{2+x} . In case of NbB_{2+x} , *c* parameter increases continuously up to $x=0.4$ and then saturates further with negligible up and downs, but in TaB_{2+x} , *c* parameter increases considerably but only up to $x=0.2$ sample and saturates thereafter. The structural information is in well confirmation with the literature.^{6,14–16} Although the *a* and *c* values for TaB_{2+x} samples match quantitatively with the earlier reports^{5,16} but differ in respect to corresponding compositions. MgB_2 is found to be a superconductor with T_c of about 39 K while AlB_2 is a nonsuperconductor.^{11,13}

Magnetization versus temperature (*M-T*) plot including both zero field cooled and field cooled curves is shown in the main panel of Fig. 2(a) for $\text{NbB}_{2.4}$ sample in the temperature range of 5–12 K. The $\text{NbB}_{2.4}$ sample shows a clear diamagnetic signal at about 9.5 K, implying that it is a superconductor. The lower inset in Fig. 2(a) shows magnetization versus temperature curves for NbB_2 sample in the temperature

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FIG. 1. (Color online) Rietveld refined plots for (a) MgB_2 and AlB_2 samples and (b) NbB_2 , $\text{NbB}_{2.4}$, TaB_2 , and $\text{TaB}_{2.4}$ samples. X-ray experimental diagram (dots), calculated pattern (continuous line), difference (lower continuous line), and calculated Bragg position (vertical lines in middle).

range of 5–300 K. The sample exhibits no diamagnetic signal and hence possesses no bulk superconductivity. The magnetization measurement with varying field at a fixed temperature of 5 K is also done for the superconducting $\text{NbB}_{2.4}$ sample and is shown in the upper inset. Thus, $\text{NbB}_{2.4}$ is a type-II superconductor with the H_{c1} and H_{c2} values of 500 and 1600 Oe, respectively. In this way boron excess increases the c parameter and induces superconductivity in niobium boride sample. All boron excess samples are found to possess superconductivity with different T_c values.¹²

The main panel of Fig. 2(b) shows the ρ - T measurement for $\text{NbB}_{2.4}$ sample, while the inset shows the same for NbB_2 sample. The $\text{NbB}_{2.4}$ sample shows a sharp transition with a T_c onset of 7.5 K. On the other hand the stoichiometric NbB_2 sample just shows metallic behavior from 300 K to about $T=80$ K. After that resistivity becomes almost constant and shows no superconducting transition down to 5 K. Thus ρ - T measurement is in confirmation with the M - T measurement showing that only boron excess sample is superconducting, while the stoichiometric NbB_2 is a nonsuperconductor although T_c onset obtained from magnetization measurement for $\text{NbB}_{2.4}$ is comparatively higher.

After inducing superconductivity in NbB_{2+x} , the same is tried for TaB_{2+x} sample. The magnetization versus temperature measurements (M - T) are shown in Fig. 3 for TaB_{2+x} samples in the temperature range of 5–20 K. The samples do not exhibit any diamagnetic signal confirming that there is no superconductivity below to 5 K. The magnetic moment increases with the decrease in temperature for all the samples. The inset shows the magnetic behavior of $\text{TaB}_{2.4}$ and $\text{TaB}_{2.6}$

samples with varying field at a fixed temperature of 5 K. The magnetic moment increases with the applied field and then saturates at a field of about 4 kOe and a hysteresis is obtained in decreasing direction of field. In this way, a paramagnetic type behavior is shown by both the samples. The magnetic moment of $\text{TaB}_{2.6}$ sample is more than the $\text{TaB}_{2.4}$ sample at a particular field value, which might be due to some magnetic impurity in the boron powder.

Now the point to be discussed is that if increase in c parameter induces superconductivity in NbB_{2+x} , why it does not happen in TaB_{2+x} case? Actually, if we see the values of c parameter in NbB_{2+x} case, it has increased from 3.264 Å for pure NbB_2 to 3.320 Å for $\text{NbB}_{2.4}$ and saturates thereafter. For TaB_2 , c parameter is 3.238 Å, which is less than that of pure NbB_2 . With boron excess, c parameter increases in TaB_{2+x} case also but slightly, i.e., only up to 3.278 Å for $\text{TaB}_{2.2}$. After that, no increase in c parameter is noticed, which implies excess boron cannot be accommodated in the TaB_2 lattice after this limit. Excess boron actually creates metal vacancies in the system as discussed in many theoretical studies.^{17,18} So, we come to the conclusion that although c parameter increases in TaB_{2+x} case, but it is not sufficient to create enough metal vacancies to introduce superconductivity in this system.

Figure 4 shows the variation in thermoelectric power (TEP) of MgB_2 , AlB_2 , NbB_2 samples with temperature. The $\text{Al}^{+3}/\text{Nb}^{+5}$ substitution at Mg^{2+} provides extra electrons and hence filling of the hole type sigma band and resulting electron type conductivity while MgB_2 is a hole type conductor. As mentioned before, the nonsuperconducting behavior of

TABLE I. Lattice parameters and c/a values for NbB_{2+x} and TaB_{2+x} samples with $x=0.0, 0.2, 0.4, 0.6$, and 0.8 .

x	TaB_{2+x}			NbB_{2+x}		
	$a(\text{\AA})$	$c(\text{\AA})$	c/a	$a(\text{\AA})$	$c(\text{\AA})$	c/a
0.0	3.0899(1)	3.2378(2)	1.048	3.1103(1)	3.2640(2)	1.049
0.2	3.0739 (1)	3.2776(2)	1.066	3.1013(1)	3.3051(2)	1.066
0.4	3.0732 (1)	3.2762 (2)	1.066	3.1041(2)	3.3202(1)	1.069
0.6	3.0741(1)	3.2775(1)	1.066	3.1018(1)	3.3195 (1)	1.070
0.8	3.0746(1)	3.2771(2)	1.066	3.1040(2)	3.3172(2)	1.069

FIG. 2. (Color online) (a) The magnetization vs temperature (M - T) plot for superconducting $\text{NbB}_{2.4}$. The lower inset shows the same for NbB_2 while the upper inset shows the M - H plot for $\text{NbB}_{2.4}$ sample. (b) Variation of resistivity with temperature for $\text{NbB}_{2.4}$ sample. The inset shows the same for NbB_2 .

FIG. 3. (Color online) The M - T plot for TaB_{2+x} sample with $x=0.0, 0.2, 0.4$, and 0.6 . The inset shows the M - H plot for $\text{TaB}_{2.4}$ and $\text{TaB}_{2.6}$ samples.

FIG. 4. (Color online) Thermoelectric power vs temperature plots for MgB_2 , AlB_2 , and NbB_2 samples.

NbB_2 and AlB_2 is seemingly due to two facts, i.e., changed carrier density and the c parameters. The detailed analysis of TEP data on the basis of two-band model is done earlier for MgB_2 and AlB_2 .¹³ It is discussed theoretically that the presence of vacancies in the Niobium sublattice of NbB_2 brings about considerable changes in the density of states in the near Fermi region and hence affects the superconductivity.¹⁹

In summary, the c is stretched for nonstoichiometric NbB_{2+x} and TaB_{2+x} samples. Excess boron creates metal vacancy in the lattice and induces superconductivity in niobium boride case, but the increase in c parameter is not sufficient in TaB_2 case and hence the superconductivity is not achieved. The thermoelectric power measurement shows the different types of carriers in different borides.

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