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Ta_5GeB_2: New T_2 superconductor phase

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ABSTRACT

We report superconductivity at T_C ~3.8 K in the new ternary phase Ta_5GeB_2. Bulk superconductivity is confirmed by magnetization, electrical resistivity and heat capacity measurements, the results showing conventional bulk superconductivity. Ta_5GeB_2 is a further example of a stoichiometric T_2 phase with Cr_5B_3 prototype structure.

1. Introduction

M_5Si_3B and M_5Ge_3B (where M is a transition metal) are well known through a variety of physical properties with potential for application at high temperatures [1–3] as well as being an interesting class of materials for more fundamental investigations regarding, for example, superconductivity. Nb_5Si_3 and Nb_5Ge_3 are known to superconduct at 0.7 K [4] and 0.5 K [5] respectively which with doping with small radii elements such as B, N or C have T_C's enhanced to 7.8 K for Nb_5Si_3 and 15.3 K [6] for Nb_5Ge_3 [7]. In this case, the superconducting properties are related to a high temperature phase (with a prototype Mn_5Si_3 with hexagonal symmetry) that is stabilized by the interstitial doping mentioned previously.

The Nb–Si–B ternary system also shows that it is possible to stabilize another phase, the so called T_2 phase, by substitution of Si for B. Samples with composition $Nb_5Si_{3-x}B_x$ show maximum solubility for x = 1 (Nb_5Si_2B) [8]. Interestingly, $Nb_5Si_{3-x}B_x$ was found to be superconducting at certain B doping and it was the first Cr_5B_3 prototype structure phase to be found as a superconductor. Many others compounds with the same prototype structure (Cr_5B_3) were found to be superconductors as well, such as W_5SiB_2, $(W, Ta)_5SiB_2$ and Mo_5SiB_2 [9–11]. In Mo_5SiB_2 the T_2 phase is stoichiometric, and its superconducting properties emerge with just one critical temperature. However, it is common to find other boron-silicide compounds that have the T_2 phase with a solubility limit of Si for B without nucleation of secondary phases and that it is possible to observe that the critical temperature is strongly dependent on the substitution level of Si.

Recent reports propose the existence of stoichiometric phases in the binary system Ta – Ge such as Ta_3Ge, Ta_5Ge_3, [12–15]. Ta_3Ge, for instance, can assume two different prototype structures, α-Ta_3Ge and β-Ta_3Ge. α-Ta_3Ge is stable below 1550 °C and has the Ni_3P prototype structure with tetragonal symmetry [13], while β-Ta_3Ge is stable above 1550 °C and in spite of also having also tetragonal symmetry has the Ti_3P prototype structure [14]. Another interesting phase is Ta_5Ge_3, also known to exhibit two different crystal structures, one stable at high temperatures in the W_5Si_3 prototype with tetragonal symmetry and another stable at room temperature in the Mn_5Si_3 structure with hexagonal symmetry [16]. However when B is add to this system, the new Ta-Ge-B ternary shows two different equilibrium phases with composition of Ta_5Ge_3B and Ta_5Ge_2B at 700 °C [17]. The first one consists of a Mn_5Si_3 prototype with interstitial doping of B, the doping causing this phase to be stable at room temperature. The second one is the so called T_2 phase with Cr_5B_3 prototype structure that is stabilized by substituting Ge for B at the 8 h Wyckoff position. Here we report results of resistivity, magnetization and specific heat as function of temperature that confirms the existence of superconductivity in

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Ta_5GeB_2 as another example of the T_2 phase with Cr_5B_3 prototype structure.

2. Experimental procedure

Polycrystalline samples of $Ta_5Ge_{3-x}B_x$ with $0.2 \leq \times \leq 1.0$ were prepared by solid state reaction. High purity powders of Ta, Ge and B were weighted in stoichiometric amounts, homogenized, pressed into pellets, sealed in a quartz ampoule under Ar atmosphere, heat treated at 1200 °C for 100 h and finally quenched in ice water. Another heat treatment at 1800 °C for 24 h was necessary to achieve a single phase sample within the X-ray diffraction resolution. This treatment was carried out in a resistive furnace (tubular Ta heating element) under argon. X-ray powder diffraction patterns were performed at room temperature with 40 kV–30 mA, Cu-Kα radiation, and Ni filter. The 2θ data were collected from 10 to 90° using a step of 0.05°. X-ray diffraction data were analyzed using Rietveld refinement with the software PowderCell [18] Vesta Crystallography [19] and GSAS [20].

Physical properties were obtained using a commercial VSM-PPMS EverCool II from Quantum Design. Magnetization as a function of temperature was determined with zero field cooling (ZFC) and field cooling (FC) in an applied magnetic field of 50 Oe. Electrical resistivity as a function of temperature was measured using the standard four-probe method from 1.8 to 300 K. Here, we define the superconducting transition temperature (TC) as the temperatures corresponding to a 2% resistivity drop, a 1% magnetization drop in the ZFC measurements and a 1% heat capacity anomaly start from normal state. These measurements were done both without and in applied magnetic field in order to estimate the upper critical field. Specific heat of a polished flat sample with Ta_5GeB_2 composition was measured in the range of 2 K–20 K.

3. Results

Fig. 1 shows the X-ray diffraction pattern of a sample annealed at 1800 °C for 24 h, along with the results of the Rietveld refinement and simulated structure.

The refinement was stable and converged fast. The parameter goodness-of-fit (χ^2) and weighted-profile reliability factor (R_{wp}) were 1.838 and 8.68% respectively, reasonable values for a reliable X-ray diffraction refinement. The peaks observed in 36.4 (in overlap with a peak of the T_2 phase), 40.9 and 63° (pointed out by stars in the Fig. 1) were indexed as Ta_3Ge phase published elsewhere [12] (space group 86 and Ni3P prototype structure). A sample with this composition was prepared in other to investigate if this phase could give rise to the superconductivity observed and regarding the low temperature properties this compound is found to be a Pauli-paramagnetic (data not shown). The difference between the experimental data and the refinement is shown by the blue line in Fig. 1(a). The refinement was performed by starting with the structure published for the Ta_5Ge_3 as α-$TaGe_{0.5}$ [16] (Fig. 1(b) shows the simulated), however, some significant differences in the intensity between the observed and calculated X-ray diffraction patterns were only diminished by considering that B atoms were occupying the 8 h Wyckoff position (Fig. 1(c) shows the simulated structure). It is important to mention that neutron diffraction would be necessary to confirm that the B atoms are truly in these sites. The published data for Ta_5Ge_3 a = b = 6.599 Å, c = 12.01 Å for the lattice parameters and space group I4/mcm (140). Our results for the same space group are a = b = 6.239 Å, c = 11.578 Å for the lattice parameters and with B atoms in the 8 h Wyckoff position one expects to see smaller lattice parameters since the B ionic radius is smaller than the Ge ionic radius. It is important to mention that this was the only composition where single phase tetragonal T_2 phase

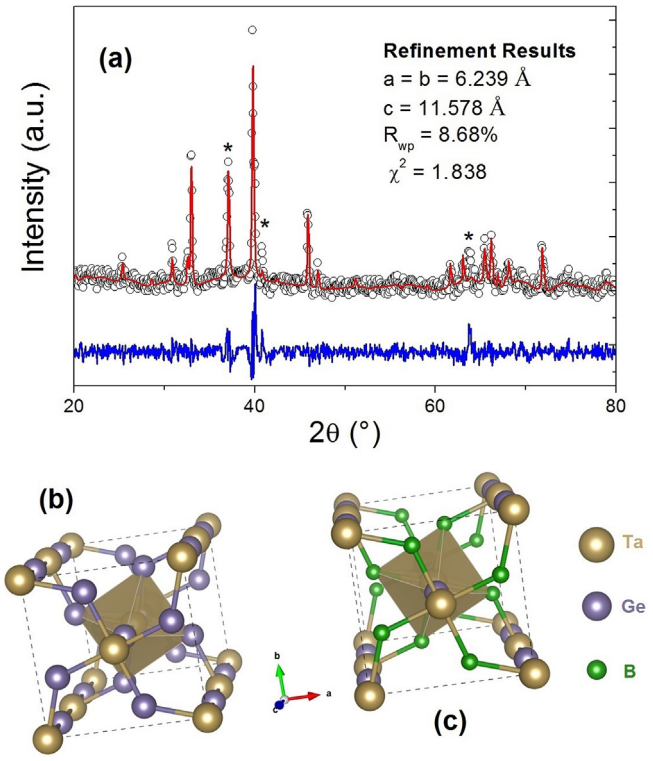

Fig. 1. (a) X-ray diffraction data. (b) Simulated structure of Ta_5Ge_3 and (c) simulated structure considering B atoms substituting Ge atoms of the 8 h Wyckoff position.

with Cr_5B_3 prototype structure were observed. This behavior also occurs in the Mo–Si–B system [11]. These results suggest that this is another example of a stoichiometric T_2 phase. In Nb–Si–B system the solubility range of the substitution of Si for B is relatively large [6].

Magnetization as a function of temperature is shown in Fig. 2.

Fig. 2 displays a superconducting transition close to 3.8 K in the ZFC and FC regimes and the inset shows the M vs H dependence at 2.0 K with the typical signature of type II superconductivity. The estimated superconducting volume by ZFC curve is around 80% which strongly suggests a bulk superconducting state. Resistivity as a function of temperature measurements also shows a

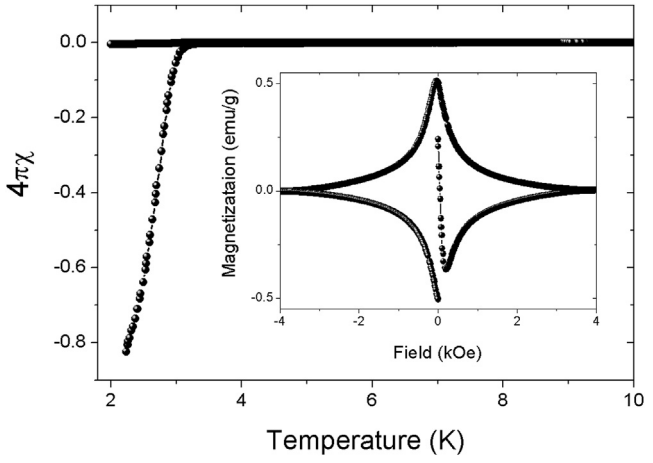

Fig. 2. Magnetization as a function of temperature in the ZFC and FC regimes showing the superconducting critical temperature close to 3.8 K. The inset shows the M vs H at 2.0 K suggesting a type II superconductor.