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Effect of Pt on the superconducting and magnetic properties of $ErNi_2B_2C$☆

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ABSTRACT

We show that the variation of T_c in $Er(Ni_{1-x}Pt_x)_2B_2C$ ($T_c \sim 10.6$ K and $T_N \sim 5.7$ K for $x = 0$) as a function of x proceeds in two steps: strong decrease of T_c for initial values of x ($0 \le x < 0.10$, $T_c = 7.3$ K at $x = 0.1$) and, thereafter, a relatively much weaker drop (almost a plateau) of T_c with further increase of x. T_N exhibits a slight, almost linear, decrease over the entire range of x studied here; $T_N = 4.7$ K for $x = 0.2$. Our results for $x = 0.10$ are in sharp disagreement with the results, namely, $T_c < T_N$, as reported by Felner et al. [I. Felner, D. Schmitt, B. Barbara, C. Godart, E. Alleno, J. Solid State Chem. 133 (1997), 5].

1. Introduction

The quaternary borocarbide superconductors RNi_2B_2C (R = rare earth atoms) exhibit a variety of phenomena associated with superconductivity, magnetism and their interplay [1,2]. Nonmagnetic borocarbide superconductors YNi_2B_2C and $LuNi_2B_2C$ have a highly anisotropic superconducting energy gap (point nodes) [3,4]. Introduction of Pt-atoms at the Ni-sites creates remarkable modifications of the superconducting properties: the superconducting gap becomes isotropic and the material is pushed from the clean to the dirty limit. For example, variation of electronic specific heat C_{el} as a function of temperature in the superconducting state in pure YNi_2B_2C follows a power law, $C_{el} \sim (T/T_c)^3$ (anisotropic energy gap) whereas it exhibits an exponential T-dependence in the material $YNi_{1.8}Pt_{0.2}B_2C$ (isotropic energy gap) [5,6]. T_c is not affected significantly with the introduction of Pt ($T_c \sim 12.2$ K in $YNi_{1.8}Pt_{0.2}B_2C$; $T_c = 15.5$ K in YNi_2B_2C); however, variation of the upper critical field H_{c2} as a function of x clearly shows that the material passes from the clean limit to the dirty limit with the increase of x [7].

Felner et al. [8] studied the effect of Pt-doping in magnetic superconductors $ErNi_2B_2C$ and $TmNi_2B_2C$. In $Er(Ni_{1-x}Pt_x)_2B_2C$ for $x = 0.1$, T_c was observed to decrease to ∼8 K whereas T_N was found to *increase* to ∼11 K. They emphasized the significance of their observation $T_c < T_N$ in this material, namely, the magnetic transition temperature T_N (∼11 K) was found to be *higher* than the superconducting transition temperature T_c (∼8 K). They also pointed out that they observed similar phenomenon in $Er(Ni_{1-x}M_x)_2B_2C$ with M = Pd and Co. As there are not many materials known with $T_c < T_N$ ($DyNi_2B_2C$ is one of them, see Refs. [1,2]), we were motivated by such considerations to undertake the investigations of the system $Er(Ni_{1-x}Pt_x)B_2C$.

2. Experimental

Polycrystalline samples of $Er(Ni_{1-x}Pt_x)_2B_2C$ ($x = 0.0$, 0.05, 0.10, 0.15, 0.20) were synthesized by standard arc-melting under protective argon atmosphere. The alloys were melted several times, turning them over before each melting as is usually done. For annealing, the samples were wrapped in tantalum foil and then sealed in evacuated quartz tubes. The samples were annealed at 1000 °C for 10 days and at 1100 °C for 1 day followed by quenching in cold water. The phase purity was checked by X-ray diffraction. Superconducting transition temperature T_c was measured by ac-susceptibility. Specific heat measurements were carried out to determine the magnetic ordering temperature T_N. Both measurements were performed in a Physical Properties Measurement System (PPMS) with a 14-T magnet from Quantum Design.

3. Results and discussion

Our samples $Er(Ni_{1-x}Pt_x)_2B_2C$ ($x = 0.0$, 0.05, 0.10, 0.15, 0.20), as characterized by X-ray diffraction, are essentially single phase materials. Fig. 1 shows the plot of a and c lattice constants as a function of x. It is interesting to note that both a and c increase almost linearly with x. In the series RNi_2B_2C, as the rare earth radius decreases (from La to Lu), c increases and a decreases. This shows

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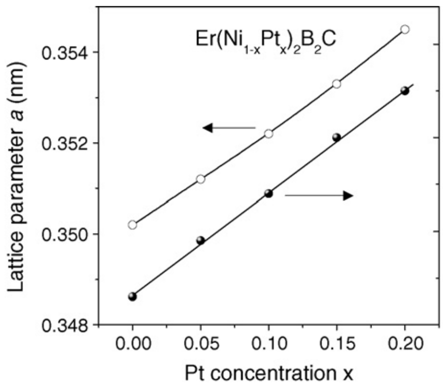

Fig. 1. Lattice constants *a*, *c* for the series $Er(Ni_{1-x}Pt_x)_2B_2C$ ($x = 0.00, 0.05, 0.10, 0.15, 0.20$). Notice that both *a* and *c* increase linearly with *x*.

that the RNi_2B_2C-lattice responds *differently* as to the variation of the R-size and the variation of the effective M-size.

Fig. 2a and b shows the results of ac-susceptibility and specific heat measurements of our Pt-free sample $ErNi_2B_2C$, respectively. The superconducting transition is sharp, suggesting good quality of the sample. The specific heat measurements shows not only the antiferromagnetic transition at $T_N \sim 5.7$ K, but also an anomaly at $T_c \sim 10$ K. Figs. 3 and 4 show the ac-susceptibility and specific heat data of the samples $Er(Ni_{1-x}Pt_x)_2B_2C$ ($x = 0.0, 0.05, 0.10, 0.15, 0.20$), respectively. ac-Susceptibility measurements suggest quite sharp superconducting transitions in all these samples, indicating their good quality. It may be noted that there is no discernible feature in the ac-susceptibility curve which can be associated with magnetic ordering. This is because of the shielding of magnetic signal due to the superconducting state of the material. This implies, and it is well

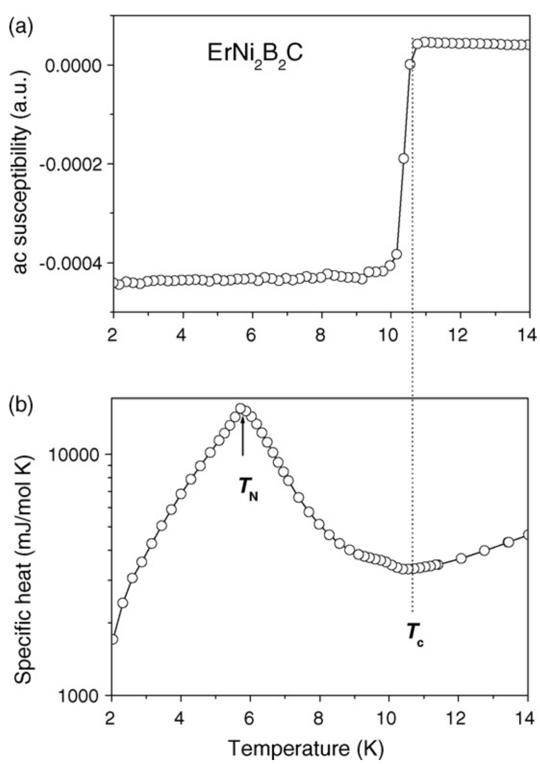

Fig. 2. (a) ac-Susceptibility measurements of the diamagnetic (superconducting) response of the sample $ErNi_2B_2C$ and (b) specific heat of $ErNi_2B_2C$, displayed in a semi-log plot, as a function of temperature. There is a huge anomaly at T_N. A weak anomaly corresponding to the superconducting transition is also discernible in the figure.

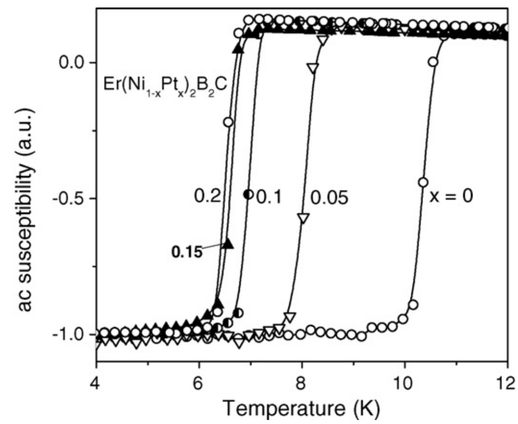

Fig. 3. Temperature dependence of ac-susceptibility in $Er(Ni_{1-x}Pt_x)_2B_2C$ ($x = 0.00$, 0.05,0.10, 0.15, 0.20) showing their T_c clearly. Notice that T_c decreases relatively rather slowly for the last two samples, $x = 0.15$ and 0.20.

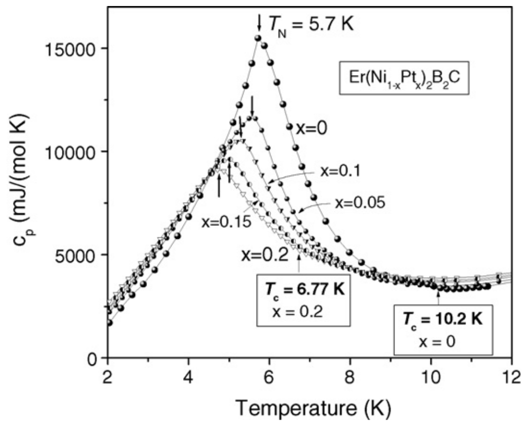

Fig. 4. Temperature dependence of specific heat c_p in $Er(Ni_{1-x}Pt_x)_2B_2C$ ($x = 0.00$, 0.05,0.10, 0.15, 0.20). T_N is defined by the peak of c_p marked by thick arrows.

known, that low field magnetization measurements are not good probe for the antiferromagnetic transition in the superconducting state. Antiferromagnetic ordering temperature T_N is taken as the temperature of the peak of the specific heat curve in each case.

Fig. 5 shows the plot of T_c and T_N as a function of *x*. From this plot, it is clear that T_c decreases rather rapidly for $0 < x < 0.1$. This is in agreement with the reported results [9] and is understood as a consequence of lattice expansion. There is no substantial decrease in T_c for higher values of *x*. This is particularly noteworthy in view of the fact that the lattice constants increase steadily as a function of

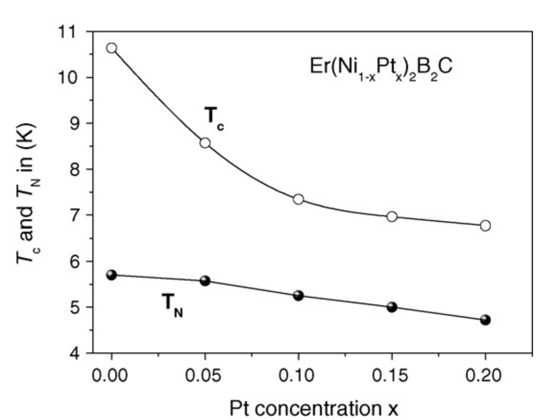

Fig. 5. T_c and T_N in $Er(Ni_{1-x}Pt_x)_2B_2C$ as function of the Pt concentration *x*.

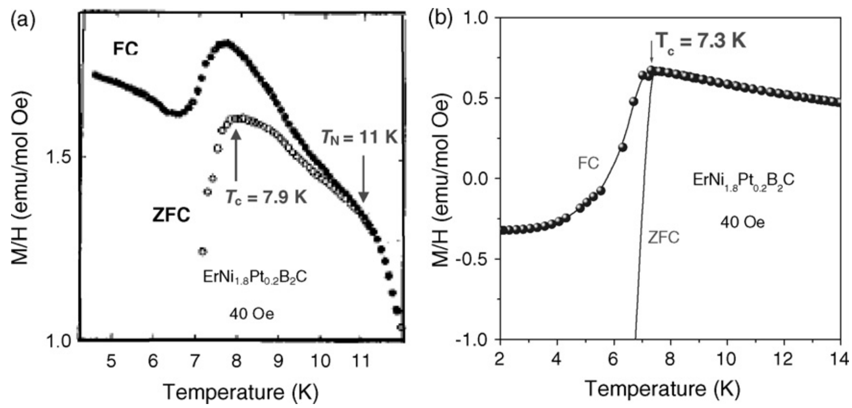

Fig. 6. dc-Magnetization in an applied field of 40 gauss as a function of temperature in $ErNi_{1.8}Pt_{0.2}B_2C$. (a) Reproduces Fig. 9 of Ref. [8]. Several features, such as no diamagnetic response in the superconducting state, etc. in this figure are commented upon in the text. (b) Shows, for comparison, results of measurements of magnetization under the same physical conditions as that in (a).

x and that should affect the density of states at the Fermi level. We consider this behaviour as suggesting that the hypothetical phase $ErPt_2B_2C$ (hypothetical because so far $ErPt_2B_2C$ has not been reported) would have $T_c \sim 7$ K. The plateau-like behaviour observed for $x > 0.1$ seems to be a trade-off between the decrease of T_c due to the reduction of the density of states and the increase of T_c due to the reduced Abrikosov–Gorkov pair-breaking which is expected for a reduced density of states. Our data of T_N, Fig. 5, unambiguously show that T_N does *decrease*, *though slightly*, *with increasing Pt-concentration.*

Now we comment upon the measurements of T_N in the samples $Er(Ni_{0.9}Pt_{0.1})_2B_2C$ as reported by Felner et al. [8]. Fig. 6a reproduces Fig. 9 of their publication [8] which shows the results of their dc magnetization in zero field cooled (ZFC) and field cooled (FC) conditions. Strangely, there is no diamagnetic response below T_c, both in FC- as well as ZFC-conditions. They identify T_c as the temperature that corresponds to the peak of the magnetic response. The FC- and the ZFC signals merge at a temperature higher than T_c and they call this temperature as the antiferromagnetic ordering temperature T_N, the onset temperature of the irreversibility which they considered arose due to magnetic ordering. Obviously, this “T_N” would be higher than T_c. Fig. 6b shows our results of dc magnetization carried out under identical conditions. We see a clear diamagnetic response in the superconducting state. The FC- and ZFC-signals are identical above T_c as expected and no irreversibility between FC- and ZFC-responses above T_c. We have no ambiguity in identifying T_c. We measured T_N through specific heat studies which revealed T_N clearly. Our measurements show that $T_N < T_c$, ruling out the exotic possibility $T_c < T_N$. Their assertion ‘that in the $ErNi_{1.8}M_{0.2}B_2C$ system, the same $T_N = 11$ K is obtained regardless of M (Pt, Pd, Co)’ makes one imagine that their samples had some common impurity phases influencing their findings. Felner et al. [8] reported that their samples were annealed at 800 °C for about 2 weeks, whereas our samples were annealed at 1000 °C for 10 days and at 1100 °C for 1 day as mentioned above. Thus, the different annealing temperatures used to prepare these samples might explain the differences in the dc susceptibility data found for these samples (see Fig. 6). Possibly, our annealing procedure results in samples of better quality.

In conclusion, we have measured T_c and T_N of several members of the system $Er(Ni_{1-x}Pt_x)_2B_2C$ ($x = 0.0, 0.05, 0.10, 0.15, 0.20$). For the early members of this series, T_c decreases rapidly as function of x and then it reaches a plateau. Our results on T_c are in agreement with those published in literature [9]. Our results regarding T_N are in strong disagreement with those of Felner et al., namely, in our case $T_c > T_N$ whereas Felner et al. reported $T_c < T_N$ for $x = 0.10$. This disagreement perhaps arises due to an inappropriate method of measuring T_N and the presence of some impurity phases in their samples. We also suggest the hypothetical compound $ErPt_2B_2C$ to have $T_c \sim 7$ K.

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