Competition of multiband superconducting and magnetic order in $ErNi_2B_2C$ observed by Andreev reflection.

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Abstract. - Point contacts (PC) Andreev reflection dV/dI spectra for the antiferromagnetic ($T_N \simeq 6 \text{ K}$) superconductor ($T_c \simeq 11 \text{ K}$) $\text{ErNi}_2\text{B}_2\text{C}$ have been measured for the two main crystallographic directions. Observed retention of the Andreev reflection minima in dV/dI up to T_c directly points to unusual superconducting order parameter (OP) vanishing at T_c . Temperature dependence of OP was obtained from dV/dI using recent theory of Andreev reflection including pair-breaking effect. For the first time existence of a two superconducting OPs in $\text{ErNi}_2\text{B}_2\text{C}$ is shown. A distinct decrease of both OPs as temperature is lowered below T_N is observed.

Introduction. – The family of quaternary nickel borocarbides superconductors RNi_2B_2C , where R is a rare-earth element or Y, has attracted worldwide attention both because of a relatively high critical temperature T_c , up to 16 K for R=Lu, and especially from the point of view of competition between superconducting and magnetic ordered states in the case of R=Tm, Er, Ho, Dy, where energy scales for the antiferromagnetic and superconducting order can be varied over a wide range (see, e.g., Refs. [1,2] and further Refs. therein). The compound with R=Er and $T_c \simeq 11$ K is interesting for two reasons [1]: below $(T_N \simeq 6 \text{ K})$ incommensurate antiferromagnetic order with spin density wave occurs and weak ferromagnetism develops below $T_{WFM} \simeq 2 K$ [3]. Both phenomena are, in general, antagonistic to superconductivity, so that competition between superconducting and the magnetic state should take place in this compound. Additionally, the superconducting ground state in borocarbide superconductors is expected to have a multiband nature [4, 5] with a complex Fermi surface and different contributions to the superconducting state by different Fermi surface sheets. Therefore, determining the influence of these magnetic states on a possible multiband superconducting ground state or multiband order parameter (OP) in $ErNi_2B_2C$ is

a challenge.

Previous tunneling (STM/STS) and point contact (PC) spectroscopy results have left some open questions regarding the coexistence of superconductivity and magnetism in ErNi₂B₂C. STM/STS measurements of [6] show a small feature, namely, decreasing of the superconducting gap below T_N nearly within error bars, which was not reproduced in subsequent experiments [7]. Early PC data on polycrystalline samples [9] indicated that the superconducting gap has roughly a BCS dependence with only a shallow dip around $T_N \simeq 6 \text{ K}$. Very recent laser-photoemission spectroscopy data [8] show the SC gap decrease (with remarkably large error bars) below the Neel temperature, but, at present the laser-photoemission spectroscopy has not enough resolution to go deeper in details.

In this paper we report our detailed directional PC Andreev reflection measurements on single crystal $\text{ErNi}_2\text{B}_2\text{C}$ along the c-axis and in the ab-plane. Our results show for the first time the presence of two dominating OPs in $\text{ErNi}_2\text{B}_2\text{C}$, which differ by a factor of about two, and appreciable depression of both OPs by the antiferromagnetic transition is found.

Experimental details. – We have used single crystals of ErNi₂B₂C grown by the Ames Laboratory Ni₂B high-temperature flux growth method [10]. PCs were established both along the c axis and in perpendicular direction by standard "needle-anvil" methods [11]. The ErNi₂B₂C surface was prepared by chemical etching or cleavage as described in [12]. As a counter electrode, edged thin Ag wires ($\otimes = 0.15 \,\mathrm{mm}$) were used to improve mechanical stability of PCs in comparison to use of a bulk Ag piece. We have measured the temperature dependence of dV/dI(V) characteristics of such N-S PCs (here N denotes a normal metal and S is the superconductor under study) in the range between $1.45 \,\mathrm{K}$ and T_c for several contacts oriented both along the c-axis and in the ab-plane. In the paper we demonstrate results of analysis of 60 dV/dI(V)along the ab-plane measured for the same PC at different temperatures between 1.45 K and 11 K and of 46 dV/dI(V)along the c-direction for another PC 1 in the same temperature range.

Results and discussion. - To determine the OP from the measured differential resistance curves dV/dI(V)we used recent theory [13] of Andreev reflection in PC, which includes the pair-breaking effect by magnetic impurities. The last assumption is reasonable, because of the presence of the local magnetic moments of Er ions. The fit of the measured curves using equations as (1) in [14]has been performed. As fit parameters the superconducting OP Δ^2 , the pair-breaking parameter $\gamma = 1/(\tau_s \Delta)$ (here τ_s is the spin-flip scattering time) and the dimensionless barrier parameter Z have been used. Although the dV/dI curves shown in Fig. 1 exhibit one minimum for each polarity, as in the case of ordinary one gap superconductors [11,15], to fit dV/dI in full we had to use a two-OP(gap) approach ³, adding the corresponding conductivities as made in [12] for LuNi₂B₂C. The contributions of these conductivities account for the part of the Fermi surface containing a particular OP. Thus, for the two-OP model, experimental curves are fitted 4 by the



Fig. 1: (Color online) Raw dV/dI curves of ErNi₂B₂C–Ag PCs in the ab-plane and in the c-direction at indicated temperatures. For clarity, only several representative curves from the total 60 for the ab-plane and 46 for the c-direction measured at different temperatures are shown.

following expression:

$$\frac{dV}{dI} = \frac{S}{K\frac{dI}{dV}(\Delta_1, \gamma_1, Z_1) + (1 - K)\frac{dI}{dV}(\Delta_2, \gamma_2, Z_2)}$$
(1)

Here, the coefficient K reflects the contribution of the part of the Fermi surface having the OP Δ_1 , S is the scaling factor to match the amplitude of the calculated and the experimental curves.

Before discussing the fitting results, we point out the unusual specific behavior of the measured dV/dI ⁵ (see

¹The PC resistance is 36 Ω along the c-axis and 10.5 Ω in the ab-plane. The PC diameter estimated by Wexler formula (see [11], pages 9, 31) is about 7 nm and 14 nm, respectively, using $\rho l \simeq 10^{-11} \Omega \text{ cm}^2$ [4]. At the same time a mean free path *l* is 28 nm, using $\rho \simeq 3.5^{-6} \Omega$ cm [10] just above T_c. Therefore mentioned PCs are close to the ballistic limit d < l.

²Assuming that pair breaking is by magnetic impurities, the energy gap Δ_0 and the OP Δ are related as follows: $\Delta_0 = \Delta(1 - \gamma^{2/3})^{3/2}$ [13].

³Not only does the one OP approach give a worse fit (especially at minima position and at maximum, see insets in Fig. 2) of the experimental data such that the rms deviation is 2-3 times higher compared to the two OP fit, it also requires a varying Z parameter. On the contrary, at two OP fit Z parameters remain constant, equal to 0.77 and 0.6 for the ab-plane and c-direction, respectively. This is important because there is no physical reason for the barrier parameter Z to be temperature dependent.

⁴Before the fit, dV/dI curves were normalized to the dV/dI curve measured above T_c and symmetrized. The fit was done between \pm 8 mV, to avoid contribution from the phonons seen, e.g., as an inflection point around 10 mV for some curves in Fig. 1. The fit was done in two stages. At first we keep both $\gamma_{1,2}$ coefficients equal to

zero. As a result of such fit $\Delta_{1,2}$ values shown in Fig. 3 were obtained, while variation of Z and K was within 10%. If we will hold K on this stage strictly constant, it results only in more scatter (noise) for $\Delta_{1,2}$, but their overall behavior remains the same. To improve the fit on the second stage, we used obtained $\Delta_{1,2}$ and variate $\gamma_{1,2}$ and K. As a result, obtained K and $\gamma_{1,2}$ are shown in Figs. 4 and 5. After this the theoretical curves were almost indistinguishable from the experimental ones (see insets in Fig. 2).

⁵Presented in the paper PCs have survived about 36 hours (c-dir) and 50 hours (ab-plane) of measurements. The dV/dI temperature series for these PCs are the most full, therefore they presented in



Fig. 2: (Color online) Reduced position of the minima in the raw dV/dI curves for ErNi₂B₂C and that for LuNi₂B₂C from [12]. Insets: comparison of two OPs (solid line) and one-OP (dashed line) fitting of the reduced experimental dV/dI(symbols) at 1.45 K.

Fig. 1). First, the distance between dV/dI-minima shown in Fig. 2, which is often taken as a rough estimation of the superconducting gap value, increases with temperature before decreasing on approaching T_c – quite different behavior from the nonmagnetic LuNi₂B₂C. Second, the dV/dI-minima for ErNi₂B₂C persist up to temperatures close to T_c (see Fig 2, upper panel) though with a small amplitude, leading to a supposition of the presence of the second OP. From these direct observations, a nontrivial behavior of the superconducting OP parameters are expected in ErNi₂B₂C.

Indeed, from the two-band model fitting both OPs Δ_1 (large) and Δ_2 (small) diminish on entering the antiferromagnetic state around 6 K (see Fig. 3). Qualitatively the same behavior has OP determined by one OP fit (Fig. 3, triangles). This is qualitatively consistent with the temperature dependence of the superconducting gap determined by tunneling in [6], by laser-photoemission spec-



Fig. 3: (Color online) Temperature dependence of the large OP Δ_1 (circles), small OP Δ_2 (squares) and OP Δ determined by one OP fit (triangles) for ErNi₂B₂C for the two main crystallographic directions. In the upper panel closed (open) symbols show OPs determined during increasing (decreasing) temperature. The same meaning have closed (open) symbols in Figs. 4 and 5.

troscopy data [8] and also with the upper critical field [2, 16] and the superconducting coherence length behavior [17] in the vicinity of T_N . The theories of coexistence of superconductivity and antiferromagnetic state also predict such OP suppression below T_N , ⁶ e. g., by antiferromagnetic molecular field [18].

Further, the large OP $\Delta_1(T)$ may be described by a BCS dependence above $T_N \simeq 6 \text{ K}$ in the paramagnetic state with extrapolated $T_c^* \simeq 14.5 \text{ K}$, close to that of nonmagnetic $R \text{Ni}_2 \text{B}_2 \text{C}$ (R=Lu, Y). On the other hand retention of the Andreev reflection minima in dV/dI up to T_c (see Fig. 2) result in unconventional abrupt $\Delta_1(T)$ vanishing near T_c . Note, that to fit experimental curves, not only OP but also large OP relative contribution K(see Eq. (1)), must be temperature dependent (see Fig. 4). Shown in Fig. 4, the decrease of K with temperature points

the paper. Of course, there were other PCs with dV/dI of lower quality or which did not survive temperature sweep in the whole range between 1.45 K and T_c. Nevertheless, there were a few of PCs which had dV/dI similar to the presented in the paper, supporting our observations.

 $^{^6}$ Various theories of antiferromagnetic superconductors including the affect of spin fluctuations, molecular field, and impurities on the Δ behavior (see e. g. H. Chi and A. D. S. Nagi, J. Low Temp. Phys. **86**, 139 (1992) and Refs. therein) in support of our observation will be discussed in the forthcoming extended publication.



Fig. 4: (Color online) Temperature dependence of the contribution K (see Eq. 1) of the larger OP to the dV/dI spectra. Curves represent a polynomial fit simply to guide the eye.

to diminution of the "superconducting" part of the Fermi surface with the large OP by approaching T_c , which results in collapse of the large OP at this point.

The mentioned decrease of K correlates with behavior of the pair-breaking parameter γ shown in Fig. 5. It appears that γ_1 is always larger than γ_2 above 2 K, that is the pair-breaking effect is stronger in the band with the larger OP. This is in line with the conclusion, that the different bands are differently affected by magnetic order, made in [5, 19] by band structure analysis of the coexistence of superconductivity and magnetism in the related antiferromagnetic superconductor DyNi₂B₂C, *i.e.*, some bands provide a basis for superconductivity while others are important for the magnetic interactions. Here we should add, that the S parameter in (1) has a maximal value of 0.5 at low temperature, the same value as the normalized zero-bias tunnel conductivity obtained by STS study of $ErNi_2B_2C$ [7]. So, both observations suggest that nearly half of the total Fermi surface (or bands) is nonsuperconducting in ErNi₂B₂C. Formation of a superzone gap at antiferromagnetic transition seen in transport measurements [16] may be responsible for this.

From Fig. 5 it is also seen that γ has a maxima at temperature close to $T_{\rm N}$ and also close to the appearance of weak ferromagnetism around 2 K, which is reasonable. At both of these temperatures an increase in pair breaking is expected due to increasing spin fluctuations accompanying the corresponding transitions.

Conclusion. – This study demonstrates that the two-band approximation with two OPs, including pairbreaking effects, suits better for describing the PC Andreev reflection spectra in $\text{ErNi}_2\text{B}_2\text{C}$ pointing for the first time to presence of multiband superconducting OP in this compound. The values and the temperature dependencies of the large and the small OPs have been estimated for the



Fig. 5: (Color online) Temperature dependence of the pairbreaking parameter γ . Curves represent polynomial fit simply to guide the eye.

ab-plane and in the c- direction. It is found that in the paramagnetic state both OPs can be described by BCS dependence, but formation of the antiferromagnetic state below $T_N \simeq 6 \,\mathrm{K}$ leads to a decrease of both OPs. The pair-breaking effect is found to be different for the large and the small OP indicating that the different bands are affected differently by magnetic order. This may be the reason for the observed abrupt vanishing of the larger OP at T_c . It is interesting that extrapolation of the larger OP by "conventional" BCS behavior above T_N results in $\mathrm{T}_{c}^{*}\simeq\!\!14.5\,\mathrm{K},$ similar to nonmagnetic YNi_2B_2C, so that $2\Delta^{\scriptscriptstyle\rm BCS}(0)/k_{\scriptscriptstyle\rm B}T_c^*\,\simeq\,4.25$ and 4.7 for the ab-plane and cdirection, respectively. BCS extrapolation gives for the small OP $2\Delta^{\text{BCS}}(0)/k_{\text{B}}T_c \simeq 4.1$ (ab-plane) and 3.5 (cdir), while for the one OP fit $2\Delta^{\scriptscriptstyle\rm BCS}(0)/k_{\scriptscriptstyle\rm B}T_c\simeq 4.6$ (abplane) and 4.5 (c-dir) pointing, in general, to moderately anisotropic and strongly coupled superconducting state in $ErNi_2B_2C.$

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