

Laser-excited ultrahigh-resolution photoemission spectroscopy of borocarbide superconductor $\text{RNi}_2\text{B}_2\text{C}$ ($\text{R} = \text{Y}$ and Er)

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Abstract

We present laser-excited ultrahigh-resolution photoemission spectroscopy of $\text{YNi}_2\text{B}_2\text{C}$ and $\text{ErNi}_2\text{B}_2\text{C}$. The extremely high energy resolution enables us to discuss the superconducting (SC) gap function of $\text{YNi}_2\text{B}_2\text{C}$ and to measure the density of states which is coexisted between superconductivity and antiferromagnetism in $\text{ErNi}_2\text{B}_2\text{C}$. For $\text{YNi}_2\text{B}_2\text{C}$, s + g-wave SC gap symmetry cannot reproduce the experimental data, implying importance of taking the complicated electronic structures into consideration. For $\text{ErNi}_2\text{B}_2\text{C}$, the observed coherent peak is very broad and finite density of state exists at Fermi level.

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1. Introduction

The discovery of the rare earth nickel borocarbides $\text{RNi}_2\text{B}_2\text{C}$ ($\text{R} = \text{Y}$ and rare earth) has attracted a great deal of interest [1,2]. For non-magnetic borocarbides ($\text{R} = \text{Y}$ and Lu), various experiments indicate superconducting (SC) gap anisotropy [3–6], although phonon-mediated superconductor [7]. But the mechanism of SC gap anisotropy has not been clarified yet.

On the other hand, magnetic rare earth compounds $\text{RNi}_2\text{B}_2\text{C}$ ($\text{R} = \text{Tm}$, Er , Ho and Dy) exhibit superconductivity coexisting with antiferromagnetism [2]. Their relatively high superconducting transition temperature T_c ($T_c = 11$, 10.5, 8.6 and 6 K, respectively) and Néel

temperature T_N ($T_N = 1.5$, 6.0, 6.3 and 10.6 K, respectively) allow us to investigate the interplay between superconductivity and antiferromagnetism easily. However, study of electronic structures for magnetic superconductor by photoemission spectroscopy (PES) has not been performed because of constrains of energy resolution of photoemission spectrometer and cooling temperature.

Recently, for advance in energy resolution of a photoemission spectrometer by using an ultraviolet laser as a photon source, we can measure the electronic structure near Fermi level (E_F) with a sub-meV energy resolution. Thanking to the long escape depth of photoelectron, the obtained spectra reflect bulk electronic structures [8]. In addition, the improvement of cooling system enable us to measure the spectral at about 3 K.

In this paper, we report laser-excited ultrahigh-resolution photoemission results on anisotropic s-wave

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superconductor $\text{YNi}_2\text{B}_2\text{C}$ and antiferromagnetic superconductor $\text{ErNi}_2\text{B}_2\text{C}$. The SC gap function for $\text{YNi}_2\text{B}_2\text{C}$ and that coexisting with magnetism for $\text{ErNi}_2\text{B}_2\text{C}$ are discussed.

2. Experimental

Single crystals of $\text{Y}(\text{Ni}_{1-x}\text{Pt}_x)_2\text{B}_2\text{C}$ ($x = 0.0$ and 0.2) were prepared by a floating zone method [9]. The dc susceptibility measurement indicated that SC transition temperature T_c of the crystals ($x = 0.0$ and 0.2) are 15.4 K and 12.1 K, respectively. Polycrystals of $\text{ErNi}_2\text{B}_2\text{C}$ were prepared by arc-melting high purity constituents. The dc susceptibility measurement indicated $T_c = 11.0$ K and $T_N = 6.0$ K. The energy resolution of all PES measurements was set to 1.5 meV to obtain good signal-to-noise ratio. All the photoemission measurements have been done for in situ fractured surfaces.

3. Results and discussion

Fig. 1 shows laser-excited PES spectra in the vicinity of E_F for (a) $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$, (b) $\text{YNi}_2\text{B}_2\text{C}$ and (c) $\text{ErNi}_2\text{B}_2\text{C}$.

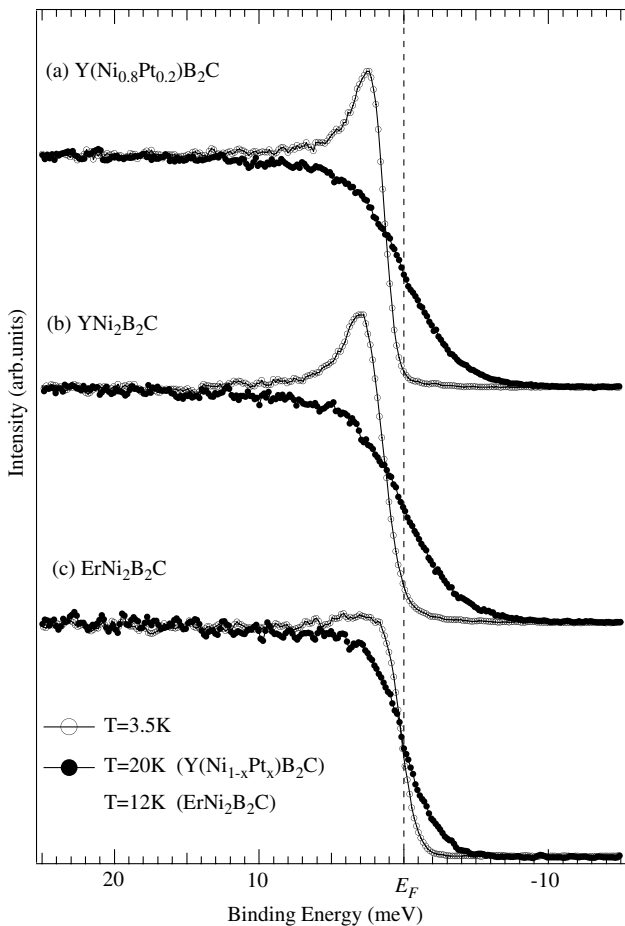


Fig. 1. Laser-excited PES spectra in the vicinity of E_F for (a) $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$, (b) $\text{YNi}_2\text{B}_2\text{C}$ and (c) $\text{ErNi}_2\text{B}_2\text{C}$ measured at 3.5 K (filled circles) and 20 K ($\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$, $\text{YNi}_2\text{B}_2\text{C}$), 12 K ($\text{ErNi}_2\text{B}_2\text{C}$) (open circles).

$\text{ErNi}_2\text{B}_2\text{C}$. The open circles indicate the spectra at superconducting state measured at 3.5 K and the filled circles indicate the normal state measured at 20 K for $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$ and $\text{YNi}_2\text{B}_2\text{C}$, 12 K for $\text{ErNi}_2\text{B}_2\text{C}$, respectively. At the normal state, the spectra have a edge whose midpoint is located at E_F . The spectral shape is well reproduced by the Fermi Dirac (FD) distribution function. In contrast, at 3.5 K, below T_c and T_N , the midpoint of the spectral edge shifts toward higher binding energy with a piling up of a prominent peak in $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$ and $\text{YNi}_2\text{B}_2\text{C}$, while observed superconducting spectral shape is broad in $\text{ErNi}_2\text{B}_2\text{C}$. These indicate the opening of a SC gap below T_c .

3.1. $\text{YNi}_2\text{B}_2\text{C}$

To see the difference between $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$ and $\text{YNi}_2\text{B}_2\text{C}$, we show enlarged spectra near E_F at 3.5 K, in Fig. 2. The spectrum of $x = 0.0$ has a lower peak intensity and a gentler slope of the leading edge with a larger intensity at E_F compared with those of $x = 0.2$. The variation of SC spectra is observed more clearly than those of our previous study [4] owing to the increase of energy resolution.

To verify SC gap function of $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$ and $\text{YNi}_2\text{B}_2\text{C}$, we have tried to fit the spectrum at 3.5 K using Dynes function including a smearing parameter (Γ) [10]. Dynes function were multiplied by the FD function of 3.5 K and convolved with a Gaussian having a full width at half maximum (FWHM) of the known resolution, which is 1.5 meV in this study. Here we suppose the electronic structure is isotropic for simplicity. In Fig. 3(a), we show $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$ spectrum at 3.5 K (open circles) and fitting results with an isotropic s-wave SC gap function (solid curve). The broken lines are the Dynes functions before being multiplied by the FD function and convolved by the Gaussian. With the values of $\Delta = 1.75$ meV and $\Gamma = 0.14$ meV, the fitting result well reproduces the spectrum, indicating that $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$ has an isotropic

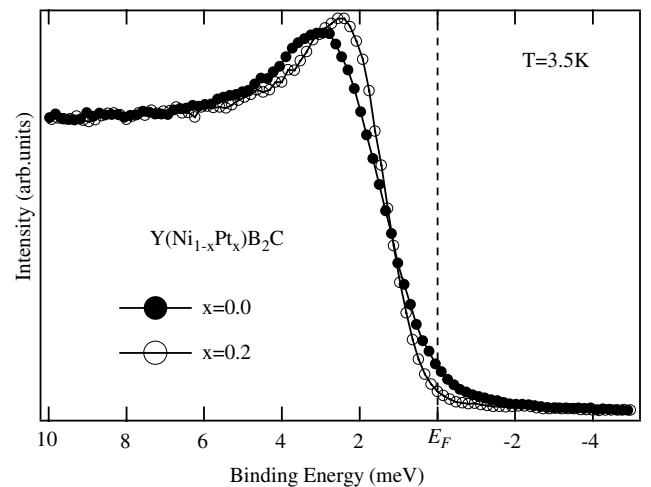


Fig. 2. Photoemission spectra of $\text{YNi}_2\text{B}_2\text{C}$ (filled circles) and $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$ (open circles) at superconducting state.

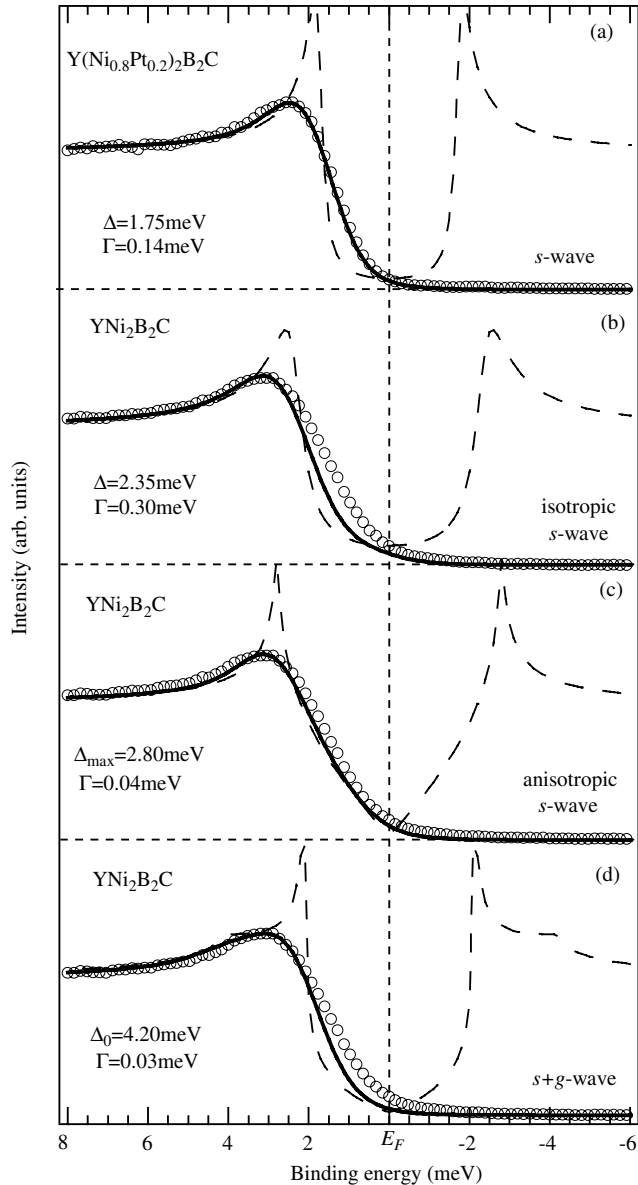


Fig. 3. Numerical calculation results of $Y(Ni_{1-x}Pt_x)_2B_2C$ for (a) $x = 0.2$ and (b)–(d) $x = 0.0$ using (a), (b) isotropic s-wave, (c) anisotropic s-wave, and (d) s + g-wave Dynes functions (connected line) together with the raw spectra of $Y(Ni_{0.8}Pt_{0.2})_2B_2C$ and YNi_2B_2C (open circle). The broken lines are the original Dynes functions.

s-wave SC gap symmetry. In Fig. 3(b)–(d), YNi_2B_2C spectra at 3.5 K are shown together with numerical calculation results with (b) isotropic s-wave, (c) anisotropic s-wave ($\Delta(\theta) = \Delta_{\min} + (\Delta_{\max} - \Delta_{\min})|\cos 2\theta|$ ($\Delta_{\min} = 0$)) and (d) s + g-wave ($\Delta(\theta, \phi) = 1/2\Delta(1 + \sin^4 \theta \cos(4\phi))$) [11]. The fitting parameters were determined to reproduce the spectral shape around the peak. For isotropic s-wave with $\Delta = 2.35$ meV and $\Gamma = 0.30$ meV, one can see the leading edge part is not well reproduced. As to anisotropic s-wave with $\Delta = 2.80$ meV and $\Gamma = 0.04$ meV, the leading edge part is not reproduced very well though the spectrum is better fitted than the isotropic s-wave case. The fact that we can fit the spectrum with anisotropic s-wave better than

with isotropic s-wave in YNi_2B_2C indicates a reduction of SC gap anisotropy in $Y(Ni_{0.8}Pt_{0.2})_2B_2C$ due to disorder and thus provides experimental evidence for anisotropic s-wave SC gap symmetry of borocarbide superconductors [12], consistently with our previous study [4]. For the case of s + g-wave function, we found that we cannot reproduce the spectral edge since it has two peaks, as in Fig. 3(d) by a broken line and Ref. [11]. As an example, we show a fitting for reproducing the peak part with $\Delta = 4.20$ meV and $\Gamma = 0.03$ meV in Fig. 3(d). This suggests that the s + g symmetry might not be a suitable for describing over all SC gap electronic structure of borocarbides. This implies importance of considering the complicated electronic structures of borocarbides, as ignored here, when the s + g wave symmetry is used for analyzing the anisotropic superconducting gap. At least YNi_2B_2C should have an anisotropy on the SC gap. Angle-resolved photoemission spectroscopy will give us more conclusive evidence for the SC gap symmetry.

3.2. $ErNi_2B_2C$

Fig. 4 shows PES spectra of $ErNi_2B_2C$ (filled circles connected with a line) and $Y(Ni_{0.8}Pt_{0.2})_2B_2C$ (open circles) at 3.5 K (SC states) in the vicinity of E_F . For $ErNi_2B_2C$, pronounced coherent peak is not observed and the shift of the midpoint of the leading edge is a little and finite density of state (DOS) exists at E_F . This behavior is in quite contrast to that of $Y(Ni_{0.8}Pt_{0.2})_2B_2C$ which is well fitted by an isotropic s-wave SC gap function. Similar behavior are also observed in STS for single crystal [13]. So, we think that the observed spectral shape is not due to sample quality. According to the Abrikosov–Gor’kov theory [14], a Cooper pair has a finite lifetime τ_s by magnetic impurity scatter-

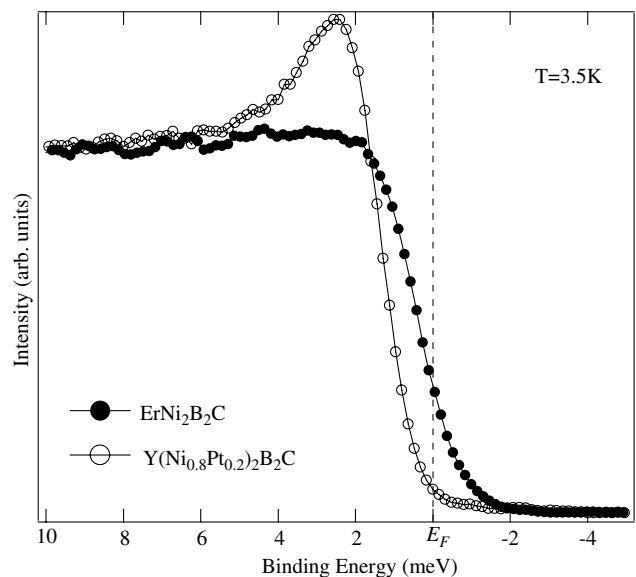


Fig. 4. Photoemission spectra of $ErNi_2B_2C$ (filled circles) and $Y(Ni_{0.8}Pt_{0.2})_2B_2C$ (open circles). Both the spectra are obtained at superconducting state.

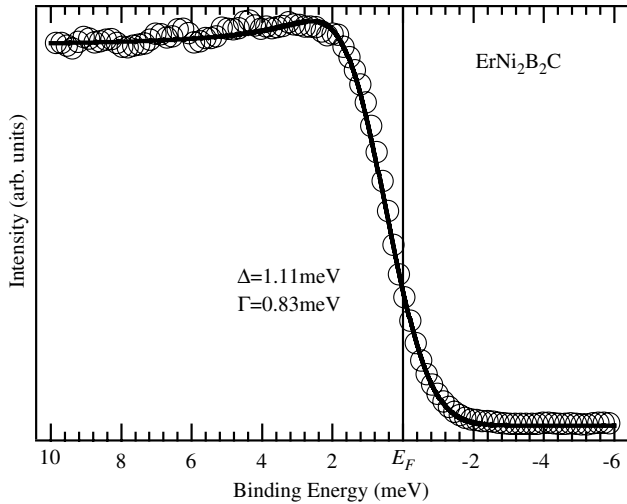


Fig. 5. Numerical calculation results using isotropic s-wave Dynes functions (solid curve) together with the raw spectra of $\text{ErNi}_2\text{B}_2\text{C}$ (open circle).

ing, and therefore this finite lifetime is responsible for an energy spread $\Gamma = \hbar/\tau_s$. For $\text{ErNi}_2\text{B}_2\text{C}$, magnetic ions are arrayed regularly on lattice and density of magnetic ions is higher than that of SC alloys. Thus, we think that the origin of the broadened spectral shape in $\text{ErNi}_2\text{B}_2\text{C}$ is strong magnetic pair breaking effect.

To estimate the values of order parameter Δ and smearing parameter Γ , we have carried out numerical simulations. In Fig. 5, we show $\text{ErNi}_2\text{B}_2\text{C}$ spectrum at 3.5 K (open circles) and the fitting result with an isotropic s-wave Dynes function (solid curve). The fit curve agrees well with experimental data with the values of $\Delta = 1.11$ meV and $\Gamma = 0.83$ meV. The value of Δ is smaller than that of the BCS theory ($\Delta(0) = 1.67$ meV). Theoretically the order parameter Δ in antiferromagnetic superconductors can be reduced below T_N due to the rapid evolution of AF molecular field [15]. Thus, the small value of Δ indicates that superconductivity is suppressed by AF magnetism. And large value of Γ compared to $\text{Y}(\text{Ni}_{0.8}\text{Pt}_{0.2})_2\text{B}_2\text{C}$ might be due to strong magnetic pair breaking effect discuss above.

4. Conclusions

We have performed laser-excited ultrahigh-resolution photoemission spectroscopy to study the SC gap anisotropy on $\text{Y}(\text{Ni}_{1-x}\text{Pt}_x)_2\text{B}_2\text{C}$ ($x = 0.0$ and 0.2) and the coexisting state spectral shape on magnetic superconductor

$\text{ErNi}_2\text{B}_2\text{C}$. For $\text{Y}(\text{Ni}_{1-x}\text{Pt}_x)_2\text{B}_2\text{C}$, we observed the reduction of SC gap anisotropy for $x = 0.2$ in comparison to the $x = 0.0$ case. This result indicates the anisotropic s-wave SC gap is realized in $\text{YNi}_2\text{B}_2\text{C}$. We also found that the spectrum of $x = 0.0$ cannot be fitted well with the s + g SC gap function, implying importance of taking the complicated electronic structures of borocarbides into consideration when the s + g wave symmetry is used for analyzing the anisotropic superconducting gap. For $\text{ErNi}_2\text{B}_2\text{C}$, very broad coherent peak and finite DOS at E_F are observed due to strong magnetic pair breaking effect. Small value of Δ indicates that superconductivity is suppressed by AF magnetism.

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References

- [1] R.J. Cava, H. Takagi, B. Batlogg, H.W. Zandbergen, J.J. Krajewski, W.F. Peak Jr., R.B. van Dover, R.J. Felder, T. Siegrist, K. Mizahashi, J.O. Lee, H. Eisaki, S.A. Carter, S. Uchida, *Nature* 367 (1994) 146.
- [2] K.H. Müller, V.N. Narozhnyi, *Rep. Prog. Phys.* 64 (2001) 943.
- [3] M. Nohara, M. Isshiki, H. Takagi, R.J. Cava, *J. Phys. Soc. Jpn.* 66 (1997) 1888.
- [4] T. Yokoya, T. Kiss, T. Watanabe, S. Shin, M. Nohara, H. Takagi, T. Oguchi, *Phys. Rev. Lett.* 85 (2000) 4952.
- [5] K. Izawa, K. Kamata, Y. Nakajima, Y. Matsuda, T. Watanabe, M. Nohara, H. Takagi, P. Thalmeier, K. Maki, *Phys. Rev. Lett.* 89 (2002) 137006.
- [6] T. Watanabe, M. Nohara, T. Hanaguri, H. Takagi, *Phys. Rev. Lett.* 92 (2004) 147002.
- [7] D.D. Lawrie, J.P. Franck, *Physica C* 245 (1995) 159.
- [8] T. Kiss, F. Kanetaka, T. Yokoya, T. Shimojima, K. Kanai, S. Shin, Y. Onuki, T. Togashi, C. Zhang, C.T. Chen, S. Watanabe, *Phys. Rev. Lett.* 94 (2005) 057001.
- [9] H. Takeya, T. Hirano, K. Kadowaki, *Physica C* 256 (1996) 220.
- [10] R.C. Dynes, V. Aarayanamurti, J.P. Garno, *Phys. Rev. Lett.* 41 (1978) 1509.
- [11] K. Maki, P. Thalmeier, H. Won, *Phys. Rev. B* 65 (2002) 140502(R).
- [12] L.S. Borkowski, P.K. Hirschfeld, *Phys. Rev. B* 49 (1994) 15404.
- [13] T. Watanabe, K. Kitazawa, T. Hasegawa, Z. Hossain, R. Nagarajan, L.C. Gupta, *J. Phys. Soc. Jpn.* 69 (2000) 2708.
- [14] A.A. Abrikosov, L.P. Gor'kov, *Sov. Phys. JETP* 12 (1961) 1243.
- [15] K. Machida, K. Nokura, T. Matsubara, *Phys. Rev. B* 22 (1980) 2307.