Effect of magnetic order on the superfluid response of single-crystal ErNi$_2$B$_2$C: A penetration depth study

Elbert E. M. Chia, W. Cheong, Tuson Park, and M. B. Salamon
Department of Physics, University of Illinois at Urbana-Champaign, 1110 W. Green Street, Urbana, Illinois 61801, USA

Eun-Mi Choi and Sung-Ik Lee
National Creative Research Initiative Center for Superconductivity and Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Republic of Korea

(Received 15 August 2005; revised manuscript received 18 October 2005; published 7 December 2005)

We report measurements of the in-plane magnetic penetration depth $\Delta \lambda(T)$ in single crystals of ErNi$_2$B$_2$C down to $\sim 0.1$ K using a tunnel-diode based, self-inductive technique at 21 MHz. We observe four features: (1) a slight dip in $\Delta \lambda(7)$ at the Néel temperature $T_N=6.0$ K, (2) a peak at $T_{WFM}=2.3$ K, where a weak ferromagnetic component sets in, (3) another maximum at 0.45 K, and (4) a final broad drop down to 0.1 K. Converting to superfluid density $\rho_s$, we see that the antiferromagnetic order at 6 K only slightly depresses superconductivity. We seek to explain some of the above features in the context of antiferromagnetic superconductors, where competition between the antiferromagnetic molecular field and spin fluctuation scattering determines increased or decreased pair breaking. Superfluid density data show only a slight decrease in pair density in the vicinity of the 2.3 K feature, thus supporting other evidence against a pure ferromagnetic state in this temperature range.

DOI: 10.1103/PhysRevB.72.214505

The magnetic members of the rare-earth (RE) nickel borocarbide family, RENi$_2$B$_2$C (RE = Ho, Er, Dy, etc.) have attracted much interest due to the interplay between magnetism and superconductivity. ErNi$_2$B$_2$C, in particular, is a good candidate for the study: superconductivity starts at $T_c \approx 11$ K, before antiferromagnetic (AF) order sets in$^1$ at $T_N=6$ K. In the AF state the Er spins are directed along the $b$ axis, forming a transversely polarized, incommensurate spin-density-wave (SDW) state, with modulation vector $\delta =0.553a^* \left( a^*=2\pi/a \right)$, before squaring up at lower temperatures.$^3$ Below $T_{WFM}=2.3$ K the Er ordering becomes commensurate, a net magnetization appears, superimposed on a modulation with a periodicity of $20a^*$, confirming the microscopic existence of spontaneous weak ferromagnetism (WFM) with superconductivity.$^3,4$

The fact that $T_N < T_c$ enables us to study the influence of magnetism on superconductivity. In particular, in this paper we study, via the penetration depth, the pair-breaking effects of the various magnetic orders on the superfluid response of this material. There have been several previous penetration depth measurements on ErNi$_2$B$_2$C. Jacobs$^5$ measured the microwave surface impedance of single-crystal ErNi$_2$B$_2$C from $T_c$ down to 4 K, but did not see the AF transition at 6 K. They concluded that the AF transition is not accompanied by changes in pair breaking in a zero field. Andreone$^6$ et al. measured the microwave properties of ErNi$_2$B$_2$C thin films—microwave surface resistance$^6$ down to 2 K, and the change in penetration depth$^7$ from 2 to 5 K. They too saw no feature at $T_N$, and attributed its absence to the smearing of the susceptibility $\chi(T)$. Gammel$^8$ et al. performed small-angle neutron scattering measurements$^5$ (SANS) on single-crystal ErNi$_2$B$_2$C down to 4 K, finding a decrease of $\lambda$ below $T_N$ that they could not account for quantitatively. In this paper we present high-precision measurements of the in-plane magnetic penetration depth of single-crystal ErNi$_2$B$_2$C down to 0.1 K. We see features at $T_N$ and $T_{WFM}$, and attribute these to the pair-breaking effects of AF order at $T_N=6$ K and the weak ferromagnetic ordering at $T_{WFM}=2.3$ K. We also observe a peak at 0.45 K, which we attribute to the presence of a spontaneous vortex phase$^9$ (SVP), expected to occur in superconductors where ferromagnetism and superconductivity coexist.$^{10,12}$ The superfluid density graph indicates that these three magnetic orderings coexist with superconductivity, i.e., they do not destroy superconductivity in this material.

Various theories of antiferromagnetic superconductors have been proposed.$^{13-19}$ We shall follow those of Chi and Nagi,$^{19}$ which is an extension of the mean-field model by Nass$^{17,18}$ to the regime where the superconducting gap $\Delta$ is finite, and it includes the effects of spin fluctuations, molecular field, and impurities. In the Chi-Nagi (CN) model, which applies specifically to superconductors with $T_N<T_c$, two temperature regimes are separately considered. First, in the paramagnetic regime ($T_N<T<T_c$), the depression of $T_c$ with respect to the nonmagnetic counterparts, LuNi$_2$B$_2$Co or YNi$_2$B$_2$C, is due to the exchange scattering of the conduction electrons from the spins of the RE Er ions. Assuming that the exchange interaction is weak, this paramagnetic phase of ErNi$_2$B$_2$C can be accounted for by the Abrikosov-Gorkov (AG) pair-breaking theory.$^{20}$ Second, in the AF phase ($T<T_N$), the effect of pair breaking depends on the competition$^{21}$ between the temperature-dependent AF molecular field [with parameter $H_F(T)$] and spin-fluctuation scattering of the conduction electrons, the latter by both magnetic RE ions (parameter $1/\tau^2$) and nonmagnetic impurities (parameter $1/\tau_\perp$). The molecular field opens AF gaps on parts of the Fermi surface (FS), hence destroying the superconducting gap in those areas. The nonmagnetic impuri-
ties do not affect the BCS state for an s-wave superconductor, but weaken the effect of the AF field by destroying the pairing state for charge density waves or spin density waves—the nonmagnetic impurities promote the recovery of superconductivity. Moreover, the effect of the magnetic field and spin fluctuations is governed by a sum rule, and the competition between them determines whether the AF phase gives increased or decreased pair breaking below $T_N$. The total electronic effective magnetic scattering rate $1/\tau^{\text{eff}}_2$ is temperature-dependent and decreases with decreasing temperature (as the magnetic moments become more and more frozen). The assumptions of the CN model are: (1) the effect of inelastic scattering, which is relevant only for $T \ll T_N$, can be ignored; (2) BCS s-wave pairing, and (3) a one-dimensional (1D) electron band that satisfies the nesting condition $\epsilon_k = -\epsilon_{k+Q}$.

The following equations of the CN model were used. The temperature dependence of the superconducting gap

$$\rho_s(T) = \left[ \frac{\lambda^2(0)}{\lambda^2(T)} \right]^2 = \left[ \pi T \sum_{n=0} A(\omega_n) \right],$$

where

$$A(\omega_n) = \frac{\Delta_{n+} - \omega_n^2}{4\epsilon_1} + \frac{\Delta_{n-} - \omega_n^2}{4\epsilon_2} + \frac{1}{4\epsilon_1} + \frac{1}{4\epsilon_2} + \frac{1}{\epsilon_1 + \epsilon_2}$$

$$+ \frac{\Delta_{n+}\Delta_{n-} - \omega_n^2\omega_{n'}}{\epsilon_1\epsilon_2(\epsilon_1 + \epsilon_2)}.$$

In the paramagnetic phase ($T_N < T < T_c$), $\rho_s$ is given by

$$\rho_s(T) = \frac{2\pi T \sum_{n=0} \frac{1}{1 + \epsilon_n}}{\epsilon_0^2},$$

where $\epsilon_0 = |(\omega_n^2 + \Delta_{n+})^{1/2}|, \epsilon_n = |(\omega_n^2 + \Delta_{n-}^{2})^{1/2}|$.

The AF molecular field is given by

$$H_0(T) = H_0(0)F(T),$$

where

$$H_0(0) = n_R g_J \sqrt{J + 1}.$$
We denote $/H9004$ of filamentary superconductivity. $gJ$ 

bulk superconductivity occurs at 10.1 K. A comparison of the three measurements show that $\Delta/\lambda/\Gamma/hi$ is expected to decrease below $\Delta/\lambda/\Gamma/hi$ = 0.227. For ErNi$_2$B$_2$C, using the values $T_c$ = 11.3 K and zero resistivity at 9.6 K. The superfluid data lead to some important observations. First, the data in the paramagnetic phase $\Delta/\lambda/\Gamma/hi$ from $T_c$ = 11.3 K to 13 K. Inset: $\Delta/\lambda/\Gamma/hi$ below 6.5 K. The arrows show features at 6 K AF phase, 2.3 K (WFM), and 0.45 K (SVP). 

FIG. 1. (C) Temperature dependence of the penetration depth $\Delta/\lambda/\Gamma/hi$ from 0.1 K to 13 K. Inset: $\Delta/\lambda/\Gamma/hi$ below 6.5 K. The arrows show features at 6 K AF phase, 2.3 K (WFM), and 0.45 K (SVP). 

The parameters of this model are determined as follows. First, the data in the paramagnetic phase $\Delta/\lambda/\Gamma/hi$ from $T_c$ = 11.3 K to 13 K. Inset: $\Delta/\lambda/\Gamma/hi$ below 6.5 K. The arrows show features at 6 K AF phase, 2.3 K (WFM), and 0.45 K (SVP). 

plane penetration depth $\Delta/\lambda/\Gamma/hi$. The onset of superconductivity, $T_c$, is 11.3 K, showing that this is a high-quality single crystal. We also see the following features: (1) a slight dip in $\Delta/\lambda/\Gamma/hi$ at $T_N$ = 6.0 K, (2) a peak at $T_{WFM}$ = 2.3 K, (3) another maximum at 0.45 K, and (4) a final broad drop down to 0.1 K. We attribute the last two features to the presence of the SVP. For dirty AF superconductors, the penetration depth is expected to decrease below $T_N$ by both $\Delta/\lambda/\Gamma/hi$ and mean free path (l) as $\Delta/\lambda/\Gamma/hi$ = $\lambda/\Gamma/hi$ (1 + 4$\pi/hi$/l), where $\lambda/\Gamma/hi$ = $\lambda/\Gamma/hi$ (1 + $\xi/hi$/l). Neither effect, however, explains our data. First, using the mean-field expression for $\chi$, in order to reproduce the experimental dip, the peak in $\chi$ at $T_c$, has to be at least an order of magnitude larger than that suggested by magnetization measurements. Second, from our resistivity data we obtained $H_{SVP}(T)$, and hence we calculated $\xi_0(T)$, $\xi_0(T)$, and lastly, $\lambda/\Gamma/hi$. Our values of $\lambda/\Gamma/hi$ also are unable to explain the magnitude of the drop of $\lambda$ below $T_N$ — a conclusion also shared by Gammel et al. from their SANS data. Since the CN model does not take into account the effect of the SVP on the superfluid density, we neglect it when we convert to superfluid density $\rho_s$. First, we assume $\Delta/\lambda/\Gamma/hi$ follows a power-law temperature dependence at low temperatures from the combined freezing of the gap minima observed in magnetic borocarbides and the increased pair breaking as Er spins disorder. Consequently, we set $\lambda_{low}(T) = \lambda(0)(1 + bT^2)$ with $b$ = 0.036 K$^{-2}$ from Ref. 9. Next we offset $\lambda_{low}$ until it matches the data at 1.3 K, the local minimum in $\Delta/\lambda/\Gamma/hi$ in the vicinity of 1.5 K. Finally we convert $\Delta/\lambda/\Gamma/hi$ to $\rho_s$ in Fig. 2. 

The superfluid data lead to some important observations. First, the data in the paramagnetic phase ($T > T_N$) fit the theoretical curve based on an isotropic superconducting gap (solid squares), and not that based on nodes. The solid line shows a superfluid calculation based on a $d_{x^2-y^2}$ order param-

FIG. 2. (Color online) (C) Experimental superfluid density $\rho_s(T) = [\lambda(0)/\lambda(T)]$ calculated from the $\Delta/\lambda/\Gamma/hi$ data in Fig. 1, from 0.1 K to $T_c$. Solid squares, calculated $\rho_s(T)$ assuming paramagnetic phase from $T = 0$ to $T_c$. Note that $1/\tau/\Gamma/hi = 24, 26, 28$ gives virtually the same theoretical curve. Solid line: Theoretical curve for the $d_{x^2-y^2}$ order parameter. The arrow denotes Néel temperature at 6 K.
electron. Second, the superconductivity is only slightly depressed in the AF phase below $T_N$. The best fit to the data above $T_N$ (solid squares) is obtained when $\lambda(0) = 740 \, \text{Å}$ — here we assume paramagnetism from $T = 0$ to $T_c$, neglecting AF order, with parameter $1/\tau\Delta_0 = 24$. The paramagnetic curve is almost unchanged if one uses $1/\tau\Delta_0 = 26$ or 28. We see that the paramagnetic curve fits the data above $T_N$ and overestimates the data in the AF phase. We will show below that, as one crosses $T_N$ from above, the AF phase leads to increased or decreased superfluid density depending on the combined effects of the following three factors: (1) the AF molecular field, which decreases the magnitude of the superconducting gap and hence decreases the superfluid density, (2) freezing out of spin fluctuations, leading to decreased pair breaking and hence increased superfluid density, and (3) scattering from nonmagnetic impurities, which reduces the suppression of the gap by the molecular field. The parameters of ErNi$_2$B$_2$C are such that these three effects result in a slight decrease in superfluid density below $T_N$.

Figure 3 shows the calculated superconducting gap amplitude $\Delta(T)$ in the presence of $H_{c2}(T)$, for various values of $1/\tau\Delta_0$, as well as the paramagnetic curve. The normalized paramagnetic gap (open circles) agrees excellently with the BCS gap (solid line). Tunneling measurements$^{31}$ also show that $\Delta(T)$ follows the BCS curve above $T_N$. Next, as shown in Fig. 3, in the AF phase, as $1/\tau\Delta_0$ increases, superconductivity is gradually recovered, as evidenced by the increase of $\Delta(T)$ (solid squares). Figure 4 shows $\rho_s(T)$ for various values of $1/\tau\Delta_0$. Notice that when $1/\tau\Delta_0 = 26$, (1) $\rho_s$ decreases only slightly at $T_N$, and (2) $\Delta$ is only slightly depressed below the BCS value, in agreement with tunneling data.$^{31,32}$ This value of $1/\tau\Delta_0$ corresponds to a mean free path (mfp) of 45 Å. Although this curve still overestimates the experimental data below $T_N$, it at least fits the data better than the paramagnetic curve.

To see if this value of $1/\tau\Delta_0$ is reasonable, we take $\Delta(0) = 1.837T_c$ for ErNi$_2$B$_2$C,$^{26}$ for which the BCS coherence length $\xi_{0\text{BCS}} = h v_F / \pi \Delta(0) = 470 \, \text{Å}$, where $v_F = 3.6 \times 10^5 \, \text{m/s}$ is taken from band-structure calculations$^{33}$ for LuNi$_2$B$_2$C and YNi$_2$B$_2$C. Using the relation

$$ H_{c2}(0) = 0.693T_c \left( \frac{dH_{c2}}{dT} \right) \bigg|_{T_c} , $$

and $dH_{c2}/dT|_{T_c} = 0 = 2.67 \, \text{kOe/K}$, we obtain the coherence length

$$ \xi_0 H_{c2} = \sqrt{2 \xi_0 H_{c2} \phi_0 / 2mH_{c2}(0)} = 130 \, \text{Å}. $$

Finally, using the relation

$$ \xi_0 H_{c2} = 0.85(\xi_0)_{BCS}^{3/2}, $$

we obtain the mfp $\ell = 42 \, \text{Å}$. On the other hand, from the resistivity value just above $T_c$, $\rho_s(T_c) = 5.8 \, \mu\Omega \, \text{cm}$, we get $\ell = 56 \, \text{Å}$. These two values agree well with the value of 45 Å calculated from $1/\tau\Delta_0 = 26$ obtained earlier, implying that this particular value of the nonmagnetic scattering rate needed to explain our $\rho_s$ data are consistent with other measurements. Note that this value of mfp calculated from the $H_{c2}$ data does not depend on the exact value of $T_c$. Also, the prefactors 0.693 and 0.85 in the above relations are for materials in the dirty limit. Here $\ell < \xi_0$, so our ErNi$_2$B$_2$C sample may be considered as “quasidirty.” It is puzzling that our sample has a high $T_c$ and be considered quasidirty, yet this is consistent with the results of other papers. Also, in this sample the nonmagnetic scattering rate (1/$\tau$) is at least two orders of magnitude larger than the effective magnetic scattering rate (1/$\tau^{MB}$), thus the mfp value is largely determined by 1/$\tau$. Our mfp value, however, is smaller than the 90 Å obtained from resistivity measurements just above $T_c$ in Ref. 35. The CN model is thus able to explain our superfluid density data, both qualitatively and quantitatively. Our data, in agreement with others, also show that AF order coexists with superconductivity below $T_N$.

It is also interesting to note that according to Ref. 21, a near-exact cancellation of spin-fluctuation and molecular-field effects occur at a critical value of $N(0) F^2 = 1 \times 10^{-2}$.
where $F^f = I_f |g_f| - 1$ is the conduction-electron local spin exchange. For the case of ErNi$_2$B$_2$C, we obtain $N(0)F^f = 1.7 \times 10^{-3}$, which explains the small change in pair breaking at $T_N$.

We turn next to an alternative explanation for the change of $\rho_s$ at $T_N$. Ramakrishnan and Varma$^{19}$ predicted that for materials with a nested FS, since the peak in susceptibility and the joint density of states (defined as the difference between the susceptibility in the superconducting state and the normal state) occur at the same $Q$ value, one should expect an increase in pair breaking at $T_N$. Conversely, a nonnested FS will give rise to decreased pair breaking at $T_N$. Two-dimensional angular correlation of electron-positron annihilation radiation measurements shows that only one out of the three FS sheets in LuNi$_2$B$_2$C possesses nesting properties, thereby accounting for the propensity for magnetic ordering found in the other magnetic members of the RE nickel borocarbides.$^{27}$ Also, Dugdale et al.$^{25}$ estimated that the fraction of the FS that would be able to participate in nesting is only 4.4%. Contrast this with the CN model, which assumes perfect 1D nesting. Hence the increased pair breaking due to partial nesting on one FS sheet is partially compensated by decreased pair breaking by the other two sheets, resulting in only a slight increase in pair breaking at $T_N$.

As temperature further reduces below $T_N$, the theoretical curve in Fig. 2 overestimates the experimental curve below 3 K ($\sim 0.3T_c$). This is due to an additional pair-breaking effect of the ferromagnetic moments in the WFM phase, which shows up as a small peak near 2.3 K (see Fig. 1). The small dip in superfluid density shows that this WFM slightly depresses, but does not completely destroy, superconductivity, demonstrating the coexistence of WFM and superconductivity. We model this WFM by including a temperature-dependent magnetic impurity scattering rate $1/r_2^{WFM} = 1/20(g_J - 1)/(J + 1)(1 - T/T_{WFM})\nu'$ with the same value of $g_J$ and $J$ as before, and adding this to the previous effective magnetic scattering rate, i.e., $1/\tau^{total} = 1/\tau^{eff} + 1/r_2^{WFM}$ when $T < T_{WFM}$. The prefactor 1/20 arises from the fact that one out of every 20 spins contributes to the WFM,$^{36}$ giving rise to a weak magnetization. The temperature dependence $(1 - T/T_{WFM})\nu'$ is analogous to the molecular field formulation. We obtain $\nu' \approx 2$ from Jensen’s calculation$^{37}$ or Choi and Canfield’s data.$^{3,4}$ Note that $\rho_s$ already starts to flatten out at 3 K, consistent with neutron-scattering data, which shows that this weak ferromagnetic component already shows up at 3 K.$^3$ Hence we choose $T_{WFM} = 3$ K in this WFM calculation. Figure 5 shows $\rho_s$ when one accounts for WFM. The calculated $\rho_s$ does flatten out below 3 K, but does not increase below 2.3 K as the data did. There is as yet no direct measurement of superconducting gap amplitude at this temperature range, though our model predicts a drop in $\Delta$ there.

One may need to include the effect of inelastic spin-fluctuation scattering in this low-temperature region, which was ignored by the CN model.

In conclusion, we present in-plane penetration depth data of single-crystal ErNi$_2$B$_2$C down to 0.1 K. The small increase in pair breaking at $T_N$ can be attributed to the interplay between the effects of the AF molecular field and spin-fluctuation scattering. It could also be due to the combined effects of nonperfect nesting on one piece of the FS and nonnesting on other pieces of the FS. The increased pair breaking at $T_{WFM}$ is modeled by a magnetic impurity scattering parameter, and both magnetic orders coexist with superconductivity. Finally, we wish to point out that we also have data where the AC field is applied parallel to the basal plane, and they show the same features as those presented in this paper.

E.E.M.C wishes to thank Y. C. Chang for teaching the computational aspects of the Chi-Nagi model, as well as C. Varma and J. Thompson for encouraging discussions on the interpretation of data. This material is based upon work supported by the U.S. Department of Energy, Division of Materials Sciences under Grant No. DEFG02-91ER45439, through the Frederick Seitz Materials Research Laboratory at the University of Illinois at Urbana-Champaign. Research for this publication was carried out in the Center for Microanalysis of Materials, University of Illinois at Urbana-Champaign. This work in Korea was supported by the Ministry of Science and Technology of Korea through the Creative Research Initiative Program.