Tunneling spectroscopy in the magnetic superconductor

\textbf{TmNi$_2$B$_2$C}

H. Suderow$^1$, P. Martinez-Samper$^2$, S. Vieira$^2$
N. Luchier$^3$, J.P. Brison$^3$
P. Canfield$^4$

$^1$Instituto de Ciencia de Materiales de Madrid (ICMM)
Consejo Superior de Investigaciones Científicas
Campus de Cantoblanco 28049 Madrid-Spain

$^2$Laboratorio de Bajas Temperaturas, Departamento de Física de la Materia Condensada
Instituto de Ciencia de Materiales Nicolás Cabrera, Facultad de Ciencias, C-III
Universidad Autónoma de Madrid, 28049 Madrid-Spain

$^3$Centre des Recherches sur les Tres Basses Temperatures
CNRS, BP 166, 38042 Grenoble Cedex 9, France

$^4$Ames Laboratory and Department of Physics and Astronomy
Iowa State University, Ames, Iowa 50011

(January 7, 2022)

Abstract

We present new measurements about the tunneling conductance in the borocarbide superconductor TmNi$_2$B$_2$C. The results show a very good agreement with weak coupling BCS theory, without any lifetime broadening parameter, over the whole sample surface. We detect no particular change of the tunneling spectroscopy below 1.5K, when both the antiferromagnetic (AF) phase and the superconducting order coexist.

PACS numbers: 74.70.Dd, 87.64.Dz, 73.40.Gk
Electron tunneling spectroscopy is a very powerful tool to investigate the fundamental properties of conducting materials which has been widely applied to the study of superconductivity [1]. The results have verified several of the most important predictions of the BCS theory. More recently, Scanning Tunneling Microscopy and Spectroscopy (STM/STS) has given the possibility to measure local variations of the superconducting density of states. For example, the vortex structure of several superconductors, such as NbSe$_2$, some of the High $T_c$ oxides or the borocarbide superconductors, could be studied in detail [2–6]. Nevertheless, it remains very difficult to get accurate information from the most simple measurement one can do with STM/STS, the zero field tunneling conductance curves. Indeed, the fitting of the measured curves by any kind of model requires the introduction of an ad hoc broadening parameter [7] which is supposed to account phenomenologically for uncharacterized pair-breaking effects, and whose magnitude is often comparable to that of the gap itself. So only the zero field curves obtained in the most simple superconductors (as e.g. Pb, Nb or Al [8–11]) have been successfully fitted by BCS or Eliashberg theory. The broadening parameter appears also when data are be obtained using other related techniques such as break-junctions, point contact spectroscopy, or planar junctions in the more complex materials (as for example heavy fermions or borocarbidies [12–15]).

Here we present new data in TmNi$_2$B$_2$C. This compound belongs to the family of the borocarbide superconductors (RENi$_2$B$_2$C where RE is a rare earth; RE = Lu, Y, Tm, Er, Ho…) [16–19]. These systems present rich phase diagrams showing coexistence or competition between superconductivity and a magnetic order of the RE spins (when they have a magnetic moment) [19]. In the Tm compound the superconducting and Neel critical temperatures are well separated ($T_c = 10.5 K$, and $T_N = 1.5 K$) [20–18,19] and in the antiferromagnetic phase, the spins of the Tm ions order in a transversely polarized spin density wave with an incommensurate modulation of the magnetic moments [18,21]. To our knowledge, no previously published tunneling experiment is available in this compound. The tunneling experiments reported up to now in nonmagnetic compounds and the point contact spectroscopy data in magnetic compounds [3,4,14,15] have never shown a simple BCS density
of states $N_{BCS}(E)$ and the conductance curves were all severely broadened. Therefore, the possible presence of low energy excitations as well as the form of the superconducting gap in the coexistence region were masked by extrinsic broadening. Here we present tunneling spectra that do not show any additional broadening, even in the antiferromagnetic phase. These data present a significant advance in tunneling spectroscopy and they should allow further progress on the detailed study of the superconducting ground state of borocarbides.

We have used a STM with an x-y table that permits coarse movement in a 2x2 mm$^2$ region in a $^3$He insert, and where the sample holder is cooled down to 0.8K. The tip is prepared from a gold wire cut with a clean blade. We measured several samples (of dimensions about 1x1x1 mm$^3$), prepared by breaking the same single crystal platlet grown by a flux technic described in [16,22]. The best results were obtained with the samples which were mounted on the STM and cooled down immediately after breaking, so that the surface remained no more than several minutes at ambient pressure (about 10 in most cases). They show good quality, highly reproducible spectra over the whole surface. The topographic images (Fig.1) are always of good quality, and independent of the tunneling resistance. They show an irregular structure with cluster-like forms, with a diameter of the order of 20-30 nm on planes of about 30-60 degrees. In spite of intensive search with the x-y table, we were unable to find sufficiently flat zones to obtain atomic resolution. These results are similar to the observations made in other borocarbides [3].

It is well known that proper RF filtering is essential to do tunneling or point contact spectroscopy in superconductors [1]. This is specially important in an STM set-up as the tunneling current is of the order of nA or lower (we use tunneling resistances between 1MΩ and 10MΩ): this is several orders of magnitude smaller than in a normal planar junction experiment. In TmNi$_2$B$_2$C, we have found an important difference between measurements obtained at the same temperature and with the same surface preparation but in two different cryostats. The same low noise STM electronics was used on both setup but one had filters and the other none. In the unfiltered setup we find a fictitious finite conductance at zero bias and we need to introduce a broadening parameter $\Gamma \approx 0.3\Delta$ [7], comparable to the
values obtained in previous works [6,14,15]. On the contrary, the curve measured with
appropriate shielding can be accurately fitted to the conventional BCS expression, using
the superconducting gap $\Delta$ and the temperature $T$ as the only fitting parameters. We
use room temperature RF feedthrough filters and thermocoaxial cables for all the electrical
connections of the STM. The results presented in this paper have been verified on three
different samples measured in the filtered set-up, two of them with the surface parallel to
the a-b plane, and one of them with the surface parallel to the c axis of the tetragonal crystal
structure. No significant differences were found. Note nevertheless that the surface (Fig.1)
consists of inclined planes that do not correspond to a clear crystallographic direction.

For the curve in Fig.2a, the best fit to the experiment (line on Fig.2a ) is obtained with
$\Delta = 1.4mV$ and $T = 2K$. The upper limit to any broadening parameter is ($\Gamma < 0.001\Delta$).
Note that the temperature obtained from the fit is larger, by 0.2 K, than the actual sample
temperature. Indeed, the quasiparticle anomaly at $\Delta$ is not as peaked as expected from BCS
theory and adding a finite $\Gamma$ does not give an appropriate fit, as it immediately results in
increasing the zero bias conductance. It could be that we still need a better filtering, but our
measurements in Pb and Al samples down to lower temperatures (400 mK [23]) have shown
that our (relative) spectral resolution is sufficiently good to resolve such details. A more
likely explanation is that the superconducting gap has a small anisotropy. Indeed, a marked
anisotropy or the presence of different gaps results in the appearance of different maxima
in the density of states at $T = 0K$ and therefore in a broadened quasiparticle peak at
finite temperature, without adding additional conductance at zero bias [1,11]. For instance,
the measured tunneling conductance in the superconductor NbSe$_2$ shows a quasiparticle
anomaly that is much less peaked than expected from BCS theory. It also has a marked
structure possibly corresponding to the very large anisotropy of the superconducting gap
found in that compound [2,4,11]. In our case, the decrease in peak intensity is much smaller
so that a slight dispersion in the value of the measured gap of the order of several percent of
$\Delta$ could already account for the observed broadening. However, a quantitative calculation
is by now not realistic, because it requires knowledge of the different values of the gap $\Delta(k)$.
for the relevant set of $k$ vectors.

Let us note that there are several other experimental indications of gap anisotropies, or equivalently of multiband (each having a proper gap amplitude) effects in borocarbide superconductors. The upward curvatures of the upper critical field in LuNi$_2$B$_2$C and YNi$_2$B$_2$C also present in TmNi$_2$B$_2$C (for H in the basal plane) have been explained first by non-local effects \[24\], backed by the structural changes in the vortex lattice \[25–29\]. But an alternate (or complementary) interpretation to the positive curvatures involves a two band model with different coupling constants and so different gap amplitudes \[30\]. The $T^3$ dependence of the zero field electronic part of the specific heat at low temperatures in YNi$_2$B$_2$C or LuNi$_2$B$_2$C, together with its strong field dependence could also point toward reduced gap regions on the Fermi surface \[31\], although our measurements indicate that the reduction of the gap in these regions and in the case of Tm is not larger than several percent.

When cooling down towards the antiferromagnetic phase ($T_N = 1.5K$), we do not observe any change in the spectroscopy, other than the temperature reduction (in Fig.2b we use $T = 1K$ and $\Delta = 1.45mV$). What is more, the whole temperature dependence of the superconducting gap shown in Fig.3 is in good agreement with conventional BCS theory, within experimental error. The value of the superconducting gap gives $\Delta/k_BT_c = 1.55$ (taking $\Delta = 1.45mV$ and $T_c = 10.5K$). Due to our error bars on the determination of $T_c$ ($\pm 15\%$) this ratio is in agreement with the BCS weak coupling value of 1.73. This value raises the same problems as those discussed in Ref. \[14\]. In this work, tunneling spectroscopy has been performed on the Y and Lu compounds with break-junctions. The curves show significant broadening, and yield a large range of ratios of $\Delta/k_BT_c$ (depending on the surface or junction quality). But the weak-coupling regime was put forward because these ratios are systematically lower or equal than the BCS value. Our results on the Tm compound also strongly support weak coupling superconductivity, thanks both to the absence of broadening and to the reproducibility of the results over very large surface areas.

On the other hand, analysis of the specific heat jump at $T_c$, and of the low temperature regime of the specific heat, implies a rather small ratio of $T_c/\omega$ where $\omega$ is some average
frequency of the phonon spectrum [1], meaning strong to intermediate coupling. Typically, for the Y and Lu compound, values for \( \omega \) between 150K and 200K are needed to explain the specific heat results [2]. They imply values of \( \Delta/k_B T_c \approx 2 \) that are clearly incompatible to the value we have found in Tm, which is not expected to have a very different phonon spectrum. In addition, \( \omega \) between 150K and 200K implies the appearance of anomalies characteristic for strong coupling superconductivity clearly within our experimental resolution on the tunneling density of states between 13 and 17mV [1], which neither we nor the authors of Ref. [14] have observed. By contrast \( \omega \approx 490K \), proposed for the two band model of the upper critical field in Ref. [30], fits much better our results. But this leaves the problem of the discrepancy with the thermodynamic data unsolved. The solution to these contradictions may lie in the complex phonon spectrum (see references and a model spectrum in [32]), and again on the multiband structure of the Fermi surface in borocarbide superconductors. They may yield different results for the average frequency involved in the specific heat, the gap, or the upper critical field [33]. This looks somewhat paradoxical in these compounds which, except for their magnetic properties, show rather isotropic behavior in the normal phase.

As regard the lack of signature of the antiferromagnetic order in the tunneling spectra, it might be explained more intuitively. For example, one could invoke that the period of the incommensurate magnetic modulation is of the order of 2.5nm, which is small compared to the superconducting coherence length \( \xi_0 = 12 \) nm [19,34]. This means that the variation of the local magnetic moment is averaged out on the superconducting coherence length or, in q-space, that the superconducting order does not make any change on the susceptibility \( \chi(q) \) at the antiferromagnetic wave-vector \( Q \gg 1/\xi_0 \). In order to test more exotic possibilities, like a local depression of the superconducting gap or the presence of new magnetic excitations of longer period, not detected by other techniques, we made local spectroscopy measurements. An I-V curve is done at a given set of pixels in a topography image. From the difference \( dI/dV(V > \Delta) - dI/dV(V < \Delta) \) we can test the local appearence of low energy excitations. But we obtained essentially flat images giving \( dI/dV(V > \Delta) - dI/dV(V < \Delta) = 1 \) within
5%, as shown in Fig.4 for a 240x240\textit{nm} scan.

We do not know of any previous measurements of the tunneling conductance in an antiferromagnetic superconductor. Indeed, it is well known, mainly through macroscopic measurements as resistivity or specific heat, that an antiferromagnetic order coexists with superconductivity in this compound \[19,20\]. But their mutual influence is a hotly debated issue (see \[21\] and \[35\]) and the predictions remained ambiguous. For instance, the authors of Ref. \[36\] point towards the possible existence of a line of nodes in the superconducting gap in the antiferromagnetic phase of TmNi$_2$B$_2$C. We have now ruled out this possibility.

In conclusion, we have studied the superconducting gap of TmNi$_2$B$_2$C as a function of temperature. The results can be fitted to conventional, BCS, weak-coupling theory in the whole temperature range. We did not detect any change of the superconducting phase when the antiferromagnetic order appears.
REFERENCES

[1] E.L. Wolf, ”Principles of Electron Tunneling Spectroscopy”, Oxford University Press (1989).

[2] H.F. Hess, R.B. Robinson, J.V. Waszcak, Phys. Rev. Lett. 64, p. 2711 (1990).

[3] I. Maggio-Aprili, Ch. Renner, A. Erb, E. Walker, O. Fisher, Phys. Rev. Lett. 64, p. 2711 (1990).

[4] S.H. Pan, E.W. Hudson, K.M. Lang, H. Eisaki, S. Uchida, J.C. Davis, Nature 403, p. 746 (2000).

[5] H. Sakata, M. Oosawa, K. Matsuba, N. Nishida, H. Takeya, K. Hirata, Phys. Rev. Lett. 84, p. 1583 (2000).

[6] Y. De Wilde, M. Iavarone, U. Welp, V. Metlushko, A.E. Koshelev, I. Aranson, G.W. Crabtree, P.C. Canfield, Phys. Rev. Lett. 78, p. 4273 (1997).

[7] R.C. Dynes, V. Narayanamurti, J.P. Garno, Phys. Rev. Lett., 41, p. 1509 (1978).

[8] S.H. Pan, E.W. Hudson, J.C. Davis, Applied Phys. Lett., 73, p. 2992 (1998).

[9] H. Suderow, A. Izquierdo, S. Vieira, Physica C, 332, p. 327 (2000).

[10] A. Yazdani, B.A. Jones, C.P. Lutz, M.F. Crommie, D.M. Eigler, Science, 275, 1767 (1997).

[11] P. Martinez Samper, E. Bascones, H. Suderow, J.G. Rodrigo, F. Guinea and S. Vieira, to be published.

[12] M. Jourdan, M. Huth, H. Adrian, Nature 398, 47 (1999).

[13] See e.g. H.v. Loehneysen, Physica B, 218, p.148 (1996) and References therein.

[14] T. Ekino, H. Fujii, M. Kosugi, Y. Zenitani, J. Akimitsu, Phys. Rev. B, 53, p. 5640 (1996).
[15] I.K. Yanson, N.L. Bobrov, C.V. Tomy, D. McK. Paul, Physica C, 334, p.33 (2000).

[16] P.C. Canfield, P.L. Gammel, D.J. Bishop, Phys. Today (1998), 51, pp. 40-46.

[17] R.J. Cava et al. Nature, 367, 146 (1994); R. Nagarajan et al. Phys. Rev. Lett. 72, 274 (1994).

[18] J.W. Lynn, S. Skanthakumar, Q. Huang, S.K. Sinha, Z. Hossain, L.C. Gupta, R. Nagarajan, C. Godart, Phys. Rev. B, 55, p. 6584 (1997).

[19] B.K. Cho, M. Xu, P.C. Canfield, L.L. Miller, D.C. Johnston, Phys. Rev. B, 52, p. 3676 (1995)

[20] R. Movshovich, M.F. Hundley, J.D. Thompson, P.C. Canfield, B.K. Cho and A.V. Chubukov, Physica C, 227, 381-6 (1994)

[21] K. Norgaard, M.R. Eskildsen, N.H. Andersen, J. Jensen, P. Hedegard, S.N. Klausen, P.C. Canfield, Phys. Rev. Lett. 84, p. 4982 (2000).

[22] B. K. Cho, P. C. Canfield, L. L. Miller, D. C. Johnston, W. P. Beyermann and A. Yatskar, Phys. Rev. B 52, p. 3684 (1995)

[23] H. Suderow, S. Vieira, to be published.

[24] V. Metlushko, U. Welp, A. Koshelev, I. Aronson, G.W.Crabtree and P.C. Canfield, Phys. rev. Lett. 79, 1738 (1997).

[25] D. McK. Paul, C.V. Tomy, C.M. Aegerter, R. Cubitt, S.H. Lloyd, E.M. Forgan, S.L. Lee and M. Tethiraj, Phys. Rev. Lett. 79, 1738 (1997).

[26] M.R. Eskildsen, P.L. Gammel, N.P. Barber, U. Yaron, A.P. Ramirez, D.A. Huse, D.J. Bishop, C. Bolle, C.M. Lieber, S. Oxx, S. Sridhar, N.H. Andersen, K. Mortensen, P.C. Canfield Phys. Rev. Lett. (1997), 78, 1968-1971.

[27] M.R. Eskildsen, P.L. Gammel, B.P. Barber, A.P. Ramirez, D.J. Bishop, N.H. Andersen,
K. Mortensen, C.A. Bolle, C.M. Lieber, P.C. Canfield, Phys. Rev. Lett. (1997), 79, 487-490.

[28] K.O. Cheon, I.R. Fisher, V. Kogan, P.C. Canfield, P. Miranovic, P.L. Gammel, Phys. Rev. B: Condens. Matter Mater. Phys. (1998), 58, 6463-6467.

[29] P.L. Gammel, D.J. Bishop, M.R. Eskildsen, K. Mortensen, N.H. Andersen, I.R. Fisher, K.O. Cheon, P.C. Canfield, V.G. Kogan, Phys. Rev. Lett. (1999), 82, 4082-4085

[30] S.V. Shulga, S.-L. Drechsler, G. Fuchs, K.H. Müller, K. Winzer, M. Heinecke and K. Krug, Phys. Rev. Lett. 80, 1730 (1998).

[31] M. Nohara, M. Isshiki, F. Sakai and H. Takagi, J. Phys. Soc. Jpn. 68, 1078 (1999).

[32] H. Michor, T. Holubar, C. Dusek and G. Hilscher, Phys. Rev. B 52, 16165 (1995).

[33] Carbotte J.P., Rev. Mod. Phys. 62, 1027 (1990).

[34] M.R. Eskildsen, K. Harada, P.L. Gammel, A.B. Abrahamsen, N.H. Andersen, G. Ernst, A.P. Ramirez, D.J. Bishop, K. Mortensen, D.G. Naugle, K.D.D. Rathnayaka, P.C. Canfield, Nature 393, p. 242 (1998).

[35] L.N. Bulaevski, A.I. Buzdin, M.L. Kulic, S.V. Panjukov, Adv. in Phys. (1985), 34, p. 175.

[36] M.L. Kulic, A.I. Buzdin, L.N. Bulaevskii, Phys. Lett. A, p. 285 235 (1997)
FIG. 1. The figure shows typical topographic images on TmNi$_2$B$_2$C. Sometimes terraces of some tens nm height can be observed. The typical image is however an inclined surface with "bumps" of 20-30 nm in height. The bumps are visualized in the figure between both images.
FIG. 2. The superconducting gap at 1.8 and 0.8 K as measured with tunneling spectroscopy (tunneling resistance about 10MΩ). In a. we show the important improvement of the quality of the I-V curves between the measurements done in an unfiltered (full points) and in a filtered setup (open circles). In b. we show the result in the Antiferromagnetic phase (T_{N}=1.5K). No changes are observed. The lines are fits to the BCS weak coupling theory using the parameters given in the figures.
FIG. 3. The temperature dependence of the superconducting gap as followed by tunneling spectroscopy. Fitting the experimental curves gives the gap shown in the inset. The scattering in the results increases above 6K, possibly due to the experimental uncertainty.
FIG. 4. Here we show two images done by doing an I-V sweep at a set of 16x16 points in a surface of 240x240 nm. Note that no changes are observed below and above the Neel temperature. Some of the dI/dV(V) curves are shown in the left side. Note that the experimental resolution is less as compared to the curves shown in Figs.2 and 4. In order to obtain the image in a reasonable time (45 minutes).