Measurements of the superconducting fluctuations in optimally doped BaFe$_{2-x}$Ni$_x$As$_2$ under high magnetic fields: probing the 3D-anisotropic Ginzburg–Landau approach

R I Rey$^1$, A Ramos-Álvarez$^1$, C Carballeira$^1$, J Mosqueira$^1$, F Vidal$^1$, S Salem-Sugui Jr.$^2$, A D Alvarenga$^3$, Rui Zhang$^4$ and Huiqian Luo$^4$

$^1$LBTS, Facultade de Física, Universidade de Santiago de Compostela, E-15782 Santiago de Compostela, Spain
$^2$Instituto de Física, Universidade Federal do Rio de Janeiro, 21941-972 Rio de Janeiro, RJ, Brazil
$^3$Instituto Nacional de Metrologia Qualidade e Tecnologia, 25250-020 Duque de Caxias, RJ, Brazil
$^4$Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, People’s Republic of China

E-mail: j.mosqueira@usc.es

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Abstract
The superconducting fluctuations well inside the normal state of Fe-based superconductors were experimentally studied through the in-plane paraconductivity in several high-quality, optimally doped BaFe$_{2-x}$Ni$_x$As$_2$ crystals. These measurements were performed in magnetic fields with amplitudes up to 14 T, and different orientations relative to the c-axis of the crystals ($\theta = 0^\circ$, $53^\circ$, and $90^\circ$). The results allowed a stringent check of the applicability of a recently proposed Ginzburg–Landau approach for the fluctuating electrical conductivity of three-dimensional (3D) anisotropic materials in the presence of finite applied magnetic fields.

Keywords: Fe-based superconductors, transport properties, superconducting fluctuations

1. Introduction
The high critical temperatures ($T_c$) of Fe-based superconductors (FeSC) and the unconventional mechanism for their superconductivity (with a pairing probably mediated by spin fluctuations and involving several bands) have generated enormous interest for these materials in the last few years [1]. A central aspect of their phenomenology is the effect of superconducting fluctuations around $T_c$ [2]. Mainly due to the short coherence length and high-$T_c$ values of these materials [1], these effects are enhanced with respect to conventional low-$T_c$ superconductors. In fact, the Ginzburg number, which characterizes the width of the critical fluctuation region around $T_c$, is in FeSC at half the level found in conventional low-$T_c$ superconductors and high-$T_c$ cuprates [3].

In addition to their intrinsic interest, superconducting fluctuation effects are a very useful tool for characterizing the nature of a superconducting transition and obtaining material parameters [2]; different works have already addressed their study in FeSC through observables like magnetization, specific heat, or electric conductivity [3–23]. However, some fundamental aspects of the phenomenology of the fluctuation effects in these materials are still debated. One of them is their dimensionality. In these materials, the transverse coherence length amplitude $\xi(0)$ is close to the Fe layer’s periodicity length, $s$. Thus, depending on the particular compound...
studied, some works report a two-dimensional (2D) behavior [3, 10, 20] similar to the one found in highly anisotropic high-
$T_c$ cuprates [24], while others find three-dimensional (3D) characteristics [4–7, 11, 12, 14–16, 19, 21–23] or even a
3D–2D transition [8, 13, 18] when increasing the temperature above $T_c$ (as in optimally doped YBa$_2$Cu$_3$O$_{6.9}$) [24]$^5$. Besides, it was recently reported that the fluctuating electrical conductivity above $T_c$ of clean LiFeAs crystals seems to follow a well defined 2D behavior in both its amplitude and reduced-temperature dependence, despite the fact that for this compound, $\xi(0) \approx 1.6$ nm is much larger than the periodicity
length of the Fe-layers ($s = 0.636$ nm) [20]. This surprising result led the authors of [20] to propose that in these
multiband superconductors, the fluctuating pairs above $T_c$ may be driven by a single 2D band. Other interesting issues that
deserve attention are the possible presence of phase fluctuations (whose effect was possibly observed near $T_c$ at low field
amplitudes in the SmFeAsO$_{0.6}$F$_{0.2}$ [17], and also in members of the less anisotropic 122 family as Ba$_{1-x}$K$_x$Fe$_2$As$_2$ [5] and Ba(Fe$_{1-x}$Rh$_x$)$_2$As$_2$ [25]), or the behavior of fluctuation effects in the short wavelength regime appearing at high reduced
magnetic fields or temperatures.

To contribute to the understanding of the above mentioned issues, we present detailed measurements of the fluctuation-induced in-plane electric conductivity ($\Delta \rho_{ab}$) in several high-quality BaFe$_{2-x}$Ni$_x$As$_2$ crystals with near-optimal doping levels ($x \approx 0.1$). These experiments were performed in magnetic fields ($H$) up to 14 T applied with different angles $\theta$ relative to the crystal’s $c$-axis ($\theta = 0$, 53, and 90 degrees), thus extending previous measurements in the same compound with $H \perp ab$ up to 9 T [21, 22]. The large fields used here allow deep penetration into the so-called Prange fluctuation regime. These fields also allow us to perform a stringent check of the applicability of a recently proposed generalization of the classic Aslamazov–Larkin (AL) results to finite fields through a 3D-anisotropic Ginzburg–Landau (GL) approach [21]. In turn, the use of different magnetic field orientations provides an important consistency test of the analysis, and allows us to obtain precise information about the system dimensionality, basic superconducting parameters (as the coherence lengths and the anisotropy factor), and the angular dependence of the upper critical field, which is currently another debated issue in these materials [26].

2. Experimental details and results

We studied three BaFe$_{2-x}$Ni$_x$As$_2$ single crystals with nominal doping levels near the optimum one, two with
$x = 0.096$ (#1 and #2), and one with $x = 0.098$ (#3). Their sizes are typically $1.5 \times 1.0 \times 0.3$ mm$^3$, being the $c$-axis of the tetragonal structure ($a = b = 3.96$ Å, $c = 12.77$ Å)

$^5$ A theory for the effect of critical fluctuations around the $T_c(H)$ line on different observables in superconductors with intermediate 2D–3D characteristics was developed in [9].
for the three samples studied are compiled in table 1. The small $\Delta T_c/T_c$ values (about $10^{-2}$) confirm the excellent stoichiometric quality of the crystals. An overview of $\rho_{ab}(T)$ in the absence of a field and up to $\approx 2T_c$ is presented in the inset of figure 1(c) for all samples studied. As we can see, $\rho_{ab}$ is almost temperature-independent from a few degrees above $T_c$ up to $2T_c$. This is an important experimental advantage to determine the conductivity induced by superconducting fluctuations (or paraconductivity), which is given by

$$\Delta \sigma_{ab}(T, H) = \frac{1}{\rho_{ab}(T, H)} - \frac{1}{\rho_{ab, \parallel}(T, H)},$$

where $\rho_{ab, \parallel}$ is the normal-state or background contribution. Figure 2 illustrates the procedure to estimate $\rho_{ab, \parallel}$. In the region 26–30 K (corresponding to 1.3–1.5 $T_c$, where fluctuation effects are expected to be negligible [21]), the resistivity is linear with the temperature up to the largest field used in the experiments. Besides, as it is shown in the inset of that figure, the magnetoresistivity in the normal state is roughly quadratic in the applied magnetic field. This allows us to parametrize the background resistivity as

$$\rho_{ab, \parallel}(T, H) = \alpha(H) + \beta(H)T,$$

where

$$\alpha(H) = a_1 + a_2H^2 \quad \beta(H) = b_1 + b_2H^2.$$ 

The coefficients $a_1$, $a_2$, $b_1$, and $b_2$ were obtained by linear fittings to the $\rho_{ab}(T)$ curves measured with $\mu_0H = 0$ and 14 T. An example (corresponding to crystal #1) of the resulting $\Delta \sigma_{ab}$ dependence on the reduced temperature, $\varepsilon \equiv \ln(T/T_c)$, is presented in figure 3.

3. Data analysis

3.1. In-plane paraconductivity in the low-field limit

In the absence of a magnetic field and for temperatures close to $T_c$, it is expected that $\Delta \sigma_{ab}$ will follow the classical AL result, which for 3D superconductors may be written as [28]

$$\Delta \sigma_{ab} = \frac{e^2}{32\xi^2} e^{-\varepsilon^{-1/2}},$$

where $e$ is the electron charge, $h$ is the reduced Planck constant, and $\xi$ is the $c$-axis coherence length amplitude. As we see in the inset of figure 3, for reduced-temperatures below $\varepsilon \approx 0.1$, a critical exponent close to $-1/2$ is observed, in agreement with equation (4). Above this $\varepsilon$-value, a rapid falloff of the fluctuation effects is observed and a well-defined critical exponent is no longer observed, a behavior that may be attributed to short-wavelength fluctuation effects [2, 29, 30]. For completeness, in the same inset we present the prediction of the 2D-AL result

$$\Delta \sigma_{ab} = \frac{e^2}{16\xi^2} e^{-1},$$

where $s = 6.38 \text{Å}$ is the periodicity length of the Fe-As layers. As we can clearly see, it overestimates the experimental data by almost two orders of magnitude, which is well beyond the experimental uncertainties, including those associated with the determination of the normal-state background.

In the presence of a finite magnetic field, roughly above the so-called ghost critical field $H^{\ast}\parallel(T)$ (which is the symmetric above $T_c$ of the corresponding $H^{\ast}\perp(T)$ line [31]), $\Delta \sigma_{ab}$ is expected to be significantly reduced with respect to equation (4) [2, 32, 33]. As we see in figure 3, particularly in the $\Delta \sigma_{ab}(H)$ representation of panels (d–f), such a reduction is clearly observed with the field amplitudes used in our experiments and, as expected, is more prominent for temperatures close to $T_c$ (i.e., for $\varepsilon \to 0$). We also notice the dependence on the field orientation relative to the crystal’s $c$-axis, which is a direct consequence of the anisotropy of the upper critical field in the studied compound (see below).
3.2. Comparison with the GL approach for the finite-field or Prange regime

We will now analyze the experimental data in terms of the 3D-anisotropic GL approach developed in [21]. This approach adapts to the present dimensional case, the model proposed by A Schmid, which is based on a combination of the standard Gaussian GL-expression of the thermally-averaged current density with the generalized Langevin equation of the order parameter [34]. Since the energy of the fluctuation modes increases with $H$, this finite-field approach includes an energy cutoff in the fluctuation’s spectrum as proposed in [29, 30]. For $H$ perpendicular to the $ab$ layers, it leads to

$$ \Delta\sigma_{ab} = \frac{e^2}{32\hbar^2\xi_c^2} \left[ \frac{1}{2} \int_0^{\frac{C-h}{2\hbar}} dx\left( \psi(\frac{\epsilon + h}{2\hbar} + x^2) - \psi\left(\frac{C + h}{2\hbar} + x^2\right) \right) \right], $$

where $h = H/H_z^2$, $H_z$ being the upper critical field (linearly extrapolated to $T = 0$ K for an arbitrary angle $\theta$ between $H$ and the crystal $c$-axis).

Figure 3. Example for crystal #1 of the $\Delta\sigma_{ab}$ dependence on the reduced temperature (a–c) and on the magnetic field amplitude (d–f). The indicated $\theta$ values represent the angle between the applied magnetic field and the crystal $c$-axis. The lines are the best fits for equation (6) using only three free parameters for the entire set of data of each field orientation: $\xi_c$, $H_z(\theta)$, and $C$. The dashed lines in (d–f) represent the crossover to the Prange regime, according to the criterium $\epsilon = h$. Inset in (a): log-log plot of the $\epsilon$-dependence of $\Delta\sigma_{ab}$ in the absence of an applied field. Solid and dotted lines are the best fits for equation (6) and, respectively, the 3D-AL approach, equation (4) (this last for $\epsilon < 0.1$, where short-wavelength effects are expected to be negligible). The dashed line is the prediction of the 2D-AL approach, equation (5).
Equation (6) is fitted to the complete set of data for each field orientation with only three free parameters: the upper critical field $H_{c2}(\theta)$, the amplitude (directly related to $\xi_c$), and the cutoff constant $C$. As we see in figure 3(a–c), the agreement is excellent, extending down to a field-dependent temperature below $T_c$ that may be close to the upper bound of the critical region\(^6\). The agreement is also excellent in the $\Delta\sigma_{ab}(H)$ representation of figure 3(d–f), which is focused on temperatures above $T_c$. The resulting $H_{c2}(\theta)$ values are presented in figure 4 for all samples studied. These data follow the behavior expected for 3D-anisotropic materials (solid lines) \([38]\), which represents an important consistency check of the present results. The $H_{c2}(0^\circ)$ and $H_{c2}(90^\circ)$ values are within the ones obtained in the literature in the same material from the shift of the resistive transition induced by the field \([39–42]\), although the rounding associated with fluctuation effects makes this procedure strongly dependent on the criterion used (generally a given % of the normal-state resistivity). In the present case, as we see in figure 5, the 50% criterion gives $H_{c2}$ values in good agreement with the ones resulting from the analysis of fluctuation effects.

As the $\Delta\sigma_{ab}$ amplitude may be affected by the uncertainties associated with both the finite size of the electrical contacts and the crystal’s geometry, the amplitude term in equation (6) is not used to determine $\xi_c$. Instead, the GL coherence length amplitudes are obtained from the $H_{c2}(\theta)$ values in figure 4, according to

$$\xi_{c0} = \left[\frac{|\phi_0|}{2\pi \mu_0 H_{c2}(0^\circ)}\right]^{1/2},$$

which is evaluated by using the parameters in table 1.

Figure 4. Dependence of $H_{c2}$ on the angle between the applied field and the $c$-axis of the crystals. These data result from the analysis of $\Delta\sigma_{ab}(T, H)$ in the normal state in terms of equation (6). The lines correspond to the 3D-anisotropic GL expression, equation (9), evaluated by using the parameters in table 1.

![Figure 4](https://via.placeholder.com/150)

Figure 5. Temperature dependence of the upper critical field for the three field orientations studied, resulting from the analysis of fluctuation effects (lines) and from the 50% criterium (data points). The data of this example correspond to crystal #1. See the main text for details.

values in figure 4, according to

$$\xi_{c0} = \left[\frac{|\phi_0|}{2\pi \mu_0 H_{c2}(0^\circ)}\right]^{1/2},$$

and

$$\xi_c = \xi_{c0}/\gamma,$$

where the anisotropy factor $\gamma$ is obtained from the ratio

$$\gamma = \frac{H_{c2}(90^\circ)}{H_{c2}(0^\circ)}.$$
Fe₂⁻, and in particular in experiments on the paraconductivity at high-ρ values of the same compound [21], and on the precursor diamagnetism in optimally-doped Ba₁₋ₓKₓFe₂As₂ [14]. It is also close to the cutoff constant found in other superconducting families, including high-Tc cuprates [43–46], low-Tc metallic elements and alloys [47, 48], and compounds like MgB₂ or NbSe₂ [49, 50]. Our results confirm the proposal in [29, 30] about a universal C value close to ~0.5. This value is associated with the limits encountered at high-ρ or h to the shrinkage of the superconducting wavefunction to lengths of the order of the pair size.

Let us finally comment on the applicability of a GL approach to a two-band superconductor as BaFe₂−ₓNiₓAs₂. In principle, the analysis of fluctuation effects in multiband superconductors, in particular when they involve two or more weakly coupled bands with different anisotropy, would require a specific multiband functional that takes into account the nonlocal effects arising from having an effective coherence length in one of the crystallographic directions of the system that is much smaller than the one associated with one of the bands. It has been proposed that this is the case with MgB₂ [51], although a good description of fluctuation effects in terms of GL approaches was also found for this compound [49]. The applicability of a GL approach to BaFe₂−ₓNiₓAs₂ would suggest that the interband coupling in this compound is larger than that in MgB₂. This is consistent with the fact that the relative band interaction constant defined in [51], S₁₂, is in MgB₂ of the order of 0.035, while in optimally doped BaFe₂−ₓNiₓAs₂ we find S₁₂ ≃ 0.134 (i.e., four times larger) by using the coupling parameters reported in [52].

3.3. H–T phase diagram for Δσ_{ab}

The large number of measured Δσ_{ab} isofields allowed us to plot detailed H–T phase diagrams of the Δσ_{ab} amplitude for the three field orientations studied. An example for crystal #1 is shown in figure 6. The solid line is the upper critical field, as obtained from the Tc and Hc₂ values in figure 4 by assuming a linear temperature dependence close to the transition. The dotted line represents the experimental limit of applicability of equation (6), and roughly separates the Gaussian and critical fluctuation regimes. The solid line is the upper critical field resulting from the analysis of Δσ_{ab} (H, T) in terms of equation (6). The dot-dashed line is the so-called ghost field (the symmetric above Tc of the upper critical field), above which finite field effects are expected to be relevant.

\[
T_{irr}(H) = T_{irr}\left(\frac{H}{H_{c2}(0)}\right)
\]

Figure 6. Experimental H–T phase diagram for crystal #1, showing the Δσ_{ab} amplitude for the three H orientations studied. The circles indicate where the resistivity vanishes. The dotted line is the observed limit of applicability of equation (6), and roughly separates the Gaussian and critical fluctuation regimes. The solid line is the upper critical field resulting from the analysis of Δσ_{ab} (T, H) in terms of equation (6). The dot-dashed line is the so-called ghost field (the symmetric above Tc of the upper critical field), above which finite field effects are expected to be relevant.

Then, taking into account equation (9), the H_{irr}(T, θ) lines should scale when normalized by \( (\cos^2 θ + χ^2 \sin^2 θ)^{-1/2} \). As we see in figure 7, such scaling is observed when using the χ values in table 1. Just for completeness, note that the irreversibility line for θ = 0° follows the temperature dependence predicted in [53], which was obtained within a 3D-disordered
3.4. Comparison with recent works

In a recent work by Rullier-Albenque et al [20], it is reported that the paraconductivity of clean LiFeAs samples is 2D in nature, in spite of the fact that the superconducting parameters of this compound (similar to the ones of BaFe$_2$(As$_{1-x}$Sb$_x$)$_2$) would suggest a 3D behavior; $\xi \sim 1.6$ nm is much larger than the interlayer distance, $s = 0.636$ nm. Here we show that this conflicting result may be an artifact associated with the procedure used to determine the normal-state contribution. First of all, let us note that in clean crystals the same fluctuation effects (i.e., the same $\Delta\sigma_{ab}$) lead to a much weaker resistivity rounding than in dirty samples (with a much larger background resistivity). The reason is that not too close to $T_c$, the change in the electrical resistivity due to superconducting fluctuations may be approximated by

$$\Delta\rho_{ab} \approx \rho_{ab,0}\Delta\sigma_{ab}.$$  \hspace{1cm} (15)

In the clean crystals used in [20], $\rho_{ab,0} \approx 4 \times 10^{-8}$ $\Omega$m just above $T_c$. Subsequently, in the case of 3D fluctuations, at intermediate reduced temperatures (e.g., $\varepsilon = 0.1$) the relative change in $\rho_{ab}$ is expected to be about 0.06%. Even in the case of 2D fluctuations, for which $\Delta\sigma_{ab}$ is given by equation (5), the relative change in $\rho_{ab}$ is expected to be smaller than 1%. Indeed, the observation of fluctuation effects in the resistivity of clean crystals would require extraordinary precision in the determination of the background, whatever the procedure used. In [20], $\rho_{ab,0}$ is estimated by allegedly quenching fluctuation effects with magnetic fields typically above 10 T. However, it has been shown that fluctuations above $T_c$ survive up to fields of the order of $H_{c2}/d$ \cite{33}. For LiFeAs this quantity is about 28 T \cite{54}, and it is likely that fluctuation effects are still present above 10 T.

Nevertheless, in [20] a good agreement (without free parameters) is found between equation (5) and the data obtained in one of the samples (named FP2). However, there is a large difference with the results obtained in the other sample (FP1): the $\Delta\sigma_{ab}$ data at 18.8 K ($\varepsilon \approx 0.09$), which is not included in figure 4 of [20] but is available from the data in figure 2(b), is a factor 2.5 larger than the one for sample FP2. The difference cannot be attributed to a wider superconducting transition (as is shown in figure 1(a), both samples present similar transition widths), and suggests that the
agreement of equation (5) with sample FP1 could be accidental. If the fluctuation effects in LiFeAs were actually 3D in nature, the arguments given in [20] supporting a pure $H^2$ behavior for transverse magnetoresistivity in this compound should be revised.

4. Conclusions

We have presented detailed measurements of the conductivity induced by superconducting fluctuations just above the superconducting transition of three high-quality, optimally doped BaFe$_{2−x}$Ni$_x$As$_2$ single crystals. These measurements were performed with magnetic fields up to 14 T, which allowed us to deeply penetrate into the finite-field (or Prange) fluctuation regime. The magnetic field was applied with different orientations with respect to the crystal’s c-axis ($\theta = 0°$, $53°$, and $90°$), allowing us to investigate the anisotropy of fluctuation effects. The analysis of the experimental data leads to solid evidence that a recently published Gaussian GL approach for fluctuation effects in LiFeAs were actually 3D in nature, the arguments given in [20] supporting a pure $H^2$ behavior for transverse magnetoresistivity in this compound should be revised.

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