Gapless state in low- and high-\(T_c\) superconductors: evidence for thermal phase fluctuations

G. Lamura\(^a,^*\), J. Le Cohec\(^a\), A. Gauzzi\(^b\), J.C. Villégier\(^c\), F. Licci\(^b\), B. Plaçais\(^d\), D. Di Castro\(^e\), A. Bianconi\(^e\), J. Bok\(^a\)

\(^a\) Laboratoire de Physique du Solide, ESPCI, 10 rue Vauquelin, F-75231 Paris, Cedex 05, France
\(^b\) MASPEC-CNRS Institute, Area delle Scienze, 43010 Parma-Fontanini, Italy
\(^c\) CEA-Grenoble, SPSMS, Laboratoire de Cryophysique, 17 rue des Martyrs, 38054 Grenoble, France
\(^d\) LPMC, Ecole Normale Supérieure, UMR 8551, 24 rue Lhomond, 75231 Paris, France
\(^e\) Unità INFN and Dipartimento di Fisica, Università di Roma “La Sapienza”, P.le Aldo Moro 2, 00185 Roma, Italy

Abstract

We have carried out a comparative study of the low-temperature dependence of the magnetic penetration depth \(\lambda\) in (1) superconducting NbN films grown on \(R\)-plane sapphire and MgO substrates and (2) Bi2212 single crystals with different doping levels ranging from under- to over-doping. The NbN films display textured or epitaxial microstructure depending on growth conditions. The thermally activated dependence of \(\lambda(T)\) characteristic of conventional s-wave superconductors was found only in the epitaxial or thin textured NbN films. All thick textured films exhibit below 4 K a linear dependence \(\Delta\lambda \sim T\) characteristic of a gapless superconducting state. This is quantitatively explained by a model of granularity-induced thermal phase fluctuations of the order parameter. As for Bi2212, we have found in all crystals a linear low-temperature dependence \(\Delta\lambda_{ab} \sim T\) in the \(ab\)-plane. The slope \(\Delta\lambda_{ab}/\Delta T\) displays a pronounced minimum \(\sim 8\) A/K at optimum doping. This value is in good agreement with the d-wave model proposed for cuprates. However, this model fails to account for the large slope values \(> 40\) A/K observed at both under- and over-doping. These values are quantitatively explained by the same model of phase fluctuations adopted for the textured NbN films, in spite of the difference between these two superconducting systems. © 2001 Published by Elsevier Science B.V.

Keywords: Superconductors; NbN; Bi2212; Phase fluctuations

1. Introduction

It has been established that lines of nodes in the superconducting gap, as in the d-symmetry gap proposed for cuprates, determine a gapless state. The presence of lines of nodes appears as a linear low-temperature dependence of the variations of the magnetic penetration depth \(\Delta\lambda(T)\) [1]. The opposite is not true, i.e. this linear dependence does not imply lines of nodes. The same dependence can be caused by other phenomena, such as magnetic impurities [2], proximity effects [3] or thermal phase fluctuations of the gap [4,5]. To elucidate the origin of the gapless state in cuprates, here we report on \(\Delta\lambda(T)\) measurements in the s-wave superconductor NbN and in Bi2212 single crystals at various doping levels. We show that the same linear dependence can be found in both
systems depending on the granular structure or doping level respectively. Our analysis of the data suggests that the same simple model of thermal phase fluctuations [4,5] quantitatively accounts for the above dependence in both systems, except in NbN films with epitaxial structure or optimally doped Bi2212.

2. Experimental

We have measured seven NbN films and three Bi2212 single crystals. The NbN films were grown epitaxially or textured, depending on growth conditions, on R-plane sapphire and MgO substrates by DC magnetron sputtering (see Table 1), as described elsewhere [6]. As for Bi2212, we have studied one underdoped sample [7] with \(T_c = 73.8\) K and one optimally doped with \(T_c = 92\) K. The latter was subsequently overdoped by annealing it at 550°C in flowing oxygen [8–11]. The resulting \(T_c\) after this treatment was 80.4 K. The variations \(\Delta\lambda\) were measured at low temperatures by using a novel single coil mutual inductance technique with resolution better than 0.1 Å, as described in Ref. [12].

3. Results

In Figs. 1 and 2 we show the experimental curves \(\Delta\lambda(T)\) obtained respectively in the epitaxial NbN films and thin MgO buffered textured one and in the thick textured films. Note that the

![Graph 1](image1)

**Fig. 1.** Low-temperature dependence of \(\Delta\lambda(T) = \lambda(T) - \lambda(1.6\) K) in the epitaxial and the thin (100) textured NbN films (see Table 1). All experimental data are well fitted by the exponential dependence of conventional s-wave superconductors (—).

![Graph 2](image2)

**Fig. 2.** The same as in Fig. 1 for the textured NbN films. The legend refers to Table 1.

<table>
<thead>
<tr>
<th>Sample</th>
<th>A1043</th>
<th>A1013</th>
<th>A1044</th>
<th>A1057</th>
<th>A1060</th>
<th>A1063</th>
</tr>
</thead>
<tbody>
<tr>
<td>Structure</td>
<td>E(100)</td>
<td>E(100)</td>
<td>T(100)</td>
<td>T(111)</td>
<td>T(111)</td>
<td>T(111)</td>
</tr>
<tr>
<td>Substrate</td>
<td>Al₁O₃</td>
<td>MgO</td>
<td>Al₂O₃⁺</td>
<td>Al₂O₃</td>
<td>Al₂O₃</td>
<td>Al₂O₃</td>
</tr>
<tr>
<td>thickness (Å)</td>
<td>100</td>
<td>9500</td>
<td>100</td>
<td>3600</td>
<td>6300</td>
<td>14000</td>
</tr>
<tr>
<td>(J_c(0)) (MA/cm²)</td>
<td>3.0 ± 0.7</td>
<td>—</td>
<td>—</td>
<td>1.3 ± 0.7</td>
<td>1.1 ± 0.3</td>
<td>0.5⁺</td>
</tr>
<tr>
<td>(T_c) (K)</td>
<td>14.2 ± 0.2</td>
<td>15.2 ± 0.2</td>
<td>14.4 ± 0.1</td>
<td>13.3 ± 0.2</td>
<td>16.2 ± 0.2</td>
<td>16.7 ± 0.2</td>
</tr>
<tr>
<td>(\Delta\lambda/\Delta T) (Å/K)</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>2.1 ± 0.5</td>
<td>3.3 ± 0.2</td>
<td>5.6 ± 0.5</td>
</tr>
<tr>
<td>(\Delta\lambda/\Delta T) (fluctuation) (Å/K)</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>1.6</td>
<td>2.2</td>
<td>4.2</td>
</tr>
</tbody>
</table>

*Indicates the presence of a buffer MgO layer. Film thicknesses were measured within ±5% by stylus profilometry.

This value is estimated from Ref. [15].
thermally activated behavior characteristic of s-wave superconductors is found only in the former case (Fig. 1). In the latter case (Fig. 2), a linear dependence is found in the range 1.6–4 K. In Table 1 we report the corresponding slope values $\Delta \lambda / \Delta T$. In Fig. 3 we show the results of $\Delta \lambda_{ab}(T)$ found in the Bi2212 crystals. In agreement with previous studies, in all three samples $\Delta \lambda_{ab} \sim T$ with different slopes depending on doping level (see Table 2). The minimum value $\sim 8$ A/K is found in the optimally doped sample, in agreement with previous reports [13]. Both the under- and over-doped samples exhibit a much larger values $\geq 40$ A/K.

4. Discussion

4.1. NbN

Clearly, the linear dependence shown in Fig. 2 cannot be ascribed either to any non-s gap sym-

Table 2

Summary of the characteristics of the Bi2212 single crystals

<table>
<thead>
<tr>
<th>Sample</th>
<th>$T_c$ (K)</th>
<th>$\Delta \lambda_{ab}$ (Å)</th>
<th>$\Delta \lambda_{th}$ (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Optically (under)</td>
<td>73.8</td>
<td>92</td>
<td>80.4</td>
</tr>
<tr>
<td>Optimally (over)</td>
<td>59.4 ± 0.1</td>
<td>7.8 ± 0.1</td>
<td>47 ± 0.2</td>
</tr>
<tr>
<td>Experimental (Å/K)</td>
<td>10.5</td>
<td>7.6</td>
<td>9.2</td>
</tr>
<tr>
<td>Phase fluctuations (Å/K)</td>
<td>51</td>
<td>41</td>
<td>46</td>
</tr>
</tbody>
</table>

*Experimental and predicted values of $\Delta \lambda_{ab}/\Delta T$. Values of $\Delta \theta$ are calculated from $2\Delta \theta = 5k_B T_c$. We can also rule out any proximity effect, since in NbN grain boundaries are insulating. The only valid model left is that based on granularity-induced thermal fluctuations. A granular model is indeed supported by our TEM study [14] and previous reports [6], which clearly show the presence of parallelepipeds with the main axis orthogonal to the substrate. Within a granular model, the $T$-dependence of $\lambda$ is linear according to the following expression by Coffey [4]:

$$\frac{\Delta \lambda}{\Delta T} \approx \frac{k_B}{h \lambda_c} \frac{k_B}{2e} \approx \frac{2ek_B}{hJ_c(0)a}$$

where $\lambda_c$ is the Josephson critical current, $a$ is the mean size and $e$ the electron charge. At low temperatures $(T/T_c \ll 1)$, $\lambda_c$ can be expressed as $\lambda_c \approx J_c(0) \lambda(0)$, $\lambda(0)$ is the coherence length. In our samples $a$ is estimated to be $\sim 200$ Å [14].

In Table 1 we report the theoretical predictions for the slope values based on Eq. (1). We notice a good agreement with the experimental values within the uncertainty arising from the experimental values of $J_c$ and $a$, suggesting that the proposed phase fluctuation model is satisfactory to account for the gapless behavior of Fig. 2. Note that our simple description based on this model also explains the increase of the slope with increasing film thickness $d$, since $J_c(0)$ is known to decrease with $d$ [15].

4.2. Bi2212

In Table 2 we report the theoretical predictions for the slope values of two models: (1) d-wave; (2) thermal phase fluctuations (see Eq. (2)). We recall that the prediction of the first model requires the knowledge of $\lambda_{ab}(0)$ and the zero-temperature gap $\Delta_0$ (not the pseudogap). As to the prediction of (2), we must modify the formula proposed by Rodrick and Stroud [5] to the case of a strongly anisotropic continuous medium suited to describe cuprates. Within the Gaussian approximation of phase fluctuations, it is obtained:

$$\frac{\Delta \lambda_{ab}(T)}{\Delta T} \approx \frac{\mu_B k_B J_c}{\Phi_0^2} \lambda_{ab}^2(0)$$
where $\Phi_0$ is the flux quantum, $ab$ and $c$ indicate the $ab$-plane and $c$-axis respectively, $\gamma$ is the anisotropy factor and $\kappa_c$ is the Ginzburg–Landau parameter. To apply Eq. (2), we should measure $\gamma$ and $\kappa_c$ as a function of doping in our samples. Instead we take the values at optimum doping ($\gamma \sim 160$ [13,16,17] and $\kappa_c \sim 100$ [16]) and introduce a correction to $\lambda_{ab}(0)$ depending on doping $\delta \approx |p - p_{\text{max}}|$, where $p_{\text{max}}$ is the number of holes per Cu in the CuO$_2$ planes at the optimum doping: using the experimental relations by Presland et al. [18–20] we obtain: $\lambda_{ab}(0) = \lambda^{\text{best}}_{ab}(1 - 82.6 \delta^2)^{-1/2}$. The value $\lambda^{\text{best}}_{ab}(0) \sim 2500$ Å is taken from Ref. [13]. In Table 2 we note a good agreement between the predictions of the d-wave model and the experimental value obtained in the optimally doped sample. However this model does not account for the large values measured in both the under- and over-doped samples for any realistic values of $\lambda_{ab}(0)$ and $\Delta_0$. Such large slopes are rather compatible with the phase fluctuation model of Eq. (2).

In conclusion, only at optimum doping the d-wave model satisfactorily accounts for the presence of low-energy excitations in Bi2212. In non-optimally doped samples, our experimental results indicate that the number of low-energy excitations largely exceeds the number predicted by this model. Another mechanism causing such excitations must then appear and dominate over the d-wave effect. Our analysis of the data supports a simple thermal phase fluctuations model, which quantitatively accounts for the slope values measured in both under- and over-doped samples.

Our results on NbN films support the above scenario of phase fluctuations in cuprates, since the same gapless property found in Bi2212 also appears in thick textured films of s-wave NbN. We propose that the granularity associated with the above textured structure in the 10 nm scale favors phase fluctuations across the grains. In our Bi2212 crystals we have no such direct evidence of granularity. However, a chemical or electronic phase separation has been suggested by many authors [21] for cuprates, thus raising the question of the intrinsic granularity in the nm scale. A systematic study of the doping dependence of the slope $\Delta \lambda_{ab}/\Delta T$ in Bi2212 and in other families of cuprates should help to verify this hypothesis and is the subject of our future work.

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References