Nernst Effect and Disorder in the Normal State of High-T_c Cuprates

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We have studied the influence of disorder induced by electron irradiation on the Nernst effect in optimally and underdoped $YBa_2Cu_3O_{7-\delta}$ single crystals. The fluctuation regime above T_c expands significantly with disorder, indicating that the T_c decrease is partly due to the induced loss of phase coherence. In pure crystals the temperature extension of the Nernst signal is found to be narrow whatever the hole doping, contrary to data reported in the low-T_c cuprate families. Our results show that the presence of intrinsic disorder can explain the enhanced range of the Nernst signal found in the pseudogap phase of the latter compounds.

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The nature of the pseudogap phase remains a key issue in understanding superconductivity in high-T_c cuprates. Among the variety of scenarios that have been proposed [1], an important one is to consider the pseudogap as a precursor to superconductivity, its opening being attributed to a phase-incoherent pairing, the long range coherence occurring only at T_c [2]. An experimental support for this description of the pseudogap regime has been set out recently by the occurrence of a substantial Nernst signal in the normal state of some underdoped cuprates well above the transition temperature T_c [3–5]. As this signal is known to be associated with vortex motion in the mixed state of superconductors, it has been suggested that these Nernst effect experiments reveal the existence of vortexlike excitations surviving in the normal state. In any case, this Nernst signal can hardly be explained without invoking superconducting fluctuations [4,6–9]. This has recently been reinforced by new measurements of a diamagnetic response that has been found to track the Nernst signal [10].

It has been considered somewhat independently by Emery and Kivelson [11] that in a sufficiently bad metal classical and quantum phase fluctuations of the superconducting order parameter can depress T_c well below its mean-field value. We have previously shown that the defect induced decrease of T_c could be partly explained within this scenario [12]. One might wonder whether this results as well in a large range of incoherent phase fluctuations above T_c. In order to test this possibility, we have undertaken a systematic study on the influence of defects on the Nernst effect. We have chosen to perform experiments in optimally doped $YBCO_7$ and underdoped $YBCO_{6.6}$ compounds, which are known to be very homogeneous systems with little intrinsic disorder [13]. The controlled introduction of defects has been achieved by using electron irradiation at low temperature, which results in the creation of point defects such as Cu and O vacancies in the CuO_2 planes [14]. We demonstrate here for the first time that the presence of defects induces the apparition of a Nernst signal in a large temperature range above T_c in both compounds. This is a strong confirmation that phase fluctuations do play a role in the decrease of T_c induced by disorder. Moreover, we find that the onset temperature of the Nernst effect is not much dependent on the defect content and remains close to that of the pure system. We discuss the implications of these results on the analysis of the existing data on systems with lower intrinsic T_c such as LaSrCuO or La-doped Bi2201 [4].

The single crystals used in this study were grown using the standard flux method. Very small contacts with low resistance ($<0.1\ \Omega$) were achieved by evaporating gold pads on the crystals on which gold wires were attached later with silver epoxy. Subsequent annealings have been performed in order to obtain crystals with oxygen content $\sim$7 and $\sim$6.6. The T_c values are defined here as the zero resistance temperatures. The irradiation were carried out with 2.5 MeV electrons in the low temperature facility of the Van de Graaff accelerator at the LSI (Ecole Polytechnique, Palaiseau). During irradiation, the samples were immersed in liquid H_2 and the electron flux was limited to $10^{14} e/\mathrm{cm}^2/\mathrm{s}$ to avoid heating of the samples during irradiation. The thicknesses of the samples (20 to 40 μm) are very small compared to the penetration depth of the electrons, which warrants homogeneous damage throughout the samples. We report here data taken on three $YBCO_7$ samples: a pure one with $T_c = 92.6$ K and two irradiated ones at different electron fluences with respective $T_c = 79.5$ and 48.6 K, and four $YBCO_{6.6}$ samples: pure with $T_c = 57$ K and irradiated with $T_c = 45.1$, 24.2, and 3 K.

The Nernst signal E_y is the transverse electrical response to a thermal gradient $\nabla_x T \parallel x$ in the presence of a perpendicular magnetic field $B \parallel z$. For the measurements the sample was attached on one end to a copper block with the other end free. The temperature gradient was created with a small RuO_2 resistance attached to the free end. The measurements were performed under vacuum (10^{-2} to 10^{-1} mbar), and a heater power ranging from 0.01 to

0.02 mW was used to create temperature gradients from 0.5 to 0.8 K/mm depending on the temperature of measurement. The thermal gradient was measured with a differential chromel-constantan thermocouple. The data were taken at fixed T with magnetic field sweeps from 0 to 8 T. At some given value of the magnetic field, the thermal gradient is removed, which allows us to subtract offset voltages due to contact misalignment or an eventual contribution of the wires.

As described by Wang *et al.* [4], the Nernst coefficient $\nu = E_y/(-\nabla_x T)B$ is the contribution of two terms:

$$\nu = \frac{E_y}{(-\nabla_x T)B} = \left[\frac{\alpha_{xy}}{\sigma} - S\tan\theta\right]\frac{1}{B}, \qquad (1)$$

where α_{xy} is the off-diagonal Peltier conductivity $[J_y = \alpha_{xy}(-\nabla_x T)]$, $\theta = \sigma_{xy}/\sigma$ is the Hall angle, and S is the thermopower. The quantity of interest is the off-diagonal term α_{xy}, which involves the normal state term α_{xy}^n and the vortex contribution α_{xy}^s. In order to probe the influence of disorder on the latter, it is very important to determine as well the influence of disorder on $S\tan\theta$. We have therefore measured E_y, S, and $\tan\theta$ separately in each sample by using the same electrodes for measuring resistivity and Hall effect in one setup and the thermopower and Nernst coefficients in another one.

Let us present first the results obtained on underdoped crystals. Figure 1 shows several curves of the Nernst signal $e_y = E_y/(-\nabla_x T)$ as a function of magnetic field for the pure crystal. For $T < 55$ K, e_y is zero as long as the magnetic field does not exceed the "melting" field $B_m(T)$ necessary to depin vortices (around 2 and 1 T, respectively, at 35 and 45 K). Then the rapid increase of e_y above B_m reflects the motion of vortices induced by the thermal gradient. As T is increased across T_c, the Nernst

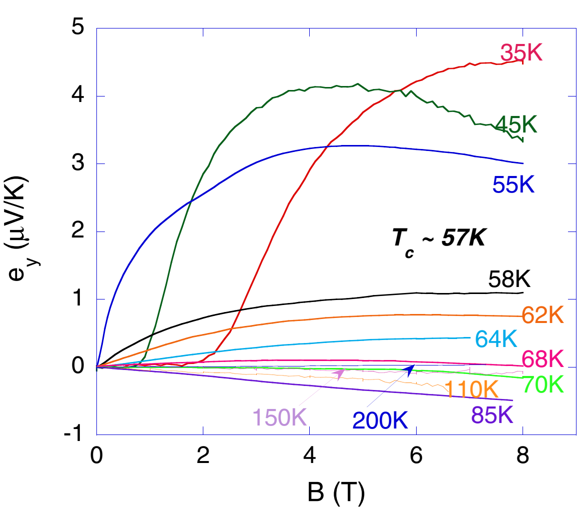

FIG. 1 (color online). Nernst signals $e_y = E_y/|\nabla T|$ versus magnetic field in the pure underdoped $YBCO_{6.6}$ crystal for T ranging from 35 to 200 K.

signal initially drops rapidly (curves at 55 and 58 K) and then decreases gradually approaching a straight line with negative slope. This behavior is clearly displayed in Fig. 2(a) in which we have plotted the T variation of the Nernst coefficient ν determined as the initial slope of e_y versus B. This negative contribution, which has been previously observed near T_c [15], is found to display a minimum at 85 K and to vanish around 200 K. This behavior, which is quite different from that observed in the other underdoped cuprates, will be seen below to result naturally from the high value of $S\tan\theta/B$ in this clean system.

The effect of electron irradiation is recalled in Fig. 2(c) where the resistivity curves $\rho(T)$ are plotted for the pure and two irradiated samples. As reported previously [16], Matthiessen's rule is well obeyed at high T, which indicates that the hole doping of the CuO_2 planes and the pseudogap temperature T^* are not significantly modified [17]. The initial parts of the low T upturn of $\rho(T)$ have been associated with a Kondo-like scattering [16]. As for the Nernst coefficient, one observes in Fig. 2(a) that the pronounced minimum that is present for the pure sample is

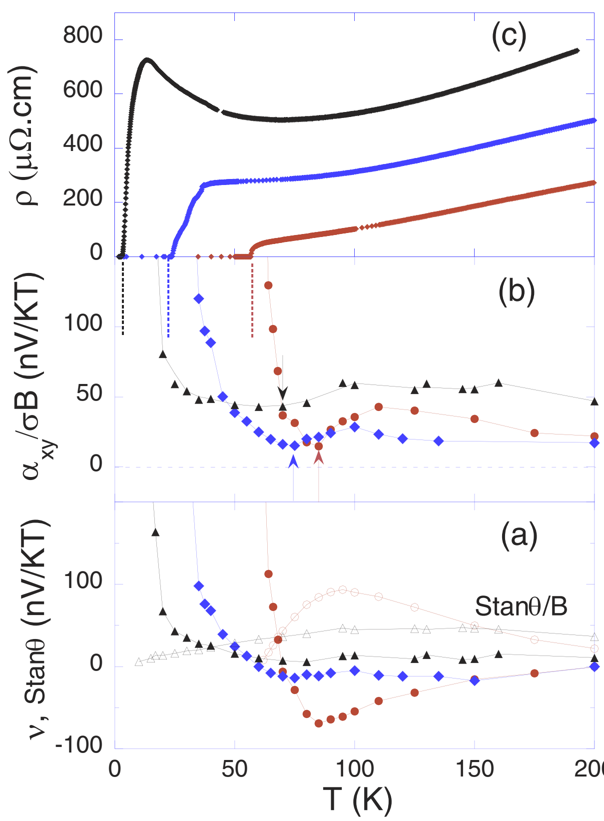

FIG. 2 (color online). Temperature evolution for the pure and two irradiated underdoped $YBCO_{6.6}$ samples of (a) the Nernst coefficient determined by the initial slope of e_y versus B (the T dependence of $S\tan\theta$ is also plotted for the pure and the most irradiated samples), (b) $\alpha_{xy}/\sigma B$ determined from Eq. (1), and (c) the resistivity. The arrows in (b) indicate the onset temperature of the vortex Nernst contribution.

smoothed out by the introduction of disorder. We have reported on the same graph the temperature dependence of $S\tan\theta/B$ for the pure and the most irradiated sample. In the pure sample with both S and $\tan\theta$ being quite large [18], $-S\tan\theta/B$ dominates in Eq. (1). Such a negative value of ν has been predicted theoretically in the framework of the Boltzmann theory by taking into account the role of the Fermi surface shape at two dimensions [20]. When increasing the defect content x, we find that S varies slightly while $\tan\theta/B$ decreases roughly as $1/x$ [21], resulting in a decrease of $S\tan\theta/B$ when T_c decreases. The T variation of the total off-diagonal Peltier term $\alpha_{xy}/\sigma B$ obtained by combining the data for $S\tan\theta$ and ν is plotted in Fig. 2(b). In all samples the normal state contribution α_{xy}^n presents a broad peak around 110 K and then decreases with temperature. Such a behavior has been quite generally observed in underdoped cuprates [4] and might therefore be characteristic of the normal state quasiparticles. As $\alpha_{xy}^n/\sigma B$ corresponds to a carrier-entropy current, one expects that it should decrease to zero at $T \rightarrow 0$. Therefore it seems legitimate to interpret any deviation from this tendency as a manifestation of a vortex contribution. We have thus indicated by the arrows in Fig. 2(b) the best estimate of the onset temperature T^ν of the superconducting contribution. This determination leads, in fact, to values that nearly coincide with those of the minimum of the Nernst coefficient. Two important results can be deduced from this plot. First, it is clearly seen that the onset temperature does not exceed 85 K in pure $YBCO_{6.6}$, showing that the fluctuation regime is quite narrow (~25 K) in this compound despite the fact that the pseudogap temperature T^* is $\gtrsim$300 K whatever the experimental probe [24]. Second, we find that T^ν is nearly the same for all the samples while T_c has been decreased down to 5 K by irradiation. This is a strong indication that the presence of defects plays a prominent role in the observation of a Nernst signal in the normal state of these samples.

As for $YBCO_7$, the Nernst coefficients are reported in Fig. 3 for the three samples studied. In the pure crystal the magnitude of the negative value of ν is much smaller than in the underdoped case. This results from the fact that $S\tan\theta/B$ is also smaller as shown by the decomposition displayed in the inset of Fig. 3. This $S\tan\theta/B$ term varies very little with defect content, and the estimate of T^ν is about the same whether we use the raw data for ν or the corrected values $\alpha_{xy}/\sigma B$. For all the samples the drop of the Nernst signal is very rapid at T_c but while it vanishes at ~10 K above T_c in the pure sample, it persists up to ~85 K, that is to say 35 K above T_c, in the most irradiated one. The fairly narrow fluctuation range found in the pure sample is similar to the one deduced from the paraconductivity in the $\rho(T)$ curves.

In order to compare results obtained on $YBCO_7$ and $YBCO_{6.6}$, we have reported in Fig. 4 the values of T^ν as a function of T_c for the different samples. In both compounds we observe that the Nernst signal extends in a

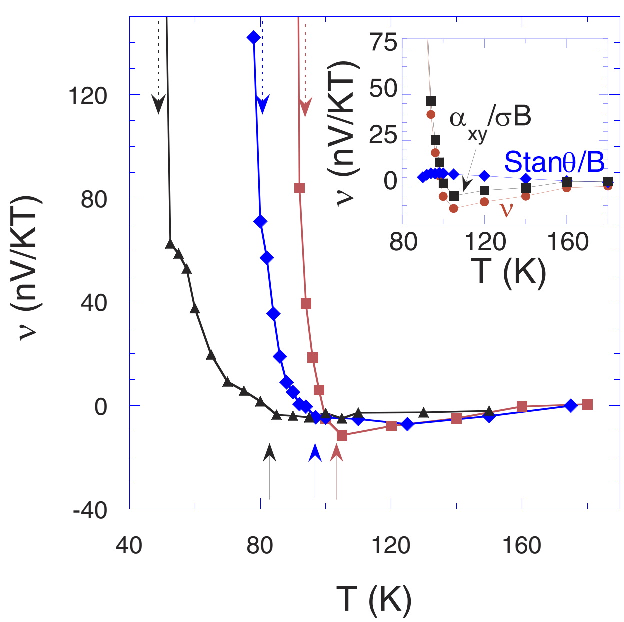

FIG. 3 (color online). The Nernst coefficient ν is plotted versus temperature for pure and irradiated single crystals of $YBCO_7$. The values of T_c and T_ν are, respectively, indicated by dashed and full arrows. The T dependences of ν (circles), $S\tan\theta/B$ (diamonds), and $\alpha_{xy}/\sigma B$ (squares) are shown in the inset for the pure crystal.

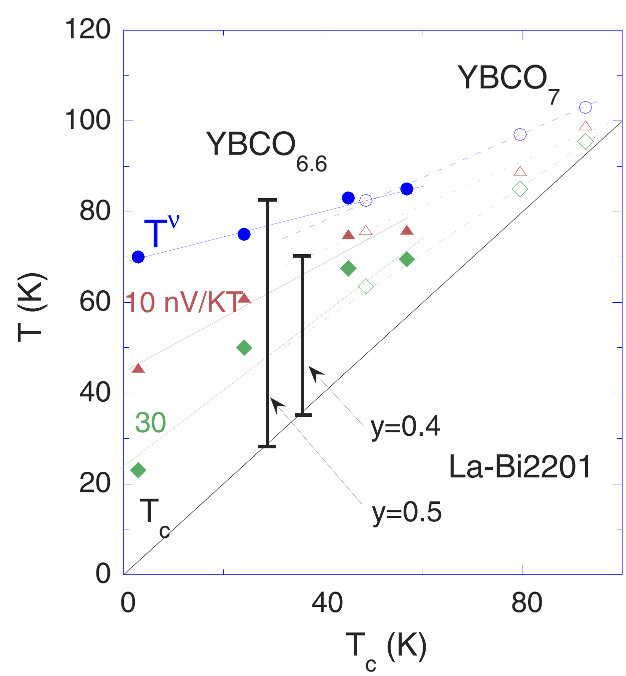

FIG. 4 (color online). The values of T^ν are plotted versus T_c together with the T values corresponding to vortex Nernst contributions of 10 and 30 nV/KT for $YBCO_7$ (empty symbols) and $YBCO_{6.6}$ (closed symbols). These data are compared to the temperature ranges of the Nernst signal measured in pure single crystals of $Bi_2Sr_{2-y}La_yCuO_6$ with $y = 0.4$ and $y = 0.5$ [4].

larger temperature range when decreasing T_c. Let us point out that this effect corresponds to very small values of the vortex Nernst signal as one can see that the temperature corresponding to a value of the vortex Nernst signal of 30 nV/KT nearly follows the T_c decrease.

These results clearly show that superconducting fluctuations survive in the normal state of both optimally doped and underdoped YBCO when T_c is decreased by the introduction of disorder. As T^ν can be considered as the characteristic temperature below which local pairing remains significant, the T_c decrease induced by disorder can be explained only by taking into account both phase fluctuations and pair-breaking effects. This gives strong support to our previous interpretation of the quasilinear decrease of T_c with defect content that is observed down to $T_c = 0$ [12]. It is worth mentioning here that the role of quantum phase fluctuations has also been invoked to explain the Nernst effect observed in the normal state of low T_c cuprates when superconductivity is suppressed by magnetic fields [25,26].

One striking point that can be seen here is the small range of superconducting fluctuations observed in the pure $YBCO_{6.6}$ and $YBCO_7$ compounds. This is much smaller than the corresponding observations done in other "pure" cuprates such as LaSrCuO or La-doped Bi2201 [3,4]. Let us recall here that the presence of extended vortex fluctuations in these underdoped cuprates has been invoked as a strong indication that d-wave superconductivity is closely related to the pseudogap state [4,10]. Our results show that this argument fails in pure underdoped $YBCO_{6.6}$, suggesting that the energy scales of T^ν and T^* are not connected. We can even see that in these clean systems T^ν and T_c increase with increasing hole doping while T^* definitely decreases.

It has previously been suggested that the low T_c in some cuprate families could be due to the presence of intrinsic defects as deduced from the analysis of ^{17}O NMR data [27]. One can wonder whether this might also explain the magnitude of the Nernst effects. We have therefore compared in Fig. 4 the temperature extensions of the Nernst signal of our irradiated samples with those obtained in the $Bi_2Sr_{2-y}La_yCuO_6$ family [4] for $y = 0.4$ ($T_c \sim 36$ K) which corresponds to optimal doping and for $y = 0.5$ ($T_c \sim 29$ K) with $T^* \sim 300$ K comparable to that of $YBCO_{6.6}$ [28]. The quite good agreement between the ranges of vortex Nernst signals found in these different samples indicates that the "intrinsic" disorder in $Bi_2Sr_{2-y}La_yCuO_6$ could also be responsible for the enhanced Nernst signal. Indeed, cation disorder on the Sr site has recently been identified [29,30]. It is then natural to conclude that the "anomalously" high values of T^ν with respect to T_c found in La-doped Bi2201 are indicative of the values of T_c that these materials should display if they were grown without local inhomogeneities. Such a conclusion might as well apply to other cuprate families [31] and is reinforced by the fact that the maximum of T^ν is in most cases of the order of 100 K [4]. Moreover, our results reveal that defects induce more phase fluctuations in the underdoped phase than for optimal doping, which might be due to lower phase stiffness and less efficient screening. It is therefore our opinion that the link between the large range of Nernst signal and the pseudogap phase has to be found in priority in the occurrence of defects and in the large sensitivity to disorder of the superconducting-pseudogap phase.

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