Influence of the Spin Gap on the Normal State Transport in $YBa_2Cu_4O_8$

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(Received 2 October 1992)

The anisotropic resistivity components of $YBa_2Cu_4O_8$ have been measured from 0 to 900 K. $YBa_2Cu_4O_8$ is an underdoped, genuinely untwinned, and stoichiometric high temperature superconductor. The CuO_2 plane resistivity ρ_a exhibits a strong correlation with the spin dynamics, i.e., with Knight shift and spin-lattice relaxation time T_1. The resistivity along the b axis (plane plus chain) is consistent with Bloch-Grüneisen theory. The cotangent of the Hall angle is quadratic in temperature above the spin gap, and linear below.

PACS numbers: 74.72.Bk, 72.15.Qm, 72.15.Gd

Since the revolution of high temperature superconductivity (HTSC), initiated by the report of Bednorz and Müller [1], the unusual normal state transport properties of the HTSC's have been suspected of giving clues to the basic mechanism responsible for superconductivity.

Especially the normal state resistivity has revealed an anomalous response [2]: The optimal doped samples as, e.g., $YBa_2Cu_3O_7$ (1:2:3) have shown a linear behavior in temperature from T_c up to some hundreds degrees Kelvin. In the overdoped regime, the exponent α of $\rho \propto T^{\alpha}$ changes from $\alpha = 1$ to 2; i.e., a Fermi-liquid behavior becomes dominant. The underdoped samples exhibit a nearly T linear dependence with deviations at low temperatures.

In the overdoped region, the basic mechanism responsible for superconductivity is hidden due to the excess carrier density. Hence, as concerns the basic mechanism of HTSC, the underdoped region, which is mainly covered by nonstoichiometric samples, should be explored. Unfortunately, nonstoichiometry is accompanied by distortions, nonhomogeneity, oxygen defects, etc., which, in addition, are modestly screened, due to the low carrier density. As a result, the intrinsic conductivity of the low doped CuO_2 plane is not easy to uncover (see also Fig. 1).

A fingerprint of underdoped samples is an opening of a spin gap above T_c and a magnetic susceptibility increasing with temperature. Both signatures have been found in $YBa_2Cu_4O_8$ (1:2:4) by means of the spin Knight shift [3] and the spin-lattice relaxation time T_1 [4]. 1:2:4 has a structure similar to 1:2:3 except the single chains along the b direction are replaced by double chains. $YBa_2Cu_4O_8$ is a bonus from nature since it is simultaneously stoichiometric, underdoped and, in addition, genuinely untwinned. Because of that, the unvarnished conductivity component of the low doped CuO_2 plane can be determined.

In this Letter, we present the conductivity components for the a axis representing the CuO_2 plane as well as for the b axis containing a plane and a chain contribution. Furthermore, we have measured the Hall effect for the CuO_2 plane. The results prove the intimate relationship between dynamic spin susceptibility and normal state transport which seems to be the clue to understanding HTSC's [5].

1:2:4 single crystals were grown with a high-pressure flux method using a BaO-CuO-rich flux. The growing parameters were 640 bars O_2 at a temperature of 1040°C. For transport measurements, gold pads were sputter deposited with the contacts aligned with the crystal axes. After an annealing at 500°C, gold wires were attached by gold epoxy. The contact resistance was less than 1 Ω.

In Fig. 2 the resistivity along the a direction is depicted. The resistivity ρ_a is nearly linear from 200 to 900 K with a positive intersection at zero temperature. Zimmermann *et al.* [4] have found a spin gap in polycrystalline, c axis oriented 1:2:4 that opens below the temperature $T_D^* \approx 160$ K. As the spins are frozen below T_D^* the scattering channel closes and, hence, the resistivity should decrease faster. This explains the signature of ρ_a in Fig. 2.

The reasons that the above-mentioned relationship has been hidden by now are numerous. A spin gap opening above T_c has been detected by nuclear quadrupole reso-

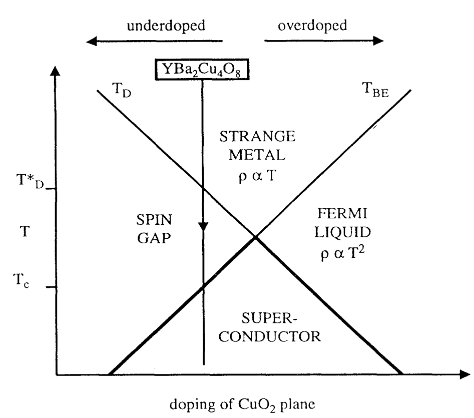

FIG. 1. Scheme of the mean field phase diagram for the CuO_2 plane as proposed by Nagaosa and Lee [26]. The thin lines indicate the spin gap opening at T_D and a Bose-Einstein condensation T_{BE} of the charge carrier, respectively. Also depicted is the temperature trajectory for the underdoped $YBa_2Cu_4O_8$ with the spin gap opening at T_D^* and the transition to a superfluid at T_c.

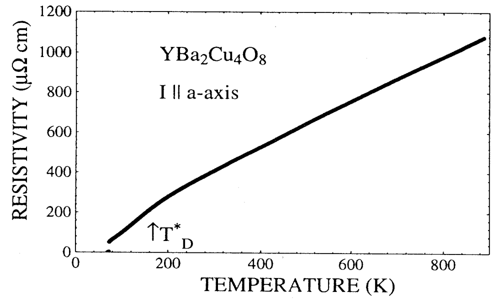

FIG. 2. Resistivity ρ_a of $YBa_2Cu_4O_8$ measured up to 900 K. Also quoted is the spin gap temperature as measured by Zimmermann *et al.*

nance (NQR) in the (LaSr)CuO family [6] only about 10 K above T_c and, therefore, the effect in question is disguised by fluctuation effects. Results of neutron scattering on $La_{1.85}Sr_{0.15}CuO_4$ have also been interpreted as a spin gap [7]. A spin gap above T_c is established in weakly doped 1:2:3 by NQR [8,9] at 130 K and by neutrons around 180 K [10]; but, as the underdoped 1:2:3 samples are nonstoichiometric compounds, the resistivity is strongly affected by the oxygen vacancies; Anderson localizations due to the random oxygen vacancies produce a resistivity increase at low temperature in contrast to stoichiometric 1:2:4. The TlBaCuO compounds have shown [6] a spin gap that coincides with T_c since most of them are in the overdoped regime where the spin gap opens at T_c.

In Fig. 3 the resistivity ρ_b is presented from 0 to 900 K. It reflects the combined response of the double chain and the CuO_2 plane. At room temperature, the conductivity is about 3 times higher than for the pure plane conductivity ρ_a; hence, the chains are metallic conductors as indeed proved [4] by the Korringa-like law for the spin-lattice relaxation time T_1 of the chain copper Cu(1). The temperature behavior is completely different with respect to ρ_a and may be assumed to be predominantly of a Bloch-Grüneisen form for the metallic chains and an additional parallel path for the plane contribution:

$$\rho(T)=\rho_0+\left\{\left[R_1\left(\frac{T}{\Theta_D}\right)^5\int_0^{\Theta_D/T}\frac{x^5\,dx}{(e^x-1)(1-e^{-x})}\right]^{-1}+(G\rho_a)^{-1}\right\}^{-1}. \tag{1}$$

The fit with Eq. (1) is depicted in Fig. 3. The parameters are the effective transport Debye temperature $\Theta_D^{\rm tr}=845$ K (compared to the thermodynamic value $\Theta_D^{\rm thermo}=470$ K [11]), the factor $R_1=2450$, the residual resistivity $\rho_0=15$ $\mu\Omega$ cm, and the geometric factor $G=2.8$. If plane and chain had independently contributed a conduction channel one might have expected $G\approx 1$; in reality, the conductivity of the plane seems to be suppressed if the path along the chain is open [12]. The high $\Theta_D^{\rm tr}$ does not sound physical; but in $\Theta_D^{\rm thermo}$ all types of phonons are involved whereas $\Theta_D^{\rm tr}$ considers only acoustic modes [13].

The applicability of the Bloch-Grüneisen (BG) theory for the b direction explains the successful fits with BG of the $\langle ab\rangle$ averaged resistivity of thin film [14] and polycrystalline 1:2:4 [15] since the 3 times higher conductivity of the b direction dominates the temperature dependence.

In Fig. 4 we have shown R_H with $I \| a$ axis. The value is about a factor of 3 smaller than for untwinned 1:2:3 [16]. Band structure calculations [17] of 1:2:4 have given such a low R_H although a similar value has been predicted for 1:2:3. Again at the spin gap temperature T_D^* ≈ 160 K, a change in the response is evident; the curve reveals a point of inflection at around 160 K.

To cope with the Hall effect in the magnetic systems of the HTSC's, Anderson has proposed [18] to evaluate the Hall angle Θ_H rather than R_H. The idea of this approach is the cancellation of the normal longitudinal transport scattering in R_H:

$$\cot\Theta_H=\frac{\sigma_{xx}}{\sigma_{xy}}=\frac{1}{\omega_c\tau_H}\propto\frac{1}{\tau_H}. \tag{2}$$

The transverse Hall relaxation rate τ_H is due to scattering between spin excitations alone and goes as T^2 (like fermion-fermion scattering). Additional magnetic moments implant an additive term to $1/\tau_H$. The predict-

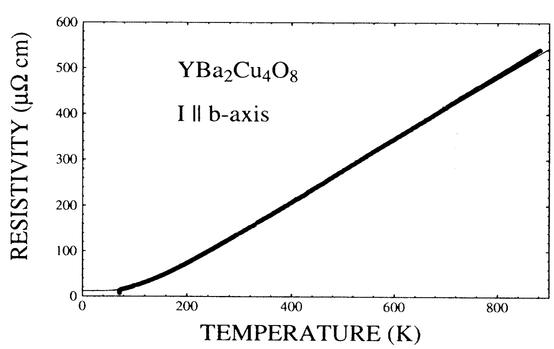

FIG. 3. Resistivity ρ_b of $YBa_2Cu_4O_8$. The thin line represents the Bloch-Grüneisen fit (see text).

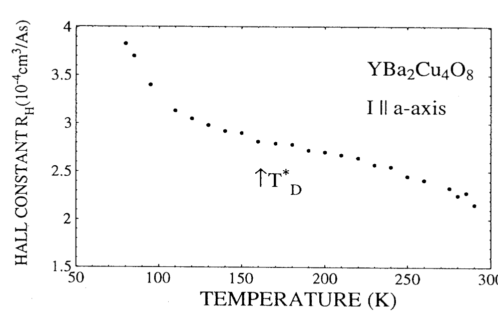

FIG. 4. Hall constant R_H of 1:2:4 with the current $I \| a$ axis. Also depicted is the spin gap temperature T_D^*.

ed $1/\tau_H \propto T^2+C$ has indeed been found by Chien, Wang, and Ong [19] for $YBa_2Cu_{3-x}Zn_xO_7$ and by Rice *et al.* [16] for untwinned $YBa_2Cu_3O_7$.

In Fig. 5 the cotangent of Θ_H vs T^2 of 1:2:4 is plotted for $I \| a$ and $I \| b$, respectively. $\cot(\Theta_H)$ is intimately connected with the resistivity, e.g., for $I \| a$ axis $\cot(\Theta_H^a) \propto \rho_b/R_H$ and vice versa [20]. As R_H for $I \| a$ and $I \| b$ do not differ significantly [21] the resistivity is responsible for the difference of the two Hall angles.

For $I \| a$ the transverse scattering channels are chain and plane. The response can be perfectly modeled by the form $1/\tau_H = AT^2+C$ with $A=0.07$ and $C \approx 0$. An influence of the spin gap is not seen as with ρ_b; the metallic double chain may short circuit the Hall conductivity perpendicular to $I \| a$, i.e., along the b direction. In comparison, the values of $\cot\Theta_H$ of untwinned 1:2:3 are $A=0.025$ and $C \approx 0$ [16].

For $I \| b$ the influence of the spin gap is evident; a Hall conductivity emerges from a feasible deviation of the charge carrier inside the plane but not perpendicular to the chain. The fitted values are $A=0.24$ and $C \approx 3500$ at the magnetic field of 1 T. The analysis of the data is subtle since the current is divided, in a first approximation as 2:1, into a chain and a plane part. The constant C may be due to the chain. As the current in the plane is overestimated by a factor 3, the parameter A renormalizes to ≈ 0.08 yielding the same order as for 1:2:3.

In Fig. 6 we have plotted the Knight shift K_{spin} and $1/T_1T$ of 1:2:4. In a metal, the Knight shift should be constant as a function of temperature. The steep decrease of K_{spin} below 260 K could be assigned to the opening of a spin gap. The spin-lattice relaxation rate $1/T_1T$ reveals a spin gap around 160 K. Hence the pseudospin gap opens at different temperatures inside the Brillouin zone. In a nearly antiferromagnetic Fermi liquid, it has been shown that ρ/T is constant [22,23]. The plot ρ_a/T clarifies the deviation from the linear behavior due to, e.g., a condensation phenomenon.

The phase diagram shown in Fig. 1 has been proposed as a phenomenological logical approach to the experimental fact of a spin gap above T_c by means of two independent, macroscopic order parameters, involving spin-charge separation with separate condensation of the spin degree of freedom below T_D. A microscopic explanation of the reduced resistivity is then based on an increased mobility of the doped holes which may be described, e.g., as a Zhang-Rice singlet [24] or more heuristically as a spin polaron [25].

The magnetic scattering mechanism of the charge carrier is still debated. Nagaosa and Lee [26] have predicted a low-**q** scattering of charge carriers on chiral spin excitations which are situated around the origin of the dispersion scheme of the spin excitations. The Knight shift K_{spin} of the spins is the appropriate quantity to probe the origin of the $\omega(\mathbf{q})$ dispersion relation as a function of temperature. If the resistivity is determined by a low-**q** scattering the spin gap temperature of the resistivity coincides with the onset temperature of the spin gap in the Knight shift (see Fig. 6).

High-**q** scattering is due to AF fluctuations at the antiferromagnetic point $\mathbf{q} \approx (\pi,\pi)$ of the Brillouin zone. The spin dynamics at (π,π) of the HTSC's is dominated by the response of Cu(2) inside the CuO_2 plane. Hence, $1/T_1T$ of ^{63}Cu(2) represents mainly the magnetic behavior at (π,π). If the resistivity is determined by high-**q** scattering the spin gap of the resistivity resembles the spin gap behavior of $1/T_1T$.

Figure 6 does not clearly favor either the high-**q** or the low-**q** scattering mechanism although the gap opening of the resistivity is closer to $1/T_1T$. A difference of the spin gap temperatures T_D^* may also be due to the fact that the resistivity is measured in single crystals whereas the magnetic probes are determined on polycrystalline samples.

The relationship of spin dynamics and dc conductivity of the CuO_2 plane extends even to optical frequencies; the optical conductivity $\sigma_a(\omega)$ of untwinned 1:2:3 also has revealed [27] a correlation with the NMR Korringa prod-

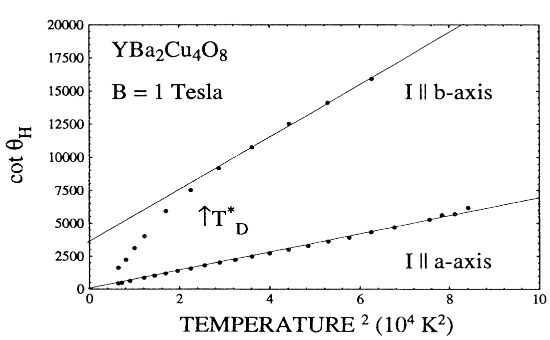

FIG. 5. Cotangent of the Hall angle Θ_H of $YBa_2Cu_4O_8$ with the current I parallel to the a and b directions, respectively, and the magnetic field $H \| c$. The thin lines are parabolic fits to the data above the spin gap.

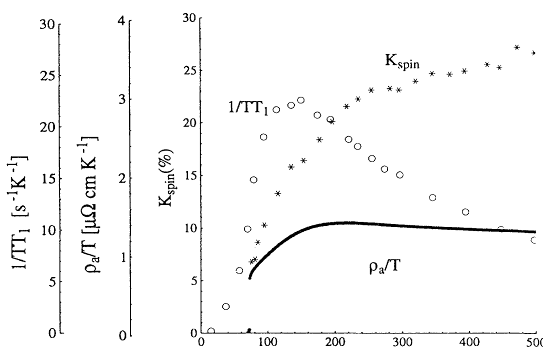

FIG. 6. Connection of the different magnetic probes of the CuO_2 plane in 1:2:4. The plot ρ_a/T clarifies the relationship of the spin gap temperature T_D^* of the resistivity to Knight shift K_{spin} and spin-lattice relaxation time T_1 (magnetic data after Zimmermann *et al.*).

uct $1/T_1T$.

The slight decrease of ρ/T with increasing temperature above T_D^* is an effect of the resistivity offset at zero degrees Kelvin; to exhibit a temperature dependence as $1/T_1T$, the resistivity must show a positive intersection at 0 K. The question arises as to whether the resistivity onsets of the underdoped compounds are intrinsic features due to antiferromagnetic spin fluctuations. Indeed, theoretical calculations of the resistivity of nearly antiferromagnetic itinerant electron systems have predicted an intrinsic offset at 0 K in the low doped regime. However, in this doping region an antiferromagnetic long-range order is expected; on the other hand, the zero crossing of $1/T_1$ and ρ_a around 40 K may be interpreted within the context of lifetime effects of AF spin fluctuations as a magnetic ordering which is impeded by the superconducting transition.

In conclusion $YBa_2Cu_4O_8$ is a suitable compound to investigate underdoped HTSC's. The CuO_2 plane transport in the normal state is governed by the spin dynamics; the spin gap below 160 K results in a diminished scattering of the charge carrier. ρ_b (chain plus plane) is dominated by the metallic transport of the double chains and can be fitted by the Bloch-Grüneisen function. The Hall data can be understood by means of the idea of a Luttinger liquid.

We are very indebted to Professor T. M. Rice for crucial hints and discussions. P. Sutter and K. Mattenberger are acknowledged for technical assistance.

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