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Anisotropy and double sign change of the Hall resistivity in $YBa_2Cu_4O_8$ single crystals

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The electrical resistivity and the Hall effect have been measured on oriented single crystals of $YBa_2Cu_4O_8$ in magnetic fields up to 10 T and temperatures from 2 to 300 K. In the normal state the anisotropic, transverse relaxation rates determined from the Hall angle for $\mathbf{I}\|\mathbf{a}$ and $\mathbf{b}$ agree with those obtained from NQR for Cu(2) and Cu(1), respectively, and indicate the formation of a spin gap. In the mixed phase the Hall effect changes its sign twice, but only when $\mathbf{I}$ is parallel to the chains. An interpretation in terms of a two-band model is given.

Since the discovery of high-temperature superconductivity in cuprates,[1] several authors have reported on Hall-effect measurements and nearly as many models have been proposed to explain different anomalous features. Besides the temperature dependence in the normal state, the temperature and field dependence in the mixed state has received particular attention. In the normal state $YBa_2Cu_3O_7$ (1:2:3) shows a $1/T$ temperature dependence of the Hall coefficient R_H (Refs. 2–8) which together with the $1/T$ temperature dependence of the conductivity σ gives for the tangent of the Hall angle $\tan\theta_H = R_H \sigma B$ a proportionality to $1/T^2$. This dependence of the transverse relaxation time $\tau_{\rm trv}$ was explained by Anderson[9] within a two-dimensional Luttinger-liquid model as an effect of spinon-spinon scattering. On the other hand, Hall-effect measurements on $YBa_2Cu_4O_8$ (1:2:4) films[10] and ceramics[5] showed a temperature-independent behavior in the normal state, raising the question whether this is a material or an anisotropy effect. To answer this question unambiguously measurements on untwinned oriented single crystals are necessary.

A second puzzle occurs in the mixed state, i.e., for temperatures a few degrees below $T_c(B=0)$, when a magnetic field is applied. In this regime, $YBa_2Cu_3O_7$ (Refs. 4 and 5) and $Bi_2Sr_2CaCu_2O$ single crystals[4,11] as well as $YBa_2Cu_3O_7$ and $ErBa_2Cu_3O_7$ epitaxial films[12–14] show at constant temperature with decreasing magnetic field a sign change from positive to negative Hall resistivity, before going to zero in the flux-creep region. $Tl_2Ba_2CaCu_2O_8$ films may even show two sign changes as a function of temperature for a constant field of the order of 2 T.[15,16] Also n-type superconducting $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ shows a sign change, but from negative to positive Hall resistivity if the temperature is decreased in the mixed phase.[17] Judging from the only existing measurements on ceramics,[5] $YBa_2Cu_4O_8$ appeared to be the exception among the high-T_c cuprates in showing no sign change of the Hall effect in the mixed state.[17]

In this paper we will show that $YBa_2Cu_4O_8$ also shows a sign change of the Hall effect. However, a novelty is that this sign change is limited to the case where the transport current flows along the $\mathbf{b}$ axis, i.e., along the chains, and the magnetic field $\mathbf{B}$ is applied parallel to $\mathbf{c}$. In this configuration the Hall resistivity reverses its sign twice, while for the current direction along the $\mathbf{a}$ axis and the magnetic field along $\mathbf{c}$ the Hall effect shows a pronounced positive peak. We will show that this very peculiar behavior can be interpreted in terms of anisotropic temperature and field dependences of flux-line motion and different signs of the Hall contributions from the planes and the chains.

From the Hall and the resistivity data the anisotropy of the Hall angle is computed. For $\mathbf{I}\|\mathbf{b}$, $\cot\theta_H^b$, i.e., $\gamma_{\rm trv}^b$ follows a $T\exp(-\Delta/kT)$ law between 180 and 300 K which we associate with spin fluctuations. For $\mathbf{I}\|\mathbf{a}$, $\gamma_{\rm trv}^a$ displays three approximately linear sections with changes of the slope at 120 and 220 K. The low-temperature kink is associated to the opening of a gap and a consecutive Bose condensation of the quasiparticles. It modifies the extrapolation to zero damping from $T=0$ to $T\cong 55$ K.

The growth of 1:2:4 single crystals by a high-pressure flux method has been described by Karpinski *et al.*[18] After an electron dispersive analysis by x ray (EDAX) control of the composition, about a dozen crystals from different batches with dimensions along the $\mathbf{a}$ and $\mathbf{b}$ axes ranging from 0.4 to 1 mm were selected for the transport measurements. The crystals were mechanically thinned to reach a thickness along the $\mathbf{c}$ axis between 0.02 and 0.04 mm. Four gold contacts were evaporated through a mask produced by a photolithographic technique either near the corners of the $\mathbf{ab}$ surface (resistivity setup)[19] or in the middle of the $\mathbf{a}$ and $\mathbf{b}$ edges (Hall setup). After annealing, 25 μm thick gold wires were either ultrasonically bonded or attached with gold paste. Two of the three investigated Hall samples showed very similar values in the normal state, while a third crystal gave a 50% higher value. We will present the Hall data for one of the crystals of the first group for which we have, after replacing the Hall contacts with resistivity contacts, also performed resistivity measurements in zero field.

Compared to earlier measurements[20] we find an overall reduction of the resistivity by about a factor of 2, i.e., $\rho_a(300\ \mathrm{K}) \simeq 490\ \mu\Omega\,\mathrm{cm}$, $\rho_a(75\ \mathrm{K}) \simeq 90\ \mu\Omega\,\mathrm{cm}$, $\rho_b(300\ \mathrm{K}) \simeq 145\ \mu\Omega\,\mathrm{cm}$, and $\rho_b(75\ \mathrm{K}) \simeq 27\ \mu\Omega\,\mathrm{cm}$, indicating the greater perfection of the present crystal. The anisotropy of the resistivity is about 20% larger than before, in agreement with out previous identification of mirioriented parts in the crystal.[20] $T_c^{\rm onset}$ derived from the $\rho(T)$ data for the present sample is 74 K.

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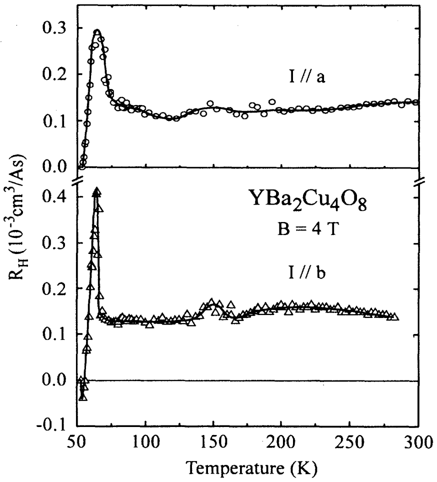

FIG. 1. Temperature dependence of the Hall coefficient of $YBa_2Cu_4O_8$ in a field of 4 T parallel to the **c** axis and for the current flowing parallel to either the **a** axis (top) or to the **b** axis (bottom). The lines are guides to the eye.

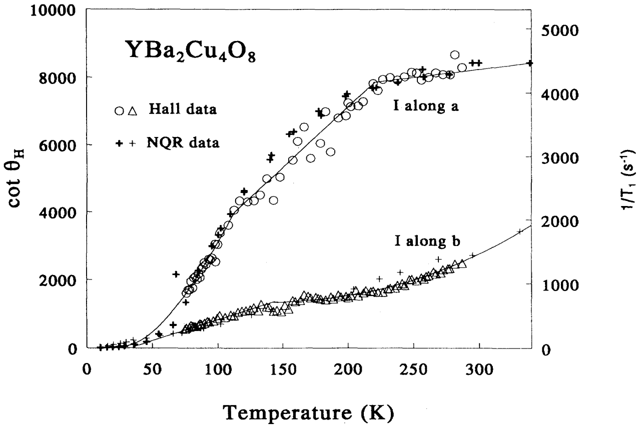

FIG. 2. Temperature dependence of cotθ_H in the normal state of YBa_2Cu_4 ($B=4$ T) for the current flowing along the **a** axis and the **b** axis (closed symbols) and comparison with NQR relaxation rates determined by Zimmermann *et al.* (Ref. 26) for Cu in the planes and the chains (crosses), respectively. The lines are fits to cotθ_H.

Figure 1 displays the temperature dependence of the Hall coefficient for **B**||**c** and the transport current flowing parallel to either the **a** axis (top) or the **b** axis (bottom). Except for some fine structure, we observe in the normal state the same temperature-independent value of $(0.14\pm0.02)10^{-3}$ cm^3/A s for both current directions. This temperature independence was found in all three samples, including the one which had $R_H=0.22\times10^{-3}$ cm^3/A s. Temperature independence of the Hall effect in the normal state is one of the characteristics of 1:2:4 as has been reported in all but one of the previous studies. Thus Char *et al.*[10] found a temperature-independent value of 0.26×10^{-3} cm^3/A s for **c**-axis oriented $YBa_2Cu_4O_8$ films. Affronte *et al.*[5] reported a temperature-independent value of 0.15×10^{-3} cm^3/A s for $YBa_2Cu_4O_8$ ceramics and van Woerden *et al.*[21] found a temperature-independent value of $\cong0.1\times10^{-3}$ cm^3/A s for polycrystalline $ErBa_2Cu_4O_8$. The only exception to this temperature independence has been claimed recently by Bucher *et al.*[22] These authors reported a decrease of the Hall coefficient with increasing temperature, which is very reminiscent in shape and absolute value of the curve reported by Char *et al.*[10] for $Y_2Ba_4Cu_7O_{15}$, which is known to form under quite similar external conditions than $YBa_2Cu_4O_8$.[18]

The simplest meaningful model to discuss the Hall effect of 1:2:4 is not a one-, but a two-band model. Due to the approximately one-dimensional character of the chain bands, these bands contribute less to the Hall effect but they dominate the diagonal conductivity. If the current flows along the **b** axis more than $\frac{2}{3}$ of it is carried by the chains and, in first approximation, will not contribute to R_H. Thus R_H is reduced by a factor of $\simeq3$. A more elaborate estimate of the Hall effect in $YBa_2Cu_4O_8$ has been given by Massida *et al.*[23] These authors have calculated the Hall coefficient of 1:2:4 by solving the Boltzmann equation using the band structure obtained with the full potential linearized augmented plane wave method within the local density approximation. For **B**||**c**, Massida *et al.*[23] obtain $R_H=0.21\times10^{-3}$ cm^3/A s which is comparable to our experimental values.

In the normal state $\omega_c\tau\ll1$ and the Hall data can give information on the anisotropy of the scattering mechanism. The tangent of the Hall angle is defined as the quotient of the Hall field E_H and the longitudinal field E_L. If the transport current flows along the **a** axis, we have $\tan\theta_H^a=E_H/E_L=\sigma R_H B=\sigma_{xx}\rho_{xy}$. From $\underline{\rho}\,\underline{\sigma}=\underline{1}$ we obtain $\rho_{xy}=-\sigma_{xy}/(\sigma_{xx}\sigma_{yy}-\sigma_{xy}\sigma_{yx})$ and $\sigma_{xx}=\rho_{yy}/(\rho_{xx}\rho_{yy}-\rho_{yx}\rho_{xy})$. Because in the normal state of 1:2:4 the off-diagonal resistivities and conductivities are two orders of magnitude smaller than the diagonal resistivities and conductivities, respectively, $\cot\theta_H^a=(\sigma_{xx}\rho_{xy})^{-1}\cong\rho_{xx}/\rho_{xy}$ and we have plotted in Fig. 2, ρ_{xx}/ρ_H(**I**||**a**) and ρ_{yy}/ρ_H(**I**||**b**). The differences between these two curves are remarkable and they also differ from all Hall-damping curves published so far for 1:2:3 (Refs. 6 and 7) which shows a T^2 dependence for $T\gg T_c$ or 1:2:4 (Ref. 22), which was claimed to also show a T^2 dependence. In contrast, for **I**||**a** and $T>75$ K the overall temperature dependence in Fig. 2 is sublinear. The remarkable similarity of $\cot\theta_H^a(T)$ with the relaxation rate observed by Zimmermann *et al.*[24] in a NQR experiment on ^{63}Cu(2) in the plane of $YBa_2Cu_4O_8$ (shown as crosses in Fig. 2) indicates that, in contrast to the longitudinal scattering probed by the resistivity, the transverse scattering is dominated, like NQR, by magnetic spin scattering. It also indicates that the transverse scattering within the **ab** plane is a high-momentum scattering. The strong decrease of the magnetic scattering with decreasing temperature in the ^{63}Cu(2) nuclear quadrupole resonance (NQR) data has been associated with the formation of a spin or a pseudospin gap,[25] first evoked by Rice.[26] While the overall temperature dependence of $\cot\theta_H^a$ corroborates this view, a closer look at the data leads us to believe that the curve consists of at least two linear sections separated by a transition region. The section between 75 and 120 K extrapolates to zero at 55 K. Above 230 K the slope of $\cot\theta_H^a$ is about 16 times smaller than between 75 and 120 K. In the transition region the linear approximation extrapolates to zero at 0 K. The decrease of the damping at lower temperatures below this extrapolation indicates a freezing of scattering channels. A fit of $\cot\theta_H^a$ in Fig. 2 with the expression $\cot\theta_H^a = \alpha T \exp(-T^*/T)$ indicates a gap temperature $T^* = (120 \pm 10)$ K.[27] In the resonating-valence-bond-state model of Nagaosa and Lee for underdoped high-temperature superconductors[28] the kink near 120 K would correspond to the spin-gap opening (spinon pairing), which depending on whether inelastic lifetime effects are included or not, coincides or occurs at higher temperature than the Bose condensation of the quasiparticles (holons). Above the spin-gap temperature the model of Nagaosa and Lee predicts a strange metal, in agreement with $\gamma_{\mathrm{trv}}^a \sim T$ up to 220 K. The decrease of the slope above this temperature may indicate a localization of the magnetic moments of the quasiparticles.

For $\mathbf{I} \| \mathbf{b}$ the Hall damping shows a linear behavior below 130 K extrapolating to zero damping at 32 K and a superlinear increase above 180 K. The agreement with $1/T_1$ from NQR is again remarkable (note the identical scales for both Cu sites), but our data show clearly deviations from linearity. To fit the superlinear part we have used the expression $\cot\theta_H^b = 1300 + 83T \exp(-850/T)$. We associate $T^* = 850$ K with the antiferromagnetic exchange coupling. Although the quasi-one-dimensional chains will not order, antiferromagnetic fluctuations are to be expected. Below about 120 K the chains couple more effectively with the planes and both dampings become approximately linear functions of T, until at T_c the damping drops to zero. Thus our measurements indicate that the chains become superconducting in 1:2:4 through the coupling with the planes.

In the last part of this paper we like to discuss the mixed state of 1:2:4. Coming back to Fig. 1, we find in a field of 4 T with decreasing temperature a strong rise of the Hall coefficient below $T_c(0)$ with a maximum near 64 K. The onset of the rise and the value of the maximum depend on the current direction. On lowering further the temperature, R_H goes directly to zero for $\mathbf{I} \| \mathbf{a}$, while for $\mathbf{I} \| \mathbf{b}$ the Hall coefficient makes an excursion to negative values before also becoming zero.

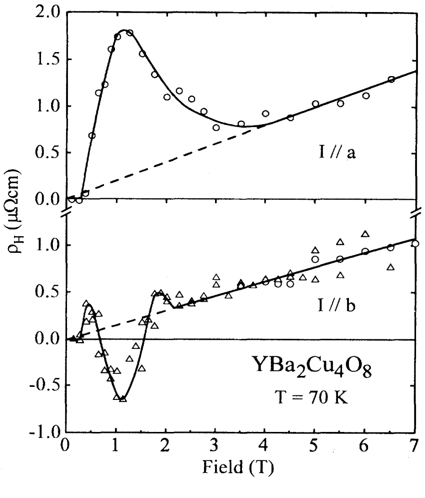

FIG. 3. Field dependence of the Hall resistivity at 70 K for the current flowing either along the **a** axis (top) or the **b** axis (bottom). The lines are guides to the eye.

Figure 3 displays the Hall resistivity ρ_H at 70 K as function of field. At high fields, ρ_H is proportional to the external field and the extrapolations to zero field give $\rho_H = 0$. However, below 3–4 T, corresponding to B_{c2}, we find remarkable deviations from linearity and an extreme anisotropy. For $\mathbf{I} \| \mathbf{a}$, ρ_H reaches at $B = 1.1$ T a maximum value of 1.8 $\mu\Omega$ cm, which is 8.5 times larger than the linear extrapolation would indicate. For $\mathbf{I} \| \mathbf{b}$ the field dependence is even more complex. With decreasing field, ρ_H increases below B_{c2} to peak near 1.8 T. It then decreases strongly, reverses its sign at 1.5 T, and has a negative extremum near 1.1 T. Then ρ_H becomes positive again at 0.7 T, makes a second positive peak at 0.4 T, and, finally, vanishes at 0.25 T. As mentioned in the Introduction sign changes are a quite common property in high-T_c superconductors, but to our knowledge, such a complex and anistropic behavior as shown in Fig. 3 has never been observed before. The classical theories to intepret anomalies of the Hall effect in the mixed state start from the flux-flow model, which has been successful in describing the longitudinal resistivity. However, neither the local hard-core model of Bardeen and Stephen[29] nor the soft-core model of Nozières and Vinen[30] predict a sign change, which would require that the flux lines flow against the transport current direction. To circumvent this shortage several modifications and new models have been proposed. The majority of them can be divided into two classes. In a first class the drag force of the vortices is altered.[11,16,17,31] The second class of models is the two-band models, which assume that negative and positive carriers contribute with different temperature and field dependences.[12,32,33] The anisotropy depicted in Figs. 1 and 3 and particularly the fact that for $\mathbf{I} \| \mathbf{b}$ the Hall resistivity shows the same positive increase at the transition from the flux-creep to the flux-flow regime than for $\mathbf{I} \| \mathbf{a}$ and again a positive excursion near the transition from the flux-flow to the normal state bears witness that the negative Hall resistivity in between the positive maxima is an effect directly related to the Hall effect of the chains. A negative Hall contribution of the chains in the normal state has been predicted by the theory of Massida *et al.*[23] who found for one of the chain bands $\hbar\sigma_{xyz}/(e^3\tau^2) = -0.27 \times 10^{-3}$ a.u. and for the other $+0.18 \times 10^{-3}$ a.u., while the corresponding values for the plane bands are $+1.49$ and $+1.22 \times 10^{-3}$ a.u. Thus, if the flux-flow-induced increase of the negative Hall resistivity of the chain band is larger than that of the other bands and occurs in a narrower field range we understand the complex behavior depicted in Fig. 3. The first prerequisite is likely to be fulfilled as the mobility in the chains is larger than in the planes and it tends faster to infinity in the superconducting phase. The second prerequisite is fulfilled as checked by resistivity measurements in fields on another sample of $YBa_2Cu_4O_8$.

In conclusion we have presented evidence from Hall data in the normal state for the formation of a spin gap in an underdoped high-T_c superconductor and Bose condensation of the quasiparticles as a mechanism of superconductivity. The first observation of a strong anisotropy of the Hall resistivity in the mixed state with two sign reversals for **I** flowing along the chains and no sign change for **I** flowing in the planes indicates that at least two bands with opposite carrier signs contribute to the sign reversal.

Note added in proof. In two recent publications Samoilov[34] and Vinokur [35] have shown in experiments on $Bi_2Sr_2CaCu_2O_x$ and in a scaling theory, respectively, that $\rho_{xy} \sim \alpha \rho_{xx}^2$. The coefficient α relies on the microscopic model and determines the sign of ρ_{xy}. In the temperature and field regime where thermally activated flux flow dominates, α is nearly temperature independent and ρ_{xy} should be proportional to ρ_{xx}^2. This regime corresponds to the temperature range from 64 to 55 K in Fig. 1, where R_H drops rapidly with decreasing temperature.

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