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Limits on deviations from Onsager-Casimir symmetry in the resistance of $YBa_2Cu_3O_{7-\delta}$

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The normal-state resistance $R_{12,34}$ of a $YBa_2Cu_3O_{7-\delta}$ thin film (measured with current contacts 1,2 and voltage contacts 3,4) is found to obey the symmetry relation $R_{12,34}=R_{34,12}$ to within the experimental resolution of a few parts in 10^4. This result sets a limit on the anomalous zero-field Hall effect, caused by spontaneously broken time-reversal symmetry above T_c, which has been proposed in connection with anyon models for high-T_c superconductivity.

Time-reversal symmetry (TRS) constrains the resistivity $\vec{\rho}$ to be a symmetrical tensor,

$$\rho_{\alpha\beta}=\rho_{\beta\alpha}\,. \tag{1}$$

This is an example of an Onsager-Casimir symmetry relation.[1] It has recently been suggested[2-4] that in high-T_c superconductors, TRS is broken spontaneously—in the absence of an applied magnetic field. The violation of time-reversal symmetry and two-dimensional reflectional symmetry (parity) is a consequence of the fractional statistics of the quasiparticles ("anyons") in Laughlin's model for high-T_c superconductivity.[5,6] The temperature T_{tp} below which these symmetries are broken is expected to coincide or to be larger than the critical temperature T_c for superconductivity.[3] A violation of TRS would lead to the appearance of an asymmetric contribution to $\vec{\rho}$—at least if TRS is broken macroscopically (which requires that adjacent CuO_2 layers break the symmetry in the same way). Such an experimental test was suggested in Refs. 3 and 4, and is the subject of the present paper. Experimental investigations on TRS violation in equilibrium properties have been reported by Kiefl *et al.*[7] and by Lyons *et al.*,[8] as discussed towards the end of this paper.

A current-voltage measurement yields a resistance rather than a resistivity. The four-terminal resistance $R_{12,34}\equiv V/I$ refers to a measurement configuration in which the current I flows from contact 1 to contact 2, and the voltage V is measured between contacts 3 and 4. The symmetry of $\vec{\rho}$ implies[1,9]

$$R_{12,34}=R_{34,12}\,, \tag{2}$$

that is to say, the resistance is invariant under interchange of the pairs of current and voltage contacts. The reciprocity relation (2) is a more general consequence of time-reversal symmetry than Eq. (1), as it does not require the existence of a local resistivity tensor.[10] Moreover, even if a local $\vec{\rho}$ exists, Eq. (2) is a more sensitive test for violations of TRS than Eq. (1), since to extract the components of the resistivity tensor from resistances requires precise alignment of the current and voltage contacts and preferably a truly homogeneous sample. For these reasons the reciprocity relation (2) forms the basis for our search for deviations from Onsager-Casimir symmetry.

We report on precise resistance measurements of a thin $YBa_2Cu_3O_{7-\delta}$ film in the 90–160 K temperature range. The film was deposited at the University of Twente by pulsed laser ablation. An excimer laser in the Xe-Cl mode was used (wavelength 308 nm) with a pulse duration of 20 ns. The energy density of the focused beam on the target was 2 J/cm^2 and the repetition frequency was 2 Hz. The $YBa_2Cu_3O_{7-\delta}$ target, at the correct stoichiometry, was prepared by the citrate pyrolisis method. The $SrTiO_3$ (100) substrate was heated up to 720 °C by a thermo-coax heater. The ablation time was 15 min in an oxygen pressure of 30 Pa, resulting in a c-axis-oriented film of 100-nm thickness. Afterwards the film was cooled down to room temperature in an oxygen atmosphere within 1 h. Contacts were made on the film by evaporating four 99.9999%-pure Au spots, on which Cu wires were attached using In spheres. The four contacts were in a rectangular arrangement, a few mm apart (see inset to Fig. 1). The film was mounted on a sample holder and put into a ^{4}He flow cryostat. Temperature was monitored with a carbon-glass resistor, and temperature stabilization was typically better than 0.05 K. Resistance measurements were done using an ac technique with a current amplitude down to 1 μA, giving rise to a power dissipation as low as 10^{-11} W. ac voltages were measured using two PAR 113 low-noise preamplifiers, the output of which was fed into two lock-in detectors (PAR 5204 and 5209). One of these detected the amplified voltage over a calibrated resistor and the other one the voltage drop over the sample. The

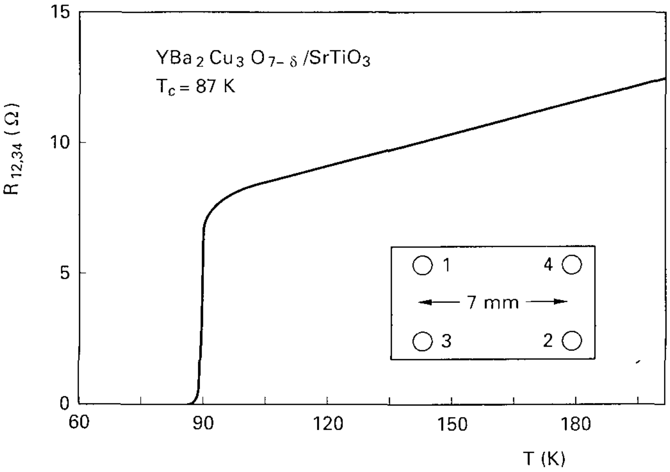

FIG. 1. Temperature dependence of the resistance $R_{12,34}$ of the $YBa_2Cu_3O_{7-\delta}$ film studied. The inset shows the measurement configuration.

output of the lock-in detectors was connected to a HP 2345 digital voltmeter. After sampling and averaging, film resistance could be determined with a total experimental resolution of 0.02%, a precision mainly determined by temperature fluctuations (see further). To maximize contributions to the resistance from off-diagonal components of $\overleftrightarrow{\rho}$, we measured the four-terminal resistance in a configuration where the line between the current contacts crosses the line between the voltage contacts. This corresponds to the $R_{12,34}$ and $R_{34,12}$ measurement (see inset of Fig. 1).

Figure 1 shows the temperature dependence of the resistance $R_{12,34}$. The zero-resistance critical temperature $T_c = 87$ K. Above 100 K the resistance is only weakly temperature dependent ($dR/dT \simeq 0.039$ Ω/K). On approaching T_c, the temperature variation of the resistance is much stronger. The uncertainty in the temperature of 0.05 K leads to a potential error in the resistance measurement of 0.039 Ω/K×0.05 K≃2 mΩ above 100 K. Hence temperature fluctuation is the main factor determining the observed experimental resolution. Below 100 K the experimental error becomes much larger due to the stronger temperature dependence of the resistance.

In Figs. 2 and 3 we show the difference in resistance $R_{12,34} - R_{34,12}$ on interchanging current and voltage contacts, as a function of current and temperature. We have plotted the absolute rather than the relative values of $R_{12,34} - R_{34,12}$ because we do not expect the zero-field Hall resistance to scale with the diagonal resistance, by analogy with the conventional Hall effect. The error bars in Fig. 3 are omitted for clarity, but are of the same magnitude as in Fig. 2. Only for the data points at 93 K is the uncertainty much larger (due to the larger value of dR/dT). Since the Onsager-Casimir symmetry relations are valid only in the regime of small currents and voltages, it is of importance to use the lowest possible currents. We found a small current dependence of the resistance down to the lowest currents of 1 μA. Typically, $dR/dI \simeq 2$ mΩ/μA around 100 K. As shown in Fig. 2 the reciprocity relation is obeyed within the experimental resolution for for currents below 4 μA. Deviations from reciprocity at higher currents can be attributed to joule heating and other nonlinear effects. In Fig. 3 the resistance difference is shown over the whole temperature range for three low values of the current. We find no systematic temperature dependence of $R_{12,34} - R_{34,12}$ above 100 K. The large scatter of the points at $T = 93$ K, on approaching T_c, impedes a reliable estimate for the resistance difference but is still consistent with a null result.

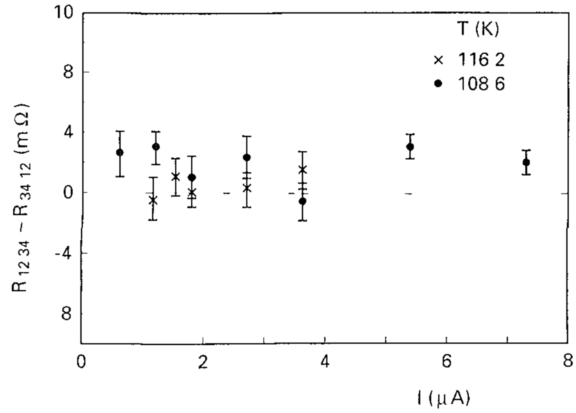

FIG. 2. Current dependence of the difference in the resistance on interchanging current and voltage contacts, at two different temperatures.

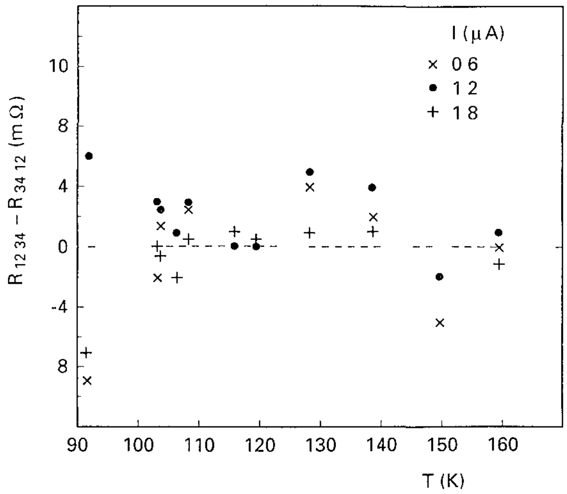

FIG. 3. Resistance difference as a function of temperature, for three values of the current.

As we discussed in the introduction, reciprocity of the resistance is a more general consequence of time-reversal symmetry than symmetry of the resistivity tensor. Therefore we do not measure the individual components of $\overleftrightarrow{\rho}$. Nevertheless, approximately one has $R_{12,34} - R_{34,12} \simeq (\rho_{xy} - \rho_{yx})/t$, where t is the film thickness and x and y are the coordinates in the film plane. Given the estimated experimental uncertainty of 2 mΩ in $R_{12,34} - R_{34,12}$ and the film thickness $t = 100$ nm, we can deduce an upper bound for the zero-field Hall resistivity

$$\rho_0 \equiv \tfrac{1}{2}(\rho_{xy} - \rho_{yx}) \simeq \tfrac{1}{2}t(R_{12,34} - R_{34,12}) < 1\times10^{-10}\ \Omega\,\mathrm{m}\,. \tag{3}$$

This value is equivalent to an upper bound of about 0.1 Ω for the zero-field Hall resistance of a CuO_2 layer—under the assumption that adjacent layers couple ferromagnetically with respect to the time-reversal symmetry breaking. The effect per layer could be much larger in the case of antiferromagnetic coupling: in that case TRS is broken within each layer but not macroscopically.

Less extensive reciprocity measurements on another $YBa_2Cu_3O_{7-\delta}$ film prepared in a different way[11] are consistent with the results reported here.

We do not know of a reliable estimate of the asymmetric component of the resistivity tensor within the anyon model. Chen *et al.*[4] have calculated the electrostatic potential perpendicular to the current due to vortices in the superconducting phase of the anyon gas. They estimate 0.03 volts per ampere of supercurrent in a single layer. This electrostatic potential is canceled by the chemical potential so that zero resistance results, as it should in a superconductor. Their result is therefore not directly applicable to the present experiment Impurity scattering causing TRS violation will presumably play an important role above T_c Aronov and Shelankov[12] have incorporated such a term in the Ginzburg-Landau equations, but did not calculate the normal-state properties

In conclusion we find no evidence in the reciprocity of the resistance for macroscopically broken time-reversal symmetry above T_c This finding is, on the one hand, consistent with the negative result of the search by Kiefl *et al* [7] for an anomalous internal magnetic field in high-T_c superconductors Lyons *et al*,[8] on the other hand, have observed circular dichroism above T_c in reflection from $YBa_2Cu_3O_{7-\delta}$ films (and from other high-T_c superconductors), indicative of macroscopically broken TRS invariance with onset temperature $T_{tp} \gtrsim 150$ K Two more recent searches for broken time reversal symmetry, by Spielman *et al* [13] and Weber *et al*,[14] in optical circular effects have yielded respectively a negative[13] and a positive result [14]

We see two ways to reconcile our negative result with the positive results of Lyons *et al* and Weber *et al* The first would be that the effect of TRS breaking on the reciprocity of the resistance is too small for the sensitivity of our experiments The second, more interesting, way would be that the circular dichroism is due not to TRS breaking but to the helicity of the material (a possibility suggested by Lyons *et al*) The resulting breaking of reflection symmetry would have no effect on the Onsager-Casimir symmetry, which holds regardless of any spatial symmetry

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