Test of the Wiedemann-Franz law in an optimally-doped cuprate

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We present a study of heat and charge transport in Bi\textsubscript{2+x}Sr\textsubscript{2−x}CuO\textsubscript{6+δ} focused on the size of the low-temperature linear term of the thermal conductivity at optimal doping level. In the superconducting state, the magnitude of this term implies a d-wave gap with an amplitude close to what has been reported. In the normal state, recovered by the application of a magnetic field, measurement of this term and residual resistivity yields a Lorenz number \( L = \kappa \rho_0 / T = 1.3 \pm 0.2 L_0 \). The departure from the value expected by the Wiedemann-Franz law is thus slightly larger than our estimated experimental resolution.

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In spite of many years of intense research by a sizeable fraction of the condensed-matter physics community, high \( T_c \) superconductivity remains a mystery. A central question is the extent of the validity of Landau’s Fermi liquid picture to describe the elementary excitations of the ground state. High-\( T_c \) cuprates are doped Mott insulators and are host to a particularly strong Coulomb repulsion neglected in the Fermi liquid picture. It remains to be established, however, to what extent this becomes an obstacle for the formation of Landau quasi-particles in the \( T=0 \) limit. A recent attempt to answer this question has been made by measuring the subkelvin thermal conductivity of the normal state in order to check for the validity of the Wiedemann-Franz law which is a robust signature of a Fermi liquid.\textsuperscript{1-3} The validity of this universal law relating the magnitude of thermal and electrical conductivities is expected in the \( T=0 \) limit, disregarding the fine details of electronic scattering and Fermi surface. On the other hand, various scenarios based on electron fractionalization lead to its violation.

In this paper, we present a first experimental study to check the validity of the Wiedemann-Franz(WF) law in a hole-doped cuprate at optimal-doping concentration. Recovering the normal state of Bi\textsubscript{2+x}Sr\textsubscript{2−x}CuO\textsubscript{6+δ} (Bi-2201) by the application of a strong magnetic field, we found a thermal conductivity slightly larger than what is expected by the WF law. The departure appears to be genuine as it is significantly larger than the experimental precision obtained in the verification of the WF law in a simple metal.

In-plane thermal conductivity (\( \kappa \)) in presence of a magnetic field applied along the c-axis was measured with a standard two-thermometers-one-heater set-up which permitted to measure the in-plane electric resistivity in the same conditions. Bi-2201 single crystals, with typical dimensions of (2-8) \( \times \) (400-800) \( \times \) (600-900) \( \mu \)m\textsuperscript{3}, were grown in a gaseous phase in closed cavities of a KCl solution melt as detailed elsewhere.\textsuperscript{4} Doping level in stoichiometric Bi\textsubscript{2}Sr\textsubscript{2}CuO\textsubscript{6+δ} can be modified by replacing Sr\textsuperscript{2+} either with Bi\textsuperscript{3+} or with La\textsuperscript{3+}.\textsuperscript{5,6} We succeeded to make single-phase high quality crystals of Bi\textsubscript{2+x}Sr\textsubscript{2−x}CuO\textsubscript{6+δ} in the range of 0.17\(<x<0.7. The maximum [resistive] \( T_c \) found in this system was \( \sim 10.5 \) K in agreement with previous studies.\textsuperscript{1,7} In Bi\textsubscript{2}Sr\textsubscript{2−x}La\textsubscript{x}CuO\textsubscript{6+δ}, on the other hand, the maximum \( T_c \) is 38K.\textsuperscript{8} Thus, the disorder associated with (Sr,Bi) substitution is apparently stronger than the one due to (Sr,La) doping and leads to a lower \( T_c \) and a higher residual resistivity in Bi\textsubscript{2+x}Sr\textsubscript{2−x}CuO\textsubscript{6+δ} compared to Bi\textsubscript{2}Sr\textsubscript{2−x}La\textsubscript{x}CuO\textsubscript{6+δ}.\textsuperscript{9}

An unambiguous determination of carrier concentration, \( p \), in high-\( T_c \) cuprates other than La\textsubscript{2−x}Sr\textsubscript{x}CuO\textsubscript{4}(La-214) is not straightforward. However, by comparing Hall coefficients of various families of cuprates, Ando \textit{et al.}\textsuperscript{8} have shown that the magnitude of renormalised Hall coefficient(\( R_H e / V_0 \), where \( V_0 \) is the volume associated with each Cu atom) is an appropriate measure of carrier concentration. In order to check...
the doping level of our samples we measured the Hall coefficient in a number of them and found that the magnitude of $R_{H}V_{0}/r_{0}$ is close to what is expected for an optimal doping cuprate. As seen in Fig. 1 which presents a typical curve, the magnitude of renormalised Hall coefficient is somewhere between the values obtained for La-214 at $p=0.15$ and $p=0.18$. A similar result has been recently reported by Ono and Ando. Together with the linear resistivity observed from room temperature down to $T_{c}$ (see the inset of Fig.1), this provides compelling evidence that the physics of cuprates at optimal doping level can be explored in Bi-2201, which contrary to other hole-doped cuprates, presents a resistive upper critical field within the range of available DC magnetic fields.

As the required field ($\sim 25$ T), however, still exceeds the range provided by available commercial magnets, the experiment was performed in a resistive magnet at Grenoble High Magnetic Field Laboratory. Measuring subkelvin thermal conductivity in this context faced two major technical challenges. The first was to cool down the sample and thermometers held in vacuum to low temperatures in spite of strong mechanical vibrations associated with the circulation of cooling water. This problem was partially solved by designing a new insert with a vacuum chamber containing the entire thermal conductivity set-up which was placed inside the mixing chamber of a Kelvinox top-loading dilution fridge. The second was to measure accurately the temperature at high magnetic fields given the non-negligible magneto-resistance of the RuO$_2$ thermometers used in the set-up and the absence of any zero-field zone. This second problem was resolved by using Coulomb Blockade Thermometry (CBT). An array of 100 tunnel junctions provided by Nanoway (Finland) was attached to the cold finger and used as a field-independent thermometer for calibrating the shift in the resistance of RuO$_2$ thermometers with the application of magnetic field.

Zero magnetic field: Fig. 2(a) displays the temperature dependence of thermal conductivity for three crystals of Bi-2201 at zero magnetic field. Since the lattice thermal conductivity is expected to display a cubic temperature dependence at very low temperatures, ($\kappa/T$) is plotted as a function of $T^2$ in order to extract a linear term, $\kappa_0/T$, associated with the electronic contribution. The magnitude of this linear term varies from 0.19 in the most underdoped sample ($p=0.14$) to 0.40 mW/K$^2$cm in the most overdoped sample ($p=0.19$) (See fig. 2(c)). Doping level in these samples was estimated using the reported empirical relationship between $T_c$ and carrier concentration in Bi-2201. Fig. 2(b) presents the temperature dependence of resistivity in the three samples. c) A comparison between the doping dependence of $\kappa_0/T$ (open circles) and of $L_0/\rho_0$ (solid circles). Lines are guides for eye.

| Compound | $T_c$ (K) | $\kappa_0/T$ ($\sigma$) | $\Delta_0$ (meV) | $\Delta_{narrow}$ (meV) |
|----------|-----------|------------------------|------------------|------------------------|
| Bi-2212  | 89        | 0.15                   | 34               | 30                     |
| Bi-2201  | 11        | 0.33                   | 35               | 16                     |

TABLE I: A comparison of two Bi-based cuprates at optimal doping level. $\kappa_0/T$ is expressed in mW/K$^2$cm units. $\Delta_0$ was calculated using $v_F=250$ km/s and $k_F=0.7A^{-1}$ obtained in ARPES studies on Bi-2212.

FIG. 2: a) Subkelvin thermal conductivity in three different Bi-2201 single crystals. b) Temperature dependence of the zero-field resistivity in the three samples. c) A comparison between the doping dependence of $\kappa_0/T$ (open circles) and of $L_0/\rho_0$ (solid circles). Lines are guides for eye.
purity concentration up to the highest scattering rates investigated for optimally-doped samples. On the other hand, according to recent studies on La-214 and on Y-123, $\kappa_0/T$ is a monotonously increasing function of doping concentration.

Fig. 2(c) confirms the latter behavior in the case of Bi-2201. In the limited range of our exploration, the magnitude of $\kappa_0/T$ increases with the increase in doping level. This allows us to determine the magnitude of $\kappa_0/T$ in Bi-2201 at optimal doping level ($p=0.17$) to be 0.33 mW/K cm. This value is more than twice the magnitude reported for Bi-2212, which has a substantially higher $T_c$. This is not surprising, since $\kappa_0/T$ is a zero-energy probe inversely proportional to the superconducting gap. Now, it is tempting to forget that Bi-2201 is not in the clean limit (as defined above) and compute $v_F/v_2=35$, using the procedure already used for other cuprates. Since $v_2$ is proportional to the angular slope of the gap at a nodal position, this number can be related to the magnitude of the superconducting gap assuming a regular $d$-wave angular dependence ($\Delta = \Delta_0 \cos 2\phi$). This yields $\Delta_0 = \hbar v_F v_2/2 = 16$ meV comparable with the magnitude reported by tunnelling studies.

Now, we turn our attention to the isomeric state: $L = \kappa_N\rho_0/T$, should be equal to Sommerfeld’s value:

$$L_0 = \frac{\pi^2 k_B^2}{3e^2} = 24.4 \times 10^{-9} V^2/K^2 \quad (1)$$

Here $\kappa_N/T$ and $\rho_0$ are thermal conductivity and electric resistivity of the normal state in the $T=0$ limit. Before presenting the data on heat conductivity in the normal state, let us note that in our samples the residual thermal conductivity in the superconducting state exceeds the magnitude of the thermal conductivity in the normal state expected by the magnitude of the resistivity (either $\rho(T_c)$ or $\rho_0$) and according to the WF law. In other words, $\kappa_0/T > L_0/\rho_0$. This surprising inequality was first reported in the case of underdoped La-214.

In Bi-2201, as seen in Fig. 2(c), it persists at optimal doping, but gradually disappears with increasing $p$. In this context, a field-induced enhancement of $\kappa_0/T$, like the one observed in optimally-doped Bi-2212, would lead to a strong violation of the WF law. The application of a magnetic field, strong enough to destroy any trace of superconductivity in charge transport, has little effect on low-temperature thermal conductivity of Bi-2201. In the slightly overdoped sample ($p=0.19$), the magnetic field leads to a slight increase in the residual linear term. No change in $\kappa/T$ is observed for $H=15$ T and $H=20$ T. The few available data points for $H=25$ T confirms that a reliable linear term for this field can be extracted from the data obtained at lower fields. Assuming that the magnetic field does not affect the slope of $\kappa/T$ in the ballistic regime, we estimate $\kappa_N/T = 0.46\pm0.04$ mW/K cm by extrapolating the $H=20$ T data to $T=0$ along a line parallel to the $H=0$ slope. In the case of the optimally-doped sample ($p=0.17$), a magnetic field of 25 T leaves the thermal conductivity virtually unchanged in the explored temperature range. Thus, the zero-field extrapolation for this sample ($\kappa_0/T = 0.32\pm0.03$ mW/K cm) may be considered as the magnitude of $\kappa_N/T$. As seen in the figure, for both samples, $\kappa_N/T$ exceeds $L_0/\rho_0$. The excess is about 20(30) percent for the $p=0.19(0.17)$ sample.

In order to estimate our resolution in the verification of the WF law in a simple metal, we used the same set-up to measure the low-temperature thermal and electric conductivities of a 17 µm-diameter gold wire at $H=0$ and $H=25$ T. The application of the magnetic field led to a three-fold increase in $\rho_0$ and a concomitant decrease in $\kappa/T$ of the gold wire. Using Coulomb blockade thermometry, we succeeded in verifying the WF law with a precision of 1(3) percent at $H=0$ (25 T). Thus, the deviation observed in Bi-2201 is much larger than our experimental precision on the absolute magnitude of $\kappa/T$. Besides absolute thermometry, other sources of error may arise in the case of sub-millimetric and anisotropic samples. The geometric factor, assumed identical for $\kappa$ and $\rho$ in our analysis, may be different for electric and thermal transport. Since we did not observe any correlation between the magnitude of thermal conductivity and the width of gold electrodes, we can exclude the finite width of the contacts as a significant source of discrepancy. An eventual $c$-axis contamination of the presumably in-plane conductivity would lead to an overestimation of $\rho_0$. Moreover, as the electric transport is much more anisotropic than heat transport, the measured $\kappa/T$ would be less affected by such a contamination. We estimate that in the case of $p=0.17$ sample (with a thickness of 2.5µm), this can lead to an error of 6 percent in the absolute magnitude of residual resistivity.

The sum of the three identified sources of experimental error (extrapolation to zero temperature, absolute thermometry and overestimation of residual resistivity due to a $c$-axis contamination) yield an uncertainty of 17 percent. We can therefore conclude that for the $p=0.17$ sample, the Lorenz number exceeds Sommerfeld’s value by a slight yet significant margin, that is $L = 1.3 \pm 0.2 L_0$.

Prior to this work, compelling evidence for the validity of the WF law in the overdoped regime was reported for overdoped Tl-2201 ($p=0.26$) and for heavily overdoped La-214 ($p=0.30$). Together with the detection...
of a purely $T^2$ temperature-dependence of the resistivity in the latter compound, this seems to establish that the ground state in the overdoped limit is indeed a Fermi liquid. A previous study performed on an electron-doped cuprate at optimal doping level, pointed to a different conclusion\cite{1}. Note, however, that in the latter experiment the extraction of $\kappa N / T$ was complicated by the presence of an intriguing downturn in thermal conductivity below 0.3 K. For temperatures above 0.3 K, Hill et al., extracted a $\kappa N / T$ which exceeded the expected WF value by a factor of 1.7\cite{1}. Our results do not suffer from the presence of this downturn which was also observed in overdoped La-214\cite{18}. They yield a $\kappa N / T$, still larger than, but closer to, the WF value. Very recently, we have found that the slight deviation observed here is considerably enhanced with underdoping and/or disorder\cite{28}. This indicates that the slight departure from the WF law at optimal doping level is not due to an experimental imperfection.

The outcome of this study has interesting implications for the debate on the origin of the anomalous transport properties of cuprates. In theories invoking electron fractionalization in cuprates\cite{29}, the zero-temperature excitations of the normal state are not Landau quasi-particles and the WF law is not expected to be valid. In the case of a Luttinger liquid, for example, a strong violation of the WF law has been theoretically predicted\cite{51}. On the other hand, if the anomalous properties of the normal state is due to the existence of a competing hidden order, then the validity of the WF law at $T=0$ is still expected\cite{51}.

In summary, we have measured heat transport in the normal and superconducting state of Bi-2201. In the normal state, we have found a Lorenz number slightly but significantly larger than Sommerfeld’s value.

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\begin{figure}[h]
\centering
\includegraphics[width=0.8\textwidth]{fig3.png}
\caption{Upper and lower panels: The effect of magnetic field on subkelvin thermal conductivity in two Bi-2201 samples with different doping levels. Solid lines represent the extrapolated low-temperature behavior and arrows show the expected WF magnitude. The insets display the resistivity data for the two samples.}
\end{figure}

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