Editors' Suggestion

Thermal diffusivity above the Mott-Ioffe-Regel limit

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We present high-resolution thermal diffusivity measurements on several near optimally doped electron- and hole-doped cuprate systems in a temperature range that passes through the Mott-Ioffe-Regel limit, above which the quasiparticle picture fails. Our primary observations are that the inverse thermal diffusivity is linear in temperature and can be fitted to $D_Q^{-1} = aT + b$. The slope a is interpreted through the Planckian relaxation time $\tau \approx \hbar/k_B T$ and a thermal diffusion velocity v_B , which is close, but larger than the sound velocity. The intercept b represents a crossover diffusion constant that separates coherent from incoherent quasiparticles. These observations suggest that both phonons and electrons participate in the thermal transport, while reaching the Planckian limit for relaxation time.

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The standard paradigm for transport in Fermi-liquid metals relies on the existence of well-defined quasiparticles. Transport coefficients such as electrical and thermal conductivities can then be calculated using Boltzmann theory, where the electrons are treated semiclassically [1]. However, such an approach fails when the quasiparticle mean free path becomes comparable to its de Broglie wavelengths. Beyond this so-called Mott-Ioffe-Regel (MIR) limit [2], the material is dubbed a "bad metal" [3], and transport becomes "incoherent," as the notion of momentum eigenstate quasiparticles fails. A new theoretical framework is needed to describe this regime, while new experiments, complementary to the extensively studied electrical resistivity, are needed to provide an additional perspective on the problem.

In the absence of a microscopic transport theory, one may still use thermodynamics supplemented by continuity equations for the charge and energy as conserved quantities. In the absence of thermoelectric effects, this approach leads to Einstein relations for the electrical conductivity $\sigma = \chi D_e$ and the thermal conductivity $\kappa = cD_Q$, which are expected to hold regardless of the presence of quasiparticles. Here, $\chi = e^2(dn/d\mu)$ is the charge susceptibility and is proportional to the density of states, c is the specific heat, and D_e and D_Q are charge and thermal diffusivity, respectively. It is therefore interesting to explore transport in a nonquasiparticle regime by studying the diffusivities directly. Indeed, in recent studies diffusivity was singled out as a key observable for incoherent nonquasiparticle transport, possibly subject to fundamental quantum mechanical bounds [4], particularly a minimum

In this Rapid Communication we report high-resolution thermal diffusivity measurements on single crystals of nearly optimally doped electron-doped cuprates Sm_{2-x}Ce_xCuO₄ (SCCO), Nd_{2-x}Ce_xCuO₄ (NCCO), Pr_{2-x}Ce_xCuO₄ (PCCO), and near optimally doped Bi₂Sr₂CaCu₂O₈ (BSCCO) in the temperature range of 100–600 K. These materials exceed the MIR limit in resistivity [2] above ~250–300 K [7]. Unlike the hole-doped cuprates, the electron-doped cuprates do not lose oxygen upon heating and thus can be studied to relatively high temperatures. In addition, comparing "as grown" with "annealed" samples allows for a comparison of disorder effects on the high-temperature thermal transport. Our primary observation is that for all samples we measured the inverse diffusivity is linear in temperature, and can be fitted to

$$D_{Q}^{-1}(T) = aT + b = \left(\alpha v_{B}^{2} \frac{\hbar}{k_{B}T}\right)^{-1} + D_{0}^{-1}.$$
 (1)

The slope a is interpreted as a result of the product of a diffusion velocity v_B , a Planckian relaxation time $\tau \approx \hbar/k_B T$, and

[&]quot;Planckian" relaxation time $\tau \sim \hbar/k_BT$ [5], which leads to the iconic linear resistivity that persists beyond the MIR limit. Such a behavior has been observed in numerous resistivity measurements on strongly correlated materials including the hole-doped [6] and electron-doped [7] cuprate superconductors. However, no sufficient studies have been performed for thermal transport in these materials, despite evidence from early studies by Allen *et al.* [8], and more recent data from Zhang *et al.* [9], suggesting that thermal transport in the high-temperature regime of the strongly correlated cuprate superconductors must involve incoherent electrons and phonons.

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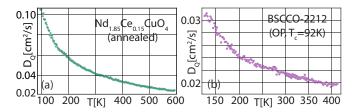


FIG. 1. Examples for thermal diffusivity in the ab plane measured as a function of temperature using the optical setup. (a) Nd_{1.85}Ce_{0.15}CuO₄ annealed, and (b) optimally doped Bi₂Sr₂CeCu₂O_{8+x} with $T_c=92$ K. Statistical error is smaller than the data points. A systematic error of $\sim 5\%$ is estimated as a result of calibration of the optical paths, and the finite size of the focused laser spots.

an order-unity constant α . The constant $b=D_0^{-1}$ represents a quantum-diffusion constant separating incoherent transport from a regime with well-defined quasiparticles, and will be discussed further below. In the absence of exact theoretical guidance we set $\alpha=1$, and estimate v_B to be about twice to three times the sound velocity in that material. In comparison to literature data for diffusivity of undoped or lightly doped insulators of similar materials, we observe that b=0 and the extracted velocity is close to the sound velocity of the respective material. These observations unambiguously establish the substantial participation of electrons in the thermal transport and suggest that also the phonons reach the Planckian limit for relaxation time.

See Supplemental Material [10] (see also Refs. [11,12]) for sample preparation methods. For the highresolution thermal diffusivity measurements we use a photothermal microscope previously used in Ref. [9], described in detail in the Supplemental Material [10]. Using this apparatus, the thermal diffusivity is obtained directly, without the need to measure the thermal conductivity and specific heat separately. An advantage of this apparatus, exploited in our previous study of underdoped YBCO, is the ability to measure the full in-plane anisotropy of the thermal diffusivity by orienting the pair of laser spots at any orientation with respect to the crystal axes. The mobility in the optics is further used for diagnostics of spatial uniformity of the thermal diffusivity values. Electrical resistivity was measured in the *ab* plane on some of the samples for comparison.

The stability of oxygen in the electron-doped cuprates allows for measurements up to 600 K. The ab-plane thermal diffusivities of two single-crystal Sm_{1.84}Ce_{0.16}CuO₄, two $Nd_{1.85}Ce_{0.15}CuO_4$, an as-grown and an annealed sample for each material, and a Pr_{1.87}Ce_{0.13}CuO₄ annealed sample were measured from 100-350 K (SCCO as-grown) and 100-600 K(all others). The thermal diffusivity of BSCCO was only measured to ~420 K, above which the crystal loses oxygen in vacuum. Examples of our diffusivity results are shown in Fig. 1, where the rest of the data is shown in the Supplemental Material [10]. These ranges of temperature capture the behavior below and above the resistive MIR limit. To appreciate the quality of the data, we refer to the Supplemental Material [10], which shows a typical frequency response of the phase shift from which we determine the value of D_O . The single coefficient fit means that for timescales of $(20 \text{ kHz})^{-1}$

to $(200 \text{ Hz})^{-1} D_Q$ is unique, indicating thermal transport of a uniform "fluid." We emphasize that all data presented in this Rapid Communication show similar behavior.

We first note that above ~ 200 K, the thermal diffusivities of all these materials are very low, indicating that in addition to being bad electrical conductors, they are also bad thermal conductors, similar to complex insulators such as perovskites [13]. An initial estimate of the electronic contribution to the thermal conductivity based on resistivity measurements and the Wiedemann-Franz law suggests that electrons contribute only about 10%–20% of the total thermal transport in the relevant temperature range (see, e.g., [8,14]). Thus, assuming thermal diffusivity that is dominated by phonons, we can estimate a thermal phonon mean free path $\ell_Q^{\rm ph}$ from the measured thermal diffusivity D_Q using the speed of sound $v_{\rm s}$ as a typical phonon velocity: $D_Q \sim v_{\rm s} \ell_Q^{
m ph}$. Following similar estimates for complex insulators [13], we use compressional sound velocity [15], with the rationale that much of the heat is transported by the longitudinal acoustic (LA) mode, since it involves excursions of atoms along the direction of heat propagation. We estimate at room temperature $\ell_Q^{\rm ph}(295~{\rm K})\sim 6~{\rm \AA}$ for the electrondoped cuprates, $\ell_Q^{\rm ph}(295~{\rm K})\sim 5~{\rm \AA}$ for near optimally doped Bi₂Sr₂CaCu₂O₈, and $\ell_O^{ph}(295~{\rm K})\sim 3~{\rm \AA}$ for underdoped YBa₂Cu₃O_{6+x} [9]. All these estimates yield mean free paths that are of the order of the lattice constant $(\sim 4 \text{ Å})$, which leads us to conclude that phonons are very strongly damped, with many modes poorly defined in this temperature range. Furthermore, the fact that the "as-grown" and "annealed" samples show similar results suggests that disorder does not play a major role in the thermal transport in this high-temperature regime. In fact, a similar conclusion was already reached by Allen et al. [8] in their study of thermal conductivity in the hole-doped Bi₂Sr₂CaCu₂O₈ and insulating Bi₂Sr₂YCu₂O₈ cuprates. Moreover, a recent comprehensive study of the LA branch corresponding to the compressional sound velocity in BSCCO crystals ($v_s \approx 4.37 \times 10^5$ cm/s) shows strong deviation from a simple harmonic mode, accompanied by interaction with optical modes already at 240 K [16]. A more detailed comparison of the length scales is given in the Supplemental Material [10].

Another notable feature of the data is that the inverse thermal diffusivities $D_O^{-1}(T)$ at high temperatures are linear in T. Figure 2 shows the same data of Fig. 1 plotted as inverse diffusivity, together with a high-temperature fit to the form in Eq. (1). For comparison, we also show resistance measurements on the NCCO and BSCCO samples in the same temperature range, where, as was previously established, they cross the MIR limit. These observations further support our previous assumption that underdoped YBCO crystals would have reached a similar linear dependence if they could be measured above room temperatures (the temperature range was limited to below room temperature to assure the same crystal structure and oxygen ordering and doping [17]). Thus, for comparison we also show in Fig. 2 our previously published data on underdoped YBCO crystals [9], together with a high-temperature linear fit asymptote. Unique to this system was the similar behavior of the inverse diffusivity and the resistivity anisotropies, which was taken as proof that both phonons and electrons participate in entropy transport above

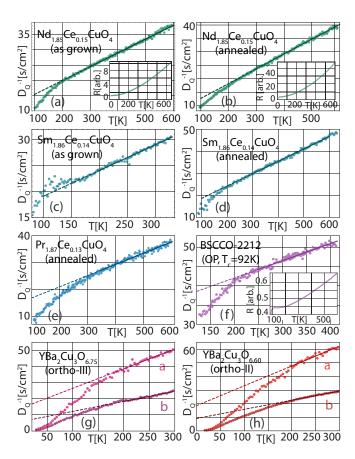


FIG. 2. Inverse thermal diffusivity $D_Q^{-1}(T)$ as a function of temperature of optimal electron-doped NCCO, SCCO, PCCO, optimal hole-doped BSCCO crystal, and two underdoped YBCO crystals from [9]. Solid lines show linear fit in the form $D_Q^{-1} = aT + b$ to the data above 300 K for electron-doped cuprates, above 250 K for BSCCO, and above 200 K for YBCOs. Dotted lines show the same fit extended to lower temperatures. Insets in (a), (b), and (f) show four-terminal resistance measured on the same crystals in arbitrary units.

the MIR limit, possibly moving as a "soup" of overdamped electron-phonon fluid.

A T^{-1} temperature dependence of the phonon relaxation time is ubiquitous in highly crystalline insulators (silicon, sapphire, etc.) at very high temperatures, traditionally explained as a consequence of umklapp scattering of phonons with a scattering rate that decreases as θ_D/T [18], often commencing well below the Debye temperature [19]. However, in the presence of impurities or other forms of disorder that limit the mean free path, notably in glasses, as the temperature increases the thermal conductivity either saturates or weakly increases, approaching the pure crystals' high-temperature limit (i.e., umklapp limit) from below [20,21]. In most of these cases umklapp scattering alone is unlikely to be sufficiently strong to account for the observed small mean-free path [22,23]. Other relaxation channels, such as higher order phonon scattering and, in particular, scattering of acoustic phonons by interaction with optical phonons may dominate the thermal transport, particularly in complex material systems. In the presence of electrons, phonon-electron scattering rate may be the dominating high-temperature cause of the observed short phonon mean free path (see, e.g., [22,24]).

Relevant to the present study are complex insulators similar in structure to the cuprates, which typically exhibit low thermal conductivity, attributed to their complex structure and a variety of local disorder effects. For example, perovskites exhibit tilting of octahedra, distortions of octahedra sites, disorder of the cation inside the octahedra, and when doped, antisite disorder [13,23]. A recent study of insulating oxide perovskites at elevated temperatures [13] revealed inverse thermal diffusivity where $\ell_Q^{\rm ph} \lesssim a$, that is dominated by a T-linear term. More recently, Martelli et~al.~[25] published a detailed study of thermal transport in SrTiO3, where at high temperatures a $D_Q^{-1} \propto T$ behavior was observed, and the prefactor was set by the sound velocity and Planckian relaxation time $\tau \approx \hbar/k_BT$ [26]. Behnia and Kapitulnik [27] further examined results on many similar complex systems ranging from high-symmetry cubic systems to glasses, and found similarly that the thermal diffusion constant is bound by the square of sound velocity times the Planckian relaxation time

The above discussion motivates us to interpret the observed $D_Q^{-1}(T) = aT + b$ behavior in a similar way to the complex insulators. Apart from the constant, "b," the thermal diffusion is characterized by a velocity v_B , a Planckian relaxation rate $\tau \approx \hbar/k_BT$, and an order-unity constant α , which depends on the dimensionality of the system and the precise coefficient in the expression of τ . The intercept, D_0^{-1} , which is the extrapolated $T \to 0$ limit of $D_Q^{-1}(T)$, will be interpreted below as a quantum diffusion constant that separates the coherent from incoherent regimes of the electronic part of the thermal transport. This constant is zero for insulators (see, e.g., [25]), which further demonstrates the fact that it is an incoherent current of both electrons and phonons that transport entropy in the high-temperature regime.

Since a prefactor for a Plankian relaxation time has not been rigorously calculated for these systems, choosing an appropriate α is somewhat arbitrary. A reasonable choice would be to take $\alpha=2.0$, which is based on a direct comparison with similar (nonmagnetic) insulators where the sound velocity is the only relevant velocity [further discussion on the choice of α is given in the Supplemental Material [10] (see also Ref. [28])]. The results for the diffusion velocity v_B for the different materials are given in Table I. For comparison we also show the respective speed of sound v_s and Fermi velocity v_F for each of these material systems. Note that the fitted v_B are very similar in magnitude, indicating very similar slope of D_Q^{-1} in the T-linear regime. Also note that a more common choice of $\alpha=1$ [4] will yield an even larger v_B .

We now turn to the residual diffusion constant D_0 . Since this $T \to 0$ intercept of the inverse diffusivity is observed only for the doped systems with appreciable carrier density, we conclude that this term is a consequence of the electronic contribution to the thermal diffusivity. However, since this term is extracted from above the MIR limit, where quasiparticles are not well defined, it will not make sense to interpret this term as a true zero-temperature limit obeying Matthiessen's rule. This empirical rule relies on coherent quasiparticles and its extrapolation to zero temperature yields the impurity scattering. However, in the spirit of this rule,

TABLE I. Effective thermal diffusion velocity v_B and effective diffusion mass m_D (in terms of free electron mass m_e) extracted from the fitting form Eq. (1) of measured thermal diffusivity using $\alpha=2.0$ (see text and Supplemental Material [10]). Here "(n)" denotes "annealed" and "(g)" denotes "as-grown." For YBCO, "(a)" denotes a axis and "(b)" denotes the chain b axis. We also list the experimentally determined speed of sound v_s [15,29] and Fermi velocities v_F [30–32].

Sample	v_s (10 ⁵ cm/s)	$v_B(\alpha = 2.0)$ (10^6 cm/s)	v_F (10 ⁷ cm/s)	m_D (m_e)
$NCCO_{0.15}(g)$	7.0	1.2	2.5	4.0
$NCCO_{0.15}(n)$	7.0	1.13	2.5	2.6
$SCCO_{0.16}(g)$	5.9	1.2	2.0	5.2
$SCCO_{0.16}(n)$	5.9	1.06	2.0	5.0
$PCCO_{0.13}(g)$	6.25	1.3	2.1	4.6
BSCCO	4.37	1.2	2.4	12
$YBCO_{6.75}(a)$	6.05	0.8	2.25	5.3
$YBCO_{6.75}(b)$	6.5	1.06		2.2
$YBCO_{6.60}(a)$	6.05	0.7	2.1	6.8
$YBCO_{6.60}(b)$	6.5	1.0		3.3

where the full scattering rate of quasiparticles is the sum of all scattering rates, we can interpret the constant $b = D_0^{-1}$ as a limiting rate for incoherent quasiparticles. This then must be the quantum of diffusion at the MIR limit, where we substitute $k_F \ell = 1$ into $D_{\rm MIR} = \frac{1}{3} v_F \ell$, yielding $D_0 = \frac{1}{3} \frac{\hbar}{m_D}$, where m_D is an effective mass associated with the diffusion process. This term also describes the limiting rate of spread of a wave packet of momentum-state quasiparticles of mass m_D in time (see, e.g., [33]), which should cease to be applied above the MIR limit. For example, effective mass extracted from quantum oscillations measurements on NCCO with Ce doping x = 0.15 yielded $m^* \approx 3m_e$ [34], while the effective mass extracted from specific heat and Raman measurements on a similar PCCO, with Ce x = 0.15 is in the same range [35,36]. On the other hand, single particle mass enhancement extracted from optical conductivity data on PCCO and NCCO with similar doping, and at room temperature, yielded $m^* \approx$ $6m_e$ [37]. For BSCCO2212 OP, m^* is estimated from specific heat to be $8.4 \pm 1.6 m_e$ [38], while for YBCO_{6.60} optical conductivity [39] and quantum oscillation was [40] found to be $m^* \sim 2-3m_e$. While these m^* values have been extracted in different temperature regimes, and using different techniques, they all seem to be quite similar to m_D , which further supports our conjecture about D_0 .

Finally we briefly discuss the resistivity above the MIR limit. As we observe from the insets in Fig. 2, as well as previous studies on similar material systems [7,41], the

$$\chi = (\rho D_e)^{-1} \approx (\rho D_Q)^{-1} \propto T^{1-y}. \tag{2}$$

While such estimation is in not an exact quantitative prediction, the decrease in χ as T increases is qualitatively consistent with the loss of carrier density at high temperature, which could be one interpretation of optical conductivity measurements in both hole-doped and electron-doped cuprates [43,44]. Recent theoretical arguments for possible reduced electron susceptibility at high temperatures were discussed by Perepelitsky *et al.* [45] in their high-temperature expansion studies of the Hubbard model, and by Werman and Berg [46], for a specific model with strong electron-phonon interaction.

On the other hand, the near optimally doped Bi₂Sr₂CeCu₂O₈ displays *T*-linear resistivity with the typical slope found in many hole-doped cuprates (see, e.g., [47]), thus allowing for a direct comparison of D_Q and D_e . In particular, we are interested in the velocity that controls the two diffusion constants above the MIR limit where the relaxation time reaches the Planckian limit. While for thermal diffusion we obtained a velocity $v_B \approx 1.7 \times 10^6$ cm/s, the slope of the resistivity yields $v_e \approx v_F$, that is, the charge diffusivity seems to be controlled by the Fermi velocity as is also evident from the data of Bruin et al. [47]. Note that here we assume a constant charge susceptibility χ . We believe that this result is particularly important for any attempt to construct a complete description of transport above the MIR limit as we show that the two processes, thermal and charge transports, can be governed by the same Planckian relaxation time but different velocities.

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resistivity of the electron-doped cuprates behaves as $\rho \propto T^y$, with $y \geqslant 1$. The case of T^2 resistivity has been discussed in detail recently [42], pointing out that a T^2 behavior at high temperatures is inconsistent with the standard Fermi-liquid theory without hydrodynamic effects. Assuming in the limit where all relaxation times are bounded by $\sim \hbar/k_BT$, thermal and charge diffusivities are expected to be equal, which, by using the Einstein relations, implies that the electronic susceptibility obeys

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^[2] The Mott-Ioffe-Regel limit has been expressed in different ways in the literature, e.g., as $k_F \ell \approx 1$, where k_F is the Fermi wave vector and ℓ is the mean free path, or $\ell/a \approx 1$ where a is the

lattice constant. These approaches typically produce the same order of magnitude estimate. In this Rapid Communication we use the criterion proposed by Emery and Kivelson in [3] of $\ell/\lambda_F \approx 1$ where $\lambda_F = 2\pi/k_F$.

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