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Thermal transport measurements through the charge density wave transition in CsV_3Sb_5

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We study thermal transport and thermalization in single crystals of CsV_3Sb_5 through the CDW transition by directly measuring thermal diffusivity (D), thermal conductivity (κ), specific heat (c) and resistivity (ρ). Commensurate with previous reports, we observe a sharp, narrow anomaly in specific heat associated with a first order transition that results in a CDW state below ~ 94 K. While a corresponding sharp anomaly in thermal diffusivity is also observed, resistivity and thermal conductivity only exhibit small steps at the transition, where the feature is sharp for resistivity and broader for thermal conductivity. Scrutinizing the thermal Einstein relation $\kappa = cD$, we find that this relation is generally satisfied, except in the narrow two-phase regime of the putative first order transition. The Wiedemann-Franz law as well seems to work outside the two-phase regime, where strong resemblance between the specific heat and the resistivity derivative below the transition may point to a concurrent emergence of a secondary electronic order parameter.

Introduction.— Understanding transport in correlated quantum materials is a subject of intense intellectual effort, with newly discovered material systems helping to stimulate fresh and original theoretical work that in particular examine the validity of common assumptions based on kinetic theory. Focusing on thermal transport, this includes Fourier’s law of heat transfer and the Wiedemann-Franz (WF) law that connects the coefficients of thermal and electrical conductivities of electrons in metals. Where correlations are pronounced and the quasi-particle picture breaks down, both assumptions may fail as was previously demonstrated in studies of thermal transport beyond the Mott-Ioffe-Regel (MIR) limit in cuprates [1–3]. There, phonons were shown to play a key role, which was strikingly evident from parallel studies of diffusivity bounds in similar complex insulators [4].

Another fertile ground to examine non-quasiparticle transport is in the vicinity of a charge density wave (CDW), which is driven by an interplay of both strong electron-electron and electron-phonon interactions. For example, thermal diffusivity, resistivity, and specific heat measurements on the CDW material ErTe_3 exhibit a sharp decrease in thermal conductivity both parallel and perpendicular to the primary CDW at the CDW transition temperature, while the resistivity changes more gradually, implying a strong breakdown of the WF law in the critical regime of the CDW transition [5]. Furthermore, assuming Fisher-Langer (FL) theory [6] applies to the continuous CDW transition in ErTe_3 , large anomalies observed in the temperature derivative of the resistivity

stands in sharp contrast to the small anomaly observed in heat capacity measurements [7]. These results are suggestive of a phenomenological recount of a strongly coupled electron-phonon critical ‘soup’ [5].

A particularly exciting new material system, where a CDW state appears as a forceful effect, is the class of quasi-two-dimensional kagomé metals AV_3Sb_5 , which exhibit charge order transitions at ~ 80 K, 103 K and 94 K for $A = \text{K}, \text{Rb}$ and Cs respectively [8]. Focusing on CsV_3Sb_5 , the CDW transition is associated with a reconstruction of the Fermi surface pockets linked to the vanadium orbitals and the kagomé lattice framework [9]. Nuclear magnetic resonance (NMR) studies on the different vanadium sites are consistent with orbital ordering at $T_{\text{CDW}} \sim 94$ K induced by a first order structural transition, accompanied by an electronic charge density wave (CDW) that appears to grow gradually below T_{CDW} , with possible intermediate subtle stacking transitions perpendicular to the Kagomé planes [10]. With superconductivity appearing at lower temperatures (~ 4 K), CsV_3Sb_5 is a prime example of a system exhibiting “intertwined order” [11, 12], where multiple phases emerge out of a primary phase, starting from the first order transition at ~ 94 K. Understanding the consequences of this unique electron-phonon landscape on thermal transport is the primary objective of the present study.

In this letter we examine the dynamics of the CDW phase transition by means of independent measurements of the specific heat, thermal conductivity, thermal diffusivity and electrical resistivity. Our primary result is that thermal transport in a critical regime near T_{CDW}

shows anomalous behavior inconsistent with quasiparticle transport. In addition, we also observe: *i*) The specific heat shows a strong singularity at T_{CDW} , although no hysteresis could be detected; *ii*) Despite it being a first order transition, a finite drop in the resistivity at T_{CDW} yields a strong peak in its temperature derivative, which seems to correspond to the specific heat anomaly similar to continuous CDW transition materials [6]. *iii*) The thermal conductivity is almost temperature independent above the transition, but starts to increase below the transition; *iv*) Thermal diffusivity through the CDW transition shows a sharp decrease, which cannot be fully explained by the ratio $D = \kappa/c$; *v*) except for the region of ~ 2 K around the CDW transition, the relation $\kappa = cD$ is satisfied quantitatively and applying the WF law to the electrical resistivity, the difference thermal conductivity gives a reasonable result for the phonon contribution to the heat transport.

Results.— CsV₃Sb₅ samples were grown at MPI Dresden following the procedure described in [13], [8], and in related publications. Here a self-flux method was used, ensuring melt purity, and producing large crystals with a high degree of structural order. For the thermal diffusivity measurements we used a photothermal microscope [1]. Fig. 1 shows specific heat, resistivity, thermal diffusivity and thermal conductivity measurements on same-batch CsV₃Sb₅ crystals. Technical details of these measurements are described in the Supplementary Material (SM)[14] (see also references [15–17] therein).

Figure 1(a) shows specific heat of a same-batch CsV₃Sb₅ crystal (see SM [14] for determination of geometrical factors), matching previous work [8, 18, 19] featuring a strong anomaly at T_{CDW} with the magnitude of the anomaly over 70%. We note we measure a larger magnitude in the specific heat anomaly compared to previous work with a measured anomaly between 20%-50%. Fitting the specific heat to a Debye model, it shows an increase in Debye temperature from $\theta_D = 160$ K at 10 K to $\theta_D = 260$ K at 80 K and above through the CDW transition, and saturates at the high temperature Dulong-Petit value. The specific heat at T_{CDW} almost doubles its value, showing a sharp anomaly that rises to $\Delta c_p \approx 0.8$ J/cm³·K within ~ 3 K of the transition. However, despite its first-order nature, no hysteresis or latent heat (down to 6.6×10^{-4} J/cm³) was detected through the CDW transition (see Fig. 1(a) inset).

CsV₃Sb₅ resistivity was measured previously. However, due to the strong anisotropy of this layered structure, it is difficult to quantitatively determine the resistivity from resistance measurements. Indeed, scale of resistivity reported in the literature varies between 31 $\mu\Omega$ ·cm and 300 $\mu\Omega$ ·cm [8, 13, 18, 20–26], although all resistivity measurements can be scaled to show the same temperature dependence, including a small “jump” at T_{CDW} . Fig. 1(b) shows resistivity measurements on

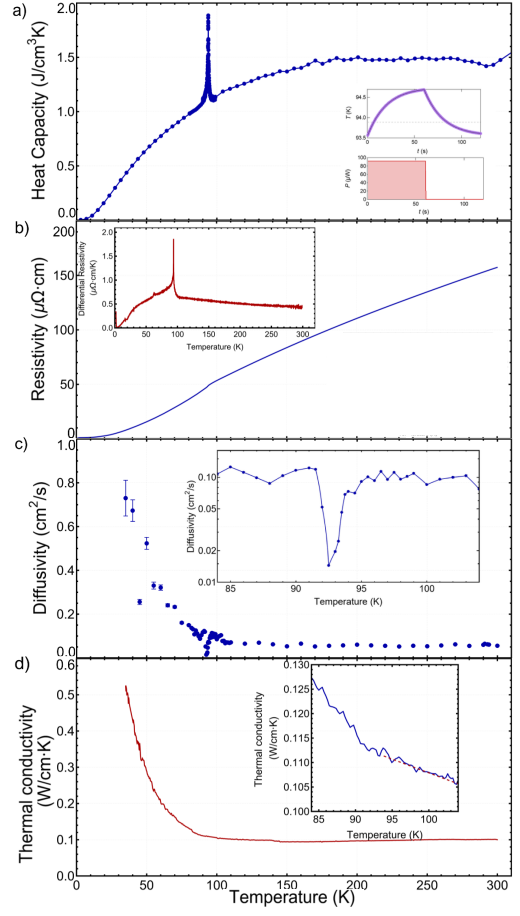


FIG. 1. Electrical and thermal measurements on CsV₃Sb₅: (a) Rescaled specific heat vs. temperature. The only anomaly in specific heat is at T_{CDW} . Inset: temperature vs. time through T_{CDW} (gray dashed line) with heater on ($t < 60$ s) and off ($t > 60$ s). No plateau of temperature indicates no latent heat. Black solid lines indicate relaxation time fit to the time evolution of the sample temperature $T(t)$. (b) Rescaled a-b plane resistivity vs. temperature. Inset: differential resistivity vs. temperature. Note large anomalies at 2.5 K (superconductivity) and 94 K (CDW). There are smaller anomalies at 62 K possibly indicating a transition to a $2 \times 2 \times 4$ supercell and at 18 K. (c) Thermal diffusivity measured using photothermal microscope with CDW transition at $T_{CDW} \approx 94$ K. Scatter of data primarily associated with one-pixel control of measurement position and relative distance between heating and probing laser spots. Note the sharp drop of diffusivity at the CDW transition and the strong increase below the transition. (d) Direct measurement of total thermal conductivity κ vs. temperature rescaled to match known room temperature values. See [14] for details. Inset: thermal conductivity in vicinity of T_{CDW} .

our crystals exhibiting 160 $\mu\Omega$ ·cm at 300 K and a RRR between 150 and 220, resulting in very low residual resistance and indicating high sample quality. We can also use the WF law and our thermal diffusivity and conductivity measurements to put a lower bound of $\sim 40\%$ for the heat conducted by electrons at room temperature, while the

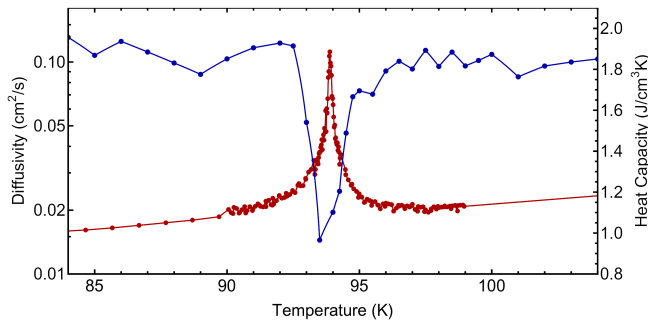


FIG. 2. CsV_3Sb_5 thermal diffusivity (left axis) measured using photothermal microscope near $T_{CDW} \approx 94$ K (blue points). Diffusivity values shifted up by 1 K from different same-batch crystals in different apparatuses to match T_{CDW} with specific heat. Scatter of data primarily associated with one-pixel control of measurement position and relative distance between heating and probing laser spots. Additional 5% systematic uncertainty, see SM [14] not included. Note the increase in diffusivity below T_{CDW} and the very sharp drop of diffusivity by over an order of magnitude at the CDW transition. Red curve shows specific heat (right axis). Note that while the diffusivity decreases by 90%, specific heat only increases by 70%.

remainder, i.e. phononic, component of thermal conductivity ~ 0.06 W/cm·K is similar to other layered chalcogenide materials [5]. Using WF to extract electronic thermal conductivity below T_{CDW} yields a sharp upturn, before it turns down at low temperatures. Finally, the small resistivity jump at T_{CDW} yields a very sharp singularity in the derivative $\partial\rho/\partial T$ shown in Fig. 1(b) inset. A weaker anomaly seen at ~ 62 K may correspond to a previously observed transition to a $2 \times 2 \times 4$ supercell [9], while an observed anomaly at ~ 18 K was not previously reported.

Thermal diffusivity, shown in Fig. 1(c), was directly measured using a photothermal microscope [1] from 30 K to 300 K, showing three different regions: below T_{CDW} , around T_{CDW} , and above T_{CDW} . Above the CDW transition temperature, the diffusivity is approximately constant with a slight increase at lower temperatures. Lowering the temperature towards the transition, the thermal diffusivity sharply decreases by at least a factor of $\times 20$ as it is limited by the resolution of the measurement. This anomaly is followed by a recovery to roughly the value just above T_{CDW} , with a trend of increasing thermal diffusivity below the transition. Fig. 2 depicts an expanded region of the CDW transition, showing that within the larger uncertainty in diffusivity data, the specific heat and diffusivity anomalies are roughly similar in width, about $\lesssim 3$ K around T_{CDW} , similar to previously reported NMR measurements [10]. Below T_{CDW} a sharp increase in thermal diffusivity together with a decrease in electrical resistivity is consistent with decrease in electron-phonon scattering.

Thermal conductivity in the a - b plane has been measured previously with though there is a spread in the quantitative conductivities throughout the entire temperature range [19, 23, 27]. Therefore, thermal conductivity in the a - b plane was also measured as shown in Fig. 1(d) with the aim to compare this direct measurement to the measurements of specific heat and diffusivity on the same growth of crystals. Here a known heat current was introduced to the sample and the measured temperature gradient across the sample was measured, showing values between 0.05 K and 0.8 K. Additional details of thermal conductivity measurements are described in the Supplementary Material (SM): [14]. Our measurements are in agreement with previous literature. While not as pronounced as in other measurements, we also see a slight decreases in thermal conductivity from room temperature to around 140 K similar to the glasslike thermal transport previously observed [19, 27]. The most striking observation here is that the thermal conductivity does not show any singularity at the CDW transition, similar to previously reported results in [19, 23]. The inset in Fig. 1(d) further indicates a tendency to an increase below T_{CDW} , again similar to [19, 23]. Assuming as in Eqn. 4 a global thermal “Einstein relation” for the thermal conductivity $\kappa = cD$, it is surprising that vestiges of the first order transition singularity at T_{CDW} are absent in κ .

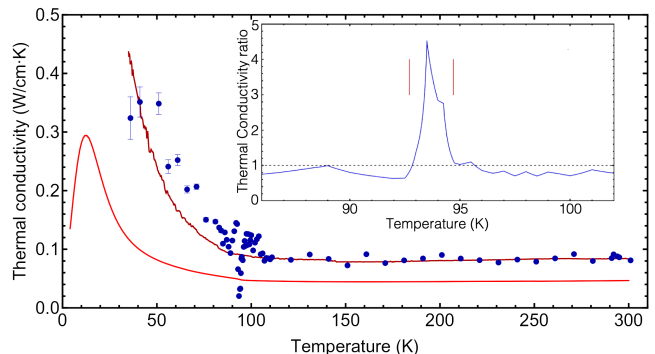


FIG. 3. Direct measurement of total thermal conductivity κ , of CsV_3Sb_5 (dark red and dark orange), electronic component κ_{el} computed from ρ assuming WF law (red), and calculated conductivity with 1 K shift in diffusivity to match T_{CDW} with specific heat $\kappa = Dc$ (dark blue). Inset shows the ratio: $R_q = \kappa/cD$ as a function of temperature indicating a strong deviation from the simple diffusion regime ($R_q = 1$) in the vicinity of the CDW transition (marked with two vertical lines). Note the otherwise good agreement between the directly measured and calculated conductivities.

Comparing the direct measurement of thermal conductivity with the aforementioned thermal Einstein relation, Fig. 3 shows an overall excellent correspondence of the two approaches above the critical region, a strong deviation in the critical region and a small deviation below the CDW transition. An instructive way to demonstrate this

observation is shown in the inset of Fig. 3 where we plot the ratio: $R_q \equiv \kappa/cD$ using the experimentally measured quantities. Where the kinetic approach prevails, we expect $R_q = 1$, which seems to hold above T_{CDW} . We find that $R_q \gg 1$ in the ~ 3 K around T_{CDW} , which is the two phase regime as was also found in the NMR experiments of Song *et al.*, [10]. We also include in Fig. 3 the electronic thermal conductivity calculated from the measured resistivity and assuming the WF law, which shows similar trend outside the CDW transition region, with the difference from the measured thermal conductivity indicating a reasonable phonon contribution, at least above T_{CDW} [5]. Below T_{CDW} the difference $\Delta\kappa = \kappa - \kappa_e$ shows increase as well, presumably due to increase in both phonon and electron thermal conductivities due to decrease in electron-phonon scattering. It is thus reasonable to assume that $\Delta\kappa \equiv \kappa_{ph}$ outside the critical regime. We note that previous work suggests that the Lorenz number from the Wiedemann-Franz law begins to increase from $L_0 = \pi^2 k_B^2 / 3e^2 \approx 2.44 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$ around 40 K to $3 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$ at 30 K and continuing to rise to $6.9 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$ at 4.2 K [19]. This change in Lorenz number would slightly lower the $\Delta\kappa \equiv \kappa_{ph}$ below 40 K. Combining the increase in the Lorenz number with our sample quality and low residual resistivity, we expect and do measure a larger low temperature total thermal conductivity than has been previously reported.

Discussion.— Heat conduction in solids is mediated by the thermal motion of quasiparticles and various elementary excitations, primarily phonons, which also serve to locally thermalize the solid. Where both electrons and phonons are well defined, the thermal conductivity is the sum of both electrons and phonons thermal conductivities $\kappa = \kappa_e + \kappa_{ph}$, such that a temperature gradient applied to the sample results in a heat flux density

$$\vec{j}_q = -\kappa \nabla T, \quad (1)$$

transferring entropy from the hotter to the colder end of the sample. This so-called Fourier’s law considers heat as a “fluid” and together with the first law of thermodynamics lead to the heat equation. Applying energy conservation, the local molar energy density $u(\vec{r}, t)$ will satisfy a continuity equation

$$\frac{\partial u}{\partial t} + \nabla \cdot \vec{j}_u = 0. \quad (2)$$

Since most experiments are done at constant pressure conditions, it is advantageous to consider the first law in the form $dh = Tds + vdP + \sum_i \mu_i dn_i$, where $h(s, P, \{n_i\})$ is the molar enthalpy of the system, s is the molar entropy, v is the molar volume, P is the pressure, μ_i is the chemical potential of the i^{th} component (if any) and n_i is the respective density. For a one-component system such as in our present experiment and under constant pressure and density conditions, it is advantageous to consider the

continuity equation for the enthalpy. This can be shown to yield the heat equation (written in components convention):

$$c_p \left(\frac{\partial T}{\partial t} \right)_P - \kappa_{ij} \partial_i \partial_j T - (\partial_i \kappa_{ij}) \partial_j T = 0 \quad (3)$$

For most cases the thermal conductivity is uniform in any particular direction in space and thus the second gradient term vanishes yielding a simple diffusion equation with $D_{ij} = \kappa_{ij}/c_p$. If the electrons and phonons are in equilibrium and well defined quasiparticles, the kinetic approach (e.g. for the isotropic case) implies:

$$\kappa = c_p D = \kappa_e + \kappa_{ph} = c_e D_e + c_{ph} D_{ph} \quad (4)$$

where c_e and c_{ph} are the electronic and phononic specific heats and D_e, D_{ph} are the corresponding thermal diffusivities. Furthermore, when transport is dominated by weakly interacting quasi-elastic scattering processes, κ_e is related to electrical conductivity by the Wiedemann-Franz law, i.e. $\kappa_e/\sigma = L_0 T$, where $L_0 = \pi^2 k_B^2 / 3e^2 \approx 2.44 \times 10^{-8} \text{ W}\Omega\text{K}^{-2}$ is a universal constant. Observing this ratio indicates “standard” transport in a given electronic system, while significant violations of the WF law may indicate a breakdown of the quasiparticle description (see e.g. [28]).

The data presented above clearly shows that while the kinetic approach, which follows Eq. 4 holds in most of the temperature regime, it strongly fails in a narrow temperature range around the CDW anomaly. We noted earlier that $R_q = 1$ holds except for a ~ 3 K range around T_{CDW} . In fact, this is the exact temperature range where a two-phase region is observed in the NMR experiments of Song *et al.*, [10]. Since the thermal conductivity does not show any significant anomaly at T_{CDW} (similar to the resistivity), we would a-priori expect that the anomaly in the specific heat will be compensated by the anomaly in the thermal diffusivity. The fact that R_q shows a strong singularity at the transition indicates the breakdown of simple heat diffusion. This can happen if e.g. the nonlinear term in Eqn. 3 becomes significant in the two-phase regime where the system exhibits randomly distributed puddles of the two phases, thus κ_{ij} varies in space, causing the $\partial_i \kappa_{ij}$ term to be finite. In addition to the spatial inhomogeneity in the two-phase regime, the latent heat of the transition [29] will create local temperature variations, which will be time dependent as per the first term in Eqn. 3. Thus, at a given time scale the term $(\partial_i \kappa_{ij}) \partial_j T$ may dominate over simple diffusion, yielding a strong anomaly in the measured diffusivity. For a slow measurement of the thermal conductivity the electrons and phonons are in equilibrium and κ may not acquire any singularity through the transition. However, the diffusivity is a dynamical property that depends on the time scale of the measurement, the associated relative fraction of the two phases through the transition as well as their spatial distribution.

While we measured thermal transport in the *ab*-plane, Pang *et al.* measured thermal conductivity in the stacking *c*-axis [27]. There they measured glass-like thermal conductivity along the *c*-axis and a sharp increase in thermal conductivity from 0.0014 W/cm² to 0.0029 W/cm². Interestingly, this doubling of thermal conductivity is only slightly larger than the almost doubling of the specific heat we measure. Thus, it is possible that the Einstein ratio $R_q \approx 1$ does hold along the *c*-axis through the CDW transition and two-phase regime and the violation only occurs along the *ab*-plane.

Focusing on our experimental results, both the specific heat (Fig. 1a) and the thermal conductivity (Fig. 1d) were measured quasi-statically as is evident from the lack of hysteresis in these two measurements. On the other hand the thermal diffusivity was measured at a finite frequency (1 to 5 kHz), exhibiting a very strong dip at the transition and accompanied by a large scatter of the diffusivity value in different runs, presumably due to difference in local distribution of the two phases. Below the transition the system is anisotropic and whether $R_q = 1$ may depend strongly on the nature of the CDW state. Indeed, close examination of directly measured κ vs. the combination cD below T_{CDW} may indicate the latter exhibiting a slightly larger value and $R_q \lesssim 1$. However, as seen in Fig. 3, the steep increase of the thermal conductivity below the transition prevents an accurate determination of this value. In particular we note the large variation in resistivity among measured samples, which if WF is used implies similar variation in the electronic part of the thermal conductivity.

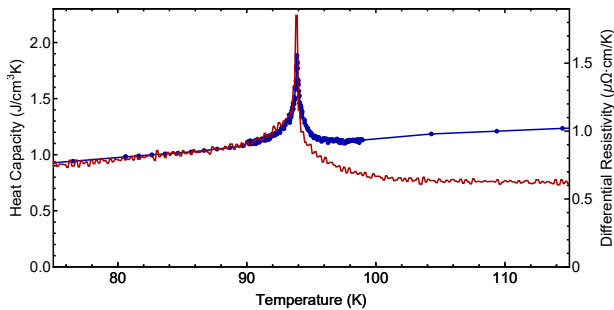


FIG. 4. Differential resistivity $d\rho/dT$ vs. temperature and specific heat vs. temperature near T_{CDW} . Note the strong correspondence below T_{CDW} but not above.

Turning to the anomaly of the electronic response through the transition, it is instructive to plot the derivative of the resistivity, which exhibits a sharp drop at the transition. We notice that while above the transition the correspondence of the two anomalies is poor, below the transition it is remarkably similar, persisting well below the narrow two-phase regime. For a continuous transition Fisher-Langer (FL) theory [6] states that the dominant contribution to the scattering near the phase transition of a metal is due to the short-range order-parameter fluctu-

ations, which in turn cause the temperature derivative of the resistivity through the transition to simulate the specific heat anomaly. However, here we observe a first order transition, for which the FL theory is not applicable. A possible explanation to the correspondence of the anomalies in $d\rho/dT$ and c_p below the transition is that a corresponding continuous order parameter emerges, which is triggered by the first order distortion transition at T_{CDW} . This idea is further supported by NMR studies, which suggest that the observed transition originates from orbital ordering at $T_{CDW} \sim 94$ K induced by a first order structural transition, accompanied by electronic charge density wave (CDW) that appears to grow gradually below T_{CDW} [10]. In that case we may expect two different effects that will affect the resistivity through the transition. While the first order structural transition may primarily induce the finite step in the resistivity as a result of a sharp change in the density of states, the accompanied continuous formation of CDW below the transition may be responsible for the enhanced scattering, conforming to the FL theory.

In conclusion by comparing direct measurements of thermal conductivity, specific heat, thermal diffusivity and electrical resistivity we could test the validity of the kinetic approximation, where heat is considered a diffusive fluid, as well as the Wiedemann-Franz law for the electronic part of the thermal conduction. We find that both hallmarks of standard transport in metals hold except in the very narrow regime of the first-order transition, where a two-phase regime appears.

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