Kinks in the electronic specific heat of strongly correlated systems

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We find that the heat capacity of a strongly correlated metal presents striking changes with respect to Landau Fermi liquid theory. In contrast with normal metals, where the electronic specific heat is linear at low temperature (with a $T^3$ term as a leading correction), a dynamical mean-field study of the correlated Hubbard model reveals a clear kink in the temperature dependence, marking a rapid change from a low-temperature linear behavior and a second linear regime with a reduced slope. Experiments on LiV2O4 support our findings, implying that correlated materials are more resistive to cooling at low $T$ than expected from the intermediate temperature behavior.

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If we trace from low to high temperatures the specific heat capacity $c_V = \partial E / \partial T$ of a solid, it provides for a rich variety of information. For a metal it increases linearly, $c_V = \gamma_0 T$, with the prefactor $\gamma_0$ proportional to the density of the electronic states, i.e., $\gamma_0 \sim N(E_F)$. This result is also valid for correlated systems that maintain a normal metallic behavior. In this case we can rely on Landau’s normal Fermi liquid (FL) theory, which describes the low-energy excitations of correlated (interacting) electrons as “quasiparticles” (QP) which are adiabatically connected to the non-interacting electrons. As a result, only a QP renormalization factor $Z_{FL}$ needs to be included in comparison to non-interacting electrons so that $c_V = \gamma_{FL} T$ with $\gamma_{FL} = \gamma_0 / Z_{FL}$. This description is so universally applicable that special attention is paid to any deviation occurring in the vicinity of special points (e.g., Quantum Critical Points), where the specific heat shows a logarithmic $T$-dependence.

Turning back to the normal case, the common understanding is that the next electronic contribution to the specific heat is cubic, $\sim T^3$. This is of the same order as the contribution from the lattice degrees of freedom, where the prefactor is given by the stiffness of the lattice and the mass of its ions. This makes the “lattice” prefactor much larger than the electron contribution, so that the cubic phonon contribution is usually dominant. At higher temperature, finally, the specific heat saturates with a value proportional to the number of degrees of freedom in the system (law of Dulong and Petit).

In this paper we show that the above described common understanding of the low-temperature specific heat of a metal needs to be markedly corrected, if the movement of the electrons is strongly correlated because of their mutual Coulomb interaction. Our finding is based on numerical solution of the Hubbard model using Dynamical Mean-Field Theory (DMFT), combined with a field theory formula for the specific heat calculation given by Abrikosov et al. and recent results for the energy-momentum dispersion relation.

As mentioned above, starting point of our consideration is the half-filled single band Hubbard model, the minimal model which describes strongly correlated electrons on a lattice. This model is solved numerically using DMFT for a semicircular DOS with bandwidth $W$, and exact diagonalization (ED) as impurity solver with 7 energy levels in the bath.

Fig. 1 shows the total energy $E_{tot}$ as a function of $T$ and the specific heat $c_V$ obtained through numerical differentiation for a ratio Coulomb interaction ($U$) to band-
width \((W) = 0.8\) (top panels) in the temperature range where \(c_v(T)\) is monotonically increasing. Thanks to the extremely dense temperature mesh, our results clearly show a rapid but continuous change of slope (kink) of \(c_v\) at \(T^* \sim 0.015W\), a feature entirely unexpected for a normal FL. This kink becomes more and more pronounced when electronic correlations are further enhanced by increasing the Coulomb interaction (we show in the bottom panels of Fig. 1 the case of \(W = 1\)), i.e., when moving towards the metal-to-insulator (phase) transition. At the same time, the value of \(T^*\), where the kink appears, is reduced, displaying a clear relation with the increasing correlations which reduce \(Z_{FL}\).

A proper fit to the numerical data hence needs to consist of two slopes (renormalization factors) \(\gamma_{FL}\) and \(\gamma_2\) instead of a single one: \(c_v = \gamma_{FL}T\) for \(T < T^*\) and \(c_v = B + \gamma_2T\) for \(T > T^*\) with a rather sharp crossover in between. This has been achieved through fitting \(E_{tot}(T) = \left[E_{tot}(0) + \gamma_{FL}T^2/2\right] f(T - T^*) + \left[E_{tot}(T^*) + BT + \gamma_2T^2/2\right][1 - f(T - T^*)]\), using a Fermi-function-like change \(f(x) = 1/(1 + e^{\beta x})\) for the crossover at \(T^*\). Note that this fit (solid red line in left panels of Fig. 1) is valid in the temperature range where \(c_v(T)\) is monotonically increasing, i.e., approximatively \(0 < T < T^*\).

To assess the reliability of our impurity solver, we compared our results with precise DMFT(QMC) data [11] (green dots in Fig. 1 second row, first panel). The comparison of the total energy shows an excellent agreement with our DMFT(ED) calculations. Notice that a direct comparison of the total energy shows an excellent agreement with our DMFT(ED) calculations. Notice that a direct comparison of the total energy shows an excellent agreement with our DMFT(QMC) calculations. Notice that a direct comparison of the total energy shows an excellent agreement with our DMFT(QMC) calculations.

In the case of a FL, where just one renormalization factor \(Z_{FL}\) is present for the low-frequency behavior of the self-energy, the standard FL formula \((c_v(T) = \gamma_0/Z_{FL}T)\) is easily recovered.

The same Eq. (2), however, yields completely different results for strongly correlated metals: When the interactions are strong enough, the spectral function displays a typical “three-feature” structure (the QP peak, and the two Hubbard subbands), which survives to moderate doping. In this situation two distinct renormalization factors can be identified in the low-frequency regime with a kink in the real part of \(\Sigma(\omega)\) in between [8]. Specifically, Ref. [8] shows that while the lowest frequencies follow the FL behavior \(\text{Re}\Sigma(\omega) = (1 - 1/Z_{FL})\omega\) there is a rapid (but continuous) change of slope (kink) in \(\text{Re}\Sigma(\omega)\) at frequency \(\omega^* < W\) (e.g., \(\omega^* \simeq (\sqrt{2} - 1)Z_{FL}W/2\) in the case of the semicircular DOS). For larger frequency, \(\text{Re}\Sigma(\omega) = -b + (1 - 1/Z_{CP})\omega\) with a reduced slope \(Z_{CP} > Z_{FL}\) (typically by about a factor 2), see inset of Fig. 2 and Ref. [8]. The constant \(b = (1/Z_{FL} - 1/Z_{CP})\omega^*\) ensures the continuity of \(\Sigma(\omega)\).
As a consequence of this self energy kink, the Green functions and their temperature derivatives appearing in Eq. (2) have to be written separately for the two regimes, namely $G_R = (yT/Z_{FL} - \epsilon + i0^+)^{-1}$ and $\frac{dy}{dT}G_R^{-1}(\epsilon, yT) = y/Z_{FL}$ for $\omega < \omega^*$, while for frequencies larger than $\omega^*$ one has $G_R = (yT/Z_{CP} - \epsilon + b + i0^+)^{-1}$ and $\frac{dy}{dT}G_R^{-1}(\epsilon, yT) = y/Z_{CP}$ with $Z_{CP} > Z_{FL}$. The parameters have been extracted from fitting $\Sigma(\omega)$ of Ref. [16] (the use of the numerical renormalization group as an impurity solver allowing for very accurate low-frequency results). In the inset of Fig. 2 we show $\Sigma(\omega)$ and the fit (blue line). We recall in passing that it is $Z_{CP}$, which controls the width of the “quasiparticle” peak in the interacting DOS; while $Z_{FL}$ characterizes only the asymptotic properties in the limit $\omega \rightarrow 0$ (or $T \rightarrow 0$) [6].

The evaluation of Eq. (2) has been performed by splitting explicitly the integral over $y$ in the two regions:

$$c_V(T) = T \left[ \frac{1}{Z_{FL}} \int_{|y| < \omega^*} N\left(\frac{yT}{Z_{FL}}\right) + \frac{1}{Z_{CP}} \int_{|y| > \omega^*} N\left(\frac{yT}{Z_{CP}} + b\right) \right] \times dy \frac{y^2 e^{y}}{(e^{y} + 1)^2}$$

This equation is the final result of our analytical calculation. It allows us to compute, through a simple integral, the specific heat from the non-interacting density-of-states $N(\epsilon)$, the two renormalization factors $Z_{FL}$ and $Z_{CP}$ and the kink frequency $\omega^*$. Using the parameters extracted from Ref. [16] we obtain the solid line shown in Fig. 2.

It is easy to verify that the standard Fermi-liquid behavior is recovered from Eq. (3) in the limit of large $\omega^*$ (i.e., when only one low-frequency scale is present). In the opposite limit $\omega^* \rightarrow 0$ a standard Fermi-liquid behavior is also recovered, though with a different renormalization factor $\gamma = \gamma_0/Z_{CP}$. More interesting is the intermediate situation, which we are considering here, where $\omega^*$ lies in the low-frequency range. In this case, the specific heat behavior shows a kink at a temperature $T^* \propto \omega^*$ (with a proportionality factor of about 1/5 for the case of the semicircular DOS). The agreement with the AGD formula is particularly remarkable if we notice that, strictly speaking, the AGD formula is only applicable to the linear-$T$ regime, since it does not include all additional terms leading to the aforementioned $T^3$ contribution. However, this term is small at low temperatures. Hence, if the correlation is strong enough, it can push the kink in the very small $T$ regime, where the AGD formula is expected to work. This explains why our analytical calculation is able to reproduce our numerical results to a very good accuracy in Fig. 2. Let us emphasize that it was not at all clear a priori whether an AGD-like calculation was possible beyond the regime of Landau’s QP, i.e., after the kink in the energy-momentum dispersion which indicates the basic excitations are no longer Landau QP.

The theoretical evidence of a low-temperature kink in the electronic specific heat of strongly correlated systems poses the question of its experimental observation, which was -so far- still lacking. The main problem is obviously the phonon contribution $c_V \sim T^3$ which, because of its large prefactor, usually overshadows the much smaller electronic contribution to the specific heat, already at temperatures of few ten Kelvin. This restricts the choice to materials which show the kink at a very low $T^*$. That means in turn compounds with a strong renormalization ($Z_{FL} \ll 1$), i.e., heavy Fermion systems. Given our starting point, the Hubbard model, the ideal material is LiV$_2$O$_4$, the first d-electron system where heavy Fermion behavior was found [17]. Indeed, recent LDA+DMFT calculations [18], which take into account the realistic three-d-band structure of LiV$_2$O$_4$, have demonstrated that an effective description in terms of the single-band Hubbard model (very close to half-filling) is particularly appropriate for this compound.

In Fig. 3, we show that our theory nicely describes the
FIG. 3: (Color Online) Kink in the low temperature specific heat of LiV$_2$O$_4$ (open blue circles) visible at $T^* \sim 5 - 6$K, and well reproduced by our analytical theory (red solid line) experimental results for LiV$_2$O$_4$. We compare the data of Ref. 19 (displayed in a magnified low-$T$ range with respect to the original publication) with our analytical formula, fitting the free parameters to the experimental data. Indeed, a kink is clearly visible, as at the curve rapidly changes its slope at a temperature $T^*$ of 5 – 6K. The three fitting parameters ($Z_{FL} = 0.054; Z_{CP} = 0.092, \omega^* = 0.0035W$ with $W = 600$ meV for LiV$_2$O$_4$) assume very reasonable values. This clearly confirms the strong-correlation origin of the kink in the specific heat of this material.

We notice there are also kinks in the specific heat of $f$-electron heavy Fermions such as YbRh$_2$Si$_2$[20] or YbCu$_{5-x}$Al$_x$[21]. However these materials are close to a quantum critical point, at which additional physical processes become important. In some systems also long range magnetic order leads to additional structures in the specific heat. Hence, at present, it is less clear in how far these kinks are connected to our theory. Another material with strongly correlated Fermions showing similar kinks in the specific heat is $^3$He (Ref. 22,23) for which however the application of a lattice model such as the Hubbard model represents certainly quite a crude approximation.

In conclusion, we have demonstrated numerically, analytically and experimentally that the textbook knowledge of the electronic specific heat at low temperatures needs to be modified for strongly correlated electrons. In the proximity of the Mott transition the leading correction to the linear Fermi-liquid temperature behavior is a quite rapid change of slope, i.e., a kink, which takes place well before (at smaller $T$) the standard $T^3$ behavior becomes relevant. Let us emphasize the reported kink is a generic feature of strongly correlated electron systems, in very contrast to existing theories for kinks stemming from the coupling to (potentially present) bosonic degrees of freedom. Since the slope of the specific heat is reduced after the kink, the behavior of $c_V/T$ is just opposite to what one would expect from the standard theory, i.e., $c_V/T$ is decreasing with increasing temperature instead of the expected increase due to the cubic term. Hence, if one extrapolates from the behavior at intermediate temperatures (i.e., after the kink) without taking into account the kink, a much lower specific heat at low temperatures is obtained with respect to the actual result. In other words, a material with strongly correlated electrons can be unexpectedly resistant against cooling at low temperatures. Moreover, depending on the temperature range considered in the experiments, only one of the two regimes of linear behavior of $c_V(T)$ may be accessible. This can easily lead to remarkable inconsistencies in the analysis of the experimental data for strongly correlated materials.

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