Crossover to non-Fermi-liquid spin dynamics in cuprates

J. Bonča\textsuperscript{1,2}, P. Prelovšek\textsuperscript{1,2}, and I. Sega\textsuperscript{1}

\textsuperscript{1}J. Stefan Institute, SI-1000 Ljubljana, Slovenia and
\textsuperscript{2}Faculty of Mathematics and Physics, University of Ljubljana, SI-1000 Ljubljana, Slovenia

(Dated: 22nd March 2022)

The understanding of the phase diagram of cuprates continues to exemplify one of the major theoretical and experimental challenges [1]. Besides superconductivity (SC) and antiferromagnetic (AFM) ordering, several regimes with distinct electronic properties have been identified within the normal metallic phase. The behavior of spin degrees of freedom, which are the subject of this paper, has been intensively studied using the inelastic neutron scattering (INS) [2,3] and NMR relaxation experiments [4]. They clearly reveal that in underdoped cuprates magnetic properties are not following the usual Fermi-liquid (FL) scenario within the metallic state above the SC transition $T > T_c$. Within a normal FL one expects a dynamical spin susceptibility $\chi''_Q(\omega)$ to be $T$-independent at low $T$, $\omega$. On the contrary, INS results show that $q$-integrated spin susceptibility exhibits in a broad range of $\omega$ and $T$ an anomalous, but universal behavior $\chi''_q(\omega) \propto f(\omega/T)$, first established in La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) at low doping [3,5]. This behavior can be even followed to lowest $T$ in YBaCu$_2$O$_{6+x}$ (YBCO), where $T_c$ has been suppressed by Zn doping [4]. At the same time, low-energy INS reveals at low $T$ the saturation of the inverse AFM correlation length $\kappa = 1/\xi$, at least in YBCO [2] and in LSCO at low doping [3,5]. Anomalous $T$-dependence of $^{63}$Cu NMR spin-lattice relaxation rate $1/T_1$ and of the spin-spin relaxation rate $1/T_2$ in overdoped cuprates is in general compatible with INS [4], in particular $1/(T_1T) \propto \chi''_L(\omega,T)/\omega|_{\omega \rightarrow 0} \propto 1/T$, in contrast to a $T$-independent value (Korringa law) in a normal FL.

On the other hand, cuprates at optimum doping and, moreover, in the overdoped regime show a strong reduction of the spin response at low energies $\omega$. This is evident from the loss of INS intensity in the normal state (as well as in a weak resonant peak for $T < T_c$) and low NMR relaxation rates $1/T_1$, $1/T_2$. At the same time, NMR confirms the approach to the normal FL behavior, $1/(T_1T) \sim \text{const}$, and $1/T_2 \sim \text{const}$. There are other indications that the normal FL behavior is approached in the overdoped regime. Recently, the angle-resolved photoemission spectroscopy (ARPES) on BiSr$_2$CaCuO$_6$ (BSCCO) system gave evidence for the existence of coherent electronic excitations for $T < T_X$ at higher doping [7], i.e., the FL-like phase is found in the normal state only in the overdoped regime where $T_X$ shows a steep increase with hole doping $c_h$. Intimately related to the onset of the FL-like spin response is also the observation that in cuprates doped with nonmagnetic Li and Zn the impurity-induced spin susceptibility varies as $1/(T + T_K)$, i.e., with a Kondo-like behavior with a characteristic temperature $T_K(c_h)$ [8], where $T_K \sim 0$ in the underdoped regime, whereas it shows a strong increase in the overdoped regime.

From the point of theoretical understanding, an approach to a FL behavior in the overdoped regime far from a metal-insulator transition seems plausible, nevertheless a solid theoretical evidence is still missing. A crossover from a strange metal to a coherent metal phase is, e.g., predicted within the slave-boson approach [9]. Frequently invoked interpretation is given in terms of the quantum critical point (QCP) at optimum doping $c_h^o$ (masked, however, at low $T$ by the SC phase), dividing the FL phase at $c_h > c_h^o$ and a (singular) non-Fermi-liquid (NFL) metal at $c_h < c_h^o$. While such a concept is well established in spin systems [10], its application to metallic cuprates is controversial due to the absence of a critical length scale (e.g., $\xi(T \rightarrow 0) \rightarrow \infty$). Low energy spin dynamics as emerges from INS and in particular from NMR experiments has been extensively analysed within the phenomenological theory [11], describing a FL close to an AFM instability. In the latter approach the spin-fluctuation energy in fact plays the characteristic FL scale, as discussed furtheron.

The present authors recently showed that an anomalous $\omega/T$ scaling, as observed at low doping, emerges from a general approach to $\chi_q(\omega)$ under a few basic requirements [12]: a) collective $Q = (\pi, \pi)$ AFM mode in the normal state is overdamped $\gamma > \omega_Q$, b) equal-time correlations $S_Q = \langle S^z_Q S^z_Q \rangle$ and the corresponding inverse correlation length $\kappa$ are finite and saturate at low $T$. A nontrivial $\omega_Q(T)$ dependence then follows from the fluctuation-dissipation relation,

$$\frac{1}{\pi} \int_0^\infty d\omega \, \text{cth} \frac{\omega}{2T} \chi''_q(\omega) = S_Q,$$

(note that we use $\hbar = 1$ and define $\chi_q(\omega)$ in units of $g^2\mu_B^2$).
leading to a $\omega/T$ scaling for $T > \omega_p \sim \gamma e^{-2\zeta}$ where $\zeta \propto \gamma/r^2$. Within such an approach it is natural that $\omega_p > 0$ is finite within the whole paramagnetic regime. Nevertheless, due to strong dependence on parameters, in particular on $\zeta$, $\omega_p(c_h)$ can show quite a sharp crossover from very small values in the underdoped regime to a large increase in overdoped systems, consistent with experimental indications.

In this paper we present numerical results for the doping dependence of the FL scale $\omega_{FL}$ within models relevant to cuprates, i.e., the planar $t$-$J$ model and the Hubbard model. We furthermore compare these quantities with the ones extracted directly from NMR-relaxation and INS experiments on cuprates. One possibility is to get $\omega_{FL}$ from the full $T$-dependence of various magnetic quantities, in particular from static $\chi_Q(T)$ and $S_Q(T)$. It is evident that in the NFL regime $T > \omega_{FL}$ a relation follows from Eq. \ref{eq:1},

$$\frac{S_Q}{T\chi_Q} = \left[1 - \frac{\Delta}{S_Q}\right]^{-1},$$

which evolves into the ‘classical’ relation for $\Delta \ll S_Q$. Note that $\Delta(T)$ arises from Eq. \ref{eq:1} as the integral over the large-$\omega$ tail $\chi_Q^\nu(\omega > T)$. We are interested in the low-$T$ regime in the paramagnetic phase where $S_Q(T)$ already saturates. The saturation is quite evident from the numerical analysis of various models \cite{1,13}. Eq. \ref{eq:2} indicates that even constant $S_Q$ can be compatible with strongly $T$-dependent $\chi_Q(T)$ which seems to be the essence of the NFL regime in cuprates. In contrast, one expects a finite $\chi_Q(T \to 0)$ within the FL regime.

The characteristic energy scale of spin fluctuations is given by $\omega_{FL}(T) = S_Q/\chi_Q(T)$ with the corresponding $T = 0$ limit $\omega_{FL}(0)$. The latter can be calculated from $T = 0$ numerical results. Note that $\omega_{FL}(0) = \langle \omega \rangle$ is just the first frequency moment of the shape function $\chi_Q^\nu(\omega, T = 0)/\omega$ for $\omega > 0$. On the other hand, we extract $\omega_{FL}$ also from experiments, in particular from NMR $1/T_2\gamma$ relaxation data, which give rather straightforward information on $\chi_Q(T)$.

Let us first consider the $t$-$J$ model,

$$H = -t \sum_{\langle ij \rangle} c_{i\sigma}^\dagger c_{j\sigma} + J \sum_{\langle ij \rangle} (S_i \cdot S_j - \frac{1}{4} n_i n_j),$$

with the nearest neighbor hopping on a square lattice, which we analyse for $J/t = 0.3$, as relevant for cuprates (for comparison with cuprates we use also $t \sim 400$ meV). Results for $S_Q(T)$ and $\chi_Q(T)$ are evaluated using the finite-$T$ Lanczos method (FTLM) \cite{14}. In this way we analyse systems with $N = 18$ sites for arbitrary hole doping $c_h = N_h/N$, and with $c_h \leq 3/20$ for $N = 20$. It should be also noted that FTLM results are rather insensitive to finite-size effects for $T > T_{fs}$, whereby for systems considered $T_{fs} \sim 0.1 t$ \cite{14}.

In Fig. 1 we present results for $\chi = 4T\chi_Q$ as a function of $c_h$ for various $T > T_{fs}$. Note that the limiting value within the $t$-$J$ model is $\chi(T \to \infty) = 1 - c_h$. Two distinct regimes become immediately evident from Fig. 1. The crossing of curves $\chi(c_h)$ with different $T$ can be used as the definition of the ‘optimum’ doping $c_h \sim 0.16$, whereby it is indicative that the same value is obtained analysing cuprates with highest $T_c$ \cite{13}. In the underdoped regime $\chi$ increases by lowering $T$ (down to reachable $T \sim T_{fs}$) and appears to saturate to the NFL behavior, Eq. \ref{eq:2}, consistent with the anomalous $\omega/T$ scaling \cite{14}. On the other hand, at $c_h > c_h^*$ the tendency of $\chi(T)$ is opposite. I.e., $\chi_Q(T)$ saturates for $T < J$, indicating a ‘normal’ FL behavior. If $\chi(c_h)$ curves would, even for lowest $T$, indeed cross at $c_h = c_h^*$, we would have been dealing with a singularity resembling a QCP with diverging $\chi_Q(T \to 0) \propto 1/T$. Moreover $\chi_Q(T \to 0)$ would be divergent in the whole regime $c_h < c_h^*$. Although present results cannot exclude this possibility, the deviation visible at lowest $T = 0.1 t$ is more in accord with a crossover between FL and NFL regimes.

In Fig. 2 we show corresponding FTLM results for $\omega_{FL}(c_h)$ at various $T \leq J$. Note that in this regime $S_Q(T)$ is essentially $T$-independent, and the values agree very well with the $T = 0$ results obtained via the usual Lanczos technique for the ground state (g.s.). The latter approach allows the calculation of $S_Q(T = 0)$ and $\chi_Q(T = 0)$ also for somewhat larger systems, i.e., for $N = 20$ at all $N_h$ and for $N = 26$ at $N_h \leq 2$. $T = 0$ results for $S_Q$ are shown in the inset of Fig. 2 and overall follow surprisingly well the linear variation $1/S_Q = K c_h$ with $K \sim 15$. In contrast to $S_Q(c_h)$, the FL scale $\omega_{FL}$ reveals a nonuniform variation with doping. Again, for $c_h > c_h^* \omega_{FL}$ is already $T$-independent for $T < J$, or at least approaching finite $\omega_{FL}(0)$. In the regime $c_h < c_h^* \omega_{FL}(T)$ is decreasing with $T$, so that we can establish only an upper bound for $\omega_{FL}$. In the same Fig. 2 we plot also results for $\omega_{FL}(0)$, evaluated directly via the $T = 0$ calculation for largest available systems. In the overdoped regime the general agreement with the FTLM is evident. As expected, in the underdoped region obtained $\omega_{FL}(0)$ seem to be consistently smaller that $\omega_{FL}(T > 0)$ values, whereby a decrease of $\omega_{FL}(0)$ with system size is also observed (e.g., values obtained for $N = 26$ systems are smaller than those for $N = 18, 20$). So we can
summarize results in Fig. 2 as follows: a) in the overdoped regime $\omega_{FL} \sim \alpha (c_h - c_{h0})$ with $c_{h0} \sim 0.12$ and a large slope $\alpha \sim 3.9t$ in 1.6 eV, b) in the underdoped regime our results seem to indicate on a smooth crossover to very small $\omega_{FL} \ll J$.

The alternative relevant model is the Hubbard model on a square lattice,

$$H = -t \sum_{<ij>}(c^\dagger_{ia} c_{jb} + \text{H.c.}) + U \sum_i n_{ia} n_{ib},$$  
(4)

which in the case of strong Coulomb repulsion $U \gg t$ and close to half-filling maps onto the $t$-$J$ model with $J = 4t^2/U$. We calculate $S_{Q}$ in the g.s. as a function of hole doping $c_h$ within the Hubbard model on a square lattice and at $U = 8t$ using the constrained-path quantum Monte Carlo method (CPMC) [16]. In this method, the ground state wave function is projected from a known initial wave function by a branching random walk in the overcomplete space of Slater determinants. Since the method is most efficient in the closed-shell cases, we extend our calculations to various tilted square lattices where the number of sites $N$ is ranging between $N = 34$ to $N = 164$. The susceptibility $\chi_{Q} = \partial m_{Q}/\partial B_{Q}$ is calculated by computing sublattice magnetization $m_{Q}$ induced by a small staggered magnetic field $B_{Q}$.

Our results for $1/S_{Q}$ again reveal a linear variation $\sim K c_h$ with $K \sim 14$. Such results are in qualitative agreement with previous QMC calculations for $U/t = 4$ [1], where in the latter case $K \sim 14.3$. In Fig. 3 we present corresponding $\omega_{FL}(0)$. The qualitative behavior of $\omega_{FL}$ is very similar to the result within the $t$-$J$ model, Fig. 2. In the overdoped regime one can again approximate the variation of $\omega_{FL}$ as linear, with $c_{h0} \sim 0.1$ and $\alpha \sim 4.8t$, while for $c_h < c_{h0}$ $\omega_{FL}$ becomes very small. Altogether, obtained $\omega_{FL}$ do not differ much from that within the $t$-$J$ model, in spite of plausibly weaker correlations within the Hubbard model for $U/t = 12$. In Fig. 3 we display also the corresponding free fermion result. We notice that on approaching the empty band $c_e = 1 - c_h \rightarrow 0$ both curves converge. However, close to half-filling there is a huge qualitative difference.

Figure 2: (color online) FL scale $\omega_{FL}/t$ vs. $c_h$, obtained for the $t$-$J$ model using the FTLM for $T > 0$ and the usual Lanczos method for $T = 0$. The inset shows $T = 0$ results for $1/S_{Q}$ vs. $c_h$. Thin lines are guide to the eye only.

Figure 3: (color online) FL scale $\omega_{FL}/t$ vs. doping $c_h$, as obtained via the CPMC method for the Hubbard model with $U/t = 8$, where the dashed line is a guide to the eye.

Let us finally estimate $\chi_{Q}(T)$ and consequently $\omega_{FL}$ directly from experiments on cuprates. Within the normal state we use the results for the NMR spin-spin relaxation time $T_{2G}$, obtained from the $^{63}$Cu spin-echo decay, related to static $\chi_{Q}$ as [11],

$$1/T_{2G}^2 = 0.69 \left[ \frac{1}{N} \sum_{q} (F(q)\chi_{Q})^2 - \frac{1}{3} \sum_{q} (F(q)\chi_{Q})^2 \right].$$  
(5)

Assuming that $\chi_{Q}$ is peaked at commensurate $q = Q$ and can be described by a Lorentzian form $\chi_{Q} = \chi Q (\kappa^2/[(q - Q)^2 + \kappa^2])$ (e.g., consistent with INS in YBCO) with $\kappa \ll \pi$, the second term in Eq. (5) can be neglected and the form factor replaced by $F(Q)$. This leads to the relation $1/T_{2G}^2 \sim 0.083\kappa F(Q)\chi_{Q}$. $1/T_{2G}$ relaxation rates have been measured and summarized in Ref. [4], i.e., from underdoped to optimally doped YBCO with $0.63 < x < 1$, underdoped YBa$_2$Cu$_3$O$_8$, nearly optimum doped Tl$_2$Ba$_2$Ca$_2$Cu$_2$O$_{10}$ (Tl-2223) and the overdoped Tl$_2$Ba$_2$CuO$_{8+\delta}$ (Tl-2201), whereby the normalization with corresponding $F(Q)$ has been already taken into account (see Fig. 8b in Ref. [4]). Note that $\kappa$ relevant to $\chi_{Q}$ is the one appropriate for low-$\omega$ spin dynamics, as measured directly by INS (plausibly $\kappa \ll \delta$). For YBCO $\kappa(x)$ has been summarized in Ref. [13]. For cuprates considered here appropriate hole concentrations $c_h$ have been estimated in Ref. [15]. Assuming a continuous variation of $\kappa(c_h)$ we determine also $\kappa$ for YBa$_2$Cu$_3$O$_8$, Tl-2223 and Tl-2201 (for the latter we take $\kappa = 1.2/a_0$), not available experimentally. In this way, we evaluate $\chi_{Q}(T)$. Equal-time correlations $S_{Q}$ are so far not directly accessible by INS. As shown before they are
nearly model independent, so we assume here the $t$-$J$ model results to finally extract corresponding $\omega_{FL}(T)$ as presented in Fig. 4 for various cuprates.

![Graph](image)

**Figure 4:** (color online) $\omega_{FL}$ vs. $T$, as evaluated from the NMR relaxation rate $1/T_{2\omega}$ and the INS width $\kappa$ for various cuprates. The inset shows the extrapolated scales $\omega_{FL}(0)$ and $T$ vs. doping $c_h$.

For $T$ well above $T_c$, $\chi_Q(T)$ extracted from $1/T_{2\omega}$ follows the Curie-Weiss behavior, i.e., $\chi_Q(T) \propto 1/(T + \Theta)$. Such a behavior emerges also within our analytical approach in the scaling regime. Hence, for $T > 150K$, we can well parametrize $\omega_{FL}(T) = \omega_{FL}(0)(1 + T/\Theta)$ and present in the inset of Fig. 4 the doping dependence of $\omega_{FL}(0)$ and $\Theta$. It is evident that both $\omega_{FL}(0)$ and $\Theta$ reveal a similar behavior, which qualitatively and to some extent even quantitatively follow our result within the $t$-$J$ and Hubbard models. In particular, there is a clear change of scale between the underdoped and overdoped cuprates.

In conclusion we present evidence, based both on numerical results within $t$-$J$ and Hubbard models as well as on the analyses of NMR and INS experimental data on cuprates, that the FL scale $\omega_{FL}$ exhibits a rather sharp crossover between a steep increase in the overdoped regime and very low $\omega_{FL} \ll \omega$ in the underdoped regime for $c_h < c_h^\ast$. Note that in the latter regime within cuprates one can easily reach values $\omega_{FL}(0)$ smaller than $T_c$. This can explain why anomalous NFL scaling of the spin response as well as of other quantities is observed throughout the normal phase at $T > T_c$. On the other hand, the transition to the normal FL is quite abrupt in the overdoped regime, at least with respect to the spin response discussed here.

Our results are well in agreement with other experimental evidence for the existence of transition to the FL behavior in cuprates. The FL scale $T_X$, as revealed by recent ARPES experiments on BSCCO, in particular its doping dependence $T_X(c_h)$ in the overdoped regime, is close to our results for $\omega_{FL}(c_h)$ with an extrapolated $c_h^\ast \sim 0.1$. Similar doping dependent scale $T_K$, analogous to a Kondo scale in metals, arises from the analysis of the local-moment susceptibilities in YBCO with in-plane nonmagnetic Li and Zn impurities. Experiments show an abrupt and steep increase of $T_K$ on approaching the optimum doping. It is plausible that the impurity-induced uniform susceptibility is related to local $\chi_L$ (and to staggered $\chi_0$) in an uniform system, hence $T_K$ seems to be related to $\Theta$. Needless to say such a relation requires a theoretical justification.

Still, our numerical results cannot exclude the possibility of the existence of a QCP. From our analysis, the latter can be present at the point where $\omega_{FL}(0)$ vanishes on approaching the overdoped side, i.e., in our model systems at $c_h \sim c_h^\ast < c_h^\ast$. An analogous interpretation might follow also from experimental values in Fig. 4, as well as from results on the Kondo temperature $T_K(c_h)$ in YBCO. However, the main obstacle to such a scenario is that there is no evidence for an ordered AFM phase for $c_h < c_h^\ast$, neither from calculated $S_Q$ within the $t$-$J$ and Hubbard models nor from experiments. As our analysis shows $\omega_{FL}(0)$ remains finite throughout the normal phase at all dopings $c_h$ down to the onset of the ordered AFM phase at $c_h^{AFM} < c_h^\ast$. The experimental distinction between the QCP and the present crossover scenario is that in principle $\omega_{FL}(0) > 0$ in the normal phase even in the heavily underdoped regime, hence one should be able to detect this experimentally by suppressing the SC phase, e.g., as investigated with INS on YBCO system. However, the theory reveals that $\omega_{FL}(0) \propto \omega_p$ can be extremely small in the overdoped regime.

---

[1] M. Imada, A. Fujimori, and Y. Tokura, Rev. Mod. Phys. 70, 1039 (1998).
[2] L. P. Regnault et al., Physica B 213-4, 48 (1995).
[3] M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. Endoh, Rev. Mod. Phys. 70, 897 (1998).
[4] M. A. Kastner, R. J. Birgeneau, G. Shirane, and Y. Endoh, Adv. Phys. 49, 1 (2000).
[5] J. Bonča and P. Prelovšek, Phys. Rev. B 65, 121405 (2002).
[6] J. Bonča and P. Prelovšek, Adv. Phys. 50, 859 (2001).
[7] J. Bonča and P. Prelovšek, J. Phys. Soc. Jpn. 71, 1356 (2002).
[8] P. A. Lee and N. Nagaosa, Phys. Rev. B 48, 12972 (1993).
[9] P. A. Lee and N. Nagaosa, Rev. Mod. Phys. 70, 1039 (1998).
[10] J. Bonča and P. Prelovšek, Phys. Rev. B 67, 085103 (2003).
[11] A. V. Balatsky and P. Bourges, Phys. Rev. Lett. 82, 5337 (1999).