Freezing of spin dynamics in underdoped cuprates

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The memory-function approach to spin dynamics in doped antiferromagnetic insulator combined with the assumption of temperature-independent static spin correlations and constant collective-mode damping leads to ω/T scaling in a broad range. The theory involving a nonuniversal scaling parameter is used to analyze recent inelastic neutron-scattering results for underdoped cuprates. Adopting modified damping function also, the emerging central peak in low-doped cuprates at low temperatures can be explained within the same framework.

I. INTRODUCTION

It is experimentally well established that magnetic static and dynamical properties of high-T_c cuprates are quite anomalous. Even at intermediate doping the magnetic response does not follow the Fermi-liquid phenomenology in the metallic state and reveals the remarkable resonant peak within the superconducting (SC) state. With decreasing doping the magnetic response approaches in a novel way that of the reference undoped materials—the antiferromagnetic (AFM) insulator. It has been first observed by inelastic neutron-scattering (INS) experiments on low-doped $La_{2-x}Sr_xCuO_4$ (LSCO) (Refs. 1 and 2) that local (**q**-integrated) spin dynamics in the normal state exhibits anomalous ω/T scaling, not reflected in the AFM correlation length ξ which shows no significant T dependence. Similar behavior has been found also in $YBaCu_3O_{6+x}$ (YBCO), initially at $x=0.5$ and 0.6 (Ref. 3) with the regime restricted to $T>T_c$, and in Zn-doped YBCO (Ref. 4) with reduced $T_c \sim 0$.

Recent more detailed INS experiments on heavily underdoped (UD), including Li-doped LSCO,[5] YBCO with $x=0.35$ (Refs. 6 and 7) and $x=0.45$,[8,9] as well as electron-doped $Pr_{1-x}LaCe_xCuO_{4-\delta}$ (PLCCO) with $x=0.12$,[10,11] confirm universal features of anomalous normal-state spin dynamics. Latter materials exhibit no SC (Li-doped LSCO) or have very low T_c (YBCO and PLCCO) so that the ω/T scaling is found in a broad range both in **q**-integrated local susceptibility $\chi''_L(\omega)$ (Refs. 5, 7, and 9) and in $\chi''_{\mathbf{q}}(\omega)$ at the commensurate AFM $\mathbf{q}=\mathbf{Q}=(\pi,\pi)$.[5,7,10,11] Typically, $\chi''_{\mathbf{Q}}(\omega)$ is a Lorentzian with the characteristic relaxation rate scaling as $\Gamma = \alpha T$ but with a nonuniversal α. Similarly, normalized $\chi''_L(\omega)$ has been fitted to the scaling form $f(\omega,T)=\chi''_L(\omega,T)/\chi''_L(\omega,T=0) \sim (2/\pi)\tan^{-1}[A_1\omega/T+A_2(\omega/T)^3]$ (Refs. 7 and 9) with material dependent $A_{1,2}$. It is characteristic that the normal-state-like scaling is found even below T_c without any clear sign of the presence of the resonant peak. On the other hand, it has been observed that at low T the spin dynamics gradually transfers into a quasielastic peak for $T<T_g$,[5,6] whereas the inelastic response saturates. However, this *freezing* mechanism appears to be of entirely dynamical origin since ξ as well as the integrated intensity are unaffected by the crossover.

In order to describe the anomalous scaling of spin dynamics as well as of other electronic properties of cuprates, the concept of marginal Fermi liquid has been introduced.[12] Whereas the deeper origin of such behavior is often given in terms of proximity to a quantum critical point (QCP),[13,14] the above systems clearly lack the criticality of $\xi(T)$. It is thus quite remarkable that a similar behavior is also found in heavy-fermion metal system CeCuAu,[15] which possesses a well-defined QCP (unlike in cuprates) in the phase diagram, nevertheless exhibits similar ω/T scaling without the criticality in $\xi(T)$.

The present authors introduced a theory of spin dynamics in doped AFM,[16] which describes the scaling behavior as a dynamical phenomenon. Namely, assuming that static quantities—the equal-time spin correlations in particular—are unaffected (or weakly dependent) by T and that the system is metallic with finite spin collective-mode damping, the system close to AFM naturally exhibits ω/T scaling in a wide energy range, but unlike at usual QCP, with a saturation at low-enough T. It should be noted that such a scenario is close to the theory of freezing of a liquid, which is as well a dynamical phenomenon reflected only weakly in equal-time correlations.

The aim of this Rapid Communication is to analyze recent INS experiments on different cuprates mentioned above within this framework and show that observed differences (nonuniversality) in scaling behavior can be made consistent by taking into account actual parameter regimes. Moreover, the analysis can be generalized to the emergence of the CP assuming the same static quantities but the change in collective-mode damping.

Within the memory-function approach[16,17] the dynamical spin susceptibility can be generally expressed as

$$\chi_{\mathbf{q}}(\omega) = \frac{-\eta_{\mathbf{q}}}{\omega^2 + \omega M_{\mathbf{q}}(\omega) - \omega_{\mathbf{q}}^2}. \quad (1)$$

The spin stiffness $\eta_{\mathbf{q}}$ is the first frequency moment of the dynamical susceptibility $\eta_{\mathbf{q}}=-i\langle[S^z_{-\mathbf{q}},\dot{S}^z_{\mathbf{q}}]\rangle$ and can be within any relevant model expressed in terms of equal-time correlations only, whereas $\omega_{\mathbf{q}}=(\eta_{\mathbf{q}}/\chi^0_{\mathbf{q}})^{1/2}$ is an effective collective-mode frequency where $\chi^0_{\mathbf{q}}=\chi_{\mathbf{q}}(\omega=0)$ is the static susceptibility. Since we have in mind a doped AFM the actual evaluation in relevant models such as the t-J model[16,17] reveals that $\eta_{\mathbf{q}}$ is only weakly **q** dependent for $\mathbf{q}\sim\mathbf{Q}$ whereby $\eta_{\mathbf{q}}\sim\eta\sim2J$.

Form (1) is particularly suited for a description of damped collective modes close to an ordered magnetic state. $M_{\mathbf{q}}(\omega)$ is the (complex) memory-function incorporating information on the collective-mode damping $\gamma_{\mathbf{q}}(\omega) = M_{\mathbf{q}}''(\omega)$. Since here we will be dealing with doped cuprates in the normal state which are (anomalous) paramagnetic metals (PMs), low-frequency collective modes at $\mathbf{q} \sim \mathbf{Q}$ are generally over-damped, $\gamma_{\mathbf{q}} > \omega_{\mathbf{q}}$ consistent with INS results.

Underdoped cuprates close to the AFM phase represent a system of low charge-carrier concentration but large spin fluctuations whose dynamics is quite generally restricted by the sum rule

$$\frac{1}{\pi}\int_0^\infty d\omega \, \text{cth}\frac{\omega}{2T}\chi_{\mathbf{q}}''(\omega) = \langle S_{-\mathbf{q}}^z S_{\mathbf{q}}^z \rangle = C_{\mathbf{q}}. \tag{2}$$

In doped AFM $C_{\mathbf{q}}$ is strongly peaked at $\mathbf{Q}$ with a characteristic width $\kappa_T = 1/\xi$. Moreover, the total sum rule for a system with local magnetic moments (spin 1/2) is given by $(1/N)\Sigma_{\mathbf{q}}C_{\mathbf{q}} = (1 - c_h)/4$, where c_h is an effective hole doping.[17]

While the formalism so far is very general, we now introduce approximations specific to UD.[16] INS experiments listed above indicate that within the normal state the effective $\mathbf{q}$ width of $\chi_{\mathbf{q}}''(\omega)$, i.e., dynamical $\kappa(\omega)$, is only weakly T and ω dependent even on entering the regime with the CP response which implies that $C_{\mathbf{q}}$ is T independent or at least not critical for $T \to 0$. This has also been confirmed by the present authors[16] in numerical investigations of the relevant t-J model, where κ_T has been found to be rather large even for the lowest doping. Since $\eta_{\mathbf{q}} \sim \eta$ is also nearly constant the ω/T scaling and the freezing mechanism must emerge from spin dynamics not reflected in equal-time correlations. Within further analysis we will assume the commensurate AFM response at $\mathbf{Q}$ and the double-Lorentzian form $C_{\mathbf{q}} = C/[(\mathbf{q}-\mathbf{Q})^2 + \kappa_T^2]^2$ close to INS experiments, although qualitative results at low ω do not depend on a particular form of $C_{\mathbf{q}}$.

II. PARAMAGNETIC METAL

Even at rather low doping cuprates behave as PMs, as manifest, e.g., by their metallic resistivity $\rho(T)$ and quasiparticlelike excitations on (the parts of) the Fermi surface as revealed by angle-resolved photoemission spectroscopy. This is underlying our assumption that the (Landau) damping $\gamma_{\mathbf{q}}(\omega)$ is dominated by particle-hole excitations[17] being weakly $\mathbf{q}$ and ω dependent; hence $\gamma_{\mathbf{q}}(\omega) \sim \gamma$, as has been found also in the numerical analysis of model results,[16] whereby a noncritical dependence $\gamma(T)$ can still play some role. We treat γ here as a phenomenological parameter, although it can be estimated from model results[16] or even analytically.[17] It could also be extracted from high-energy INS experiments following the crossover to underdamped modes $\omega_{\mathbf{q}} \sim \gamma$. However, most INS data on UD cuprates are restricted to energies $\omega < 50$ meV being too low to observe the crossover.
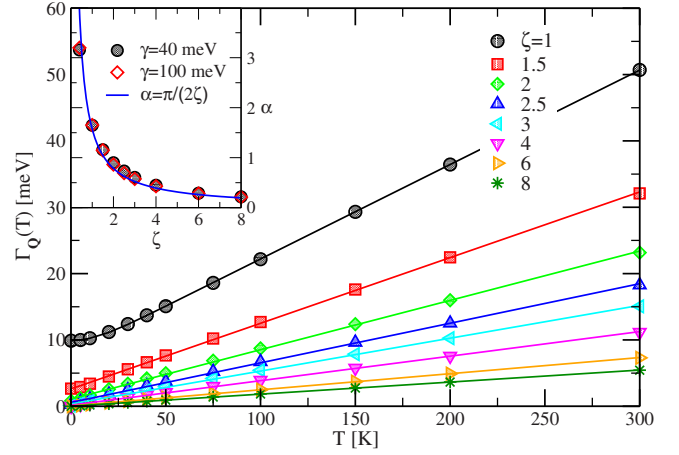
The assumption of constant γ in Eq. (1) leads to

FIG. 1. (Color online) Relaxation rate $\Gamma_{\mathbf{Q}}$ vs T for different parameters ζ and $\gamma = 40$ meV. Inset: dependence of slope parameter α on ζ for two different γ. For comparison, an estimate for α based on Eq. (6) is included.

$$\chi_{\mathbf{q}}''(\omega) = \frac{\eta\gamma\omega}{(\omega^2 - \omega_{\mathbf{q}}^2)^2 + \gamma^2\omega^2}, \tag{3}$$

which in the overdamped regime, $\omega_{\mathbf{q}} < \gamma/2$ and $\omega \ll \gamma$, results in

$$\chi_{\mathbf{q}}''(\omega) \sim \chi_{\mathbf{q}}^0 \frac{\omega\Gamma_{\mathbf{q}}}{(\omega^2 + \Gamma_{\mathbf{q}}^2)}, \quad \Gamma_{\mathbf{q}} = \frac{\eta}{\gamma\chi_{\mathbf{q}}^0}. \tag{4}$$

While such form appears plausible, it can be explicitly tested experimentally. Recent INS data obtained for $\chi_{\mathbf{Q}}''(\omega)$ have been fitted with Eq. (4) (Refs. 5 and 7) to extract $\Gamma_{\mathbf{Q}}(T)$. Indeed, plotting the interdependence of data for $\chi_{\mathbf{Q}}^0$ and $\Gamma_{\mathbf{Q}}$ reported in Ref. 7 (see Fig. 12) shows that $\chi_{\mathbf{Q}}^0\Gamma_{\mathbf{Q}}(T) = \text{const}(= \eta/\gamma)$ to within experimental error over the *whole* span of $\Gamma_{\mathbf{Q}}$, confirming in this way our assumption of constant $\gamma(\omega) \sim \gamma$.

Let us now concentrate on the behavior of $\chi_{\mathbf{Q}}''(\omega)$ as enforced by the sum rule, Eq. (2). It has been noticed that the variation in $\Gamma_{\mathbf{Q}}$ is mainly determined by the parameter

$$\zeta = C_{\mathbf{Q}}\pi\gamma/(2\eta), \tag{5}$$

and only marginally on, e.g., γ provided that we are discussing the regime $T \ll \gamma$ which is experimentally relevant.

In Fig. 1 we first present results for $\Gamma_{\mathbf{Q}}(T)$ in the experimentally relevant regime and units for a range of $\zeta = 1 - 8$. It is evident that for any ζ there exists a finite limiting value $\Gamma_{\mathbf{Q}}^0 = \Gamma_{\mathbf{Q}}(T \to 0)$ that is strongly dependent on ζ, i.e., $\Gamma_{\mathbf{Q}}^0 \sim \gamma \exp(-2\zeta)$.[16] On the other hand, for $T > \Gamma_{\mathbf{Q}}^0$ the variation is nearly linear with $\Gamma_{\mathbf{Q}} \sim \alpha T$ being a manifestation of the ω/T scaling. It is also seen that the linear-in-T dependence of $\Gamma_{\mathbf{Q}}$ extends well beyond the limit $\omega \sim \gamma/2$ provided, of course, that κ_T remains weakly dependent on T in the respective temperature region.

As seen in Fig. 1, the slope α is not universal but depends on ζ as well as on γ, where the latter dependence is very weak (logarithmic), as is also manifest from the collapse of

the data for two values of γ onto a single set (inset of Fig. 1). In fact, from Eqs. (2) and (3) and for $\omega_q < \gamma/2$ one obtains

$$\frac{\Gamma_\mathbf{Q}}{T} \cong \pi \left[2\zeta + \Psi\left(1 + \frac{\Gamma_\mathbf{Q}}{2\pi T}\right) - \ln\frac{\gamma}{2\pi T} \right]^{-1}, \qquad (6)$$

provided that $\gamma \gg 2\pi T$ and $\Psi(\cdot)$ is the digamma function. Hence, in the regime $\Gamma_\mathbf{Q} \sim \alpha T$ and for moderate $\alpha < 2\pi$, we get $\alpha \sim \pi/(2\zeta)$ within leading order, which is seen to agree very well with the exactly calculated values (inset of Fig. 1).

Obtained results can be directly put in relation to INS experiments on UD cuprates. Several results have been recently reported for $\chi_\mathbf{q}''(\omega)$, scanned in both $\mathbf{q}$ and ω for different T. In particular, it has been observed that $\chi_\mathbf{Q}''(\omega)$ can be well described by the simple Lorentzian, Eq. (4), and $\Gamma(T) \sim \alpha T$ has been extracted. For UD YBCO with $x=0.35$ the authors[7] give $\alpha=0.7$, whereas for LSCO that is weakly doped with $x=0.06$ Li scaling is well described by $\alpha=0.18$.[5] Electron-doped PLCCO with doping $x=0.12$ also exhibits scaling[11] with $\alpha=0.44$. It is therefore plausible that different α can be, within our theory, explained with different ζ, Eq. (5). Since $C_\mathbf{Q} \propto 1/\kappa_T^2$ the strongest dependence results from κ_T and experimental results on $\alpha \propto 1/\zeta$ are consistent with the ranking of measured (low-frequency) κ, i.e., $\kappa \sim 0.03$ (in r.l.u.) for YBCO,[7] $\kappa \sim 0.01$ for LSCO,[5] and $\kappa \sim 0.02$ for PLCCO.[10,11] Since η is a robust quantity and rather well known from model calculations, only unknown γ prevents so far more quantitative statements.

The above experimentally deduced α all require quite large ζ; hence one can expect, following Fig. 1, very low saturation $\Gamma_\mathbf{Q}^0$ for $T \to 0$. On the other hand, the mentioned INS data for YBCO and LSCO seem to indicate quite substantial $\Gamma_\mathbf{Q}^0$. However, saturation of $\Gamma_\mathbf{Q}$, setting in for $T < T_g$, is accompanied by the simultaneous appearance of the CP. This effect goes beyond here presented (simple) explanation and requires within the PM the introduction of additional damping, as will be discussed later on.

Recent detailed INS measurements by Hinkov *et al.*[8,9] on UD YBCO with $x=0.45$ allow also for a more complete analysis of $\chi_\mathbf{Q}''(\omega)$ as a function of T. Data normalized at $T=0$, i.e., $\chi_\mathbf{Q}''(\omega,T)/\chi_\mathbf{Q}''(\omega,T=0)$ are presented in Fig. 2 and compared with theoretical ones, following from Eqs. (2) and (3), where the only relevant parameter is ζ. The overall agreement is quite satisfactory and is achieved for $\zeta \simeq 1.8$, implying $\Gamma_\mathbf{Q}^0 \simeq 1.3$ meV. The marked discrepancy between theory and experimental data at $\omega=3$ meV is tentatively associated with a possible saturation of $\Gamma_\mathbf{Q}^0 [\chi_\mathbf{Q}''(\omega,T=0)]$ to a larger (smaller) value than predicted by theory. Namely, a better fit to experimental data based on ζ *and* $\Gamma_\mathbf{q}^0$ as independent parameters yields unchanged ζ but substantially larger $\Gamma_\mathbf{Q}^0 \sim 4$–5 meV, which would correspond to a crossover temperature $T_g \sim \Gamma_\mathbf{Q}^0/\alpha \sim 50$ K. On the other hand, the poor agreement with the 32.5 meV data should be attributed to the breakdown of ω/T scaling since $\omega > \gamma/2$.

Experimental results for $\mathbf{q}$-integrated $\chi_L''(\omega)$ in the same regime also reveal the ω/T scaling and have been several times fitted with the ansatz involving $\tan^{-1}(x)$, as already noted in Sec. I. However, the fit did not find so far a deeper theoretical background. It has been nevertheless shown that
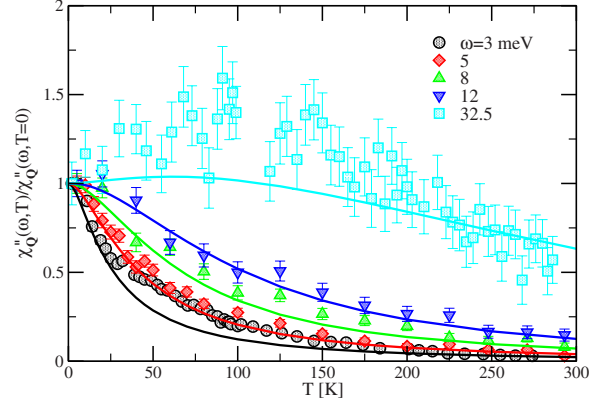

FIG. 2. (Color online) Temperature evolution of $\chi_\mathbf{Q}''(\omega,T)$ for UD YBCO with $x=0.45$ (Ref. 9) (symbols) compared with theoretical result, Eq. (4), with $\zeta=1.8$ and $\gamma=40$ meV (lines).

results of the present theory are quite close to this description.[16] In fact, assuming a Lorentzian dependence of $\chi_\mathbf{q}^0$ on $\mathbf{q}$ and invoking the relation $\Gamma_\mathbf{q} = \eta/(\gamma\chi_\mathbf{q}^0)$, a simple calculation yields $\tan^{-1}(A_1\omega/T)$ with $A_1 \sim 1/\alpha$ provided that $\kappa \ll 1$ but $\kappa^2\chi_\mathbf{Q}^0 \sim$ const. Note, however, that quite generally $\chi_\mathbf{q}^0$ obtained from Eq. (2) decays at least as a double Lorentzian, so that $\chi_L''(\omega,T)$ is a more complicated function, allowing even for nonanalytic behavior in ω for $\zeta \gg 1$.

In Fig. 3 we next present the theoretically obtained data for the scaling function $f(\omega/T)=\chi_L''(\omega,T)/\chi_L''(\omega,T=0)$ for several different ω and the temperature range 0–300 K based on the same set of data for γ and ζ as discussed in relation to Fig. 2. While the ω/T scaling is quite remarkable it breaks down at higher energies, as expected. We also plot the scaling function $f(x)=2/\pi \tan^{-1}(A_1x+A_2x^3)$ often employed in fits to experimental data; the "best-fit" coefficients $A_1 \sim 0.56$ and $A_2 \sim 0.09$ are in reasonable agreement with values $A_1 \sim 0.49$ and $A_2 \sim 0.12$ determined directly from the experimental data.[9]

III. CP RESPONSE

The advantage of the memory-function formalism is that the emergence of the CP at $T < T_g$ in the spin response can be
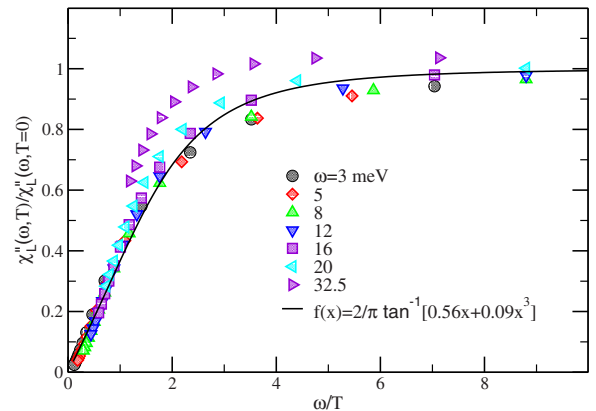

FIG. 3. (Color online) Scaling of theoretical normalized $\chi_L''(\omega,T)$ based on ζ and γ as in Fig. 2. For comparison, the scaling function $f(x)$ as frequently used in fits to experiments is also plotted.

as well treated within the same framework. One has to assume that unlike in a PM the mode damping $\gamma_{\mathbf{q}}$ is not constant but may acquire an additional low-frequency contribution. In particular we can take

$$\widetilde{M}_{\mathbf{q}} \sim i\gamma - \delta^2/(\omega + i\lambda), \qquad (7)$$

with T-dependent δ and λ, which via Eq. (3) leads to $\chi_{\mathbf{q}}''(\omega)$ of the form used also to analyze experimental INS data for YBCO with $x=0.35$.[6] It has been first employed for the analysis of the CP in ferroelectrics,[18] assuming a coupling of a damped mode to a slowly fluctuating object.

For $\lambda \rightarrow 0$ but $\delta^2/\lambda \gg \gamma$ the modified $\widetilde{M}_{\mathbf{q}}$ leads to two distinct energy scales and hence to two contributions to spin dynamics, i.e., the CP part $\chi_{\mathbf{q}}^c(\omega)$ and the regular contribution $\chi_{\mathbf{q}}^r(\omega)$,

$$\chi_{\mathbf{q}}^c(\omega) \sim \frac{\chi_{\mathbf{q}}^0 \Gamma_c}{\Gamma_c - i\omega}, \quad \chi_{\mathbf{q}}^r(\omega) \sim \frac{\chi_{\mathbf{q}}^{r0} \Gamma_r}{\Gamma_r - i\omega}, \quad \mathbf{q} \sim \mathbf{Q}, \qquad (8)$$

valid for $\omega < \lambda$ and $\lambda < \omega \ll \gamma$, respectively. Thus, below T_g the new scales are set by $\Gamma_r = \Omega_{\mathbf{q}}^2/\gamma$ and $\Gamma_c = (\eta/\delta^2)\lambda/\chi_{\mathbf{q}}^0$, with $\chi_{\mathbf{q}}^{r0} = \eta/\Omega_{\mathbf{q}}^2$ and $\Omega_{\mathbf{q}}^2 = \omega_{\mathbf{q}}^2 + \delta^2$. If one assumes that δ saturates at low T, as is manifest by the saturation of Γ_r,[7] $\Gamma_c \propto \lambda/\chi_{\mathbf{q}}^0$ becomes the smallest energy scale, resulting in a quasielastic peak which is, according to Eq. (8), a Lorentzian of width Γ_c, with the static value $\chi_{\mathbf{q}}(\omega \rightarrow 0) = \chi_{\mathbf{q}}^0$, as required by Eq. (1). However, the real part drops for $\omega > \lambda$ to the value $\chi_{\mathbf{q}}^{r0}$; hence the CP contribution is $\Delta\chi_{\mathbf{q}} = \chi_{\mathbf{q}}^0(\delta/\Omega_{\mathbf{q}})^2 \approx \chi_{\mathbf{q}}^0$.

It should be pointed out that within such a formalism a modified $\widetilde{M}_{\mathbf{q}}$ does not (necessarily) induce a change in equal-time correlations as $C_{\mathbf{q}}$, in particular the characteristic low-ω width κ remains unchanged, a fact observed experimentally.[6,7] Moreover, as the CP is too narrow to affect the moments of $\chi_{\mathbf{q}}''(\omega)$, $\eta_{\mathbf{q}}$ is determined solely by the regular part. It is then plausible to assume that $\chi_{\mathbf{q}}''(\omega)$ also remains constant on lowering $T < T_g$, as also observed experimentally in Li-doped LSCO,[5] where below $T_g \sim 50$ K the $\mathbf{q}$-integrated regular part takes the form $\chi_L''(\omega) \propto \omega/(\omega^2 + \Gamma^2)$ with T-independent Γ. The CP contribution then follows from the sum rule

$$T\Delta\chi_{\mathbf{q}} = C_{\mathbf{q}}^c(T) = C_{\mathbf{q}} - C_{\mathbf{q}}^r(T), \qquad (9)$$

with the regular part given by[19]

$$C_{\mathbf{q}}^r(T) = \frac{\eta}{\pi\gamma}\ln\frac{\gamma}{\Gamma_r} + \frac{\pi T}{\gamma\Gamma_r}, \quad T \ll T_g, \qquad (10)$$

again consistent with experiment on $x=0.35$ YBCO,[7] where below $T \sim 50$ K the (integrated) intensity transfers from a broad inelastic scattering to a CP while conserving the (total) sum rule.

From the above considerations it follows that the onset of CP can be explained as a dynamical freezing phenomenon. However, it is evident that the appearance of the anomalous (singular at low ω) damping $\widetilde{M}_{\mathbf{q}}(\omega)$ is not consistent with normal metallic particle-hole excitations. We can speculate that to obtain such a form one has to invoke an inhomogeneous (disordered) system with predominantly localized charge carriers preventing low-ω scattering, resulting in a nearly diverging $\widetilde{M}_{\mathbf{q}}(\omega \rightarrow 0)$. In any case the transition to CP-type dynamics can be associated with a significant softening of λ ($T < T_g$).

In conclusion, we have shown that the approach presented gives a consistent explanation of the ω/T scaling both in $\chi_{\mathbf{q}}''(\omega)$ as well as in $\chi_L''(\omega)$. It is based on two well-established experimental facts: a) the overdamped nature of the response, and b) the saturation of effective inverse correlation length $\kappa(\omega)$ at low ω and T, leading to the anomalous slowing down, i.e., gradual freezing of the dynamic spin response, a phenomenon of entirely quantum origin. The theory can as well account for the INS results in UD cuprates, yielding an explanation for the nonuniversal parameter $\alpha = \Gamma_{\mathbf{Q}}/T$.

The appearance of the CP for $T < T_g$ is easily incorporated into the formalism via the (almost) singular ω- and T-dependent damping $\widetilde{M}_{\mathbf{q}}(\omega)$. However, the question as to the nature of CP, whether of intrinsic origin, based on spin-glass-like arguments, as suggested, e.g., in Ref. 6, or of some other, possibly even extrinsic origin, remains to be settled.

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[19] Note that this estimate follows from Eq. (6) on assuming that $\Gamma_{\mathbf{q}(T)}$ saturates to (almost) T-independent Γ_r below T_g.