

Magnetic Response of Nonmagnetic Impurities in Cuprates

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A theory of the local magnetic response of a nonmagnetic impurity in a doped antiferromagnet, as relevant to the normal-state in cuprates, is presented. It is based on the assumption of the overdamped collective mode in the bulk system and on the evidence that equal-time spin correlations are only weakly renormalized in the vicinity of the impurity. The theory relates the Kondo-like behavior of the local susceptibility to the anomalous temperature dependence of the bulk magnetic susceptibility.

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One of the open theoretical questions in cuprates is the understanding of a well established experimental fact that nonmagnetic impurities have a strong effect on superconducting as well as on the normal-state properties of cuprates [1]. A prominent example is the substitution of Cu in CuO₂ planes by Zn²⁺ which acts as a spinless impurity. In contrast to the naive picture of a weak scatterer, the Zn²⁺ impurity already depresses superconductivity at a small concentration and represents a strong scatterer for transport properties. In this contribution we address the magnetic effects of an impurity in the nonsuperconducting phase. NMR experiments show that Zn induces local magnetic moments on the nearest neighbor (n.n.) copper sites [2], as well as on more distant copper neighbors [3,4]. The observed local susceptibility [5,6] is well accounted for by the Curie-Weiss form $\chi_{\text{loc}} \propto 1/(T + \Theta)$, whereby Θ is independent of the impurity concentration but reveals a clear dependence on doping. Namely, in the underdoped YBa₂Cu₃O_{6+x} (YBCO) the behavior is nearly Curie-like, with $\Theta \sim 0$, whereas in the overdoped YBCO Θ shows a strong increase with doping [7]. In analogy with the Kondo effect in metals Θ has been interpreted as a relevant Kondo temperature. Analogous effects in doped cuprates have been established for a Li⁺ impurity which is also nonmagnetic [7]. By multi-nuclei NMR imaging it has become also increasingly clear that the spatial distribution of the magnetization around the impurity reflects the antiferromagnetic (AFM) correlations of the bulk system [3].

From the point of a theoretical description, it seems well established that a nonmagnetic impurity can be incorporated into a microscopic electronic model of CuO₂ planes in cuprates by introducing, e.g., an inert empty site into the *t*-*J* model relevant to cuprates [8], or a local site with a very different local energy within the Hubbard model [9,10]. Impurity-induced moments and a Curie-like susceptibility have been established in spin ladders [11], in the presence of the gap in the spin excitation spectrum. An analogous treatment, assuming a spin gap in underdoped cuprates, also leads to the appearance

of a Curie-type susceptibility of an unpaired spin [12]. The Kondo effect based on the *t*-*J* model has been used also to explain strong transport scattering on the impurity site [13]. The 2D Hubbard model with impurities has been analyzed using a renormalized random-phase approximation (RPA) [9]. Numerical studies confirmed the existence of an induced Curie susceptibility at low doping [10], whereas variational Monte Carlo simulation established equal-time AFM correlations around the impurity [14].

Nevertheless, the understanding of magnetic effects of such an impurity is far from satisfactory. It is clear that in the normal state of cuprates there is at most a pseudogap in the underdoped regime, whereas at higher doping spin excitations are gapless [12,13]. There is also no explanation for the finite and large Kondo temperature $\Theta > 0$ in the overdoped regime. Lacking also is the theoretical answer to the evident question whether the local magnetic response around the impurity is just the reflection of the bulk (as well anomalous) magnetic response, as evidenced by recent experiments [3].

To address the above issues we present a generalization of the theory of spin response applied to explain the anomalous bulk magnetic response [15]. The novel input is the observation that equal-time spin correlations around the impurity are to a large extent unrenormalized. The consequence is that the local magnetic response is an image of the anomalous staggered susceptibility in a homogeneous system. Consequently, the onset of a finite Kondo scale Θ is related to the doping-driven crossover of the bulk spin dynamics from a non-Fermi-liquid spin dynamics to a Fermi-liquid one [16].

In a homogeneous doped AFM the dynamical spin susceptibility $\tilde{\chi}_{\mathbf{q}}(\omega) = -\langle\langle S_{\mathbf{q}}^z; S_{\mathbf{q}}^z \rangle\rangle_{\omega}$ can be expressed within the memory function formalism as [15]

$$\tilde{\chi}_{\mathbf{q}}(\omega) = \frac{-\eta_{\mathbf{q}}}{\omega^2 + \omega M_{\mathbf{q}}(\omega) - \omega_{\mathbf{q}}^2}, \quad (1)$$

where $\eta_{\mathbf{q}} = -i\langle\langle [S_{-\mathbf{q}}^z, \dot{S}_{\mathbf{q}}^z] \rangle\rangle$, $\omega_{\mathbf{q}}^2 = \eta_{\mathbf{q}}/\chi_{\mathbf{q}}$, and $\chi_{\mathbf{q}} = \tilde{\chi}_{\mathbf{q}}(\omega = 0)$ is the static susceptibility. $\omega_{\mathbf{q}}$ represents the collective-mode frequency in the case of low damping

$\gamma_{\mathbf{q}} \sim M_{\mathbf{q}}''(\omega_{\mathbf{q}}) < \omega_{\mathbf{q}}$. However, in cuprates the collective mode is found to be overdamped throughout the normal phase, i.e., $\gamma_{\mathbf{q}} > \omega_{\mathbf{q}}$. Following the evidence from the analysis of the t - J model [15,17], we assume also constant $\eta_{\mathbf{q}} \sim \eta$ and $\gamma_{\mathbf{q}} \sim \gamma$ in the region of interest near the AFM wave vector $\mathbf{q} \sim \mathbf{Q} = (\pi, \pi)$. η is closely related to energy, so it is quite T independent and only smoothly dependent on doping. In a doped AFM damping γ emerges from the decay of a spin collective mode into electron-hole excitations and is also assumed to remain finite and large at low $T \rightarrow 0$ in the normal state [17]. Hence, the main T variation enters Eq. (1) via $\chi_{\mathbf{q}}$.

The central idea of the theory of the anomalous scaling in doped AFM [15] is that a nontrivial dependence $\chi_{\mathbf{q}}(T)$ is driven by the fluctuation-dissipation relation

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

Within the t - J model there is firm evidence at finite hole concentration $c_h > 0$ [18] that $C_{\mathbf{q}-\mathbf{Q}} \sim C/(\kappa^2 + \tilde{q}^2)$, $\tilde{\mathbf{q}} = \mathbf{q} - \mathbf{Q}$, and that the AFM correlation length $\xi = 1/\kappa$ saturates at low T . Similar conclusions emerge from an analysis of inelastic neutron scattering on YBCO [19,20]. The relation (2) then leads to a strongly T dependent $\chi_{\mathbf{q}-\mathbf{Q}}(T)$, in contrast to the usual Fermi liquid where $\tilde{\chi}_{\mathbf{q}}''(\omega)$ is essentially T independent. We note that there could also be a high-frequency free-fermion-like component in $\tilde{\chi}_{\mathbf{q}}''(\omega)$ contributing to $C_{\mathbf{q}}$. However, from our previous [21] and present improved calculations it follows that $\omega > t$ range contributes less than 10% to the sum rule in the regime of low to optimum doping, i.e., $c_h \leq 0.15$.

Let us first consider the behavior of $\chi_{\mathbf{q}}(T)$. Performing the high- T expansion in Eq. (2) we get

$$C_{\mathbf{q}} \sim T\chi_{\mathbf{q}} + \frac{1}{\pi} \int_0^{\infty} d\omega \frac{\omega}{6T} \tilde{\chi}_{\mathbf{q}}''(\omega) = T\chi_{\mathbf{q}} + \frac{\eta_{\mathbf{q}}}{12T}, \quad (3)$$

where we have taken into account that $\eta_{\mathbf{q}}$ is the second moment of the shape function $\tilde{\chi}_{\mathbf{q}}''(\omega)/\omega$. The high- T expansion is thus consistent with the Curie-Weiss behavior $\chi_{\mathbf{Q}} = C_{\mathbf{Q}}/(T + \Theta_{\mathbf{Q}})$, where $\Theta_{\mathbf{Q}} = \eta_{\mathbf{Q}}/12C_{\mathbf{Q}}$. Since in a doped AFM, as represented, e.g., by the t - J model, $C_{\mathbf{Q}} \propto 1/c_h$ [16] we get quite a small scale $\Theta_{\mathbf{Q}} \propto c_h$. On the other hand, the expansion is valid only for $T > \Theta_{\mathbf{Q}}$. At $T = 0$ we get $\chi_{\mathbf{Q}}(T = 0) = \eta/(\gamma\omega_p)$ [15], where $\omega_p \sim \gamma e^{-2\xi}$ and $\xi = \pi\gamma C_{\mathbf{Q}}/2\eta$.

In Fig. 1 we present $C_{\mathbf{Q}}/\chi_{\mathbf{Q}}(T)$ as follows from the solution of Eqs. (1) and (2) for fixed $\eta = 0.6t$ and $\gamma = 0.5t$ [15] (note that $t \sim 400$ meV for cuprates) but varying $\kappa = 0.5$ – 1.5 (in units of inverse lattice spacing). By way of Eq. (2) κ also determines $\tilde{\kappa}$ which enters the low- ω behavior of $\tilde{\chi}_{\mathbf{q}}''(\omega)$. For the range of κ considered, $\kappa/\tilde{\kappa} \sim 2$ so that $\tilde{\kappa}$ roughly corresponds to the experimental range of values in underdoped to overdoped YBCO [19]. Note that the Curie-Weiss law is obeyed down to $T \sim \Theta_{\mathbf{Q}}$.

$\Theta_{\mathbf{K}} = C_{\mathbf{Q}}/\chi_{\mathbf{Q}}(T = 0)$ can be interpreted as the relevant Kondo temperature and its variation with κ is presented in the inset of Fig. 1. On increasing κ (note that $\kappa \propto \sqrt{c_h}$) we are facing quite an abrupt transition from a Curie behavior at $T \rightarrow 0$ to a Curie-Weiss variation with finite and rapidly increasing $\Theta_{\mathbf{K}} > 0$ [16]. Such a behavior is also consistent with experimental results for $1/\chi_{\mathbf{Q}}(T)$ [16].

Let us turn to a doped AFM with an added nonmagnetic impurity. To be more specific we have in mind the planar t - J model, where the impurity is represented as an empty site at the origin $i = 0$. In analogy with the homogeneous system, Eq. (2), we consider the equal-time local correlations $C_{ij} = \langle S_i^z S_j^z \rangle$ as an essential input. In general, C_{ij} differ from correlations in a homogeneous system without any impurity where $C_{ij}^0 = C^0(\mathbf{R}_j - \mathbf{R}_i)$. However, it appears characteristic for strongly correlated electrons in low-doped AFM that at least shorter-range C_{ij} close to the impurity deviate modestly from unperturbed C_{ij}^0 . The insensitivity of C_{ij} around the impurity can be interpreted as an effective spin-charge decoupling, where the empty site represents just a free spinon [12,13].

In support of the above conjecture we perform an exact-diagonalization calculation of C_{ij} within the t - J model with an empty site. We present in Fig. 2 results for a system $N = 20$ sites at $T = 0$ with $N_h = 2, 3$ mobile holes and $J/t = 0.3$. We show some nonequivalent n.n. and next n.n. correlations C_{ij} around the impurity. We notice that even on sites neighboring the impurity at least shorter-range C_{ij} are nearly the same as in the bulk, or even enhanced [14,22].

It is therefore plausible to treat a system with an impurity by assuming frozen correlations; i.e., we take that $C_{ij} = 0$ for either $i = 0$ or $j = 0$, while $C_{ij} = C_{ij}^0$ elsewhere. Note that at $T = 0$ and in a homogeneous system (with even N, N_h) the ground state is a singlet ($S_{\text{tot}} = 0$). Since $\sum_j C_{ij}^0 = \langle S_i^z S_{\text{tot}}^z \rangle = 0$ this leads to

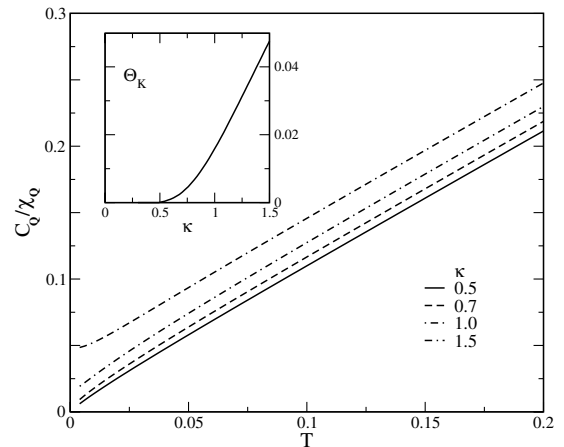


FIG. 1. $C_{\mathbf{Q}}/\chi_{\mathbf{Q}}$ vs T (both in units of t) for various κ . The inset shows the variation of Kondo $\Theta_{\mathbf{K}}$ with κ .

$$\sum_{i,j \neq 0} C_{ij} = \sum_{i,j} C_{ij}^0 - 2 \sum_j C_{ij}^0 + C_{00}^0 = C_{00}^0 = \frac{1}{4} - c_h. \quad (4)$$

We further study the real-space local susceptibilities $\tilde{\chi}_{ij}(\omega) = -\langle\langle S_i^z; S_j^z \rangle\rangle_\omega$ in analogy to the homogeneous case. The dynamical matrix $\tilde{\chi}(\omega)$ can be generally expressed in terms of corresponding matrices in real-space $\underline{\chi}$, $\underline{\eta}$, \underline{M} as

$$\underline{\chi}(\omega) = -[\omega^2 \underline{1} + \omega \underline{M}(\omega) - \underline{\delta}]^{-1} \underline{\eta}, \quad (5)$$

where $\underline{\delta} = \underline{\eta} \underline{\chi}^{-1}$. Again, the local fluctuation-dissipation relation is used to fix $\underline{\chi}$,

$$\frac{1}{\pi} \int_0^\infty d\omega \text{cth} \frac{\omega}{2T} \tilde{\chi}_{ij}''(\omega) = C_{ij}. \quad (6)$$

The next step in an inhomogeneous system is to diagonalize the correlation matrix $C_{ij} = \sum_\lambda C_\lambda (v_\lambda^i)^* v_\lambda^j$, where $\underline{C} \underline{v}_\lambda = C_\lambda \underline{v}_\lambda$. From the sum rule (6) it then follows that one can simultaneously diagonalize the susceptibility matrix $\tilde{\chi}_{ij}(\omega) = \sum_\lambda \tilde{\chi}_\lambda(\omega) (v_\lambda^i)^* v_\lambda^j$ with $\tilde{\chi}_\lambda(\omega) = -\eta_\lambda / (\omega^2 + i\gamma_\lambda \omega - \delta_\lambda)$, where we have assumed the diagonal form of $(M_{ij}, \eta_{ij}) \sim \sum_\lambda (i\gamma_\lambda, \eta_\lambda) (v_\lambda^i)^* v_\lambda^j$. In analogy with the homogeneous system, the local magnetic response around the impurity will be determined by the behavior around $\lambda \sim \Lambda$ for which $C_\Lambda = \max$. In this region we assume constant $\gamma_\lambda \sim \gamma$ and $\eta_\lambda \sim \eta$. Clearly, this is based on the assumption that damping is quite local.

In a system with a nonmagnetic impurity a spin polarization around the impurity induced by a homogeneous magnetic field gives rise to the susceptibility χ_i on site i ,

$$\chi_i = \sum_j \chi_{ij} = \sum_\lambda \chi_\lambda (v_\lambda^i)^* v_\lambda, \quad v_\lambda = \sum_j v_\lambda^j. \quad (7)$$

In the case of a n.n. site χ_i is directly related to the Knight shift on the ^7Li impurity and of ^{89}Y near the Zn impurity

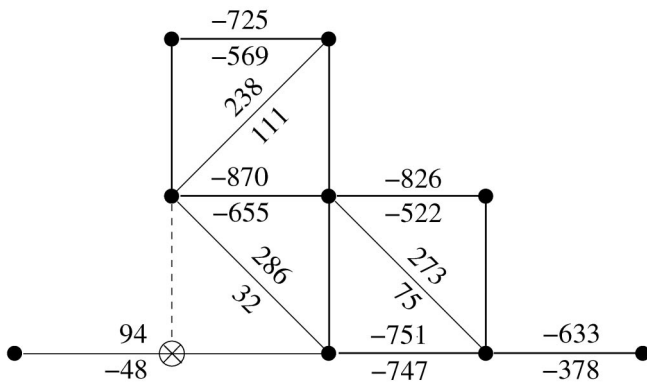


FIG. 2. Equal-time correlations C_{ij} around the impurity (denoted by \otimes) within the t - J model for $J/t = 0.3$ and dopings $c_h = 2/20$ (upper value) and $c_h = 3/20$ (lower value) in units of 10^{-4} .

[9]. Another relevant quantity is the (average) uniform susceptibility $\bar{\chi} = \sum_\lambda \chi_\lambda |v_\lambda|^2 / N$ which yields the impurity-induced contribution $\Delta\chi = \bar{\chi} - \chi_{q=0}^0$.

We first give an approximate solution to the impurity problem via the perturbation calculation. Here, $\underline{C} = \underline{C}^0 + \underline{C}'$ with the perturbative part $C'_{00} = -1/4$ and $C'_{0j} = C'_{j0} = -C_{0j}^0$. The unperturbed eigenvectors are the homogeneous ones, $v_{\mathbf{q}}^i = \exp(i\mathbf{q}\mathbf{r}_i) / \sqrt{N}$, and the lowest order calculation gives

$$\Delta \underline{v}_{\mathbf{q}} = \frac{1}{N} \sum_{\mathbf{q}' \neq \mathbf{q}} \underline{v}_{\mathbf{q}'} \frac{\mu^2 - C_{\mathbf{q}}^0 - C_{\mathbf{q}'}^0}{C_{\mathbf{q}}^0 - C_{\mathbf{q}'}^0}, \quad (8)$$

where $\mu = \sqrt{1/4 - c_h}$ is the effective local moment. The impurity-induced correction to the local susceptibility is

$$\Delta\chi_i = \sum_{\mathbf{q}} [\Delta\chi_{\mathbf{q}} v_{-\mathbf{q}}^i v_{\mathbf{q}} + \chi_{\mathbf{q}}^0 (\Delta v_{-\mathbf{q}}^i v_{\mathbf{q}} + v_{-\mathbf{q}}^i \Delta v_{\mathbf{q}})] \quad (9)$$

and can be expressed as $\Delta\chi_i = \Delta\chi / N + \tilde{\chi}_i$, i.e., $\Delta\chi = N \Delta C_{q=0, q=0} / T = \mu^2 / T$, whereas

$$\tilde{\chi}_i = \sum_{\mathbf{q}} e^{i\mathbf{q}\mathbf{r}_i} \chi_{\mathbf{q}}^0 \left[\frac{\mu^2}{C_{\mathbf{q}}^0} - 1 \right]. \quad (10)$$

Nontrivial is the spatial distribution of $\tilde{\chi}_i$. Since the main contribution in Eq. (10) arises from $\mathbf{q} \sim \mathbf{Q}$ and $C_{\mathbf{Q}}^0 \gg \mu^2$, we obtain from Eq. (10) $\tilde{\chi}_i \sim -\chi_{0i}^0$; i.e., the local response is just the intersite susceptibility of a homogeneous system. Observe that Eq. (10) can as well apply to impurity-induced local magnetism in an undoped AFM insulator [23] since the basic assumption of a local perturbation in C_{ij} remains valid there [22].

In Fig. 3(a) we present results for $1/\tilde{\chi}_1$ for the n.n. site. We use Eq. (10) with $\chi_{\mathbf{q}}^0$ determined via a self-consistent solution to Eqs. (1) and (2). To be consistent with the t - J model on a 2D square lattice we take here the form $C_{\mathbf{q}}^0 = a / (\kappa^2 + \zeta_{\mathbf{q}}) - b$ with $\zeta_{\mathbf{q}} = 2[\cos(qx) + \cos(qy) + 2]$, whereas a, b are chosen such that $C_{q=0}^0 = 0$ and $C_{ii}^0 = 1/2$. As in the case of homogeneous $\chi_{\mathbf{Q}}^0$ in Fig. 1, we notice that the behavior is close to the Curie-Weiss form $1/\tilde{\chi}_1 \propto T + \Theta_1$ with a qualitative transition from $\Theta_1 \sim 0$ for $\kappa < 0.7$ to finite and large Θ_1 for $\kappa > 1$. Deviations from the Curie-Weiss dependence are understandable since Eq. (10) includes contributions from all \mathbf{q} where $\Theta_{\mathbf{q}} > \Theta_{\mathbf{Q}}$. Nevertheless, the behavior at $\mathbf{q} \sim \mathbf{Q}$ is dominant since considered κ are quite large [15].

Alternatively, we can solve the impurity problem as defined by Eqs. (5) and (6) directly, without relying on the perturbation expansion. We first find eigenvalues C_λ and corresponding \underline{v}_λ numerically. For the homogeneous doped system we assume that C_{ij}^0 correspond to the 2D lattice $C_{\mathbf{q}}^0$ as discussed above. The final results for local $\tilde{\chi}_i$, as obtained from Eq. (7) for the same parameters as in the perturbation calculation, are shown in Fig. 3(b). Note that

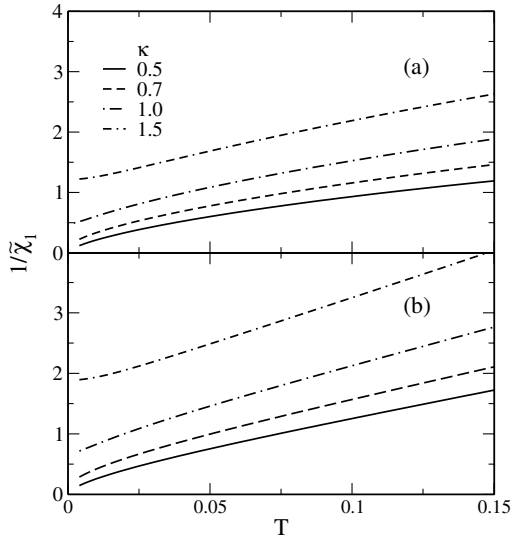


FIG. 3. Local susceptibility $1/\tilde{\chi}_1$ for the site n.n. to the impurity vs T (both in units of t) for different κ : (a) evaluated via the perturbative calculation and (b) with the diagonalization procedure.

results presented in Figs. 3(a) and 3(b) are quite similar, even quantitatively. However, the full calculation yields results even closer to the Curie-Weiss behavior.

Of interest is also the spatial distribution of the local susceptibilities $\tilde{\chi}_i$ around the impurity. It is evident from Eq. (10) that it reflects the bulk χ_{0i}^0 . Moreover, if we consider only the high- T local Curie constant \mathcal{A}_i , it follows from $\chi_{\mathbf{q}} \sim C_{\mathbf{q}}^0/T$ that $\mathcal{A}_i \sim -C_{0i}^0$. Thus \mathcal{A}_i measures the equal-time AFM correlations, changing sign in accordance with the sign of C_{0i}^0 .

In summary, we have presented a theory for the magnetic response of nonmagnetic impurities in doped AFM. It is based on the presented evidence that correlations C_{ij} are not substantially modified in the vicinity of an impurity. The same can be shown for η_{ij} , since the exchange part is directly related to C_{ij} [17]. Note that such a behavior would not apply to a normal Fermi liquid but is rather the consequence of strong correlations; i.e., C_{ij} exhibit a kind of charge-spin separation. Within the present theory the local spin response around the impurity clearly reflects the one in the homogeneous system which is also anomalous.

We have also shown that $\chi_{\mathbf{Q}}$ in a doped AFM follows quite well the Curie-Weiss behavior. Consequently, also local $\tilde{\chi}_i$ exhibit similar behavior, although with some deviations due to contributions from $\mathbf{q} \neq \mathbf{Q}$. From the theory it is clear that \mathcal{A}_i is related to AFM correlations

C_{0i}^0 . Even more important, the transition from the regime with Kondo scale $\Theta_i \sim 0$ to a finite and fast increasing Θ_i reflects the crossover from a non-Fermi liquid to a more normal Fermi-liquid regime.

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