Long-range order and low-energy spectrum of diluted 2D quantum AF

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The problem of diluted two-dimensional (2D) quantum antiferromagnet (AF) on a square lattice is studied using spin-wave theory. The influence of impurities on static and dynamic properties is investigated and a good agreement with experiments and Monte Carlo (MC) data is found. The hydrodynamic description of spin waves breaks down at characteristic wavelengths $\Lambda \gtrsim \exp(\pi/4x)$, $x$ being an impurity concentration, while the order parameter is free from anomalies. We argue that this dichotomy originates from strong scattering of the low-energy excitations in 2D.

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The interest in magnetic properties of high-$T_c$ compounds has been a major driving force of intensive studies of low-dimensional magnetic systems during the last decade [1]. One of such systems is the LA$_2$Cu$_{1-x}$Zn(Mg)$_x$O$_4$, a parental compound doped with static vacancies, which shows greater stability of the AF order against doping than its mobile hole doped counterpart LA$_2$-Sr$_2$CuO$_4$ [2,3]. It represents a fine example of a diluted 2D quantum $S = 1/2$ AF whose properties are the subject of this study. The problem of diluted spin systems has attracted much attention in the past [4,5] and their physics is well understood. The general understanding is that the low-energy excitations of these systems are weakly damped spin waves which belong to the infinite cluster [5] and are well defined up to the percolation threshold. In this work we show that quantum effects and low dimensionality prevent the excitations spectrum to be defined in these hydrodynamic terms [2] at arbitrarily small doping and lead to a paradoxical situation where the long-range order is preserved but the long-wavelength spectrum is not ballistic.

The site-diluted AF on a square lattice is described by:

$$\mathcal{H} = J \sum_{\langle ij \rangle} p_ip_js_i \cdot s_j,$$

with $p_i = 1(0)$ for the magnetic (non-magnetic) site. The pure system at long wavelengths can be described by the nonlinear $\sigma$-model [4]. Its applicability in the presence of a quenched disorder has been questioned [4] since impurities destroy the Lorentz invariance of the model. Some generalizations of the $\sigma$-model have nevertheless been proposed with parameters modified according to MC data [4] and the classical percolation theory [5].

In this work we study the problem of disorder using the $t$-matrix approach combined with a configurational average over the random positions of impurities. It also leads to description of the system in terms of an effective medium with renormalized parameters. In an earlier work Harris and Kirkpatrick [6] have shown that in 2D a non-hydrodynamic term appears in the spin-wave self-energy which explicitly violates Lorentz invariance: $\Sigma_k(\omega) \propto xk(\ln |\omega| - i\pi/2)$. A recent study by Wan et al. [2] has confirmed this result. One can show that the spectrum is overdamped at wavelengths $\Lambda \gtrsim \exp(\pi/4x)$ which implies the existence of a new lengthscale in the system. The absence of a well-defined long-wavelength mode also suggests that the true order should be unstable under an infinitesimal impurity doping, or that $d = 2$ is the upper critical dimension for this problem [6].

In this work we study the effect of impurities on the spectrum, staggered magnetization, $M(x)$, and Néel temperature, $T_N(x)$, to clarify the problem of stability of a (quasi)-2D AF order. We show that neither $M(x)$ nor $T_N(x)$ possesses anomalous contributions which would imply an instability. Thus, we have a somewhat paradoxical situation when the spectrum is ill-defined while the order parameter is not affected. Such a dichotomy comes from the strong influence of disorder in the low-energy excitations in 2D. The averaging procedure, which effectively restores translational invariance, fails to recover the long-wavelength excitation spectrum of this effective medium [4]. The low-frequency modes do exist in some form but they cannot be classified in terms of an effective wave-vector and thus the long-wavelength propagation is entirely diffusive. This scheme implies an existence of a disorder-induced energy scale which restricts the applicability of the continuum approach.

We start with the Hamiltonian (1) in the spin-wave approximation $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_A^{\text{imp}} + \mathcal{H}_B^{\text{imp}}$:

$$\mathcal{H}_0 \simeq 4SJ \sum_k \omega_k (\alpha_k^\dagger \alpha_k + \beta_k^\dagger \beta_k),$$

$$\mathcal{H}_A^{\text{imp}} \simeq -4SJ \sum_{l \in A, k,k'} e^{i(k-k')R_l} \left[ V_{A,k,k'}^\alpha \alpha_l^\dagger \alpha_l + V_{A,k,k'}^\beta \beta_l^\dagger \beta_l + V_{A,k,k'}^{\alpha\beta} (\alpha_k^\dagger \alpha_l^\dagger - h.c.) \right],$$

where the quadratic part of the pure host Hamiltonian $\mathcal{H}_0$ is diagonalized using Bogolyubov transformation: $S_k^A/\sqrt{2S} \simeq \gamma_k = u_k \omega_k + v_k \beta_{-k}^\dagger$, $S_k^B/\sqrt{2S} \simeq \gamma_k = u_k \beta_k + v_k \alpha_{-k}$. $A$ and $B$ denote the sublattices, with $u_k^2 - v_k^2 = 1$, $2u_kv_k \gamma_k = \gamma_k^2/\omega_k^2\gamma_k = (\cos(k_x) + \cos(k_y))/2$, and bare spin-wave frequency $\omega_k = \sqrt{1-\gamma_k^2}$. All momenta belong to the magnetic Brillouin zone $k_x + k_y \leq \pi$. $\mathcal{H}_B^{\text{imp}} = \mathcal{H}_A^{\text{imp}}(A \rightarrow B)$, interactions are
given by $V_{A,i}^{\alpha \alpha} = V_{B,i}^{\beta \beta} = u_1 v_1 + \gamma_1 v_1 u_1 + \gamma_1 u_1 v_1 + \gamma_2 u_2 v_2 + u_1 v_2 + \gamma_2 v_2 u_1 + \gamma_1 v_2 u_1 + \gamma_2 v_2 u_1$. 

\[ V_{A,i}^{\alpha \beta} = V_{B,i}^{\beta \alpha} = x v_1 v_2 + \gamma_1 v_1 u_2 + \gamma_1 u_1 v_2 + \gamma_2 v_2 u_2, \]

\[ V_{A,i,j}^{\alpha \beta} = V_{B,i,j}^{\beta \alpha} = u_1 v_{i,j} + \gamma_1 v_{i,j} u_1 + \gamma_1 u_{i,j} v_1 + \gamma_2 v_{i,j} u_1 + \gamma_2 v_{i,j} u_1, \]

$t$ runs over the impurity sites. As a first step we solve the scattering problem for the single impurity. Since the impurity potential is short-ranged one uses the $t$-matrix approach. The advantage of this method is that the single impurity problem can be solved exactly (within the spin-wave approximation) \[1\]. Therefore, the results of this approach become exact in the dilute limit $x \to 0$.

In our case the square lattice site-defect scatters only in $s-, p_x-, p_y-$, and $d$-symmetric channels and one needs to solve the $t$-matrix for each harmonic. The $t$-matrix equations are:

\[ \Gamma_{A,i}^{\alpha \alpha} = -V_{A,i}^{\alpha \alpha} - V_{A,i}^{\alpha \beta} C_{A,i}^{\alpha \beta} - V_{A,i}^{\beta \alpha} C_{A,i}^{\beta \alpha}, \]

\[ \Gamma_{A,i}^{\alpha \beta} = -V_{A,i}^{\alpha \beta} - V_{A,i}^{\beta \alpha} C_{A,i}^{\alpha \beta} - V_{A,i}^{\beta \beta} C_{A,i}^{\beta \beta}, \]

\[ \text{with } V_{i}^{s}, \text{ where } i = s, p_x, p_y, \text{ and } d \text{-components of } V^s \text{. We suppress } \kappa \text{ and } \omega \text{ in vertex functions } \Gamma_{i}^{\kappa \kappa}, \text{ bare Green's functions } C_{i}^{\kappa \kappa} = C_{i}^{\kappa \kappa}(\kappa, -\omega) = (\omega - \omega_k + i\eta)^{-1}, \text{ and interactions } V_{kk}, \text{ Products VGT involve summation over the internal momenta, all energies are in units of } 4SJ. \text{ An equivalent set of equations gives the vertices } \Gamma_{A,i}^{\alpha \beta}. \text{ Since the impurity potential in each partial wave is separable: } V_{A,i}^{\alpha \beta} = u_{k} \omega k u_{k'} + \phi_{k}, V_{A,i}^{\alpha \beta} = \phi_{k}, \omega k u_{k'} + \phi_{k'}, V_{A,i}^{\alpha \beta} = \phi_{k}, \omega k u_{k} + \phi_{k}, \omega k u_{k'}, \phi_{k}, \omega k u_{k} + \phi_{k}, \omega k u_{k'}, \text{ and } V_{A,i}^{\alpha \beta} = 1/(2\pi) \ln((\omega - \omega_k)/4 - i), \text{ Eqs. (4) can be readily solved: } \Gamma_{A,i}^{\alpha \beta}(k, \kappa, \omega) = -V_{A,i}^{\alpha \beta}(k, \kappa, \omega) \hat{\Gamma}_{i}^{\kappa \kappa}(\kappa, \omega), \]

\[ \hat{\Gamma}_{i}^{\kappa \kappa}(\omega) = -1/\omega - (1 + \omega)p(\omega)/[1 - \omega(1 + \omega)p(\omega)], \]

\[ \hat{\Gamma}_{p}^{\kappa \kappa}(\omega) = 2/[1 + \omega + (1 - \omega)\omega^2(\omega - \rho_d)/2], \]

\[ \hat{\Gamma}_{\rho}^{\kappa \kappa}(\omega) = 1/[1 + (1 - \omega)\rho_d(\omega)], \]

with $\rho = \sum_{p} 1/(\omega^2 - \omega_p^2)$ and $\rho_d = \sum_{p} (\gamma_p^2)/(\omega^2 - \omega_p^2)$, which can be expressed through the complete elliptic integrals \[2\]. For the impurity in $B$ sublattice $\Gamma_{B,i}^{\alpha \beta} = -V_{B,i}^{\alpha \beta} \hat{\Gamma}_{i}^{\kappa \kappa}(\omega)$. The $s$-wave scattering \[6\] reveals a zero-frequency mode which originates from the oscillations of the fictitious degrees of freedom at the impurity site. Roughly speaking, since in the spin-wave approximation spins are quantized through bosons even if $S^z$ is set to zero there is still $\omega_0 = 0$ in $S^z$ (for discussion see Ref. \[1\]). This gives rise to the unphysical zero-frequency mode which has to be projected out. We do so by introducing magnetic fields at the impurity sites (similar to Refs. \[13\]) $\Delta H = H_z \sum_{i} a_i^\dagger a_i$. Within this approach after straightforward algebra \[13\] in the limit $H_z \to \infty$ one obtains: $\Gamma_{A,i}^{\alpha \beta}(k, \kappa, \omega) = -V_{A,i}^{\alpha \beta}(k, \kappa) \hat{\Gamma}_{s}^{\kappa \kappa}(\omega) + \Delta \Gamma_{A,s}^{\alpha \beta}(k, \kappa, \omega), \]

\[ \hat{\Gamma}_{s}^{\kappa \kappa}(\omega) = -(1 + \omega)p(\omega)/[1 - \omega(1 + \omega)p(\omega)], \]

which is free from the zero-frequency pole. $\Delta \Gamma_{A,s}^{\alpha \beta}(k, \kappa, \omega) = \omega_k \omega_k - \omega_k, \Delta \Gamma_{A,s}^{\alpha \beta}(k, \kappa, \omega) = -\omega_k \omega_k - \omega_k, \Delta \Gamma_{B,s}^{\alpha \beta}(k, \kappa, \omega) = \Delta \Gamma_{A,s}^{\alpha \beta}(k, \kappa, \omega)$. Therefore, the results of this approach become exact in the dilute limit $x \to 0$.
the expansion in $x$. One can also show that the summation of the Green’s function series and dressing of the inner lines in self-energies using self-consistent equation on $\rho(\omega)$ and $G^{\sigma\sigma}$ actually enhance the anomaly.

In a complementary problem of a quasi-2D AF a small inter-plane coupling $\alpha = J_1/J$ cuts off the log-singularity: $\rho_{3D}(\omega) \approx (1/\pi) \ln(\alpha/32) + i\mathcal{O}(\omega) \text{ at } \omega < \sqrt{2}\alpha$. Therefore a “safe” range of concentrations $x < x^* \sim \ln^{-1}(1/\alpha)$ can be found where the long-wavelength quasiparticles are still well defined deep in the 3D region of the $k$-space ($\Lambda^{-1} < \sqrt{\alpha}$). However, one should be able to observe a nonlinearity of the spectrum and an abnormal damping of the spin waves in the 2D long-wavelength region ($\sqrt{\alpha} < \Lambda^{-1} < 1$) similar to the quasi-1D problem.

For the real materials $\alpha \sim 10^{-4}$ giving $x^* \sim 20\%$. Above the concentration $x^*$ all the low-energy excitations are incoherent because the 2D disorder-induced scale $\omega_0^{-1}$ is shorter than the 3D length $1/\sqrt{\alpha}$ so the spin waves lose coherence before they can propagate in 3D. The same consideration applies to the case of small anisotropies introducing gaps in the spectrum with a modified $\alpha = \alpha_{\text{eff}}$ accumulating the total effect of the gaps and 3D coupling. It should be noted that the incoherence comes from the averaging procedure which converts the dissipation of momentum into the dissipation of the energy. Therefore, the overdamped excitations should be understood as diffusive. It is interesting that it requires 2D and “strong” disorder to restrict the number of Euclidean paths for spin waves and to break down the description of the problem in terms of an effective medium.

Now we proceed with quantities whose expansion in $x$ can be shown to be reliable. Staggered magnetization at the magnetic site $M(x) = M_0 - \delta M(x)$, $M_0 = S - \delta \lambda$, 

$$
\delta M(x) = \sum_k [(\alpha^\dagger_k \alpha_k) - \gamma_k (\sigma^\dagger_k \sigma_k)]/\omega_k = \sum_k \int_{-\infty}^{\infty} \frac{n(\omega) d\omega}{\pi \omega_k} \text{Im} [G_{R,k}^{\sigma\sigma}(\omega) - \gamma_k G_{R,k}^{\sigma\sigma}(\omega)],
$$

where $\delta \lambda = \sum_k \frac{\alpha^2_k}{\omega_k} \simeq 0.1966$ is from zero-point fluctuations, $n(\omega) = [e^{\omega/\beta} - 1]^{-1}$ is the Bose occupation number, index $R$ denotes retarded. At $T = 0$ $n(\omega) = -\theta(-\omega)$ and integrals in (11) with $G^{\sigma\sigma}$ from (9) can be taken numerically. It can be shown that the expansion in $x$ for $G^{\sigma\sigma}$ can be performed before the integration $G^{\sigma\sigma} \approx G_0^{\sigma\sigma} + G_0^{\sigma\sigma} \Sigma^{\sigma\sigma} G_0^{\sigma\sigma}$, $G^{\sigma\sigma} \approx G_0^{\sigma\sigma} \Sigma^{\sigma\sigma} G_0^{\sigma\sigma}$ and that all integrals for the linear in $x$ term are convergent giving $\delta M(x) = 0.209(8) \cdot x + \mathcal{O}(x^2)$. For $S = 1/2$ $M(x)/M_0 \simeq 1 - 0.691(5) \cdot x$. This linear slope together with the results of calculations using $G^{\sigma\sigma}$ from Eq. (9), MC [10] and NMR data [17] are shown in Fig. 2. Note that Fig. 2 and Eq. (10) are showing the reduction of the magnetic moment by the quantum fluctuations induced by impurities. In order to extract the same quantity from the MC data [10], which are averaged over all sites, one needs to deduce a probability of finding a spin-occupied site from them. One can see a very good agreement of our results with numerical data up to high concentrations.

The oxidation of the crystals can be the reason of different behavior is $T_N$ for quasi-2D problem. The thermal corrections to the staggered magnetization $\delta M_T \sim T \ln(T/\omega)\big|_{\omega = \sqrt{\alpha}}$ yield a finite $T_N \sim \ln(1/\alpha)^{-1}$ for a pure system. Since the original spectrum is bent from the linear form this should manifest itself in the finite temperature part of Eq. (10) as a stronger divergency. One indeed finds such corrections in Eq. (10) ($\sim T^2 \ln^2(\omega)$) already in a perturbative limit ($x \ln |\omega| / \omega \ll 1$). However, these anomalous terms from diagonal and off-diagonal parts cancel each other. From the mean-field equation $M_{TN}(x) = 0$ after some algebra one obtains (at $\alpha \rightarrow 0$): $T_N(x)/T_N(0) \simeq 1 - xA$ with $A = \pi - 2/\pi + 0.209(8)/M_0$. For $S = 1/2$, $A = 3.196(5)$ [18], which fits very well experimental data [13,21], see Fig. 3.

Thus the order is preserved up to a high $x$ value [13] while the long-wavelength spectrum is not well defined at any $x$. Experimentally one would need to probe directly the spin-wave spectrum close to the AF ordering vector and look for anomalous broadening and nonlinearity. The actual overdamped part of the spectrum is associated with a small energy scale (and small momenta, e.g. at $x = 0.2$, $k_0 \sim 0.01\pi/a$) and it is not clear if it is observable within the range of doping where conclusions of this work are reliable. The observed deviation from the simple exponential behavior of the correlation length $\xi(T, x)$ v.s. $1/T$ [13] might be related to the character of the low-energy spectrum we discuss.

Another aspect relates our problem to the other problems of disorder in 2D. We observe that some weight from every mode is transferred to the low energies $\omega \sim \omega_0$ which can be understood as some sort of localization, though the localization criteria for this problem is unclear. Altogether these low-energy states result in a peak which can be understood as some sort of localization, while the long-wavelength spectrum is not well defined for every mode is transferred to the low energies $\omega \sim \omega_0$ which can be understood as some sort of localization, though the localization criteria for this problem is unclear. Altogether these low-energy states result in a peak which can be understood as some sort of localization, while the long-wavelength spectrum is not well defined for every mode is transferred to the low energies $\omega \sim \omega_0$ which can be understood as some sort of localization, though the localization criteria for this problem is unclear. Altogether these low-energy states result in a peak which can be understood as some sort of localization, while the long-wavelength spectrum is not well defined for every mode is transferred to the low energies $\omega \sim \omega_0$ which can be understood as some sort of localization, though the localization criteria for this problem is unclear. Altogether these low-energy states result in a peak which can be understood as some sort of localization, while the long-wavelength spectrum is not well defined.
doped with static vacancies. At the same time the long-wavelength excitation spectrum is shown to be diffusive for any value of doping, restricting a hydrodynamic approach to the problem from low energies.

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FIG. 1. Belyaev diagram series for the diagonal, $G^\alpha\alpha$, and off-diagonal, $G^\alpha\beta$, Green’s functions. Self-energies $\Sigma^\alpha\alpha$ (circle) and $\Sigma^\alpha\beta$ (square) are the configurational averages of $\Gamma^\alpha\alpha$ and $\Gamma^\alpha\beta$, respectively.

FIG. 2. Left axes: reduction of the local magnetic moment by the quantum fluctuations induced by impurities $M(x)/M(0)$ from NMR (•, Ref. [1]) and MC (○, Ref. [4]) studies. Lines show our results from Eq. (10). Right axes: the absolute value of $\delta M(x)$ from Eq. (10) (lines) and MC data, Ref. [10], for $S = 1/2$ (○) and $S = 1$ (●).

FIG. 3. Néel temperature v.s. $x$ from $\mu$SR and susceptibility measurements Refs. [17,24] for the La$_2$Cu$_{1-x}$Zn(Mg)$_x$O$_4$, and the linear slope from our theory.