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10 August 1999; accepted 21 September 1999

REPORTS

Quantum Impurity in a Nearly Critical Two-Dimensional Antiferromagnet

Subir Sachdev,* Chiranjeeb Buragohain, Matthias Vojta

The spin dynamics of an arbitrary localized impurity in an insulating two-dimensional antiferromagnet, across the host transition from a paramagnet with a spin gap to a Néel state, is described. The impurity spin susceptibility has a Curie-like divergence at the quantum-critical coupling, but with a universal effective spin that is neither an integer nor a half-odd integer. In the Néel state, the transverse impurity susceptibility is a universal number divided by the host spin stiffness (which determines the energy cost to slow twists in the orientation of the Néel order). These and numerous other results for the thermodynamics, Knight shift, and magnon damping have important applications in experiments on layered transition metal oxides.

The recent growth in the study of quasi–two-dimensional transition metal oxide compounds (*1*) with a paramagnetic ground state and an energy gap to all excitations with a nonzero spin (the "spin-gap" compounds such as $SrCu_2O_3$, $CuGeO_3$, and NaV_2O_5) has led to fundamental advances in our understanding of low-dimensional, strongly correlated electronic systems. These systems are insulators and thus are not as complicated as the cuprate high-temperature superconductors (which display a plethora of phases with competing magnetic, charge, and superconducting orders); this simplicity has exposed the novel characteristics of the collective quantum spin dynamics.

One of the most elegant probes of these spin-gap compounds is their response to intentional doping by nonmagnetic impurities, such as Zn or Li, at the location of the magnetic ions. Such experiments were initially undertaken on the cuprate superconductors (*2, 3*), but their analogs in the insulating spin-gap compounds have proved to be a fruitful line of investigation (*4*). They have demonstrated a remarkable property of the paramagnetic ground state of the host compound: Each nonmagnetic impurity has a net magnetic moment of spin ½ located in its vicinity (for the case in which the host compound has magnetic ions with spin ½). The confinement of spin is a fundamental defining property of the host paramagnet and is a key characterization of the quantum-coherent manner in which the host spins form a many-body, spin zero ground state; this confining property was predicted theoretically (*2, 5*) for the paramagnetic states of a large class of two-dimensional antiferromagnets.

We describe here the quantum theory of an arbitrary localized deformation in such antiferromagnets; examples of deformations are (i) a single nonmagnetic impurity, along with changes in the values of nearby exchange interactions, and (ii) a change in sign of a localized group of exchange interactions from antiferromagnetic to ferromagnetic. Our main concern is the behavior of the impurity as the host antiferromagnet undergoes a bulk quantum phase transition from a paramagnet to a magnetically ordered Néel state; we show that the spin dynamics of any deformation is universally determined by a single number—an integer or half-odd integer valued spin S.

Apart from applications to experiments on materials intentionally driven across a quantum phase transition, our results also lead to new insights and predictions about the behavior of impurities in existing spin-gap compounds. The traditional view of the spin-gap paramagnet is based on strong local singlet formation between nearest-neighbor spins (Fig. 1A); the resulting picture of doping by a nonmagnetic impurity is that the partner spin of the impurity site is essentially free. To obtain any nontrivial dynamics, one performs an expansion about such a decoupled limit, and this yields simple localized spin behavior with nonuniversal details, depending on the specific microscopic couplings. In practice, however, spin-gap systems are usually well away from the local singlet regime, and strong resonance between different singlet pairings leads to appreciable spin correlation lengths: Their spin gap, Δ, is significantly smaller than J, a typical nearest-neighbor exchange. A systematic and controlled approach for analyzing such a fluctuating singlet state, which we advocate here, is to find a quantum-critical point to a magnetically ordered state somewhere in parameter space and then to expand away from it into the spin-gap state. The coupling between the bulk and impurity excitations becomes universal in such an expansion, and all dynam-

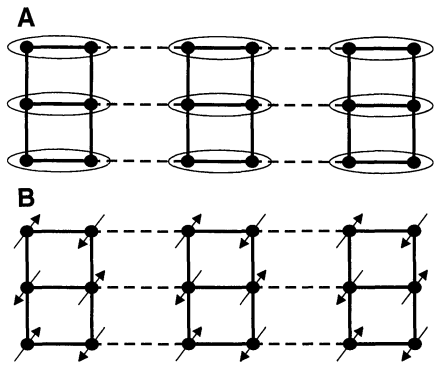

Fig. 1. The coupled-ladder antiferromagnet. The *A* links are solid lines and have exchange J; the *B* links are dashed lines and have exchange λJ. The paramagnetic ground state for $\lambda < \lambda_c$ is schematically indicated in (**A**): The ellipses represents a singlet valence bond, $(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle)/\sqrt{2}$ between the spins on the sites. The Néel ground state for $\lambda > \lambda_c$ appears in (**B**).

Department of Physics, Yale University, Post Office Box 208120, New Haven, CT 06520–8120, USA.

*To whom correspondence should be addressed. E-mail: subir.sachdev@yale.edu

ical properties depend only on the bulk parameters, Δ and a velocity c.

For clarity, we state our main results in the context of a simple, explicit theoretical model; however, they are more general and apply quantitatively to a broad class of experimentally realizable systems. We begin by reviewing the properties of the regular antiferromagnet described by the Hamiltonian (*6, 7*)

$$H = J\sum_{i,j\in A} \mathbf{S}_i \cdot \mathbf{S}_j + \lambda J \sum_{i,j\in B} \mathbf{S}_i \cdot \mathbf{S}_j \qquad (1)$$

where $\mathbf{S}_i$ are spin-½ operators on the sites of the coupled-ladder lattice shown in Fig. 1, with the A links forming two-leg ladders and the B links coupling the ladders. The ground state of H depends only on the dimensionless coupling λ, and we restrict our attention to $J > 0$, $0 \le \lambda \le 1$. At $\lambda = 0$, the ladders are decoupled, and each forms a spin singlet quantum paramagnet (Fig. 1A). This paramagnetic state continues adiabatically for small nonzero λ until the quantum-critical coupling $\lambda = \lambda_c \approx 0.3$, where the spin gap vanishes as $\Delta \sim (\lambda_c - \lambda)^{\nu}$, where ν is a known exponent (*7*) (the symbol $\sim$ indicates that the two quantities are asymptotically proportional). For $\lambda > \lambda_c$, the ground state has long-range Néel order (Fig. 1B) characterized by the nonzero spin stiffnesses ρ_{sx} and ρ_{sy}, which determine the energy cost of twists in the order parameter orientation in the x and y directions [we also define $\rho_s \equiv (\rho_{sx}\rho_{sy})^{1/2}$]. The low-lying excitations above the Néel state are spin waves that travel with velocities c_x and c_y in the x and y directions [with $c_x^2/c_y^2 = \rho_{sx}/\rho_{sy}$; we define $c \equiv (c_x c_y)^{1/2}$]. As λ approaches the critical value λ_c from above, all the stiffnesses vanish as $(\lambda - \lambda_c)^{\nu}$, whereas the velocities remain finite and noncritical.

Introducing a nonmagnetic impurity in H by removing the spin at site $i = X$ (Fig. 2), the modified Hamiltonian H_X has the same form as H, but all links connected to site X do not appear in the sums in Eq. 1. The system can be probed by examining its total linear susceptibility (χ) to a uniform magnetic field $\mathbf{H}$ (under which the Hamiltonian becomes $H_X - g\mu_B \Sigma_{i\neq X} \mathbf{H} \cdot \mathbf{S}_i$, where μ_B is the Bohr magneton and g is the gyromagnetic ratio of the ion). This susceptibility may be written as $\chi = (g\mu_B)^2(\mathbf{A}\chi_b + \chi_{\rm imp})$ where $\mathbf{A}$ is the total area of the antiferromagnet, χ_b is the bulk response per unit area of the antiferromagnet without the impurity, and $\chi_{\rm imp}$ is the additional contribution due to the nonmagnetic impurity. We will now describe the behaviors of χ_b and $\chi_{\rm imp}$ as the temperature T approaches 0 ($T \to 0$) and λ moves across λ_c.

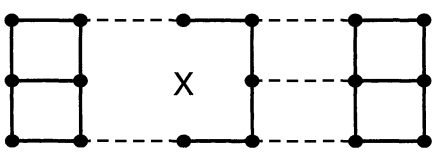

Fig. 2. The impurity Hamiltonian H_X in which the spin and links on site $i = X$ have been removed.

In the quantum paramagnet, $\lambda < \lambda_c$, the presence of the spin gap implies that the bulk response is exponentially small, $\chi_b = (\Delta/\pi\hbar^2c^2)e^{-\Delta/k_BT}$ (*7*). The confinement of a magnetic moment in the vicinity of the impurity site implies that there will be a Curie-like contribution, and so

$$\chi_{\rm imp} = \frac{S(S+1)}{3k_BT} \qquad (2)$$

where $S = $ ½ for the model under consideration here (*8*); for a general local deformation, we consider Eq. 2 as the definition of the value of S, which, naturally, must be an integer or a half-odd integer. These expressions for χ_b and $\chi_{\rm imp}$ are exact as $T \to 0$ for all $0 < \lambda < \lambda_c$. Another way of characterizing the confinement of the magnetic moment near X is by looking at the time autocorrelation function of a spin at a site $i = Y$ close to X (say, its nearest neighbor); at $T = 0$, this obeys

$$\lim_{\tau\to\infty} \langle \mathbf{S}_Y(\tau) \cdot \mathbf{S}_Y(0)\rangle = m_Y^2 \neq 0, \qquad (3)$$

where τ is imaginary time and m_Y is the local remnant magnetic moment on site Y, which is usually significantly smaller than the total impurity moment S appearing in Eq. 2.

Next, we turn to the behavior as $T \to 0$ at the critical point $\lambda = \lambda_c$ [more generally, the $T > 0$ results here will apply for $\Delta < T < J$ ($\rho_s < T < J$) for $\lambda < \lambda_c$ ($\lambda > \lambda_c$)]. We expect that as the spin gap in the quantum paramagnet disappears, the bulk magnon excitations will proliferate and their screening will eventually quench the impurity moment, so m_Y approaches 0 as λ approaches λ_c from below. We can anticipate a power-law decay of the spin autocorrelations (*9–11*), with

$$\langle \mathbf{S}_Y(\tau) \cdot \mathbf{S}_Y(0)\rangle \sim 1/\tau^{\eta'} \qquad (4)$$

for large τ, $T = 0$, and $\lambda = \lambda_c$; and our result for the new universal exponent η' is given below. Standard scaling arguments also imply that m_Y vanishes as $m_Y \sim (\lambda_c - \lambda)^{\eta'\nu/2}$. The behavior at the critical point therefore appears analogous to that in the overscreened multichannel Kondo problem (*12, 13*); in that case, the impurity spin is screened by a bath of conduction electrons carrying multiple "flavors" and also exhibits a power-law decay in its autocorrelation. Furthermore, in the multichannel Kondo case, the T dependence of $\chi_{\rm imp}$ is given essentially by the Fourier transform of Eq. 4; that is, by $\chi_{\rm imp} \sim T^{-1+\eta'}$ (*13*). This result is a consequence of a compensation effect (*14*) because the magnetic response of the screening cloud of conduction electrons is negligible: The local Fermi levels of up and down electrons adjust themselves to the local magnetic field, and hence the susceptibility is not very different from the bulk susceptibility except in the immediate vicinity of the impurity spin (*15*). In more technical terms, $\chi_{\rm imp}$ vanishes in the strict continuum limit, and corrections to scaling have to be considered, which lead eventually to $\chi_{\rm imp} \sim T^{-1+\eta'}$. Our computations show that the behavior of H_X at $\lambda = \lambda_c$ is dramatically different: The magnon excitations do not have an exact compensation property, and their response is nonzero already in the scaling limit. So in a sense, the present problem is simpler than the overscreened Kondo case, and naïve scaling arguments always work, without inclusion of irrelevant operators. The scaling dimension of χ is that of inverse energy (*7*), and so we have one of our central results

$$\chi_{\rm imp} = \frac{C_1}{k_BT} \qquad (5)$$

at $\lambda = \lambda_c$, where C_1 is a universal number independent of microscopic details (as are all the C_i introduced below). We computed C_1 in the expansion in $\epsilon = 3 - d$, where d is the spatial dimension, and obtained

$$C_1 = \frac{S(S+1)}{3}\left[1 + \left(\frac{33\epsilon}{40}\right)^{1/2} - \frac{7\epsilon}{4} + O(\epsilon^{3/2})\right] \qquad (6)$$

The omitted higher order corrections in Eq. 6 will, in general, depend on S. Comparing with Eq. 2, we can define an effective impurity spin $S_{\rm eff}$ at the quantum-critical point by $C_1 = S_{\rm eff}(S_{\rm eff} + 1)/3$; it is evident that $S_{\rm eff}$ is a universal function of S, is neither an integer nor a half-odd integer, and is almost certainly irrational at $\epsilon = 1$. The leading corrections in the ϵ expansion are quite large, and this is a feature of all the results obtained below; accurate numerical estimates require some resummation scheme, but we do not discuss this here. For completeness, we note that at $\lambda = \lambda_c$, the bulk response (*16*) $\chi_b = C_2(k_BT)/(\hbar c)^2$, a T dependence that is also different from the bulk response in the overscreened Kondo problem.

Finally, we describe the situation for $\lambda > \lambda_c$. The presence of Néel order at $T = 0$ implies that the response is anisotropic. Parallel to the Néel order, there is a total magnetic moment quantized precisely at S (*8*), and this does not vary under a small longitudinal field (there is also a staggered local moment in zero field, as defined by Eq. 3, which obeys $m_Y \sim |\lambda - \lambda_c|^{\eta'\nu/2}$). Orthogonal to the Néel order, there is a linear response to a transverse field, $\chi_\perp$. For the bulk response, we have the well-known result that $\chi_{b\perp}$ is proportional to the spin stiffness, $\chi_{b\perp} = \rho_s/(\hbar c)^2$. In contrast, the same scaling arguments leading to Eq. 5 imply that $\chi_{{\rm imp}\perp}$ is inversely proportional to ρ_s, the

latter being the only energy scale characterizing the ground state as λ approaches λ_c from above; so another key result is

$$\chi_{\mathrm{imp}\perp} = \frac{C_3}{\rho_s} \tag{7}$$

In general d, this relationship is $\chi_{\mathrm{imp}\perp} = C_3(\hbar c)^{(2-d)/(d-1)}/\rho_s^{1/(d-1)}$, and the ϵ expansion of C_3 is

$$C_3 = \frac{15S}{\sqrt{22}}\left(\frac{11S_{d+1}}{2\epsilon}\right)^{1/(d-1)}[1 - (0.937 + 1.390S + 1.256S^2)\epsilon + O(\epsilon^2)] \tag{8}$$

where $S_d = 2/[\Gamma(d/2)(4\pi)^{d/2}]$. ρ_s vanishes, and so $\chi_{\mathrm{imp}\perp}$ diverges, as λ approaches λ_c. Turning to $T > 0$ but very small, in $d = 2$ and in the absence of any spin anisotropy, strong angular fluctuations cause the Néel order to vanish at any nonzero T. Then the susceptibility takes the rotationally averaged value $\chi_{\mathrm{imp}} = S^2/(3k_BT) + (2/3)\chi_{\mathrm{imp}\perp}$, where the first term is the contribution of the net moment noted earlier [this term has a coefficient S^2 and not $S(S + 1)$, because the locking of the moment orientation to the local Néel order makes it behave classically]. In practice, this averaged χ_{imp} will not be observable as even an extremely small anisotropy will pin the Néel order below a small $T > 0$. Our results for χ are summarized in Fig. 3.

The next two paragraphs outline the field-theoretic derivation of the results above—details appear elsewhere (*17*). We describe the bulk-ordering transition by a $d + 1$-dimensional field theory with action S_b of a field $\phi_\alpha(x,\tau)$ ($\alpha = 1\ldots3$) representing the collinear Néel order parameter (*7*). This is coupled by the action S_{imp} to an impurity spin at $x = 0$ with orientation given by the unit vector n_α. The partition function is $\int D\phi(x,\tau)Dn(\tau)\exp(-S_b - S_{\mathrm{imp}})$ with

$$S_b = \int d^dx d\tau\left[\frac{1}{2}[(\nabla_x\phi_\alpha)^2 + c^2(\partial_\tau\phi_\alpha)^2 + r\phi_\alpha^2] + \frac{g_0}{4!}(\phi_\alpha^2)^2\right] \tag{9}$$

$$S_{\mathrm{imp}} = \int d\tau\left[iSA_\alpha(n)\frac{dn_\alpha}{d\tau} - \gamma_0 S n_\alpha(\tau)\phi_\alpha(x = 0,\tau)\right] \tag{10}$$

where $\epsilon_{\alpha\beta\gamma}\partial A_\beta/\partial n_\gamma = n_\alpha$, and the term proportional to $A(n)$ is a Wess-Zumino form representing the Berry phase of the impurity spin. The bulk transition in S_b is driven by tuning the coupling r through a critical value r_c, which therefore plays a role similar to λ; the $\lambda < \lambda_c$ ($\lambda > \lambda_c$) region of the lattice antiferromagnet H maps onto the $r > r_c$ ($r < r_c$) region of the field theory S_b. Quite generally, any local deformation of the antiferromagnet is described by the action $S_b + S_{\mathrm{imp}}$, where S, defined as the integer or half-odd integer appearing in Eq. 2, is (roughly) the net local imbalance of spin between the two sublattices. Changes in exchange constants lead to additional terms such as $\int d\tau\phi_\alpha^2(x = 0,\tau)$ which are all strongly irrelevant under the renormalization group (RG) analysis in powers of ϵ. The $r = 0$, $g_0 = 0$ case of Eqs. 9 and 10 was considered earlier by Sengupta (*10*) [and related models in (*9, 11*)] in a nonlocal formulation in which $\phi_\alpha(x \neq 0,\tau)$ was integrated out. However, such a model has a pathological response to even an infinitesimal field **H** (the energy is unbounded below), and the quartic g_0 coupling is essential to stabilize the system and to all the results obtained here. Further, the local formulation here facilitates development of the RG to all orders.

The RG analysis of $S_b + S_{\mathrm{imp}}$ is carried out by the methods of boundary-critical phenomena (*18*) of a $(d + 1)$–dimensional system with a 1-dimensional boundary at $x = 0$, which constitutes a dimensional reduction of $d > 1$ [contrast this with the case of a $(d + 1)$–dimensional system with a d-dimensional boundary, with a dimensional reduction of 1, which has been invariably (*13, 19*) considered earlier, as in all the Kondo problems]. The irrelevance of the boundary "mass" term $\phi_\alpha^2(x = 0,\tau)$ implies that there is only an ordinary transition at the position of the bulk critical point (*20*) (this has been implicit in our earlier discussion), and there are no analogs of the surface, special, and extraordinary transitions (*18*). The RG analysis of the bulk action S_b is now standard textbook material—we will not reproduce it here and will follow the notation of (*21*). We introduce renormalized fields $\phi = \sqrt{Z}\phi_R$, $n = \sqrt{Z'}n_R$, and renormalized couplings by $g_0 = (\mu^\epsilon/c)(Z_4/Z^2S_{d+1})g$, $\gamma_0 = (\mu^\epsilon c)^{1/2}(Z_\gamma/\sqrt{ZZ'}\tilde{S}_{d+1})\gamma$, where μ is a renormalization inverse length scale, $\tilde{S}_d = \Gamma(d/2 - 1)/(4\pi^{d/2})$, and the bulk renormalization factors Z and Z_4 are specified in (*21*). For the new boundary renormalization factors, we obtain two loops, $Z' = 1 - 2\gamma^2/\epsilon + \gamma^4/\epsilon$ and $Z_\gamma = 1 + \pi^2[S(S + 1) - \frac{1}{3}]g\gamma^2/(2\epsilon)$. These lead to the β function for g found in (*21*) and the new β function for the boundary coupling

$$\beta(\gamma) = -\frac{\epsilon\gamma}{2} + \gamma^3 - \gamma^5 + \frac{5g^2\gamma}{144} + \pi^2[S(S + 1) - 1/3]g\gamma^3 + O[(\gamma,\sqrt{g})^7] \tag{11}$$

The critical fluctuations at the boundary are therefore controlled by the fixed point values $\gamma = \gamma^*$, $g = g^*$ (both nonzero) at which both β functions vanish, and canonical methods then imply the exponent

$$\eta' = \epsilon - \left[\frac{5}{242} + \frac{6\pi^2}{11}[S(S + 1) - 1/3]\right]\epsilon^2 + O(\epsilon^3) \tag{12}$$

Equation 6 can now be obtained by the methods of (*22*), whereas Eq. 8 follows directly from a renormalized perturbation theory in the ordered phase at $T = 0$. We conclude our technical interlude by noting that our RG

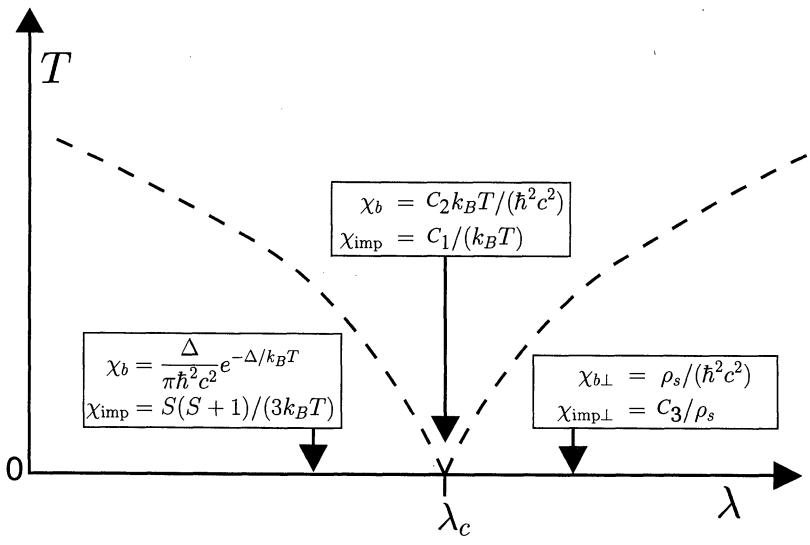

Fig. 3. Summary of the results for the bulk and impurity susceptibilities of H_X. The constants C_{1-3} are universal numbers, insensitive to microscopic details such as variations in the magnitude or sign of the exchange constants in the vicinity of the impurity, or the presence of additional nearby vacancies or impurity ions with different spins. The constants C_1 and C_3 depend only on the integer/half-odd integer valued S, and we can view Eq. 2, the $T \to 0$ limit of χ_{imp} in the paramagnet ($\lambda < \lambda_c$), as the experimental definition of S. For the case in which nonmagnetic impurities are added in a localized region, with no modification of exchange constants, S is the net imbalance of spin between the two sublattices. The constant C_1 defines the effective spin at the quantum-critical point by $C_1 = S_{\mathrm{eff}}(S_{\mathrm{eff}} + 1)/3$, and S_{eff} is neither an integer nor a half-odd integer.

scheme shows directly that the only graphs that contribute to the renormalization of γ_0, beyond those arising from the wave-function renormalization Z', must include a factor of the bulk interaction g_0; this implies that $Z_\gamma = 1$ for $g = 0$, and shows that for the models of (*9, 10*) the one-loop exponent $\eta' = \epsilon$ is exact.

The above methods can be extended to determine the behavior of other observables in the regimes of Fig. 3. We mention a few as follows.

1) Entropy: In the paramagnetic phase ($\lambda < \lambda_c$), there is clearly a residual entropy of $\ln(2S + 1)$ as $T \to 0$. At $\lambda = \lambda_c$, the ϵ expansion shows that this is modified to $\ln(2S + 1) - S(S + 1)(33\epsilon/160)^{1/2} + O(\epsilon^{3/2})$, whereas in the Néel state ($\lambda > \lambda_c$, the Néel order pinned by some small spin anisotropy) the impurity entropy vanishes as T^d at low T.

2) Knight shift: We restrict the discussion here to the intermediate quantum-critical region of Fig. 3, $T > |\lambda - \lambda_c|^\nu$. The shift in the nuclear magnetic resonance frequency is proportional to the local response in the presence of a uniform external field, $\chi(x)$. In the vicinity of the impurity (for example, at site $i = Y$), $\chi(x) \sim T^{-1+\eta'/2}$. Well away from the impurity ($|x| \to \infty$), apart from the bulk response of the antiferromagnet, there are staggered and uniform contributions that decay exponentially with $|x|$ on a scale $\sim \hbar c/(\sqrt{\epsilon} k_B T)$.

3) Magnon damping: In the quantum paramagnet ($\lambda < \lambda_c$), and at $T = 0$, the pure antiferromagnet has a pole in the dynamic spin structure factor $\sim 1/(\Delta - \hbar\omega)$ at the antiferromagnetic ordering wavevector from the triplet magnon excitations. In the presence of a dilute concentration of impurities, n_i, this pole will be broadened on an energy scale Γ; scaling arguments and the structure of the fixed point found here imply the exact form (*23*) $\Gamma \sim n_i(\hbar c)^d \Delta^{1-d}$. We argue that this damping mechanism is the main ingredient in the broadening of the resonance peak observed recently in Zn-doped $YBa_2Cu_3O_7$ (*24*). Using the values $\hbar c = 0.2a$ eV (a is the lattice spacing), $\Delta = 40$ meV, and $n_i = 0.005/a^2$, we obtain the estimate $\Gamma = 5$ meV, which is in excellent accord with the observed line width of 4.25 meV (*24*). We have also studied the line shape of the magnon peak (*17*) and find that it is asymmetric at very low T, with a tail at high frequencies; it would be interesting to test this in future experiments.

We have described the highly nontrivial, collective, quantum spin dynamics of a single impurity in a strongly correlated, low-dimensional electronic system. The problem maps onto a new boundary quantum field theory (Eqs. 9 and 10) and is therefore also of intrinsic theoretical interest. Unlike previously studied quantum impurity problems, there is a complicated interference between bulk and boundary interactions, and its proper description is the key to the physical results we have obtained. Our theoretical results for the magnon damping in the spin-gap phase are in good agreement with existing experiments (*24*). Studies of materials exhibiting other aspects of the regimes of Fig. 3 appear possible, and we hope they will be undertaken; spin-gap compounds can be driven across the transition by, say, application of hydrostatic pressure or by doping with other impurities that have the same spin as the host ion they replace and do not change the sign of the exchange constants (*25*). Quantum Monte Carlo simulations should also allow more accurate determination of the universal constants C_1 and C_3.

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23 September 1999; accepted 10 November 1999

Ion Penetration of the Water-Oil Interface

Kai Wu, Martin J. Iedema, James P. Cowin*

Ions typically pass with difficulty from water into organic phases because of water's superior solvation power. This inhibits such processes as ion transport in batteries or in lipid bilayers of cells. Ion penetration across such an interface was studied with unusual structural control. Hydronium ions were soft-landed at 1 electron volt on cold films of 3-methylpentane ("oil") on a metal substrate. The field produced by these ions drove them through the films when warmed. Coadsorption of water (0.14 to 35 bilayers) inhibited the ion penetration by creating a solvation energy trap. A Born solvation model successfully predicted the trapping energies (0 to 38 kilojoules per mole).

Transport of ions across the interface of two immiscible phases is fundamentally important in biological membranes, electrochemistry, phase transfer catalysis, fuel cells, extraction of nuclear waste, and groundwater contamination (*1–6*). Typically, hydration and solvation strongly affect both the energetics and kinetics of the ion transport. The Born equation (*7*) led to a qualitative understanding of the ion-transfer energetics derived from the difference in the dielectric constants of the two phases. Attempts to improve this understanding (*8, 9*) have been experimentally complicated by constraints of charge neu-

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