Quantum Impurity in a Nearly Critical Two Dimensional Antiferromagnet

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We describe 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. The impurity spin susceptibility has a Curie-like divergence at the quantum-critical coupling, but with a universal, effective spin which 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 significant applications to 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 non-zero spin (the 'spin gap' compounds, like $SrCu_2O_3$, $CuGeO_3$ and NaV_2O_5) has led to fundamental advances in our understanding of low dimensional, strongly correlated electronic systems. These systems are insulators and are so not as complicated as the cuprate high temperature superconductors (which display a plethora of phases with competing magnetic, charge, and superconducting order), and this simplicity has clearly 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 non-magnetic impurities, like 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 most fruitful line of investigation *(4)*. They have demonstrated a remarkable property of the paramagnetic ground state of the host compound: each non-magnetic impurity has a net magnetic moment of spin 1/2 located in its vicinity (for the case where the host compound has magnetic ions with spin 1/2). 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 will describe the quantum theory of an arbitrary localized deformation in such antiferromagnets; examples of deformations are (i) a single non-magnetic impurity, along with changes in the values of nearby exchange interactions; (ii) change in sign of a localized group of exchange interactions from antiferromagnetic to ferromagnetic. Our main concern will be 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 will 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 for 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 (see Fig 1A below); the resulting picture of doping by a non-magnetic impurity is that the partner spin of the impurity site is essentially free. To obtain any non-trivial dynamics one performs an expansion about such a decoupled limit, and this yields simple localized spin behavior with non-universal details depending upon 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 to then expand away from it into the spin gap state. As we shall discuss below, the coupling between the bulk and impurity excitations becomes *universal* in such an expansion, and all dynamical properties depend only upon the bulk parameters, Δ and a velocity c (defined below).

For clarity, we will 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)*

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

where $\mathbf{S}_i$ are spin-1/2 operators on the sites of the coupled-ladder lattice shown in Fig 1, with the A links forming two-leg ladders while the B links couple the ladders. The ground state of H depends only on the dimensionless coupling λ, and we will restrict our attention to $J > 0$, $0 \leq \lambda \leq 1$. At $\lambda = 0$ the ladders are decoupled, and each forms a spin singlet quantum paramagnet (Fig 1A). This paramagnetic state continues adiabatically for small non-zero 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 the two quantities are asymptotically proportional). For $\lambda > \lambda_c$, the ground state has long range Néel order (Fig 1B) characterized by the non-zero spin stiffnesses, ρ_{sx}, ρ_{sy}, which determine the energy cost to twists in the order parameter orientation in the x, 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 which travel with velocities c_x, c_y in the x, 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 $(\lambda - \lambda_c)^\nu$, while the velocities remain finite and non-critical.

Introducing a non-magnetic impurity in $\mathcal{H}$ by removing the spin at site $i = X$ (Fig 2), the modified Hamiltonian $\mathcal{H}_X$ has the same form as $\mathcal{H}$ 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 $\mathcal{H}_X - g\mu_B \sum_{i \neq X} \mathbf{H} \cdot \mathbf{S}_i$ where μ_B is the Bohr magneton and g is the gyromagnetic ratio of the ion). This susceptiblity may be written as $\chi = (g\mu_B)^2(\mathcal{A}\chi_b + \chi_{\rm imp})$ where $\mathcal{A}$ 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 non-magnetic impurity. We will now describe the behaviors of χ_b and $\chi_{\rm imp}$ as the temperature $T \to 0$, and λ moves across λ_c.

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

$$\chi_{\rm imp} = \frac{S(S+1)}{3k_B T}, \tag{2}$$

where $S = 1/2$ 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

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

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, 10, 11)*, with

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

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 χ_{imp} is given essentially by the Fourier transform of Eq 4, that is by $\chi_{\text{imp}} \sim T^{-1+\eta'}$ *(13)*. This result is a consequence of a 'compensation' effect *(14)*, as 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, χ_{imp} vanishes in the strict continuum limit, and corrections to scaling have to be considered, which lead eventually to $\chi_{\text{imp}} \sim T^{-1+\eta'}$. Our computations show that the behavior of $\mathcal{H}_X$ at $\lambda = \lambda_c$ is dramatically different: the magnon excitations do not have an exact compensation property, and their response is non-zero already in the scaling limit. So in a sense, the present problem is simpler than the overscreened Kondo case, and naive 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:

$$\chi_{\rm imp} = \frac{\mathcal{C}_1}{k_B T} \tag{5}$$

at $\lambda = \lambda_c$, where $\mathcal{C}_1$ is a universal number independent of microscopic details (as are all the $\mathcal{C}_i$ introduced below). We computed $\mathcal{C}_1$ in the expansion in $\epsilon = 3 - d$, where d is the spatial dimension, and obtained

$$\mathcal{C}_1 = \frac{S(S+1)}{3}\left[1 + \left(\frac{33\epsilon}{40}\right)^{1/2} - \frac{7\epsilon}{4} + \mathcal{O}\left(\epsilon^{3/2}\right)\right]; \tag{6}$$

the omitted higher order corrections in Eq 6 will, in general, depend upon S. Comparing with Eq 2 we can define an effective impurity spin, $S_{\rm eff}$, at the quantum-critical point by $\mathcal{C}_1 = S_{\rm eff}(S_{\rm eff}+1)/3$; 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 $\epsilon = 1$. Also notice that the leading corrections in the ϵ-expansion are quite large, and this will be a feature of all the results obtained below; accurate numerical estimates require some resummation scheme, but we will not discuss this here. For completeness, let us note that at $\lambda = \lambda_c$, the bulk response *(16)* $\chi_b = \mathcal{C}_2(k_B T)/(\hbar c)^2$, a T-dependence that is also different from the bulk response in the overscreened Kondo problem.

Finally, we describe the situation for $\lambda > \lambda_c$. Now 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 of this paper is

$$\chi_{\text{imp}\perp} = \frac{\mathcal{C}_3}{\rho_s}. \tag{7}$$

In general d, this relationship is $\chi_{\text{imp}\perp} = \mathcal{C}_3(\hbar c)^{(2-d)/(d-1)}/\rho_s^{1/(d-1)}$, and the ϵ-expansion of $\mathcal{C}_3$ is

$$\mathcal{C}_3 = \frac{15S}{\sqrt{22}}\left(\frac{11S_{d+1}}{2\epsilon}\right)^{1/(d-1)}\left[1-(1.193+0.553S+0.419S^2)\epsilon+\mathcal{O}(\epsilon^2)\right], \tag{8}$$

where $S_d = 2/(\Gamma(d/2)(4\pi)^{d/2})$. Note that ρ_s vanishes, and so $\chi_{\text{imp}\perp}$ 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 non-zero T. Then the susceptibility takes the rotationally averaged value $\chi_{\text{imp}} = S^2/(3k_BT) + (2/3)\chi_{\text{imp}\perp}$, where the first term is the contribution of the net moment noted earlier (note that 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 contain a technical interlude which outlines 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 $\mathcal{S}_b$ of a field $\phi_\alpha(x,\tau)$ ($\alpha = 1\ldots3$) representing the collinear Néel order parameter *(7)*. This is coupled by the action $\mathcal{S}_{\text{imp}}$ to an impurity spin at $x = 0$ with orientation given by the unit vector n_α. The partition function is $\int \mathcal{D}\phi(x,\tau)\mathcal{D}n(\tau)\exp(-\mathcal{S}_b - \mathcal{S}_{\text{imp}})$ with

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

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

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 $\mathcal{S}_b$ 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 $\mathcal{H}$ maps onto the $r > r_c$ ($r < r_c$) region of the field theory $\mathcal{S}_b$. Quite generally, any local deformation of the antiferromagnet is described by the action $\mathcal{S}_b + \mathcal{S}_{\rm imp}$, 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 like $\int d\tau \phi_\alpha^2(x=0,\tau)$ which are all strongly irrelevant under the renormalization group (RG) analysis in powers of ϵ. The $r=0$, $g_0 = 0$ case of Eqs 9,10 was considered earlier by Sengupta *(10)* (and related models in *(9, 11)*) in a non-local 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 $\mathbf{H}$ (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 $\mathcal{S}_b + \mathcal{S}_{\rm imp}$ 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 $\mathcal{S}_b$ 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 $g_0 = (\mu^\epsilon/c)(Z_4/Z^2 S_{d+1})g$, $\gamma_0 = (\mu^\epsilon c)^{1/2}(Z_\gamma/\sqrt{ZZ'\widetilde{S}_{d+1}})\gamma$ where μ is a renormalization inverse length scale, $\widetilde{S}_d = \Gamma(d/2-1)/(4\pi^{d/2})$, and the bulk renormalization factors Z, Z_4 are specified in *(21)*. For the new boundary renormalization factors, we obtained to two loops $Z' = 1 - 2\gamma^2/\epsilon + \gamma^4/\epsilon$ and $Z_\gamma = 1 + \pi^2(S(S+1) - 1/3)g\gamma^2/(6\epsilon)$. These lead to the β function for g found in *(21)*, and the new β function for the boundary coupling

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

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

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

Eq 6 can now be obtained by the methods of *(22)*, while 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 scheme shows directly that the only graphs which contribute to the renormalization of γ_0, beyond those arising from the wavefunction 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:

(i) 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 $\ln(2S+1) - S(S+1)(33\epsilon/160)^{1/2} + \mathcal{O}(\epsilon^{3/2})$, while 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.

(ii) 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 NMR resonance frequency is proportional to the local response in the presence of a uniform external field, $\chi(x)$. In the vicinity of the impurity (*e.g.* 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 which decay exponentially with $|x|$ on a scale $\sim \hbar c/(\sqrt{\epsilon} k_B T)$.

(iii) 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 linewidth of 4.25 meV *(24)*. We have also studied the lineshape 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 non-trivial, 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 9,10, 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 which 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 $\mathcal{C}_1$ and $\mathcal{C}_3$.

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[23] More precisely, the theory in Eqs 9,10 predicts that $\Gamma = \mathcal{C}_4 n_i (\hbar c)^d \Delta^{1-d}$ where $\mathcal{C}_4$ is a universal number, of order unity in $d = 2$. Note that this result is *independent* of the coupling, γ_0, between the bulk and the impurity.

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FIGURES

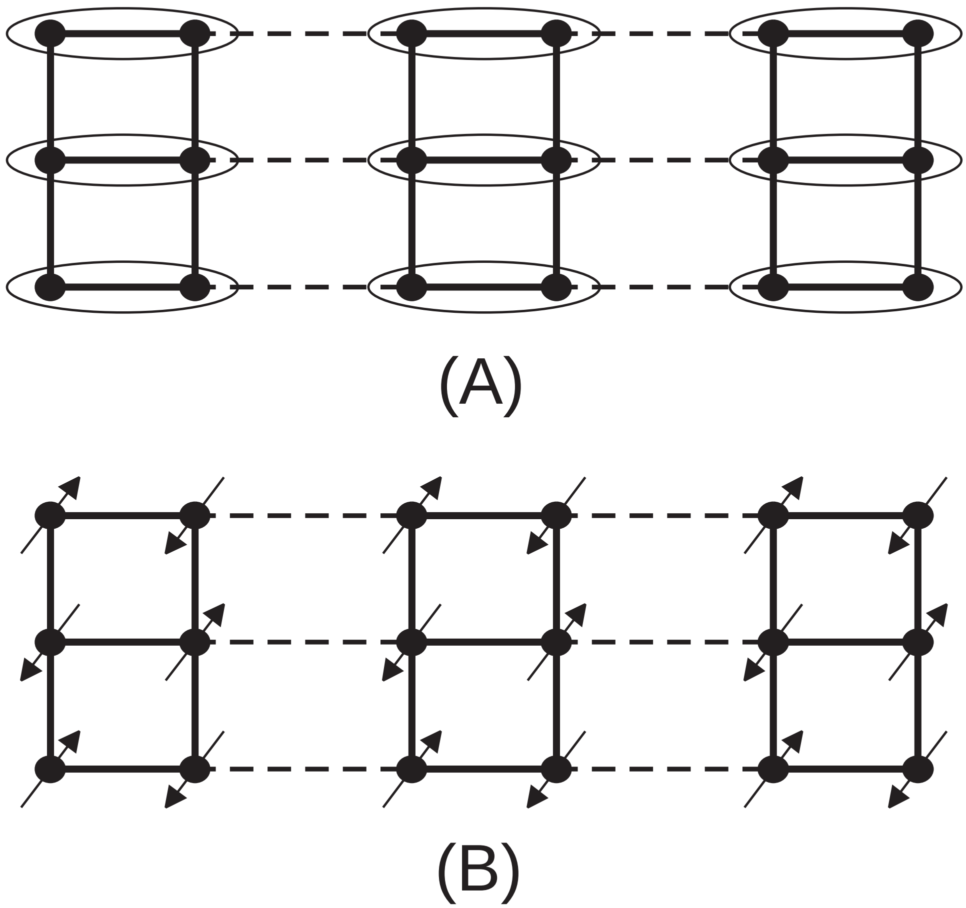

FIG. 1: The coupled ladder antiferromagnet. The A links are full lines and have exchange J, while the B links are dashed lines and have exchange λJ. The paramagnetic ground state for $\lambda < \lambda_c$ is schematically indicated in (A): the ellipse represents a singlet valence bond, $(|\uparrow\downarrow\rangle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

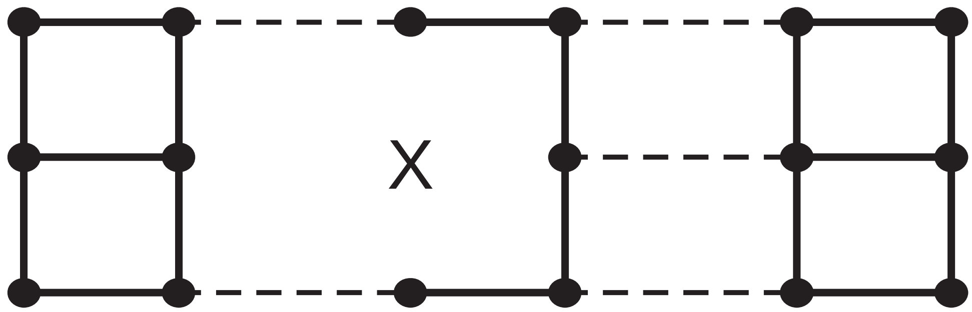

FIG. 2: The impurity Hamiltonian $\mathcal{H}_X$ in which the spin and links on site $i = X$ have been removed.

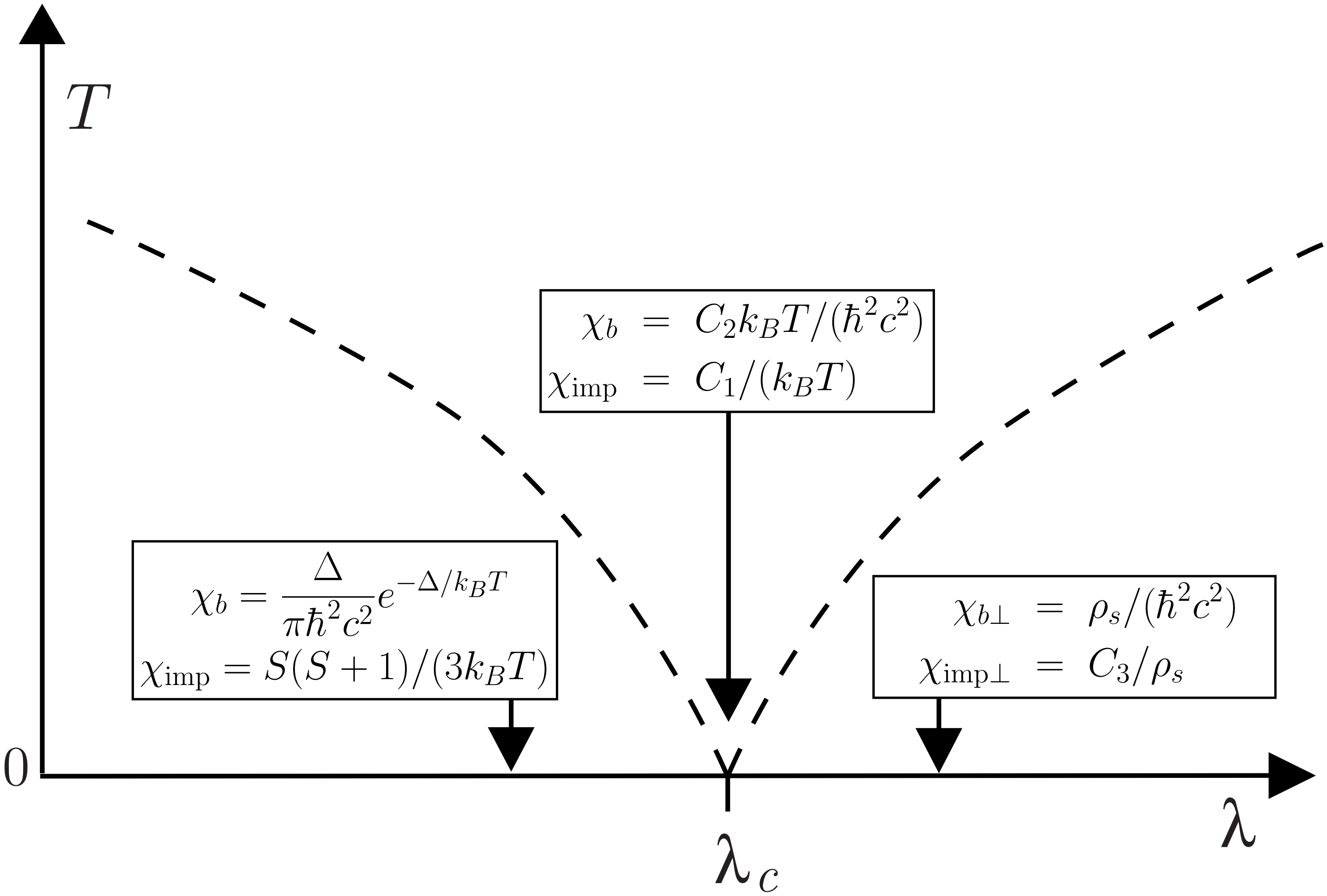

FIG. 3: Summary of the results for the bulk and impurity susceptibilities of $\mathcal{H}_X$. The constants $\mathcal{C}_{1-3}$ are universal numbers, insensitive to microscopic details like variations in the magnitude or sign of the exchange constants in the vicinity of the impurity, or presence of additional, nearby, vacancies or impurity ions with different spins. The constants $\mathcal{C}_1$ and $\mathcal{C}_3$ depend only on the integer/half-odd-integer valued S, and we can view Eq 2, the $T \to 0$ limit of $\chi_{\rm imp}$ in the paramagnet ($\lambda < \lambda_c$), as the experimental definition of S. For the case in which non-magnetic 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 $\mathcal{C}_1$ defines the effective spin at the quantum-critical point by $\mathcal{C}_1 = S_{\rm eff}(S_{\rm eff}+1)/3$, and $S_{\rm eff}$ is neither an integer nor a half-odd-integer.