Nonlinear spectroscopy of random quantum magnets

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We study nonlinear response in quantum systems near strong-randomness critical points. Nonlinear dynamical probes, such as 2D coherent spectroscopy, can diagnose the nearly localized character of excitations in such systems. We focus on the random transverse-field Ising model and derive exact results for nonlinear response, from which we extract information about critical behavior that is absent in linear response. Our analysis incorporates realistic channels for dissipation in random magnets; we present exact scaling forms for the resulting distribution functions of relaxation times.

The use of strong electromagnetic fields to probe solid-state systems and pump them into exotic states has been a fruitful research direction. One paradigm for these experiments has been “pump-probe spectroscopy,” where a system is pumped with an intense field, creating a far-from-equilibrium state, whose response to a weaker “probe” field is subsequently measured [1–11]. Two-dimensional coherent spectroscopy (2DCS) [12–16] is a conceptually similar multi-pulse technique, but operates in a regime where the pump changes the state of the system only weakly. Instead of creating and characterizing far-from-equilibrium states, 2DCS probes multitime correlation functions of a given equilibrium state, which can often extract qualitative information not captured by linear response. For instance, they can distinguish between “inhomogeneous” broadening (i.e., a continuum of response due to many separate sharp modes with a wide frequency spread) and “homogeneous” broadening (i.e., broadening due to finite excitation lifetimes). Consequently, 2DCS can isolate interaction effects in settings where linear response does not diagnose the central phenomena of interest [17–24], such as in systems exhibiting fractionalization [25, 26] or localization [6, 27, 28].

In the present work we construct a theory for the response of random quantum systems probed using 2DCS (or, more generally, pump-probe spectroscopy). We focus on systems near infinite-randomness quantum critical points (IRQCPs) [29–32], for which we can make explicit, asymptotically exact predictions for the broadening and intensity of spectral lines. A system near an IRQCP can be modeled as an ensemble of weakly interacting two-level systems (TLSs); the properties of this ensemble are determined by scaling exponents associated with the IRQCP. Anomalous exponents persist away from the critical point itself, due to strong Griffiths effects [30]. Some information about the TLS distributions can be extracted from linear response [33] and nuclear magnetic resonance relaxation times [34, 35]. We argue here that nonlinear response reconstructs the full TLS distribution function as well as the residual interactions among TLSs.

Our central results concern the lifetimes of TLS’s at IRQCPs and in quantum Griffiths phases. At IRQCPs, these lifetimes are infinite; lifetimes in realistic experiments are therefore governed by the system-bath coupling. Although the system-bath coupling is irrelevant in the renormalization group sense, it is dangerously irrelevant, and causes the TLSs to have finite lifetimes. We relate the relaxation mechanisms and rates to critical data by computing the full distributions of relaxation times. For random quantum magnets coupled to phonons (or other non-magnetic environments), we find that the TLS relaxation times are power-law distributed both at criticality and into the Griffiths phase. Local probes measure the relaxation of a typical TLS, which is exponential with a rate we compute. However, the spatially averaged response probed by most optical experiments picks up the entire broad spectrum of relaxation times. We show that a 2DCS response in the frequency-time plane extracts exponents characterizing both the relaxation-time and resonance-frequency distributions. We argue that the phenomenology of the averaged response is generic to a large class of random quantum magnets, not just those described by infinite-randomness physics.

Random TFIM.— We focus on the one-dimensional random transverse field Ising model (RTFIM),

$$H_{RTFIM} = -\sum_i (h_i \sigma^z_i + J_i \sigma^x_i \sigma^{x}_{i+1}),$$

(1)

where the $h_i$, $J_i$ are positive i.i.d. random variables. (We swap $\sigma^z$ and $\sigma^x$ relative to convention; this leads to a more natural TLS basis later.) The RTFIM can be iteratively diagonalized by an asymptotically exact real-space renormalization-group (RSRG) method [30, 31, 36]. For the ground state, the RSRG rules are as follows. One picks the strongest coupling. If it is a bond $J_i$, one eliminates site $i$ by placing it into its $|\uparrow\downarrow\rangle$ state, creating an effective perturbative coupling $J_i J_{i+1}/h_i$. These steps are iterated until all spins have been decimated. Under this procedure, any finite amount of initial disorder in the bare couplings flows to extremely broad (power-law) distributions of renormalized couplings: this is what is meant by ‘infinite random-
ness’. This leads to the following emergent low-energy properties: (1) The RTFIM has an IRQCP controlled by the parameter $2\delta \equiv (\ln \tilde{t} - \ln J)I$, where \((\cdots)\) denotes an average over the disorder distribution. The IRQCP is at $\delta = 0$. (2) At the IRQCP, spatial and temporal fluctuations are infinitely anisotropic, and scale via the relation $\ln t \sim \sqrt{\ell(t)}$. At criticality, the typical magnetic moment of a superspin at scale $\ell$ is $\ell^{\phi/2}$, where $\phi$ is the Golden mean. (3) Distributions of physical observables are extremely broad, so that (for example) average and typical correlation lengths diverge with different exponents away from criticality. (4) The IRQCP is flanked by Griffiths phases on both paramagnetic (PM) and ferromagnetic (FM) sides. In the PM Griffiths phase, for example, the system as a whole is not magnetically ordered, but has rare FM regions which locally appear to be on the “wrong side” of the transition, which can be viewed as effective TLS’s that dominate the response.

**Rare TLSs in the Griffiths phase.**—For concreteness we specialize to the IRQCP and the PM regime near it. Recall that a field decimation effectively decouples a superspin (cluster) from the rest of the system, whereas a bond decimation grows a FM cluster. A cluster that decouples at energy scale $\varepsilon$ contributes to dynamics at $\omega = \varepsilon$, but freezes out at lower frequencies. Clusters that are slow compared to the probe frequency are also unimportant to response, as they can only contribute via higher-order perturbative processes. Thus, to understand response at $\omega$, we must characterize a typical TLS generated by field decimations occurring at scale $\varepsilon = \omega$.

In the PM, the system initially looks critical on short distances, but eventually on coarse-graining out to the correlation length $\xi \sim (\ln \varepsilon)^2 \sim \delta^{-2}$, the energy scale reduces below $\varepsilon^* \sim e^{-1/\delta}$. The RG then crosses over into the off-critical PM regime where it is overwhelmingly likely to decimate fields rather than bonds. For frequencies $\omega < \varepsilon^*$, the system can therefore be viewed as a set of TLSs that are weakly coupled by the residual bond terms. Such clusters contribute anomalous power laws to low-frequency response: a locally FM cluster of $l$ sites has an exponentially small probability $p^l$ of occurring. Each such cluster has two parity eigenstates $|\pm\rangle = \frac{1}{\sqrt{2}}(|0\rangle^{\otimes l} \pm |1\rangle^{\otimes l})$ separated from the other states by an energy gap. The resonant frequency for tunneling between these obeys $\log(1/\omega) \propto l$. Thus, at any frequency $\omega$, the TLSs that are flippable at frequency $\omega$ are of size $\sim \ln \omega$, and so are only power-law rare in $\omega$.

Precise scaling forms can be computed by running the RG from a microscopic energy scale $\Omega_I$ (set to 1 throughout) to the probing scale $\Omega$, where the remaining degrees of freedom will be the rare regions discussed above. The bond and field distributions are

$$ P_{\Omega}(J) = \frac{u_\Omega}{\Omega} \left( \frac{J}{\Omega} \right)^{u_\Omega - 1}, \quad \rho_{\Omega}(h) = \frac{\tau_\Omega}{\Omega} \left( \frac{h}{\Omega} \right)^{\tau_\Omega - 1} $$

with $u_\Omega = \frac{2\delta}{\varepsilon^* + 1}$ and $\tau_\Omega = \frac{2\delta}{1 - \varepsilon^*}$, where $\Gamma = \ln \Omega_I/\Omega$.

As $\delta \to \Omega$, $u_\Omega \sim \tau_\Omega \sim 1/\Gamma$, and both distributions tend to $P(J) = \frac{1}{\Omega} \left( \frac{J}{\Omega} \right)^{1-1/\Gamma}$ which broadens as the RG flows to $\Gamma \to \infty$, as is characteristic of IRQCPs. Thus, $\rho_{\Omega}(\varepsilon)$ is density of TLS’s with splitting $\varepsilon < \Omega$ when the RG is at scale $\Omega$. The effective size of a rare TLS at energy scale $\varepsilon$ is obtained by running the RG to $\Omega = \varepsilon$ and then using the rare region arguments above; this yields

$$ \ell_{\varepsilon} = \frac{1}{\tau_{\varepsilon}} |\ln \varepsilon| \sim \frac{1 - \varepsilon^{-2\delta}}{2\delta} |\ln \varepsilon|, $$

Note that usually the $\varepsilon$-dependence in the prefactor is ignored (the difference amounts to replacing the bare cutoff $2\delta$ by its renormalized value $\sim \tau_{\varepsilon}$). We have retained the full dependence, since it allows us to access both the $\ell_{\varepsilon} \sim |\ln \varepsilon|^2$ scaling at criticality (by taking $\delta \to 0$ before $\varepsilon \to 0$) as well as finite-$\delta$ behavior (the opposite order of limits). Similarly, we may estimate the magnetic moment of a TLS by viewing it as composed of $n \sim 1/\xi$ critical clusters of size $\xi$ and moment $\mu_{\varepsilon} \sim \xi^{\phi/2}$; using $\xi \sim \delta^{-2}$ and replacing $\delta$ by its renormalized value:

$$ \mu_{\varepsilon} \sim |\ln \varepsilon| \left( \frac{2\delta}{1 - \varepsilon^{2\phi}} \right)^{1-\phi}, $$

which becomes $\mu \sim |\ln \varepsilon|^{\phi}$ at criticality (as noted above).

**Relaxation processes.**—The RSRG generates TLS’s that are infinitely sharp, corresponding to strictly localized excitations. In realistic systems, these TLS’s eventually relax. Relaxation can occur either because the system is coupled to an extrinsic reservoir, or because interactions cause the system to act as its own bath [37]; we focus on extrinsic baths, but our results should also generalize, with some modifications, to intrinsic baths treated self-consistently [37–39]. Two other key distinctions in terms of relaxation dynamics are: (i) between magnetic baths that couple directly to the order parameter $\sigma^z$ (and therefore involve magnetic degrees of freedom, e.g., nuclear spins) and non-magnetic baths that do not (e.g., phonons), and (ii) between baths with rapidly vanishing low-energy spectral density $J(\omega) \sim \omega^s$, $s > 1$ (superohmic baths, defined more precisely below) and those with $s \leq 1$ (i.e., ohmic or subohmic) [40]. Our central new results involve non-magnetic baths; before turning to these, we briefly comment on magnetic ones [41–46]. A magnetic bath always has a matrix element ($\mu_{\varepsilon}$) to flip a single TLS, and in the ohmic/subohmic cases couples strongly to TLS’s and destroys the IRQCP [41–46]. For superohmic baths, however, the IRQCP survives, and one can straightforwardly compute excitation lifetimes using Fermi’s Golden Rule, as $\tau^{-1} \sim \mu_{\varepsilon}^2 \varepsilon^s$. This sets timescales for both energy relaxation $(T_1)$ and dephasing $(T_2)$.

We now turn to the more delicate relaxation channel due to the coupling of TLS’s to phonons, which modulate the distance (and therefore the coupling) between nearby spins. We can incorporate phonons through the change $J_i \mapsto J_i(1 + X_i)$ to the bond terms in (1),...
I case, due to the paucity of bath modes as \( \Omega \) \( s \) subohmic bath with is similar and evolves smoothly out of instead, it can only couple diagonally the effective bath spectral density. at energy \( \epsilon \) bond is coupled to its own bath, an effective superspin so the RG proceeds as in the closed system. Since each TLSs are always weakly coupled to the bath, \( J \) \( \lambda \) \( \Theta \) \( \Theta(\epsilon_\gamma - \gamma) \),

\[
\gamma_{\text{typ}} = e^{\ln \gamma} = \gamma_{\epsilon}^{\ast} e^{-\frac{t}{\tau_\gamma}} = \frac{1 - e^{-\frac{t}{\tau}}}{\Sigma} \ln |e^{\epsilon/2^s} \frac{e^{\epsilon/2^s}}{2^s}.
\]

This quantity has a stretched-exponential suppression in the PM Griffiths phase (\( \delta > 0 \)) that is absent at criticality [Fig. 1(b)]. Consequently, local response at energy \( \epsilon \) sharpens on moving off-criticality into the PM.

**2DCS Response.**—We now discuss how these line-shapes can be probed using nonlinear spectroscopy, focusing on 2DCS for concreteness [12, 13, 28]. We consider the following spectroscopic protocol: initialize the TLS in its ground state; apply two sharp pulses \( A \) and \( B \), separated by a time \( \tau \), that couple to the magnetization; wait a time \( \tau \) and finally measure the magnetization of the the TLS. In terms of the Pauli matrices \( \sigma^x, \beta, \gamma \), the TLS has
Hamiltonian \( H_0 = -\frac{\gamma}{2} \sigma^z \), \( |0\rangle = |\pm \rangle \) and the pulses are \( \delta \)-function kicks that couple to \( \sigma^z \). As these rotate the TLS by some angle \( \theta_{A,B} \) in the \( yz \) plane, corresponding to action \( R(\theta) = e^{i\frac{\pi}{2} \sigma^z} \), the protocol prepares the state (setting \( h = 1 \)) \( |\psi(t; \tau)\rangle = e^{-iH_0t}R(\theta_A)e^{-iH_0\tau}R(\theta_B)|0\rangle \), and we measure \( P^x(t, \tau) = \langle \psi(t; \tau) | \sigma^x | \psi(t; \tau) \rangle \). In the perturbative limit, \( R_0 \approx 1 + i \bar{\theta} \sigma^z + \cdots \), and only odd powers contribute to \( P^x \). The leading response is linear in \( \theta \) (i.e., one factor of \( \theta \sigma^z \) appears either on the bra or the ket side), is proportional to the autocorrelation function, and is usually extracted out. The next set of contributions are cubic in \( \theta \). There are a number of distinct cubic contributions; a key insight of 2DCS is that these oscillate at different frequencies with respect to \( t \) and \( \tau \) [13]. The one of interest here is the “pump-probe” (PP) contribution, in which both the bra and ket are flipped by the initial pulse, yielding the response

\[
P^x_{PP}(t, \tau) \propto \text{Im} \langle 0 \rangle \sigma^x e^{iH_0(\tau + t)} \sigma^z e^{-iH_0t} \sigma^x e^{-iH_0\tau} \sigma^z |0\rangle
\]

\[
\sim \sin \varepsilon t
\]  

which looks like a linear-response correlator measured in the state \( \sigma^z |0\rangle \) driven out of equilibrium by the drive.

In the presence of relaxation, the PP response is subject to longitudinal relaxation between the pulses, and transverse relaxation after the second pulse:

\[
P^x_{PP}(t, \tau) \sim e^{-\tau/T_1} e^{-\tau/T_2} \sin \varepsilon t,
\]

and hence distinguishes homogeneous broadening due to longitudinal relaxation from inhomogeneous broadening: only the former depends on \( \tau \). However, in a 2DCS experiment the PP response is convolved with those from other excitation sequences (i.e., processes in perturbation theory where the pulses act at different sequences of times). To separate out the excitation sequences, one usually Fourier transforms the full nonlinear response of interest scaling data. Thus, for example, linear response 2DCS focuses on spectral diffusion [48], which is a distinct mechanism from the quenched disorder operational here.

**Discussion.**— Linear response near random quantum critical points probes a particular subset of the critical scaling data. Thus, for example, linear response at the Ising IRQCP is a convolution of effects due to anomalous scaling of magnetic moments and density of states; it is entirely insensitive to the lifetime of excitations at an IRQCP, and also largely insensitive to whether one is at the IRQCP or near it. By contrast, nonlinear response captures and separates out these features: the magnetic moment sets the strength of the nonlinearity, the density of states sets the overall spectral intensity (which can be measured in linear response), and the typical excitation lifetimes diagnose whether the system is in the critical or the Griffiths regime. While the Ising IRQCP is quite well studied, the same techniques extend directly to other IRQCPs and can be used to probe nontrivial aspects of these critical points, like the distribution of residual couplings, that would otherwise be inaccessible. A key feature of the low-frequency spatially-averaged lineshapes we computed is that they depend only on the broad distribution of couplings between low-energy degrees of freedom. Therefore, they are relevant to random quantum magnets in arbitrary spatial dimensions (including those not described by infinite-randomness fixed points), as long as two criteria are satisfied: (i) the low-energy degrees of freedom probed by the response are localized, with residual couplings exponentially sensitive to the following: (i) the density of TLS splittings with splitting \( \varepsilon \) is \( \rho_0(\varepsilon) \sim 1/\varepsilon^{1-2\delta} \); (ii) such TLSs relax with \( T_2 \approx 2T_1 \sim \gamma^{-1} \) distributed according to (5); and (iii) each TLS couples to a probe field via its moment \( \mu_\varepsilon \) (4), with the nonlinear response of interest scaling \( \sim \mu_\varepsilon^2 \). Accordingly, the averaged mixed pump-probe response for \( \omega_i > 0 \) scales as

\[
P^x_{PP}(\omega_i, \tau) \sim \frac{1}{\omega_i} e^{-\gamma \tau/2} \int_0^{\Omega} \! d\varepsilon \rho_0(\varepsilon) \int_0^{\gamma} \! d\gamma P_x(\gamma) e^{-\gamma \tau/2} (\varepsilon - \omega_i + i\gamma) \]

Since the response is dominated by small \( \gamma \), we can use the Sokhotski-Plemelj identity \( \text{Im} \frac{1}{\varepsilon - \omega_i} = \delta(\varepsilon) \) to decouple the \( \gamma \) and \( \varepsilon \) integrals, leading to

\[
\text{Im} P^x_{PP}(\omega_i, \tau) \sim \rho_0(\omega_i) e^{-\gamma \tau/2} \sim \frac{\mu_\varepsilon^2}{\omega_i^{1-2\delta}} \int_\varepsilon^{\infty} \! d\xi e^{-\xi - \varepsilon} \delta(\xi) \]

where \( f_\varepsilon(x) = \frac{\omega_\varepsilon^2}{\omega_i^{1-2\delta}} \int_\varepsilon^{\infty} \! d\xi e^{-\xi - \varepsilon} \delta(\xi) \) describes the averaged decay profile at frequency \( \varepsilon \) [cf. Fig. 1(a)]. Fig 1c shows such a mixed-2DCS portrait of the pump-probe response of the RTFIM at criticality; note the rapid sharpening of the response at low frequencies. A fixed \( \omega_i \)-slice corresponds to \( f_\varepsilon \) and hence allows us to extract \( \gamma_i \), whereas the evolution of the peak height at \( \tau = 0 \) allows us to extract information on the energy-dependent distributions and renormalized moments. We note that previous work on anomalous power-law relaxation in 2DCS focused on spectral diffusion [48], which is a distinct mechanism from the quenched disorder operational here.
the spacings between them, which are exponentially distributed; and (ii) the bath does not couple directly to the magnetic degrees of freedom, but only modulates their interactions. The former behaviour is generic in random quantum magnets [49], while the latter is typical of several realistic bath mechanisms, including phonons. However, the behaviour of typical lifetimes and their sharpening off-criticality are characteristic of IRQCPs, rather than being applicable more generally.

We took each bond to couple to its own phonon bath. More realistically, phonons have spatial structure, with a dispersion $\omega \sim k$. Thus, a TLS at energy $\varepsilon$ couples to a phonon wavepacket that is correlated over a range $\sim \varepsilon$ (which is much larger than the TLS size). For the many effective TLS’s in the same “phonon volume” the spin-phonon coupling is of the form $X \sum_i J_i \sigma_i^z \sigma_{i+1}^z$. Integrating out phonons can thus generate many-spin interactions. Crucially, however, phonons are non-magnetic and cannot mediate flip-flops among distinct TLS’s. Residual magnetic couplings between an effective TLS and its neighbors still involve bond terms $J_i$; provided that $P(J)$ is a broad distribution, relaxation rates are still broadly distributed, despite the spatial structure of phonons. Therefore, our conclusions continue to apply (up to prefactors) for realistic phonon baths.

The frequency range that is readily probed by existing THz spectroscopic methods ($\sim 0.1 – 10$ THz $\sim 1 – 20$ K) lines up well with the intrinsic energy scales of various experimentally realized Ising magnets such as cobalt niobate [50, 51]. Further, the analysis we presented above applies with essentially no changes [49] to random Heisenberg magnets, which exhibit similar phenomenology. Examples include BaCu$_2$Si$_{1-x}$Ge$_x$O$_7$ [34] where the doping dependent exchange scale $\sim 400$ K, and the organic salt quinolinium-(TCNQ)$_2$, where “random singlet” physics has been reported [52, 53] at temperatures $\lesssim 20$ K. The challenge in present-day spectroscopic experiments is to drive the system with enough laser power to induce a measurable optical nonlinear magnetic response; this is a major current objective in the field [54], which we expect to be achieved in near-term experiments.

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