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Quantifying and controlling entanglement in the quantum magnet Cs_2CoCl_4

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The lack of methods to experimentally detect and quantify entanglement in quantum matter impedes our ability to identify materials hosting highly entangled phases, such as quantum spin liquids. We thus investigate the feasibility of using inelastic neutron scattering (INS) to implement a model-independent measurement protocol for entanglement based on three entanglement witnesses: one-tangle, two-tangle, and quantum Fisher information (QFI). We perform high-resolution INS measurements on Cs_2CoCl_4, a close realization of the $S = 1/2$ transverse-field XXZ spin chain, where we can control entanglement using the magnetic field, and compare with density-matrix renormalization group calculations for validation. The three witnesses allow us to infer entanglement properties and make deductions about the quantum state in the material. We find QFI to be a particularly robust experimental probe of entanglement, whereas the one- and two-tangles require more careful analysis. Our results lay the foundation for a general entanglement detection protocol for quantum spin systems.

Introduction.—Quantum entanglement is increasingly considered a vital resource for novel effects and applications [1]. Entanglement is also central to our understanding of many-body systems [2, 3], where it forms a deep connection between condensed matter physics and quantum information. Phenomena such as quantum spin liquids [4], topological order [5], quantum criticality [6, 7], and thermalization in quantum systems [8], are all inherently related to entanglement properties. It is crucial to develop experimental protocols to detect and quantify entanglement in the solid state, in order to allow unambiguous and rapid identification of quantum materials suitable for new applications, and novel insights into complex quantum phenomena.

Due to the rich structure of many-body states, a number of different entanglement measures have been introduced. The most important example in condensed matter theory is entanglement entropy (EE), used to quantify bipartite entanglement. Yet there is no "EE observable" that can be probed directly, which makes experimentally quantifying entanglement in many-body systems challenging [2, 9]. Although EE has been measured in cold-atom [10, 11] and photonic systems [12], neither approach is suitable for probing entanglement in macroscopic condensed matter systems.

In special cases, entanglement can be inferred through neutron scattering experiments. For instance, two-spin entanglement within and between dimers [13, 14], and entanglement between two molecular magnet qubits [15] have been characterized with neutrons. Also, certain low-dimensional spin systems can be shown to have entanglement via close comparison with theory [16–18]. However, these approaches rely on tractable models, with either small Hilbert spaces or special ground states, which are limited to a handful of systems. For most strongly correlated systems, such methods are not applicable, calling for model-independent approaches.

A promising approach, which we explore in this Letter, is using entanglement witnesses (EWs) [2, 3, 9], i.e. observables that can be used to identify some *subset* of entangled states. We consider (i) one-tangle (τ_1) [19–21], (ii) concurrence or two-tangle (τ_2) [13, 15, 19, 20, 22], and (iii) quantum Fisher information (QFI) [23–25]. These EWs witness (i) entanglement between a spin and the rest of the system, (ii) pairwise entanglement, and (iii) multipartite entanglement, respectively, and thus provide complementary information. All three EWs are accessible to inelastic neutron scattering (INS) experiments. τ_1 and τ_2 can be obtained from ordered moments and spatial spin-spin correlations [21], while QFI can be expressed as an integral of the dynamical spin structure factor (DSF) [25], $S(k, \hbar\omega)$. This powerful formulation of QFI has been applied to experiments on Heisenberg spin chains [26, 27], but remains otherwise largely unexplored.

We contrast EE and the mentioned EWs in the spin-1/2 transverse-field XXZ chain. The one-dimensional setting confers an enhanced susceptibility to fluctuations and a higher degree of theoretical tractability, making it an excellent proving ground for our EW protocol. The system hosts two distinct quantum critical points (QCPs) and a classical, minimally entangled point, and thus provides a range of interesting behaviors. We study this model numerically using the density matrix renormalization group (DMRG) [28–30]. We also report high-resolution INS data on the chain compound Cs_2CoCl_4, known to be an excellent realization of the XXZ model [31–37]. We find that QFI values extracted from experiment and

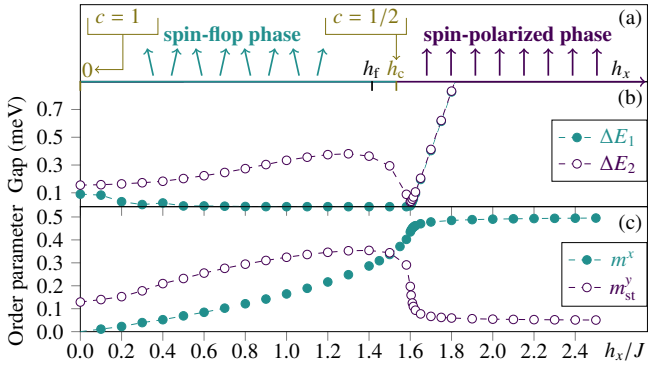

FIG. 1. (a) Schematic phase diagram for $-1 \le \Delta \le 1$. There are two quantum critical points at $h_x = 0$ and h_c, and a classical point at a factoring field h_f, close to h_c. For $\Delta = 0.25$, $h_f \approx 1.58J$ and $h_c \approx 1.6J$. The distance $h_c - h_f$ is exaggerated for clarity. (b) Energy gap, and (c) magnetization and staggered magnetization [38] from DMRG for $\Delta = 0.25$, $J = 0.23$ meV. Non-vanishing ΔE_i, m^y_{st} at $h_x = 0$, and $m^y_{st} \neq 0$ at $h_x \ge h_c$, are due to a finite-size effect in the DMRG calculation.

simulation show good agreement, demonstrating it is an experimentally viable probe of entanglement. We also find the experimental one-tangle to deviate from theory in a potentially revealing manner, whereas the two-tangle extraction requires spin-polarization-resolved experiments.

Transverse-field XXZ chain.— A particularly rich yet simple system is found in the XXZ chain,

$$H = \sum_{i=1}^{L} \left[J\left(S^x_i S^x_{i+1} + S^y_i S^y_{i+1} + \Delta S^z_i S^z_{i+1}\right) + h_x S^x_i \right], \qquad (1)$$

where S^α_i, $\alpha \in \{x, y, z\}$, are spin-1/2 operators, Δ represents exchange anisotropy, and h_x is a uniform magnetic field in the transverse ($\hat{x}$) direction. For Cs_2CoCl_4 we take the parameters $J = 0.23$ meV and $\Delta = 0.25$ [34], but we note $\Delta \approx 0.12$ has been proposed elsewhere [36, 37]. (We consider other Δ values in the Supplemental Material (SM) [38].) The model is also relevant to quantum simulation using cold atoms in optical lattices [39].

For $h_x = 0$, the model can be solved exactly using the Bethe ansatz [40–42]. However, a finite transverse field breaks integrability, and induces a new source of fluctuations when $\Delta \neq 1$. The model is particularly nontrivial in the spin-flop region, $-1 < \Delta < 1$ [43], where its phase diagram contains two QCPs, as shown in Fig. 1(a). At the first QCP, $h_x = 0$, it is equivalent to a gapless Luttinger liquid, which is described by a conformal field theory (CFT) with central charge $c = 1$ [6]. At $h_x > 0$, a gapped, long-range Néel order develops, with a staggered magnetization component, m^y_{st}, along $\hat{y}$, and a magnetization component, m^x, along $\hat{x}$ [38]. This order remains up to a critical field, h_c, where the system is described by a $c = 1/2$ CFT. Above h_c a gapped, nondegenerate spin-polarized (paramagnetic) phase develops and m^x saturates asymptotically. There also exists a "classical" or factoring field, $h_f(\Delta) = J\sqrt{2(1+\Delta)}$, where the ground state is exactly of the classical spin-flop Néel type [44, 45]. At h_f, quantum fluctuations are precisely balanced by the field, and entanglement estimators indicate an entanglement transition [21, 46–48].

The model has previously been studied using Jordan-Wigner fermion mean-field theory [43, 49], exact diagonalization [50], DMRG [49, 51, 52], and quantum Monte Carlo methods [21]. The real-frequency dynamics were studied in Refs. [49, 52], where the mean-field theory [49] was found to give qualitatively different spectra to the DMRG calculation [52] at $h_x \le h_c$. Here we use a $T = 0$ DMRG method described in SM [38]. Care is taken to relate our finite-size ($L = 100$ unless stated otherwise) results in the spin-flop region to the thermodynamic limit [38, 49, 52]. There is a finite-size gap between a unique ground state and the first excited state, $\Delta E_1 = E_1 - E_0$, where E_n is the energy of the nth state. The physical excitation gap is given instead by $\Delta E_2 = E_2 - E_0 > \Delta E_1$, as shown in Fig. 1(b). Magnetization is plotted in Fig. 1(c).

Experimental method.—INS data on a high-quality 9 g solution-grown Cs_2CoCl_4 single crystal were collected using the direct-geometry time-of-flight spectrometer IN6 at Institut Laue-Langevin, with monochromatic incident neutrons of 2.35 meV. Cooling was provided by a dilution refrigerator, and data was collected at 70 mK ($\approx 0.026J \approx 6\ \mu$eV). The sample was oriented with crystallographic b, c-axes in the horizontal scattering plane. Magnetic fields up to 2.5 T were applied along the a-axis using a vertical field cryomagnet. For more details about the experiments, see Ref. [53]. Raw neutron counts were normalized by the integrated quasielastic incoherent scattering to account in a first approximation for neutron absorption from the sample. The nonmagnetic background was modeled and subtracted, and resulting counts were divided by the squared spherical magnetic form factor for Co^{2+}, so resulting intensities are proportional to the purely magnetic scattering cross section.

The Co^{2+} ions in Cs_2CoCl_4 form a Kramers doublet, which can be described by an effective spin $S = 1/2$. Magnetic interactions between Co^{2+} ions are quasi-1D along the b-axis, with exchange interaction much lower than the energy gap to higher crystal field levels, resulting in an effective spin-1/2 Hamiltonian with strong XXZ anisotropy. Finite 3D interchain couplings (estimated to be at least an order of magnitude smaller than J [33, 37, 54, 55]) stabilize long-range order below $T_N = 0.212$ K with ordering wavevector $\boldsymbol{q} = (0, 1/2, 1/2)$, where spins point near the b-axis. Transverse magnetic fields applied along the a-axis suppress this order at $h_c^{exp} = 2.10(4)$ T [34]. This field direction is at an $\approx 40°$ angle to the xy easy plane of the spins. This angle—along with interchain couplings [55, 56]—is expected to renormalize transition fields compared to the in-plane field case considered in Eq. (1), but not to change the qualitative content of the phase diagram. To compare experimental and DMRG results, we scale fields such that $h_c^{DMRG} \approx 1.604J = h_c^{exp}$.

Spectra.—Figure 2 compares INS spectra for Cs_2CoCl_4 with spectra calculated for Eq. (1). For more field strengths

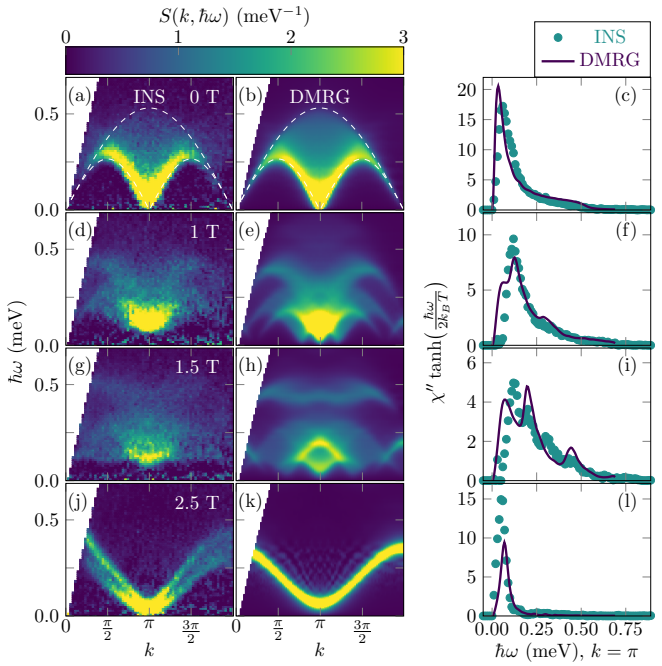

FIG. 2. Left column: INS spectra for Cs_2CoCl_4 at four representative field strengths. Center column: Calculated spectra for the XXZ chain at matching fields, accounting for the experimental polarization factor. Right column: QFI integrand at $k = \pi$. White dashed lines in (a),(b) bound the two-spinon continua. Throughout we designate the wavevector component k along the chain in units of $1/b$.

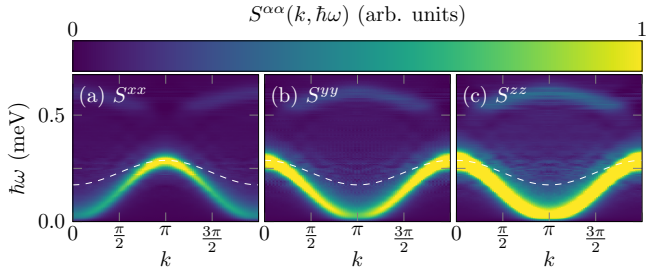

FIG. 3. Theoretical DSF for $J = 0.23$ meV, $\Delta = 0.25$, and $h_x = 1.58114J \approx 2.07$ T $\approx h_f$. Dashed white lines show linear spin wave energy predictions [45]. Agreement with DMRG is excellent at $k = \pi$ in (a), and $k = 0$ in (b),(c). Elsewhere in the Brillouin zone the dispersion is significantly modified by anharmonic terms in the full spin-wave Hamiltonian. Note that the experimental polarization factor has not been applied, see [38, Eq. (S5)].

and processing details, see SM [38]. At low fields the data qualitatively agrees with simulations of the ideal chain model, Eq. (1). Interchain couplings become qualitatively important near h_c, where the field-dependent gap is of similar strength to the interchain exchange. Interchain couplings also produce a band splitting at high fields, as seen in Fig. 2(j)–(k), whereas the DMRG spectrum reduces to a single magnon branch. Hence, we conclude that Cs_2CoCl_4 is 1D-like for weak and intermediate fields. Precise modeling of interchain effects is beyond the scope of this work.

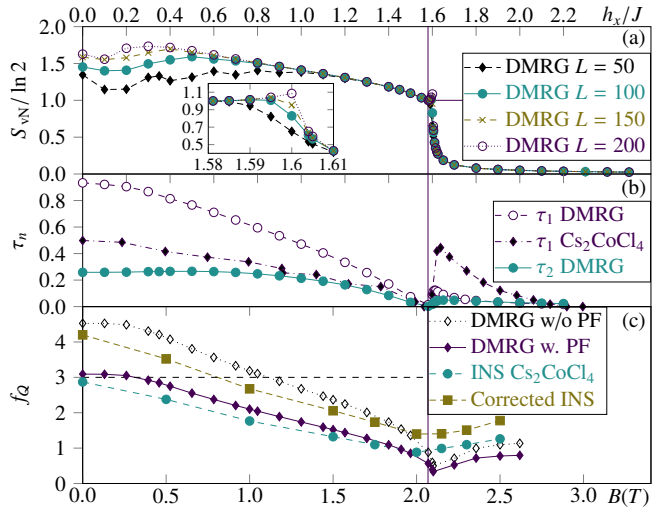

FIG. 4. (a) Entanglement entropy, S_{vN}, from DMRG as a function of h_x. The vertical line indicates the factoring field, where $S_{vN} \approx \ln 2$ (horizontal line). For $h_x > h_c$ there is a steep drop in entropy as the system enters a polarized phase with a non-degenerate ground-state. The inset shows EE near h_c. (b) The approximate one- (τ_1) and two-tangles (τ_2) reach a minimum at h_f. (c) QFI from INS and DMRG $S(k,\hbar\omega)$. Above the horizontal dashed line QFI indicates the presence of *at least* bipartite entanglement. Below it QFI cannot be used to distinguish separable and entangled states. The polarization factor (PF)-corrected INS f_Q line is obtained by scaling f_Q^{INS} by the ratio between the two DMRG f_Q values [38].

At zero field the main contributions to the DSF come from the two-spinon continuum [57, 58], bounds of which are drawn in Fig. 2(a)–(b). At finite field the excitation branches begin to split [Fig. 2(d),(e)], eventually decoupling the upper branch from the low-energy excitations, forming a high-energy feature at $\hbar\omega \geq 0.4$ meV. As Ref. [52] noted, this feature is beyond the mean-field prediction [49]. Here we see it is present in the experimental material [panels (d),(g)] and DMRG [panels (e),(h)]. The intensity of this high-energy feature weakens as h_c is approached from below, and as Fig. 2(j),(k) show, it disappears above h_c. To understand its origin, it is instructive to consider the factoring field. While the ground state at h_f is classical, the dynamics cannot be fully described using linear spin-wave theory (LSWT) [45, 59]. For Eq. (1) the dynamics is LSWT-like only near $k = \pi$ for $S^{xx}(k,\hbar\omega)$, and near $k = 0$ for $S^{yy/zz}(k,\hbar\omega)$ [45]. As Fig. 3 shows, this behavior agrees well with DMRG. The high-energy feature vanishes at k points where LSWT is exact, heavily suggesting its origin is in multi-magnon physics, as proposed in Ref. [52].

Entanglement.—We now investigate the quantum phase transition (QPT) of Eq. (1) and Cs_2CoCl_4 using entanglement measures. Figure 4(a) shows half-chain von Neumann EE. At QCPs, in a system of length L with open boundaries, it is expected to follow the CFT expression [7], $S_{vN} = \frac{c}{6}\ln\left[\frac{L}{\pi}\right] + C$, where C is a non-universal correction. We observe approximately logarithmic scaling at the QCPs, and saturation of S_{vN}

for most non-critical fields [6]. Notably, we find at h_f that $S_{vN} = \ln 2$ to good approximation, consistent with a two-fold classical ground-state degeneracy.

Another sharp signature of the classical state has previously been found using entanglement estimators [21, 47, 48]. We consider one-tangle, τ_1, which quantifies entanglement between a single site and the rest of the system, and two-tangle, τ_2, which quantifies the total pairwise entanglement in the system, and satisfies $\tau_2 < \tau_1$ [19, 60]. For translation-invariant $S = 1/2$ systems, τ_1 can be defined in terms of spin expectation values at a given site j, $\tau_1 = 1 - 4\sum_\alpha \left(\langle S^\alpha_j \rangle\right)^2$. It is useful for interpreting experiments, with the caveat that it is only strictly defined at $T = 0$. We approximate τ_1 by keeping only ferro- and antiferromagnetic ordered moments [38]. The theoretical prediction is shown in Fig. 4(b), along with experimental results obtained [38] using 80 mK ($\approx 0.03J$) data from Ref. [34]. At zero field the experimental τ_1 is reduced from the theoretical value due to magnetic ordering at low temperature, but still indicates substantial entanglement. We discuss the high τ_1 at $B > 2$ T later.

Next, two-tangle is defined as $\tau_2 = 2\sum_{r\neq 0} C_r^2$, where C_r is the concurrence for separation r. C_r for the $S = 1/2$ XXZ model absent spontaneous symmetry breaking ($m^y_{\rm st} = 0$) can be defined [20–22, 46] $C_r = 2\max\{0, C'_r, C''_r\}$, where

$$C'_r = \left|\langle S^y_i S^y_{i+r}\rangle + \langle S^z_i S^z_{i+r}\rangle\right| - \sqrt{\left(\frac{1}{4} + \langle S^x_i S^x_{i+r}\rangle\right)^2 - (m^x)^2}, \tag{2}$$

$$C''_r = \langle S^x_i S^x_{i+r}\rangle + \left|\langle S^y_i S^y_{i+r}\rangle - \langle S^z_i S^z_{i+r}\rangle\right| - \frac{1}{4}. \tag{3}$$

This definition acts as a lower bound for pairwise entanglement in the symmetry-broken state [61, 62]. While such correlation functions are straightforward to compute theoretically, for anisotropic systems they require spin-polarization-resolved techniques to measure experimentally. Since we have not conducted such experiments, we plot only the theoretical τ_2 in Fig. 4(b). (In [38] we simulate a polarized INS experiment by using DMRG to correct for polarization factors (PFs), and estimate concurrence and τ_2 from unpolarized data. We find rough agreement between experiment and theory at low fields, suggesting τ_2 could be a reliable EW with carefully performed experiments.)

Finally, we come to the quantum Fisher information. The QFI density, f_Q, can be expressed [25]

$$f_Q(k,T) = \frac{4}{\pi}\int_0^\infty \mathrm{d}(\hbar\omega)\tanh\left(\frac{\hbar\omega}{2k_BT}\right)\chi''(k,\hbar\omega,T), \tag{4}$$

where the dynamical susceptibility, χ'', is linked to the DSF through the fluctuation-dissipation theorem, $\chi''(k,\hbar\omega,T) = \tanh\left(\hbar\omega/2k_BT\right) S(k,\hbar\omega)$, and $S(k,\hbar\omega)$ is normalized per site (i.e. intensive) according to the the sum rule $\sum_{\alpha\in\{x,y,z\}}\int_{-\infty}^{\infty}\mathrm{d}(\hbar\omega)\int_0^{2\pi}\mathrm{d}k\, S^{\alpha\alpha}(k,\hbar\omega) = S(S+1)$. We are interested in $f_Q(k=\pi)$, which witnesses entanglement associated with the AFM ordering [38]. Importantly, one can derive bounds for f_Q that can only be met by certain classes of entangled states [63–65]. For unpolarized neutron scattering and $S = 1/2$ systems, $f_Q > 3n$, with n a divisor of L, indicates the system is *at least* $n+1$-partite entangled [38].

Figure 4(c) shows QFI determined from INS data normalized against DMRG [38], and from DMRG with and without PF applied. All QFI integrals used $T = 70$ mK. In all cases, maximal f_Q occurs at $h_x = 0$. Unlike τ_1, QFI is insensitive to zero-field magnetic order since elastic peaks are suppressed by the tanh factor in Eq. (4). The results indicate the experimental PF suppresses QFI below the lower bound required to observe bipartite entanglement. Using DMRG intensities we can obtain PF-corrected values [38], which do witness at least bipartite entanglement at the lowest measured fields. This finding highlights that it is easy to underestimate the underlying QFI due to resolution effects, and calls for higher resolution in future experiments. Additionally, it would be valuable to derive tighter bounds on f_Q, even if they do not apply in general [66, 67].

There is qualitative and reasonably quantitative agreement between DMRG and INS QFI at intermediate fields ($\lesssim 1.75$ T), but not at high fields, where interchain coupling causes deviations from ideal 1D behavior. In particular, interchain coupling raises the field required for full polarization, which may explain the observed increase in f_Q above h_c. As $h_x \to \infty$ we expect f_Q to vanish. In addition, the $f_Q^{\rm DMRG}$ minimum occurs at h_c, while the $f_Q^{\rm INS}$ minimum appears at a lower field, likely due to deviations from ideal 1D behavior.

Another deviation from 1D behavior is seen in the large τ_1 at $B > 2$ T [Fig. 4(b)], which could naively be interpreted as a sign that the system has entered a highly entangled state. However, this scenario seems unlikely given the observed f_Q behavior, and suppression of magnetic fluctuations at high field. Instead, τ_1 is likely overestimated in this region due to spin correlations not captured by the ferro- and antiferromagnetic ordered moments used to evaluate τ_1, induced by the small, but finite interchain couplings, which become relevant for fields near h_c where the spin gap is small and the system is near-critical. Capturing such 3D effects is beyond the scope of the current paper.

On its face, the low f_Q values at h_c may seem incompatible with the prediction that CFTs have both large bipartite and multipartite entanglement [68]. QFI near h_c is low because the Néel order parameter becomes vanishingly small near the transition to the polarized state, such that there is little spectral weight available for entanglement [69], and so $f_Q(k=\pi)$ is not an effective witness at this QCP. We generically expect QFI associated with antiferromagnetic ordering vectors to demonstrate significant entanglement only away from paramagnetic transitions. This illustrates a general limitation of EWs: they are not universal [9]. Thus additional EWs would be required to experimentally characterize the entanglement properties of the transverse-field XXZ chain in the entire field range.

Conclusion.—We have contrasted several entanglement measures by applying them to Cs_2CoCl_4 and the transverse-field XXZ chain. Although we are unable to directly witness

genuine multipartite entanglement in Cs_2CoCl_4, the strong agreement between DMRG and INS QFI suggests QFI is already a useful tool for qualitative investigations of entanglement properties, potentially even for topological phases [70–72]. With improved resolution and bounds it can also prove valuable for directly quantifying *local* entanglement in materials. QFI can be used in combination with other EWs, to infer entanglement properties as a control parameter is tuned. Such combinations may be required since paramagnetic QPTs remain inaccessible to $f_Q(k = \pi)$. We find both one-tangle and QFI to be useful measures for inelastic experiments in general, while two-tangle requires polarization analysis. We expect the model-independent approach we outline here, which applies to many spectroscopic techniques and also to higher-dimensional systems, will prove useful in identifying materials with entangled states and highly quantum properties. As the search for materials realizing exotic quantum states continues, EWs can allow clear discrimination between truly entangled, and disordered non-entangled states.

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