Letter

Experimental verification of many-body entanglement using thermodynamic quantities

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The phenomenon of quantum entanglement underlies several important protocols that enable emerging quantum technologies. Entangled states, however, are extremely delicate and often get perturbed by tiny fluctuations in their external environment. Certification of entanglement is therefore immensely crucial for the successful implementation of protocols involving this resource. In this Letter, we propose a set of entanglement criteria for multiqubit systems that can be easily verified by measuring certain thermodynamic quantities. In particular, the criteria depend on the difference in optimal global and local works extractable from an isolated quantum system under global and local interactions, respectively. As a proof of principle, we demonstrate the proposed scheme on nuclear spin registers of up to 10 qubits using the nuclear magnetic resonance architecture. We prepare a noisy Bell diagonal state and noisy Greenberger-Horne-Zeilinger class of states in star-topology systems and certify their entanglement through our thermodynamic criteria. Along the same line, we also propose an entanglement certification scheme in many-body systems when only partial or even no knowledge about the state is available.

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Introduction. Quantum entanglement, identified as a puzzling feature of multipartite quantum systems [1–4], plays the pivotal role in a number of important quantum information protocols [5–11] (see also Ref. [12]). In quantum systems involving more than two parts, entanglement appears in different inequivalent and exotic forms [13,14], that have been proved to be useful in several distributed protocols [15–24]. However, entangled states are fragile and easily lost by external perturbations. The successful implementation of the protocols involving entanglement therefore demands the faithful certification of entanglement. Although the generic *separability problem* is known to be extremely hard even for bipartite systems [25], the negative-partial-transposition (NPT) criterion [26,27] and sometimes measurement of the entanglement witness operator [28] become useful for certifying entanglement. On the other hand, there exist entropic quantities that also serve the purpose of entanglement certification [29,30]. However, these entropic quantities are not directly measurable in experiments, and calculating the value of the witness operator and evaluating the NPT-ness of a state demand complete tomographic knowledge which is practically impossible when a large number of subsystems are involved.

Of late, in a completely different approach, researchers have been trying to identify operationally motivated thermodynamic quantities that can capture the signature of entanglement in multipartite quantum systems [31–37]. In this Letter we show that suitably defined functions of such a thermodynamic quantity, namely the ergotropic work, can serve as bona fide entanglement certifiers for generic N-qubit systems. The optimal amount of work extractable from an isolated quantum system by keeping its entropy unchanged is known as ergotropic work [38]. Depending upon whether a many-body quantum system is addressed globally or its parts are addressed separately, different kinds of ergotropic works can be extracted. Interestingly, entanglement of the initially prepared multipartite state keeps it footprints in the difference of these global and local ergotropic works. Furthermore, while extracting work one might infer the spectral of the state in question. Depending on the available information about the spectral of the global state and its marginals we propose several entanglement certifiers. As proof of principle, we implement the proposed thermodynamic entanglement criterion on nuclear spin registers of up to ten qubits via nuclear magnetic resonance (NMR) architecture. In particular, the star-topology systems allow preparation of a Greenberger-Horne-Zeilinger (GHZ) class of states in large registers [39–41]. We prepare a two-qubit Bell diagonal state and noisy states comprising a singlet/GHZ state and white noise, and certify their entanglement through our proposed criteria.

Theory and framework. The state of an N-qubit system is described by a density operator $\rho_{A_1\cdots A_N} \in \mathcal{D}[(\mathbb{C}^2)^{\otimes N}]$, where $\mathcal{D}(\mathbb{H})$ denotes the set of positive trace-one operators acting on the Hilbert space $\mathbb{H}$. A state is called fully separable if it is a probabilistic mixture of a fully product state, i.e., $\rho_{A_1\cdots A_N} = \sum_i p_i (\bigotimes_{j=1}^N |\psi^i_{A_j}\rangle\langle\psi^i_{A_j}|)$, with $|\psi^i_{A_j}\rangle \in \mathbb{C}^2_{A_j} \equiv \mathbb{C}^2$. States lying outside the set of fully separable states are entangled. However, different kinds of entanglement are possible in multiqubit systems. Let $\mathcal{S}[X|X^c]$ denote the set of states separable across the X-vs-X^c bipartite cut, where X contain κ parties together and X^c contains the remaining parties; $\kappa \in \{1,\dots,N-1\}$. States lying outside $\mathcal{S}[X|X^c]$ contain entanglement across X-vs-X^c bipartition.

When such an isolated system evolves from an initial state ρ to a lower-energy state σ, the difference in energies can be extracted as work. The study of this topic dates back to the

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late 1970s [42,43] and recently it has gained renewed interest [44–47]. Consider an N-qubit system governed by a noninteracting Hamiltonian $H = \sum_{l=1}^{N} \tilde{H}_l$, where $\tilde{H}_l := \mathbf{I}_1 \otimes \cdots \otimes \mathbf{I}_{l-1} \otimes H_l \otimes \mathbf{I}_{l+1} \otimes \cdots \otimes \mathbf{I}_N$, with $H_l = \sum_{i=0}^{1}(E_l + i\alpha_l)|i\rangle\langle i|$ and $|0\rangle$ and $|1\rangle$ being the energy eigenkets with respective eigenvalues E_l and $E_l + \alpha_l$; $\mathbf{I}_j$ is the identity operator on the jth qubit. Evolution from the initial state to the final state is governed through a cyclic unitary $U(\tau)$ generated by switching on a time-dependent interaction.

The optimally extractable work, called ergotropy, amounts to $W(\rho) = \mathrm{Tr}[\rho H] - \min_{U(\tau)} \mathrm{Tr}[U(\tau)\rho U^\dagger(\tau)H]$, where optimization is considered over all unitaries. As it turns out during optimal work extraction the system evolves to the passive state ρ^P, and accordingly we have $W(\rho) := E(\rho) - E(\rho^P) = \mathrm{Tr}[\rho H] - \mathrm{Tr}[\rho^P H]$ [42,43]. The passive state is the lowest energetic state with a spectral identical to the initial state. Moreover, it is diagonal in the energy basis where higher-energy states are less populated. In the multipartite scenario different parts of the system can be probed separately leading to several inequivalent configurations for work extraction. For instance, in the X-vs-X^c configuration, with X containing κ parties together, the optimal extractable work from the X subsystem is given by $W_{[\kappa]}(\rho_X) := \mathrm{Tr}[\rho_X H_X] - \mathrm{Tr}[\rho_X^P H_X]$, where $\rho_X := \mathrm{Tr}_{X^c}(\rho) \in \mathcal{D}[(\mathbb{C}^2)^{\otimes\kappa}]$, $\rho \in \mathcal{D}[(\mathbb{C}^2)^{\otimes N}]$, H_X is the Hamiltonian of the subsystem X, and ρ_X^P is the passive state corresponding to ρ_X, with $W_{[N]}(\rho)$ simply denoted as $W(\rho)$.

We will denote the spectral for a generic N-qubit state ρ as $\vec{t}_\rho \equiv \{t_j\}_{j=0}^{2^N-1}$, arranged in decreasing order. The system's Hamiltonian H can be reexpressed as $H = \sum_{j=0}^{2^N-1}(E_g + n_j)|e_j\rangle\langle e_j|$, where $|e_0\rangle = |0\rangle^{\otimes N}$ is the ground state with an energy value $E_g = \sum_{l=1}^{N} E_l$, and the energy eigenvalues are arranged in increasing order, i.e., $n_{j+1} \geqslant n_j$, $\forall\, j$, with $n_0 = 0$. The highest excited state $|e_{2^N-1}\rangle = |1\rangle^{\otimes N}$ has an energy value $E_g + \sum_{l=1}^{N} \alpha_l$. The spectral of the subsystem X will be denoted as $\vec{x}_{\rho_X} \equiv \{x_j\}_{j=0}^{2^\kappa-1}$, with its Hamiltonian reexpressed as $H_X = \sum_{j=0}^{2^\kappa-1}(E_g^X + m_j)|f_j\rangle\langle f_j|$. While extracting work in the X-vs-X^c configuration, we can evaluate the thermodynamic quantity

$$\Delta_{X|X^c} := W(\rho) - W_{[\kappa]}(\rho_X) - E(\rho_{X^c}) + E_g^{X^c}. \tag{1}$$

Here, the first three terms are state dependent and their values can be evaluated through experiment; the last term designates the ground-state energy of the Hamiltonian of the X^c part. We are now in a position to provide our thermodynamic entanglement criteria (for a proof we defer to the Supplemental Material [48]).

Theorem 1. An N-qubit state separable across the X-vs-X^c bipartition satisfies

$$\Delta_{X|X^c} \leqslant \sum_{i=1}^{2^\kappa-1}(m_i - m_1)x_i + \sum_{i=1}^{2^N-1}(m_1 - n_i)t_i := \delta_{X|X^c}^{GL}, \tag{2a}$$

$$\Delta_{X|X^c} \leqslant \sum_{i=1}^{2^\kappa-2}(m_i - n_i)t_i + \sum_{i=2^\kappa-1}^{2^N-1}(m_{2^\kappa-1} - n_i)t_i := \delta_{X|X^c}^{G}, \tag{2b}$$

where $m_{i+1} \geqslant m_i$ for $i \in \{0, 2^\kappa - 1\}$, $n_{i+1} \geqslant n_i$ for $i \in \{0, 2^N - 1\}$, and $m_{2^\kappa-1} = \sum_{i=1}^{\kappa} \alpha_i$.

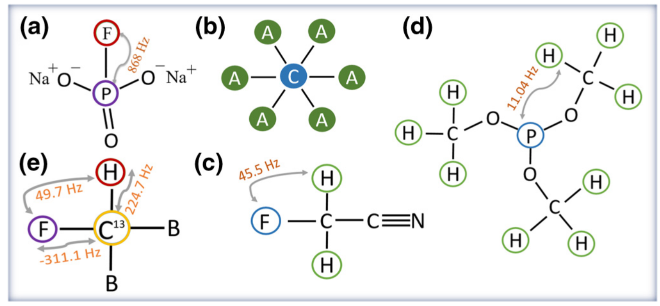

FIG. 1. (a) Two-qubit system sodium fluorophosphate (NAFP, with ^{19}F and ^{31}P being first and second qubits) used in experiment I. (b) The star-topology configuration, wherein each ancillary spin A interacts with the central spin ^{13}C. (c), (d) Three-qubit star-system fluoroacetonitril (FAN) and ten-qubit star-system trimethylphosphate (TMP) used in experiment II. (e) Dibromofluoromethane (DBFM, with ^{1}H, ^{13}C, and ^{19}F being first, second, and third qubits) used in experiment III.

The violation of any of the conditions in Theorem 1 certifies entanglement across X-vs-X^c bipartition. Please note that examining condition (2a) requires knowledge of the global and local spectrals $\vec{t}_\rho$ and $\vec{x}_{\rho_X}$. Quite interestingly, this thermodynamic criterion turns out to be a special case of the Nielsen-Kempe entanglement criterion [49] (see Remark 1 in the Supplemental Material [48]). The separability bound $\delta_{X|X^c}^{G}$ in (2b) depends only on the global spectral of the given state (and hence the superscript G) and generally turns out to be a weaker than (2a). One can also come up with a thermodynamic entanglement criterion that does not involve the knowledge about the state, albeit it will be weaker than the state-dependent criteria (see Remark 2 in the Supplemental Material [48]). The state-independent separability bound will be denoted as $\delta_{X|X^c}^{I}$. For instance, for the identical three-qubit noisy GHZ states $\rho_\lambda[3] := (1-\lambda)\mathbb{I}/8 + \lambda\,|\psi_3\rangle\langle\psi_3|$, where $|\psi_3\rangle := (|000\rangle + |111\rangle)/\sqrt{2}$ and all the subsystems are governed through identical Hamiltonian, criterion (2b) can detect entanglement for $\lambda > 3/7$, whereas a state-independent criterion detects entanglement for $\lambda > 0.66$. In the remaining part of this Letter we investigate the aforesaid thermodynamic entanglement criteria by preparing specific classes of entangled states in NMR setup.

Experiment I: Two-qubit Bell diagonal states. In our first experiment we deal with two-qubit Bell diagonal states prepared using the system NAFP [Fig. 1(a)] dissolved in D_2O. All the experiments were carried out on a 500-MHz Bruker NMR spectrometer at an ambient temperature of 300 K. The Hamiltonian of the system consisting of the internal part and the rf drive reads as $H_{12} = H_{12}^{\mathrm{int}} + H_{12}^{\mathrm{rf}}$, where $H_{12}^{\mathrm{int}} = -\omega_F I_{1z} - \omega_P I_{2z} + 2\pi J I_{1z} I_{2z}$ and $H_{12}^{\mathrm{rf}} = \Omega_F(t) I_{1x} + \Omega_P(t) I_{2x}$, with $I_{ix} = \sigma_{ix}/2$, $I_{iy} = \sigma_{iy}/2$, $I_{iz} = \sigma_{iz}/2$. Here, (ω_F, ω_P) and (Ω_F, Ω_P) respectively denote the Larmor frequencies and rf amplitudes of (^{19}F, ^{31}P), $\hbar = 1$, and J is the scalar coupling constant. We prepare Bell diagonal states with two independent controllable parameters β and γ. We start with the thermal state, which under a high-field, high-temperature approximation reads as $\rho_{\mathrm{th}} = \mathbf{I}/4 + \epsilon_P(\frac{\gamma_F}{\gamma_P} I_{1z} + I_{2z})$, where $\epsilon_P = \gamma_P B_0/4k_B T$. The method of spatial averaging yields us pseudopure state (PPS) $|11\rangle\langle 11|^{\mathrm{pps}} = (1-\epsilon)\mathrm{I}/4 +$

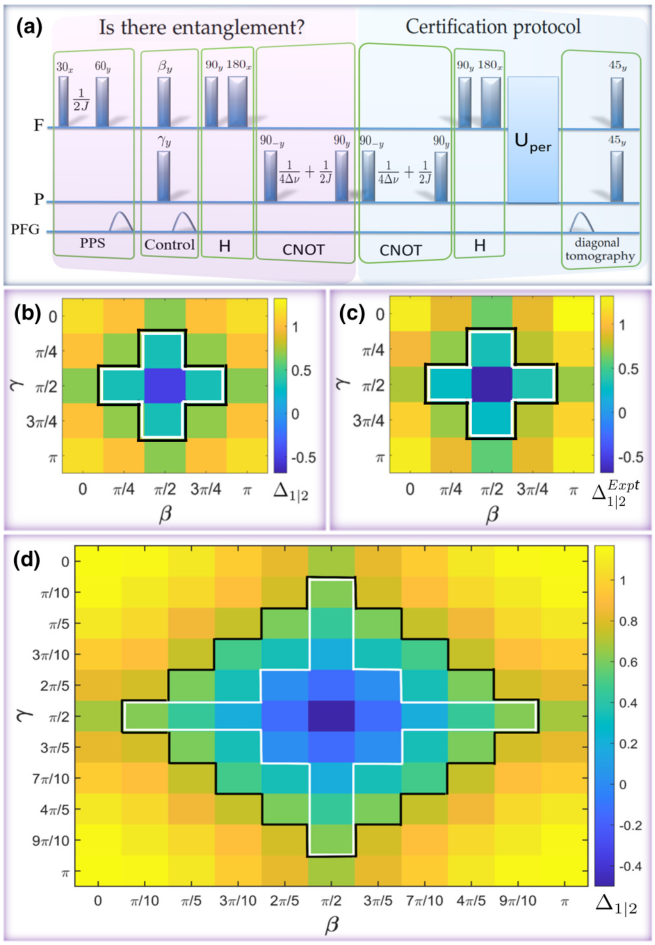

FIG. 2. (a) The NMR pulse sequence to prepare the Bell diagonal state with control parameter β and γ. Here, PFG is the pulsed-field gradient and $\Delta\nu$ is the resonance offset of both ^{19}F and ^{31}P. One of the operations ($U_{\rm per} \circ H \circ$ CNOT) takes the Bell diagonal state to its passive state. A detailed explanation of this sequence is given in the Supplemental Material [48]. (b) Gradient-color plot for theoretical values of $\Delta_{1|2}$ (in units of ω_P) vs the control parameters β and γ. States outside the white line (inner perimeter) are entangled as $\Delta_{1|2} > \delta^G_{1|2}$. For the states outside the black line (outer perimeter) $\Delta_{1|2} > \delta^I_{1|2}$ and hence they are also entangled. (c) Gradient-color plot for the experimental values of $\Delta^{\rm Expt}_{1|2}$ with estimated errors of $\pm 0.1\omega_P$. Here, the error originates both from the spin system as well as the NMR hardware, so accordingly we have estimated the random error from the experimental NMR spectrum corresponding to the least signal-to-noise ratio providing a useful upper bound for errors. (d) 11×11 pixel theoretical gradient-color plot of $\Delta_{1|2}$: Evidently the state-independent certification scheme is weaker than the state-dependent scheme.

$\epsilon\,|11\rangle\langle 11|$, with $\epsilon \ll 1$ [Fig. 2(a)]. Within the paradigm of PPS we set $\epsilon = 1$ and realize the effective state $|11\rangle\langle 11|$ [50]. Subsequently we prepare the two-parameter Bell diagonal state

$$\rho_{12} = \sum_{i,j=0}^{1} p_{ij}\, |\mathcal{B}_{ij}\rangle\langle \mathcal{B}_{ij}|\,, \tag{3}$$

$$|\mathcal{B}_{0j}\rangle := (|00\rangle + (-1)^j\,|11\rangle)/\sqrt{2}, \quad p_{00} := [\mathrm{S}(\beta/2)\mathrm{S}(\gamma/2)]^2,$$

$$|\mathcal{B}_{1j}\rangle := (|01\rangle + (-1)^j\,|10\rangle)/\sqrt{2}, \quad p_{01} := [\mathrm{S}(\beta/2)\mathrm{C}(\gamma/2)]^2,$$

$$p_{10} := [\mathrm{C}(\beta/2)\mathrm{S}(\gamma/2)]^2, \quad p_{11} := [\mathrm{C}(\beta/2)\mathrm{C}(\gamma/2)]^2,$$

where $\mathrm{S}(\star) := \sin(\star)$ and $\mathrm{C}(\star) := \cos(\star)$. Further details on the preparation circuit are provided in the Supplemental Material [48]. To evaluate the quantity in Eq. (1), we evolve the state ρ_{12} into its passive state, the lowest energetic state. While this requires an optimization over all possible unitary operations, in the Supplemental Material [48] we argue that for a generic two-qubit Bell diagonal state this can be achieved by considering only 24 permutation operations ($U_{\rm per}$). This way we obtain the value of the quantity $\Delta^{\rm Expt}_{1|2}$ experimentally. Notably, as the number of qubits increases, the optimization over the set of possible unitary operations expands significantly, and hence the scalability issues persists. Nonetheless, for two-qubit case, arranging $\{p_{ij}\}$ in descending order and denoting the resulting vector as $\vec{t} \equiv \{t_k\}_{k=0}^{3}$, theoretically we have

$$\begin{Bmatrix} \Delta_{1|2} = (1.162 - 2.324t_2 - 3.324t_3 - t_1)\omega_P, \\ \delta^G_{1|2} = (1.324t_1 - t_3)\omega_P, \quad \delta^I_{1|2} = 0.662\omega_P \end{Bmatrix}. \tag{4}$$

Note that the evaluation of the quantity $\delta^G_{1|2}$ in Eq. (4) demands knowledge of the global spectral, whereas $\delta^I_{1|2}$ is state independent. Entanglement is certified whenever $\Delta_{1|2}$ is strictly greater than any one of these quantities. Varying the parameter β and γ we show the entanglement certification in Fig. 2 through a gradient-color plot. As expected and also evident from the plot, the state-independent certification scheme turns out to be weaker than the state-dependent scheme. For instance, the specific values of $\beta = 2\pi/5$ and $\gamma = 3\pi/10$ yield $\Delta_{1|2} = 0.338\omega_P$, which is strictly less than $\delta^I_{1|2} = 0.662\omega_P$, but greater than $\delta^G_{1|2} = 0.292\omega_P$. This is not visible in Figs. 2(b) and 2(c) due to limited pixel resolution, but can be seen in Fig. 2(d). It is important to note that our entanglement certification scheme does not require tomographic knowledge of the state, but rather it is obtained by evaluating the expected energies of the given state and the unitarily evolved state. More specifically, our thermodynamic entanglement criteria can certify entanglement in a given state without requiring the information of the population frequencies for different energy eigenstates.

Experiment II: Multiqubit systems. Multiqubit entangled states within the NMR architecture can be prepared in a star-topology register (STR) [40]. STR involves a central qubit C (first qubit) uniformly interacting with a set of $N-1$ identical satellite qubits A [see Fig. 1(b)]. The central qubit can be selectively addressed as it is realized by a different nuclear isotope. The ancillary qubits being indistinguishable can be addressed globally only. STR allows the efficient preparation of the entangled GHZ state [39]. The STR Hamiltonian along with the pulse sequence dynamics is described in Supplemental Material [48].

We carry out experiments on the following two systems: (i) three-qubit STR using FAN, wherein ^{19}F spin is the central qubit and two ^{1}H spins are the satellite qubits, with $J_{\rm CA} = 45.5$ Hz; (ii) ten-qubit STR using TMP, wherein ^{31}P spin is the central qubit and nine ^{1}H spins are the satellite qubits, with $J_{\rm CA} = 11.04$ Hz (see Fig. 1). After preparing the noisy state

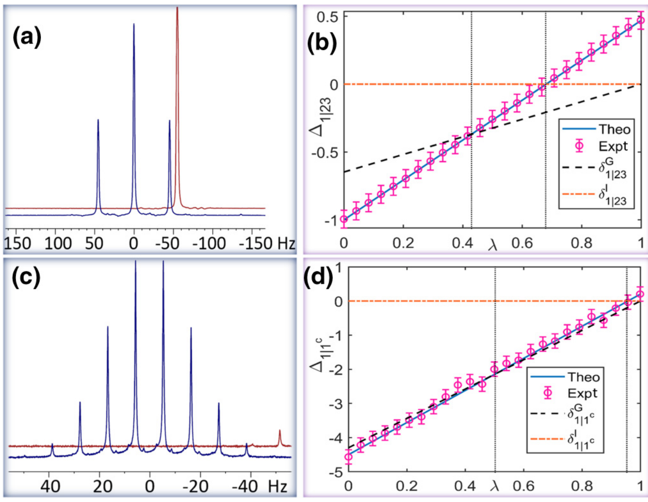

FIG. 3. (a) ^{19}F spectra of FAN corresponding to the one-pulse experiment on a thermal state (front) and to the three-qubit GHZ state (back). (b) Plot $\Delta_{1|23}$ (in the unit of ω_H) vs purity λ for three-qubit noisy GHZ states. Comparing the values of $\Delta_{1|23}$ and $\delta^G_{1|23}$, $\delta^I_{1|23}$, we identify the threshold values marked by the dotted lines: $\lambda = 3/7$ and $\lambda = 0.68$, respectively. Above these thresholds, the state exhibits entanglement. (c) ^{31}P spectra of TMP corresponding to the one pulse experiment (front), and to GHZ (back). (d) $\Delta_{1|1^c}$ vs purity λ for the ten-qubit GHZ class. Here, $\lambda = 0.499$ and $\lambda = 0.957$ for $\delta^G_{1|1^c}$ and $\delta^I_{1|1^c}$ marks the entanglement threshold boundary. The error bar represents the estimated random error due to noise.

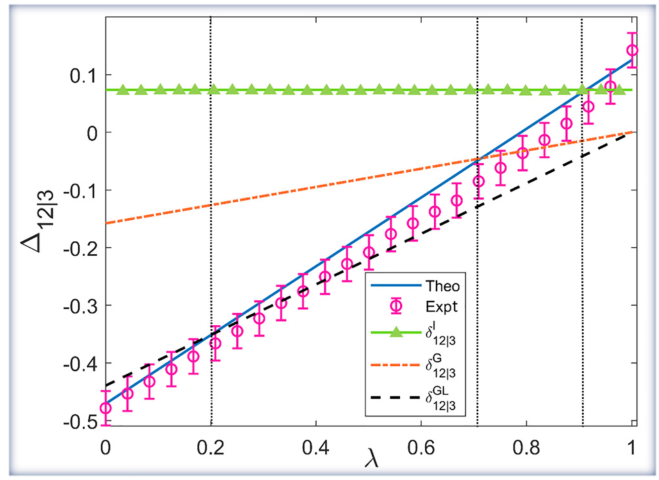

FIG. 4. Plot of $\Delta_{12|3}$ (experimental as well as theoretical), $\delta^{GL}_{12|3}$, $\delta^G_{12|3}$, and $\delta^I_{12|3}$ against the noise parameter λ. The state-independent, the global, and the global-local separability conditions certify entanglement for the parameter ranges $\lambda > 0.91$, $\lambda > 0.71$, and $\lambda > 1/5$, respectively, establishing hierarchy among these conditions.

$\rho_\lambda[N] = (1-\lambda)\mathbf{I}/2^N + \lambda\,|\psi_N\rangle\langle\psi_N|$, with $|\psi_N\rangle := (|0\rangle^{\otimes N} + |1\rangle^{\otimes N})/\sqrt{2}$, we test entanglement across the C-vs-A bipartition considering $N = 3$ and 10, respectively, for FAN and TMP. In doing that, we experimentally determine the thermodynamic quantity $\Delta_{1|1^c}$ along with global spectral-dependent separability bound $\delta^G_{1|1^c}$ and state-independent bound $\delta^I_{1|1^c}$. Subsequently, we find out the ranges of λ for which $\Delta_{1|1^c}$ exceeds these bounds, and accordingly entanglement across C vs A gets certified. Experimental results are shown in Fig. 3, and the detailed analysis is deferred to the Supplemental Material [48]. Importantly, separability bounds generally rely on the Hamiltonian of the individual systems. This is noteworthy for the state-independent bound in particular. As the central qubit has a greater energy gap between the ground and excited states, the range of entanglement proportionally expands (see Remark 3 in the Supplemental Material [48]).

Experiment III: Global versus global-local separability bounds. Here, we experimentally establish the difference between the global-local separability bound of condition (2a) and the global separability bound of condition (2b) as stated in Theorem 1 using DBFM [see Fig. 1(e)], dissolved in Acetone-d6 as the three-qubit spin system. Here, ^{1}H, ^{13}C are together treated as X, whereas ^{19}F is treated as X^c. The procedure to obtain global and local spectral is discussed in the Supplemental Material [48]. In Fig. 4 we plot $\Delta_{12|3}$ (experimental as well as theoretical), $\delta^{GL}_{12|3}$, $\delta^G_{12|3}$, and $\delta^I_{12|3}$ against the noise parameter λ.

Discussion. Manipulating entanglement efficiently in a multipartite system is essential for emerging quantum technologies that involve distributed quantum information protocols. Continuous research effort is going on to this aim with different quantum architectures [51–55]. Certifying entanglement is a crucial step for the successful implementation of many quantum protocols. Along with a device-independent certification scheme through Bell tests [56–61], there exists a device-dependent witness-based method of entanglement certification [62,63]. However, implementing those methods is quite challenging in practice when the quantum systems are composed of many subsystems.

In that respect our proposed thermodynamic criteria are less demanding. It provides a way to certify entanglement by measuring global and local ergotropic works. We experimentally validate the proposed thermodynamic entanglement criterion in NMR architecture by considering particular classes of two-qubit, three-qubit, and ten-qubit noisy entangled states. In comparison with Ref. [64] that relies on the full density state tomography of pure states and Ref. [65] that is limited to bipartite systems, our method achieves certification for multiqubit mixed states with three different bounds based on the thermodynamic quantifiers of the system. Our thermodynamic approach opens up an easy avenue to certify entanglement even when the knowledge about the state in question is not available. While we have invoked the pseudopure paradigm for our ensemble architecture, similar protocols can be easily set up for other architectures with access to different degrees of state purity. For instance, entanglement-enhanced quantum sensing by optical probes [11] or NV centers [66] may be benefited from prior certification of entanglement. At this point, we would like to point out that the study of ergotropy is constantly advancing with different quantum architectures, such as optical mode [67] and bosonic Gaussian models [68,69]. It will be therefore interesting to test entanglement in those physical systems using our proposed criteria. The recent work of Ref. [70] wherein the coarse-grained measurement scheme is proposed is worth mentioning at this point. Our study welcomes a number of other questions for future research. For instance, generalizing our criteria for systems with arbitrary local dimensions and generalizing to capture more exotic kinds of entanglement, such as genuine multipartite entanglement, would be quite important. While the local passivity in our case is studied under local unitary operations, a more general notion of strong local passivity is introduced by considering a more general local quantum operation [71–73]. Obtaining entanglement certification criteria under this generic consideration could also be quite interesting.

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