Letter

Grüneisen parameter as an entanglement compass and the breakdown of the Hellmann-Feynman theorem

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The Grüneisen ratio Γ, i.e., the singular part of the ratio of thermal expansion to the specific heat, has been broadly employed to explore both finite T and quantum critical points (QCPs). For a genuine quantum phase transition (QPT), thermal fluctuations are absent and thus the thermodynamic Γ cannot be employed. We propose a quantum analog to Γ that computes entanglement as a function of a tuning parameter λ and show that QPTs take place only for systems in which the ground-state energy depends on λ nonlinearly. Furthermore, we demonstrate the breakdown of the Hellmann-Feynman theorem in the thermodynamic limit at any QCP. We showcase our approach using the quantum one-dimensional Ising model with a transverse field and Kane's quantum computer. The slowing down of the dynamics and thus the "creation of mass" close to any QCP/QPT is also discussed.

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The investigation of the thermodynamic response close to a finite-T critical end point is a widely explored branch [1]. A key physical quantity related to the critical behavior of matter is the correlation length ξ. As the temperature T and tuning parameter approach their critical values, $\xi \to \infty$ [1]. This is because of the emergence of large fluctuations of the order parameter and thus intrinsic entropy accumulation in this regime [2,3]. Such enhancement of the entropy in the vicinity of critical points can be used to attain giant caloric effects [3]. Interestingly enough, critical phenomena can be observed in various fields, including complex biological systems, such as the brain and the cognition development [4,5], which has provided a new foundation for modern thermodynamics. Apart from ξ, an appropriate tool to explore critical phenomena is the so-called Grüneisen ratio Γ_g [3,6],

$$\Gamma_g = \frac{\alpha_g}{c_g} = -\frac{1}{T}\frac{\left(\frac{\partial^2 F}{\partial T \partial g}\right)}{\left(\frac{\partial^2 F}{\partial T^2}\right)_g} = -\frac{1}{T}\frac{\left(\frac{\partial S}{\partial g}\right)_T}{\left(\frac{\partial S}{\partial T}\right)_g} = \frac{1}{T}\left(\frac{\partial T}{\partial g}\right)_S, \quad (1)$$

where α_g and c_g are, respectively, the thermal expansivity and heat capacity, both at a constant value of a tuning parameter g; F and S are, respectively, the Boltzmann free energy and entropy. For the case in which g is pressure, Γ_g quantifies the barocaloric effect. Stress, magnetic, and electric field can be used as g, being, respectively, each of them associated with the elastic-, magnetic-, and electric-Grüneisen parameters [3,7].

The peculiar behavior of Γ_g close to finite-T critical points, namely an enhancement and sign change upon varying g, is well established in the literature [8,9]. The same holds true for quantum critical points (QCPs) *near* absolute zero [2,10]. In this context, it is tempting to ask, "what is the expression for Γ_g at exactly $T = 0$ K?" This is the key question we have addressed in the present Letter. Note that as $T \to 0$ K, both α_g and $c_g \to 0$ because of the third law of thermodynamics [1], being clear that in such a case, Γ_g is undetermined. At first glance, one could consider that $T \to 0\,\text{K} \Rightarrow \Gamma_g \to \infty$ is a mere consequence of the vanishing of the energy scale associated with the thermal energy [cf. Eq. (1)]. It turns out that at $T = 0$ K, Eq. (1) no longer holds true and the classical concept of caloric effects is no longer applicable. Regarding QCPs, the behavior of Γ_g has been investigated, in practice, at ultralow temperatures since typical order parameter fluctuations associated with QCPs are accessible in the T range of a few mK [2,10]. However, as pointed out previously, the expression for Γ_g is no longer valid exactly at $T = 0$ K, thus preventing the analysis of a genuine QCP.

Here, we propose an analogous expression for Γ_g which is universal, valid at absolute zero temperature, and might be relevant for quantum computing. To this end, we start by considering a generalized Hamiltonian given by

$$H = H_0 + H_1(h) + H_2(g), \quad (2)$$

where H_0 is the unperturbed Hamiltonian, and $H_1(h)$ and $H_2(g)$ are perturbed terms whose eigenenergies E_i depend on the tuning parameters h and g, e.g., magnetic or electric fields. Also, our proposal covers the case of h and g being

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two competing energy scales. Since we are dealing with a quantum system, we propose, in analogy to α_g, to consider the numerator of the first part of Eq. (1) as $\frac{\partial^2 E_0}{\partial h \partial g}$, where E_0 is the ground-state energy, and for the denominator, we propose, in analogy to c_g, to consider $-h\frac{d^2E_0}{dh^2}$, following similar arguments presented in Ref. [11]. The g and h derivatives of E_0 are related to the Hellmann-Feynman theorem, namely $[dE(\nu)/d\nu] = \langle\psi_n|\frac{dH(\nu)}{d\nu}|\psi_n\rangle$, where ν is a general tuning parameter and ψ_n a single-particle wave function [12]. Hence, upon connecting the dots, a mathematical expression for the Grüneisen parameter that is valid at $T = 0$ K reads

$$\Gamma^{0\,\mathrm{K}} = -\frac{\left(\frac{\partial^2 E_0}{\partial h \partial g}\right)}{h\left(\frac{\partial^2 E_0}{\partial h^2}\right)}, \tag{3}$$

whose dim is $[\mathrm{g}]^{-1}$. Although the second and cross derivatives of E_0 with respect to the tuning parameters have been employed to investigate quantum phase transitions (see, e.g., Ref. [13]), to the best of our knowledge, a proper discussion about its genesis, i.e., its correspondence with the Grüneisen parameter, was still lacking. In a real physical system, it is natural to expect that E_0 might be more sensitive with respect to either h or g depending on the particular physical aspects of the investigated system. Upon considering that E_0 is changed as a result of the variation of h and/or g, the quantum entropy of the system is affected. More precisely, changing E_0 leads to a variation of the von Neumann entropy $S_N = -\mathrm{Tr}(\rho \ln \rho)$ [14], where $\rho = \sum_j p_j|\psi_j\rangle\langle\psi_j|$ is the density matrix and p_j is the probability of each state $|\psi_j\rangle$. This can be demonstrated upon making use of the average energy of the system $\langle H\rangle$ in a state $|\psi\rangle$ in terms of ρ, i.e., $\langle H\rangle = \mathrm{Tr}(\rho H)$ [15]. Considering that the system lies in the ground state, its average energy is E_0 and thus its variation due to a change of h and g means that ρ is also changed. As a consequence, if ρ is varied, S_N is varied as well. Hence, we write $\Gamma^{0\,\mathrm{K}}$ in terms of S_N [see Eq. (1)], namely

$$\Gamma^{0\,\mathrm{K}} = -\frac{\left(\frac{\partial S_N}{\partial g}\right)_h}{h\left(\frac{\partial S_N}{\partial h}\right)_g} = -\frac{\left(\frac{\partial\,\mathrm{Tr}(\rho\ln\rho)}{\partial g}\right)_h}{h\left(\frac{\partial\,\mathrm{Tr}(\rho\ln\rho)}{\partial h}\right)_g}. \tag{4}$$

In our case, we have considered S_N as the global entanglement, which is defined as $S_N = -(\partial E_0/\partial h)$, in direct analogy to the thermodynamic case [1]. Since S_N quantifies the entanglement of a quantum system [16], the definition of $\Gamma^{0\,\mathrm{K}}$ in Eq. (4) can be regarded as an *entanglement compass* to explore QCPs upon varying h and g.

The detection of genuine quantum phase transitions (QPTs) and QCPs is key in the context of quantum computing, since close to both QPTs and QCPs, the system's states are highly entangled [17]. This is corroborated by the so-called concurrence, which quantifies the degree of entanglement between two qubits [16–18]. Hence, $\Gamma^{0\,\mathrm{K}}$ can also compute the concurrence between two qubits upon varying h and/or g [cf. Eq. (4)]. In Ref. [17], the field switching of the entanglement for the canonical one-dimensional (1D) Ising model under a transverse field (IMTF) is discussed, demonstrating that close to the QCP the entanglement is strongly magnetic field dependent. Hence, we consider that $\Gamma^{0\,\mathrm{K}}$ is the appropriate physical quantity to quantify the latter. It turns out that applying $\Gamma^{0\,\mathrm{K}}$ [Eq. (3)] to the $S = 3/2$ Hamiltonian with a spin-orbit interaction for the case of a Brillouin-like paramagnet [19,20], a hydrogen atom under an external magnetic field [21], a Kondo lattice [22], and Majorana nanowires [23] leads to $\Gamma^{0\,\mathrm{K}} \to \infty$ for $h, g \to 0$, which is the trivial case, i.e., no critical features show up for finite h or g. Also, their cross and second derivatives of E_0 with respect to h and g do not present any divergence, which suggests that QCPs/QPTs are absent for these systems.

Regarding the case in which E_0 is nonlinear with respect to the tuning parameter, $\Gamma^{0\,\mathrm{K}}$ can be employed to probe QCPs and QPTs. This is the case of the 1D IMTF, whose Hamiltonian is given by [24,25]

$$H = -B\sum_i S_i^x - J\sum_i S_i^z S_{i+1}^z, \tag{5}$$

where B is the modulus of the transverse magnetic field, S_i^x is the spin operator at the i site along the x axis, J is the exchange coupling constant between nearest-neighbor magnetic moments, and S_i^z and S_{i+1}^z are the spin operators along the z axis at sites i and $i+1$, respectively. Before computing $\Gamma^{0\,\mathrm{K}}$, E_0 must be obtained for this case. At this point, it is worth mentioning that Γ, but not $\Gamma^{0\,\mathrm{K}}$, was already computed for this case in the vicinity of the QCP showing a divergentlike behavior when the magnetic energy matched J, i.e., for $\lambda = J/B = 1$ [26]. To compute E_0 for Eq. (5), a textbook Jordan-Wigner transformation can be employed and the Hamiltonian is diagonalized, so that [27]

$$H = B\sum_k \Lambda_k \eta_k^\dagger \eta_k - \frac{B}{2}\sum_k \Lambda_k, \tag{6}$$

where $\Lambda_k = 2\sqrt{1+\lambda^2-2\lambda\cos(k)}$ and $\eta_k^\dagger$ and η_k are, respectively, the creation and annihilation operators, being then E_0 per site given by $E_0 = -(B/\pi)\sum_k \Lambda_k$ [25], which in turn can be rewritten in terms of an elliptical integral in the thermodynamic limit [28],

$$E_0 = -\frac{B}{\pi}\int_0^\pi \Lambda_k dk = -\frac{4B\sqrt{\lambda^2+2\lambda+1}\,\mathrm{Ell}\left[-\frac{4\lambda}{(\lambda-1)^2}\right]}{\pi\sqrt{\frac{\lambda^2+2\lambda+1}{(\lambda-1)^2}}}, \tag{7}$$

where $\mathrm{Ell}[-4\lambda/(\lambda-1)^2]$ is the so-called complete elliptic integral of the second kind [29]. Hence, upon choosing $h = B$ and $g = J$, $\Gamma^{0\,\mathrm{K}}$ can be computed exactly. It turns out that for the 1D IMTF both E_0 derivatives in the numerator and denominator of Eq. (3) are exactly the same, which is in contrast with the thermodynamic case [cf. Eq. (1)]. This is merely a consequence of the fact that E_0 has the same dependence on J and B. Figure 1 depicts the B and J derivatives of E_0 as a function of λ. Remarkably, for $\lambda = 1$, $\frac{\partial^2 E_0}{\partial J \partial B}$ and $-B\frac{\partial^2 E_0}{\partial B^2} \to \infty$, a fingerprint of a QCP governing the change from a ferromagnetic to a quantum paramagnetic phase [26]. Also, the result of S_N as a function of λ is depicted in Fig. 1. An enhancement in S_N is observed for $\lambda = 1$ analogously to the thermodynamic case of a phase transition, which in turn can be discussed in terms of either a first or second order one [1]. It is notorious that for $\lambda = 1$, S_N is very sensitive to subtle changes of the tuning parameter. Hence, the system's entanglement in that region is enhanced analogously to the case of caloric effects close to finite-T critical points [3]. It is

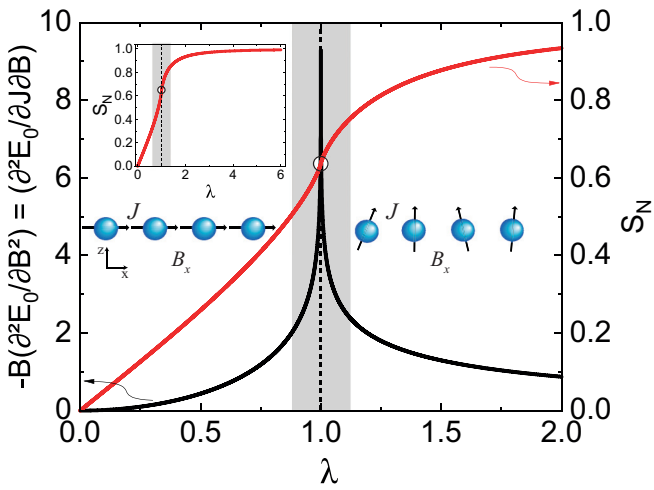

FIG. 1. Cross $(\partial {E_0}^2/\partial J\partial B)$ and second $-B(\partial^2 E_0/\partial B^2)$ derivatives of the ground-state energy E_0 (black line) and the von Neumann entropy S_N as a function of $\lambda = J/B$. The gray region indicates the λ range where both E_0 derivatives and S_N vary more expressively with λ. The spin configurations depict the ferromagnetic phase for $\lambda < 1$ and the quantum paramagnet close to $\lambda = 1$. Inset: S_N vs λ for large values of λ, i.e., for $S_N \to 1$, the so-called Greenberger-Horne-Zeilinger-like state is achieved [30]. The white bullet depicted in S_N vs λ both in the main panel and inset indicates $S_N \to \infty$ for $\lambda = 1$.

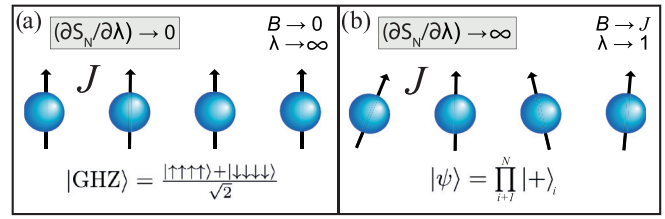

FIG. 2. Schematic representation of the here-proposed optimization of quantum information processing for the 1D IMTF. (a) For $B \to 0 \Rightarrow \lambda \to \infty$, the ferromagnetic phase state is the Greenberger-Horne-Zeilinger-like state (GHZ) [30], i.e., a superposition of $\uparrow$ and $\downarrow$ states. The von Neumann entropy S_N is insensitive to λ changes. (b) Upon applying the transverse field, the system is brought to the quantum paramagnetic phase, i.e., $\lambda \to 1$, where the system state is defined as the product of each eigenstate and $|+\rangle = 1/\sqrt{2}(|\uparrow\rangle + |\downarrow\rangle)$ [35], which corresponds to the simplest entangled state. In this regime, S_N dramatically changes upon varying λ. For a real system, one has to properly set the initial state [(a)] before applying a transverse B to achieve the here-discussed optimal conditions for quantum computing.

clear that in both cases the adiabatic character is crucial [31], considering that real applications to quantum computing take place at finite T. Note that we propose that upon considering the tuning parameter dependence of E_0, S_N can be computed straightforwardly by making the first derivative of E_0 with respect to the tuning parameter, i.e., a reduced density matrix is not needed in our case to compute global entanglement as it is done in the frame of the Meyer-Wallach approach [32]. This is corroborated by the fact that our result for S_N vs λ is in perfect agreement with the literature results employing the Meyer-Wallach approach to compute global entanglement in the thermodynamic limit (cf. Fig. 1 of Ref. [30]). The enhancement of $(\partial S_N/\partial\lambda)$ in the immediate vicinity of the QCP for the 1D IMTF was already reported [30]. However, our proposal of $\Gamma^{0\,\mathrm{K}}$ universalizes the behavior of S_N in terms of E_0 changes due to the tuning of either h or g for any quantum system, including the 2D and 3D versions of the IMTF [30]. Such an enhancement of $(\partial S_N/\partial\lambda)$ close to the QCP is associated with a violation of the Hellmann-Feynman theorem [12], which in turn reflects the emergence of low-energy spin-wave excitations for $\lambda \simeq 1$ [33]. As it is known in the literature, S_N scales logarithmically with N at the QCP and diverges for an infinite chain $(N \to \infty)$ [17,34], enhancing $(\partial S_N/\partial\lambda)$ in the vicinity of the QCP and diverging right at the QCP. Hence, we recognize that $S_N = -(\partial E_0/\partial\lambda)$ is connected to the Hellmann-Feynman theorem. The divergence of the first derivative of E_0 with respect to λ, i.e., S_N, at the QCP indicates a breakdown of the Hellmann-Feynman theorem for an infinite chain. Such a breakdown is not expected to take place for finite chains (cf. Refs. [17,34]). Figure 2 depicts our proposal of a controlled tuning of S_N for the 1D IMTF. For $B \to 0\,(\lambda \to \infty) \Rightarrow S_N \to 1$ (cf. the inset of Fig. 1), and thus $(\partial S_N/\partial\lambda) \to 0$, meaning that S_N is insensitive to B variations in this regime, corresponding to the Greenberger-Horne-Zeilinger-like (GHZ) state [30]. For $B \to J$ $(\lambda \to 1)$, a quantum paramagnetic phase is established, meaning S_N is dramatically affected by small changes of B. Hence, it is clear that in the quantum paramagnetic phase, i.e., in the regime $\lambda \simeq 1$, the system's entanglement can be easily manipulated upon varying the tuning parameter, which is B in the case of the 1D IMTF.

It turns out that entanglement is a key ingredient for quantum computing [36]. Hence, the here-proposed $\Gamma^{0\,\mathrm{K}}$ can be considered as an *entanglement compass* [37], having direct implications for Kane's quantum computer. The latter is based on the Hamiltonian for two interacting nucleus-electron ^{31}P atoms embedded in a Si matrix, namely [38],

$$H = H(B) + A_1\sigma^{1n}\cdot\sigma^{2e} + A_2\sigma^{2n}\cdot\sigma^{2e} + J'\sigma^{1e}\cdot\sigma^{2e}, \quad (8)$$

where $H(B)$ is the Zeeman interaction, A_1 and A_2 are, respectively, the hyperfine coupling constants between the first and second ^{31}P nuclear and electronic magnetic moments, and J' the coupling constant between electronic magnetic moments. Upon considering $A_1 = A_2 = A$, the energy E difference between states $|10-01\rangle$ and $|10+01\rangle$ is given by [38]

$$E = h\nu_{J'} = 2A^2\left(\frac{1}{\mu_B B - 2J'} - \frac{1}{\mu_B B}\right), \quad (9)$$

where $\nu_{J'}$ is the nuclear spin exchange frequency and μ_B the Bohr magneton. Thus, Eq. (3) can be employed straightforwardly to compute $\Gamma^{0\,\mathrm{K}}$ for Kane's quantum computer considering $h = J'$ and $g = B$. Figure 3 shows that, analogously to the case of the 1D IMTF, when $\mu_B B = 2J'$, S_N is dramatically enhanced, which in turn means that Kane's quantum computer achieves a condition in which the information processing capacity is increased. Also, upon analyzing the results depicted in Fig. 3, it is remarkable that both derivatives, namely $-J'(\partial^2 E/\partial J'^2)$ and $(\partial^2 E/\partial J'\partial B)$, are not exactly the same. This is because the J' and B dependencies of E are distinct [cf. Eq. (9)]. A sign change of both derivatives can be observed for $J' = 0.5$ due to the distinct configurations

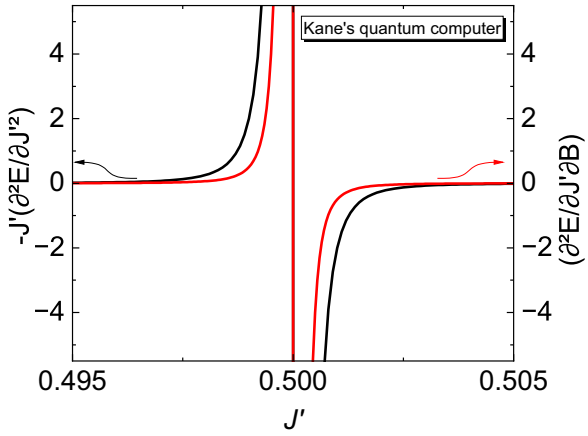

FIG. 3. Cross $(\partial E^2/\partial J'\partial B)$ and second $-J'(\partial^2 E/\partial J'^2)$ derivatives of Eq. (9) for Kane's quantum computer using $B = 1$ T. See details in the main text.

between magnetic moments. It is clear that Kane's quantum computer is a bipartite system, i.e., two qubits are considered [38]. Hence, we can extend our analysis to the so-called multipartite systems, where n qubits are considered and $\rho_n = \frac{1}{2^n} I_n$ [39], where I_n is the identity operator. The latter describes ρ for a chain of n qubits, making it possible to calculate S_N for any multipartite system where ρ_n is known. By computing S_N for a multipartite system, the derivatives incorporated in the here-proposed $\Gamma^{0\,\mathrm{K}}$ can be employed to tune the system to a range of enhanced S_N where the information processing capacity can be optimized. The process of entanglement typically involves manipulating bipartite systems aiming to maximize the entangled state. However, as the number of qubits increases, maintaining and increasing entanglement in multipartite systems becomes increasingly challenging due to the fragility of entanglement, which can be easily disrupted by environmental noise and other sources of interference. Hence, our proposal for increasing S_N upon tuning the system close to QCPs/QPTs by varying specific tuning parameters might be relevant for the field of quantum computing. Such a method could simplify the process of entanglement control, potentially leading to more efficient and reliable quantum algorithms, as well as to more robust quantum hardware.

Besides the discussions regarding $\Gamma^{0\,\mathrm{K}}$ as an entanglement compass, we now discuss the slowing down of the system dynamics and the "creation of mass" close to any QCP/QPT. Following Anderson's statement in his seminal paper that "the plasma frequency is equivalent to the mass" [40], and based on our previous work [8], we propose that symmetry breaking and fluctuations of the order parameter on the verge of any critical point/phase transition are inherently associated with the slowing down of the system's dynamics, which gives rise to low-energy excitation modes and emulates mass. This can be found, for instance, in the magnetic and electronic Griffiths phases [8,41]. In other words, as a direct consequence of the phases competition in the critical regime, the appearance of "mass" leads to a slowing down of the system's dynamic, which in turn is reflected in an enhancement of both relaxation time [8] and effective mass [42,43]. Such fascinating phenomena can be explored using $\Gamma^{0\,\mathrm{K}}$ and Γ [8,44].

In summary, we have proposed another form of the Grüneisen parameter that is valid at absolute zero temperature. The here-proposed $\Gamma^{0\,\mathrm{K}}$ can be considered as an *entanglement compass* to explore QPTs/QCPs upon varying the tuning parameters. Considering that $\Gamma^{0\,\mathrm{K}}$ quantifies the entanglement as a function of the tuning parameters, it can be employed to optimize information processing, in particular in the immediate vicinity of QCPs/QPTs. We have employed the 1D IMTF and Kane's quantum computer as workhorses to showcase our proposal. Furthermore, we have demonstrated that genuine QPTs/QCPs take place only when E_0 is nonlinear with respect to the control parameter, which is key in quantum computing regarding both bipartite and multipartite systems. Also, we propose that in the thermodynamic limit a breakdown of the Hellmann-Feynmann theorem takes place at any QCP and we anticipate that this should also occur for the Bose-Einstein condensation [44]. It is challenging to put the present concepts in practice in real systems. Yet, the "creation of mass" in the vicinity of critical points/phase transitions was discussed [45].

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