



OPEN Higgs like stiffness and fractons on the verge of phase transitions

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In condensed matter Physics, massive longitudinal Higgs modes emerge from fluctuations of the order parameter amplitude. A few years ago, the Higgs mode was *caught* experimentally in the vicinity of an insulator-to-superconductor quantum phase transition [Nat. Phys. 11, 188 (2015)]. Here, we propose, in analogy to the Higgs mode, the concept of Higgs-like stiffness (HLS), which emerges *close* to both classical and quantum phase transitions as a universal manifestation of matter in this regime. We build up a Landau free energy for the dielectric response function to demonstrate that *any* complex physical quantity can be used to infer the presence of the HLS. Our analysis is discussed in terms of experimental results of the quasi-static dielectric constant for the (TMTTF)₂SbF₆ Fabre salt. Yet, we discuss the emergence of fractons in connection with the locking of particular molecular rotational degrees of freedom and the estimation of distinct critical exponents above and below the Mott-Hubbard ferroelectric transition.

In his seminal paper about *plasmons, gauge invariance, and mass*, P.W. Anderson proposed that “... *plasma frequency is equivalent to mass*...”¹. A key question which arises is how mass can be “created” in this framework? It is well-established that a continuous symmetry breaking gives rise to two excitation modes, namely the transverse massless Nambu-Goldstone^{2,3} and the longitudinal massive Higgs modes⁴, usually identified using a potential energy density with a “Mexican hat” shape⁵. Recently, the Higgs mode was experimentally accessed in disordered thin films of NbN and InO superconductors close to a quantum phase transition by measuring the so-called excess conductivity σ_H in the THz range, being σ_H determined by subtracting the BCS contribution σ_1^{BCS} from the measured real part σ_1^{exp} , namely $\sigma_H = (\sigma_1^{\text{exp}} - \sigma_1^{\text{BCS}})$ ⁵. Here, we make use of the dielectric response as a *working horse* to demonstrate the emergence of what we propose as Higgs-like stiffness, hereafter HLS, on the verge of *any* phase transition. In analogy to the canonical massive Higgs mode, we propose that on the verge of phase transitions, fluctuations of the order parameter amplitude give rise to an increased stiffness due to the emergence of low-energy excitation modes⁶. Although P.W. Anderson has discussed the concept of generalized rigidity in a broad way⁶, we have focused on the particular case of the vicinity of phase transitions. In our analysis, using the ferroelectric transition as a case of study, the electric dipoles’ reorientation stiffness is enhanced, justifying thus the use of the term HLS. This is a similar situation to the enhancement of the spin stiffness close to a quantum phase transition, for instance, for the case of the one-dimensional Ising model under a transverse magnetic field⁷. This is because the spin stiffness $\rho_S \simeq \partial^2 E' / \partial \varphi^2$ ⁸, where E' is the energy and φ a reference angle, is linked with the quantum Grüneisen parameter proposed recently by some of us⁷. Since an enhancement of the quantum Grüneisen parameter is observed in the vicinity of quantum critical points/quantum phase transitions⁷, we can infer that the spin stiffness is also increased, which emulates the “creation of mass” in such a regime. Although the authors of Ref.⁵ report on a superconductor-to-insulator transition in which the emergence of the Higgs boson is due to the Anderson-Higgs mechanism, the latter does not work for accounting to the mass generation in the so-called type-III superconductors due to the intrinsic presence of inhomogeneous superconducting droplets⁹. However, we stress that “mass generation” is still present in such a case, which is in line with our HLS proposal. This is because the energy cost for the establishment of inhomogeneous superconducting droplets most likely leads to the enhancement of the HLS. The Higgs mode was also detected using terahertz pulse excitation in a BCS-type superconductor¹⁰. Our proposal is distinct from those cases, since it takes into account a dissipative contribution associated with the order parameter close to a phase transition of interest, a feature so far not covered in the frame of Landau theory.

To demonstrate our HLS proposal, we make use of the molecular metal (TMTTF)₂SbF₆¹¹. Such a system presents a triclinic unit cell with two donor TMTTF molecules and the SbF₆ as a counter-anion. At room temperature, this system is a metal and, due to strong electronic correlations, around 157 K the hydrogenated variant becomes both a Mott insulator and a ferroelectric, being its ground-state an antiferromagnetic phase

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achieved around 8 K¹¹. Next, we discuss our proposal in terms of the electric dipoles' reorientation stiffness enhancement in the vicinity of the second-order ferroelectric transition for the (TMTTF)₂SbF₆ salt as a case of study.

Results and discussions

The Landau and ϕ^4 theory for the second-order ferroelectric transition

To showcase our proposal, we start by recalling the phenomenon of dielectric catastrophe for frequencies $\omega \rightarrow 0$ under the light of the Bruggemann effective medium approximation (BEMA)¹². Essentially, BEMA approach considers a percolation theory background and describes the behavior of the static dielectric constant close to a phase transition, being recently employed to trace dielectric constant results under pressure close to the Mott transition for the spin-liquid candidate κ -(BEDT-TTF)₂Cu₂(CN)₃^{13,14}. Also, upon crossing the Mott transition by application of pressure, a sign-change of the dielectric constant is observed, being the dielectric response enhanced in the low-frequency regime^{14,15}. Motivated by such literature results, our goal here is to make use of the dielectric catastrophe observed at the Mott transition for the spin-liquid candidate as a *ground* to demonstrate that upon measuring the real part ε' of the complex dielectric constant $\hat{\varepsilon} = \varepsilon' - \varepsilon''i$, the HLS can be accessed, being ε'' the imaginary contribution to $\hat{\varepsilon}$. Note that the definition of $\hat{\varepsilon}$ in terms of the difference between ε' and ε'' is because we are considering a dissipative medium¹⁶. We present ε' experimental results for the Fabre salt (TMTTF)₂SbF₆, where TMTTF stands for tetramethyltetrafulvalene. The latter presents a metal-to-insulator transition coinciding with the establishment of a Mott-Hubbard ferroelectric phase^{11,17}, which we consider as an appropriate playground to explore the manifestation of the HLS as a *case of study*. On the verge of the metal to the Mott-Hubbard ferroelectric phase transition, metallic puddles coexist with an insulating matrix or vice-versa¹⁸. Let us write the free energy F expansion with pressure p as the tuning parameter considering the electric polarization P as the order parameter in the frame of the well-known Landau theory for phase transitions, namely for $T = T_c$ ¹⁹:

$$F = a_0(p - p_c)P^2 + bP^4 + \dots, \quad (1)$$

where a_0 and b are non-universal constants and p_c the critical pressure, in which the Mott transition takes place^{13,14}. Note that Eq. 1 is applicable for the spin-liquid candidate where the Mott transition is accessed by application of p ^{13,14}, being that for the (TMTTF)₂SbF₆ salt the Mott transition is driven by varying T . Next, we unprecedentedly modify Landau theory to account for a dissipative component related to the order parameter amplitude. For a linear dielectric medium $P = (\hat{\varepsilon} - \varepsilon_0)E$, where ε_0 is the vacuum permittivity and E the electric field¹⁹, being the vector notation omitted for simplicity. In terms of the real and imaginary parts of $\hat{\varepsilon}$ and considering only quadratic terms for a fixed E , Eq. 1 becomes:

$$F \simeq a_0(p - p_c)(\varepsilon'^2 + \varepsilon''^2)E^2 + b(\varepsilon'^2 + \varepsilon''^2)^2E^4. \quad (2)$$

Note that for $\varepsilon'' \rightarrow 0$ the Landau theory is nicely recovered, so that it becomes evident that our approach considers a finite $\varepsilon'' \ll \varepsilon'$. We define an excess dielectric constant $\varepsilon_H^2 = (\varepsilon'^2 + \varepsilon''^2)$ in analogy with the excess conductivity σ_H ⁵, which is pivotal to understand the manifestation of the HLS. While σ_H accounts for the deviation of the experimentally observed optical conductivity from the theory, ε_H incorporates a dissipative component, being thus reminiscent of the so-called excess function, usually employed in Thermodynamics²⁰. Note that ε_H^2 emerges naturally from the consideration of only quadratic terms upon computing F . It is remarkable that Eq. 2 has a similar mathematical structure of the two component potential energy density U embedded in the Lagrangian employed in classical field theory to describe continuous symmetry breaking, the so-called ϕ^4 theory²¹:

$$U(\phi_1, \phi_2) = -\frac{\mu^2}{2}(\phi_1^2 + \phi_2^2) + \frac{\lambda}{4!}(\phi_1^2 + \phi_2^2)^2, \quad (3)$$

where ϕ_1 and ϕ_2 are the so-called scalar fields, being ϕ_1 associated with the massive excitation and ϕ_2 with the massless one, μ is the mass, and λ the interaction strength. Note that both Eqs. 2 and 3 give rise to the well-known “Mexican hat” shape, see Fig. 1 b). In our analysis, by comparing Eqs. 2 and 3, we associate ε' with ϕ_1 and ε'' with ϕ_2 . It is to be noted that for an accumulative medium one can define $\hat{\varepsilon} = (\varepsilon' + \varepsilon''i)$ ¹⁹, so that $F \simeq a_0(p - p_c)(\varepsilon'^2 - \varepsilon''^2)E^2 + b(\varepsilon'^2 - \varepsilon''^2)^2E^4$, giving rise to a hyperbola, which does not exhibit a global minimum. Hence, it is clear that only a plot of F versus ε' versus ε'' for a given E employing Eq. 2 shall deliver a “Mexican hat”, cf. Fig. 1 b). Hence, we propose at this point a connection between the dielectric catastrophe and the natural manifestation of the HLS, i.e., pseudo “creation of mass”, when a continuous symmetry breaking takes place accompanying a particular phase transition, cf. Fig. 1 c).

We discuss now experimental results aiming to illustrate our HLS proposal. It is worth mentioning that we have measured the dielectric response ε' associated with the ionic contribution, i.e., along the c^* -axis direction, of the second-order ferroelectric transition for the (TMTTF)₂SbF₆ salt^{22,23}. This is a very important aspect since the ionic contribution can be treated as weakly coupled to the charge localization/disproportionation along the stacks formed by the TMTTF molecules. Thus, we apply the ϕ^4 theory solely for the ferroelectric transition. Our proposal is general in the sense that it is based on considering a dissipative contribution related to the order parameter amplitude of a given phase transition, which is not originally accounted for in the Landau free energy. In the particular case of the ferroelectric transition, we have rewritten P in terms of $\hat{\varepsilon}$, which in turn enabled us to rewrite F in terms of both ε' and ε'' . However, our proposal can be extended to other phase transitions of interest with their corresponding order parameters. It is well-known that the hallmark of the ferroelectric transition is

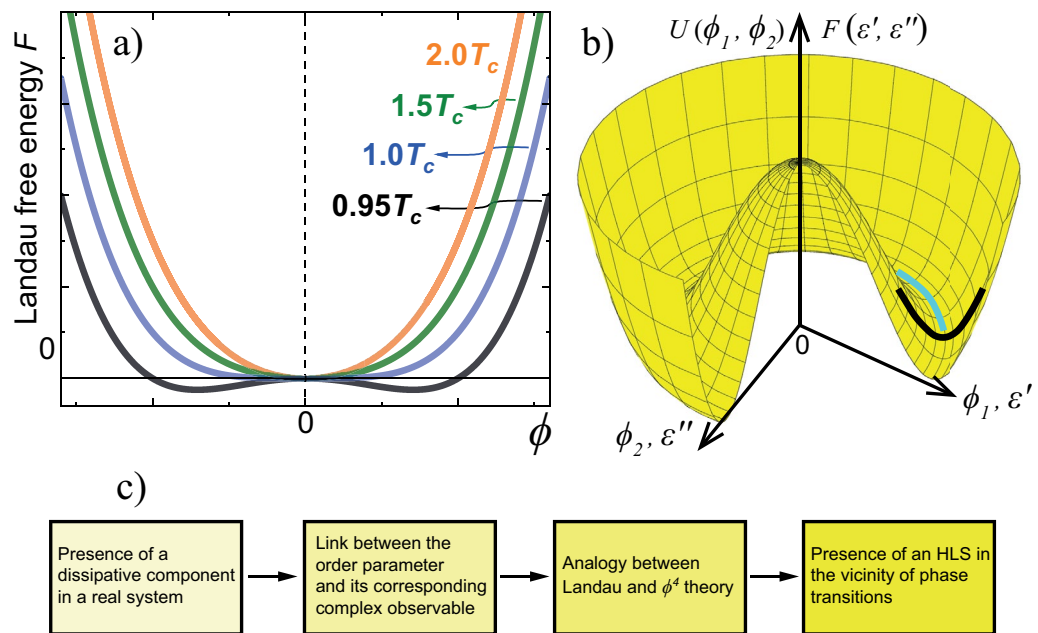


Fig. 1. (a) Landau free energy F versus order parameter ϕ for various temperatures in terms of the critical temperature T_c . (b) Potential energy density $U(\phi_1, \phi_2)$ versus ϕ_1 versus ϕ_2 , where ϕ_1 and ϕ_2 are scalar fields and the free energy $F(\epsilon', \epsilon'')$ versus ϵ' versus ϵ'' , being ϵ' the real and ϵ'' the imaginary contributions to the dielectric constant. The cyan and black thick solid lines represent, respectively, the Nambu-Goldstone and the Higgs modes. The 3D plot of $U(\phi_1, \phi_2)$ and $F(\epsilon', \epsilon'')$ is partially shown for a better visualization of the Nambu-Goldstone and the Higgs modes. (c) Flowchart indicating the manifestation of the HLS mechanism. More details in the main text.

an anomaly in ϵ' around the critical temperature, broadly discussed in textbooks¹⁹. Note that our HLS proposal is given in terms of the deviation of ϵ' from its ideal behavior, namely $(\epsilon'^2 + \epsilon''^2)$, due to the presence of a dissipative component, which is always present in real systems. In other words, the excess dielectric constant takes into account both components of the complex dielectric constant and their contribution to the free energy. Our approach is corroborated, for instance, by systematic ϵ' and loss tangent measurements versus temperature for the system $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ close to a tricritical composition point^{24,25}. Regarding the experimental results depicted in Fig. 2 c), given the high metallicity degree inherent to the Fabre salts¹¹ and intricate competition between the two order parameters close to T_{co} , ϵ'' surpasses ϵ' at 1 kHz, being highly desirable to explore the frequency-dependence of both ϵ' and ϵ'' . For the superconducting-to-insulator transition reported in Ref.⁵, the HLS in the frame of our proposal is given by $(\sigma_2^2 + \sigma_1^2)$, being σ_2 the contribution from the imaginary part to the complex optical conductivity $\hat{\sigma} = (\sigma_1 + i\sigma_2)$. This is because, for the particular case of $\hat{\sigma}$, σ_1 is the dissipative component while σ_2 is the accumulative one, being $\sigma_1 \propto \epsilon''$ and $\sigma_2 \propto \epsilon'^{26}$. It is fascinating that the presence of a dissipative component in a real system enables us to describe it in terms of Landau theory analogously to the ϕ^4 theory, which in turn makes it possible to infer the presence of an HLS in the vicinity of a given phase transition, cf. Eqs. 2 and 3 and Fig. 1 c).

Connection between the HLS and the dielectric response

In this Section, we discuss the critical exponents extracted from the fit of the dielectric constant data, cf. Fig. 2 c), the key role played by the counter-anion for the emergence of the ferroelectric phase, the HLS proposal in terms of a complex quantity related to the order parameter, and how such a proposal can be extended to other physical systems. Figure 2 c) depicts experimental results of the ionic contribution to ϵ' normalized by ϵ_0 versus T for the hydrogenated and for the partially deuterated (97.5%) variants of the molecular metal $(\text{TMTTF})_2\text{SbF}_6$. Such data set depicted in Fig. 2 c) aims to provide insights about the dielectric response ϵ' for a real system. In a previous work, we have reported on the ionic contribution of ϵ' to the ferroelectric phase for the hydrogenated variant of $(\text{TMTTF})_2\text{SbF}_6$, among other Fabre salts¹⁷. Clear peak-like anomalies are observed at the charge-ordering transition temperature $T_{co} \simeq 157$ and 162 K, respectively, for hydrogenated and partially deuterated variants²², being a similar shift in T_{co} due to deuteration effects also observed for the case of the $(\text{TMTTF})_2\text{PF}_6$ system¹⁷. Based on our analysis in the frame of the ϕ^4 theory, we propose that the peak-like anomaly in ϵ' at T_{co} is related to the emergence of an enhanced electric dipoles' reorientation stiffness into the system, i.e., “creation of mass”. Although the $(\text{TMTTF})_2\text{SbF}_6$ salt is the one that presents a behavior closer to a mean field-type (MF) transition, it deviates from MF most likely due to the presence of impurities, dislocations, and structural defects in the specimen²². Aiming to explore this feature, we have employed a fit of the ϵ' versus T data set using the protocol discussed in the “Methods” Section. We have obtained the critical exponents b_0 for both investigated salts above and below T_{co} . For the hydrogenated salt, we have obtained $b_0 = 0.996$ ($T > T_{co}$) and

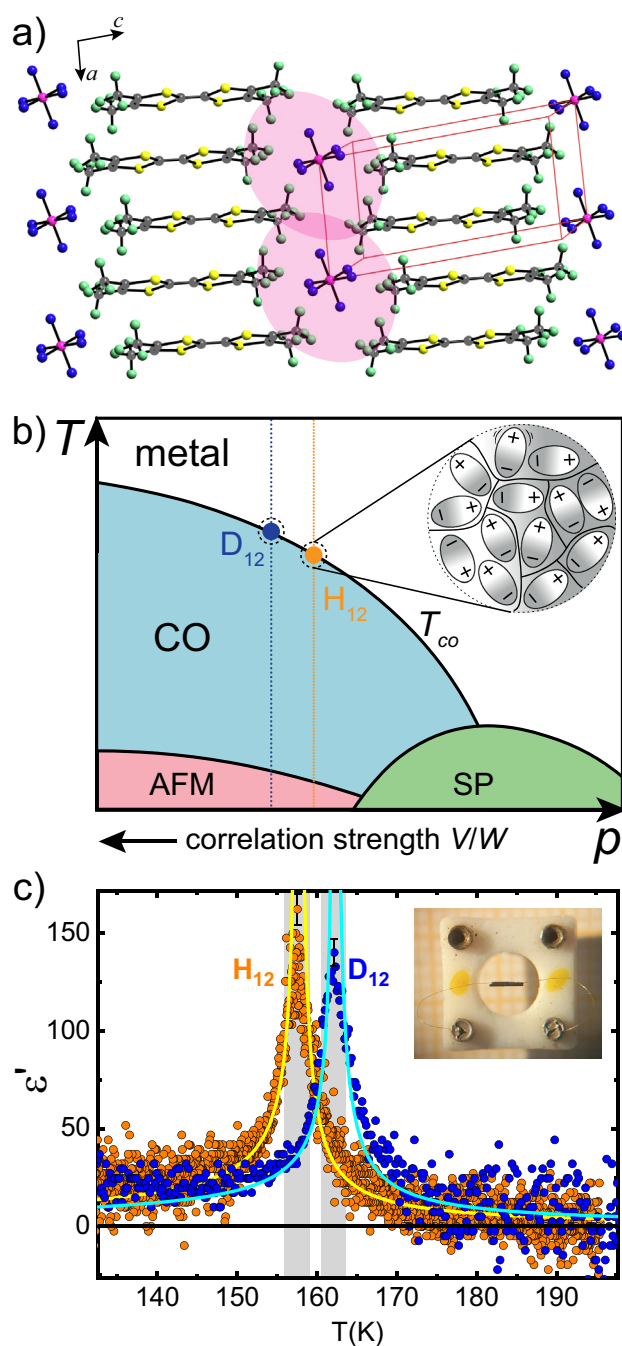


Fig. 2. (a) Crystal structure of (TMTTF)₂SbF₆, where the spheres represent hydrogen (green), carbon (gray), sulfur (yellow), fluorine (blue), and antimony (purple) atoms. The triclinic unit cell is outlined by the red solid lines. The cavities delimited by the methyl-end groups are outlined in pink color. (b) Schematic temperature T versus pressure p phase diagram of the (TMTTF)₂SbF₆ Fabre salt showing the metallic, charge-ordered (CO), antiferromagnetic (AFM), and spin-Peierls (SP) phases. The vertical dashed lines represent the corresponding charge-ordering transition temperatures T_{co} for both hydrogenated (H₁₂) and partially deuterated (D₁₂) variants. The correlation strength $V/W \propto p^{-1}$ is also depicted, where V is the inter-site Coulomb repulsion and W the bandwidth. The electric dipoles configuration is schematically depicted aiming to indicate fluctuations of the order parameter amplitude close to T_{co} . Figure based on Ref.¹⁴. (c) Quasi-static ($f = 1$ kHz) ionic dielectric constant ϵ' versus T along the c^* -axis direction for the hydrogenated (H₁₂, orange color circles) and deuterated (D₁₂, blue circles) variants of (TMTTF)₂SbF₆ from raw data set, being measurements taken every ≈ 14 s. The yellow and cyan solid lines are fittings for the data set, cf. protocol described in the “Methods” Section. The gray shaded region, except for $T = T_{co}$ and its immediate vicinity, indicates the onset where the dielectric constant is significantly enhanced and the manifestation of HLS is more expressive. In the inset, a picture of a (TMTTF)₂SbF₆ sample with gold wires attached on its surface using carbon paste is shown. The gold wires are fixed in an insulating socket with silver paint. Ge varnish is employed to fix the gold wires on the socket to avoid stress on the crystal. Details in the main text.

0.677 ($T < T_{co}$), while for the deuterated variant $b_0 = 0.918$ ($T > T_{co}$) and 0.694 ($T < T_{co}$). It is remarkable that the critical exponent b_0 is not the same above and below T_{co} . Although the Mott-Hubbard ferroelectric and the metal-to-Mott insulator transitions are considered weakly coupled, such an anomalous critical behavior and the deviation from the mean-field prediction are most likely related not only to possible local variations of T_{co} , cf. observed for other organic conductors at the Mott transition²⁷, and the presence of impurities and dislocations in the system, but also to the intricate interplay between the various degrees of freedom involved in such transitions, e.g., freezing of the counter-anion rotational degrees of freedom²⁸, lattice effects at the charge-ordering transition, and the presence of on- and inter-site Coulomb repulsion¹¹. Interestingly, the critical behavior of ε' is washed out upon irradiating the specimen, cf. discussions in Ref.¹⁷. Thus, it is natural to infer that the HLS can be suppressed by X-ray irradiation since it gradually annihilates the dielectric response¹⁷. Yet, the Curie law does not account for negative ε' , which is observed experimentally in the vicinity of T_{co} , cf. Fig. 2 c).

It is to be noted that the counter-anion plays a key role in the stabilization of the ferroelectric transition²⁸. The order-disorder character is intrinsic to the coexistence region and thus the corresponding fluctuations of the order parameter amplitude in the vicinity of T_{co} . Following Ref.²⁹, each counter-anion lies in a double-well potential and the minima are linked with the formation of hydrogen bonds between the counter-anion and the methyl-end groups. Deuteration acts like a negative pressure³⁰ making the cavity delimited by the methyl-end groups bigger³¹, increasing thus the V/t ratio, where V is the inter-site Coulomb repulsion and t the transfer-integral between nearest neighbor sites²². The potential height is increased upon deuteration and thus T_{co} is enhanced for the deuterated variant^{17,31} [Fig. 2 b)], making the system to reduce its dynamics naturally and thus its dielectric response is expected to be less pronounced when compared with the hydrogenated variant for a given frequency. A similar behavior was also observed by Nad and collaborators regarding the electronic dielectric response contribution at the Mott-Hubbard ferroelectric phase transition²², which is about three orders of magnitude higher than the ionic one¹⁷. Also, Nad demonstrated experimentally that for the TMTTF salts the dielectric response is dramatically enhanced upon reducing the frequency of the applied electric field²², a feature also observed for other molecular salts, such as the quasi-two-dimensional system κ -(BEDT-TTF)₂Cu[N(CN)₂]Cl³². Note that for $T > T_{co}$, negative values of ε' are attained due to the metallic character of the system in this temperature range²⁶. We assign the enhanced data dispersion in this T -range to the intrinsic fluctuations of the order parameter amplitude in this regime. It is clear that the maximum in ε' is associated with the canonical Landau free energy variation $\Delta F = -EP + g_0 + 1/2g_2P^2 + \dots$ close to T_c , being g_0 and g_2 the Landau coefficients, cf. Ref.⁶ and references cited therein. Since ε' is related to P , it becomes also clear that ΔF scales with ε' , being maximized at T_{co} and vanishing above and below T_{co} . In other words, fluctuations of the order parameter amplitude are mainly governed by ε' , cf. black thick line depicted in Fig. 1 b), while we associate the massless mode with ε'' indicated by the cyan line in Fig. 1 b). If the dissipative component dominates the system, it may suppress the ferroelectric transition in a particular frequency range²⁶. Even for the case of ε'' being higher than ε' , a finite peak-like anomaly in ε' around T_{co} might still be observed, so that, although out of the scope of the present work, the HLS can be also investigated^{25,33}. In this context, it is tempting to classify the Fabre salts as ferroelectric metals³³ in this regime. It is clear that for any complex physical quantity, the imaginary contribution is always finite. Upon changing the dimensionality of the system, e.g., considering a thin film of a molecular conductor, we anticipate that the HLS will become more pronounced due to the less accessible degrees of freedom, which in turn enhances the electric dipoles' reorientation stiffness. Yet, since the dielectric constant and the refractive index are related quantities, the ε' peak-like anomaly at T_{co} indicates the onset of critical opalescence across the Mott transition³⁴.

Now, we make an analysis considering the well-known complex order parameter of the form $\Psi = Ae^{i\theta}$, being the amplitude $A = (\phi_1^2 + \phi_2^2)^{1/2}$ and the phase $\theta = \arctan(\phi_2/\phi_1)$ ³⁵. Just to mention, the latter is reminiscent of the Weinberg angle θ_W in the frame of the electroweak theory³⁶. Based on our proposal of HLS, we can analogously write $\Psi = (\varepsilon'^2 + \varepsilon''^2)^{1/2} e^{i[\arctan(\varepsilon''/\varepsilon')]}$, so that it becomes evident that a finite ε'' affects both the amplitude and phase of the complex order parameter. It is clear that for a given pair of ε' and ε'' the phase remains unchanged, which is in line with the $U(1)$ symmetry. At this point, it is appropriated to recall that for the case of superconductors and superfluids, the phase ϕ bending energy $U_b \propto (\nabla\phi)^2$ ³⁶. In our case, the HLS can be thus quantified in terms of $[\nabla\arctan(\varepsilon''/\varepsilon')]^2$. Note, however, that the behavior of the imaginary contribution to the response function is system-dependent and not all response functions give rise to a finite imaginary part³⁷. The latter is finite only if it is coupled to the microscopic quantity associated with the corresponding ordering³⁷. Following discussions by P. Coleman³⁶, gauge field + phase $\rightarrow$ massive gauge field. A natural question that arises is: what is in fact the HLS? In simple words, the HLS can be understood as the energy needed to be “paid” for the establishment of the ordered phase. Analogous to the photon “swallowing” a Goldstone boson and acquiring mass²¹, ε' “swallows” ε'' , giving rise to the ferroelectric phase. In other words, in our proposal of HLS, the local electric field that emerges due to the inherent interactions between electric dipoles in the vicinity of the phase transition plays the role of the gauge field and the phase is $\theta = \arctan(\varepsilon''/\varepsilon')$, giving rise to the “creation of mass” in terms of HLS. Aiming to illustrate our HLS proposal, following discussions in Ref.³⁷, we consider the simple case of a particle with mass m under a potential $U(x) = -ax^2 + cx^4$, where a and c are arbitrary constants. For small oscillations around the equilibrium position, the frequency $\omega^2 = 2|a|/m$ ($a < 0$) and $\omega^2 = 4a/m$ ($a > 0$)³⁷. For $a \rightarrow 0$, the period of oscillation $T^* \propto |a|^{-1/2} \rightarrow \infty$, which implies a critical slowing down³⁷ that emulates our proposal of HLS as if there was a “creation of mass”. The authors of Ref.⁵ probed the Higgs mode in an insulator to superconductor quantum phase transition. In the latter, thin films of NbN and InO were tuned close to quantum criticality upon increasing disorder. Note that for the case of the (TMTTF)₂SbF₆ salt, the emergence of metallic puddles in an insulating matrix, or vice-versa, i.e., in the phases coexistence region, is analogous to the insertion of disorder. We propose that *any* complex physical quantity can be employed to probe the HLS on the verge of phase transitions, e.g., refractive index and

a.c. magnetic susceptibility, being the choice of the observable dependent on the particular investigated system. In other words, a similar analysis based on Eq. 2 and above discussions can be performed for *any* complex physical quantity by considering its relation with the order parameter of the particular investigated phase transition. It is clear that the form of Eq. 2 remains the same for any complex physical quantity. Our proposal can be also extended to complex thermodynamical quantities, such as complex specific heat^{38,39}.

Regarding the universality of our approach, note that the role played by the HLS is distinct depending on the particular phase transition considered, for instance, in the ferroelectric case, HLS is linked with the electric dipoles' reorientation stiffness, while for a magnetic phase transition, the spin stiffness plays the role of HLS. The corresponding excess function for a given phase transition should be defined accordingly²⁵. Our proposal is based on the fact that close to phase transitions, the HLS manifestation can be inferred. We propose that the HLS plays an analogous role as if "mass" was created. Examples include²⁵ the stiffness of rotating electric dipoles close to the ferroelectric transition, cf. discussed in the present work, the scattering of light right at the nematic to smectic-A transition⁴⁰, or the immobile character of the Cooper pairs accompanied by the emergence of electric strings in the superconductor to superinsulating transition⁴¹. The celebrated Landau theory does not consider the dissipative component linked with the observable, e.g., the imaginary part of ε , which in turn is associated with the order parameter. However, our proposal embodies a dissipative component that is always present in a real system, enabling us to infer the universal emergence of an HLS in phase transitions. At this point, it is worth recalling that the Landau theory is applicable to any phase transition if we have in hand the corresponding order parameter. Analogously, our HLS approach is universal in the sense that if we are able to link the order parameter with its corresponding complex observable, the HLS can be inferred. Even in particular cases that it is challenging to define an order parameter, e.g., the BKT transition, the inherent vortex binding below the BKT transition temperature is accompanied by an elastic rigidity⁴², which we anticipate as being the manifestation of our proposal of the HLS. Thus, although the BKT transition has embedded an HLS, the formal mathematical treatment is challenging because the order parameter is not defined. Yet, Landau theory does not account for spatial variations of the order parameter, which is covered in the Ginzburg-Landau theory^{25,36}. However, none of such theories incorporate the dissipative component associated with the order parameter, which is the ground of our proposal.

The HLS under the perspective of the Mott transition

Aiming to support the discussions regarding the HLS in the frame of the ϕ^4 theory, we briefly discuss in the following a few general aspects related to the Higgs mechanism in superconductors and analyze the HLS using the plasma frequency and Drude weight. Interestingly, time-dependent Ginzburg-Landau theory for superconductivity delivers the relaxation time $\tau \propto (T - T_c)^{-1}$ ⁴³, so that it becomes clear that for $T \rightarrow T_c \Rightarrow \tau \rightarrow \infty$. Such a behavior is consistent with the emulation of "creation of mass" on the verge of a superconducting transition. Following Anderson, close to T_c , fluctuations of the order parameter amplitude are enhanced, implying that excitations have become concentrated at low-frequencies⁶. A universal aspect of phase transitions is that close to T_c , given the phases competition in this regime, we are dealing with a phases coexistence region, so that τ can be computed employing Avramov/Casalini's model^{18,44}. In such cases, $\tau = \tau_0 \exp(C/T\nu^{\Gamma_{eff}})$, where τ_0 is a typical relaxation time of the system, C a non-universal constant, ν the volume, and Γ_{eff} the effective Grüneisen parameter^{18,44}. Regarding the Mott transition, the dielectric response ε' as a function of pressure is positive (negative) when the system is a Mott insulator (metal)^{13,14}. Recently, some of us have demonstrated that upon crossing the first-order transition line of the Mott transition, τ is dramatically enhanced and an electronic Griffiths-like phase sets in¹⁸, so that "mass creation" can also be inferred. It is well-known from textbooks that the electron mass m_e is related to the plasma frequency ω_p by $m_e = e^2 n_e / \varepsilon_0 \omega_p^2$ ²⁶, where e is the electron fundamental charge, and n_e the electron density. It turns out that upon tuning the system from a metal to a Mott insulator, electrons tend to localize suppressing thus charge fluctuations, i.e., $\omega_p \rightarrow 0 \Rightarrow m_e \rightarrow \infty$. The latter is corroborated by recent random phase approximation (RPA) calculations of charge collective modes at the onset of the Mott transition⁴⁵ and it is in line with our proposal of the emergence of the HLS, i.e., "mass enhancement", on the verge of a phase transition, cf. proposed by Anderson¹. In terms of the Drude weight $D = \lim_{\omega \rightarrow 0} \omega \sigma_2(\omega) \simeq (\partial^2 E_0 / \partial k^2) = \hbar^2 m^{-1}(k)$, where E_0 the ground-state energy, k the wave vector, and $m(k)$ the effective mass^{46,47}. Upon going from a metal to an insulating phase, $(\partial^2 E_0 / \partial k^2) \rightarrow 0 \Rightarrow D \rightarrow 0$, $(\partial^2 E_0 / \partial k^2) \rightarrow 0 \Rightarrow m(k) \rightarrow \infty$. The latter is reminiscent of the breakdown of the Hellmann-Feynman theorem on the verge of a quantum phase transition⁷.

About fractonic excitations and the HLS

Now, we discuss the "mass enhancement", i.e., the slow-dynamics regime, in connection with fractons, which are linked with immobile excitations⁴⁸. In our approach, such fractonic excitations can be connected with the enhancement of the electric dipoles' reorientation stiffness^{46,49}, so that the appearance of the HLS might be linked with the increased immobility of the electric dipoles on the verge of the Mott transition. The increase of the dipoles' reorientation stiffness particularly in the vicinity of a quantum critical point was already reported⁴⁹, but, to the best of our knowledge, no association with the concept of Higgs mode was discussed so far. For the Fabre salts, the methyl-end groups play a crucial role in the stabilization of the Mott-Hubbard ferroelectric phase^{11,50,51}. Regarding the particular case of the (TMTTF)₂SbF₆, the charge disproportionation $2\delta = 0.5$ ²⁸, so that the ferroelectric phase for the SbF₆ salt is one of the most robust among the Fabre salts. Based on the fact that the Mott-Hubbard charge disproportionation occurs along the TMTTF stacks⁵², it is natural to infer that the counter-anion displacement from its center-symmetric position gives rise to the appearance of an ionic contribution to the dielectric response¹⁷. The latter is most likely related to the formation of a charge imbalance between the methyl-end groups and the counter-anion. This scenario is corroborated by vibrational spectra measurements showing that a distortion of the octahedral structure of the counter-anions takes place below T_{co}

⁵³. More specifically, the low structural symmetry, i.e., triclinic $P\bar{1}$ space of the Fabre salts produces an intricate counter-anion displacement within the cavities formed by the methyl-end groups, cf. Fig. 2 a). Following discussions in Ref.¹¹, there are two possibilities of rearrangement of the counter-anions, i.e., the counter-anion displacement might occur approaching either methyl-end groups or the sulfur atom of the TMTTF molecule nearby. Assuming that the counter-anion displacement occurs towards the methyl-end groups, such entities will build up an “ionic” electric dipole. In this context, both counter-anion and methyl-end groups become rigid, i.e., their degree of immobility is enhanced, emulating thus the manifestation of fractonic excitations⁴⁸. This is corroborated by the experimental verification of a divergent relaxation time at T_{co} for the $(\text{TMTTF})_2\text{AsF}_6$ salt⁵⁴. It is also to be noted that the freezing of the methyl-groups rotational degrees of freedom around T_{co} can be inferred from high-resolution thermal expansion¹¹ and ^{19}F NMR measurements^{28,55}. Regarding thermal expansion measurements for the Fabre salts, the continuous freezing of the rotational degrees of freedom of the methyl-end groups makes such vibrational modes no longer to contribute to the thermal expansion¹¹. In this regime, the conservation of the electric dipole moment restricts the rotational degrees of freedom of both counter-anion and methyl-end groups, which we interpret as the emergence of fractons⁴⁸. It is worth mentioning that the inherent competition between various degrees of freedom in molecular conductors enables us to explore the here-discussed fascinating manifestations of matter. Hence, we propose that the evidence for the emergence of fractonic excitations associated with the ferroelectric phase for the investigated molecular conductor not only lies in the fluctuations of the order parameter amplitude in the vicinity of T_{co} , but also can be corroborated by the increasing degree of immobility of the counter-anions¹¹, a divergent-like relaxation time for the particular case of $(\text{TMTTF})_2\text{AsF}_6$ ⁵⁴, and the freezing of the methyl-end groups rotational degrees of freedom^{28,55}. Since fractonic excitations are associated with a high degree of charge immobility, our proposal of HLS perfectly resonates with such excitations, cf. previously discussed.

Conclusions

In summary, we have demonstrated that close to phase transitions the HLS appears naturally as a consequence of strong fluctuations of the order parameter amplitude. We propose that the deviation of ε' from its ideal behavior due to a finite ε'' emulates the electric dipoles' reorientation stiffness in our case of study, being extendable to any phase transition with its corresponding complex observable. Experimental results for the $(\text{TMTTF})_2\text{SbF}_6$ salt suggest that the quasi-static ionic dielectric response is linked with an increased immobility of both the counter-anion and the methyl-end groups, which we associate with the emergence of fractonic excitations, being a systematic exploration of the frequency-dependence of both ε' and ε'' part of future projects. Our proposal of HLS is universal and it is challenging to explore this intricate manifestation of matter on the verge of other phase transitions, being highly desirable to determine the gauge field responsible for the emergence of HLS in each case.

Methods

The electrical contacts were prepared using tempered gold wires provided by Cryogenics with 20 μm diameter properly attached to the specimen surfaces along the c^* -axis direction employing carbon paste. The sample dimensions are $(2.3 \times 0.6 \times 0.3) \text{ mm}^3$ and $(2.5 \times 0.4 \times 0.1) \text{ mm}^3$, respectively, for the specimens of hydrogenated and partially deuterated $(\text{TMTTF})_2\text{SbF}_6$. The measurements were carried out employing a cooling rate of -10 K/h (hydrogenated) and -40 K/h (deuterated), and an electric field of 500 mV/cm at a fixed frequency of 1 kHz . A high-resolution Andeen-Hagerling capacitance bridge and a Teslatron PT cryostat supplied by Oxford Instruments were used. For further experimental details, please refer to Ref. ¹⁷. The fittings for the ε' versus T data set shown in Fig. 2 c) for both hydrogenated and deuterated salts were obtained by employing the following protocol: i) T_{co} was estimated as the highest value of ε' and we have considered 8% of the temperature range around it; ii) we have plotted $-\log \varepsilon'$ versus $\log |T - T_{co}|$; iii) a linear fit was then performed to obtain a rough estimate of the critical exponents, both above and below T_{co} ; iv) based on the obtained critical exponents, we have fitted the data above and below T_{co} using, respectively, $\varepsilon' = 1/(a|T - T_{co}|^{b_0})$ and $\varepsilon' = 1/(2a|T - T_{co}|^{b_0})$ ¹⁹, being the parameter a a non-universal constant and b_0 the critical exponent. The figures were generated employing the softwares Wolfram Mathematica version 13.0 (License ID L8671-6484), Origin version 2024b (Registration ID SWX-9OF-FFY), and Adobe Illustrator 2024 version 28.5.

Data availability

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

Received: 29 July 2024; Accepted: 22 August 2025

Published online: 30 August 2025

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Acknowledgements

MdeS acknowledges financial support from the São Paulo Research Foundation – Fapesp (Grants number 2011/22050-4, 2017/07845-7, and 2019/24696-0), National Council of Technological and Scientific Development – CNPq (Grants number 303772/2023-9), J.P. Pouget and Pascale Foury-Leylekan for discussions, and Alec Moradpour (in memoriam) for synthesizing the investigated single-crystals. ACS acknowledges CNPq (Grants number 308695/2021-6). LS acknowledges IGCE for the post-doc fellowship. We thank the anonymous Reviewers for fruitful suggestions, which improved the quality of the manuscript.

Author contributions

L.S. and M.deS. carried out the experiments and calculations and L.S. generated the figures. L.S. and M.deS. wrote the paper with contributions from G.O.V., S.M.S., A.C.S. and R.E.L.M. All authors revised the manuscript. M.deS. conceived and supervised the project.

Declarations

Competing interests

The authors declare no competing interests.

Additional information

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