


Long-Lived Higgs Modes in Strongly Correlated Condensates

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We investigate order parameter fluctuations in the Hubbard model within a time-dependent Gutzwiller approach. While in the weak coupling limit we find that the amplitude fluctuations are short-lived due to a degeneracy with the energy of the edge of the quasiparticle continua (and in agreement with Hartree-Fock + RPA theory), these are shifted below the edge upon increasing the interaction. Our calculations therefore predict undamped amplitude (Higgs) oscillations of the order parameter in strongly coupled superconductors, cold atomic fermion condensates, and strongly interacting charge- and spin-density wave systems. We propose an experimental realization for the detection of the spin-type Higgs mode in undoped cuprates and related materials where, due to the Dzyaloshinsky-Moriya interaction, it can couple to an out-of-plane ferromagnetic excitation that is visible via the Faraday effect.

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The appearance of amplitude and phase modes in BCS superconductors is a consequence of the breaking of $U(1)$ symmetry (for a review, see Ref. [1]). The invariance of the ground state energy with respect to the phase of the complex order parameter Δ leads to the appearance of a Goldstone mode that is pushed up to the plasma frequency by the long-range Coulomb interaction [2]. This work has inspired the development of a similar theory in the Lorentz invariant case, which could account for the mass of gauge bosons in the standard model [3–5]. Recently, due to this analogy, the amplitude modes in a superconductor (SC) are also named “Higgs modes”; see, e.g., Refs. [1,6–19].

In a SC, the energy of the Higgs mode is determined from the pole in the pair correlation function $\chi_{\Delta\Delta}(\omega)$ [20]. For an s -wave SC and within BCS theory (density of states $\rho(\epsilon)$, coupling λ) this correlation function reads

$$\chi_{\Delta\Delta}(\omega) = \frac{\chi_{\Delta\Delta}^{(0)}(\omega)}{1 - \lambda\chi_{\Delta\Delta}^{(0)}(\omega)} \quad (1)$$

with the bare BCS correlations

$$\chi_{\Delta\Delta}^{(0)}(\omega) = 2 \int d\epsilon \rho(\epsilon) \frac{\epsilon^2}{\sqrt{\Delta^2 + \epsilon^2}} \frac{1}{\omega^2 - 4(\Delta^2 + \epsilon^2)}, \quad (2)$$

and Δ denotes the SC order parameter. Thus, the denominator of Eq. (1) vanishes exactly at $\omega = 2\Delta$ where it reduces to the BCS self-consistency condition

$$1 = -\frac{\lambda}{2} \int d\epsilon \frac{\rho(\epsilon)}{\sqrt{\Delta^2 + \epsilon^2}}. \quad (3)$$

As a result, the energy of the Higgs mode is identical to the spectral gap and the associated damping, due to the decay

into quasiparticle excitations, together with its property as a scalar quantity has hampered the experimental detection of the Higgs mode via conventional spectroscopy. In principle, the application of a supercurrent can make the Higgs mode infrared active [21]; however, the associated breaking of inversion symmetry also allows for optical transitions across the SC gap, even in the clean case [22,23]. Therefore, current research related to the detection of the Higgs mode mainly focuses on nonequilibrium (and) or nonlinear response techniques [7,8,17] as third-harmonic generation in time-resolved terahertz spectroscopy. However, also in this case it is difficult to disentangle Higgs and single-particle excitations across the gap [24] since both occur at the same energy and often yield comparable contributions to the response [25–27]. A notable exception is the case of NbSe_2 where due to the coexistence of s -wave SC with a charge-density wave (CDW) the Higgs mode can be pushed below the spectral gap, which allows to distinguish both excitations in Raman scattering [28–32].

It should be stressed that (in the absence of coexisting orders) the equality of Higgs mode energy and spectral gap is peculiar to s -wave BCS SCs. For other pairing symmetries this is no longer valid and, e.g., for a d -wave SC the signature of the Higgs mode in fact can appear slightly below the maximum spectral gap. However, it turns out that in this case the mode does *not* correspond to a pole in the pairing correlation function, which together with the finite density of quasiparticle excitations inside the gap leads to comparable difficulties in the experimental observability as in the s -wave case.

Here, we show that even for a pure s -wave condensate the situation changes dramatically, when a mean-field BCS picture does not apply and correlations become important. For SCs in the strongly coupled regime, we predict a shift

of the Higgs mode energy below the spectral gap leading to a long-lived collective mode. This provides a sharp tool to determine if a SC is in the strong-coupling regime, which is certainly of relevance for ultracold atom experiments where the strong-coupling regime is easily achieved [33]. Furthermore, we also analyze the appearance of amplitude (“Higgs”) modes in itinerant antiferromagnets where analogous mechanisms are at work due to the breaking of spin-rotational symmetry and the strong-coupling regime applies to a variety of condensed matter systems. We propose an experimental setup that should allow identifying these excitations in undoped (antiferromagnetic) cuprates and related materials.

As our considerations are both valid for a magnetically ordered state or a SC, we start our discussion with the *repulsive* single-band Hubbard Hamiltonian relevant for the former,

$$H = -t \sum_{(ij),\sigma} c_{i,\sigma}^\dagger c_{j,\sigma} + |U| \sum_i \left(n_{i,\uparrow} - \frac{1}{2} \right) \left(n_{i,\downarrow} - \frac{1}{2} \right), \quad (4)$$

where $\langle \dots \rangle$ restricts the sum to the nearest neighbor sites, $-t$ is the hopping amplitude, and $|U|$ parametrizes the strength of the on-site repulsive interaction. Here, $n_i = \sum_\sigma n_{i,\sigma}$ with $n_{i,\sigma} = c_{i,\sigma}^\dagger c_{i,\sigma}$.

We focus on the interaction dependence of amplitude modes in the particle-hole symmetric limit of the bipartite model (half-filling) where SC charge-ordered and spin-ordered ground states are related by canonical transformations of the Hamiltonian [34,35] (see Appendix).

It is convenient to define the staggered spin operators,

$$\hat{S}_i = \frac{1}{2} e^{i\mathbf{Q}\cdot\mathbf{R}_i} (c_{i,\uparrow}^\dagger, c_{i,\downarrow}^\dagger)^T \boldsymbol{\sigma} \begin{pmatrix} c_{i,\uparrow} \\ c_{i,\downarrow} \end{pmatrix}. \quad (5)$$

Here, $\boldsymbol{\sigma}$ is the vector of Pauli matrices and $\mathbf{Q} = (\pi, \pi)$ for a square lattice (lattice constant $a \equiv 1$).

In the repulsive case, broken symmetry appears as a spin-density wave (SDW) with uniform staggered magnetization \mathbf{m} , which can be oriented in any direction, i.e., $\langle \hat{S}_i \rangle = \mathbf{m}/2$. The mean-field Hamiltonian takes the form

$$H^{\text{MF}} = -t \sum_{(ij),\sigma\sigma'} q_{ij}^{\sigma\sigma'} c_{i,\sigma}^\dagger c_{j,\sigma'} + \sum_i \boldsymbol{\lambda} \cdot \hat{S}_i, \quad (6)$$

with $q_{ij}^{\sigma\sigma'} = \delta_{i,j} \delta_{\sigma,\sigma'}$ and $\boldsymbol{\lambda} = -|U|\mathbf{m}$. The invariance of the ground state with respect to the orientation of \mathbf{m} generates the Goldstone spin-wave modes, whereas fluctuations of $|\mathbf{m}|$ correspond to the spin amplitude mode.

From Eq. (6) it follows that within standard Hartree-Fock theory, the amplitude of the order parameter $\Delta = |\boldsymbol{\lambda}| = |U|\mathbf{m}|$ also determines the spectral gap 2Δ for single-particle excitations. As a consequence, the Higgs excitations have a minimum energy of 2Δ that can be immediately derived

from the corresponding RPA equation [Eq. (1)], cf. Ref. [36].

In order to study the relation between Higgs excitation and spectral gap beyond a mean-field + RPA scheme, the model Eq. (4) is solved within the time-dependent Gutzwiller approximation (TDGA) [37–45] based on a wave function,

$$|\Psi_G\rangle(t) = \hat{P}_G |HF\rangle, \quad (7)$$

where the time-dependent Gutzwiller projector \hat{P}_G optimizes the number of doubly occupied states in the underlying Hartree-Fock state $|HF\rangle$ (see Ref. [46] for a finite temperature extension). The solution can be obtained from a time-dependent variational principle that allows to compute the time-dependent density matrix $\rho(t)$ and variational double-occupancy parameters $D_i(t) = \langle \Psi_G | n_{i,\uparrow} n_{i,\downarrow} | \Psi_G \rangle(t)$. We have previously shown that the dynamic correlation functions for the two-dimensional Hubbard model, obtained within the TDGA, are in very good agreement with corresponding results from exact diagonalization on small lattices, Monte Carlo, and dynamical mean-field methods [37–41,45].

In case of the TDGA, the underlying quasiparticle Hamiltonian also takes the form of Eq. (6) but now the hopping renormalization $q_{ij}^{\sigma\sigma'}(n, \mathbf{m}, D)$ is a function of charge density n , magnetization \mathbf{m} , and double occupancy D [39]. More importantly, the parameter λ in the TDGA originates from a constraint that links the double-occupancy parameter to the fermion operators and in general is not directly related to the order parameter for the SDW. As a consequence, the associated dynamics is generally different, which leads to a decoupling of amplitude fluctuations and spectral gap in the TDGA.

Thanks to the repulsive-attractive transformation [34,35] (see Appendix) the above considerations apply also to the attractive Hubbard model. Indeed, applying Eqs. (A1) and (A2) one finds that an antiferromagnetic ground state with magnetization in the xy plane transforms into a SC state, whereas a state with staggered magnetization along the z direction transforms into a CDW. In the case of the SC the quasiparticle wave function in Eq. (7) becomes a BCS state, i.e., $|HF\rangle = |BCS\rangle$ [47].

Figure 1 shows the spectrum of Higgs excitations for the half-filled two-dimensional square lattice (bandwidth parameter $B = 4t$). Here, the momentum of the (imaginary part of the) order parameter correlation function $\chi_{\Delta\Delta}(q, \omega)$ corresponds to $\mathbf{q} = (\pi, \pi)$ in case of SDW and CDW, whereas $\mathbf{q} = (0, 0)$ for the SC. Clearly, in HF + RPA (red dashed) the enhancement of $\chi_{\Delta\Delta}(q, \omega = 2\Delta)$ indicates the presence of amplitude excitations at $\omega = 2\Delta$, the value of which is indicated by the vertical dashed line. On the other hand, the low energy peak in the corresponding TDGA correlations starts to shift below the spectral gap of the quasiparticle continuum (vertical solid line) when the interaction becomes larger than the bandwidth $|U|/B \gtrsim 1$.

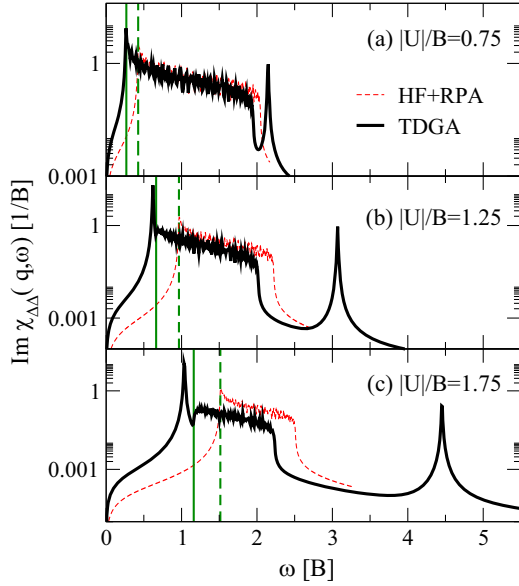


FIG. 1. Imaginary part of the amplitude correlation function $\chi_{\Delta\Delta}(q, \omega)$ for the half-filled 2D Hubbard model in case of a SC (attractive model, $\mathbf{q} = 0$), a CDW (attractive model, $\mathbf{q} = \mathbf{Q}$), or a SDW (repulsive model, $\mathbf{q} = \mathbf{Q}$) ground state. Panels (a)–(c) refer to different interactions $|U|/B = 0.75, 1.25, 1.75$, and black (red) lines correspond to the TDGA (HF + RPA) result. The green vertical solid (dashed) line indicates the spectral gap of the particle-hole continuum (particle-particle continuum in the SC case) in case of TDGA (HF + RPA).

In addition, one observes the formation of a high-energy double-occupancy (“doublon”) excitation above the continuum of single-particle excitations.

We have checked that both HF + RPA and TDGA obey the first-moment sum rule, [48,49]

$$\frac{1}{\pi} \int_0^\infty d\omega \omega \text{Im} \chi_{\Delta\Delta}(q, \omega) = 2\langle T \rangle,$$

where $\langle T \rangle$ denotes the kinetic energy in the corresponding ground state. Clearly, the Higgs mode and the high-energy doublon excitation have opposite roles in the sum rule and tend to cancel in the average kinetic energy.

Figure 2 shows the evolution of the spectral gap and the Higgs mode energy vs the interaction $|U|/B$. For $U/B \gtrsim 1$ both energy scales start to separate up to $U/B \approx 2$, corresponding to the total bandwidth where the relative shift (inset) acquires a maximum. The subsequent decrease of the relative shift for larger values of U/B is not only due to the increase of 2Δ but also due to a slight decrease of $2\Delta - \Omega_{\text{Higgs}}$ beyond $|U|/B = 2$.

As mentioned above, the splitting of Ω_{Higgs} from the continuum above a critical U_c is due to the fact that the spectral gap and the order parameter become decoupled within the TDGA. It is not influenced by an anomalous power in the real part of the (bare) amplitude correlation

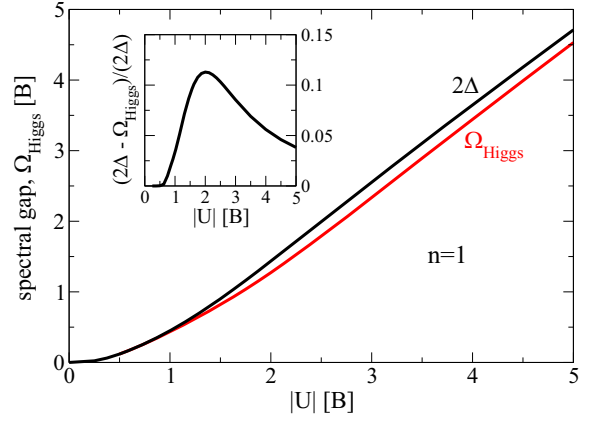


FIG. 2. Main panel: TDGA energies of the spectral gap 2Δ (black) and the Higgs mode (red) as a function of $|U|/t$ for the half-filled 2D Hubbard model. Inset: relative shift of the Higgs mode inside the spectral gap.

function, which still displays the BCS-like $1/\sqrt{(2\Delta)^2 - \omega^2}$ behavior.

We checked that the above results are not exclusive of the two-dimensional square lattice but are quite robust and also present, for example, in the Bethe lattice with coordination number $z \rightarrow \infty$ for which the exact evaluation of expectation values within the Gutzwiller wave function coincides with the Gutzwiller approximation [50]. In the following, we present results for this model, which has a particularly simple semicircular density of states, $\rho(\omega) = (1/\pi)\sqrt{B^2 - \omega^2}$.

Let us study first the density dependence of the Higgs mode for the attractive model, where away from half-filling the SC instability dominates over the CDW. Instead of evaluating the amplitude correlation function, we explicitly study the time evolution under the influence of an infinitesimal interaction quench $U \rightarrow U \pm \delta U$ ($\delta U/U = 0.005$). The energy of the Higgs mode is then obtained from a Fourier transformation of the resulting dynamics of the anomalous correlation function $J(t) = \langle d_{i,\downarrow} d_{i,\uparrow} \rangle$ of the attractive Hubbard model [Eq. (A3)]. Here, without loss of generality, we assume a real order parameter.

Transformations [Eqs. (A1) and (A2)] map the HF + RPA approximation of the repulsive Hubbard model into a bare ladder approximation in the attractive case, which becomes exact at low densities [42,51] as it coincides with Galitskii’s approach [52]. Our TDGA also converges to the correct low-density limit, where one finds that the energy of the Higgs mode in the attractive model coincides with the spectral gap in the particle-particle continuum (see Fig. 3 for $|U|/B = 2$). However, as the density increases, the Higgs mode shifts inside the spectral gap. For the present parameters, in the low-density limit, the fermions form bound states, so the ground state can be seen as a Bose condensate characterized by a chemical potential below the lower band edge. The vertical dotted line in Fig. 3 marks the

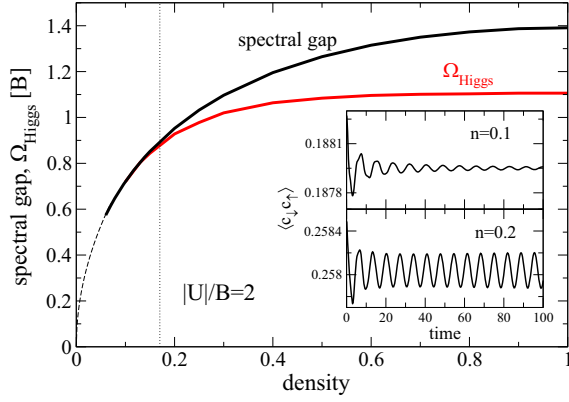


FIG. 3. Main panel: TDGA energies of the spectral gap 2Δ (black) and the Higgs mode (red) as a function of density for the attractive Hubbard model ($|U|/B = 2$) with a semielliptic density of states (bandwidth B). To the left of the vertical dotted line, the chemical potential falls below the lower band edge. Inset: time dependence of the anomalous correlations for densities $n = 0.1$ and $n = 0.2$, respectively.

density below, which the chemical potential moves below the lower band edge and defines the Bose condensation regime. We see that the divergence of the Higgs mode from the edge of the quasiparticle continuum sets in practically at the same density.

In the low-density regime, where Ω_{Higgs} coincides with the spectral gap, the decay into single-particle excitations induces the same damping $\sim 1/\sqrt{t}$ in the time evolution of the anomalous correlation as in the conventional BCS case, cf. inset $n = 0.1$. As soon as the Higgs mode is split off from the continuum the dynamics does not show any damping (cf. inset $n = 0.2$); however with increasing density a stronger admixture with double-occupancy fluctuations leads to interesting coupling phenomena [47].

SCs by definition are charged and one has to take into account the long-range Coulomb interaction that pushes the Goldstone mode to high energy [2], thus eliminating a decay channel for the amplitude excitation. On the other hand, the prerequisite of strong coupling hampers the observability of a subgap Higgs mode in superconducting materials since it requires an interaction that is at least of the order of the bandwidth, cf. Figs. 2 and 3. In principle, ultracold fermionic quantum gases can provide a platform to investigate in a controlled way the coherent modes of superfluid systems [33,53–55] where the interaction strength can be tuned via Feshbach resonances [56]. After our work was submitted, it was reported [57] that a long-lived amplitude mode, as predicted here, indeed appears in the strongly interacting regime of an ultracold quasi-2D Fermi gas.

Concerning “condensed matter systems,” we propose that the antiferromagnetic order observed in many transition metal oxide materials is an ideal playground for studying the subgap Higgs mode corresponding to the

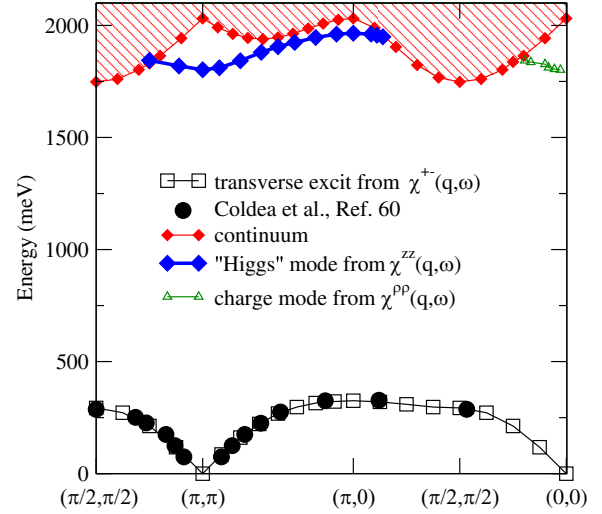


FIG. 4. Dispersion of excitations in the transverse spin channel (squares), the (spin) amplitude channel (blue diamonds), and the charge channel (green triangles) for the half-filled Hubbard model in the two-dimensional square lattice ($U/t = 8$, $t'/t = -0.2$, $t = 354$ meV). Data of the spin-wave excitations for La_2CuO_4 [60] are shown by filled, black circles. The onset of incoherent particle-hole excitations (continuum) is indicated by red circles.

associated spin amplitude fluctuations. As an example, we consider undoped high- T_c superconductors, for which the repulsive Hubbard model, including a next-nearest neighbor hopping t' , can account for the antiferromagnetic state.

For the undoped system, Fig. 4 reports previous results for the low-energy transverse magnetic excitations [58,59] in very good agreement with inelastic neutron experiments from Coldea *et al.* [60]. The same figure shows the lower bound of the continuum (red) at ~ 2 eV, which at $\mathbf{q} = 0$ would correspond to the onset of charge excitations as seen in the optical conductivity. The blue symbols report the prediction for the amplitude mode, which in the magnetic Brillouin zone would appear well below the continuum. A much weaker signal from the charge correlations (which are mixed to the longitudinal spin excitations) is also expected to appear in the “nuclear zone” (green symbols) near $\mathbf{q} = 0$. It should be noted that corresponding signatures of subgap modes have been previously obtained in the antiadiabatic limit of the TDGA [61] and from Gaussian fluctuations around slave-boson saddle points [62] without, however, linking this feature to the possibility of undamped Higgs excitations.

Also in the present case, the scalar property of subgap spin amplitude excitations makes them invisible in the optical conductivity. In principle, the scattering by impurities can provide a dipole moment; however, this would similarly broaden the onset of the continuum and therefore put in jeopardy a clear subgap feature. Polarized resonant inelastic x-ray scattering would be an ideal experimental tool for the detection of the subgap spin amplitude mode; however, in cuprates the relevant energy range is dominated

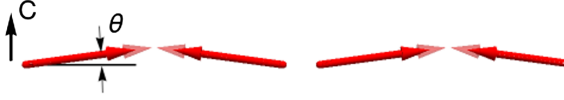


FIG. 5. Staggered spin structure with a small ferromagnetic component along the c axis as found in the LTO1 phase of La_2CuO_4 . For simplicity, we show only a Cu-O chain. Because of the Dzyaloshinsky-Moriya interaction, an amplitude excitation at the antiferromagnetic wave vector (shown as semitransparent red arrows) implies an oscillation of the ferromagnetic moment along the c direction.

by the dd excitations, which would overshadow the corresponding signatures. More promising could be a resonant inelastic x-ray scattering experiment in AgF_2 where the charge transfer excitations are at significantly higher energy than in cuprates while the energy of dd excitations is similar [63].

Here, we propose that the spin amplitude mode at $\mathbf{q} = (\pi, \pi)$ can be transferred to an out-of plane $\mathbf{q} = (0, 0)$ excitation through the Dzyaloshinsky-Moriya interaction [64], which is present in most cuprates and induces spin-canting (angle Θ , cf. Fig. 5) with a concomitant out-of plane ferromagnetic alignment. The idea, which we outline in the following, is that the amplitude (Higgs) fluctuations couple to fluctuations of this ferromagnetic moment (Fig. 5), which can be detected by magneto-optical methods.

In the low-temperature oxide LTO1 phase of La_2CuO_4 the ferromagnetic moments between adjacent planes are coupled antiferromagnetically, so that a sufficiently large magnetic field has to be applied in order to generate the spin-flop transition to a global ferromagnet [65,66]. This is not necessary in the LTO2 phase of $\text{La}_{2-x}\text{Nd}_x\text{CuO}_4$ [67–70] or in AgF_2 [71], which are globally and spontaneously weak ferromagnets even in the absence of an external field.

Fluctuations in the ferromagnetic moment M_c along the c axis can be measured using Raman scattering via the magneto-optical Faraday effect. The Raman Hamiltonian reads [72]

$$\hat{H}_R = -\frac{i}{8\pi}(E_I^a E_S^b - E_I^b E_S^a) f_{1c}^c M_c. \quad (8)$$

Here, f_{1c}^c is the relevant component of the first-order magneto-optical constant tensor and E_I^a, E_S^a are the basal-plane components of the incoming and scattered light electric field. The Raman spectra is proportional to the Fourier transform of the dynamical magnetic susceptibility $\chi_{M,M}(t) = -i\theta(t)\langle M(t), M(0) \rangle$. Because of the mixing between the amplitude mode and the magnetization (cf. Fig. 5) the Raman intensity becomes $I_R(\omega) = (E_I^a E_S^b - E_I^b E_S^a)^2 [\sin^2\theta\chi_{\Delta\Delta}(\omega) + \cos^2\theta\chi_{\perp}(\omega)]$. Here, $\chi_{\perp}(\omega)$ is a transverse susceptibility, while the term $\chi_{\Delta\Delta}(\omega)$ gives access to the Higgs mode of the antiferromagnet. By this process, an incident photon with frequency ω polarized, i.e., along the x direction is absorbed and subsequently emitted with frequency $\omega \pm \Omega$ along the y direction. Thus,

this dynamic generalization of the Faraday effect gives access to the detection of the spin amplitude mode. In contrast to the case of a SC, where the low-energy mode is shifted to the plasma frequency by long-range Coulomb interactions, this is not the case for the magnons of the antiferromagnet. Therefore, the amplitude mode, though split off from the continuum, can still decay into magnons, which, however, should only lead to minor damping due to the large energy separation between both excitations [73].

In summary, we found that in systems with an isotropic continuous order parameter, the Higgs amplitude excitation can manifest as a long-lived mode when the coupling becomes sufficiently strong. This is in stark contrast to the result from weak coupling BCS or Hartree-Fock theories, where this mode always appears at the energy of the spectral gap and is strongly damped. Besides the possibility of detecting the mode in cold atom systems, we propose that in the low-temperature LTO2 phase of $\text{La}_{2-x}\text{Nd}_x\text{CuO}_4$ and related materials the spin amplitude mode couples to the out-of-plane ferromagnetic moment, which therefore can be measured from the frequency-dependent Faraday rotated optical signal.

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Appendix: Repulsive-attractive transformation.—The sign of the interaction in the Hamiltonian Eq. (4) can be reversed by a canonical transformation that allows the mapping of different ground states [34,35]. The transformation reads

$$c_{i,\uparrow} = d_{i,\uparrow} \quad (\text{A1})$$

$$c_{i,\downarrow} = e^{i\mathbf{Q}\cdot\mathbf{r}_i} d_{i,\downarrow}^\dagger, \quad (\text{A2})$$

with $\mathbf{Q} = (\pi/a, \pi/a)$ and lattice constant $a \equiv 1$, and one obtains

$$H = -t \sum_{\langle ij \rangle} d_{i,\sigma}^\dagger d_{j,\sigma} - |U| \sum_i \left(n_{i,\uparrow} - \frac{1}{2} \right) \left(n_{i,\downarrow} - \frac{1}{2} \right) \quad (\text{A3})$$

with densities defined as before ($n_{i,\sigma} = d_{i,\sigma}^\dagger d_{i,\sigma}$). The transformation [Eqs. (A1), (A2)] also maps the staggered spin operator [Eq. (5)] to real-space Anderson pseudo-spins \mathbf{J} but with staggered charge density,

$$\mathbf{J}_i = \frac{1}{2} \begin{pmatrix} d_{i,\uparrow}^\dagger d_{i,\downarrow}^\dagger + d_{i,\downarrow} d_{i,\uparrow} \\ i[-d_{i,\uparrow}^\dagger d_{i,\downarrow}^\dagger + d_{i,\downarrow} d_{i,\uparrow}] \\ e^{i\mathbf{Q}\cdot\mathbf{r}_i} [d_{i,\uparrow}^\dagger d_{i,\uparrow} + d_{i,\downarrow}^\dagger d_{i,\downarrow} - 1] \end{pmatrix}. \quad (\text{A4})$$

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