Higgs amplitude mode in a two-dimensional quantum antiferromagnet near the quantum critical point

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Spontaneous symmetry-breaking quantum phase transitions play an essential role in condensed-matter physics1–3. The collective excitations in the broken-symmetry phase near the quantum critical point can be characterized by fluctuations of phase and amplitude of the order parameter. The phase oscillations correspond to the massless Nambu–Goldstone modes whereas the massive amplitude mode, analogous to the Higgs boson in particle physics4,5, is prone to decay into a pair of low-energy Nambu–Goldstone modes in low dimensions6–9. Especially, observation of a Higgs amplitude mode in two dimensions is an outstanding experimental challenge. Here, using inelastic neutron scattering and applying the bond-operator theory, we directly and unambiguously identify the Higgs amplitude mode in a two-dimensional $S = 1/2$ quantum antiferromagnet $\text{Cu}_2\text{H}_3\text{N}_2\text{CuBr}_4$ near a quantum critical point in two dimensions. Owing to an anisotropic energy gap, it kinematically prevents such decay and the Higgs amplitude mode acquires an infinite lifetime.

The Higgs boson appears as the amplitude fluctuation of the condensed Higgs field in the Standard Model of particle physics. Since its discovery, there has been much interest in similar Higgs boson-like particles in condensed-matter physics, such as in superconductors10–16, charge-density-wave systems17,11,12, ultracold bosonic systems13, and antiferromagnets14,15. Strictly speaking, only superconductors are analogous to the particle physics from the point that the gauge field (photon) coupling to the condensate acquires its mass (Meissner effect) by means of symmetry breaking (Anderson–Higgs mechanism). In a broad sense, nevertheless, the excitation mode of the amplitude fluctuation of the order parameter is also termed as ‘Higgs amplitude mode’ in condensed-matter physics15. These works provided new insights about the fundamental theories underlying these exotic materials.

The Higgs amplitude mode is expected in the proximity of a quantum critical point (QCP) but can decay into a pair of low-energy Nambu–Goldstone modes which makes it experimentally difficult to detect. In three-dimensional (3D) systems, where the QCP is a Gaussian fixed point, the Higgs amplitude mode is well defined near the QCP16. In contrast, in the two-dimensional (2D) case, where the longitudinal susceptibility becomes infrared divergent near the QCP, it has been debated whether the Higgs amplitude mode may not survive or it is still visible in terms of a scalar susceptibility17,18–20. Indeed, the Higgs amplitude mode in 2D was evidenced by the scalar response for an ultracold atomic gas near the superfluid to Mott-insulator transition21, although the observed spectral function is heavily damped. Note that when the Nambu–Goldstone modes become gapped, there is no such physical infrared singularity. In the following paper, we will demonstrate observation of a sharp Higgs amplitude mode through the longitudinal response being such a case in an $S = 1/2$ 2D coupled-ladder compound $\text{Cu}_2\text{H}_3\text{N}_2\text{CuBr}_4$ (abbreviated as DLCB) in the vicinity of a QCP in two dimensions.

The quantum $S = 1/2$ Heisenberg antiferromagnetic two-leg spin ladder is one of the cornerstone models in low-dimensional quantum magnetism14,22. In the one-dimensional limit of isolated spin-1/2 ladders, the ground state consists of dressed valence-bond singlets on each rung of the ladder. Interestingly, the ground state as shown in Fig. 1a can be tuned by the inter-ladder coupling from the quantum disordered (QD) state, through the QCP, to the renormalized classical regime of a long-range magnetically ordered (LRO) state23,24. In the QD phase, the magnetic excitations are triply degenerate magnons with a spin gap energy $\Delta$ which vanishes on approach to the QCP. In the LRO phase, the triplet modes evolve into two gapless Nambu–Goldstone modes reflecting spin fluctuations perpendicular to the ordered moment, accompanied by a longitudinal mode (LM) reflecting spin fluctuations along the ordered moment. The latter mode has a gap which grows continuously with the moment and is analogous to the Higgs amplitude mode. Such a LM is usually unstable and decays into a pair of transverse modes, as observed in the $S = 1/2$ coupled Heisenberg chain compound $\text{KCuF}_4$, and has a finite lifetime24,25.

In our previous work26–29, we have shown that the metal–organic compound DLCB is a unique spin ladder material where the inter-ladder coupling is sufficiently strong to drive the system into the magnetically ordered phase. Figure 1b shows the molecular two-leg ladder structure of DLCB. The collinear magnetic structure was determined by the unpolarized neutron diffraction technique and...
the U(1) symmetry is not spontaneously broken in the ordered rung interactions (Supplementary Fig. 2). In the easy-axis case, a large gap energy is expected to be ±μbμH, while the LM should remain unchanged. Consequently, if the splitting is large enough, the LM could be identified by this Zeeman effect.

Figure 2a shows the zero-field background-subtracted energy scan at the magnetic zone centre q = (0.5, −0.5, 1.5) and T = 50 mK. The spectral lineshape was modelled by a double-Lorentzian damped harmonic-oscillator (DHO) model convolved with the instrumental resolution function. The best fit yields the gap energies of TMs (&S; = ±1) and the LM (S = 0) as ΔTM = 0.34(3) meV and ΔLM = 0.48(3) meV, respectively. At μbH = 1 T in Fig. 2b, TMs (S = ±1) are split into two branches. The observed quasielastic neutron scattering hinders observation of TM (S = ±1) at μbH = 1.5 T in Fig. 2c. At this field, TM (S = ±1) is merged into the elastic line while the LM becomes clearly visible and well distinguished from TM (S = −1). Figure 2e summarizes the measured field dependences of ΔTM (S = ±1) and ΔLM (S = 0). ΔTM (S = ±1) is a function of field agree well with the Zeeman spectral splitting ±μbμHμbH using g = 2.15 and the LM is indeed field-independent within the experimental uncertainties. The small discrepancy between data and calculations at 2 T is due to the occurrence of a spin-flip transition (Supplementary Fig. 3). The analysis also indicates that the peak profile of the Higgs amplitude mode in each field is limited by the instrumental resolution within experimental uncertainty, as shown in Fig. 2f. In other words, decay of the LM/Higgs amplitude mode (S = 0) at TM (S = ±1) and another TM (S = −1) is hidden by the kinematic conditions. However, because of the limited access of the reciprocal space using a horizontal-field cryomagnet, we could not map out the excitation spectra in the Brillouin zone (BZ).

Another straightforward way to unambiguously determine the nature of spin polarization of magnetic excitations is by the polarized neutron-scattering method. In general, it is fairly challenging to carry out the polarized INS because of significant loss of neutron intensity due to neutron polarization arrangements compared with unpolarized neutron arrangements. To compensate for that, the polarized neutron data were collected using a high-flux cold-neutron spectrometer. The polarization analysis was performed using the recently developed capability of wide-angle 3He spin filters34. Figure 3a shows the θ-scans of the magnetic Bragg reflection (0, 1, −1) at T = 1.4 K with the non-spin-flip (NSF) and spin-flip (SF) configurations, respectively. The flipping ratio F can be calculated as θSF/θNSF ≃ 43(1), which corresponds to an overall polarization efficiency of (F − 1)/(F + 1) = 0.95. Furthermore, Fig. 3b shows the background-subtracted θ-scans of the magnetic Bragg reflection (0.5, 0.5, −0.5) at T = 1.4 K with the NSF and SF configurations, respectively. The scattering intensity in the SF channel dominates over that in the NSF channel, suggesting that the out-of-plane spin component is dominant, and thus confirming the determined orientation of the staggered moments.

With the high efficiency of neutron spin filters firmly established and the magnetic spin structure in DLCB well determined, we proceed to investigate the spin dynamics using polarized neutrons.
The experiment was designed in such a way that TMs and the LM can be separated from each other in the SF and NSF configurations, respectively (for further details, see Methods). For the comparison purposes, Fig. 3c shows the background-subtracted energy scan at $q = (0.5, -0.5, -0.5)$ and $T = 1.4$ K using unpolarized neutrons. Figure 3d,e shows the same energy scans with the SF and NSF configurations, respectively. And data were fitted to the same DHO model convolved with the instrumental resolution function. The spectral weight ratio between them is approximately 2.6:1.

After confirming the feasibility of such a challenging experiment, we managed to map out the excitation spectra in the BZ. Serving as a reference, Fig. 4a,b shows the false-colour maps of the spin excitation in the SF and NSF configurations, respectively. Solid lines are fits to a Gaussian function. The background-subtracted transferred energy scans at the magnetic zone centre $q = (0.5, 0.5, -0.5)$ with unpolarized neutron (SF) and NSF configurations, respectively. Solid lines are fits to a two-Lorentzian damped harmonic-oscillator model convolved with the instrumental resolution function. All experimental data were collected at $T = 1.4$ K. Error bars represent one standard deviation.

Since the one-magnon excitation of the LM is not predicted in LSWT, to analyse the experimental data with the NSF configuration, we employed the bond-operator theory (BOT) for the description of the low-energy excitations in the vicinity of the QCP on DLCB. The detailed description of the harmonic BOT can be found in refs 36–39. The ordered moment is estimated as 38.5% of the saturation value and is consistent with the experimental value of 37(5)%. The exchange interaction parameters were extracted as $J_{\text{ex}} = 0.57$ meV, $J_{\text{ex}} = 1.21$ meV, $J_{\text{ex}} = 0.11$ meV, and $\lambda = 0.95$ from the best fit. Note that the extracted parameters correspond to the renormalized parameters within the scope of harmonic BOT. Since DLCB is a weakly interacting ladder system, $J_{\text{ex}}$ and $J_{\text{ex}}$ are strongly renormalized (the former and latter are enhanced and reduced, respectively). Therefore, $J_{\text{ex}}$ is markedly larger than $J_{\text{ex}}$. The solid green lines in Fig. 4e,f show the calculated LM (Higgs amplitude mode) with a gap energy at 0.48 meV. We notice that the BOT calculation in Fig. 4f increases monotonically and deviates from the experimental data at the zone boundary, which may originate from the fact that the BOT is a mean-field treatment and application of this technique to the low-dimensional system could be limited. For the low-energy excitations, nevertheless, it works well in both QD and LRO phases. For instance, as shown in Fig. 2e, the agreement of the field dependence of the Zeeman energy term between the experimental data and the calculations by BOT is excellent.

Figure 4e,f shows the calculated excitation spectra by BOT, which reproduce the experimental data qualitatively. Thus, our conclusion that the nature of spin excitation observed in the NSF configuration is due to spin fluctuation along the staggered moment direction is fairly convincing. Note that the Higgs amplitude mode in DLCB is distinctly different from the longitudinal excitations in the $S = 1/2$ 2D Heisenberg square-lattice (HSL) antiferromagnet Cu(DCOO)$_2$$\cdot$4D$_2$O (CFTD)$_{12.5}$ because the $S = 1/2$ 2D HSL...
antiferromagnet is far from the QCP and the observed longitudinal spectra in CFTD originate from the two-magnon continuum. Consequently, the spectral lineshapes are broadened. Recent theoretical work\(^{46,45}\) suggests that there is a prominent resonance, which was proposed as a Higgs resonance with finite lifetime, inside the continuum due to the attractive magnon–magnon interaction. Moreover, the grey lines in Fig. 4ef are the calculated lower boundary of the two-magnon continuum in DLCB, which lies well above the Higgs amplitude mode. Hence, the spontaneous decay of the Higgs amplitude mode into a pair of TMs is forbidden due to the vector dependence of the intrinsic linewidth \(\Gamma\), which is limited by the instrumental resolution, as shown in Fig. 4g. It is worth pointing out that the Higgs amplitude mode is also evident from the excitation spectra with an external magnetic field applied perpendicularly to the easy-axis\(^{31}\). In that case, the Higgs amplitude mode is stable at low fields and the decay occurs beyond the crossover with the lower boundary of the two-magnon continuum at \(\sim 1.5\) T.

In summary, the unique ability of neutron scattering to probe the spin polarization of dynamic spin pair-correlation functions allows one to distinguish the Higgs amplitude mode from the dominant transverse Nambu–Goldstone modes in the two-dimensional \(S = 1/2\) antiferromagnet DLCB.

**Note added in proof:** Recently we became aware of an INS work\(^{46}\) that reports the Higgs amplitude mode in a 2D antiferromagnet Ca\(_4\)RuO\(_4\).

**Methods**

Methods, including statements of data availability and any associated accession codes and references, are available in the online version of this paper.

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Neutron-scattering measurements. Unpolarized neutron-scattering measurements using a horizontal-field superconducting magnet with a dilution fridge insert were carried out on a cold neutron-triple-axis spectrometer (FLEXX) at Helmholtz-Zentrum Berlin. The sample consists of two co-aligned deuterated single crystals with a total mass of 2.5 g and a 1.0° mosaic spread and was oriented in the (H − HL) reciprocal-lattice plane. Unpolarized inelastic neutron-scattering measurements using a standard helium-flow cryostat were carried out on a cold neutron triple-axis spectrometer (CTAX) at the High Flux Isotope Reactor, Oak Ridge National Laboratory. Polarized neutron-scattering measurements using a standard helium-flow cryostat were performed on a high-intensity multi-axis neutron crystal spectrometer (MACS) at the NIST Center for Neutron Research. The peak flux at the sample position is approximately 5 × 10^8 neutrons cm⁻² s⁻¹. The sample assembly with three co-aligned deuterated single crystals (a total mass of 3.5 g and a 1.0° mosaic spread) was oriented in the (HK − K) reciprocal-lattice plane. In all experiments, the final neutron energy was fixed at 3.0 meV and the energy resolution of FLEXX and MACS at the elastic line are 0.10 meV and 0.15 meV, respectively. The background was determined at T = 15 K under the same instrument configurations, and has been subtracted.

Polarized neutron measurements. In the experimental set-up, both incident and outgoing neutron beams were polarized by nuclear spin polarized ¹H gas cells. NMR-based inversion of the °H polarization in the polarizer cell allows polarization of the incident beam parallel or antiparallel to the vertical axis at will. The overall transmission at the beginning of a polarized neutron set-up (each run lasts about two days) is approximately 11% and reduced to approximately 5% before the °H gas cells change out. The initial flipping ratio F is about 43(1), indicating that the product of the polarizing efficiencies of the NSF cells P₄ = (F − 1)/(F + 1) is 95%. Typically, after the two-day operation, F is reduced to 20(1) and P₄ becomes 91%. Since F was always above 20, the polarization leakage effect is as small as 1/F (5%). To account for decay of the °H polarization and neutron transmission with time, the polarized neutron data were corrected by °H efficiency correction software as described in Supplementary Information.

The principles for polarized neutron scattering can be summarized as follows: phonons and structural scattering are seen in the NSF channel; components of spin fluctuations parallel to the direction of neutron polarization are seen in the NSF channel; components of spin fluctuations perpendicular to the direction of the neutron polarization are seen in the SF channel. The sample was aligned in such a way that the [0,1,1] direction in the real space is vertical. Thus, the angle α between the vertical polarization and staggered moment direction is 17.6°. In this geometry with the NSF configuration, the large fraction (cos²α ≈ 91%) is due to spin fluctuations along the direction of the staggered moment (LM) of the Higgs amplitude mode, while the remaining 9% corresponds to the spin fluctuations perpendicular to the staggered moment (TMs) and is negligible. In contrast, in the SF configuration, TMs have accounted for 91% of the contribution and the Higgs amplitude mode is negligible. Therefore, by employing polarized INS, we are able to separate the Higgs amplitude mode from the TMs in the magnetic excitation spectra. Polarized neutron measurements cover half of Brillouin zone due to the fact that the polarization efficiency becomes either significantly reduced or not available for small neutron-scattering angles.

Data analysis. The spectral lineshape in Figs 2a–d and 3d–e was fitted to the following double-Lorentzian damped harmonic-oscillator model

\[ S(E) = \frac{A}{1 + \exp(-\nu/kT)} \left[ \frac{\Gamma}{(\nu - \Delta + D^2)} + \frac{\Gamma}{(\nu + \Delta + D^2)} \right] \]

where \( k_B \) is the Boltzmann constant, \( \Delta \) is the peak position and \( \Gamma \) is the resolution-corrected intrinsic excitation linewidth, that is, half-width at half-maximum (HWHM) and convolved with the instrumental resolution function. The experimental resolution was calculated using the Reslib software. For the false-colour maps in Figs 4a–f, data were obtained by combining a series of constant-q scans along either the (H0.5 − 0.5) or (0.5 K − 0.5) direction with a step size of 0.05 r.l.u. and simulations were convolved with the instrumental resolution function where the neutron polarization factor and the magnetic form factor for Cu⁺⁺ were included. A detailed description about the determination of the lower boundary of two-magnon continuum can be found in ref. 31.

Measurement of the inter-layer dispersion. Additional unpolarized inelastic neutron-scattering measurements were performed at MACS to investigate the possible interaction between the two-dimensional layers in DLCB. For that purpose, a single crystal (~2 g) with a 1.0° mosaic spread was aligned in the (HHK) reciprocal-lattice plane. Supplementary Fig. 1 shows the measured excitation spectrum along the leg direction and the observed dispersions are fully consistent with the calculations by LSWT as described in the main text. Along the inter-layer direction, the dispersion is absent within the instrumental resolution (Supplementary Fig. 1b), indicating that DLCB is an excellent 2D spin-interacting system.

Analysis of the energy gap by tuning the inter-rung interactions. We assume that \( E_{\text{int}} \) is fixed and the inter-rung interactions vary as \( E_{\text{int}}^L = R E_{\text{int}}^L \) and \( E_{\text{int}}^R = R E_{\text{int}}^R \), where values of \( E_{\text{int}}^L \) and \( E_{\text{int}}^R \) are obtained as described in the main text. Supplementary Fig. 2 summarizes the calculations by BOT of the evolution of the spin gap energy as a function of the enhancement factor R. At small R, due to the Ising anisotropy, the triplet spin gap energy splits into a singlet (\( S = 0 \)) and a doublet (\( S = ± 1 \)). When R initially increases, the spin gap energies of both the singlet and the doublet decrease. Spin gap of the singlet closes at the QCP while the doublet remains gapless. When R further increases, the softened singlet mode acquires a spin gap again and becomes the Higgs amplitude mode. The analysis indicates that the QCP is located at \( R = 0.923 \), which is close to the case in DLCB (\( R = 1 \)) and thus confirms our conclusion in the main text. For \( R < R_c \), the quantum disordered phase is stabilized, while the long-range ordered phase is stabilized for \( R > R_c \). Calculation by BOT of emergence of the staggered moment size as a function of R is also shown in Supplementary Fig. 2.

The spin-flop transition. In DLCB, an application of a magnetic field along the easy-axis direction would lead to the spin reorientation, that is, spin-flop transition. In the spin-flop phase, \( S_z \) is no longer a good quantum number. To find out the critical field where the spin reorientation occurs, we measured the field dependence of several magnetic reflections, as shown in Supplementary Fig. 3, at FLEXX using a horizontal-field cryomagnet. The integrated-peak intensities are almost field-independent at low fields, then start to increase above 1.7 T, and finally become saturated above 2.5 T after the spin is flopped for the (0.5 − 0.5 − 1.5) and (1.5 − 1.5 − 2.5) magnetic reflections. From the fact that neutron scattering probes the components of spin fluctuation parallel to the transferring wavevector, the orientation of the ordered moment in the spin-flop phase is 90° out of the horizontal plane, with the axis of rotation approximately along \( q = (1.5, −1.5, 0.5) \). Overall, the above results confirm that in DLCB, when the field direction is aligned parallel to the easy-axis direction, \( S_z \) remains as a good quantum number at least up to 1.7 T.

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding author upon request.

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