Pressure-Induced Collapse of the Charge Density Wave and Higgs Mode Visibility in $2H$-TaS_2

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The pressure evolution of the Raman active electronic excitations of the transition metal dichalcogenides $2H$-TaS_2 is followed through the pressure phase diagram embedding incommensurate charge-density-wave and superconducting states. At high pressure, the charge-density wave is found to collapse at 8.5 GPa. In the coexisting charge-density-wave and superconducting orders, we unravel a strong in-gap superconducting mode, attributed to a Higgs mode, coexisting with the expected incoherent Cooper-pair breaking signature. The latter remains in the pure superconducting state reached above 8.5 GPa. Our report constitutes a new observation of such Raman active Higgs mode since the long-standing unique case $2H$-$NbSe_2$.

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Symmetry breaking across an electronic phase transition always occurs along with the emergence of new collective excitations, including oscillations of the amplitude of the order parameter. In charge-density-wave (CDW) systems, translational symmetry breaking gives rise to the amplitudon [1,2]. Similarly, in superconductors $U(1)$ rotational symmetry breaking gives rise to oscillations of the amplitude of the order parameter, also called the Higgs mode because of its analogy to the one found in high-energy field theories [3]. The study of these collective modes and their interaction is of great interest for the study and control of intertwined electronic orders. In the case of coexisting CDW and superconducting (SC) orders, recent studies [4–7] have shown that controlling order parameters dynamics of these coexisting orders using light pulses can induce an enhancement of the superconducting critical temperature. In the context of high T_c cuprates coexisting SC and CDW orders have also attracted great interest recently since they could lead to a nonuniform SC state, a pair density wave (PDW), with a distinctive order parameter dynamics [8,9].

Raman spectroscopy is a well-known tool for the observation of excitations in materials. It has been extensively used to study amplitudons in various systems exhibiting a CDW order [10–12]. However, the observation of a Higgs mode in a superconducting (SC) state remains elusive as it is only weakly coupled to spectroscopic probes [13] and remains short lived because of the quasiparticle continuum developing at 2Δ [14–17]. Nevertheless the detection of the Higgs mode has been recently reported using strong THz pulses in conventional [18,19] and unconventional SC [20], and also by conventional infrared (IR) spectroscopy in disordered SC [21]. However, the nature of the measured mode and the conditions of its observability are still under debate [13,17,22–26]. Early on a possible observation of the Higgs mode was also reported in the transition metal dichalcogenide (TMDC) $2H$-$NbSe_2$ using Raman spectroscopy [27–29] where the Higgs mode could be visible thanks to the coupling to the CDW amplitudon [3,16,30]. At present $2H$-$NbSe_2$ remains a unique case and other observation of Higgs modes in CDW superconductors are desirable to assess how generic is the coupling between the Higgs mode and the amplitudon.

Although in high-T_c cuprates a coexisting CDW and SC phase was recently detected [31–36], no Higgs mode has yet been identified in the Raman spectra. On the other hand, the family of the TMDCs contains few systems where SC and CDW orders coexist [37–39] but low superconducting critical temperatures prevent any Raman spectroscopy study of the SC state. For example $2H$-TaS_2, for which an incommensurate CDW develops below 77 K followed by a superconducting state below $T_c = 1$ K. In this compound, recent reports [40] have shown a dramatic increase in T_c with pressure, up to 8.5 K at 10 GPa. At such temperature an observation of the coexisting superconducting and CDW states, and of the Higgs mode, becomes accessible using Raman scattering.

In this Letter we map out the CDW phase diagram of $2H$-TaS_2 by following the CDW excitations and gap under high pressure and find that the CDW completely collapses at 8.5 GPa. In the low temperature SC state, we unravel a low energy in-gap collective mode which is attributed to a Higgs mode and whose interplay with the CDW mode points to a similar mechanism of observability as in $2H$-$NbSe_2$. Besides, in $2H$-TaS_2, this in-gap mode coexists with the usual incoherent Cooper-pair breaking peak at 2Δ, clearly differentiating both excitations, and demonstrating that the Higgs mode is a well-defined collective mode located below the continuum of quasiparticle excitations in the SC + CDW state.

Crystals of $2H$-TaS_2 were grown by chemical vapor transport from the presynthetic material, using iodine as a transport agent, as already reported [41]. Composition and phase purity were confirmed by powder x-ray diffraction, inductively coupled plasma spectrometry, and elemental analysis [42]. We have performed Raman spectroscopy measurements with a 532 nm laser line on single crystals of bulk $2H$-TaS_2 under hydrostatic pressure in a membrane diamond anvil cell using helium as a pressure-transmitting medium as described in Refs. [29,44,45] (see also the Supplemental Material [42]). We have tracked low energy excitations down to 7 cm^{-1} under extreme conditions, down to 3 K and up to 9.5 GPa. Superconductivity was accessed by performing measurements at low laser power of 0.2 mW on a 30 μm^2 laser spot (30 $W\,cm^{-2}$). We have followed simultaneously the phonons, the charge-density-wave modes, and the superconducting excitations across the pressure-temperature phase diagram.

In Fig. 1(a) we show the Raman response from $2H$-TaS_2 at ambient pressure at 13 K. Three regular phonons are measured: E_{2g}^2 (26.1), E_{2g}^1 (300.3), and A_{1g} (404.0 cm^{-1}), defined in the point group D_{6h}. The incommensurate charge-density wave (ICDW) manifests itself with amplitudons, which correspond to a soft-phonon coupled to the electronic density at Q_{CDW} and dressed by the amplitude fluctuations of the CDW order parameter [1,2]. Contrary to previous works [46] where only one amplitudon could be readily tracked, we report two well-defined amplitudons, one in each symmetry and labeled accordingly: E_{CDW} (46.5) and A_{CDW} (75.8 cm^{-1}). In addition, only in the ICDW state, we observe multiple weak peaks, labeled (*), which most probably correspond to regular phonons folded to the zone center of the Brillouin zone due to the establishment of the CDW state. Beside, as presented Fig. 1(d), a depletion of the electronic background develops in the E_{2g} symmetry below T_{ICDW}. This loss of spectral weight at low energy is attributed to the opening of a gap in the CDW state, similarly to what has been observed in rare-earth telluride prototopical CDW systems [47,48]. This gap extends up to at least $\Delta = 400$ cm^{-1}. On the contrary, no such gap is measured in the A_{1g} symmetry [see inset of Fig. 1(d)]. Finally, two peaks are also detected at ~160 (E) and ~270 cm^{-1} (A) in the E_{2g} and A_{1g} symmetries, respectively. They are measured already at 300 K in the normal state and persist up to the highest pressure. They are not associated with the CDW state and may arise from IR phonons activated by disorder.

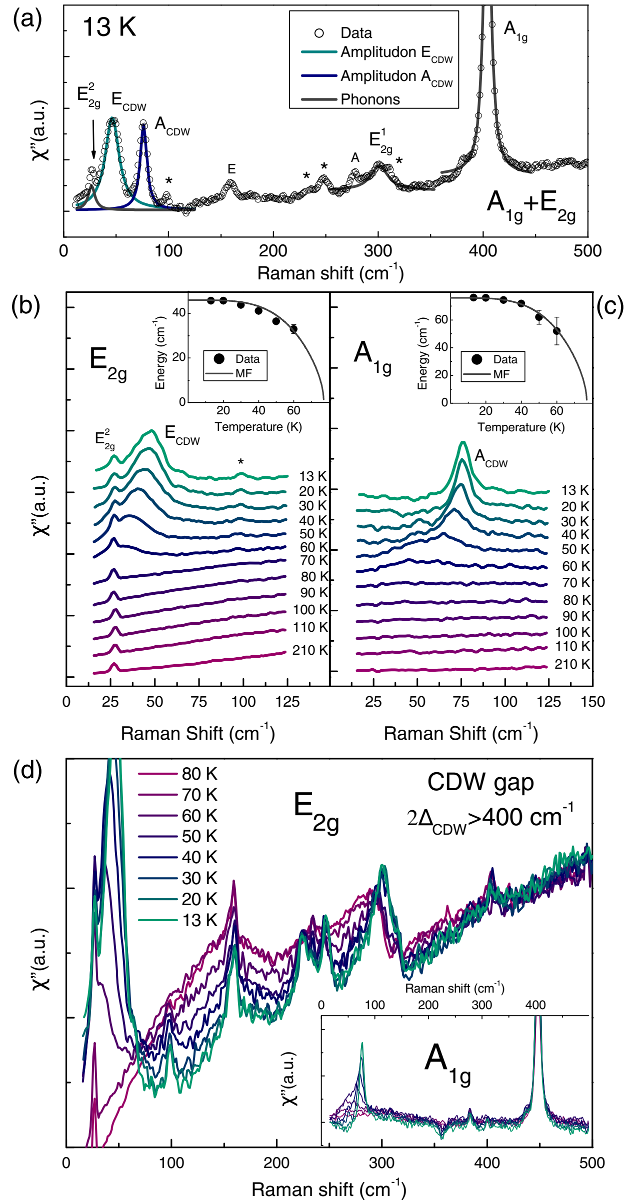

FIG. 1. (a) Raman spectra of $2H$-TaS_2 at ambient pressure at 13 K in the ICDW state and for the $A_{1g} + E_{2g}$ symmetry. We observe three phonons (E_{2g}^2, E_{2g}^1, and A_{1g}) and two CDW modes labeled according to there symmetry (E_{CDW} and A_{CDW}). Additional less intense CDW modes (denoted *) are also visible. (b), (c) Temperature dependence of the amplitudons in the pure symmetries. Insets: Energy of the amplitudons as a function of temperature. The black line corresponds to a fit using a mean-field calculation. (d) Raman spectra at various temperatures in the E_{2g} symmetry where a CDW gap opens. Inset: Raman spectra in the A_{1g} symmetry.

As shown in Figs. 1(b) and 1(c), the CDW amplitudons' behaviors at ambient pressure is typical to this kind of excitations: both lose intensity, enlarge, and soften toward the zero energy with increasing temperature toward the transition at 77 K. In the inset of Figs. 1(b) and 1(c) the amplitudons' energy is displayed as a function of temperature. They are well fitted using a mean-field-like temperature dependence [49]. This typical order-parameter-like behavior is also observed for the CDW amplitudons of $2H$-$NbSe_2$ [10]. While keeping the same energy, the folded CDW phonons (*) smoothly loose intensity and disappear at T_{ICDW}. The absence of a visible gap in the A_{1g} symmetry could be explained either because of the screening of the electronic response in this channel due to the Coulomb effect or because of a significant anisotropy of the CDW gap. Interestingly, the CDW gap in $2H$-TaS_2 appears clearly by Raman scattering whereas it remains elusive in $2H$-$NbSe_2$. This is probably due to the fact that the CDW gap is open on larger parts of the Fermi surface in $2H$-TaS_2, in good agreement with recent ARPES results [51].

Figures 2(a) and 2(b) show the pressure evolution of the Raman spectra of $2H$-TaS_2 in the $A_{1g} + E_{2g}$ symmetry at 10 and 40 K, respectively. While the E_{2g}^2 phonon hardens with pressure, its width remains stable showing a good hydrostaticity in the pressure chamber. Both A_{1g} and E_{2g} amplitudons soften and enlarge as the pressure is increased up to a complete collapse between 8 and 9.5 GPa at 10 K. (and between 6 and 7.1 GPa at 40 K). The folded CDW phonons (*) and the CDW gap are also no longer measured above 8 GPa (see Supplemental Material [42]).

Following the evolution of the amplitudons energy [see Figs. 2(c) and 2(d)], we find that the collapse of the CDW occurs at 8.5 GPa for 10 K and 7 GPa for 40 K. Hence, all three manifestations of the CDW in $2H$-TaS_2 are consistent and point toward a complete collapse of the CDW at 8.5 GPa. Hence we draw a new phase diagram for the CDW in $2H$-TaS_2 using Raman spectroscopy, as depicted in Fig. 2(e). Notable differences are obtained with previous results from transport measurement [40] where signatures of the ICDW were reported up to at least 17 GPa. This discrepancy might result from the presence of a pseudogap as reported by ARPES measurements [51] at ambient pressure above T_{CDW} and which may survive above P_c while being detected by transport measurements. Alternatively, application of high pressure could induce a loss of a long-range CDW order and a softening of the amplitudon energy [52,53], while a short-range CDW order may leave a broad signature detected by transport measurements.

We now turn to the study of the superconducting state, reached above 5 GPa by minimizing the laser heating. While entering it, new features develop above 6 GPa ($T_c > 6.5$ K) [see Fig. 3(a)]. At 6 GPa, an intense excitation can be seen at very low energy. It is visible only in the SC state vanishing completely above T_c. Its shape and position is confirmed by a second pressure run spectra reaching lower energies (~ 8 cm^{-1}). It is a narrow and intense in-gap mode (below 2Δ, See Supplemental Material [42]) and it is present, at least, in the E_{2g} symmetry [see Fig. 3(b)].

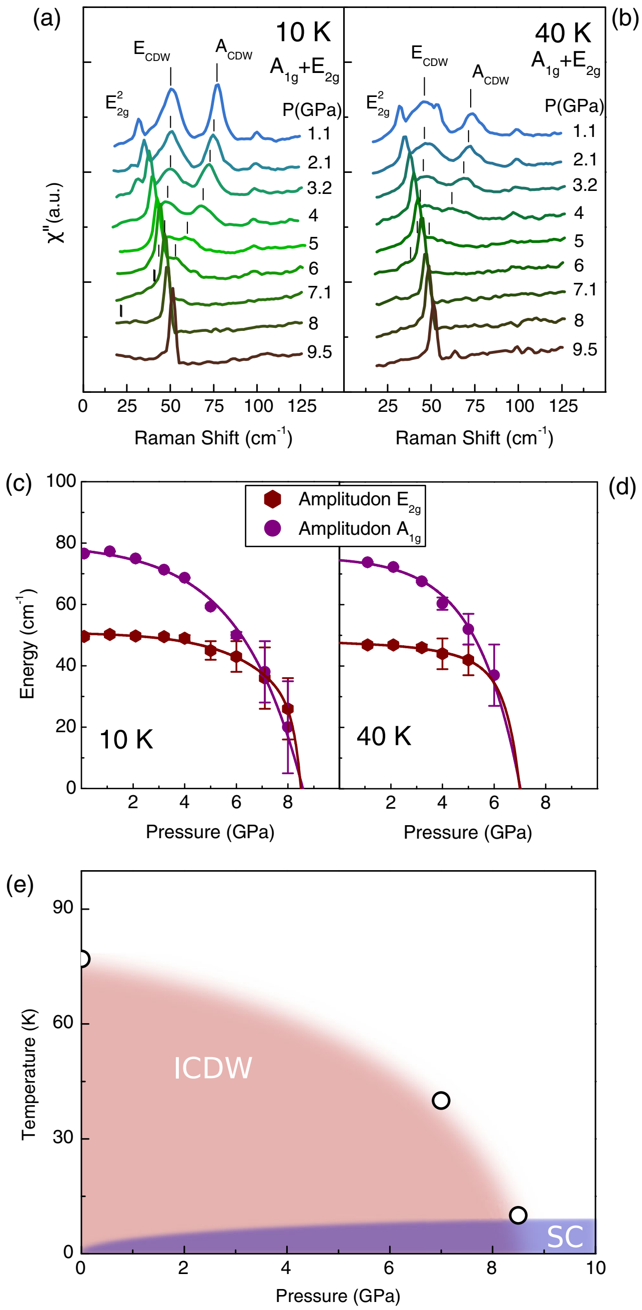

FIG. 2. (a),(b) Raman spectra of $2H$-TaS_2 in the $A_{1g} + E_{2g}$ symmetry at 10 (a) and 40 K (b) for various pressures ranging from 1.1 to 9.5 GPa. (c),(d) Pressure evolution of the energy of the CDW amplitudons at 10 K (c) and 40 K (d). The A_{1g} and E_{2g} modes soften with pressure toward zero energy (solid lines are guides for the eye). (e) Phase diagram (T,P) of $2H$-TaS_2.

While increasing further the applied pressure, an additional feature develops at 2Δ (see Supplemental Material [42]). It consists in a gap opening below ~ 20 cm^{-1} and an asymmetric peak above [see Fig. 3(c)]. This structure, sometimes observed in simple superconductors [54] and here observed up to 9.5 GPa in the pure superconducting state, is the expected incoherent Cooper-pair breaking

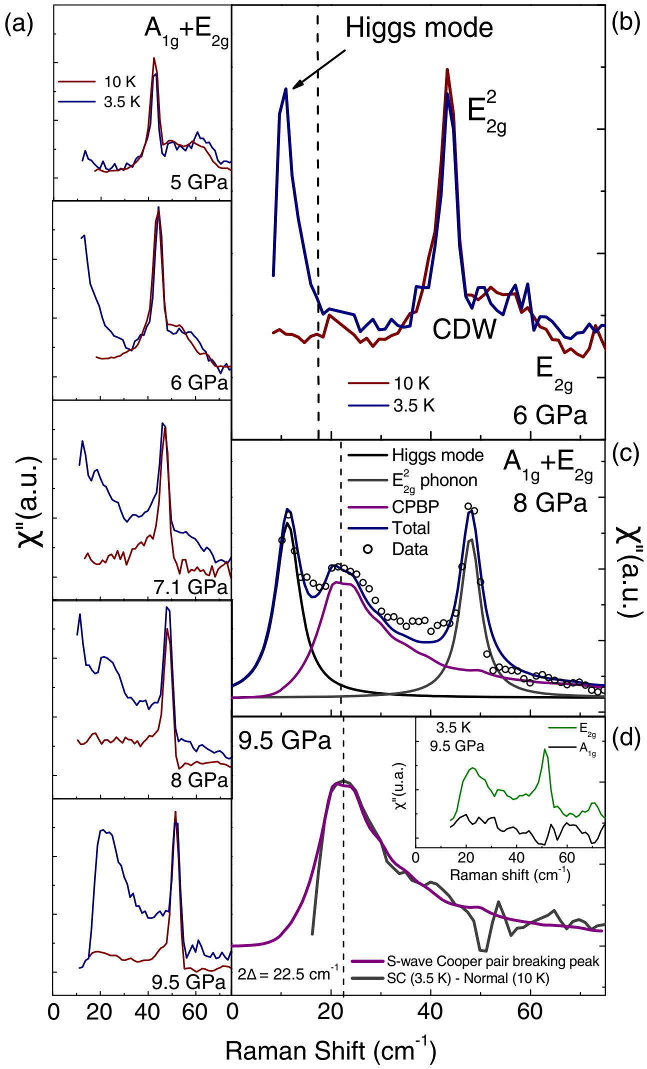

FIG. 3. (a) Raman spectra of $2H$-TaS_2 in the $A_{1g}+E_{2g}$ symmetry at 3.5 and 10 K for various pressure ranging from 5 to 9.5 GPa. (b) Low energy Raman spectra in the E_{2g} symmetry at 6 GPa below T_c. The in-gap mode is at ~10 cm^{-1}, below 2Δ (dotted line), similarly to what is observed in $2H$-$NbSe_2$. (c) Fit of the various features observed in the SC state at 8 GPa in the $A_{1g}+E_{2g}$ symmetry: a sharp in-gap Higgs mode, an incoherent Cooper-pair breaking peak (CPBP), and the E^2_{2g} phonon. (d) Subtraction of the normal state response (10 K) from the superconducting state response (3.5 K) at 9.5 GPa in the $A_{1g}+E_{2g}$ symmetry. The SC signature fits a Cooper-pair breaking peak for a BCS response in an s-wave superconductor with a gap $2\Delta = 22.5$ cm^{-1} corresponding to a T_c of 8.55 K (purple line). Inset: Raman spectra for the pure A_{1g} (black curve) and E_{2g} (green curve) symmetries. The Cooper-pair breaking peak appears only in the E_{2g} symmetry.

peak (CPBP). At the highest measured pressure (9.5 GPa), the SC transition has already reached its maximum of 8.5 K [40]. By calculating the theoretical Raman response [purple line in Fig. 3(d)] of an s-wave superconductor, a gap 2Δ of 22.5 cm^{-1} is obtained. This corresponds to a T_c of 8.85 K, using the standard weak-coupling BCS ratio, in good agreement with the T_c measured by transport measurements [40]. We note that it is only visible in the E_{2g} symmetry [see inset of Fig. 3(d)] likely due to Coulomb screening in the A_{1g} channel [55–57]. Above P_c, at 9.5 GPa, the in-gap mode disappears with the collapse of the CDW order, thus mimicking the behavior of the in-gap mode measured in $2H$-$NbSe_2$ [27–29]. Both modes in these two compounds from the same family certainly share the same nature. Up to now, the most explored hypothesis, supported by theoretical calculations and investigation under high pressure, of the nature of the in-gap modes in $2H$-$NbSe_2$, and so of this pressure-induced one in $2H$-TaS_2, is its assignment to the amplitude "Higgs" mode [3,16,30], the analogous of the Higgs boson in superconductors.

Here, the observation of both superconducting features in the SC + CDW state, the in-gap mode and the incoherent Cooper-pair breaking peak, at well separated energies is crucial. The evolution of the Cooper-pair breaking peak through the whole pressure phase diagram from the coexisting CDW + SC state to the pure SC state is gradual: the energy follows the evolution of T_c and the spectral weight continuously increases as the SC gap takes over parts of the Fermi surface previously gapped by the CDW. In particular, we do not observe any dramatic effect of the collapse of the CDW order on the incoherent CPBP. By contrast the in-gap mode intensity abruptly collapses in the pure SC state, as expected for the SC Higgs mode which couples to the Raman probe only via the CDW order. These observations rule out the interpretation of the in-gap mode as a Cooper-pair breaking peak affected by the opening of the CDW gap. They further demonstrate that the Higgs mode is a collective mode located below the continuum of quasiparticle excitations in the coexistence state consistently with theoretical work [3,16].

The proposed mechanism of observability of the Higgs mode in the presence of CDW amplitudons [3,16,30] has been shown to be consistent with the pressure dependence of the electronic excitations in $NbSe_2$ [29]. The present data suggest this mechanism is also at play in $2H$-TaS_2. This would then imply that in this compound, even if the Fermi surface is significantly gapped by the CDW [51], the superconducting and CDW gaps must overlap on some parts of the Fermi surface.

Whereas the Cooper-pair breaking peak was not observed in the coexisting SC + CDW state of $2H$-$NbSe_2$, most probably due to its reduced intensity and its proximity with the intense in-gap mode, in $2H$-TaS_2 it is possible to track the CPBP in both the pure SC and the mixed SC + CDW states. Quantitatively, it is shown that the ratio of the CPBP energy (or T_c) to the Higgs mode energy is larger in $2H$-TaS_2; i.e., the in-gap mode is pushed further at low energy. Since the electronic CDW gap is measured only in $2H$-TaS_2, we may argue that the quasiparticle states are more largely gapped by the CDW in $2H$-TaS_2 than in $2H$-$NbSe_2$ [51], giving rise to a larger overlap of the CDW and SC gaps on the Fermi surface. This would then make the Higgs mode less damped, enhancing its intensity while pushing it to lower energy (as compared to 2Δ). Alternatively this can be due to a stronger electron-phonon coupling in $2H$-TaS_2 but such a hypothesis is at present not supported by any calculation or experimental result.

In conclusion, we have reported the evolution of the excitations from the charge-density wave and the superconducting state under pressure in the dichalcogenide $2H$-TaS_2 and draw its pressure-temperature phase diagram. The pressure evolution of the amplitudons, the folded phonons, and the gap indicate a complete collapse of the incommensurate charge-density wave at about 8.5 GPa. In the coexisting charge-density-wave and superconducting states, an in-gap superconducting mode, interpreted as a Higgs mode, is reported. This constitutes a new observation of such a Raman active Higgs mode in condensed matter systems since the unique case of $2H$-$NbSe_2$. It has been clearly differentiated from the usual incoherent Cooper-pair breaking peak which survives in the pressure-induced pure superconducting state and our observations are consistent with the mechanism of observability of the Higgs mode for which an overlap of the charge-density wave and superconducting gaps on the Fermi surface is necessary.

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