

Phonon dynamics in chromium under pressure: Absence of phonon criticality at the approach of the quantum critical point

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Fermi surface nesting is key to understanding the origin of the itinerant antiferromagnetic spin density wave in chromium. This magnetic order is accompanied by a charge density wave, and both density waves disappear above the critical pressure $P_c \approx 10$ GPa at low temperatures, defining a quantum critical point. Whether this pressure-induced quantum phase transition is accompanied by a phonon instability remains an open question. Here, we use inelastic x-ray scattering to track the room-temperature acoustic phonon dispersions at pressures up to $P = 15.6$ GPa, well above P_c . We focus on the Kohn anomalies near the H and N points of the Brillouin zone, with the H point anomaly lying close to the incommensurate spin density wave ordering vector. The phonon branches harden smoothly under pressure, while the positions and wave-vector extents of both anomalies remain essentially unchanged across P_c , with no additional critical softening. *Ab initio* calculations likewise show that Fermi surface nesting remains robust above P_c . These results indicate that the pressure-induced quantum phase transition is not driven by a phonon instability and support a primarily spin density wave driven mechanism in presence of a robust Kohn anomaly.

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I. INTRODUCTION

A quantum critical point (QCP) is a phase transition at 0 K as a function of a tuning parameter that can be pressure, magnetic field, or doping [1,2]. QCPs are of great current interest because of their singular ability to influence the finite temperature properties of materials. The occurrence of this paradigmatic QCP is ubiquitous as it appears in ferromagnetic, antiferromagnetic compounds as well as in superconductor pairing systems or as a result of purely electronic interactions. Unconventional superconductivity often emerges in the proximity of a magnetically ordered phase, raising the possibility that spin fluctuations associated to the proximity of a QCP may mediate the formation of superconducting Cooper pairs.

Chromium has a simple body centered cubic crystal (bcc) structure and is considered the archetype of an antiferromagnetic spin density wave (SDW). At ambient pressure, the spin density wave develops at $T_N = 311$ K with a modulation wave vector $\mathbf{q} = (1 - \delta, 0, 0)$ and $\delta = 0.037$ at T_N . The itinerant antiferromagnetism in Cr is the result of many body effects and the value of δ is associated with the nesting wave vector of nearly octahedral electron and hole Fermi surfaces via Coulomb attraction [3]. The incommensurability arises from the slightly different sizes of the hole and electron surfaces, as it has been assessed by the change of the modulation wave vector upon doping.

Elastic neutron scattering experiments have revealed the presence of third order harmonic of the modulation at 3δ of

magnetic origin [4]. X-ray diffraction experiments sensitive to the charge modulation [5] have shown that second and fourth orders also exist at 2δ and 4δ . Importantly, the intensities of the n harmonic scale well with that of the first harmonic [4,5], $I_{n\delta}(T) \propto I_{\delta}^n(T)$. Even harmonics may result from a strain wave accompanying the onset of the SDW [6] which is parented to a charge density wave (CDW) in the language of electronic transitions. In other words, the modulation of the spin amplitude straightforwardly implies a modulation of the effective charge. The understanding of CDW phase transitions largely stems from the work on quasi-one-dimensional (Q1D) metals where the one dimensionality brings in a metal-insulator phase transition through electron-phonon coupling at a given wave vector [7]. Marked deviations from a regular phonon dispersion curve at the approach of the CDW phase transition have been reported in many of these Q1D compounds resulting in the Kohn anomalies or a highly localized softening of a phonon mode of the appropriate symmetry at the position where the incommensurability arises.

In the particular case of Cr, phonon dispersion curves measured well above room temperature have shown the presence of a phonon softening at $\mathbf{q} = (2/3, 2/3, 2/3)$ upon increasing temperature. This softening is inherent to simple metals having a bcc structure [8]. However, it has little to do with a possible electron-phonon interaction leading to the SDW phase transition at T_N . Other distinct softenings [9,10] have been observed in the phonon dispersion curve around Brillouin zone boundaries at $\mathbf{q} = (1, 0, 0)$ (or H point) and $\mathbf{q} = (1/2, 1/2, 0)$ (or N point). The dip survives in the paramagnetic phase as well. The only change observed is the overall hardening of the phonon dispersion, which is consistent with extrapolating the results of Ref. [8] to lower temperatures. These features have lent support to the oc-

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currence of a phonon-assisted mechanism at the origin of the SDW. Despite the presence of this drop, inelastic x-ray scattering studies on the temperature dependence of the phonon dispersion curves have not reported any change in the position or in the extent of the drop, thus casting doubts on the role of phonons in explaining the appearance of the SDW state [10].

Early studies on the pressure dependence of macroscopic variables in Cr have shown that $T_N \rightarrow 0$ beyond 8 GPa [11], thus supporting the idea of the presence of a QCP. A continuous disappearance of magnetic order near the critical pressure $P_c \approx 10$ GPa has been recently confirmed in x-ray diffraction studies of the P dependence of the intensity and q position of both the spin density wave and charge density wave peaks [12,13]. Close to P_c , the quantum fluctuations play a role on the disappearance of this order. The occurrence of a QCP has been confirmed in Cr-V alloys, $\text{Cr}_{1-x}\text{V}_x$ with $x_C = 0.032$, and constitutes a second road to achieve quantum criticality.

X-ray diffraction experiments under pressure of the CDW peaks have shown that the nesting at the Fermi surface improves at the approach of the QCP [13]. One might therefore expect pressure-induced phonon softening to evolve as the QCP is approached. Here, we investigate this pressure dependence in order to reveal potential soft phonon behavior.

II. EXPERIMENT

A single crystal of Cr was cut and chemically etched down to a typical thickness of several dozen micrometers. The mosaic of the sample is 0.05° on the $\mathbf{Q} = (1, 1, 0)$ and 0.05° on the $\mathbf{Q} = (2, 0, 0)$ Bragg peaks. The inelastic x-ray scattering experiment was performed at the beamline ID28 of ESRF in a backscattering configuration. The incident beam was monochromatized thanks to the (9,9,9) reflection of the Si at a wavelength of $0.6968 \text{ \AA}$, and the near backscattering configuration warrants an energy resolving power of $\approx 1.5 \times 10^{-7}$ and an ensuing Lorentzian-like energy resolution of 2.6 meV full-width half-maximum (FWHM). Energy scans were performed with a step of 0.7 meV and acquisition times of 2 min/point. Up to nine different momentum transfers can be recorded simultaneously [14].

The sample was placed in a diamond anvil cell with Helium as the transmitting pressure medium, ensuring excellent hydrostatic conditions. All the measurements reported here have been carried out at room temperature, in the paramagnetic phase. The pressure was measured by the ruby fluorescence method. The drift of the pressure was less than 2% during the entire measurements of the phonon branches (40 h). The axis of the pressure cell was placed approximately along the x-ray beam direction. The crystal orientation was chosen such that high symmetry directions of the Brillouin zone (BZ) can be accessed. Phonons were fitted with a damped harmonic oscillator (DHO) convoluted with a lorentzian energy resolution fixed to 2.6 meV. The damping of the DHO is a free parameter but is always far below the FWHM of the lorentzian, typically below 0.3 meV. The wave vectors $\mathbf{Q}$ are expressed in relative lattice units of $\|\mathbf{a}^*\|$. The lattice parameters were extracted at each pressure from the position of the $\mathbf{Q} = (1, 1, 0)$ and the

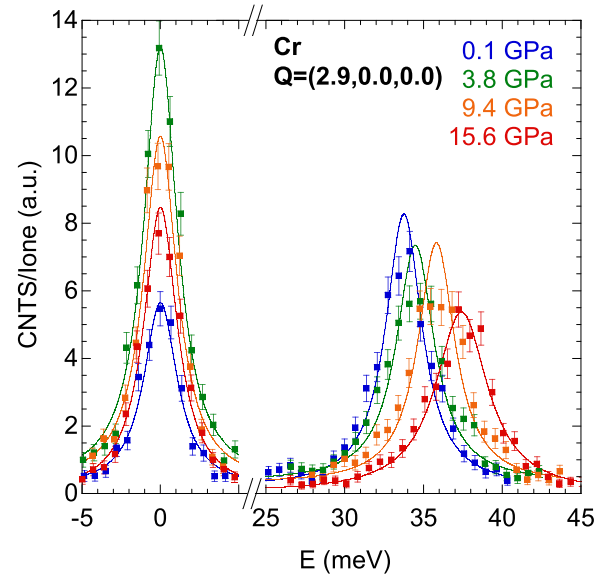


FIG. 1. Pressure dependence of the longitudinal acoustic phonon mode at 0.9Γ -H for $\mathbf{Q} = (2.9, 0, 0)$. Each point is the number of photons detected on an acquisition time of 2 minutes/point normalized by the incident flux. Solid curves are a fit of the data.

$\mathbf{Q} = (2, 0, 0)$ Bragg peaks. Even at the highest pressure, the mosaicity of these two Bragg peaks is below 0.06° .

Inelastic x-ray measurements consisted in energy scans at constant wave vector, $\mathbf{Q}$. Measurements along the ΓH direction have been performed at proximity of the H point at $\mathbf{Q} = (3, 0, 0)$: from $\mathbf{Q} = (3, 0, 0)$ to $\mathbf{Q} = (2, 0, 0)$ for the longitudinal acoustic (LA) mode, and from $\mathbf{Q} = (3, 0, 0)$ to $\mathbf{Q} = (3, 1, 0)$ for the transverse $\parallel \mathbf{a}^*$ mode. In Fig. 1 we show typical energy scans of the LA mode propagating along the $[1,0,0]$ direction at position $\mathbf{q} = (0.9, 0, 0)$, very close to the Brillouin zone boundary, and at different pressures. As expected, a hardening of this mode is clearly observed. The central line is also measured to calibrate the zero-energy position. For the ΓN direction, the T2 mode is measured close to the $\mathbf{Q} = (3, 0.5, 0.5)$, the N point along the direction $(3, 1-l, l)$. At the maximum pressure, the T1 and L modes have been also measured in the BZ close to the $\mathbf{Q} = (2, 2, 0)$ Bragg peak along $(2+h, 2-h, 0)$ and $(2-h, 2-h, 0)$ directions, respectively.

Density functional theory (DFT) calculations have been performed using QUANTUM ESPRESSO [15,16]. We use the Perdew-Burke-Ernzerhof (PBE) functional with pseudopotentials from the pseudodojo database. We use a k -point grid of $16 \times 16 \times 16$. For the calculation of electronic nesting and the Fermi surface, this grid was further interpolated using the BOLTZTRAP2 package [17]. The value of the Gaussian smearing has a large impact on the phonon frequencies, especially on the lower energy mode (T2) close to the N point (see Supplemental Material (SM) Ref. [18]). Moreover, for a large smearing value the Kohn anomalies disappear. For the calculations shown in Fig. 2, we are using a smearing of $\gamma = 0.05 \text{ Ry}$. Phonon calculations were performed using density functional perturbation theory (DFPT) on a $\mathbf{q}$ grid of $16 \times 16 \times 16$. For the calculation of the electron-phonon coupling, an interpolation of the Brillouin zone was performed.

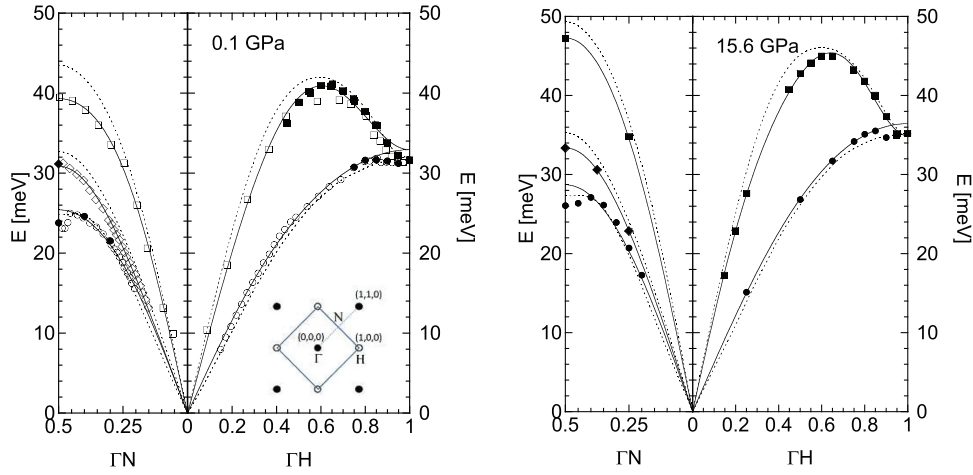


FIG. 2. Phonon spectra of the Cr along the two main Brillouin zone directions ΓH and ΓN at 0.1 GPa (left) and at 15.6 GPa (right). The open symbols are published neutron data from Ref. [9], closed symbols are from this work. The solid lines are the fit based on the Born-von Kármán calculation. The dotted lines are the *ab initio* calculation. The inset of the left panel is the representation of the first Brillouin zone of a BCC crystal. The black circles are the Bragg peaks, and open are the forbidden reflections.

III. RESULTS AND DISCUSSION

Complete phonon dispersion curves along ΓH and ΓN directions at ambient pressure and at 15.6 GPa, i.e., above the critical pressure, have been measured and are displayed in Fig. 2. The dispersion curves are fitted with a Born-von Kármán model taking into account interactions up to the fifth nearest neighbors (See SM [18]). The data from Shaw and Muhlestein [9] at ambient pressure have also been included in the fit. The force constants extracted from the fit at ambient pressure closely agree with the previously published values [8,9,19]. The *ab initio* calculation of the dispersion curves is also included. Interestingly, the different branches are qualitatively and quantitatively well described. Close to the N point, the T1 branch along the ΓN direction is slightly overestimated. It is this branch which is strongly dependent on the smearing parameter. Experimentally, this is the same phonon mode that exhibits a strong temperature dependence and is significantly affected by the diffusion jump into a nearest-neighbor vacancy [8]. This sensitivity is reproduced by the *ab initio* calculations through the electronic smearing parameter, which affects this branch most strongly. The T2 transverse modes near the N and H points are also particularly sensitive to electronic smearing because their frequencies depend on fine details of the Fermi surface (See SM Fig. S2).

As expected, all acoustic branches harden under pressure and the dispersion curves at 15.6 GPa have been fitted with the same model. The fit was initiated with the force constants obtained at ambient pressure incremented by 20%. The results are listed in the SM [18]. For the intermediate pressures, the tensor force constants are linearly interpolated and the dispersion relation calculated.

The values of the propagation velocity of the acoustic modes, and consequently of the elastic constants, are calculated from the dispersion curves. The relationship between a cubic crystal's elastic constants and sound velocities is very well known. Along the ΓH direction, $c_{11} = \rho v_L^2$ and $c_{44} = \rho v_T^2$ whereas along the ΓN direction, $c_{44} = \rho v_{T2}^2$ and

$c' = (c_{11} - c_{12})/2 = \rho v_{T1}^2$. As usual, ρ is the density and v_i the propagation velocity of the mode i . The values are shown in Table I. Elastic constants at ambient pressure and room temperature, measured by ultrasonic techniques by different groups [20,21], are $c_{11} = 350$ GPa, $c_{44} = 100.8$ GPa, and $c' = 141.1$ GPa. c' is comparable to our results, whereas c_{11} and c_{44} are stronger on the phonon dispersion. From ambient pressure to 15.6 GPa, the elastic constants increase between 22 and 34%. The anisotropy coefficient defined by the ratio c_{44}/c' is anomalously low in Cr compared to the group-VI bcc metals. At ambient temperature and pressure this ratio is below 1, the value which signs an isotropic compound at room temperature. Here, the effect of pressure is to harden the lattice and decreases the ratio c_{44}/c' . This effect is similar to that of the temperature where this ratio is increasing when the lattice is softening [8].

The pressure dependence of the longitudinal (LA) and transverse acoustic (TA) modes close to the H point, i.e., close to the positions where the SDW and CDW superstructures appear, are shown in Fig. 3. The phonon dispersion is calculated with a force tensor linearly interpolated between the ambient pressure and 15.6 GPa. As previously reported, a clear anomalous softening relative to the Born-von Kármán dispersion is visible near the H point. This softening occurs in the vicinity of the nesting vector and is therefore identified as a

TABLE I. Pressure dependence of the lattice parameter and the elastic constants. In italics, the values are calculated from the estimated force constants tensors (see text).

P (GPa)	a (Å)	c_{11} (GPa)	c_{44} (GPa)	$(c_{11} - c_{12})/2$ (GPa)
0.1	2.8885	384	127	131
3.8	2.870	<i>409</i>	<i>134</i>	<i>141</i>
9.4	2.846	<i>447</i>	<i>144</i>	<i>157</i>
15.6	2.825	489	155	176

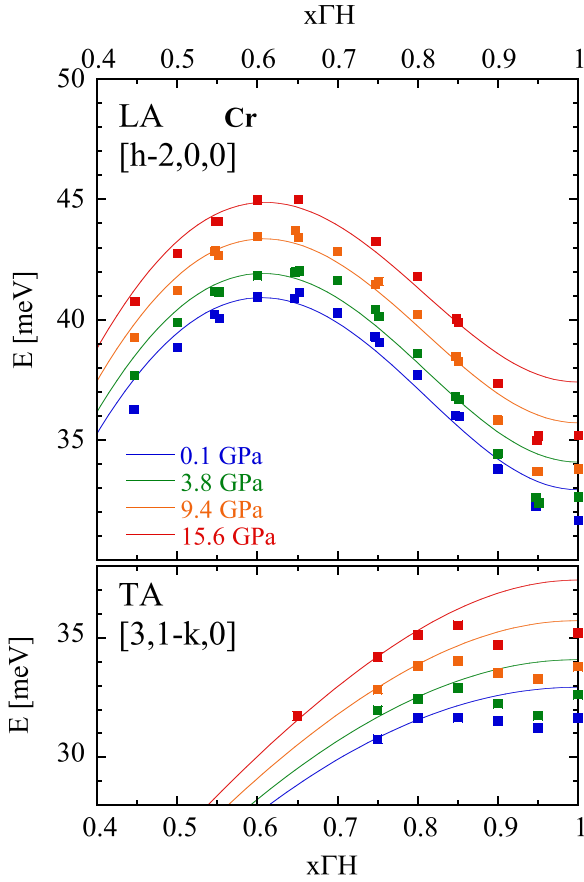


FIG. 3. Pressure dependence of the phonon dispersion of the LA (top) and TA (bottom) modes along the ΓH direction. The lines are the calculated dispersion curves with the force constants described in the paper.

Kohn anomaly [10]. The anomaly remains visible at all measured pressures, both below and above the critical pressure. Its relative magnitude and $\mathbf{q}$ -space extent remain qualitatively unchanged.

To understand the robustness of the Kohn anomalies under pressure, we calculate the Fermi surface nesting function from first principles. This function is related to the zero frequency limit of the imaginary part of the electronic susceptibility:

$$\lim_{\omega \rightarrow 0} \chi''(\mathbf{q}, \omega)/\omega = \sum_{m,n} \sum_{\mathbf{k}} \delta(\epsilon_{\mathbf{k}+\mathbf{q},m} - \epsilon_F) \delta(\epsilon_{\mathbf{k},n} - \epsilon_F) \quad (1)$$

where m and n denote the different bands, δ is the Dirac delta function, and ϵ_F is the Fermi energy. We present the pressure evolution of this function in the ΓHN plane in Fig. 4. Remarkably, the amplitudes of the nesting-function maxima change only weakly with pressure, and their positions in $\mathbf{q}$ space do not shift. Because these maxima occur at the same wave vectors as the Kohn anomalies, the persistence of the anomalies can naturally be associated with the robustness of Fermi surface nesting. Furthermore, our results are in good agreement with recent DFT simulations, which calculated the momentum dependence of the particle-hole susceptibility along specific high-symmetry directions at 10 GPa [22].

Our experimental result is in sharp contrast to other CDW systems, for example 2H-NbSe₂, where the energy shift is

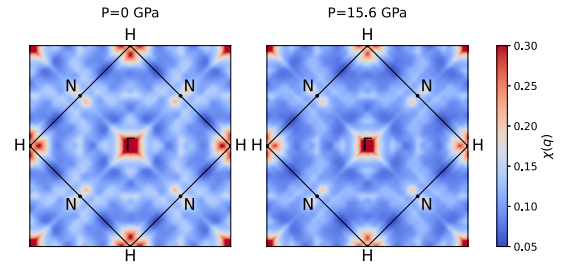


FIG. 4. Nesting function in the $(hk0)$ plane at ambient pressure (left) and at 15.6 GPa (right).

strongly pressure and temperature dependent at the proximity of the lattice instability [23].

The pressure dependence of the T2 branch near N is shown in Fig. 5. As at H , a pronounced anomaly persists over the entire pressure range. The downward deviation from the smooth Born-von Kármán dispersion signals a softening beyond conventional harmonic lattice dynamics. This softening has been attributed to enhanced electron-phonon coupling arising from interband transitions along the HN line [10]. Although no superstructure is observed near N , the anomaly coincides with a local maximum of the nesting function. Its wave vector remains unchanged between 0.1 and 15.6 GPa, while its amplitude decreases slightly.

Our first-principles calculations further connect the electronic structure to the phonon anomalies. The branches displaying the Kohn anomalies near H and N also exhibit strong electron-phonon coupling near the corresponding wave vectors (SM Fig. S1 [18]). Upon increasing the electronic smearing, which washes out fine structure near the Fermi level, both the nesting maxima and the Kohn anomalies vanish (SM Figs. S2 and S3 [18]).

Under pressure, however, the maximum deviation from the Born-von Kármán dispersion increases slightly, qualitatively consistent with the narrowing of the CDW peak near P_c reported in Ref. [13]. By contrast, the calculated nesting maxima decrease slightly over the same pressure range (Fig. 4). These contrasting trends indicate that the strength of the phonon anomalies reflects the combined influence of Fermi surface geometry and electron-phonon coupling. Neither anomaly exhibits any critical evolution across P_c . This is our main result.

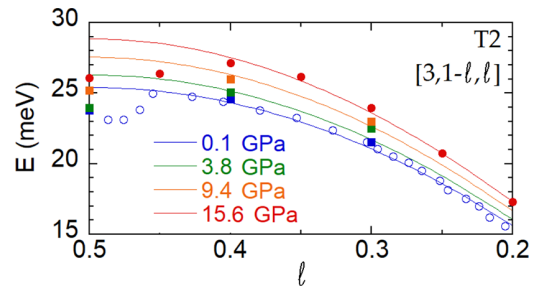


FIG. 5. Pressure dependence of the phonon dispersion of the T2 acoustic mode along the ΓN direction. $\mathbf{Q} = (3, 0.5, 0.5)$ is a N point. The blue open symbols are from Ref. [9]. The lines are the calculated dispersion curves with the force constants described in the paper.

Now, we examine the consequences of the robustness of the Fermi surface nesting on the physics of the CDW/SDW in chromium and the associated quantum phase transition. It is widely accepted that the critical pressure P_c , at which the superstructure peak associated with the CDW reaches 0 K, corresponds to the pressure at which the SDW phase of chromium is destroyed. The connection between the superstructure peaks and antiferromagnetism is established through the ambient pressure intensity and the $\mathbf{Q}$ -vector relationship between the magnetic Bragg peaks measured by neutron scattering and the charge Bragg peaks observed by x-ray diffraction. The mechanism underlying this link remains debated, with proposals including a magnetoelastic mechanism [24] or a purely electronic coupling [6]. The latter scenario could explain the uncorrelated fluctuations in the charge and spin channels [25]. Nevertheless, both mechanisms imply that the CDW is induced by the SDW, and thus the SDW ground state exists only below P_c [13]. The robustness of the Fermi surface nesting and the electron-phonon coupling above P_c evidences that another mechanism, such as, for example, a reduction in the exchange coupling, is responsible for this suppression.

Beyond chromium as a model system, numerous strongly correlated electron systems—such as cuprates [26–28] and nickelates [29,30]—exhibit both CDW and SDW phases. In these systems, the $\mathbf{Q}$ vector of the CDW can sometimes be twice that of the SDW, though this relationship is not systematic. This variability may arise from a high sensitivity to extrinsic effects, such as CDW pinning. Additionally, phonon anomalies have an unusual behavior, possibly due to a hybridization with CDW excitations [31,32]. The relationship between the Fermi surface, Kohn anomalies, and coupled density waves in weakly correlated chromium could thus serve as a reference for understanding these more complex systems.

IV. CONCLUSION

Our results for the phonon dispersion curves under pressure at room temperature, combined with *ab initio* calculations,

support the idea that a robust Fermi surface nesting and electron-phonon coupling renormalize the phonon dispersion at the H and N points. However, they do not play a direct role in the charge density wave ordering in chromium or in the critical pressure, at least not in the way that is expected from a Peierls-like mechanism where a phonon mode softens at the approach of the phase transition and this condensation leads to a superstructure at $2k_F$. Different types of (DFT-DMFT) calculations [22,33] have underlined the need of the so-called Kohn points to yield a maximum of the nonuniform magnetic susceptibility at the SDW modulation wave vector. Our data hereby support the claim that the formation of a SDW is the actual order parameter in the antiferromagnetic phase of Cr, and the formation of a CDW is collateral to the appearance of a SDW. The ensuing QCP that develops under pressure or V doping is hardly affected by the Kohn anomalies which are robust upon pressure or temperature.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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