

Multiorbital character of the density wave in trilayer nickelate superconductors

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Ruddlesden–Popper nickelates exhibit high-temperature superconductivity closely intertwined with charge and spin density waves. However, fundamental questions persist regarding the interplay between the associated density wave (DW) fluctuations and superconductivity, as well as the orbital character and symmetry underlying the DW instabilities. Here, we utilize polarized Raman scattering to investigate the phononic and electronic Raman responses of the trilayer nickelate $\text{La}_4\text{Ni}_3\text{O}_{10}$ across its concomitant charge and spin density wave transitions. In addition to distinct phonon anomalies occurring below the transition temperature, we observe a depletion of continuum spectral weight up to 114 meV and a pronounced peak centered at this energy. By combining momentum-selective information from polarized electronic Raman scattering with Raman-response model calculations based on a multiorbital Raman vertex in a reconstructed two-orbital DW state involving both $\text{Ni-}3d_{x^2-y^2}$ and $\text{Ni-}3d_{z^2}$ orbitals, we identify 114 meV as the energy scale $2\Delta_{\text{DW}}$ of the DW gap, characterized by incoherent opening and non-mean-field behavior. Furthermore, the model calculations reveal that the corresponding $2\Delta_{\text{DW}}$ peak has a multiorbital origin, requiring both orbital contributions and their mixing beyond single-orbital projections, thus shedding light on the nature of the DW instabilities in $\text{La}_4\text{Ni}_3\text{O}_{10}$.

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I. INTRODUCTION

Unconventional superconductors, including cuprates, iron pnictides, and heavy-fermion materials, host symmetry-broken phases and electronic instabilities that either compete with or facilitate superconductivity [1–6]. Raman spectroscopy has provided key insights into these intertwined phases through phonon anomalies, collective electronic modes, spectral-weight redistribution, and low-frequency scaling behavior of the electronic continuum [7–12].

Recently, Ni-based materials with either the infinite-layer or the $\text{La}_{n+1}\text{Ni}_n\text{O}_{3n+1}$ Ruddlesden–Popper (RP) crystal structure, have emerged as a distinct class of unconventional superconductors [13,14]. For RP nickelates, superconducting transition temperatures (T_c) comparable to those of several cuprate high-temperature superconductors have been reported. The bilayer compound $\text{La}_3\text{Ni}_2\text{O}_7$ exhibits superconductivity above 80 K under pressures exceeding 14 GPa

[15], while trilayer $\text{La}_4\text{Ni}_3\text{O}_{10}$ also becomes superconducting under pressure, albeit with lower T_c values between 15 and 40 K [16–19].

At ambient pressure, both $\text{La}_3\text{Ni}_2\text{O}_7$ and $\text{La}_4\text{Ni}_3\text{O}_{10}$ display charge density wave (CDW) order [20–24], which is gradually suppressed under pressure before superconductivity emerges [15,16], suggesting a competitive interplay between these phases. In $\text{La}_4\text{Ni}_3\text{O}_{10}$, a spin density wave (SDW) transition is concomitantly suppressed with the CDW under pressure [23], while residual spin fluctuations that persist in the absence of long-range order might play an essential role for superconductivity, analogous to the spin-fluctuation mediated pairing in other unconventional superconductors [1]. In $\text{La}_3\text{Ni}_2\text{O}_7$, however, moderate pressure enhances the SDW transition [25], leading to a divergence between the CDW and SDW transition temperatures prior to the onset of superconductivity, suggesting a more complex interplay among the electronic phases. More fundamentally, the basic properties of the density-wave (DW) instabilities in $\text{La}_3\text{Ni}_2\text{O}_7$ and $\text{La}_4\text{Ni}_3\text{O}_{10}$ are still under debate—including the size of the DW gaps, their momentum dependence, and the orbital character of the involved electronic states—further complicating efforts to determine how the DW orders intertwine with superconductivity.

For $\text{La}_4\text{Ni}_3\text{O}_{10}$, band structure calculations predict a multiorbital Fermi surface (FS) comprising dispersive bands with mixed $\text{Ni-}3d_{x^2-y^2}$ and $3d_{z^2}$ orbital character along with a flat band of primarily d_{z^2} character (γ band) [26–34].

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Angle-resolved photoemission spectroscopy (ARPES) studies observed a comparable FS topology but reached different conclusions regarding the involvement and position of the d_{z^2} -dominated γ band as well as the size of the DW gap [35,36]. Infrared and ultrafast optical spectroscopy, as well as scanning tunneling microscopy (STM) studies also reported conflicting DW gap magnitudes [22,37], with values as large as $2\Delta_{\text{DW}} \approx 122$ meV [38,39].

A Raman spectroscopy study extracted a gap value of $2\Delta_{\text{DW}} \approx 110$ meV and proposed that Ni $d_{x^2-y^2}$ states drive the DW order, reminiscent of the dominant role of Cu $d_{x^2-y^2}$ orbitals in the low-energy physics of cuprates [40]. This orbital-selective assessment in Ref. [40] is based on identifying FS regions with large electronic band curvature and associating them with selection rules observed in the electronic Raman response. Along similar lines, studies have overlaid Raman-vertex form factors on the FS of RP nickelates and inferred from the distribution of form-factor weight which FS sheets and orbital states participate in DW formation [41,42]. However, while such Raman-vertex form factors obtained within the effective-mass approximation and plotted on the FS of effective single-band systems, such as cuprates, can provide useful guidance for identifying the electronic states that contribute to the Raman response [7], this interpretation can be less straightforward in multiorbital systems. Indeed, recent work on Sr_2RuO_4 , a material with several FS sheets and active orbitals, has shown that the Raman vertex must be treated as a matrix in orbital space and retained explicitly in the Raman-response calculation to identify the contributing electronic states [43]. This motivates a multiorbital analysis of the Raman response of $\text{La}_4\text{Ni}_3\text{O}_{10}$, especially in view of a growing body of work emphasizing that key electronic instabilities and fluctuations in RP nickelates involve d_{z^2} or mixed $d_{x^2-y^2}/d_{z^2}$ states [26–28,30,33,34,44–51].

In this work, we use polarization-resolved Raman scattering to investigate DW order in high-quality $\text{La}_4\text{Ni}_3\text{O}_{10}$ single crystals. In the phonon sector, we observe several spectral anomalies that bear fingerprints of the DW transition. In particular, high-energy oxygen bond-stretching modes, identified via density functional theory (DFT) based phonon calculations, show clear intensity changes across T_{DW} , whereas frequency and linewidth renormalizations are subtle. Symmetric phonon line shapes indicate modest electron–phonon coupling, suggesting that DW order is primarily driven by electronic instabilities of the FS [52] rather than electron–lattice interactions. In electronic Raman scattering, we detect spectral weight depletion with decreasing temperature, indicative of a gap opening with $2\Delta_{\text{DW}} \approx 114$ meV, broadly consistent with previous Raman [40] and optical conductivity results on $\text{La}_4\text{Ni}_3\text{O}_{10}$ [38,39]. In addition, we observe a distinct polarization-dependent peak centered at the same energy. Using a Raman-response calculation that retains the multiorbital Raman vertex in a reconstructed two-orbital DW state, we identify this feature as the $2\Delta_{\text{DW}}$ peak and show that it cannot be reproduced by single-orbital projections. Instead, the spectra require both Ni- $d_{x^2-y^2}$ and d_{z^2} orbital contributions together with their mixing. We further unveil a non-BCS-like evolution of the gap opening and short-ranged DW correlations persisting above T_{DW} , and discuss implications of the multiorbital DW fluctuations for superconductivity.

II. METHODS

A. Samples

High-quality $\text{La}_4\text{Ni}_3\text{O}_{10}$ single crystals were grown using the optical floating-zone method and post-annealed in an oxygen atmosphere to optimize oxygen stoichiometry. Single-crystal x-ray diffraction (SXRD) revealed the monoclinic $P2_1/a$ structure (space group no. 14). A magnetic transition was observed in susceptibility measurements at $T \approx 140$ K. Details about the sample synthesis and characterization are given in Sec. I of the Supplemental Material [53]. Phase purity and trilayer stacking in the $\text{La}_4\text{Ni}_3\text{O}_{10}$ crystals were confirmed by scanning transmission electron microscopy (see Sec. II of the Supplemental Material [53]). The transition temperature $T_{\text{DW}} \approx 140$ K is consistent with several previous reports of the concomitant SDW and CDW ordering in monoclinic $\text{La}_4\text{Ni}_3\text{O}_{10}$ [20–24], where neutron diffraction established that the SDW emerges in the outer NiO_6 layers of the trilayer block, while SXRD indicated that the CDW modulation occurs in each of the three layers [20].

In transport measurements on crystals from the same batch used for this Raman study, a resistivity downturn was observed for pressures above ~ 20 GPa (to be published elsewhere), suggesting that our crystals exhibit high-pressure superconductivity consistent with that of $\text{La}_4\text{Ni}_3\text{O}_{10}$ samples in Refs. [16–19]. By contrast, for an orthorhombic variant, which likely corresponds to heavily oxygen-deficient $\text{La}_4\text{Ni}_3\text{O}_{10-\delta}$, the DW transition temperature deviates from 140 K [62,63], and a tetragonal variant showed neither a DW transition at ambient pressure nor superconductivity under high pressure [64].

B. Polarized Raman scattering

The Raman experiments were performed with a HORIBA Jobin Yvon LabRAM HR800 spectrometer in backscattering geometry, using a He-Ne laser (wavelength 632.8 nm) focused on the sample with a 50x magnification ultra-long working distance objective. The laser power under the microscope was less than 1 mW to minimize heating effects on the sample. The temperatures indicated in the figures of the main text were determined by Stokes/anti-Stokes intensity analysis. All plotted Raman spectra were divided by the Bose-Einstein factor. An ultralow-frequency filter (consisting of two volume Bragg-grating filters) was used to suppress elastically scattered light. For the low-temperature measurements, a KONTE helium-flow cryostat from CryoVac was used.

The Raman spectra in Figs. 2(c) and 2(d) were fitted to a superposition of Voigt profiles resulting from a convolution of the intrinsic Lorentzian line shape with the Gaussian spectrometer resolution, determined as 2 cm^{-1} from the spectrum of a Ne gas discharge lamp. For the background in the fits to Figs. 2(c) and 2(d), hyperbola and linear functions were used, respectively.

C. Phonon calculations

Details of the phonon calculations and a table of the computed Raman phonon frequencies are provided in Sec. IV of the Supplemental Material [53].

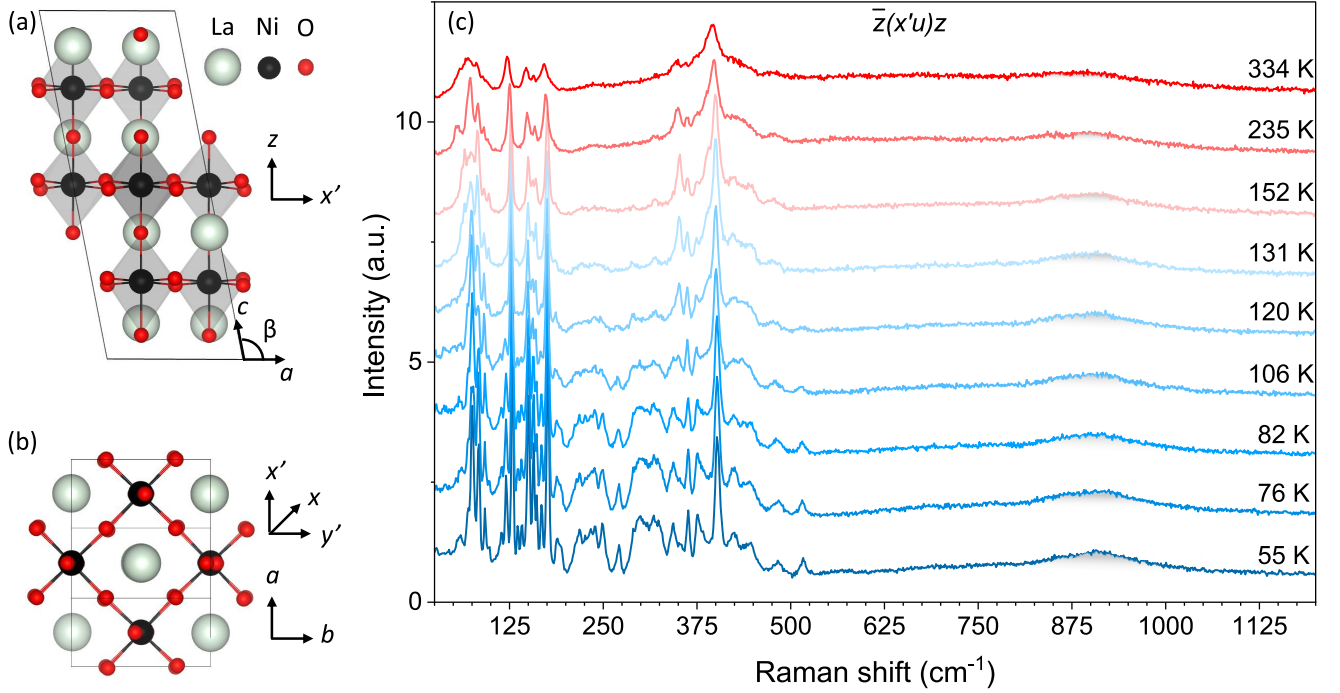


FIG. 1. Crystal structure and temperature dependence of Raman scattering in $\text{La}_4\text{Ni}_3\text{O}_{10}$. (a) Side view of the $P2_1/a$ unit cell of $\text{La}_4\text{Ni}_3\text{O}_{10}$, with the monoclinic angle β between the a and c axes indicated. The x' and z directions of the laboratory frame of the Raman experiment are also indicated. (b) Top view of the unit cell, with the a and b axes indicated, along with the x' , x , and y' directions of the laboratory frame. (c) Raman spectra at various temperatures, acquired in $\bar{z}(x'u)z$ scattering configuration (without analyzer). Spectra above (below) the DW transition are colored in red (blue). All spectra are divided by the Bose-Einstein factor and offset in the vertical direction by 1.25 a.u. for clarity.

D. Two-orbital model

The minimal model with two orbitals is defined as

$$H = H_0 + H_{\text{DW}}, \quad (1)$$

where the bare model without DW modulation is given by

$$H_0 = \sum_{\mathbf{k}} \psi_{\mathbf{k}}^\dagger H_{\mathbf{k}} \psi_{\mathbf{k}}, \quad \psi_{\mathbf{k}}^\dagger = (x_{\mathbf{k}}^\dagger, z_{\mathbf{k}}^\dagger), \quad (2)$$

where $x_{\mathbf{k}}^\dagger$ and $z_{\mathbf{k}}^\dagger$ create electrons in the $d_{x^2-y^2}$ and d_{z^2} orbitals, respectively. The 2×2 matrix $H_{\mathbf{k}}$ is the Hamiltonian in the orbital basis, with

$$H_{\mathbf{k}} = \begin{bmatrix} t_{xx}(\mathbf{k}) & t_{xz}(\mathbf{k}) \\ t_{xz}(\mathbf{k}) & t_{zz}(\mathbf{k}) \end{bmatrix}, \quad (3)$$

where the element

$$t_{xx}(\mathbf{k}) = -\frac{3}{2}t(\cos k_x + \cos k_y) + \Delta_{\text{cf}} - \mu, \quad (4)$$

describes hopping between $d_{x^2-y^2}$ orbitals,

$$t_{zz}(\mathbf{k}) = -\frac{1}{2}t(\cos k_x + \cos k_y) - \Delta_{\text{cf}} - \mu, \quad (5)$$

corresponds to hopping between d_{z^2} orbitals, and

$$t_{xz}(\mathbf{k}) = \frac{\sqrt{3}}{2}t(\cos k_x - \cos k_y), \quad (6)$$

accounts for hopping between the $d_{x^2-y^2}$ and d_{z^2} orbitals. The crystal-field splitting of the e_g orbital levels is denoted by Δ_{cf} , μ is the chemical potential, and $\mathbf{k}$ is the momentum in a

two-dimensional square lattice. Thus, the model is two dimensional in real and momentum space, while it is multiorbital in its local electronic basis.

The Hamiltonian in the band basis is diagonal

$$U_{\mathbf{k}}^\dagger H_{\mathbf{k}} U_{\mathbf{k}} = H_{\mathbf{k}}^B = \begin{bmatrix} \varepsilon_1(\mathbf{k}) & 0 \\ 0 & \varepsilon_2(\mathbf{k}) \end{bmatrix}, \quad (7)$$

with band energies

$$\varepsilon_{1,2}(\mathbf{k}) = \frac{t_{xx}(\mathbf{k}) + t_{zz}(\mathbf{k})}{2} \pm \frac{r(\mathbf{k})}{2}, \quad (8)$$

where

$$r(\mathbf{k}) = \sqrt{[t_{xx}(\mathbf{k}) - t_{zz}(\mathbf{k})]^2 + 4t_{xz}^2(\mathbf{k})}. \quad (9)$$

In the numerical calculations, we use the effective hopping parameter $t = 0.28$ eV, which is somewhat smaller than that predicted by first-principles calculations, but can be justified by considering the correlated character of $\text{La}_4\text{Ni}_3\text{O}_{10}$, which increases the effective mass. We fix $\Delta_{\text{cf}} = t$, $\mu = -1.25t$, and the density $n = 4/3$.

Within the nonresonant effective-mass approximation, the Raman vertices are obtained from second derivatives of the low-energy Hamiltonian with respect to momentum. Since the model is multiorbital, the Raman vertex is a matrix in orbital space,

$$R_{\mathbf{k}}^\gamma = \left(\frac{\partial^2}{\partial k_x^2} \pm \frac{\partial^2}{\partial k_y^2} \right) H_{\mathbf{k}} \quad (10)$$

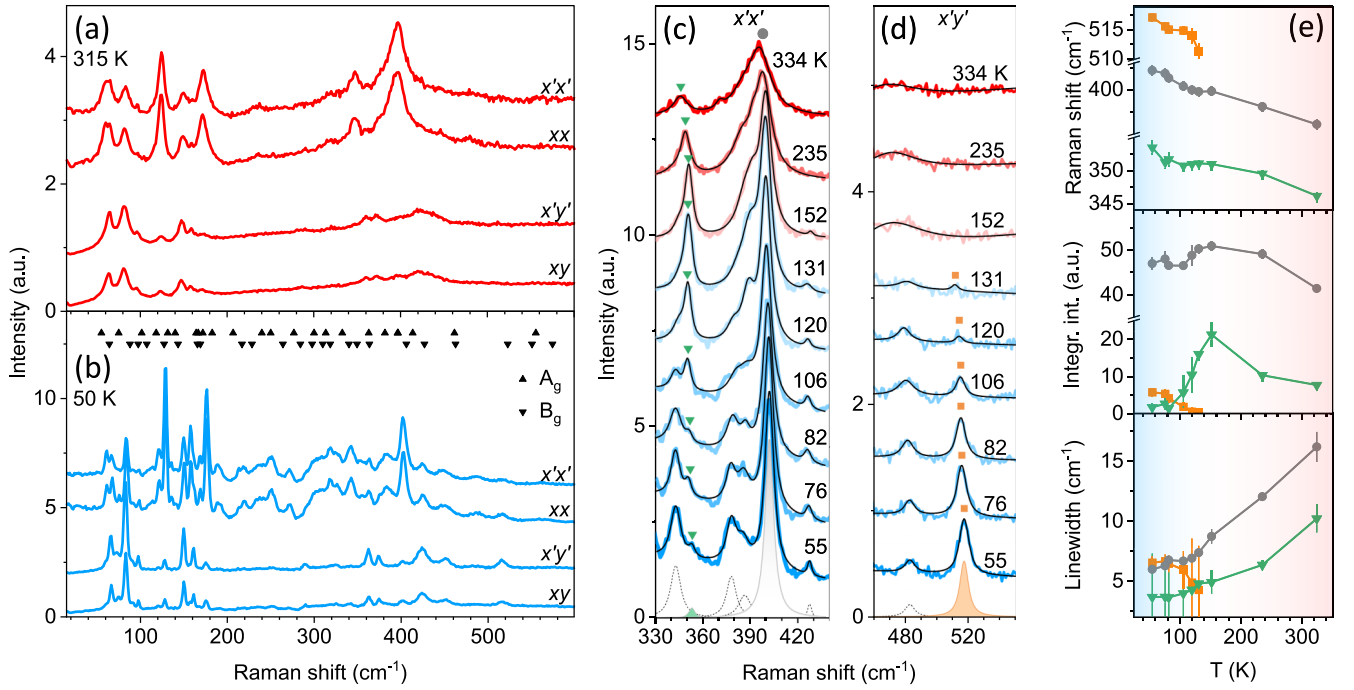


FIG. 2. Phononic Raman scattering in $\text{La}_4\text{Ni}_3\text{O}_{10}$. (a), (b) Polarization-resolved Raman spectra at 315 K (a) and 50 K (b), acquired in the $x'x'$, xx , $x'y'$, and xy scattering configurations, respectively. Black triangles in (b) indicate computed phonon frequencies of A_g and B_g symmetry, respectively. (c) Temperature dependence of selected phonon modes in the $x'x'$ channel. Solid black lines are fits of the data using Voigt profiles. The individual profiles of the 55 K fit are shown as dashed lines at the bottom of the panel. The profiles of phonon modes around 355 and 400 cm^{-1} are highlighted in green and gray, respectively. Green triangles and gray circles indicate the position of the modes at each temperature. (d) Temperature dependence of selected phonon modes in the $x'y'$ channel. The profile of a phonon mode around 515 cm^{-1} is highlighted in orange, and the orange squares indicate the position of this mode at several temperatures. Spectra in (a)–(d) are offset in the vertical direction by 0.8, 1.8, 1.5, and 0.5 a.u., respectively. (e) Temperature dependence of the energy (top panel), integrated intensity (middle panel), and linewidth (bottom panel), extracted from the fits to the modes indicated by green triangles, gray circles, and orange squares in (c) and (d). The change in background color gradient from red to blue indicates the DW transition temperature.

where $+$ ($-$) corresponds to the $\gamma = A_{1g}$ (B_{1g}) vertex. Our model does not include second-nearest neighbor hopping t' , and therefore the B_{2g} vertex is not considered. We therefore restrict the analysis to the A_{1g} and B_{1g} channels.

To describe the reconstructed phase, we introduce a DW potential with ordering vector $\mathbf{q}$,

$$H_{\text{DW}} = \delta \sum_{\mathbf{k}} (x_{\mathbf{k}+\mathbf{q}}^\dagger x_{\mathbf{k}} + z_{\mathbf{k}+\mathbf{q}}^\dagger z_{\mathbf{k}} + \text{H.c.}), \quad (11)$$

where δ is the amplitude of the DW potential, corresponding to a modulation of the local chemical potential μ . In the calculations below, we use $\delta = 0.2t$ and $\mathbf{q} = (\pi, 0)$, which is close to the dominant nesting vector of the model Fermi surface.

In the folded basis

$$\Phi_{\mathbf{k}}^\dagger = (x_{\mathbf{k}}^\dagger, z_{\mathbf{k}}^\dagger, x_{\mathbf{k}+\mathbf{q}}^\dagger, z_{\mathbf{k}+\mathbf{q}}^\dagger), \quad (12)$$

the Hamiltonian becomes

$$H = \sum_{\mathbf{k}} \Phi_{\mathbf{k}}^\dagger \mathcal{H}_{\mathbf{k}} \Phi_{\mathbf{k}}, \quad (13)$$

with

$$\mathcal{H}_{\mathbf{k}} = \begin{bmatrix} H_{\mathbf{k}} & \delta I \\ \delta I & H_{\mathbf{k}+\mathbf{q}} \end{bmatrix}, \quad (14)$$

where I is the 2×2 identity matrix and the momentum sum is over the entire Brillouin zone. The reconstructed Hamiltonian is diagonalized by a 4×4 unitary matrix $\mathcal{U}_{\mathbf{k}}$ to give four bands α with energies $E_{\alpha\mathbf{k}}$.

In the same folded orbital basis, the Raman vertex is approximated as block diagonal,

$$\mathcal{R}_{\mathbf{k}}^\gamma = \begin{bmatrix} R_{\mathbf{k}}^\gamma & 0 \\ 0 & R_{\mathbf{k}+\mathbf{q}}^\gamma \end{bmatrix}. \quad (15)$$

Transformation to the reconstructed band basis then yields

$$\mathcal{R}_{B,\mathbf{k}}^\gamma = \mathcal{U}_{\mathbf{k}}^\dagger \mathcal{R}_{\mathbf{k}}^\gamma \mathcal{U}_{\mathbf{k}}. \quad (16)$$

The Raman response is evaluated as

$$\chi^\gamma(i\omega_n) = \sum_{\mathbf{k}, i\nu_n} \text{Tr}[\mathcal{R}_{B,\mathbf{k}}^\gamma \mathcal{G}_{\mathbf{k}}(i\nu_n + i\omega_n) \mathcal{R}_{B,\mathbf{k}}^\gamma \mathcal{G}_{\mathbf{k}}(i\nu_n)], \quad (17)$$

where ν_n and ω_n are fermionic and bosonic Matsubara frequencies, respectively, and the trace is taken over the

reconstructed band indices. Here $\mathcal{G}_{\mathbf{k}}$ is the quasiparticle part of the Green's function, i.e., we account for the Raman transitions between the coherent quasiparticle α bands. Because of correlations, an energy-dependent quasiparticle weight $Z(E) \propto \eta^2/(E^2 + \eta^2)$ with $\eta = 0.5t$ is introduced, leading to

$$\mathcal{G}_{\mathbf{k},\alpha\alpha'}(i\nu_n) = \delta_{\alpha\alpha'} \frac{Z(E_{\alpha\mathbf{k}})}{i\nu_n - E_{\alpha\mathbf{k}}}, \quad (18)$$

where $E_{\alpha\mathbf{k}}$ are the eigenvalues of Eq. (14).

III. RESULTS

A. Phononic Raman scattering

High-quality $\text{La}_4\text{Ni}_3\text{O}_{10}$ single crystals with the $P2_1/a$ structure [Figs. 1(a) and 1(b)] were grown using the optical floating-zone method. A transition in the magnetic susceptibility measurements was observed at $T \approx 140$ K. For further details about the sample synthesis and characterization, see Sec. II and Secs. I and II of the Supplemental Material [53].

$\text{La}_4\text{Ni}_3\text{O}_{10}$ in the monoclinic $P2_1/a$ space group (C_{2h} point group) exhibits 99 vibrational modes at the Γ point, out of which 51 are infrared active (26 A_u + 25 B_u) and 48 are Raman active (24 A_g + 24 B_g). The corresponding Raman tensors are given in Sec. III of the Supplemental Material [53]. The relation between the x , y , and z directions of the laboratory frame of the experiment and the a , b , and c crystallographic directions is displayed in Figs. 1(a) and 1(b). Note that the monoclinic angle β introduces an offset of $\sim 11^\circ$ between the c and z directions.

Figure 1(c) provides an initial overview of the temperature dependence of the Raman spectra of a $\text{La}_4\text{Ni}_3\text{O}_{10}$ single crystal. The scattering geometry, including the photon polarizations, is given in Porto's notation, where u indicates that no analyzer was inserted in the scattered light path, i.e., $\bar{z}(x'u)z$ contains both $\bar{z}(x'x')z$ and $\bar{z}(x'y')z$ scattering contributions. In the following, we omit the labels $\bar{z}$ and z , denoting the incident and scattered photon directions in backscattering geometry, respectively, for simplicity. For technical details, see Sec. II.

In the unpolarized ($x'u$) spectrum at 334 K in Fig. 1(c), distinct phonon modes appear in the ranges 60–180 cm^{-1} and 350–450 cm^{-1} , alongside a broad hump centered around 910 cm^{-1} [shaded area in Fig. 1(c)]. Upon cooling, the phonon linewidths sharpen significantly and additional modes emerge between 60 and 515 cm^{-1} , while the broad hump shows minor temperature dependence. The most pronounced spectral changes occur between 152 K and 120 K, indicating that the DW transition at $T_{\text{DW}} \approx 140$ K affects the Raman scattering.

For a more detailed analysis, we focus in Fig. 2 on polarized spectra and features below 600 cm^{-1} , which is the typical energy range of phonons in other nickelates [54] and related oxides [7,55]. Figures 2(a) and 2(b) display Raman spectra in the $x'x'$, xx , $x'y'$, and xy polarization channels, acquired at 315 and 50 K, respectively. At both temperatures, differences between the $x'x'$ and xx spectra, as well as between the $x'y'$ and xy spectra, are subtle. This is consistent with the relatively small in-plane (orthorhombic) asymmetry between the a and b directions in $\text{La}_4\text{Ni}_3\text{O}_{10}$ (see Sec. I of the Supplemental Material [53]). By contrast, the difference between spectra in

the parallel and cross-polarization channels ($x'x'$ and $x'y'$ or xx and xy) is striking, with several modes appearing exclusively in one configuration. However, because of the monoclinic angle β of $\text{La}_4\text{Ni}_3\text{O}_{10}$, the diagonal and off-diagonal elements of the Raman tensor mix in our employed backscattering geometry, leading to the appearance of both A_g and B_g modes in each channel.

The temperature dependence of several representative phonon modes between 355 and 515 cm^{-1} is displayed in Figs. 2(c) and 2(d). Complementary analyses of lower-energy phonons are presented in Fig. S3 within the Supplemental Material [53]. In Fig. 2(c), a mode centered around 355 cm^{-1} increases in intensity upon cooling from 334 to 152 K, followed by a strong decrease in intensity for temperatures below T_{DW} [green triangles in Fig. 2(c)]. A mode around 400 cm^{-1} [gray circle in Fig. 2(c)] continuously sharpens upon cooling from 334 to 55 K, while it retains significant intensity even below T_{DW} . A mode around 515 cm^{-1} [orange squares in Fig. 2(d)] has notable intensity only below T_{DW} whereas it is absent at higher temperatures. The energy, integrated intensity, and linewidth of these three phonons, extracted from fits using Voigt profiles (see Sec. II), are summarized in Fig. 2(e).

In order to assign the phonon modes with these distinct behaviors, we perform density functional theory (DFT)-based structural relaxations of the $P2_1/a$ structure of $\text{La}_4\text{Ni}_3\text{O}_{10}$, followed by phonon frequency calculations using density-functional perturbation theory (DFPT) in combination with the PHONOPY package (see Sec. IV of the Supplemental Material [53]). The computed phonon frequencies are displayed as black triangles in Fig. 2(b), and a table of all frequencies is provided in Table S3 within the Supplemental Material [53]. The computed range, spanning from 55 to 574 cm^{-1} , agrees reasonably well with the experimentally observed phonons from 61 to 516 cm^{-1} at 50 K [Fig. 2(b)]. Note that the $P2_1/a$ symmetry persists for $\text{La}_4\text{Ni}_3\text{O}_{10}$ below T_{DW} [56,57], supporting a comparison with the 50 K spectrum.

The atomic displacement patterns of selected phonon modes are displayed in Fig. S4 within the Supplemental Material [53], including a low-energy mode around 130 cm^{-1} that involves Ni-O bending vibrations in the outer layers of the trilayer unit. Notably, this mode was identified as a coherent Raman-active phonon that can be excited by ultrashort laser pulses in Ref. [37]. The prominent phonon observed around 402 cm^{-1} in Fig. 2(c) likely corresponds to an A_g mode involving bond stretching vibrations of basal oxygen atoms in the outer Ni-O planes. This vibration pattern is reminiscent of quadrupolar modes in La_2CuO_4 , which, unlike the oxygen breathing modes, are insensitive to the on-site Coulomb repulsion U_d of the Cu $3d$ orbitals [58]. Alternatively, the observed phonon might correspond to A_g modes computed at slightly lower energies that involve quadrupolar bond-stretching vibrations of the basal oxygen atoms in the inner Ni-O plane. The phonon that emerges below T_{DW} in Fig. 2(d) near 517 cm^{-1} likely corresponds to a computed B_g mode characterized by a breathing distortion of the inner oxygen octahedra.

In regions of dense phonon clustering in Fig. 2(b), individual modes are difficult to distinguish, especially between 200 and 325 cm^{-1} in the parallel polarization channels. While

a high density of phonon modes in this range is consistent with our calculations (see Table S3 within the Supplemental Material [53]), the overlap of peaks hinders the unambiguous assignment of individual modes.

B. Electronic Raman scattering

After analyzing the phonon sector across the DW transition, we now turn to a broader energy range. In the spectra up to 1750 cm^{-1} [Fig. 3(a)], a broad peak centered around 910 cm^{-1} emerges most prominently in the $x'x'$ and xx channels. Its linewidth exceeds 150 cm^{-1} even at the lowest measured temperatures, which is substantially broader than that of the phonon modes observed up to 515 cm^{-1} . Notably, this feature appears to divide the Raman spectrum into two regimes: a high-energy region that is nearly flat above $\sim 1140\text{ cm}^{-1}$ and exhibits weak temperature dependence, and a lower-energy region with strong temperature dependence and regions of increasing or depleted spectral weight upon cooling. This separation of the Raman spectra into distinct energy scales by the 910 cm^{-1} peak suggests that it is likely associated with an electronic excitation rather than multiphonon scattering. Furthermore, the 910 cm^{-1} peak and a nearly flat background above $\sim 1140\text{ cm}^{-1}$ appear similarly in spectra acquired with both 632.8-nm (Fig. 3) and 532-nm (see Sec. V of the Supplemental Material [53]) laser excitation, confirming the robustness of the features.

The polarization dependence provides additional information on the symmetry of the electronic Raman response. The insets in Figs. 3(a)–3(d) display the temperature dependence of the Raman intensity at two representative energies (610 and 910 cm^{-1}) in the four polarization channels. In the $x'x'$ channel, the intensity at 610 cm^{-1} [Fig. 3(a), label A] decreases upon cooling, particularly below T_{DW} . This trend reflects a broader suppression of spectral weight in the surrounding energy range, at least up to 700 cm^{-1} . Below 500 cm^{-1} , the detailed behavior of any broad spectral weight distribution is masked by the rapidly growing phonon intensity. In contrast to the spectral weight depletion, the 910 cm^{-1} peak gains intensity with cooling [Fig. 3(a), label B], most strongly near T_{DW} in the $x'x'$ channel. The contrasting temperature evolution of features A and B is similarly observed in the other polarization channels (insets in Fig. 3), although it is almost absent in the xy channel.

C. Density wave in a two-orbital model

To gain insights into the origin of the 910 cm^{-1} feature and the temperature-dependent spectral weight at energies lower than 910 cm^{-1} (Fig. 3), we next develop a minimal model to compute the electronic Raman response of $\text{La}_4\text{Ni}_3\text{O}_{10}$. In constructing our model, it is crucial to take into account the multiband and multiorbital nature of $\text{La}_4\text{Ni}_3\text{O}_{10}$, in contrast to effective single-band descriptions, as often used for cuprates [2,7]. Specifically, first-principles calculations indicate that the active electronic states in $\text{La}_4\text{Ni}_3\text{O}_{10}$ near the Fermi level are composed of $d_{x^2-y^2}$ and d_{z^2} orbitals [26,27], suggesting that at least two orbitals are required to capture the essential low-energy physics and associated electronic excitations. Since both the charge- and spin-density modulations

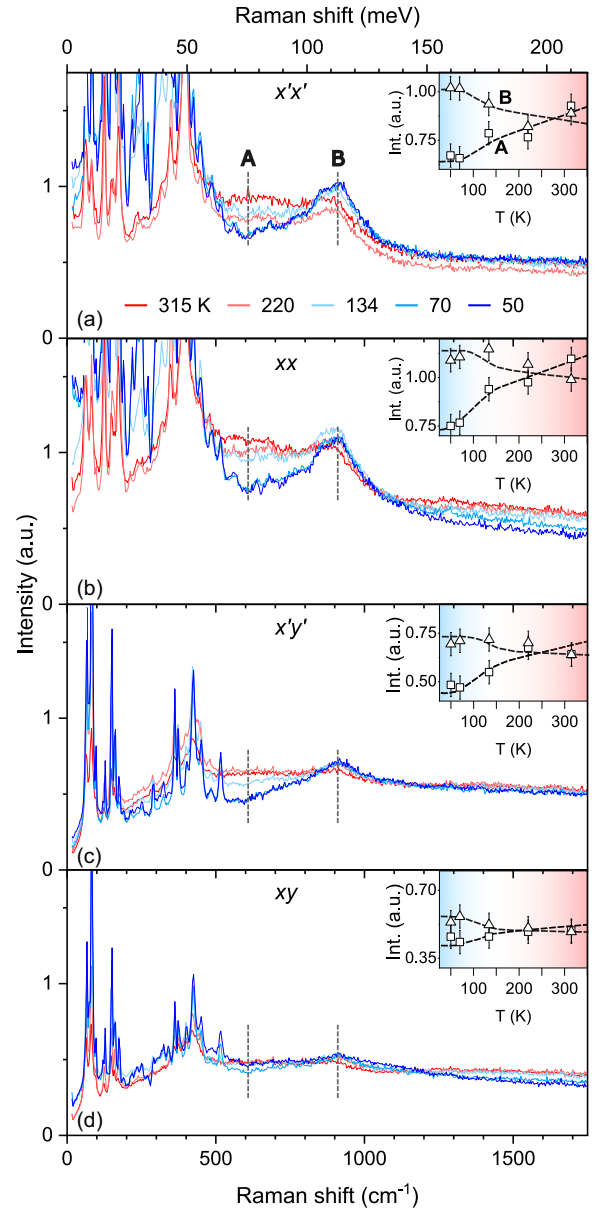


FIG. 3. Electronic Raman scattering in $\text{La}_4\text{Ni}_3\text{O}_{10}$. (a)–(d) Raman spectra across a wide energy range, acquired at different temperatures (red to blue lines) in the $x'x'$, xx , $x'y'$, and xy scattering configurations (top to bottom panels), respectively. Vertical dashed lines at 610 cm^{-1} (label A) and 910 cm^{-1} (label B) indicate regions in the spectra with a distinct temperature dependence. The insets show the corresponding temperature dependence (square symbols for A, triangle symbols for B). Dashed lines in the insets serve as a guide to the eye.

in $\text{La}_4\text{Ni}_3\text{O}_{10}$ occur primarily within the NiO_2 planes [20], we restrict our analysis to a two-dimensional model with momentum $\mathbf{k} = (k_x, k_y)$ and in-plane hopping, while retaining the two-orbital local basis. In addition, we adopt a simplified fourfold symmetric in-plane geometry for $\text{La}_4\text{Ni}_3\text{O}_{10}$, which allows for a decomposition of the Raman response into the conventional A_{1g} , B_{1g} , and B_{2g} symmetry channels, in analogy to square-lattice cuprates with CuO_2 planes. This approximation is compatible with the observed similarity of the phonon

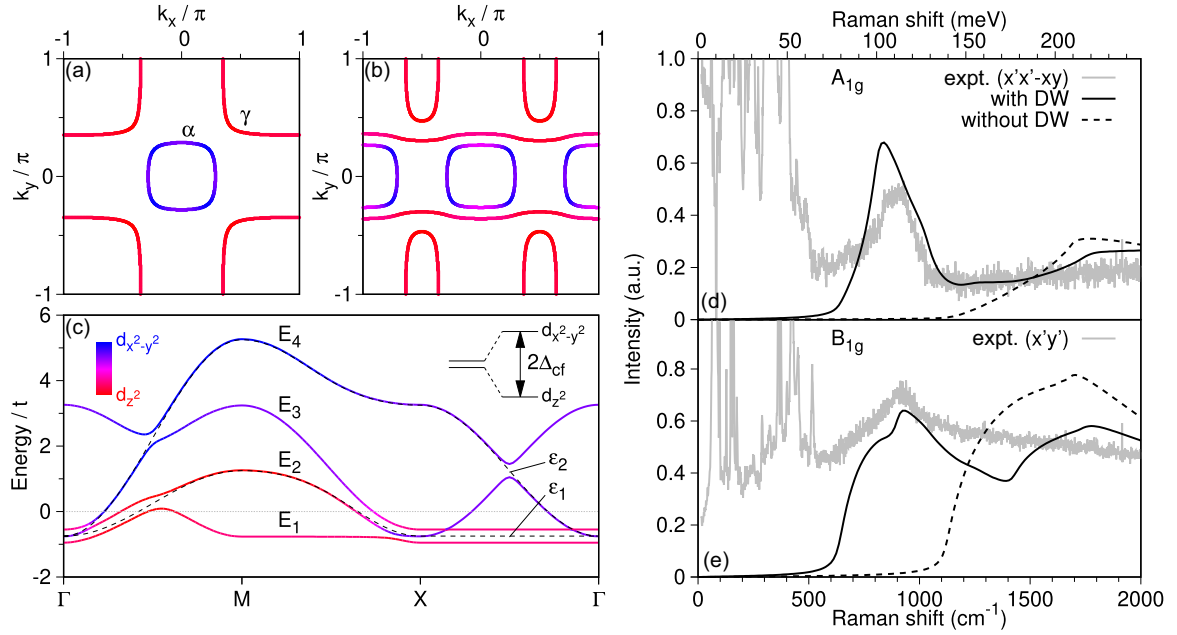


FIG. 4. Raman response in the two-orbital model with a density wave. (a) Fermi surface in the two-orbital model with α and γ sheets. The color code represents $d_{x^2-y^2}$ (blue), d_{z^2} (red), or mixed (magenta) orbital character. (b) Fermi surface in the presence of a uniaxial DW with $\mathbf{q} = (\pi, 0)$ and $\delta = 0.2t$ (see text). (c) Energy bands E_1 to E_4 of the two-orbital model in the presence of the DW, with the orbital character resolved according to the color code. For comparison, the bands ε_1 (lower black dashed line) and ε_2 (upper black dashed line) in the absence of a DW are also shown. The inset illustrates the splitting $2\Delta_{cf}$ between the $d_{x^2-y^2}$ and d_{z^2} orbital levels. (d) Raman response in the A_{1g} channel, calculated in the presence (black solid line) and absence of the DW (black dashed line). An energy-dependent quasiparticle weight was applied (see the Sec. II). Experimental data (gray line) from the $x'x' - xy$ channel are superimposed. (e) Raman response in the B_{1g} channel, calculated in the presence (black solid line) and absence of the DW (black dashed line). Experimental data (gray line) from the $x'y'$ channel are superimposed.

spectra in the $x'x'$ and xx as well as the $x'y'$ and xy channels in Figs. 2(a) and 2(b), which would not be expected in systems with pronounced orthorhombicity.

We define the model Hamiltonian as $H = H_0 + H_{DW}$, where H_0 describes electrons hopping within and between $d_{x^2-y^2}$ and d_{z^2} orbitals (for details of the model see Sec. II). The corresponding Raman vertices are obtained in the orbital basis and are therefore matrices in the $d_{x^2-y^2}/d_{z^2}$ orbital space, rather than scalar band-curvature factors. The crystal field $2\Delta_{cf}$ splits the e_g orbital manifold into two levels [inset in Fig. 4(c)], yielding dispersive bands ε_1 and ε_2 [black dashed lines in Fig. 4(c)] and two FS sheets, comprising a circular electronlike pocket with $d_{x^2-y^2}$ and minor d_{z^2} character around Γ , and a holelike pocket of predominantly d_{z^2} character centered at (π, π) [Fig. 4(a)]. This FS qualitatively reproduces the topology and orbital character reported by ARPES and first-principles calculations for $\text{La}_4\text{Ni}_3\text{O}_{10}$ [26,27,35], including the α and γ sheets as denoted in Refs. [26,27]. The β sheet, with trilayer-induced splitting into β and β' and mixed $d_{x^2-y^2}/d_{z^2}$ character [26,27], is not implemented in our minimal model.

The term H_{DW} effectively describes a density modulation within the $d_{x^2-y^2}$ and d_{z^2} orbitals, characterized by a wave vector $\mathbf{q}$ and the parameter δ , which modulates the chemical potential to simulate the presence of a DW (see Sec. II). We consider a uniaxial density modulation with $\mathbf{q} = (\pi, 0)$ and $\delta = 0.2t$, using an effective hopping parameter $t = 0.28$ eV,

reflecting effective mass renormalization because of correlations in $\text{La}_4\text{Ni}_3\text{O}_{10}$. The value of δ is chosen to reproduce the experimentally relevant DW gap scale, rather than being predicted from first principles. The resulting FS is plotted in Fig. 4(b) and the energy bands E_1 to E_4 , with their orbital character resolved, are displayed in Fig. 4(c). Furthermore, we introduce an energy-dependent quasiparticle weight (see Sec. II), which effectively reduces the intensities of high-energy interband transitions.

Figure 4(d) shows the computed Raman response in the A_{1g} channel in the presence ($\delta = 0.2t$) and absence ($\delta = 0$) of a DW. A pronounced peak emerges around 910 cm^{-1} when the DW is present. The B_{1g} response exhibits qualitatively similar behavior, with a DW-induced peak of comparable intensity, albeit with a distinct line shape [Fig. 4(e)]. Considering a simplified fourfold symmetric in-plane geometry of $\text{La}_4\text{Ni}_3\text{O}_{10}$, with the principal axes aligned along the Ni–O–Ni bond directions rather than the monoclinic setting in Fig. 1(b), the A_{1g} and B_{1g} channels correspond to the $x'x' - xy$ and $x'y'$ polarization configurations in Fig. 3, respectively. The corresponding Raman data, overlaid in Figs. 4(d) and 4(e), show good agreement with the calculated responses.

The computed Raman response in the absence of a DW term does not exhibit a peak near 910 cm^{-1} in either the A_{1g} or B_{1g} channel in Figs. 4(d) and 4(e). Instead, only a broad onset with spectral weight from interband transitions appears around 1140 cm^{-1} . Given the values of Δ_{cf} , t , and δ

used in our model calculation, a DW gap of $\Delta_{\text{DW}} = 56$ meV follows, thus strongly suggesting that the experimentally observed feature around 910 cm^{-1} (≈ 114 meV) originates from the DW and that the gap magnitude of $\text{La}_4\text{Ni}_3\text{O}_{10}$ is $2\Delta_{\text{DW}} \approx 114$ meV.

In our minimal model, we neglect second-nearest-neighbor hopping t' , which is much smaller than nearest-neighbor hopping t in $\text{La}_4\text{Ni}_3\text{O}_{10}$. The B_{2g} Raman vertex, which scales with t'^2 , is therefore vanishingly small, and a computed $2\Delta_{\text{DW}}$ peak is expected to be minor in the B_{2g} channel, consistent with the faint temperature-dependent effects observed in the electronic Raman scattering for the xy polarization channel in Fig. 3(d).

To rule out a single-orbital origin of the $2\Delta_{\text{DW}}$ peak, we next compute the Raman response, restricting the Raman vertex to either the $d_{x^2-y^2}$ or the d_{z^2} orbital sector (see Sec. VI of the Supplemental Material [53]). Notably, these single-orbital projections do not reproduce the experimentally observed intensity ratio between the $2\Delta_{\text{DW}}$ peak in the A_{1g} and B_{1g} channels, nor do they reproduce the linewidths and lineshapes. Agreement with the experiment is obtained only when both orbital contributions and their mixing are retained, demonstrating that the Raman-active DW excitation has a multiorbital character.

IV. DISCUSSION

Our Raman results in the phonon sector (Fig. 2) reveal that the lattice dynamics of $\text{La}_4\text{Ni}_3\text{O}_{10}$ are affected by the DW transition at $T_{\text{DW}} \approx 140$ K. Phonon anomalies near T_{DW} were also reported in previous Raman studies [37,40,59], although the observed phonon intensity changes in our experiment appear comparatively more pronounced [Figs. 1(c) and 2(a)–2(e)]. Moreover, the emergent clusters of densely spaced phonons between 200 and 325 cm^{-1} [see $x'x'$ and xx in Fig. 2(b)] are not apparent in Ref. [40]. On the other hand, Refs. [40,59] reported strong phonon peaks at ~ 570 and $\sim 688\text{ cm}^{-1}$, respectively, which are absent in our spectra. In particular, the $\sim 688\text{ cm}^{-1}$ peak lies far beyond the cut-off frequency of our phonon calculation for $\text{La}_4\text{Ni}_3\text{O}_{10}$ in the monoclinic $P2_1/a$ structure, possibly indicating a deviating structure because of the presence of oxygen vacancies and/or intergrown secondary phases, such as other RP or polymorph phases [60,61], in the samples of Refs. [40,59]. Orthorhombic [62,63] and nonsuperconducting tetragonal [64] variants of $\text{La}_4\text{Ni}_3\text{O}_{10}$ might also yield Raman spectra with features deviating from Figs. 1(c), 2(a), and 2(b).

Among the most striking fingerprints of the DW transition in the phonon sector is the emergence of new features below T_{DW} , including the mode around 515 cm^{-1} [orange squares in Figs. 2(d) and 2(e)] and the clusters between 200 and 325 cm^{-1} [Fig. 2(b)]. Nevertheless, we refrain from a definitive assignment of their origin to zone-folding induced by the DW, as our DFT-based phonon calculation for the ordinary $P2_1/a$ unit cell already predicts modes at these energies [triangle symbols in Fig. 2(b)]. Instead, their appearance may stem from reduced metallic screening below the metal-to-metal transition at T_{DW} [20,35] because of partial FS gapping, rendering previously overdamped $P2_1/a$ phonons observable.

Renormalization effects on the phonon self-energy, affecting both the real (frequency) and imaginary (linewidth) parts, are relatively subtle in our data compared with those of zone-center phonons of canonical DW systems [65]. The linewidth narrowing with decreasing temperature of the phonons at 355 and 400 cm^{-1} [Fig. 2(e), bottom panel] is consistent with anharmonic phonon decay, without the influence of a phase transition, and slight anomalies near T_{DW} in our data remain within the error bars. For the phonon frequencies [Fig. 2(e), top panel], we observe a subtle yet statistically significant dip below T_{DW} , interrupting an otherwise monotonic trend of anharmonic hardening. A similar dip occurs for the low-energy bending modes at 129 and 176 cm^{-1} (see Fig. S3 within the Supplemental Material [53]), involving La, Ni, and O displacements. While we cannot rule out that the frequency dip originates from a change in electron-phonon coupling across T_{DW} , the faint softening of various low- and high-energy modes, together with the decrease in the intensity of a few specific modes [orange squares in Figs. 2(d) and 2(e)], more likely reflects subtle structural changes, such as b -axis expansion [56] and small tilt-angle variations, while the $P2_1/a$ symmetry is retained [56,57].

We also note that all resolved modes exhibit symmetric peak profiles both above and below T_{DW} [Figs. 2(a)–2(d) and Sec. V of the Supplemental Material [53]], without Fano lineshapes that would signal coupling between a discrete phonon excitation and the electronic continuum [9]. Both the absence of clear self-energy renormalization and the absence of Fano lineshapes agree with theoretical predictions of weak to moderate electron-phonon coupling in RP nickelates [56,66–68]. Furthermore, this finding aligns with recent inelastic x-ray scattering (IXS) results on $\text{Pr}_4\text{Ni}_3\text{O}_{10}$, where no phonon anomalies were detected at the wavevectors associated with either CDW or SDW order [52], suggesting only a minor role for electron-phonon interactions in the DW transition. This contrasts with an unpolarized Raman study on $\text{La}_4\text{Ni}_3\text{O}_{10}$ powder [59], which proposed a crossover from an electron-phonon coupled high-temperature regime to an electron-electron correlated phase below T_{DW} .

A second energy scale, extending beyond the phonon sector up to ~ 114 meV ($\sim 910\text{ cm}^{-1}$), is characterized by temperature-dependent continuum spectral weight, with its upper bound marked by the DW peak. The distinct electronic Raman responses observed in different polarization channels (Fig. 3) show that the DW-related spectral-weight redistribution is strongly symmetry dependent. In a multiorbital system, this polarization dependence calls for an analysis based on the orbital-matrix structure of the Raman vertex [43], rather than an interpretation solely in terms of selected Brillouin-zone regions. Combined with explicit two-orbital model calculations of the Raman response, this enables the identification of the multiorbital character of the Raman-active DW excitation.

Importantly, our calculated $2\Delta_{\text{DW}}$ peaks in the A_{1g} and B_{1g} channels [Figs. 4(d) and 4(e)] align closely with the experiment, without requiring channel-dependent scaling factors, underscoring the consistency of our model. The irregular peak line shapes, including the ~ 95 meV ($\sim 760\text{ cm}^{-1}$) shoulder in B_{1g} , are also reproduced. Complementary calculations, where the Raman vertex accounts exclusively for either $d_{x^2-y^2}$ to

$d_{x^2-y^2}$ or d_{z^2} to d_{z^2} transitions, yield irregular lineshapes as well (see Sec. VI of the Supplemental Material [53]), implying that the line-shape features of the $2\Delta_{\text{DW}}$ peak are primarily governed by the FS topology rather than by distinct orbital contributions. However, without orbital mixing, the calculated relative intensity between the $2\Delta_{\text{DW}}$ peaks in the A_{1g} and B_{1g} channels as well as the linewidths deviate from the experiment (see Sec. VI of the Supplemental Material [53]), corroborating the multiorbital nature of the Raman-active DW excitation. These findings challenge the orbital-selective $d_{x^2-y^2}$ assignment of Ref. [40], whereas they align with theoretical studies emphasizing that a multiorbital basis is required to model the intertwined electronic phases and instabilities in RP nickelates [26–28,30,33,34,44,45].

As a key result of our study, the position of the $2\Delta_{\text{DW}}$ peak around ~ 114 meV marks the energy gap in $\text{La}_4\text{Ni}_3\text{O}_{10}$. This value is comparable to that determined by ultrafast optical spectroscopy on $\text{La}_4\text{Ni}_3\text{O}_{10}$ [38,39], but deviates substantially from the lower range of reported values, with Δ_{DW} between 12 and 35 meV reported by ARPES, STM, and a different Raman study [22,35,36,59]. Notably, optical conductivity measurements on bilayer $\text{La}_3\text{Ni}_2\text{O}_7$ extracted $2\Delta_{\text{CDW}} \approx 100$ meV and assigned this gap unambiguously to the CDW, since the two ordering phenomena are disjoint in that material, with $T_{\text{CDW}} \approx 110$ K and $T_{\text{SDW}} \approx 150$ K [69]. The similarity in gap magnitudes suggests that the ~ 114 meV gap in $\text{La}_4\text{Ni}_3\text{O}_{10}$ is also associated with the CDW, while the lower-energy features [35,36,59] may correspond to an SDW gap. In our spectra, the low-energy region is partly obscured by clustered phonons (Fig. 3), although we note enhanced spectral weight below ~ 50 meV in the xx channel below T_{DW} , warranting future high-resolution Raman work to clarify a possible hierarchy of DW-related energy scales.

The temperature dependence of the $2\Delta_{\text{DW}}$ peak at 114 meV and the continuum spectral weight at lower energies exhibit several unconventional characteristics: Although the peak gains most spectral weight below T_{DW} (insets in Fig. 3, label B), it is already observable above the transition temperature. This deviates from expectations based on a BCS mean-field scenario, where both the gap and the associated collective mode should vanish at $T = T_{\text{DW}}$. Instead, the persistence suggests that the CDW amplitude is already finite above T_{DW} , but that its correlation length is short-ranged and quasi-two-dimensional. In addition, the continuum depletes gradually with cooling (insets in Fig. 3, label A) rather than showing a “coherent” opening of the gap, reminiscent of pseudogap phenomena in cuprates and other correlated materials [7,70,71]. An incoherent gap opening has also been found in optical conductivity measurements on $\text{La}_3\text{Ni}_2\text{O}_7$ [69], as well as in related oxides [55]. Notably, recent Landau theory considerations predict that the CDW is subordinate to the SDW [72], and the small 12 meV gap in $\text{La}_4\text{Ni}_3\text{O}_{10}$ observed by ARPES appears to follow a conventional BCS-like temperature dependence [36].

While our minimal model is not devised to capture the full complexity of $\text{La}_4\text{Ni}_3\text{O}_{10}$, such as the three dimensionality of the DW order [35], the presence of four FS sheets [26], and structural subtleties [62–64], it nonetheless yields several robust and general insights: (i) The identification of the multiorbital character of the DW does not depend on details

of the underlying FS. Specifically, our model considers one electronlike and one holelike pocket, representing the α and γ FS sheets of $\text{La}_4\text{Ni}_3\text{O}_{10}$, with hybridized $d_{x^2-y^2}/d_{z^2}$ and nearly pure d_{z^2} orbital character, respectively. Nevertheless, similar multiorbital DW features in the Raman response can be obtained from a model that mimics the α and a β -like sheet of $\text{La}_4\text{Ni}_3\text{O}_{10}$, given that the β sheet includes at least partial d_{z^2} character (not shown here). (ii) The results are robust against the choice of the exact wave vector of the DW, as long as it connects FS regions with mixed orbital character. (iii) An upper bound for the CDW amplitude can be estimated: we obtain a modulation of ± 0.2 electrons relative to the mean, given that the canonical electron filling $n = 4/3$ and that a modulation $\delta = 0.2t$ of the chemical potential yields good agreement with the experimental data in Figs. 4(d) and 4(e), assuming that Δ_{CDW} cannot exceed the interband transitions at Γ in a fully coherent-band picture.

V. CONCLUSIONS

In summary, our polarization-resolved Raman study uncovers the phononic hallmarks of the DW transition in $\text{La}_4\text{Ni}_3\text{O}_{10}$, including the emergence of modes below T_{DW} , intensity changes of persisting modes, and subtle frequency anomalies. The combination of the momentum selectivity of electronic Raman scattering and two-orbital model calculations enables the identification of the $2\Delta_{\text{DW}}$ peak of $\text{La}_4\text{Ni}_3\text{O}_{10}$ and its multiorbital origin, involving both Ni- $d_{x^2-y^2}$ and d_{z^2} states. Spectral weight within the DW gap of approximately 114 meV depletes incoherently with decreasing temperature, while the $2\Delta_{\text{DW}}$ peak persists even above T_{DW} . The corresponding ratio $2\Delta_{\text{DW}}/k_{\text{B}}T_{\text{DW}}$ exceeds the weak-coupling BCS limit, signaling short-range correlations and the strong-coupling character of the DW instability.

Notably, our results lend strong support to theoretical models of RP nickelates that incorporate multiorbital DW instabilities, particularly those that assign prominent roles to the d_{z^2} states [27,28,30,33,34,44,45]. Given that both the orbital character and momentum structure of the hybridized Ni- $d_{x^2-y^2}/d_{z^2}$ DW correlations identified here in the ordered phase at ambient pressure exhibit close correspondence to the persisting DW fluctuations at high pressure, several constraints on superconducting pairing models can be inferred. Specifically, if these fluctuations either compete with or mediate superconductivity, their finite wavevector and interorbital nature might favor pairing states with sign-changing order parameters between distinct FS sheets, supporting extended $s^{\pm}$ -wave pairing symmetry as a viable candidate [27–29,33,73], and strongly motivating future Raman investigations not only of the pressure dependence of the DW phenomenon, but also of the potential appearance of a superconducting pair-breaking peak in RP nickelates.

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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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