

Lattice instability in the charge density wave system RPt_2Si_2 ($R = \text{Pr, Nd}$)

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Charge-density-wave (CDW) order in RPt_2Si_2 ($R = \text{Pr, Nd}$) develops through a complex sequence of structural and electronic transitions. Using atomic pair distribution function analysis and large-scale structural modeling, we show that both compounds exhibit local lattice distortions dominated by locally static Pt displacements already present at room temperature, prior to the onset of long-range CDW order. Upon cooling, these distortions become progressively correlated and evolve into superstructures characterized by alternating short and long Pt-Pt distances, forming lattice modulations along different crystallographic directions. Our results suggest a predominantly order-disorder character of the higher-temperature second-order transitions, whereas the lower-temperature first-order transitions involve a significant additional displacive component, superimposed on the preexisting Pt displacements, and further reorganization of the CDW state. These findings establish a structural connection between intrinsic lattice instabilities and the development of long-range CDW order in $PrPt_2Si_2$ and $NdPt_2Si_2$.

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I. INTRODUCTION

Charge-density waves (CDWs) represent a collective electronic state characterized by a long-range periodic modulation of the electronic charge density coupled to a static periodic distortion of the crystal lattice [1–7]. Typically, the onset of long-range CDWs is signaled by the emergence of superlattice reflections and significant changes in the electronic properties, in particular a resistivity increase. However, despite sustained research efforts over the past three decades, the role of lattice distortions in the emergence of CDWs remains unclear. Addressing this question is important because CDWs often compete or coexist with other collective electronic states, particularly magnetic order and superconductivity, giving rise to complex phase diagrams [8–13]. Recent studies have demonstrated that these phenomena can be systematically investigated by tuning parameters such as chemical composition, temperature, magnetic field, and pressure, and by examining the resulting evolution of the electronic properties and associated charge and lattice modulations. Such approaches provide a pathway toward understanding the interplay among electronic, lattice, charge, and spin degrees of freedom in materials exhibiting competing or coexisting ordered electronic states [14–18].

In this context, rare-earth intermetallics of RPt_2Si_2 family ($R = \text{La, Pr, and Nd}$) provide particularly compelling case studies. They crystallize in the CaBe_2Ge_2 type structure with tetragonal space group (S.G.) $P4/nmm$ symmetry. This structure is known as the “perovskite” of intermetallics and is adopted by many topical materials, including the 122-type AT_2Pn_2 pnictide superconductors ($A = \text{alkaline or rare earth,}$

$T = \text{transition metal, and Pn} = \text{As, Sb, or P}$) [19–21]. It features a stack of alternating $\text{Si}(1)\text{-Pt}(1)\text{-Si}(1)$ and $\text{Pt}(2)\text{-Si}(2)\text{-Pt}(2)$ layers where RE atoms occupy the open space between the layers and Pt atoms in the layers form square nets, as shown in Figs. 1(a) and 1(b). Depending on the type of rare-earth species, members of the family have been found to exhibit CDW, superconducting (SC), and/or magnetic order at low temperature [22–28]. In particular, in nonmagnetic LaPt_2Si_2 , CDW order has been suggested to emerge below $T_2 \sim 105$ K, where the electronic transport properties exhibit an abrupt change associated with a first-order transition. In addition, the material becomes a superconductor at $T_{\text{SC}} = 1.22$ K [26,29–32]. More recent single crystal diffraction studies, however, have revealed two additional phase transitions at $T_1 = 230$ K and $T_3 = 60$ K. The corresponding anomalies in the electronic transport properties are considerably weaker than those observed at T_2 , suggesting that these phase transitions are second order [33]. These findings indicate that CDW order in LaPt_2Si_2 emerges already below room temperature and evolves through a sequence of phase transitions upon cooling. In the case of nonsuperconducting PrPt_2Si_2 , a first-order CDW transition has been observed at about $T_2 \sim 95$ K, where the electronic transport properties change abruptly [34]. Two additional, apparently second-order, phase transitions at $T_3 = 60$ K and $T_1 = 250$ K have been inferred from anomalies in the thermal transport properties [34,35]. These findings indicate that CDW order in PrPt_2Si_2 also emerges already below room temperature and evolves through multiple phase transitions upon cooling. However, previous diffraction studies have not provided a clear characterization of the local lattice distortions accompanying the CDW order. Similar to LaPt_2Si_2 , single-crystal diffraction studies of nonsuperconducting NdPt_2Si_2 revealed weak satellite reflections emerging upon cooling at a

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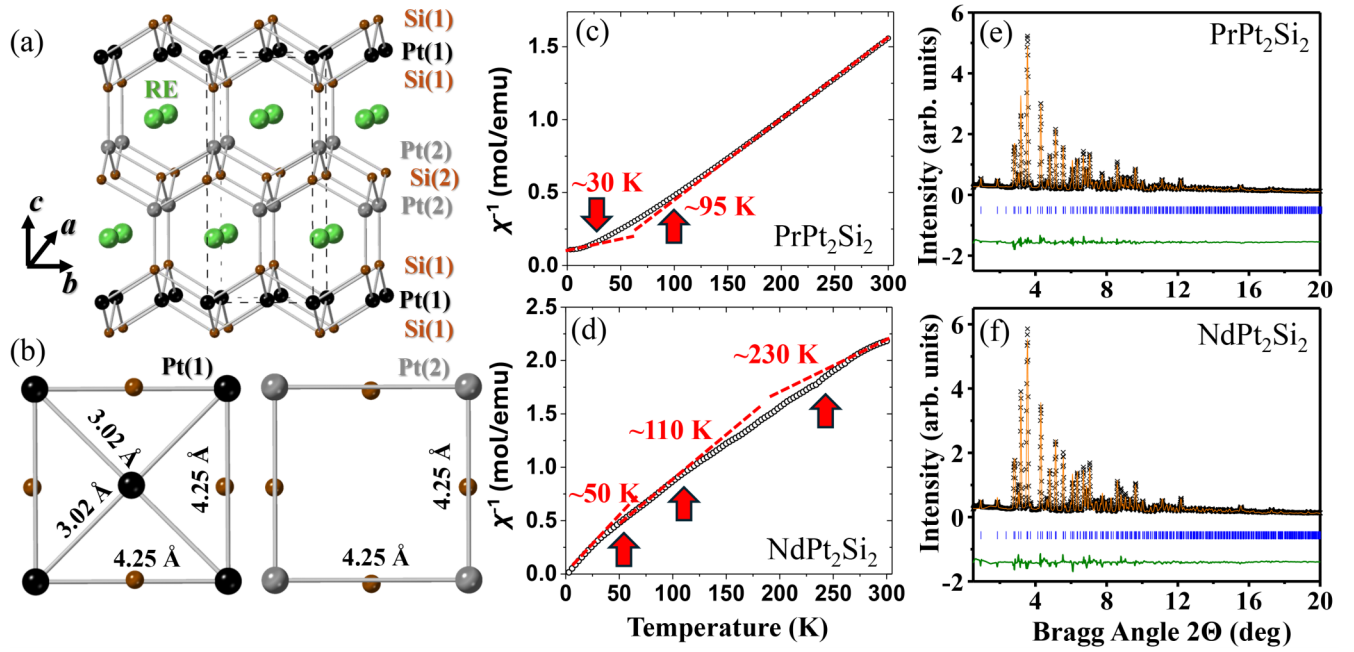


FIG. 1. (a) Fragment from the ideal tetragonal crystal structure of RPt_2Si_2 ($R = \text{Pr}, \text{Nd}$) materials featuring a stack of alternating Pt(2)-Si(2)-Pt(2) and Si(1)-Pt(1)-Si(1) layers, where RE atoms (green) sit between the layers. Pt(1), Pt(2) and Si atoms are given as black, gray, and brown circles. (b) Projections of the square planes of Pt(1) and Pt(2) atoms in the respective layers with the near-neighbor Pt-Pt distances shown. (c),(d) Inverse magnetic susceptibility, χ^{-1} , for $PrPt_2Si_2$ and $NdPt_2Si_2$ as a function of temperature. Both data sets show deviations from linearity (see red arrows), as indicated by changes in the slope of the dashed red lines used as a guide to the eye. (e),(f) Successful Rietveld fits (red line) to XRD patterns (symbols) for $PrPt_2Si_2$ and $NdPt_2Si_2$ collected at room temperature. The residual difference (green line) is shifted for clarity. Vertical blue bars show positions of Bragg peaks. The whole pattern goodness-of-fit factor is about 9% for both fits.

relatively poorly defined transition temperature, T_1 , between approximately 300 and 260 K, indicating the onset of long-range CDW order with a modulation vector close to $(1/3, 0, 0)$. Upon decreasing the temperature down to $T_2 \sim 100$ K, the intensity of the satellites has been found to increase. Further temperature decrease down to 50 K has been found to reduce their intensity, while another set of satellite reflections appeared with a modulation vector close to $(1/6, 1/6, 0.5)$. A pronounced steplike anomaly in the temperature evolution of the resistivity, ρ , has been observed at $T_3 \sim 50$ K, indicating a first-order CDW transition [36]. These observations indicate that CDW order in $NdPt_2Si_2$ also evolves through a sequence of phase transitions upon cooling, involving a progressive reorganization of the CDW state and possibly the coexistence of distinct lattice modulations. Notably, the resistivity anisotropy for both $PrPt_2Si_2$ and $NdPt_2Si_2$ has been found to be relatively small ($\sim 1 : 3$) compared with canonical quasi-two-dimensional CDW systems such as $NbSe_3$, where it reaches $\sim 1 : 20$ [37], suggesting that the CDW develops along more than one crystallographic direction. Taken together, these studies suggest that the CDW phenomena in RPt_2Si_2 ($R = \text{La}, \text{Pr}, \text{Nd}$) evolve in a complex manner as a function of both chemical composition and temperature. In $LaPt_2Si_2$, the lattice distortions accompanying the charge modulations have been suggested to occur within the Pt(1) planes and affect the contribution of Pt(1) 5d electrons to the density of states (DOS) at the Fermi level [38]. In contrast, little is known about the lattice distortions in $NdPt_2Si_2$ and $PrPt_2Si_2$, preventing a clear understanding

of their role in the CDW phenomena exhibited by these materials.

In this paper, we investigate lattice distortions in $NdPt_2Si_2$ and $PrPt_2Si_2$ using a combination of total x-ray scattering experiments and large-scale structural modeling. As demonstrated by a number of prior studies, the approach has proven effective for investigating the origin of CDWs in several families of materials exhibiting competing or coexisting ordered states [14, 15, 17, 39–45]. We find that both $NdPt_2Si_2$ and $PrPt_2Si_2$ exhibit lattice distortions within the Pt planes. One distortion involves large in-plane displacements of Pt(1) atoms from their ideal positions in the undistorted tetragonal lattice, giving rise to two distinct sets of Pt(1)-Pt(1) nearest-neighbor distances already at room temperature, prior to the onset of long-range CDW order. The displacement amplitude increases upon cooling, resulting in the emergence of a distinct pattern of four sets of Pt(1)-Pt(1) nearest-neighbor distances below approximately 100 K. A second type of distortion involves both in- and out-of-plane displacements of Pt(2) atoms, which are relatively small at room temperature but become significantly more pronounced upon cooling to 100 K. These displacements give rise to zigzag chains of alternating short and long out-of-plane Pt(2)-Pt(2) distances. Phonon dispersions calculated from the refined structural models reveal Pt-dominated soft phonon modes in the region of reciprocal space associated with the experimentally observed CDW modulation, providing complementary evidence for an underlying lattice instability associated with the structural distortions identified in the present work.

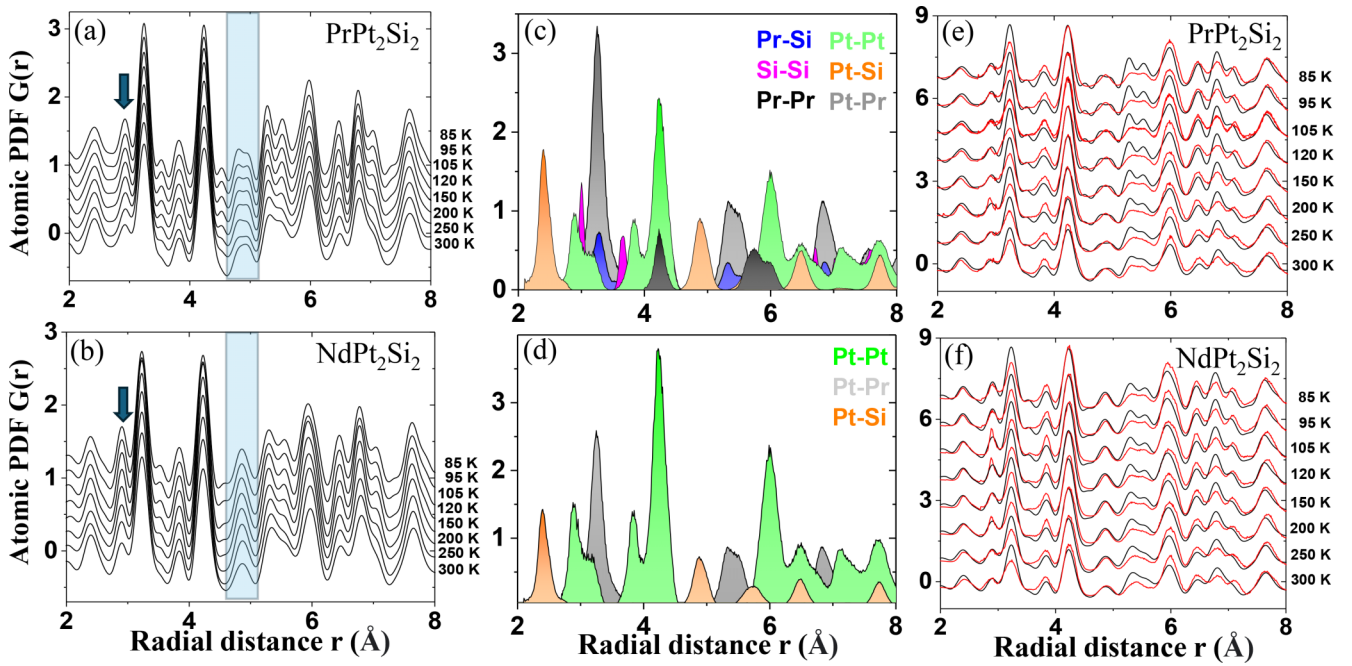


FIG. 2. (a), (b) Total atomic PDFs for $R\text{Pt}_2\text{Si}_2$ ($R = \text{Pr}, \text{Nd}$) obtained at different temperatures. PDF peaks become sharper with decreasing temperature due to diminished thermal disorder (e.g., see the behavior of the PDF peaks under the vertical arrow). Light blue rectangles highlight systematic changes in the PDFs with temperature. They also highlight the presence of a non-negligible difference between the atomic arrangements in the two materials [compare the shape of the PDF peak at 5 Å in the data sets in (a) and (b)]. (c) Partial atomic PDFs contributing to the total PDF for PrPt_2Si_2 . (d) Partial atomic PDFs contributing to the Pt-differential PDF for PrPt_2Si_2 . As shown, Pt-involving correlations (Pt-Pt, Pt-Pr and Pt-Si correlations) heavily overlap with correlations not involving Pt (Pr-Pr, Pr-Si and Si-Si correlations) in the total PDF, which is not the case with the Pt-differential PDFs where the latter are absent. (e), (f) Comparison between total (black) and Pt-differential (red) PDFs for $R\text{Pt}_2\text{Si}_2$ ($R = \text{Pr}, \text{Nd}$) obtained at different temperatures.

II. RESULTS AND DISCUSSION

A. Sample preparation

Polycrystalline NdPt_2Si_2 and PrPt_2Si_2 samples were synthesized by arc melting under an argon atmosphere. The arc-melted samples were further annealed at 900 °C for 10 days in evacuated quartz tubes. The temperature dependence of the inverse magnetic susceptibility, χ^{-1} , measured in an applied field of 10 kOe using a Physical Property Measurement System is shown in Figs. 1(c) and 1(d). The results are consistent with previous reports, confirming the absence of magnetic ordering down to 3 K [36,46]. For both materials, however, the temperature dependence of χ^{-1} exhibits subtle deviations from simple Curie-Weiss behavior. In PrPt_2Si_2 , such deviations occur near 95 and 30 K, coinciding with temperatures at which anomalies are also observed in the electronic transport properties. In NdPt_2Si_2 , χ^{-1} displays similar deviations near 230, 110, and 50 K, corresponding to temperatures at which anomalies have likewise been reported in the electronic and thermal transport properties [34–36].

B. Total x-ray scattering experiments

CDW superstructures are typically determined by single-crystal diffraction through the observation and analysis of weak satellite reflections offset from Bragg peaks by the CDW modulation vector q_{CDW} . Careful measurement of their positions and intensities enables refinement of the vector q_{CDW} , CDW symmetry, correlation length, and lattice distortion amplitude [2,47,48]. However, when satellite peaks are poorly

resolved or overlap due to short-range CDW fluctuations or the coexistence of multiple CDWs—as appears to be the case with NdPt_2Si_2 and PrPt_2Si_2 —the CDW superstructure, particularly the nature and amplitude of the lattice distortions, becomes difficult to determine. In such cases, atomic pair distribution function (PDF) analysis from total x-ray scattering provides a useful alternative. Unlike single-crystal diffraction, which largely relies on weak satellite intensities, total scattering utilizes the full signal, including Bragg, satellite, and diffuse contributions, all sensitive to lattice distortions [49]. The resulting PDFs, analyzed in real space, directly reflect interatomic distances and are therefore highly sensitive to lattice distortions across both short and long length scales, irrespective of whether these distances are periodic or not.

Total x-ray scattering experiments were carried out at the 1-ID-E beamline [50] at the Advanced Photon Source, Argonne National Laboratory, using a Pilatus3X CdTe 2M detector. An Oxford 800 series cryostream device was used to control the temperature. Data were taken at several temperatures in the range from 300 to 85 K. Representative experimental x-ray diffraction (XRD) patterns collected using x-rays with energy of 77.870 keV, which is 525 eV below the K edge of Pt are shown in Figs. 1(e) and 1(f). Note that the use of higher-energy x-rays is necessary to obtain atomic pair distribution functions (PDFs) with high real-space resolution [51]. The patterns are well fit with a structure model with S.G. $P4/nmm$ tetragonal symmetry, confirming that the samples are single phase. Total PDFs derived from the patterns using standard procedures [52] are shown in Figs. 2(a) and 2(b). As

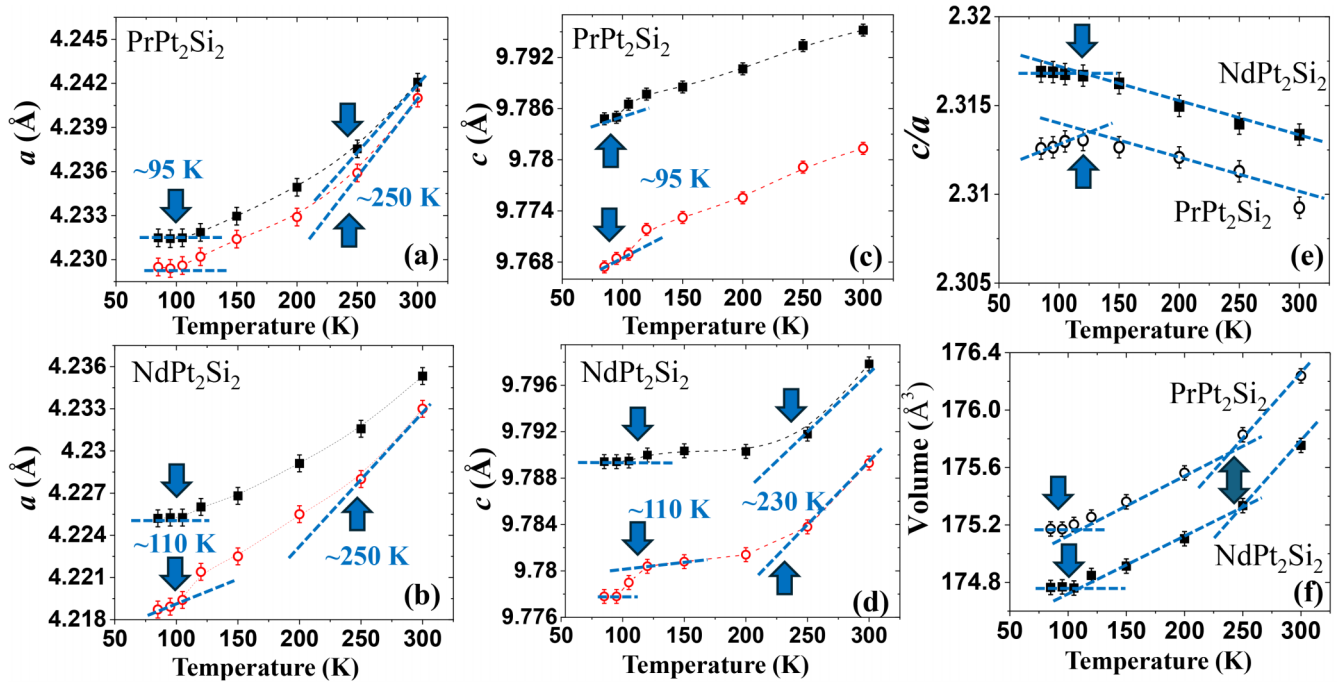


FIG. 3. (a)–(d) Temperature evolution of the a and c lattice parameters for $R\text{Pt}_2\text{Si}_2$ ($R = \text{Pr}, \text{Nd}$) obtained by Rietveld (black symbols) fits based on a perfect tetragonal structure. Also shown are results from single-unit-cell PDF (red symbols) fits where, contrary to the case of the Rietveld fits, Pt(1) atoms have been allowed to be displaced from their high-symmetry positions. Note that, as found in many CDW materials, Rietveld and PDF refined structure parameters differ significantly due to the presence of local lattice distortions. Temperature evolution of (e) the ratio of the tetragonal lattice parameters (c/a) and (f) the unit cell volume derived from the lattice parameters shown in (a)–(d). All data sets show marked deviations from linearity (see blue arrows), as indicated by changes in the slope of the dashed blue lines used as a guide to the eye.

expected, PDF peaks are seen to become increasingly sharper with decreasing temperature due to reduced thermal atomic vibrations. Also, some low- r peaks in the PDFs for NdPt_2Si_2 and PrPt_2Si_2 are seen to differ in fine details, which shows that the local atomic structure of the materials is not exactly the same.

As obtained, however, each total PDFs is a weighed sum of six distinct pair correlations between R, Pt, and Si atoms in the studied materials (see Table S1), that is, R-R, R-Pt, R-Si, Pt-Pt, Pt-Si, and Si-Si pair correlations [49,51]. The correlations overlap considerably [Fig. 2(c)], making it difficult to interpret the experimental total PDF data unambiguously. To obtain structure data with increased sensitivity to atomic correlations involving Pt atoms, which are suggested to be involved in the investigated lattice distortions [38], for each temperature, we collected an additional data set using x-rays with energy of 78.370 keV that is 25 eV below the K edge of Pt. From the difference between data sets collected 525 and 25 eV below that edge (Fig. S1), so-called Pt-differential atomic PDFs were derived, following a protocol described in Refs. [53–55]. More detailed experimental procedures are described in the Supplemental Material [56]. As obtained, Pt-differential PDFs reflect only correlations involving Pt atoms, that is, Pt-Pt, Pt-R, and Pt-Si atoms [Fig. 2(d)], which renders them significantly different from the respective total PDFs [Figs. 2(e) and 2(f)]. Hence, they provide an additional experimental constraint for the large-scale structure modeling described below.

C. Temperature evolution of the average crystal structure by Rietveld refinement

The temperature evolution of the average crystal structure for NdPt_2Si_2 and PrPt_2Si_2 was assessed by Rietveld analysis, where XRD data sets were fit with a S.G. $P4/nmm$ structure model using GSAS II software [57]. The successful fits [see Figs. 1(e) and 1(f)] yielded the tetragonal lattice parameters a and c and the corresponding unit cell volume shown in Fig. 3, in good agreement with previous studies [26,36]. Representative fit results are given in Tables S2 and S3. The parameters show a markedly nonlinear evolution with temperature. To investigate the lattice instability in NdPt_2Si_2 and PrPt_2Si_2 associated with the emergence of CDW order, as evidenced by the appearance of superlattice reflections [34,36], we determined the linear and volumetric thermal expansion coefficients from the experimentally determined lattice parameters. These quantities provide a sensitive probe of lattice dynamics because, within the quasiharmonic approximation, they are governed by the phonon spectrum through the mode Grüneisen parameters. A lattice instability associated with the softening of one or more phonon modes, such as that reported for LaPt_2Si_2 [58], can produce pronounced anomalies in the thermal expansion coefficients. The softening of specific phonon modes also determines the atomic displacement pattern of the resulting CDW superstructure. The results are presented in Fig. 4. Both compounds exhibit strong nonlinearities in the linear and volumetric thermal expansion coefficients, consistent with anomalous lattice dynamics and

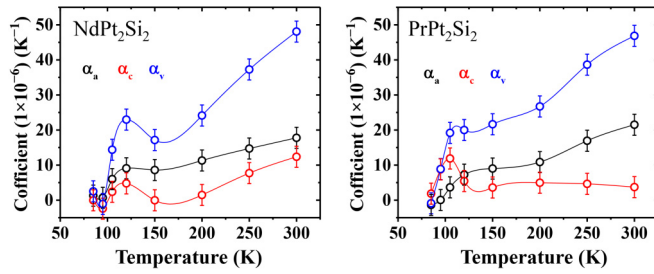


FIG. 4. Thermal lattice expansion coefficients along the a , α_a , and c , α_c , crystallographic directions in NdPt_2Si_2 and PrPt_2Si_2 . The corresponding volumetric thermal expansion coefficients, α_v , is also given. Both materials exhibit pronounced nonlinearities in their thermal expansion, consistent with anomalous lattice dynamics and an incipient lattice instability.

an incipient lattice instability. Furthermore, the pronounced anisotropy of the thermal expansion (see the nonlinearity in the c/a ratio) indicates that these anomalies are strongly direction dependent, suggesting that the lattice instability involves atomic displacement patterns that give rise to superstructures propagating along different crystallographic directions [59–63].

D. Temperature evolution of the local crystal structure by single-unit-cell PDF modeling

Previous studies have likewise related the nonlinear temperature dependence of the lattice parameters to the emergence of CDW order. However, structural analyses based solely on Bragg diffraction failed to reveal the underlying lattice distortions. To investigate these distortions, we fitted the experimental total PDFs with the S.G. $P4/nmm$

structural model using the software PDFGUI [64]. Unlike conventional Rietveld refinement, PDF analysis incorporates both the Bragg and diffuse scattering components of the diffraction data, making it sensitive to both the average crystal structure and local deviations from it. The refinements were performed over a real-space range of 2–20 Å to obtain reliable structural parameters. This fitting range enables a uniform description of lattice distortions that are local at room temperature but become progressively more correlated upon cooling, eventually giving rise to a superstructure spanning several crystallographic unit cells. Representative PDF fits are shown in Figs. 5(a) and 5(b). The fits perform poorly over the whole fitting range, with R_{wp} agreement factor $\approx 36\%$ [Figs. S2(a) and S2(b)], and fail to reproduce short-range interatomic correlations, particularly those contributing to the PDF peak at approximately 3 Å [arrows in Figs. 5(a) and 5(b)]. Comparison of the total and Pt-differential PDFs in Fig. 6, together with the partial PDF data shown in Figs. 2(c) and 2(d), indicates that this peak is dominated by Pt-related atomic correlations. This suggests that the poor fit arises primarily from an inadequate description of the Pt sublattice. Guided by these observations and by our previous study of LaPt_2Si_2 [38], we allowed both Pt(1) atoms in the tetragonal unit cell (Table S2) to independently displace from their high-symmetry positions. This modification substantially improved the description of the 3 Å PDF peak [Figs. 5(c) and 5(f)] and reduced the overall agreement factor to $R_{\text{wp}} \sim 28\%$ [Figs. S2(c)–S2(f)]. This result indicates that Pt atoms occupy locally displaced positions already at room temperature, rather than merely exhibiting thermal broadening about their high-symmetry crystallographic positions. The refined tetragonal lattice parameters a and c , together with the corresponding unit-cell volume, are shown in Fig. 3. Their temperature

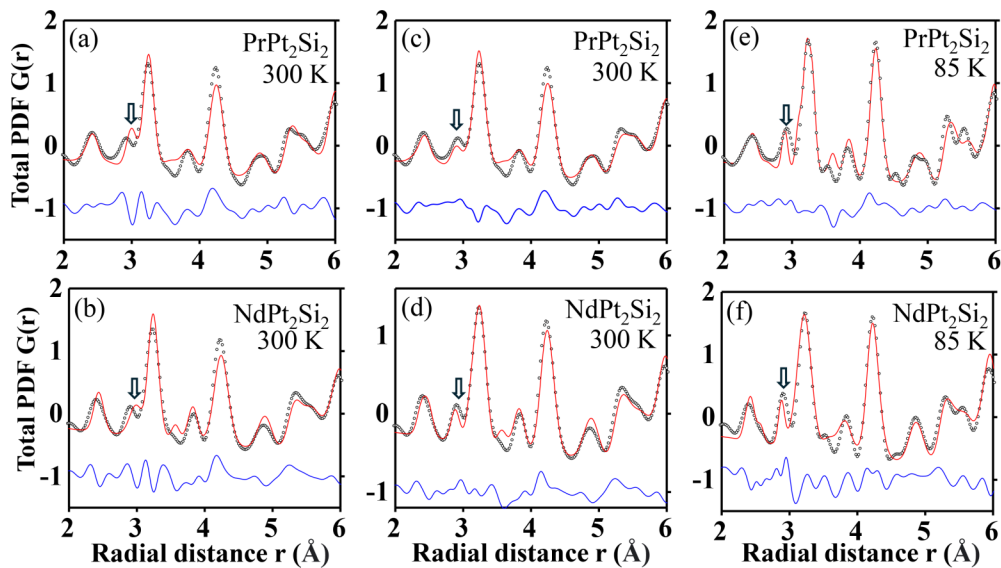


FIG. 5. (a), (b) Single-unit cell fits (red) to total PDFs (symbols) for RPt_2Si_2 ($R = \text{Pr}, \text{Nd}$) based on a perfect tetragonal structure. The residual difference (shifted for clarity) is shown in blue. Arrows point to a PDF peak (interatomic distances) appearing at about 3 Å which is not reproduced by the model. (c)–(f) Single-unit cell fits (red) to total PDFs (symbols) for RPt_2Si_2 ($R = \text{Pr}, \text{Nd}$) based on a tetragonal structure in which Pt(1) are displaced from their high-symmetry positions as discussed in the text. The residual difference (shifted for clarity) is shown in blue. The model reproduces the 3 Å PDF peak reasonably well. The data sets in (a)–(d) are obtained at 300 K while those in (e), (f) are obtained at 85 K. The goodness-of-fit factor for the fits in (a), (b) is about 36%. It is about 28% for the fits in (c)–(f).

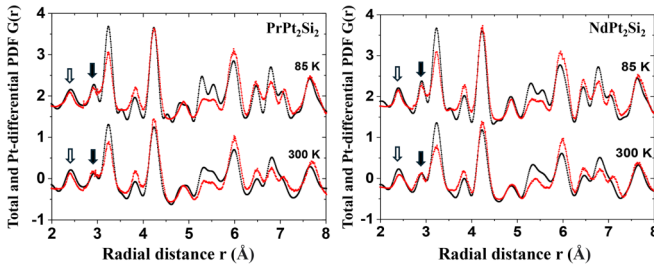


FIG. 6. Experimental total (black) and Pt-differential (red) PDFs for PrPt_2Si_2 and NdPt_2Si_2 . The close similarity between the first two peaks in the total and Pt-differential PDFs (shaded ellipses) indicates that these features are dominated by pair correlations involving Pt atoms, namely Pt-Pt, Pt-R, and Pt-Si correlations, which contribute to both PDFs. In particular, the peak at approximately 2.4 Å (white arrow) arises exclusively from nearest-neighbor Pt-Si pair correlations and exhibits little temperature dependence. The peak at approximately 3 Å (black arrow) is dominated by nearest-neighbor Pt-Pt pair correlations [see Figs. 2(c) and 2(d)]. At higher r , the total and Pt-differential PDFs diverge because the total PDF, unlike the Pt-differential PDF, also contains contributions from R-R, R-Si, and Si-Si pair correlations.

dependence is strongly nonlinear, particularly near 250 and 110 K. The nonlinear behavior is more pronounced than that obtained from the Rietveld-refined lattice parameters, indicating that the local crystal structure is more strongly distorted than the average crystallographic structure. Despite this improvement, however, the overall fit quality remains unsatisfactory, prompting us to investigate in greater detail how the complex lattice distortions in NdPt_2Si_2 and PrPt_2Si_2 evolve with temperature.

E. Temperature evolution of the local crystal structure from large-scale modeling

For this purpose, we refined large-scale structural models against the experimental total and Pt-differential PDFs using the reverse Monte Carlo (RMC) technique [14,45,65–67]. The computations were done with the help of the program full-rmc [68]. Initial 40 000-atom models ($85 \times 85 \times 100$ Å) were generated using the lattice parameters and atomic coordinates obtained from the PDF refinements that allowed the two Pt(1) atoms to independently displace from their high-symmetry positions [Figs. 5(c)–5(f) and S2(c)–S2(f)]. This provided a physically meaningful starting configuration and an overall structural constraint for the RMC refinements. The large model size allowed us to characterize lattice distortions that are local at high temperatures and become progressively correlated over longer distances upon cooling, extending across several crystallographic unit cells. It also improved the statistical reliability of the refined structural parameters. During the RMC refinements, all atoms were initially displaced with a maximum step size of 0.02 Å, which was reduced to 0.01 Å in the final stages of the refinement. Atomic displacements were not constrained to specific crystallographic directions; however, minimum interatomic distances were imposed to prevent atoms from approaching closer than the sum of their atomic radii in the corresponding elemental materials. The refinements were terminated when no further improvement in

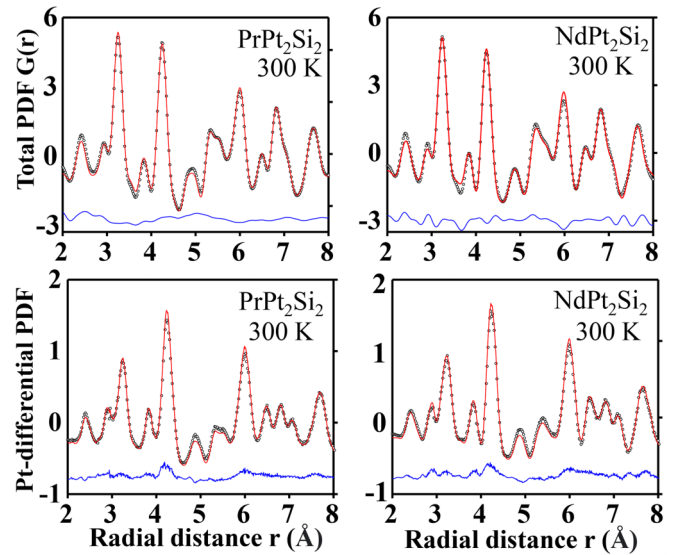


FIG. 7. (upper row) RMC fits (red) to total PDFs (symbols) for RPt_2Si_2 ($R = \text{Pr}, \text{Nd}$). (lower row) RMC fits (red) to Pt-differential PDFs (symbols) for RPt_2Si_2 ($R = \text{Pr}, \text{Nd}$). The residual difference (shifted for clarity) is shown in blue. The goodness-of-fit factor for all data sets is about 8%. The PDFs are obtained at 300 K.

the agreement with the experimental total and Pt-differential PDFs could be achieved.

To assess the robustness and reproducibility of the refinements, the RMC simulations were repeated by varying (i) the fitting ranges, (ii) the number of RMC steps, (iii) the relative weights assigned to the total and Pt-differential PDFs, (iv) the model sizes (including $60 \times 60 \times 60$ Å and $75 \times 75 \times 75$ Å models), and (v) the initial amplitudes of the Pt(1) and Pt(2) displacements from their high-symmetry positions. Several independent RMC refinements of the 40 000-atom configurations were performed using different combinations of these modeling parameters. The resulting independently converged configurations produced statistically equivalent fits to the experimental data and yielded consistent pair-distance distributions, coordination environments, bond-angle distributions, and atomic-displacement correlations. Three representative independently converged configurations were selected as the final structural models, and the reported structural parameters were obtained by averaging the corresponding quantities extracted from these configurations. Although individual RMC configurations are not expected to be mathematically unique, the reproducibility of these structural features across independent refinements indicates that they are robust and well constrained within the resolution of the experimental data and the imposed modeling constraints [69–71]. All PDF datasets collected in this work were analyzed using this procedure. Representative RMC fits to the experimental total and Pt-differential PDFs at 300, 150, and 85 K are shown in Figs. 7–9. The agreement with experiment is substantially improved compared with that obtained using the single-unit-cell model [Figs. 5(c)–5(f)]. Projections of the RMC-refined atomic configurations onto selected crystallographic planes, obtained without imposing crystallographic symmetry or

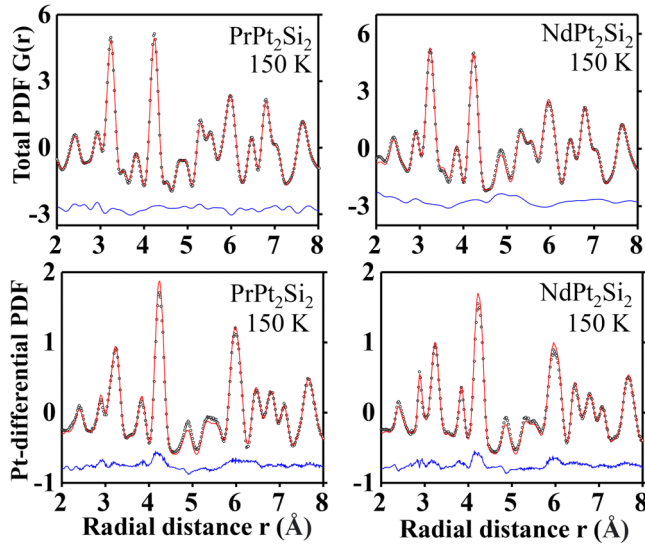


FIG. 8. (upper row) RMC fits (red) to total PDFs (symbols) for $R\text{Pt}_2\text{Si}_2$ ($R = \text{Pr}, \text{Nd}$). (lower row) RMC fits (red) to Pt-differential PDFs (symbols) for $R\text{Pt}_2\text{Si}_2$ ($R = \text{Pr}, \text{Nd}$). The residual difference (shifted for clarity) is shown in blue. The goodness-of-fit factor for all data sets is about 8%. The PDFs are obtained at 150 K.

superstructure periodicity, are presented in Figs. 10 and 11. Each black circle represents the geometric center of the projected positions of the corresponding 4000 Pt atoms within the 40 000-atom model and may therefore be interpreted as the most probable position of the Pt(1) or Pt(2) atom in the crystal lattice. The corresponding electron-density distributions in the same planes, calculated from the refined RMC models using the independent atom model (IAM) [72] are also shown.

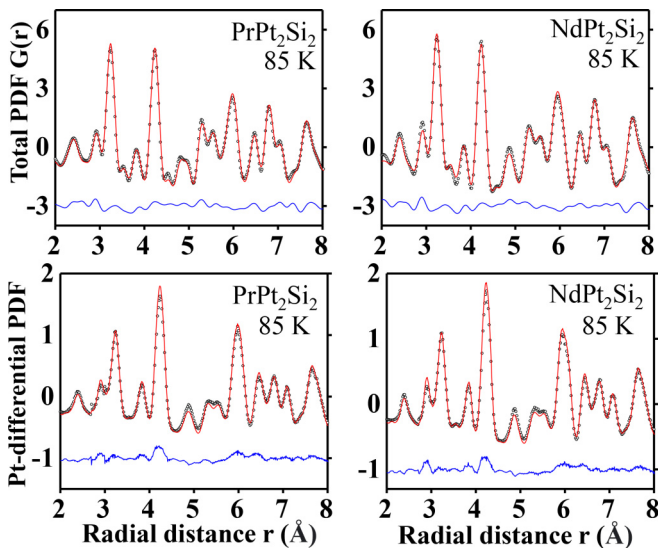


FIG. 9. (upper row) RMC fits (red) to total PDFs (symbols) for $R\text{Pt}_2\text{Si}_2$ ($R = \text{Pr}, \text{Nd}$). (lower row) RMC fits (red) to Pt-differential PDFs (symbols) for $R\text{Pt}_2\text{Si}_2$ ($R = \text{Pr}, \text{Nd}$). The residual difference (shifted for clarity) is shown in blue. The goodness-of-fit factor for all data sets is about 8%. The PDFs are obtained at 85 K.

As can be seen in Fig. 10, already at room temperature, Pt atoms in the Si-Pt(1)-Si planes in PrPt_2Si_2 are significantly displaced from their high-symmetry positions, leading to the emergence of two near-neighbor Pt(1)-Pt(1) distances of about 2.89(1) and 3.13(1) Å. In a hypothetical undistorted lattice, a single in-plane Pt(1)-Pt(1) distance of about 3.02(1) Å would be observed [Fig. 1(b)]. By contrast, Pt atoms in Pt(2)-Si(2)-Pt(2) layers are slightly displaced from their high-symmetry position and appear 4.25(1) Å apart. As can be seen in Fig. 11, the situation with NdPt_2Si_2 is similar. Here, at 300 K, the two near-neighbor of Pt(1)-Pt(1) distances are 2.89(1) and 3.14(1) Å while the unique Pt(2)-Pt(2) distance is 4.24(1) Å. As may be expected, in both PrPt_2Si_2 and NdPt_2Si_2 , in plane Pt(1)-Pt(1) and Pt(2)-Pt(2) near-neighbor distances shrink a bit (~ 0.01 Å) when the temperature is reduced from 300 to 150 K. In addition, Pt(2) atoms appear slightly more displaced from their high symmetry positions, but in-plane Pt(2)-Pt(2) distances remain uniform and close to 4.23(1) Å. When the temperature is further reduced to 85 K, Pt(1) atoms in the Si(1)-Pt(1)-Si(1) layers in PrPt_2Si_2 appear further displaced from their high-symmetry positions, leading to the emergence of four sets of in-plane Pt(1)-Pt(1) distances at about 2.83(1), 2.93(1), 3.06(1), and 3.15(1) Å. Atoms in Pt(2) planes are also further displaced from their positions, but the in-plane distances between them appear with the same length of about 4.23(1) Å. However, two sets of alternating out-of-plane Pt(2)-Pt(2) distances between adjacent Pt(2) planes are observed at approximately 3.80(1) and 3.87(1) Å (Fig. 12). In a hypothetical undistorted lattice, these distances would instead be represented by a single Pt(2)-Pt(2) distance of approximately 3.83(1) Å. A similar pattern is observed in NdPt_2Si_2 . Here, the four in-plane Pt(1)-Pt(1) distances within the Pt(1) planes are approximately 2.85(1), 2.91(1), 3.08(1), and 3.14(1) Å, whereas the in-plane Pt(2)-Pt(2) distances remain essentially uniform at approximately 4.22(1) Å. The out-of-plane Pt(2)-Pt(2) distances again split into two alternating sets, at approximately 3.80(1) and 3.86(1) Å.

The structural data obtained in this work, together with our previous studies of $R\text{Pt}_2\text{Si}_2$ compounds [38,43], suggest the following picture. Already at room temperature, i.e., prior to the onset of CDW order, the Pt(1) atoms in both PrPt_2Si_2 and NdPt_2Si_2 are significantly displaced from their high-symmetry positions in the tetragonal lattice. The Pt(2) atoms are also displaced, although to a considerably smaller extent. While these locally static displacements exhibit short-range correlations, they do not form a long-range periodic pattern, consistent with the absence or very weak intensity of superlattice reflections in this temperature range. In this sense, they may be viewed as precursors to the long-range lattice distortions that develop upon entering the CDW phase through a second order, predominantly order-disorder transition. The magnitude of the distortions (Fig. 13) changes relatively little until the temperature decreases to approximately 100 K, below which it increases abruptly as the Pt atoms undergo additional displacements. The electronic properties evolve accordingly, suggesting that the first-order structural phase transition at this temperature possesses a significant displacive component. As a result, the number of distinct Pt(1)-Pt(1) and Pt(2)-Pt(2) nearest-neighbor distances and coordination environments increases relative to the room-temperature structure

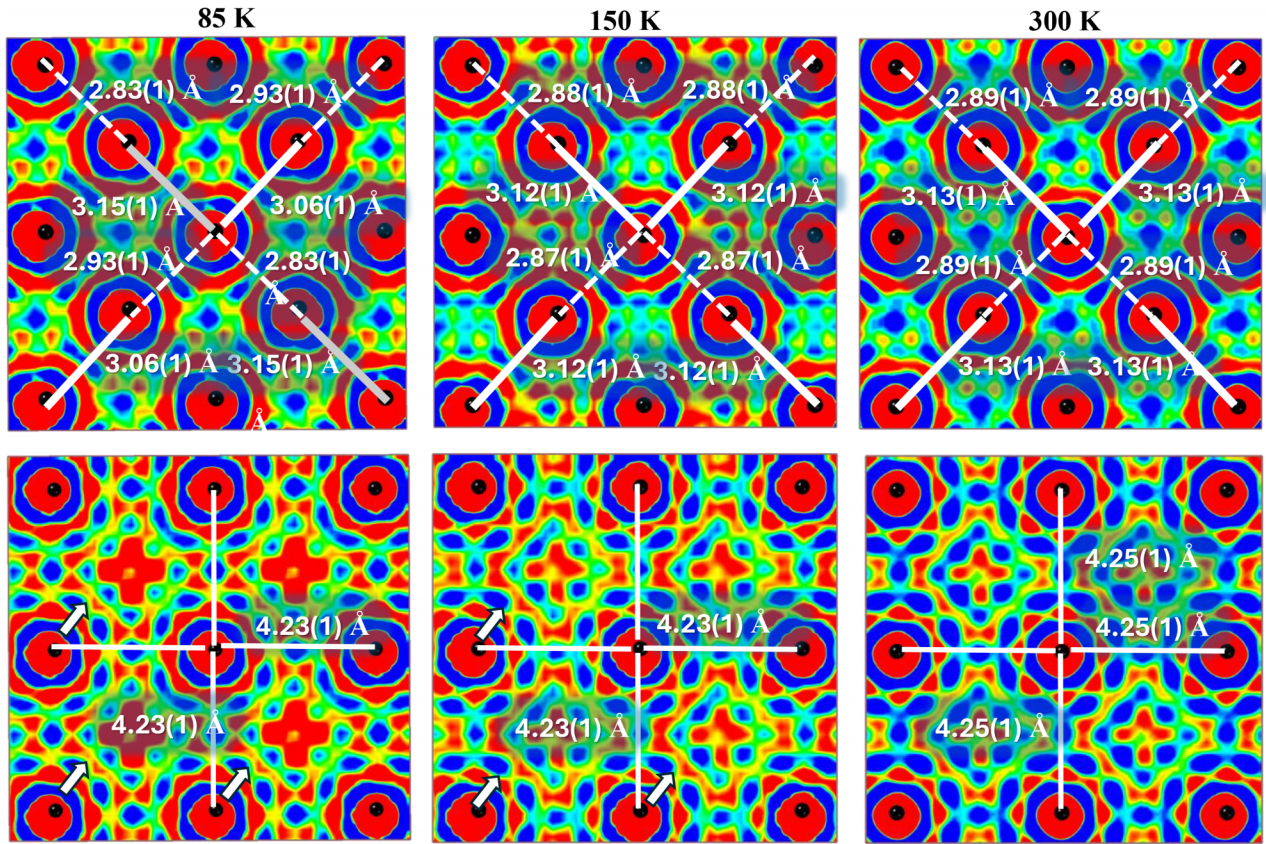


FIG. 10. (upper panel) Distribution of electron density in Pt(1) atomic planes in PrPt_2Si_2 , as derived from RMC refined structure models, at 300 and 150 K (both above the reported first-order transition at 95 K) and 85 K (below the first-order transition). Short (broken lines) and long (solid line) distances between Pt(1) atoms in the planes are also shown. The distances are seen to change with decreasing temperature due to increased displacement of Pt(1) from their high-symmetry positions. (lower panel) Distribution of electron density in Pt(2) atomic planes in PrPt_2Si_2 at the same temperatures. Although the Pt(2) atoms move away from their equilibrium positions with decreasing temperature (see the arrows), in-plane Pt(2)-Pt(2) distances remain unique. As expected, changes in the distances between near neighbor Pt atoms are accompanied by a marked redistribution of the electron density. The electron density increases linearly as the color changes from blue to red. Each atomic position (black circles) shown in panels represents the geometric center of the projected positions of 4000 corresponding Pt atoms in the 40 000-atom RMC-refined models. No crystallographic symmetry constraints are imposed on the atomic positions.

(Fig. S3). This progressive enhancement of the local lattice distortions coincides with the observed nonlinear evolution of the electronic transport properties over this temperature range, suggesting a close connection between the structural and electronic responses. Such a picture is further supported by the electron-density distributions shown in Figs. 10 and 11, which evolve in concert with the refined atomic displacements.

F. Phonon dispersion in NdPt_2Si_2 and PrPt_2Si_2 from DFT calculations based on PDF-derived structure data

To investigate the lattice dynamics of PrPt_2Si_2 and NdPt_2Si_2 , we constructed structural models for the DFT calculations directly from the experimentally constrained RMC configurations. The corresponding 40 000-atom RMC configurations were folded into single crystallographic unit cells of the same type as those used to generate the initial atomic models, without imposing additional crystallographic symmetry constraints. The atomic positions in the resulting unit cells were defined by the geometric centers of the projected positions of equivalent atoms in the RMC configurations. This procedure preserves the experimentally determined lattice

parameters refined using PDFGUI, as well as the tetragonal translation symmetry and the number and types of atoms in the unit cell (two rare-earth, four Pt, and four Si atoms), while incorporating the lattice distortions identified in Figs. 10–12. In particular, the resulting structures reflect the displacements of Pt(1) and Pt(2) from their high-symmetry (2a) and (2c) Wyckoff positions (Tables S2 and S3), respectively. As an example, the crystallographic unit cell of PrPt_2Si_2 , in which the Pt atoms occupy their high-symmetry positions, and the corresponding RMC-derived unit cell, which incorporates the Pt (1) and Pt(2) atom displacements, are presented in Tables S4 and S5, respectively. To validate the RMC-derived unit cells against the experimental data, we calculated their total PDFs and compared them with the measured PDFs. As shown in Fig. 14, the calculated PDFs reproduce the experimental data in good detail, yielding an overall agreement factor of $R_{\text{wp}} \sim 21\%$. These tetragonal unit cells, derived from the experimentally constrained RMC models and validated against the measured PDFs, were subsequently used as the structural basis for the phonon calculations.

Phonon dispersion relations were calculated using the finite-displacement method implemented in the PHONOPY

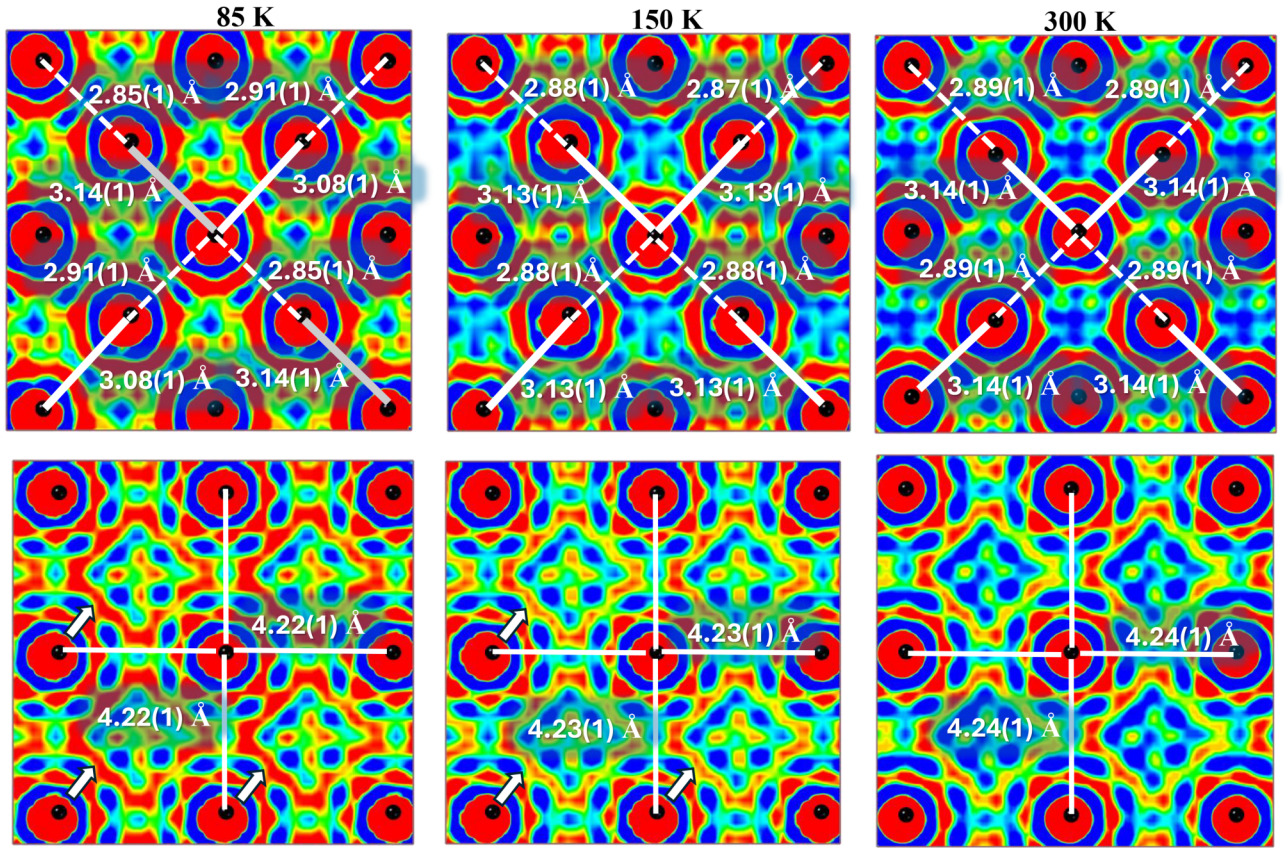


FIG. 11. (upper panel) Distribution of electron density in Pt(1) atomic planes in NdPt_2Si_2 , as derived from RMC refined structure models, at 300 K (above the inflection point in $\chi^{-1}(T)$ at 250 K), 150 K (between the inflection points in $\chi^{-1}(T)$ at 250 and 110 K), and 85 K (below the inflection point in $\chi^{-1}(T)$ at 110 K) [see Figs. 1(c) and 1(d)]. The three temperatures are above the reported first-order transition of 50 K. Short (broken lines) and long (solid line) distances between Pt(1) atoms in the planes are also shown. The distances are seen to change with decreasing temperature due to increased displacement of Pt(1) from their high-symmetry positions. (lower panel) Distribution of electron density in Pt(2) atomic planes in NdPt_2Si_2 at the same temperatures. Although the Pt(2) atoms move away from their equilibrium positions with decreasing temperature (see the arrows), in-plane Pt(2)-Pt(2) distances remain unique. As expected, changes in the distances between near neighbor Pt atoms are accompanied by a marked redistribution of the electron density. The electron density increases linearly as the color changes from blue to red. Each atomic position (black circles) shown in panels represents the geometric center of the projected positions of 4000 corresponding Pt atoms in the 40 000-atom RMC-refined models. No crystallographic symmetry constraints are imposed on the atomic positions.

package [73] in conjunction with VASP. The RMC-derived atomic coordinates were used directly as input, without further structural relaxation. This approach preserves the experimentally observed local lattice distortions and enables their influence on the lattice dynamics to be evaluated explicitly. In contrast, full DFT relaxation drives the system toward a

theoretical ground-state structure, eliminating the experimentally observed distortions within the Pt planes (Fig. S4) and producing dynamically stable structures with no imaginary phonon frequencies (Fig. S5). The phonon dispersions presented here therefore describe the lattice dynamics of the experimentally observed distorted structures rather than those

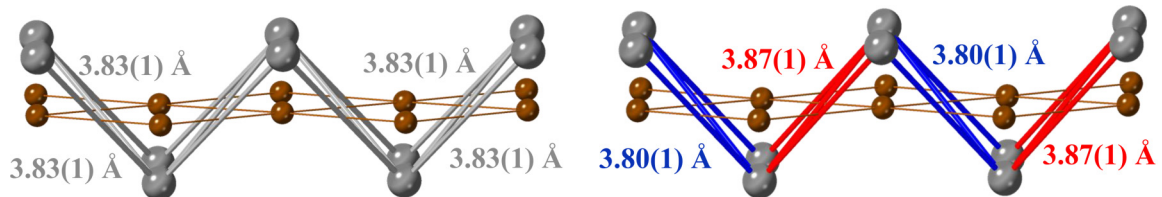


FIG. 12. (left) Out-of-plane distances between Pt(2) atoms (gray) in undistorted PrPt_2Si_2 . (right) Out-of-plane distances between Pt(2) atoms in the CDW phase of PrPt_2Si_2 (85 K), as derived from a RMC-refined model. Each atomic position represents the geometric center of the projected positions of 4000 corresponding Pt and Si atoms in the model. No crystallographic symmetry constraints were imposed during the derivation. The distribution of short (blue) and long (red) out-of-plane Pt(2)-Pt(2) distances in NdPt_2Si_2 is nearly identical to that observed in PrPt_2Si_2 . Si atoms are shown in brown.

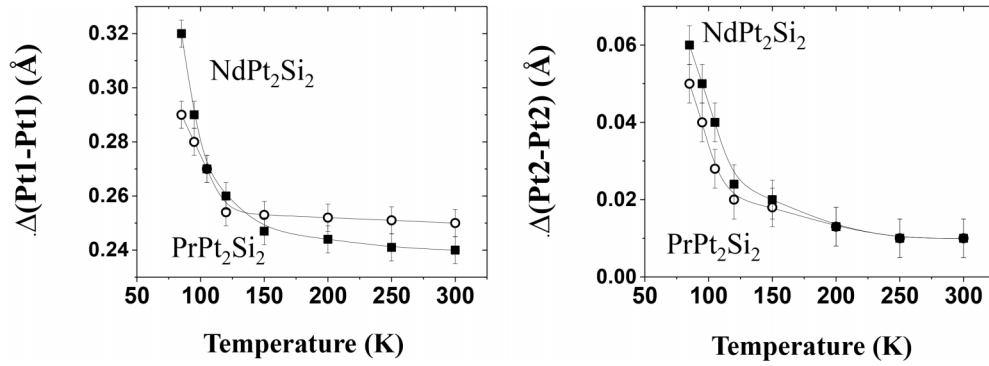


FIG. 13. (left) Difference, $\Delta(\text{Pt1-Pt1})$, between the longest and shortest nearest-neighbor Pt-Pt distances within the Pt(1) planes. (right) Difference, $\Delta(\text{Pt2-Pt2})$, between the longest and shortest out-of-plane Pt-Pt distances associated with the Pt(2) planes. The differences can serve as a measure of the degree of lattice distortions in NdPt_2Si_2 and PrPt_2Si_2 .

of the fully relaxed DFT ground state. The calculated phonon dispersions are shown in Fig. 15. A prominent feature is the presence of soft phonon instabilities (imaginary frequencies) along the Γ -X direction of the Brillouin zone. In NdPt_2Si_2 , the soft branch exhibits a shallow minimum near the CDW modulation wave vector, $q_{\text{CDW}} \sim (1/3, 0, 0)$, determined by single-crystal x-ray diffraction experiments [36,74]. In PrPt_2Si_2 , the softening is broader and involves two coupled soft branches; it begins at wave vectors somewhat below $q_{\text{CDW}} \sim (1/3, 0, 0)$ and extends through this region. Thus, although the detailed phonon dispersions differ between the two compounds, both exhibit lattice instabilities in the same region of reciprocal space as the experimentally observed

CDW modulation. A similarly broad soft-phonon anomaly has previously been reported for LaPt_2Si_2 [58,75].

The broad anomalies in the calculated phonon dispersions [76], together with the experimentally observed small resistivity anisotropy and the absence of a sharp metal-insulator transition, are consistent with lattice modulations involving multiple crystallographic directions. This picture agrees well with the strong in-plane modulation involving Pt(1) atoms and propagating within the *ab* plane (Figs. 10 and 11), as well as the weaker out-of-plane modulation involving Pt(2) atoms (Fig. 12) identified in the present work. Collectively, these findings support a close connection between the intrinsic lattice instability and the emergence and evolution of CDW order in NdPt_2Si_2 and PrPt_2Si_2 . In this picture, the locally static Pt displacements identified by the PDF/RMC analysis and the Pt-dominated soft lattice modes in the corresponding region of reciprocal space may be viewed as complementary structural and dynamical manifestations of the underlying lattice instability.

III. CONCLUSIONS

Our study suggests that charge-density-wave order in NdPt_2Si_2 and PrPt_2Si_2 develops within a lattice that is already quasistatically distorted through displacements of Pt atoms from their high-symmetry positions. Upon cooling, the initially local Pt displacement patterns become progressively correlated over longer distances, evolving toward superstructures associated with long-range CDW order. This temperature evolution provides a natural connection between the local distortions revealed by the PDF/RMC analysis and the weak, strongly temperature-dependent satellite reflections observed in previous single-crystal diffraction studies. In this picture, the evolution of the satellite reflections reflects the progressive development and reorganization of long-range correlations among Pt displacement patterns that are already present locally at higher temperatures.

Our results further suggest that the higher-temperature second-order transitions have a predominantly order-disorder character: Preexisting local lattice distortions become correlated over long-range distances rather than appearing only at the transition temperature. The emergence of satellite Bragg reflections at these transitions can therefore be understood

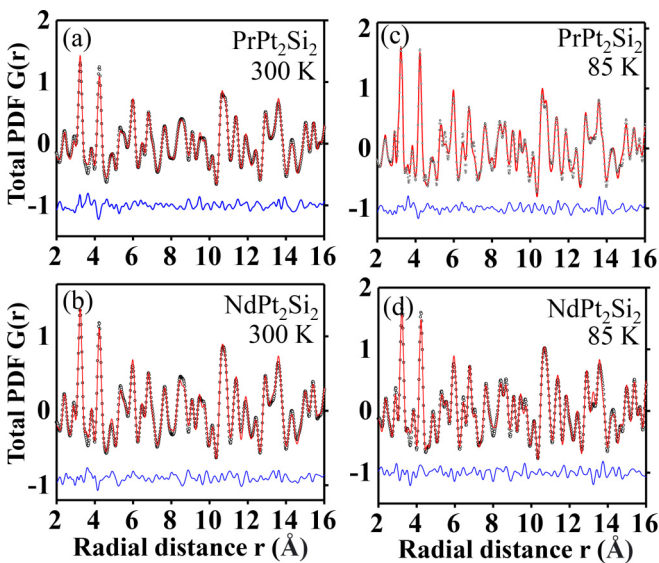


FIG. 14. (a)–(d) Single-unit-cell fits (red lines) to the experimental total PDFs (symbols) for RPt_2Si_2 ($R = \text{Pr}, \text{Nd}$), based on a tetragonal unit cell derived from the large-scale RMC models as described in the text. The residual differences (blue lines) are vertically offset for clarity. The goodness-of-fit factor is approximately 21%. The agreement is improved compared with the PDF fits shown in Figs. 5(c)–5(f) because the present structural models incorporate the unusual lattice modulations involving both the Pt(1) and Pt(2) planes, which could not be clearly resolved by the structural models used in Figs. 5(c)–5(f).

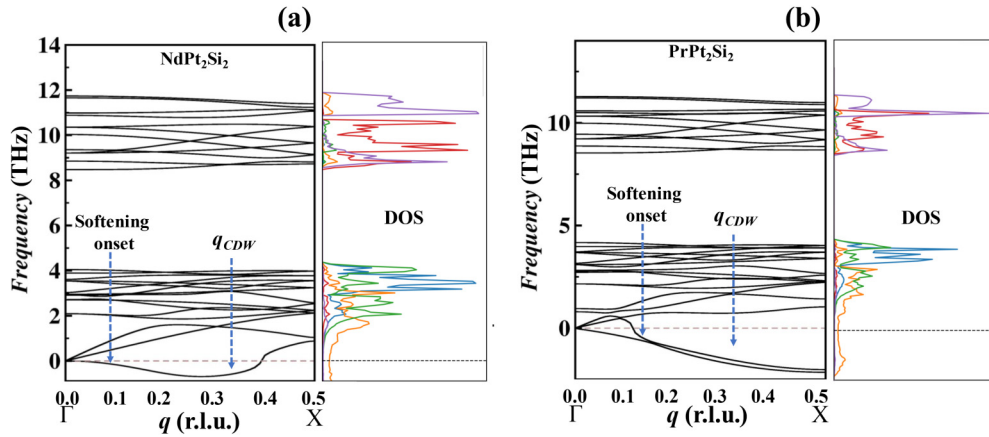


FIG. 15. Phonon dispersions for (a) NdPt_2Si_2 and (b) PrPt_2Si_2 , calculated using unit the experimentally constrained RMC models described in the text. Both compounds exhibit soft phonon instabilities (imaginary frequencies) along the Γ -X direction of the Brillouin zone. For NdPt_2Si_2 , the imaginary branch exhibits a shallow minimum near the experimentally observed CDW modulation wave vector, $q_{\text{CDW}} \sim (1/3, 0, 0)$, whereas PrPt_2Si_2 exhibits a broader softening extending through this region of reciprocal space. The contributions of Pt(1), Pt(2), RE (Pr/Nd), Si(1), and Si(2) atoms to the phonon dispersions are shown in orange, green, blue, red, and magenta, respectively. The imaginary-frequency modes are dominated by Pt displacements.

as marking the development of long-range CDW order from an already locally distorted lattice. At lower temperatures, the Pt-related distortions increase sharply near the first-order transitions, indicating a significant displacive component associated with further reorganization and strengthening of the preexisting CDW state. This picture provides a structural context for the previously reported phase transitions and the corresponding anomalies in the electronic and thermal transport properties of PrPt_2Si_2 and NdPt_2Si_2 .

The evolving Pt displacement patterns give rise to well-defined short and long Pt-Pt distances associated with structural modulations involving different crystallographic directions. A similar picture has been reported for the isostructural compound UPt_2Si_2 [38], where, in addition to the in-plane modulations involving Pt(1) atoms, U atoms are also displaced from their high-symmetry positions. The difference in the distortion patterns may, in part, be related to the difference in atomic size between U (~ 1.75 Å) and Pr/Nd (~ 1.85 Å). Our results are likewise consistent with recent studies of CDW order in the RENiC_2 (RE = Sm, Gd, Tb, Dy) family [14] and support a broader picture in which intrinsic lattice instabilities are closely connected to the development of complex electronic phases in materials exhibiting competing or coexisting ordered electronic states.

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DATA AVAILABILITY

High resolution x ray diffraction data for (Apt_2Si_2 , A = Nd, Pr) shown in Fig. 1 are available at [79]. Other data are available from the authors upon reasonable request.

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