

Probing the symmetries of nodal-line semimetal SrAl_4

Tomer Hasharoni¹, Danila Sokratov², Prathum Saraf², Johnpierre Paglione^{2,3}, and Alon Ron^{1,*}

¹*School of Physics and Astronomy, Tel Aviv University, Tel Aviv 69978, Israel*

²*Maryland Quantum Materials Center, Department of Physics, University of Maryland, College Park, Maryland 20742, USA*

³*Canadian Institute for Advanced Research, Toronto, Ontario M5G 1Z8, Canada*



(Received 27 April 2026; accepted 10 August 2026; published 5 October 2026)

Symmetry plays a central role in determining the electronic structure and topology of nodal-line semimetals. SrAl_4 is a nodal-line semimetal with tetragonal $I4/mmm$ symmetry at room temperature, an incommensurate charge-density-wave transition at $T_{\text{CDW}} \simeq 240$ K accompanied by C_4 symmetry breaking, and a low-temperature monoclinic transition near $T \simeq 90$ K. Here, we use nonlinear optical second- and third-harmonic generation to track the evolution of crystal symmetries in SrAl_4 with temperature and disorder. Temperature-dependent second-harmonic generation uncovers strong coupling between the lattice and the charge-density-wave order and shows that disorder quenches the low-temperature monoclinic phase. Room-temperature third-harmonic generation reveals broken C_4 rotational symmetry in contrast to the reported tetragonal bulk structure, possibly due to a symmetry-lowered surface reconstruction. These results establish nonlinear harmonic generation as a sensitive probe of intertwined bulk and surface symmetry breaking in SrAl_4 and as a complementary tool for refining symmetries where space group assignment through diffraction is ambiguous.

DOI: [10.1103/78p9-rjxz](https://doi.org/10.1103/78p9-rjxz)

I. INTRODUCTION

The interplay between lattice symmetry and electronic phenomena can give rise to unusual and exotic physical behavior [1–5]. Understanding the relationship between crystal structure, symmetry, and electronic phases is therefore essential for identifying the microscopic mechanisms that govern their emergence. In particular, the electronic properties of nodal-line semimetals, including the protection of their band degeneracies, depend sensitively on lattice symmetry and on the presence of mirror planes [6,7]. Charge-density-wave (CDW) order, characterized by a spatial modulation of the conduction-electron density, often occurs together with a periodic lattice distortion and the associated breaking of crystal symmetries. It is a ubiquitous electronic phase found across a wide range of material families, where it manifests in distinct yet equally intriguing forms [8–13]. CDW formation has traditionally been understood in terms of Fermi-surface nesting, namely, a Peierls-type instability, although more recent work has shown that in some systems electron-phonon coupling may instead play the dominant role [14–16]. These alternative viewpoints, together with the growing recent interest in topological materials, have renewed attention to CDW systems from a broader and qualitatively different perspective. The MAl_4 ($\text{M} = \text{Sr}, \text{Ba}, \text{Ca}, \text{Eu}$) compounds, including

SrAl_4 , belong to an isostructural family crystallizing in the tetragonal BaAl_4 -type structure. Despite their structural similarity, members of this family exhibit markedly different physical behavior under external perturbations. For example, CaAl_4 undergoes a structural transition from tetragonal ($I4/mmm$) to monoclinic ($C2/m$) upon cooling below $T = 443$ K [18,19], whereas the topological semimetal BaAl_4 [20] remains in the $I4/mmm$ structure down to low temperatures without undergoing any transition (Sec. I in the Supplemental Material [21]). In the magnetically ordered sister compound EuAl_4 , x-ray diffraction (XRD) and second-harmonic generation (SHG) measurements were used to identify two distortion patterns associated with C_2 nematic and C_4 chiral CDW states through their distinct symmetry-breaking signatures [22]. More broadly, a range of electronic and magnetic properties have been investigated across the MAl_4 family, including electrical resistivity and de Haas–van Alphen oscillations [23], revealing how these quantities evolve across their different structural phases.

SrAl_4 , a tetragonal $I4/mmm$ nodal-line semimetal [14], exhibits a rich temperature- and pressure-dependent phase diagram [24,25]. Figure 1(a) summarizes the evolution of the crystal structure of SrAl_4 with temperature and the key symmetries associated with each phase. Below $T_{\text{CDW}} \simeq 240$ K, an incommensurate CDW phase emerges [23], accompanied by a lattice distortion. Single-crystal XRD studies have further reported a transition to an orthorhombic noncentrosymmetric structure with $F222$ symmetry [26]. A second transition, observed near $T \simeq 90$ K, lowers the symmetry further to a monoclinic phase with centrosymmetric point group $2/m$. More recently, angle-resolved photoemission spectroscopy (ARPES) and scanning tunneling microscopy (STM) measurements probed the evolution of the Fermi surface and the

*Contact author: alonron@tauex.tau.ac.il

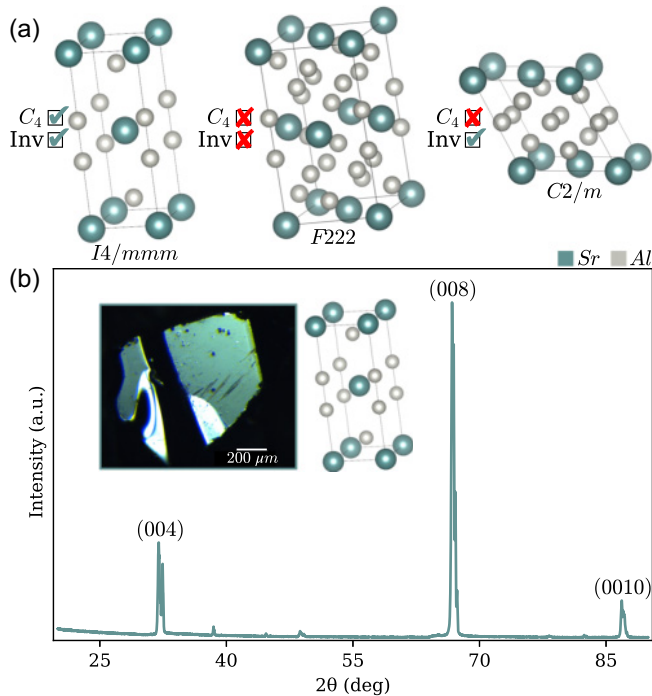


FIG. 1. (a) SrAl_4 unit cell evolution. Significant symmetries that are preserved in the lattice are indicated by a $\checkmark$, and broken by an $\times$. The leftmost is the $I4/mmm$ phase stable at room temperature. The middle illustration describes the structure reported below the CDW transition and the rightmost structure is the one reported below the monoclinic transition. (b) X-ray diffraction scan performed on single crystal of SrAl_4 oriented to the (001) plane. Inset contains image of representative SrAl_4 crystal and an illustration of SrAl_4 unit cell in the $I4/mmm$ space group—the reported crystalline structure of SrAl_4 at room temperature [17].

atomic arrangement at the surface of SrAl_4 [27]. These experiments revealed broken surface C_4 symmetry below T_{CDW} , manifested in the appearance of unidirectional replica bands in momentum space and a nematic arrangement of the surface atoms. Taken together, these results point to an unusual and still unresolved symmetry landscape in SrAl_4 , including the apparent restoration of inversion symmetry below 90 K following inversion-symmetry breaking below T_{CDW} , as well as the unclear connection between the nematicity observed by ARPES/STM and the reported bulk structural transitions.

Here, we use nonlinear second-harmonic generation and third-harmonic generation (THG) to probe the symmetries of SrAl_4 and their relationship to CDW order. By analyzing the anisotropy of the nonlinear optical response, we revisit previous symmetry assignments, extend the temperature range over which C_4 symmetry breaking can be identified, discuss the need for multiple contributions such as domains, and reveal symmetry-breaking features that have remained inaccessible to conventional diffraction-based probes.

II. EXPERIMENTAL METHODS

Our experiments were performed on the naturally grown (001) surface of flux-grown single crystals of SrAl_4 , prepared

as described in Ref. [25]. Two sets of samples with different disorder levels were studied, grown using Sr source materials of 99% and 99.99% purity. Rotational-anisotropy SHG and THG measurements were carried out in a normal-incidence polarizer-analyzer geometry. A pulsed laser beam at wavelengths $\lambda = 800$ nm and $\lambda = 1200$ nm was used to generate the second- and third-harmonic signals, respectively, using the signal and idler outputs of an optical parametric amplifier. The polarization of the incident and analyzed beams was rotated by an angle ϕ relative to the crystal axes, thereby mimicking a rotation of the sample itself. The reflected second- and third-harmonic signals were collected and bandpass filtered around $\lambda = 400$ nm, detected by a photomultiplier tube, and processed using a lock-in amplifier referenced to the laser repetition rate of 100 kHz. Measurements on both sets of crystals were performed at room temperature and at low temperature using a closed-cycle cryostat with free-space optical access. The crystals were mounted on linear, rotational, and tilting piezoelectric stages, enabling fine adjustment of the reflected beam trajectory and allowing the angle of incidence to be determined with a precision better than 1° . The beam spot diameter was on the scale of tens of micrometers, corresponding to laser fluences of order $1\text{--}10$ mJ/cm². X-ray diffraction scans, performed using a Cu $K\alpha$ source, were used to identify the (001) surface, as shown in Fig. 1(b).

III. RESULTS AND DISCUSSION

Figures 2(a) and 2(b) show the room-temperature THG response of an SrAl_4 sample grown using 99% purity Sr, measured in the parallel and crossed polarization configurations, respectively. In the parallel channel, shown in Fig. 2(a), the THG pattern exhibits a clear C_2 symmetry, with the small lobes in the pattern being 34% smaller than the large ones, indicating breaking of the C_4 rotational symmetry expected for the reported $I4/mmm$ crystal structure. A similar twofold pattern is observed in the crossed channel in Fig. 2(b). Figures 2(c) and 2(d) show the corresponding room-temperature measurements for samples grown using 99.99% purity Sr, where similarly a C_2 -symmetric pattern is observed with a 52% contrast between its small and large lobes. Since SrAl_4 is expected to remain in the $I4/mmm$ space group at room temperature, the absence of the anticipated C_4 -symmetric THG response is surprising. To identify the origin of the observed twofold anisotropy, we fit the data using the highest subgroup of $4/mmm$ that lacks a C_4 rotation axis, namely, the orthorhombic point group 222. It is unlikely that the breaking of C_4 symmetry at room temperature in SrAl_4 originates from a global symmetry lowering of the bulk, mainly due to lack of evidence for such structural transitions from diffraction-based probes [25,26].

This points toward the more plausible interpretation that the symmetry breaking observed in the THG measurements originates from symmetry breaking in the near-surface region. This could arise from inhomogeneous strain close to the surface or from atomic rearrangements at the surface, which have been observed by low-temperature STM measurements [27]. Such structural modifications may, in fact, extend beyond the immediate surface and penetrate into the bulk. Since THG is considered to be a bulk property probe, it is reasonable to

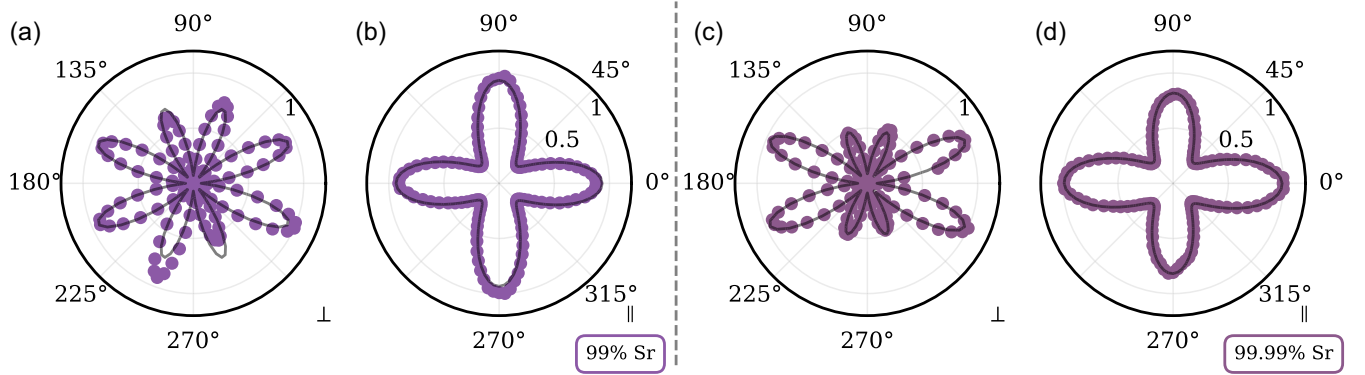


FIG. 2. Rotational-anisotropy THG measurements in the cross ($\perp$) and parallel ($\parallel$) configurations at room temperature and disorder level. Panels (a) and (b) show data for the high disorder 99% SrAl_4 and panels (c) and (d) for the low disorder 99.99% SrAl_4 crystals. All the crystals were oriented to the (001) surface. The measurements were carried out at room temperature. Purple circles are data points and the solid gray lines are best fits to the data.

conclude that the C_4 breaking order persists to a depth at least comparable to the penetration of the laser beam, which is of the order of tens of nanometers (comparable to other known topological semimetals [28,29]). Some sample-to-sample variation in the THG intensity pattern is present, likely reflecting small differences in composition or disorder near the sample surface, and is discussed further in Sec. II of the Supplemental Material [21].

To investigate the interplay between CDW order and crystal symmetry, we performed temperature-dependent rotational-anisotropy SHG measurements, which are particularly sensitive to inversion-symmetry breaking. Figure 3(a) shows the temperature-dependent SHG response measured on the (001) surface of the SrAl_4 sample grown using 99% purity Sr. Above 240 K, only a weak and nearly uniform background signal is observed. This response is isotropic, remaining unchanged as the incident and detected polarizations are rotated, and may be associated with inversion-preserving higher-order contributions allowed in the $I4/mmm$ structure. Below $T_{\text{CDW}} \simeq 240$ K, the SHG intensity

begins to increase monotonically [Fig. 3(b)] and evolves into a four-lobed pattern with apparent C_4 symmetry. These lobes become clearly distinguishable from the background below approximately 200 K. The emergence of a nonzero anisotropic SHG response indicates inversion-symmetry breaking associated here with the onset of CDW order. The highest-symmetry point group consistent with such a signal is orthorhombic. Previous XRD studies reported that below the CDW transition SrAl_4 is best described by the noncentrosymmetric orthorhombic space group $F222$ [26]. However, under normal-incidence excitation, the electric field lies entirely within the (001) plane, while the electric-dipole SHG tensor of point group 222 contains only elements that couple to out-of-plane electric fields or generate out-of-plane polarization (see Sec. III in the Supplemental Material for details [21]). Therefore, we infer that the observed SHG signal does not originate from the electric-dipole response of the orthorhombic bulk with point group 222, but instead reflects a lower symmetry. Since point group 222 is ruled out, we attempt to fit to its highest symmetry subgroup 2. Figure 3(c) shows the temperature evolution of the SHG tensor elements extracted from our fits. The expressions for the intensity incorporate χ_{yyy} and a linear combination $2\chi_{xxy} + \chi_{yyy}$ (see Sec. IV in the Supplemental Material [21]) that could be extracted independently from our data. These adopt a square-root mean-field-like functional behavior up to T_{CDW} , which goes hand in hand with the intensity's linear trend. Small deviations from it close to 240 K most likely arise due to the isotropic SHG signal that is still non-negligible at this temperature range. Since diffraction-based probes [25,26] have not detected signatures for a monoclinic phase at this temperature range, we suggest that a possible origin for the point group 2 signal is the 001 surface of the orthorhombic structure for point group 222 that has a point group 2 symmetry. Additionally, we do not observe any signature near $T \simeq 90$ K of the monoclinic structural transition previously reported for SrAl_4 in this temperature range [23].

Having found no signature of the reported low-temperature transition in the sample grown using 99% purity Sr, we turn to examine the effects of disorder on the evolution of SHG with temperature. Figure 4(a) shows the temperature-dependent SHG measurements for the sample grown using 99.99%

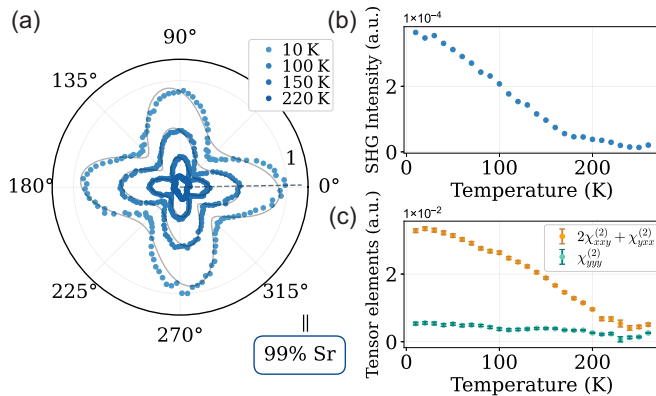


FIG. 3. (a) Rotational-anisotropy SHG measurements in the parallel configuration, taken on the 99% SrAl_4 , oriented to the (001) surface. Temperature sweep was performed at 10 K steps cooling down from room temperature to 10 K. (b) Intensity as a function of temperature plotted over a single angle [indicated by the striped gray line in panel (a)]. (c) SHG $\chi^{(2)}$ tensor element decomposition as a function of temperature.

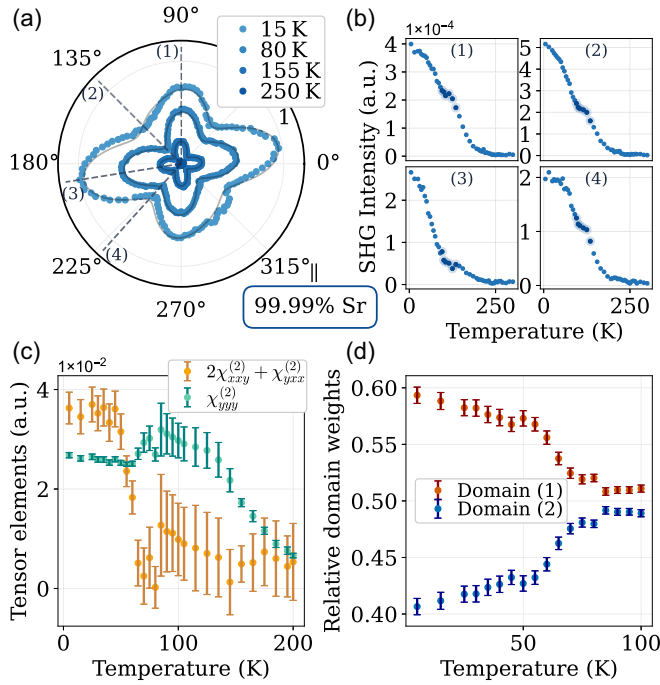


FIG. 4. (a) Rotational-anisotropy SHG measurements in the parallel configuration, taken on the low disorder 99.99% SrAl_4 , oriented to the (001) plane. Temperature sweep was performed at 10 K steps cooling down from room temperature to 10 K. Distortion is present in the different floral patterns below $T \simeq 90$ K. (b) Intensity as a function of temperature plotted over a few different angles, corresponding to the stripped gray lines numbered in panel (a). The monoclinic transition manifests as a flat disruption of the growing (cooling down) SHG intensity signal. (c) SHG $\chi^{(2)}$ tensor element decomposition as a function of temperature. (d) Relative weights of the two components as a function of temperature.

purity Sr. As in the 99% purity sample, a C_4 -symmetric SHG pattern emerges upon cooling through the CDW transition at $T_{\text{CDW}} \simeq 240$ K. In contrast, however, the pattern begins to distort below $T \simeq 90$ K, deviating from the approximate C_4 symmetry observed at higher temperatures. This distortion is accompanied by a flattening of the SHG intensity measured at a fixed angle, shown in Fig. 4(b), over the same temperature range. We associate this feature with the structural transition previously reported near $T \simeq 90$ K in SrAl_4 [23], which lowers the bulk symmetry to a monoclinic structure. Attempts to fit the C_4 symmetry breaking data (below 90 K) to point group 2 result in very poor agreements with the data. Instead of turning to a lower symmetry group, we fit the data to two independent components of point group 2. Figure 4(c) shows the $\chi^{(2)}$ tensor elements associated with monoclinic point group symmetry 2 as a function of temperature, while the corresponding weights of the two components are shown in Fig. 4(d). As soon as the temperature is lowered below $T \simeq 90$ K, the material exhibits a pronounced two-component behavior. The relatively large uncertainties in the tensor elements shown in panel (c) arise primarily from the need to employ a two-component analysis to fit the data, preventing a quantitative interpretation of their temperature dependence. In contrast, the relative weights shown in panel (d) remain sufficiently well determined to clearly identify the onset of

the two-component behavior below $T \simeq 90$ K in agreement with the monoclinic transition temperature that was observed in x-ray diffraction measurements.

The fact that the two-component regime is observed in the sample grown using 99.99% purity Sr, but not in the sample grown using 99% purity Sr, indicates that disorder suppresses or quenches the monoclinic transition that was observed in x-ray. While it may be surprising that the introduction of such mild disorder has such great structural implications, we note that it also has great effects on the residual resistivity ratios that change by almost an order of magnitude from 8 in the 99% sample to 70 in the 99.99% sample [30]. The multiple component nature we have observed could also be the reason for the absence of the lambda anomaly in heat capacity measurements [26] since the presence of multiple structural domains could significantly broaden such effects. Previous single-crystal XRD studies classified the low-temperature monoclinic phase as inversion preserving, with point group $2/m$ [26], guided in part by the behavior of isostructural CaAl_4 , which undergoes a similar tetragonal-to-monoclinic transition [18,19]. However, our SHG data show no evidence that the CDW-related inversion-symmetry breaking is suppressed across this transition. Instead, the SHG response remains finite below $T \simeq 90$ K, implying that inversion symmetry continues to be broken, in contrast to earlier interpretations. Beyond the direct implications of our results, several plausible interpretations warrant consideration. In the 99% samples, a single component was sufficient to explain the data below T_{CDW} , which is possibly attributed to the surface contribution of point group 222. The additional component detected in the 99.99% samples could originate from the monoclinic bulk below 90 K that joins the surface effect that onsets below T_{CDW} . Considering the possibility that the two components originate from the surface and the bulk, one might expect that the bulk signal would be stronger than the surface signal. Possible explanations for this discrepancy are that under reflection geometry some of the reflected photons are absorbed on the way toward the surface; alternatively, it could be that the surface signal is particularly strong due to the presence of topological or trivial surface states found in a nodal-line semimetal. Another possibility is that the two components do not originate separately from the surface and the bulk but rather from two bulk domains.

IV. CONCLUSIONS

In this work, we mapped the symmetry evolution of SrAl_4 across its tetragonal, CDW, and monoclinic phases. Room-temperature THG measurements revealed broken C_4 rotational symmetry, in contrast to the reported $14/mmm$ space group, which we propose originates from a surface proximate reconstruction that persists up to room temperature. Observing this in THG, which is a bulk probe, and not through diffraction-based techniques suggests that this symmetry breaking could be localized to a finite region near the surface of the crystal rather than to the whole bulk, or solely to the top layers near the surface. Temperature-dependent SHG measurements showed the emergence of a four-lobed pattern upon cooling below $T_{\text{CDW}} \simeq 240$ K, coincident with the onset of CDW order. Constraints imposed by the measurement geometry further indicate that this SHG

response originates from a monoclinic point group 2 whose possible origin is the monoclinic 001 surface of a crystal with an orthorhombic bulk. The comparative study of samples with different disorder levels revealed that the monoclinic phase at 90 K is suppressed when mild disorder is introduced, highlighting the fragility of the monoclinic phase to weak disorder. Finally, two orthogonal monoclinic components were required to accurately describe the low-temperature SHG response of the higher-purity samples.

ACKNOWLEDGMENTS

This research was supported by Grant No. 2022126 from the United States-Israel Binational Science Foundation (BSF).

A.R. acknowledges support from the Zuckerman Foundation and the Israel Science Foundation (Grant No. 592/25). Research at the University of Maryland was also supported by the Gordon and Betty Moore Foundation's EPIQS Initiative through Grant No. GBMF9071 and the Maryland Quantum Materials Center. We thank Judy Cha and Jason Schibler for useful discussions.

DATA AVAILABILITY

The data that support the findings of this article are openly available [31].

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