

Phase transitions and electrical conductivity of selenium disulfide at high pressure

Huixin Hu,¹ Israel Osmond,² Calum Strain,² Hannah A. Shuttleworth², Callum R. Stevens,² Andrew Huxley,² Mikhail A. Kuzovnikov², Federico A. Gorelli^{1,3,4}, Eugene Gregoryanz,^{1,2,3,5} Miriam Peña-Alvarez,² Philip Dalladay-Simpson¹, and Ross T. Howie^{1,2,*}

¹*Center for High Pressure Science & Technology Advanced Research, 1690 Cailun Road, Bldg. 6, Pudong, Shanghai 201203, People's Republic of China*

²*Center for Science at Extreme Conditions & The School of Physics and Astronomy, The University of Edinburgh, Peter Guthrie Tait Road, Edinburgh EH93FD, United Kingdom*

³*Shanghai Advanced Research in Physical Sciences (SHARPS), 68 Huatuo Road, Bldg. 3, Pudong, Shanghai 201203, People's Republic of China*

⁴*CNR-INO, Istituto Nazionale di Ottica, Consiglio Nazionale delle Ricerche, 50019 Sesto Fiorentino, Italy*

⁵*Key Laboratory of Materials Physics, Institute of Solid State Physics, Chinese Academy of Sciences (CAS), Hefei, Anhui 230031, People's Republic of China*

 (Received 25 August 2025; revised 22 January 2026; accepted 10 February 2026; published 4 June 2026)

Sulfur and selenium demonstrate one of the most complex behavior under high pressure among all elements of the periodic table. Despite being known to form interchalcogens, the properties of these compounds have not been widely explored in the dense state. Through a series of diamond anvil experiments combined with x-ray diffraction, optical spectroscopy, and electrical resistance measurements, we explore the properties of selenium disulfide (SeS₂) up to pressures of 150 GPa. At ambient pressure, SeS₂-I represents a substitutional solid solution of S and Se atoms, forming an eight-membered molecular ring arrangement analogous to γ -S. The band gap of SeS₂-I rapidly closes upon compression, and above 27 GPa, there is a transformation to a metallic tetragonal phase (SeS₂-III), in which atoms form square helical chains. Upon further compression, we observe a phase sequence from incommensurate modulated SeS₂-IV above 59 GPa, transforming to rhombohedral SeS₂-V by 116 GPa.

DOI: [10.1103/s6wf-56gj](https://doi.org/10.1103/s6wf-56gj)

I. INTRODUCTION

Sulfur and selenium are known to form a range of continuous substitutional solid solutions, with formulas Se_xS_{8-x}, whereby selenium and sulfur are placed statistically over atomic sites to form a disordered molecular structure [1]. Co-melting of S-Se mixtures yields glassy alloys that have the potential for flexible device applications, being lightweight, viscoelastic, and a high- κ dielectric despite sulfur and selenium being nonpolar and brittle [2]. In crystalline form, the alloy SeS₂ has widespread use in medicine as an antifungal agent to treat common conditions such as dandruff. While referred to as SeS₂, the compound in fact consists of eight-membered rings linked via single S-S, S-Se, and Se-Se bonds, with an average chemical composition of around 1:2 for Se and S. The application of pressure has been shown to induce novel properties in both sulfur and selenium, becoming metallic and superconducting in the dense state, albeit at vastly different conditions [3–7]. Despite such interesting

phenomena in the constituent elements, the high-pressure response of S-Se alloys has not been widely explored.

Upon compression, elemental sulfur and selenium demonstrate the most complex behavior among all elements of the periodic table [3,5,6,9–20]. Sulfur has been shown to exhibit a wide range of allotropes and remarkable polymorphism, with some 12 solid and 5 liquid phases reported below pressures of 4 GPa [9]. At ambient pressure, the most stable S-I polymorph of sulfur consists of S₈ molecules arranged in slightly puckered rings [9]. On compression above 1.5 GPa and heated to temperatures close to the melt, S-I transforms to trigonal S-II, which consists of infinite helices with a pitch of 3 sulfur atoms [14]. Above 36 GPa, there is a further transformation to tetragonal S-III, where there is a change to a 4-atom pitch of the helices [14]. Above 83 GPa, S-IV adopts a complex incommensurately modulated monoclinic structure before transforming to S-V at 153 GPa, with a primitive simple rhombohedral (β -Po type) crystal structure [13,15,19].

Selenium exhibits a similar phase sequence upon compression, albeit with the transitions occurring at different pressures. At ambient pressure, the stable phase of selenium (Se-I) is trigonal with triangular spiral chains, which is isogeometric to S-II [9,14]. Upon compression at room temperature, Se-I converts into monoclinic Se-II at 14 GPa [6,21]. In a narrow pressure region between 23 and 28 GPa, selenium adopts a triclinic structure, Se-III, which is isomorphic to Te-II [22], rather than the phases of sulfur. Se-IV is stable between

*Contact author: ross.howie@ed.ac.uk

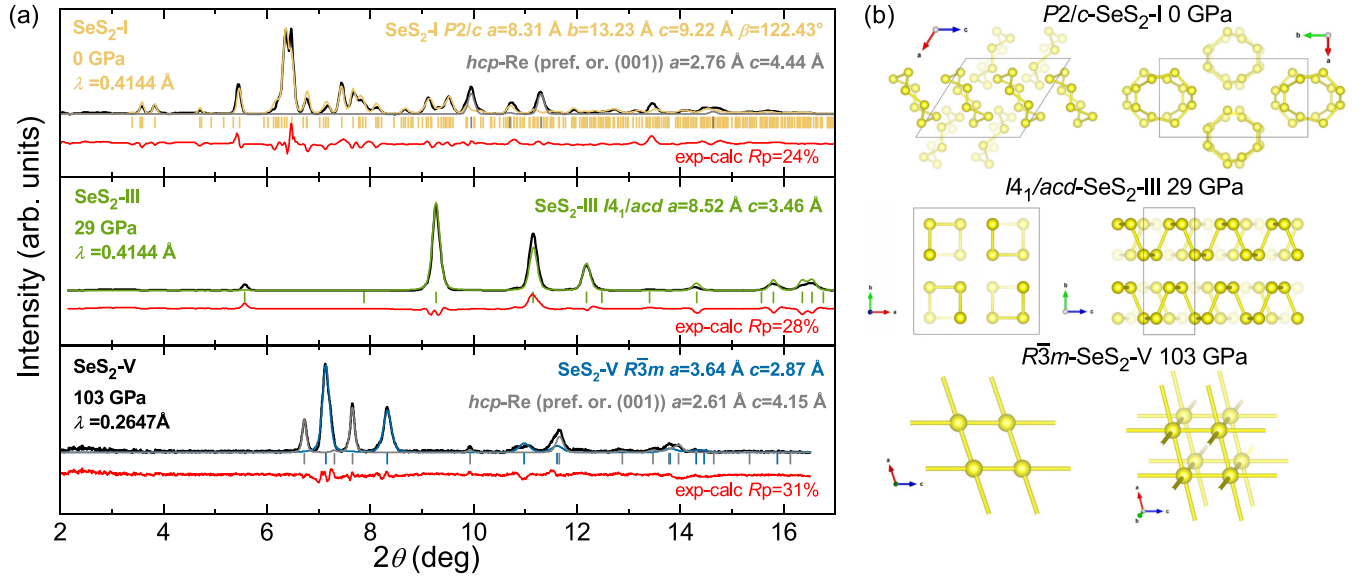


FIG. 1. (a) X-ray diffraction patterns and Rietveld refinements of SeS₂-I at ambient pressure (top), SeS₂-III at 29 GPa (center), and SeS₂-V at 103 GPa (bottom). The experimental structure factors are shown in black, the red curves show the refinement residuals, and the curves of other colors show the calculated contributions from individual phases. (b) Structural models of SeS₂-I *P2/c* structure at ambient pressure (top) illustrating distorted eight-member S/Se rings, SeS₂-III *I4₁/acd* at 29 GPa (center) illustrating helical S/Se chains, and SeS₂-V *R3m* at 103 GPa (bottom). The refined structural parameters are given in Table S1 in the SM [8].

29 and 80 GPa (isomorphic to S-IV) [16,23], above which there is a transformation to Se-V (isomorphic to S-V) [16]. At above 140 GPa, Se-VI is stable with a body-centered-cubic structure [11,15].

Both sulfur and selenium undergo transformations to metallic states upon compression [3–5,7,24]. Sulfur becomes metallic upon the transition to S-IV above 90 GPa, also exhibiting superconductivity with a $T_c = 10.1$ K at 93 GPa [3,5,7]. Upon the phase transition to S-V, the T_c increases to 17.3 K between 180 and 200 GPa [3]. Selenium becomes metallic at substantially lower pressures between 12 and 24 GPa and is dependent on the initial ambient pressure polymorph [6,24]. Superconductivity has been reported over a wide pressure range of 17–160 GPa, with T_c ranging from 4 to 7 K [3,24]. Given the difference in phases exhibited by both sulfur and selenium, together with the disparity in phase-stability pressure regimes, it is of intrigue how an interchalcogen such as SeS₂ would behave upon compression. Furthermore, there is a propensity for alloys to exhibit enhanced properties; therefore, there is potential to tune the electrical conductivity properties by combining sulfur and selenium.

In this study, we investigate the phase sequence and properties of the interchalcogen SeS₂, upon compression through a series of high-pressure diamond anvil cell experiments. Our measurements reveal that SeS₂-I is isomorphic with γ-S, which is comprised of eight-membered ring molecules. The electronic band gap of SeS₂-I gradually decreases upon compression, and eventually this phase transforms into tetragonal SeS₂-III isomorphic to S-III at 27 GPa. At pressures above 59 GPa, there is a transition to incommensurate modulated SeS₂-IV, which exhibits a soft Raman-active amplitude mode related to this charge density wave

phase. By 116 GPa, the phase transforms to rhombohedral SeS₂-V.

II. EXPERIMENTAL METHODS

SeS₂ is a solid solution, with an average S:Se ratio of 2:1. Commercial samples of SeS₂ (97% from Alfa Aesar, 99.9% from Macklin) were hermetically sealed into diamond anvil cells (DACs), both with and without a helium pressure transmitting medium. Sample chambers were formed by gaskets of rhenium for most experiments and Al₂O₃-epoxy mixture for electrical resistance measurements. Pressure was determined through the ruby fluorescence method [25] below 50 GPa in optical spectroscopy experiments and by the stressed diamond Raman edge at pressures above [26]. In x-ray diffraction experiments, the pressure was determined through the equation of state of gold [27].

Angle-dispersive x-ray diffraction experiments were performed at the BL10XU beam line [28] (Perkin Elmer XRD0822 detector, $E \sim 30.0$ keV, $\lambda = 0.4144$ Å) at SPring-8, Japan, the I15 beam line [29] (MAR345 detector, $E \sim 29.2$ keV, $\lambda = 0.4251$ Å) at the Diamond Light Source, UK, and the ID15-b [30] (EIGER2 X 9M CdTe detector, $E \sim 30.2$ keV, $\lambda = 0.4101$ Å) and ID27 beam lines [31] (EIGER2 X CdTe 9M detector, $E \sim 46.8$ keV, $\lambda = 0.2647$ Å) at ESRF, France. The sample-to-detector distance, detector tilt, and rotation were calibrated based on the diffraction patterns of either CeO₂ or Si powder standards using the Dioptas software [32]. Primary data processing, azimuthal integration, and background subtraction were performed using the Dioptas version 0.5.5 software [32]. The Powdercell 2.4 and Fullprof 7.00 programs were used for the Rietveld refinement [33,34]. To estimate the pressure domains of stability and construct the

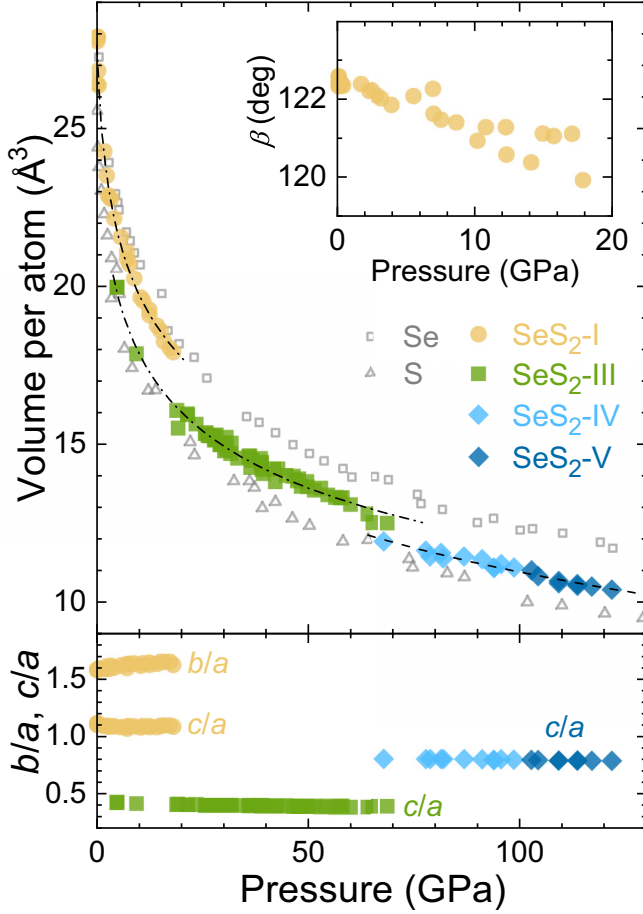


FIG. 2. Pressure dependencies of the atomic volume (top), axial ratios (bottom), and monoclinic angle (inset) of SeS_2 . The solid symbols are the experimental data and the dashed curves are the results of the $V(P)$ data fit with the Vinet equation of state, with the parameters listed in Table S2 in the SM [8]. The open symbols represent sulfur and selenium from the literature [15].

$V(P)$ equations of state for individual phases, we performed x-ray diffraction measurements during stepwise compression and decompression using membrane-type DACs. Typically, the time between two consecutive pressure points was several minutes. Figures S1–S3 in the Supplemental Material (SM) illustrate the typical x-ray diffraction patterns collected upon compression and laser heating [8].

Raman spectroscopy measurements were conducted in 180° backscattering confocal geometry with 514, 532, and 647 nm laser excitations. At pressures below 20 GPa, the laser power was kept to less than 1 mW due to sample sensitivity. An Acton SP-2500i spectrometer (Princeton Instruments) equipped with 300 and 1200 gr/mm grating coupled with PyLoN CCD (Princeton Instruments) was used for spectra collection.

Absorption measurements were conducted using a Bruker Vertex 70v FTIR coupled with a Hyperion microscope over photon energies range from 0.08 to 0.7 eV (using a KBr beam splitter and LN-MCT detector). Absorbance (A) was calculated using the function $\lg(I_r/I_s)$, where I_s is the intensity from the sample and I_r is the reference intensity from the ambient chamber filled with KBr. The energies of the direct

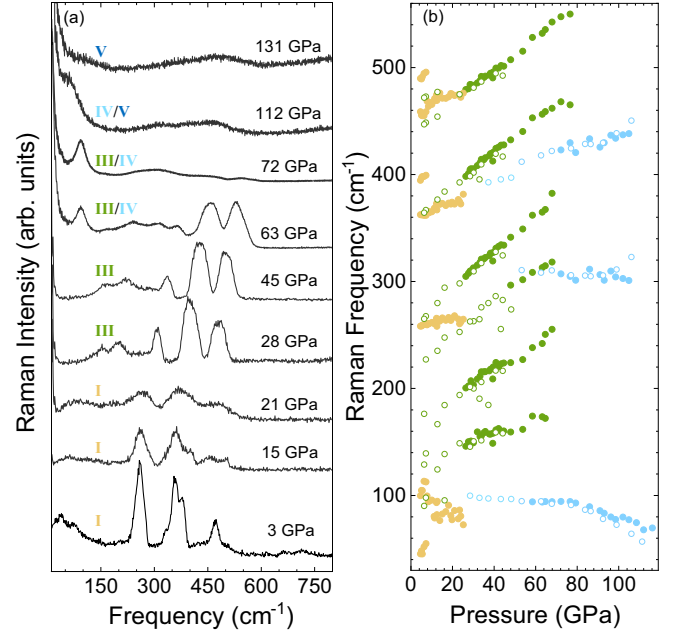


FIG. 3. (a) Representative Raman spectra of SeS_2 upon compression at 300 K. Colors represent each phase: yellow: SeS_2 -I; green: SeS_2 -III; light blue: SeS_2 -IV; dark blue: SeS_2 -V. (b) Raman frequency as a function of pressure upon compression (solid circles) and decompression (empty circles) at 300 K. The color scheme is identical to (a).

band gaps were determined using the Tauc plot method [35], which are equivalent to the intercepts of the linear regions extrapolated to the x axis in the plot of $(\alpha E)^2$ versus E , where α is the absorption coefficient and E is the photon energy. Upon compression, A is proportional to α , allowing A to be substituted for α .

Electrical resistance measurements were performed using an impedance analyzer (Hioki IM353) for two-point resistance measurements at low pressure, where the contact resistance ($\approx 100 \Omega$) is negligible compared to the sample resistance. Such measurements were taken at an excitation frequency of 4 Hz and voltage of 1 V. At higher pressure, measurements were performed in a four-probe van der Pauw configuration where a multimeter (Keithley 2001) was used to eliminate the resistance contributions from the sample leads [36], using a direct current (DC) excitation of 1 mA. In such cells, the gasket was comprised of steel foil pre-indented and electrically insulated using an Al_2O_3 -epoxy mixture. Electrodes were then formed of deposited W/Au layers leading into the sample chamber.

III. RESULTS AND DISCUSSION

A. Phase I

Upon sample loading, x-ray diffraction confirmed that SeS_2 -I is isostructural to γ sulfur with space group $P2_1/c$; [see Figs. 1(a), 2(a), and 2(b)] [37]. The structure consists of molecules in an eight-atom ring configuration, with S and Se atoms having disordered occupation with an average S:Se ratio of 2:1. Raman spectroscopy measurements of SeS_2 -I show signatures consisting of three

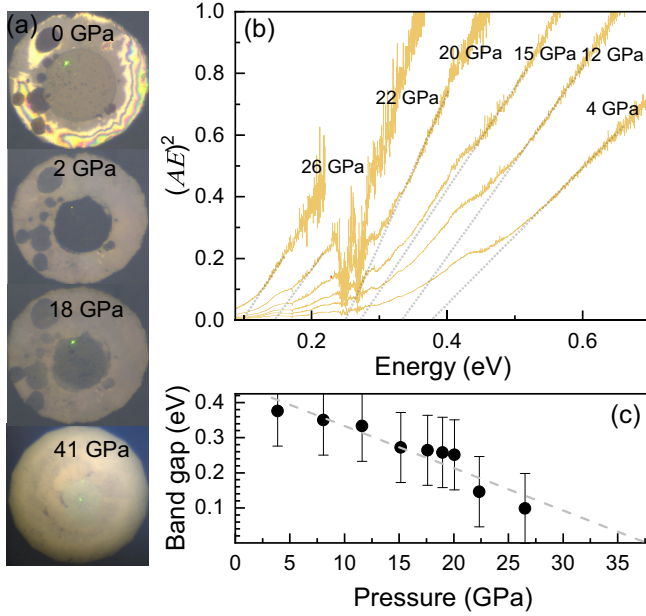


FIG. 4. (a) Photomicrographs of SeS₂ at 0, 2, 18, and 41 GPa. (b) Representative absorbance spectra of SeS₂ in the middle-infrared wavelength regime presented as $(AE)^2$ vs energy, where A is the absorbance, E is the photon energy, and the $A = \log_{10}(I_t/I_s)$ relationship is used, where I_s and I_t are the transmitted intensity through the sample and KBr, respectively. Linear extrapolations of the absorbance data give an estimated value of the band gaps. (c) Estimated electronic band gap (E_g) of SeS₂ as a function of pressure. A fit of these values gives band-gap closure at 37.7 GPa.

well-defined bands which correspond to S-S (~ 470 cm⁻¹), S-Se (~ 360 cm⁻¹), and Se-Se (~ 260 cm⁻¹) stretches at 3 GPa [Fig. 3(a)], that increase in frequency as a function of pressure [Fig. 3(b)].

Upon compression, the eight-atom rings become distorted, resulting in the intensity redistribution in the diffraction pattern. Above 17 GPa, the diffraction peaks become significantly broader compared with the ambient pressure diffraction patterns, indicative that the sample is becoming poorly crystalline. This effect upon compression is observed regardless of whether or not the samples are compressed in a He pressure transmitting medium (see Figs. S1 and S2 in the SM [8]). This is also reflected in the Raman spectroscopy measurements, whereby the intramolecular stretching bands reduce in intensity and broaden significantly [Fig. 3(a)].

At ambient pressure and temperature, SeS₂-I is visibly orange in color and, upon compression, rapidly turns red before turning black. These changes are observed in our optical absorbance measurements, whereby we see a continuous increase in absorption with pressure in the 0.1 eV to 0.7 eV energy range [see Fig. 4(b)]. Plotting the absorption data in photon energy versus $(AE)^2$ (where A is the absorbance and E is the photon energy), we can estimate direct band-gap energy from a linear extrapolation of the absorption edge. At 4 GPa, we estimate the sample to have a band gap of 0.4 eV, typical of a semiconductor, rapidly reducing upon compression to 0.1 eV at 26 GPa [Fig. 4(c)]. Linear extrapolations of the band gap as a function of pressures estimates full closure at 37.7 GPa.

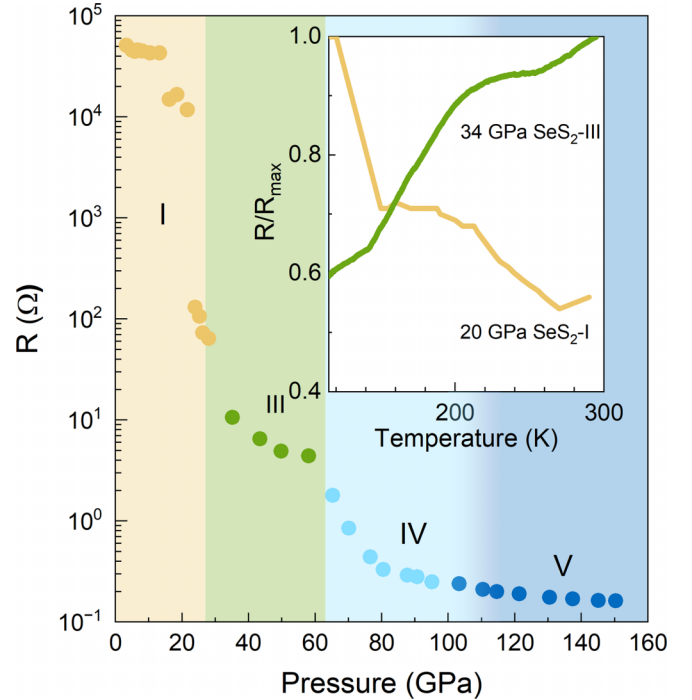


FIG. 5. Four-probe electrical measurements as a function of pressure in SeS₂. Color backgrounds indicate the pressure ranges of different phases: SeS₂-I (yellow), SeS₂-III (green), SeS₂-IV (light blue), SeS₂-V (dark blue). The inset represents normalized resistance as a function of temperature in SeS₂-I (yellow, 20 GPa) and SeS₂-III (green, 36 GPa).

To further explore changes in the electronic properties of SeS₂, we have performed electrical transport measurements. At low pressures, we observe high resistance values (~ 50 k Ω), which drops steadily upon compression to 10 k Ω at 22 GPa (Fig. 5). Upon cooling the sample at a pressure of 20 GPa, we observe that the resistance of the sample increases, which is typical behavior of a semiconductor.

B. Phase III and metallic transition

If the SeS₂-I phase is compressed to pressures exceeding 27 GPa, the texture of the x-ray diffraction pattern instantaneously transforms from barely resolvable Bragg peaks to a good powder (see Figs. S1 and S2 in the SM [8]). The transformation pressure is the same in samples both with and without the helium pressure transmitting medium. Rietveld refinements reveal a transformation to a tetragonal structure ($a = 8.52$ Å, $c = 3.46$ Å at 29 GPa) with space group $I4_1/acd$ (see Fig. 1). This phase is isostructural to S-III (and Se-VII) with squared helical chains, running parallel to the tetragonal c axis, with four atoms per turn [14]. To maintain consistent nomenclature with sulfur, we denote this phase as SeS₂-III. Upon decompression, SeS₂-III is stable down to 5 GPa, below which the sample transformed to an amorphous phase. Laser heating either the amorphous phase above 7 GPa or SeS₂-I above 18 GPa resulted in the sample transforming to SeS₂-III. The Raman spectra also markedly change upon the phase I–III transition, with two new modes emerging at ~ 150 cm⁻¹ and ~ 202 cm⁻¹ at 28 GPa [Fig. 3(a)]. The

three S-S, Se-S, and Se-Se stretching bands also become better resolved and increase in intensity. In the frequency dependence as a function of pressure, all three main bands of SeS₂-III show more significant increases compared with SeS₂-I.

Coinciding with this transition, we observe the resistance of the sample sharply drop between 20 and 30 GPa (Fig. 5). Performing cooling cycles at this pressure (and above) indeed shows that the sample has undergone a transition to a metallic state with a positive dR/dT (see the inset of Fig. 5). Furthermore, after the phase transformation, the sample appears visually metallic and difficult to distinguish from the metallic rhenium sample chamber [Fig. 4(a)].

Interestingly, while phase-III can be produced by room-temperature compression in both SeS₂ and pure S, the analogous phase in selenium (Se-VII) is synthesized at high temperature or transforms from metastable α -Se₈ at room temperature [38]. In sulfur, heating S-I does not yield S-III, but instead forms trigonal S-II or six-atom ring S-VI [14,39], while heating SeS₂-I forms SeS₂-III. Furthermore, the metallization of sulfur does not occur until 83 GPa upon the transition to the incommensurate S-IV, while we observe metallization at substantially lower pressures of 34 GPa upon the compression to SeS₂-III.

C. Transition to phases IV and V

Changes are also observed in the Raman spectrum upon the III-IV transition. Between 59 and 72 GPa, the S-S, Se-Se, and S-Se stretching bands reduce significantly in intensity, while two weak and broad peaks appear at 300 cm⁻¹ and 400 cm⁻¹ [Fig. 3(a)]. These are similar to the Raman signatures previously reported for S-IV and Se-IV [40,41], respectively. Furthermore, we observe a low-frequency Raman mode at a frequency of 94 cm⁻¹ (at 63 GPa), which we identify as a charge density wave amplitude (AMP) mode. This mode is due to the symmetry breaking caused by modulated displacements of atoms from their equilibrium positions in the incommensurate phase. Generally, amplitude modes are Raman active in incommensurate phases and merge into the wings of the Rayleigh line near the critical transition point [40–44]. This mode softens with pressure, indicative of molecular dissociation towards an atomic structure. This mode is observed in both analogous sulfur and selenium incommensurate phases [40,41,45]. Upon compression, this mode is observed up to pressures of 116 GPa, suggesting that the SeS₂-IV to SeS₂-V transition is not complete until above this pressure. Once fully transformed to SeS₂-V, we observe no further phase transformations up to 150 GPa. Interestingly, upon decompression of SeS₂-IV, we observe the AMP mode at pressures as low as 29 GPa, suggestive that SeS₂-IV and SeS₂-III coexist at such conditions.

At pressures above 68 GPa, x-ray diffraction measurements reveal a phase transition from SeS₂-III to a distorted primitive rhombohedral structure, which we denote SeS₂-IV. The average atomic volume change at the III-IV transition is 4.6%, characteristic of a typical first-order transition (Fig. 2). Upon compression, the distortion gradually disappears above 100 GPa and the crystal structure is refined with space group $R\bar{3}m$ ($a = 3.64$ Å and $c = 2.87$ Å at 103 GPa) [Fig. 1(a)]. We

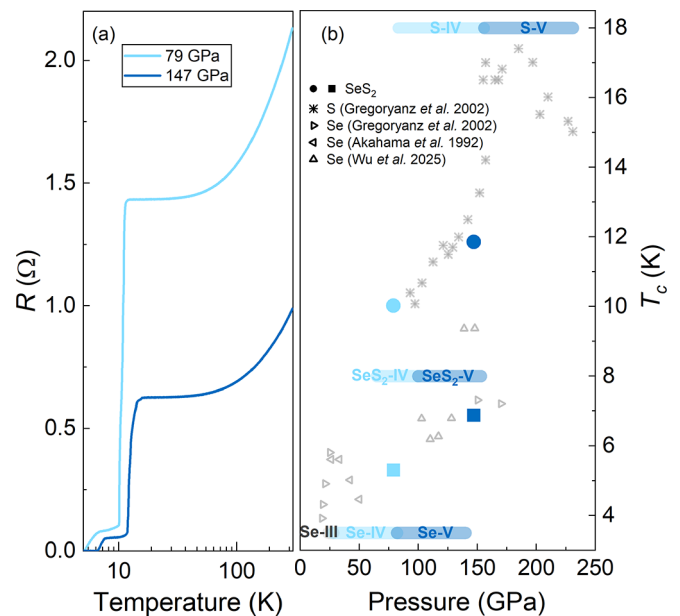


FIG. 6. (a) Four-probe electrical resistance measurements as a function of temperature in SeS₂. Light and dark blue represent pressures of 79 GPa (phase IV) and 147 GPa (phase V), respectively. (b) T_c as a function of pressure for SeS₂ (circles and squares represent temperatures corresponding to residual resistances and zero resistances, respectively) compared with the literature values of S (star-shaped symbols) and Se (triangles) [3,6,46]. Horizontal bars represent the pressure stability field of S, Se, and SeS₂ phases IV (light blue) and V (dark blue).

believe that this is analogous to the transition in the sulfur and selenium systems where incommensurate S-IV (Se-IV) transforms to S-V (Se-V).

Both sulfur and selenium exhibit superconductivity upon metallization in their respective incommensurately modulated metallic phase IV and rhombohedral phase V [3,6]. Given that SeS₂ shares properties with both constituent elements, it could be expected that SeS₂ would also exhibit superconductivity in these phases. The resistance curves presented in Fig. 6(a) show clear metallic behavior within phase IV and phase V. At 79 GPa, we observe two sharp resistance drops at 10.5 and 5.1 K [light blue curve in Fig. 6(a)], while at 149 GPa, we observe similar drops at 14 and 6.6 K [dark blue curve in Fig. 6(a)]. We tentatively assign the first drop in resistance to the superconductivity onset of trace impurities of sulfur. This assignment is supported by the agreement between previous literature T_c values of elemental sulfur [see Fig. 6(b)] together with the observation of sulfur impurities in Raman measurements [3,5]. While trace impurities of sulfur were detected through Raman measurements, no trace impurities of selenium were detected either through Raman or x-ray diffraction measurements, with the bulk sample remaining SeS₂. However, the second drop in resistance observed in our study approximately matches to the extrapolated literature T_c values of elemental selenium [3,46]. If this drop in resistance is from an Se impurity, then it would be expected that the resistance of the sample would not drop to zero due to the bulk sample being SeS₂. As it cannot be completely ruled out that the superconductivity transitions observed are from either

sulfur and selenium impurities, it poses an interesting question whether SeS_2 is superconducting with T_c values close to Se or whether superconductivity is suppressed in the alloy.

D. Conclusions

Through x-ray diffraction, Raman spectroscopy, and electrical transport measurements, we observe that the structure of SeS_2 -I, which adopts an eight-atom ring configuration, distorts and undergoes band-gap closure upon compression. At pressures above 27 GPa, we observe a phase transition to metallic phase III with a tetragonal structure where atoms are arranged in squared helical chains. Upon compression above 59 GPa, we observe a transformation to a distorted rhombohedral structure (SeS_2 -IV), which we interpret to be analogous to the incommensurate phases of sulfur (S-IV) and selenium (Se-IV). This is further evidenced by a charge density wave amplitude Raman mode observed to soften with pressure. Above pressures of 116 GPa, both the AMP mode and the lattice distortion disappear, indicative of a complete transition to β -Po-type SeS_2 -V.

ACKNOWLEDGMENTS

The work was supported by the European Research Council (ERC) under the European Union's Horizon 2020 research

and innovation programme (Grant Agreement No. 948895, MetElOne), the UKRI Future Leaders fellowship, Grant No. Mrc-Mr/T043733/1, and the National Natural Science Foundation of China (NSFC) reference code W2532012. We acknowledge DESY (Hamburg, Germany), a member of the Helmholtz Association HGF, for the provision of beam line P02.2 at PETRA-III, allocated under Proposal No. I-20230232, and we would like to thank Hanns-Peter Liermann and Timofey Fedotenko for their assistance. Part of this work was performed at BL10XU of SPring-8 with the approval of the Japan Synchrotron Radiation Research Institute (JASRI) (Proposal No. 2019A1260). We thank S. Kawaguchi for the beam-line support. We acknowledge the support of the Diamond Light Source for providing the beam time at the I15 beam line. We would like to thank M. Hanfland and G. Garbarino for their help with the ID15-b beam line at ESRF allocated under Proposal No. HC4446, and M. Mezouar and B. Wehinger for their help with ID27 beam line allocated under Proposal No. HC4878.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

- [1] R. A. Boudreau and H. M. Haendler, The isostructural γ -sulfur phase of selenium-sulfur, $\text{Se}_n\text{S}_{8-n}$, *J. Solid State Chem.* **36**, 289 (1981).
- [2] S. Susarla, T. Tsafack, P. S. Owuor, A. B. Puthirath, J. A. Hachtel, G. Babu, A. Apte, B. I. Jawdat, M. S. Hilario, A. Lerna, H. A. Calderon, F. C. R. Hernandez, D. W. Tam, T. Li, A. R. Lupini, J. C. Idrobo, J. Lou, B. Wei, P. Dai, C. S. Tiwary, *et al.*, High-K dielectric sulfur-selenium alloys, *Sci. Adv.* **5**, eaau9785 (2019).
- [3] E. Gregoryanz, V. V. Struzhkin, R. J. Hemley, M. I. Eremets, H.-k. Mao, and Y. A. Timofeev, Superconductivity in the chalcogens up to multimegabar pressures, *Phys. Rev. B* **65**, 064504 (2002).
- [4] H. Luo, S. Desgreniers, Y. K. Vohra, and A. L. Ruoff, High-pressure optical studies on sulfur to 121 GPa: Optical evidence for metallization, *Phys. Rev. Lett.* **67**, 2998 (1991).
- [5] V. V. Struzhkin, R. J. Hemley, H.-k. Mao, and Y. A. Timofeev, Superconductivity at 10–17 K in compressed sulphur, *Nature (London)* **390**, 382 (1997).
- [6] Y. Akahama, M. Kobayashi, and H. Kawamura, Pressure-induced superconductivity and phase transition in selenium and tellurium, *Solid State Commun.* **84**, 803 (1992).
- [7] S. Kometani, M. I. Eremets, K. Shimizu, M. Kobayashi, and K. Amaya, Observation of pressure-induced superconductivity of sulfur, *J. Phys. Soc. Jpn.* **66**, 2564 (1997).
- [8] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/s6wf-56gj> for detailed X-ray diffraction results.
- [9] D. A. Young, *Phase Diagrams of the Elements* (University of California Press, Berkeley, 1991), p. 129.
- [10] Y. Akahama, M. Kobayashi, and H. Kawamura, Pressure-induced structural phase transition in sulfur at 83 GPa, *Phys. Rev. B* **48**, 6862 (1993).
- [11] Y. Akahama, M. Kobayashi, and H. Kawamura, Structural studies of pressure-induced phase transitions in selenium up to 150 GPa, *Phys. Rev. B* **47**, 20 (1993).
- [12] P. Rossmanith, W. Häfner, A. Wokaun, and H. Olijnyk, Phase transitions of sulfur at high pressure: Influence of temperature and pressure environment, *High Press. Res.* **11**, 183 (1993).
- [13] H. Luo, R. G. Greene, and A. L. Ruoff, β -Po phase of sulfur at 162 GPa: X-ray diffraction study to 212 GPa, *Phys. Rev. Lett.* **71**, 2943 (1993).
- [14] O. Degtyareva, E. Gregoryanz, M. Somayazulu, P. Dera, H.-k. Mao, and R. J. Hemley, Novel chain structures in group VI elements, *Nat. Mater.* **4**, 152 (2005).
- [15] O. Degtyareva, E. Gregoryanz, H.-k. Mao, and R. J. Hemley, Crystal structure of sulfur and selenium at pressures up to 160 GPa, *High Press. Res.* **25**, 17 (2005).
- [16] O. Degtyareva, E. Gregoryanz, M. Somayazulu, H.-k. Mao, and R. J. Hemley, Crystal structure of the superconducting phases of S and Se, *Phys. Rev. B* **71**, 214104 (2005).
- [17] O. Degtyareva, E. R. Hernández, J. Serrano, M. Somayazulu, H.-k. Mao, E. Gregoryanz, and R. J. Hemley, Vibrational dynamics and stability of the high-pressure chain and ring phases in S and Se, *J. Chem. Phys.* **126**, 084503 (2007).
- [18] S. M. Arveson, Y. Meng, J. Lee, and K. K. M. Lee, High-pressure melting curve of sulfur up to 65 GPa, *Phys. Rev. B* **100**, 054106 (2019).

- [19] C. Hejny, L. F. Lundegaard, S. Falconi, M. I. McMahon, and M. Hanfland, Incommensurate sulfur above 100 GPa, *Phys. Rev. B* **71**, 020101(R) (2005).
- [20] E. Y. Tonkov and E. G. Ponyatovsky, *Phase Transformations of Elements Under High Pressure* (CRC Press, Boca Raton, FL, 2004), p. 164.
- [21] Y. Ohmura, I. Yamamoto, M. Yao, and H. Endo, Structure and electronic properties of Te–Se mixtures under high pressure, *J. Phys. Soc. Jpn.* **64**, 4766 (1995).
- [22] C. Hejny and M. I. McMahon, Complex crystal structures of Te-II and Se-III at high pressure, *Phys. Rev. B* **70**, 184109 (2004).
- [23] M. I. McMahon, C. Hejny, J. S. Loveday, L. F. Lundegaard, and M. Hanfland, Confirmation of the incommensurate nature of Se–IV at pressures below 70 GPa, *Phys. Rev. B* **70**, 054101 (2004).
- [24] Y. Akahama, M. Kobayashi, and H. Kawamura, Pressure-induced metallization and structural transition of α -monoclinic and amorphous Se, *Phys. Rev. B* **56**, 5027 (1997).
- [25] H.-K. Mao, J. Xu, and P. M. Bell, Calibration of the ruby pressure gauge to 800 kbar under quasi-hydrostatic conditions, *J. Geophys. Res.* **91**, 4673 (1986).
- [26] Y. Akahama and H. Kawamura, Pressure calibration of diamond anvil raman gauge to 310 GPa, *J. Appl. Phys.* **100**, 043516 (2006).
- [27] S. M. Dorfman, V. B. Prakapenka, Y. Meng, and T. S. Duffy, Intercomparison of pressure standards (Au, Pt, Mo, MgO, NaCl and Ne) to 2.5 Mbar, *J. Geophys. Res.* **117**, B08210 (2012).
- [28] N. Hirao, S. I. Kawaguchi, K. Hirose, K. Shimizu, E. Ohtani, and Y. Ohishi, New developments in high-pressure X-ray diffraction beamline for diamond anvil cell at SPring-8, *Matter Radiat. Extremes* **5**, 018403 (2020).
- [29] S. Anzellini, A. K. Kleppe, D. Daisenberger, M. T. Wharmby, R. Giampaoli, S. Boccato, M. A. Baron, F. Miozzi, D. S. Keeble, A. Ross, S. Gurney, J. Thompson, G. Knap, M. Booth, L. Hudson, D. Hawkins, M. J. Walter, and H. Wilhelm, Laser-heating system for high-pressure X-ray diffraction at the extreme conditions beamline I15 at Diamond Light Source, *J. Synchrotron Radiat.* **25**, 1860 (2018).
- [30] G. Garbarino, M. E. Hanfland, S. Gallego-Parra, A. D. Rosa, M. Mezouar, D. Duran, K. Martel, E. Papillon, T. Roth, P. Got, and J. Jacobs, Extreme conditions X-ray diffraction and imaging beamline ID15B on the ESRF extremely brilliant source, *High Press. Res.* **44**, 199 (2024).
- [31] M. Mezouar, G. Garbarino, S. Bauchau, W. Morgenroth, K. Martel, S. Petidemange, and H. Bureau, The high flux nano-X-ray diffraction, fluorescence and imaging beamline ID27 for science under extreme conditions on the ESRF extremely brilliant source, *High Press. Res.* **44**, 171 (2024).
- [32] C. Prescher and V. B. Prakapenka, DIOPTAS: A program for reduction of two-dimensional X-ray diffraction data and data exploration, *High Press. Res.* **35**, 223 (2015).
- [33] W. Kraus and G. Nolze, POWDER CELL– A program for the representation and manipulation of crystal structures and calculation of the resulting X-ray powder patterns, *J. Appl. Crystallogr.* **29**, 301 (1996).
- [34] J. Rodríguez-Carvajal, Recent advances in magnetic structure determination by neutron powder diffraction, *Physica B* **192**, 55 (1993).
- [35] J. Tauc, in *Optical Properties of Solids*, edited by F. Abellès (North-Holland, Amsterdam, 1972), p. 277.
- [36] L. van der Pauw, A method of measuring specific resistivity and Hall effect of discs of arbitrary shape, *Philips Res. Rep.* **13**, 1 (1958).
- [37] A. C. Gallacher and A. A. Pinkerton, A redetermination of monoclinic γ -sulfur, *Acta Crystallograph. Sec. C* **49**, 125 (1993).
- [38] H. Fujihisa, Y. Akahama, H. Kawamura, H. Yamawaki, M. Sakashita, T. Yamada, K. Honda, and T. Le Bihan, Spiral chain structure of high pressure selenium-II' and sulfur-II from powder X-ray diffraction, *Phys. Rev. B* **70**, 134106 (2004).
- [39] L. Crapanzano, W. A. Crichton, G. Monaco, R. Bellissent, and M. Mezouar, Alternating sequence of ring and chain structures in sulphur at high pressure and temperature, *Nat. Mater.* **4**, 550 (2005).
- [40] X. Li, X. Huang, X. Wang, M. Liu, G. Wu, Y. Huang, X. He, F. Li, Q. Zhou, B. Liu, and T. Cui, High-pressure dissociation of selenium and tellurium, *Phys. Chem. Chem. Phys.* **20**, 6116 (2018).
- [41] O. Moulding, L. J. Conway, I. Osmond, S. Cross, A. Hermann, J. Buhot, and S. Friedemann, Mutual stabilization of charge-density-wave and monoclinic distortion in sulfur at high pressures, *Phys. Rev. Res.* **5**, 043188 (2023).
- [42] H. Z. Cummins, Experimental studies of structurally incommensurate crystal phases, *Phys. Rep.* **185**, 211 (1990).
- [43] A. Maciel and J. F. Ryan, Observation of coupled amplitude modes in the Raman spectrum of incommensurate Na_2CO_3 , *J. Phys. C* **14**, L509 (1981).
- [44] P. Dalladay-Simpson, J. Binns, M. Peña-Alvarez, M.-E. Donnelly, E. Greenberg, V. Prakapenka, X.-J. Chen, E. Gregoryanz, and R. T. Howie, Band gap closure, incommensurability and molecular dissociation of dense chlorine, *Nat. Commun.* **10**, 1134 (2019).
- [45] O. Degtyareva, M. V. Magnitskaya, J. Kohanoff, G. Profeta, S. Scandolo, M. Hanfland, M. I. McMahon, and E. Gregoryanz, Competition of charge-density waves and superconductivity in sulfur, *Phys. Rev. Lett.* **99**, 155505 (2007).
- [46] Z. Wu, T. Fedotenko, N. Giordano, and J. Kim, Superconductivity in bcc-selenium under megabar pressure, *Sci. Rep.* **15**, 11604 (2025).