

ИНСТИТУТ ЗА ФИЗИКУ			
ПРИМЉЕНО: 06-08-2025			
Рад.јед.	б р о ј	Арх.шифра	Прилог
0801-1315/1			

Научном већу Института за физику у Београду

Београд, 06.08.2025.

Предмет:

Молба за покретање поступка за стицање звања истраживач-сарадник

На основу испуњености услова предвиђених Правилником о стицању истраживачких и научних звања, прописаног од стране ресорног Министарства, молим Научно веће института за физику у Београду да покрене поступак за мој избор у звање истраживач сарадник.

У прилогу достављам:

1. Мишљење руководиоца лабораторије са предлогом комисије за избор у звање
2. Стручну биографију
3. Преглед научних активности
4. Списак научних радова
5. Потврду о статусу студента докторских студија и пријављеној теми докторске дисертације
6. Копије диплома основних и матер студија
7. Копије научних радова

С поштовањем,

Јован Благојевић

истраживач приправник



ИНСТИТУТ ЗА ФИЗИКУ			
ПРИМЉЕНО:			
Рад.јед.	б р о ј	Арх.шифра	Прилог
	0801-1315/2	06-08-2025	

Научном већу Института за физику у Београду

Предмет: Мишљење руководиоца о избору Јована Благојевића у звање истраживач сарадник

Јован Благојевић је запослен на Институту за физику у Београду. Од децембра 2022. године ангажован је у Лабораторији за квантне материјале Центра за физику чврстог стања и нове материјале, где израду докторске дисертације обавља под мојим менторством.

С обзиром на то да испуњава све услове прописане Законом о науци и истраживањима, као и Правилником о стицању истраживачких и научних звања, који је донело Министарство науке, технолошког развоја и иновација, сагласан сам са покретањем поступка и предлажем избор Јована Благојевића у звање истраживач сарадник.

За састав Комисије за избор Јована Благојевића у звање истраживач сарадник предлажем следеће чланове:

др Ана Милосављевић, научни сарадник, Институт за физику у Београду

др Ненад Лазаревић, научни саветник, Институт за физику у Београду

др Зорица Поповић, ванредни професор, Универзитет у Београду – Физички факултет



др Ненад Лазаревић
научни саветник Института за физику
руководилац Центра за физику чврстог стања
и нове материјале

Стручна биографија

Кандидат Јован Благојевић, рођен је 10.9.1996. у Добоју. Живео и одрастао у Шамцу (Република Српска, БиХ) где је завршио основну и средњу школу. Постигнути успеси на такмичењима су му донели Вукову диплому за одличан успех и примерено владање. У периоду од 2012. до 2014. године био је полазник Истраживачке станице Петница на семинару математике, а затим и информатике. По завршетку средње школе – смер гимназија, изабран је за ученика генерације.

Уписао је Физички факултет Универзитета у Београду 2015. године, смер Б – Теоријска и експериментална физика. Дипломирао је 2021. године са просечном оценом 8,67, а исте године уписује мастер академске студије на истом смеру. У периоду од фебруара до маја 2022. године обављао је стручну праксу у Лабораторији за физику чврстог стања и нове материјале Института за физику у Београду, у оквиру које је спроведен експериментални део истраживања за мастер рад. Мастер студије је завршио 26.9.2022. године са просечном оценом 10, одбранивши рад на тему „Неуређеност и електрон-фонон интеракција у $2H-TaSe_{2-x}S_x$ ($0 \leq x \leq 2$) испитивани методом Раманове спектроскопије“, под менторством др Ненада Лазаревића.

Докторске академске студије на Физичком факултету, смер Физика кондензоване материје и статистичка физика, уписао је 2022. Тему докторске дисертације „Истраживање ефеката допирања, неуређености и напрезања на динамику решетке нискодмиеензионалних материјала методом Раманове спектроскопије“ под руководством др Ненада Лазаревића одбранио је на колегијуму Физичког факултета 18.6.2025. године.

Преглед научне активности кандидата

Главна област истраживања Јована Благојевића су квази-двостепеност материјали и њихове особине, конкретно испитивање еволуције динамике решетке са температуром и екстерним пертурбацијама – неуређеност, стрејн и допирање. Поред главне области његово истраживање укључује и проучавање колективних електронских феномена попут суперпроводности и таласа густине наелектрисања.

Од децембра 2022. године запослен је као истраживач приправник у Центру за физику чврстог стања и нове материјале, Института за физику у Београду, где своје истраживање обавља у Лабораторији за квантне материјале. Тему докторске дисертације „Истраживање ефеката допирања, неуређености и напрезања на динамику решетке нискодимензионалних материјала методом Раманове спектроскопије“ под руководством др Ненада Лазаревића одбранио је на колегијуму Физичког факултета 18.6.2025. године.

Ко-аутор је једног научног рада објављен у часопису *Physical Review Materials* 2024. године. Своје радове је представљао на неколико међународних конференција: *20th YRC – Materials Science and Engineering* (2022.), *The 21st Symposium on Condensed Matter Physics* (2023.), *22nd YRC – Materials Science and Engineering* (2024.), *International Workshop Novel Superconducting Materials* (2024.), „*Advances in Solid State Physics and New Materials*“ (2025.). Такође је учествовао је у организацији међународних конференција „*The 21st Symposium on Condensed Matter Physics*“ (2023.) и „*Advances in Solid State Physics and New Materials*“ (2025.).

Био је руководиоца интерног истраживачког пројекта Института за физику за младе истраживаче у оквиру SAIGE програма – *SEED RESEARCH GRANT*, под називом „*Visualization of strain tuned nematicity in FeSe*“ (мај–септембар 2024). Учесник је пројекта Фонда за науку Дијаспора2024 „*2D-FETPD*“ (2025.-2026.)

Учествовао је у више стручних радионица и курсева, међу којима се издвајају: *Proposal Writing Workshop* (ORNL Neutron Sciences, јануар 2025), *Visualization and Analysis of Crystal Structures Using the VESTA Program* (март 2025), *Rietveld and Pair distribution function (PDF) analysis workshop* (април 2025) и SECUF Summer School (јул 2025).

Списак објављених радова кандидата

Категорија М₂₂:

1. **Blagojević, J.**, Mijin, S. D., Bekaert, J., Opačić, M., Liu, Y., Milošević, M. V., Petrovic, C., Popović, Z. V. and Lazarević, N., „Competition of disorder and electron-phonon coupling in 2H-TaSe_{2-x}S_x (0≤x≤2) as evidenced by Raman spectroscopy“. *Physical Review Materials*, 8(2), 024004. (DOI: <https://doi.org/10.1103%2FPhysRevMaterials.8.024004>)

Категорија М₃₄:

2. M. Opačić, **J. Blagojević**, S. M. Đurđić, J. Bekaert, Y. Liu, M. V. Milošević, Č. Petrović, and N. Lazarević, “Effects of structural disorder on phonon spectra of 2H-TaSe_{2-x}S_x (0≤x≤2) single crystals, In Abstract Book of *Balkan Physical Union Congress BPU11*, 2022. (Available: <https://indico.bpu11.info/event/1/book-of-abstracts.pdf>)
3. **J. Blagojević**, S. D. Mijin, J. Bekaert, M. Opačić, M. V. Milošević, Y. Liu, C. Petrovic, Z. V. Popović, and N. Lazarević, “Effect of disorder and electron-phonon interaction on 2H-TaSe_{2-x}S_x lattice dynamics probed by raman spectroscopy”, In the Abstract Book of *20th Young Researchers’ Conference Materials Science and Engineering*, Belgrade, Serbia, 2022. (Available: https://mrs-serbia.org.rs/images/20YRC/20YRC-Book_of_Abstracts.pdf)
4. **J. Blagojević**, S. D. Mijin, J. Bekaert, M. Opačić, M. V. Milošević, Y. Liu, C. Petrovic, Z. V. Popović, and N. Lazarević, “Effect of disorder and electron-phonon interaction on 2H-TaSe_{2-x}S_x lattice dynamics”, In Abstract Book of *The 21st Symposium on Condensed Matter Physics 2023*. (Available: <https://www.sfkm2023.ipb.ac.rs/wp-content/uploads/2023/06/abstractbook.pdf>)
5. **J. Blagojević**, S. D. Mijin, J. Bekaert, M. Opačić, M. V. Milošević, Y. Liu, C. Petrovic, Z. V. Popović, and N. Lazarević, “Vibrational properties of doped 2H-TaSe_{2-x}S_x samples investigated by Raman spectroscopy”, In the Abstract Book of *22nd Young Researchers’ Conference Materials Science and Engineering*, Belgrade, Serbia, 2024. (Available: <https://www.mrs-serbia.org.rs/index.php/book-of-abstracts-22yrc>)
6. T. Belojica, **J. Blagojević**, S. M. Djurdjic, A. Šolajić, J. Pešić, B. Višić, V. Damjanović, Y. Liu, C. Petrović, Z. V. Popović, A. Milosavljević, and N. Lazarević, “Study of Crystal Phases and Temperature Dependence of InSiTe₃”, In the Abstract Book of *22nd Young Researchers’ Conference Materials Science and Engineering*, Belgrade, Serbia, 2024. (Available: <https://www.mrs-serbia.org.rs/index.php/book-of-abstracts-22yrc>)
7. **J. Blagojević**, S. D. Mijin, J. Bekaert, M. Opačić, M. V. Milošević, Y. Liu, C. Petrovic, Z. V. Popović, and N. Lazarević. “Interplay of Disorder and Electron-phonon Coupling in 2H-TaSe_{2-x}S_x (0≤x≤2) Investigated by Raman Spectroscopy”, 2024. (Available: <https://www.ifw-dresden.de/ifw-institutes/iff/events/novel-superconductors-2024>)
8. **J. Blagojević**, S. D. Mijin, J. Bekaert, M. Opačić, M. V. Milošević, Y. Liu, C. Petrovic, Z. V. Popović, and N. Lazarević, “Raman spectroscopy analysis of disorder effects in 2H-TaSe_{2-x}S_x alloys”, In Abstract Book of *Advances in Solid State Physics and New Materials*, Belgrade, Serbia, May 19 – 23, 2025. (Available: <https://www.advances25.solidstate.ipb.ac.rs/wp-content/uploads/2025/05/abstractbook.pdf>)

9. A. Kanjevac, A. Milosavljević, E. Božin, J. Lazarević, **J. Blagojević**, Q. Li, and N. Lazarević, “Symmetry-resolved Raman Study of Temperature-Induced Phonon Anomalies in ZrTe_5 ”, In Abstract Book of *Advances in Solid State Physics and New Materials*, Belgrade, Serbia, May 19 – 23, 2025. (Available: <https://www.advances25.solidstate.ipb.ac.rs/wp-content/uploads/2025/05/abstractbook.pdf>)
10. A. P. Krajnc, G. Filipič, V. L. Zupan, S. D. Škapin, **J. Blagojević**, B. Višić, N. Lazarević, and M. Remškar, “ O_2 plasma modification of MoS_2 nanotubes for photocatalytic degradation of organic water pollutants”, In Abstract Book of *Advances in Solid State Physics and New Materials*, Belgrade, Serbia, May 19 – 23, 2025.
(Available: <https://www.advances25.solidstate.ipb.ac.rs/wp-content/uploads/2025/05/abstractbook.pdf>)
11. A. Milosavljević, **J. Blagojević**, T. Belojica, B. Višić, S. Đ. Mijin, M. Opačić, A. Šolajić, J. Pešić, A. Wang, Č. Petrović, R. Hackl, and N. Lazarević, “Anisotropic strain response in FeSe ”, In Abstract Book of *Advances in Solid State Physics and New Materials*, Belgrade, Serbia, May 19 – 23, 2025.
(Available: <https://www.advances25.solidstate.ipb.ac.rs/wp-content/uploads/2025/05/abstractbook.pdf>)
12. T. Belojica, A. Milosavljević, S. Đ. Mijin, **J. Blagojević**, A. Šolajić, J. Pešić, B. Višić, V. Damljanović, M. O. Ogunbunmi, S. Bobev, Y. Liu, Č. Petrović, Z. Popović, R. Hackl and N. Lazarević. “Raman Signatures of Instabilities in InSiTe_3 ”, In Abstract Book of *Advances in Solid State Physics and New Materials*, Belgrade, Serbia, May 19 – 23, 2025.
(Available: <https://www.advances25.solidstate.ipb.ac.rs/wp-content/uploads/2025/05/abstractbook.pdf>)



Република Србија
Универзитет у Београду
Физички факултет
Д.Бр.2022/8010
Датум: 05.08.2025. године

На основу члана 161 Закона о општем управном поступку и службене евиденције издаје се

УВЕРЕЊЕ

Благојевић (Стојан) Јован, бр. индекса 2022/8010, рођен 10.09.1996. године, Добој, Босна и Херцеговина, уписан школске 2024/2025. године, у статусу: финансирање из буџета; тип студија: докторске академске студије; студијски програм: Физика.

Према Статуту факултета студије трају (број година): три.
Рок за завршетак студија: у двоструком трајању студија.

Ово се уверење може употребити за регулисање војне обавезе, издавање визе, права на дечији додатак, породичне пензије, инвалидског додатка, добијања здравствене књижице, легитимације за повлашћену возњу и стипендије.

Овлашћено лице факултета



Република Србија
Универзитет у Београду
Физички факултет
Д.Бр.2022/8010
Датум: 05.08.2025. године

На основу члана 161 Закона о општем управном поступку и службене евиденције издаје се

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Овлашћено лице факултета





ДОКТОРСКЕ СТУДИЈЕ

ПРЕДЛОГ ТЕМЕ ДОКТОРСКЕ ДИСЕРТАЦИЈЕ КОЛЕГИЈУМУ ДОКТОРСКИХ СТУДИЈА

III колпска година
2024/2025

Подаци о студенту

Име

ЈОВАН

Презиме

БЛАГОЈЕВИЋ

Број индекса

8010/2022

Научна област дисертације

ФИЗИКА КОНДЕНЗОВАНЕ МАТЕРИЈЕ

Подаци о ментору докторске дисертације

Име

НЕНАД

Презиме

ЛАЗАРЕВИЋ

Научна област

ФИЗИКА КОНДЕНЗОВАНЕ МАТЕРИЈЕ

Звање

НАУЧНИ САВЕТНИК

Институција

ИНСТИТУТ ЗА ФИЗИКУ БЕОГРАД

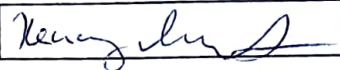
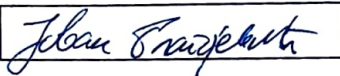
Предлог теме докторске дисертације

Наслов

ИСТРАЖИВАЊЕ ЕФЕКТА ДОПЦРАЊА, НЕУРЕЂЕНОСТИ И НАПРЕЗАЊА
НА ДИНАМИКУ РЕШЕТКЕ НИСКОДИМЕНЗИОНАЛНИХ МАТЕРИЈАЛИ
МЕТОДОМ РАМАНОВЕ СПЕКТРОСКОПИЈЕ

Уз пријаву теме докторске дисертације Колегијуму докторских студија, потребно је приложити следећа документа:

1. Семинарски рад (дужине до 10 страница)
2. Кратку стручну биографију писану у трећем лицу јединине
3. Фотокопију индекса са докторских студија

Потпис ментора	
Потпис студента	
Датум	<div>25.4.2025.</div>

Мишљење Колегијума докторских студија	
Након образложења теме докторске дисертације Колегијум докторских студија је тему	
прихватио <input checked="" type="checkbox"/>	није <input type="checkbox"/> хватио <input type="checkbox"/>
Датум	Продекан за науку Физичког факултета
<div>18.6.2025.</div>	<div>Јован Тоџић</div>



Република Србија
Универзитет у Београду

Оснивач: Република Србија

Дозволу за рад број 612-00-02666/2010-04 од 12. октобра 2011.
године је издало Министарство просвете и науке Републике Србије

Физички факултет, Београд

Оснивач: Република Србија

Дозволу за рад број 612-00-02409/2014-04 од 8. септембра 2014. године је издало
Министарство просвете, науке и технолошког развоја Републике Србије



УБ

Диплома

Јован, Стојан, Благојевић

рођен 10. септембра 1996. године, Добој, Босна и Херцеговина, уписан школске 2015/2016.

године, а дана 30. септембра 2021. године завршио је основне академске студије,
првог степена, на студијском програму Теоријска и експериментална физика, обима
240 (двеста четрдесет) бодова ЕСПБ са просечном оценом 8,67 (осам и 67/100).

На основу тога издаје му се ова диплома о стицању високог образовања и стручном називу
дипломирани физичар

Број: 12839000

У Београду, 1. новембра 2021. године

Декан
Проф. др Иван Белча

Ректор
Проф. др Владан Ђокић

00128589



Република Србија
Универзитет у Београду

Оснивач: Република Србија

Дозволу за рад број 612-00-02666/2010-04 од 12. октобра 2011.
године је издало Министарство просвете и науке Републике Србије

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Competition of disorder and electron-phonon coupling in $2H\text{-TaSe}_{2-x}\text{S}_x$ ($0 \leq x \leq 2$) as evidenced by Raman spectroscopy

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The vibrational properties of $2H\text{-TaSe}_{2-x}\text{S}_x$ ($0 \leq x \leq 2$) single crystals were probed using Raman spectroscopy and density functional theory calculations. The end members revealed two out of four symmetry-predicted Raman active modes, together with the pronounced two-phonon structure, attributable to the enhanced electron-phonon coupling. Additional peaks become observable due to crystallographic disorder for the doped samples. The evolution of the E_{2g}^2 mode Fano parameter reveals that the disorder has weak impact on electron-phonon coupling, which is also supported by the persistence of two-phonon structure in doped samples. As such, this research provides thorough insights into the lattice properties, the effects of crystallographic disorder on Raman spectra, and the interplay of this disorder with the electron-phonon coupling in $2H\text{-TaSe}_{2-x}\text{S}_x$ compounds.

I. INTRODUCTION

Transition metal dichalcogenides, a well studied family of quasi-2D materials, have attracted considerable attention in the recent years due to their rich phase diagrams, thickness-dependent transport, unique optical properties and collective electron phenomena (e.g. charge density waves and superconductivity) [1–5]. Since the experimental confirmation of the coexistence of superconductivity (SC) and charge density waves (CDW) transition metal dichalcogenides have established themselves as the ideal candidates for their investigation, given that these phenomena arise at experimentally accessible temperatures in them [6–8].

Previous experimental research has shown that at room temperature both the $2H\text{-TaS}_2$ and the $2H\text{-TaSe}_2$ crystallize into the hexagonal structure, described by the space group $P6_3/mmc$ (D_{6h}) [9, 10]. The opulent phase diagram of $2H\text{-TaSe}_2$ includes numerous charge density wave (CDW) phases at high temperatures – the incommensurate CDW (ICCDW) phase at $T_{1C} = 122$ K, the single commensurate CDW (SCCDW) phase in the temperature range from $T_{\downarrow 2C} = 112$ K to $T_{\uparrow 2C} = 90$ K, and the triply commensurate (TCCDW) phase at $T_{3C} = 90$ K [9–12]. As for $2H\text{-TaS}_2$, transition from the normal (metallic) to the ICCDW phase occurs at $T_{CDW} = 78$ K [13]. These materials exhibit unusually large Raman two-phonon scattering cross section, often correlated with the existence of CDW phase [10, 13–15]. Two-phonon fea-

ture in $2H\text{-TaS}_2$ was attributed to second-order scattering of acoustic and quasi-acoustic modes near the $q_{CDW} \cong \frac{2}{3}\Gamma\text{M}$ [16].

The latest experimental results indicate that the substitution of Se atoms with S atoms leads to a weak double dome evolution of the superconducting critical temperature T_{SC} . The T_{SC} dependence coincides with the evolution of crystallographic disorder, suggesting that the crystalline disorder favors superconductivity while suppressing the CDW phase [17]. Similarly, other types of disorder generated by etching nanopores in monolayer TaS_2 sheets [18], or by spontaneous filling of vacant sulfur sites by oxygen in few-layer $2H\text{-TaS}_2$ samples [19] enhance superconductivity in this system. In addition, first-principles calculations have revealed that the electron-phonon coupling within TaS_2 sheets is drastically boosted (by oxygenation up to 80%) [19], providing additional pathway to enhance superconductivity by doping, while suppressing the CDW state.

In this work, we present a Raman spectroscopy study of $2H\text{-TaSe}_{2-x}\text{S}_x$ ($0 \leq x \leq 2$) alloys. Obtained experimental results were found to be in good agreement with the density functional theory (DFT) calculations. The experimental Raman spectra of the end compounds host two out of the four symmetry-predicted Raman active modes. Additionally, a low-intensity overtone peak O_1 obeying pure A_{1g} selection rules can be observed only in the spectra of $2H\text{-TaS}_2$. The origin of the two-phonon structure is attributed to enhanced electron-phonon cou-

pling within phonon branches around the M - and L -points. In the spectra of doped samples additional peak and a dynamic evolution of the two-phonon structure are observed due to crystallographic disorder. Our analysis of the inverse Fano parameter $1/|q|$ of E_{2g}^2 mode indicates a weak impact of crystallographic disorder on electron-phonon coupling.

II. EXPERIMENTAL AND COMPUTATIONAL DETAILS

The preparation of the single crystal $2H$ -TaSe $_2$ - x S $_x$ alloys used in this study is described elsewhere [17]. The Raman experiment was performed using a Tri Vista 557 spectrometer with a 1800/1800/2400 grooves/mm diffraction grating combination in a backscattering configuration. As an excitation source, the 514 nm line of a Coherent Ar⁺/Kr⁺ ion laser was used. The direction of the incident (scattered) light coincides with the crystallographic c axis. Laser beam focusing was achieved through a microscope objective with 50 \times magnification. During the measurements the samples were placed inside of a KONTI CryoVac continuous helium flow cryostat with 0.5 mm thick window. All samples were cleaved in the air before being placed into the cryostat. The obtained Raman spectra were corrected by the Bose factor and presented with linear scale. The spectrometer resolution is comparable to the Gaussian width of 1 cm⁻¹.

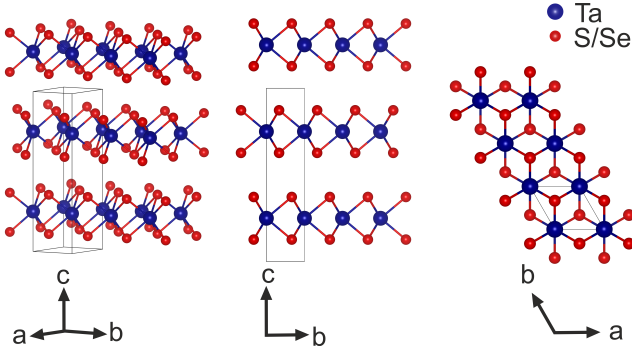


FIG. 1: Schematic representation of crystal structure of $2H$ -TaSe $_2$ ($2H$ -TaS $_2$) in various orientations.

Next, we have performed density functional theory (DFT) calculations as implemented in the ABINIT package [20]. We have used the Perdew-Burke-Ernzerhof (PBE) functional, an energy cutoff of 50 Ha for the plane-wave basis, and included spin-orbit coupling by means of fully relativistic Goedecker pseudo-potentials [21, 22], where Ta-5d³6s², S-3s²3p⁴, and Se-4s²4p⁴ states are treated as valence electrons. The crystal structure was relaxed so that forces on each atom were below 0.05 meV/Å and the total stress on the unit cell below 0.1 bar. This relaxation yields lattice parameters $a = 3.39$ Å, $c = 14.00$ Å for TaS $_2$, and $a = 3.51$ Å, $c = 14.37$ Å for TaSe $_2$. Subsequently, the phonons and

the electron-phonon coupling (EPC) were obtained from density functional perturbation theory (DFPT) calculations, also within ABINIT [23]. Here, we have used a $16 \times 16 \times 6$ \mathbf{k} -point grid for the electron wave vectors and an $8 \times 8 \times 3$ \mathbf{q} -point grid for the phonon wave vectors. For the electronic occupation we employed Fermi-Dirac smearing with broadening factor $\sigma_{FD} = 0.01$ Ha (sufficiently high to exclude unstable phonon modes related to the low-temperature CDW phases).

The phonon-wave-vector(\mathbf{q})- and mode(ν)-resolved electron-phonon coupling, plotted in Fig. 2, was evaluated as

$$\lambda_{\mathbf{q}\nu} = \frac{2}{N_F \omega_{\mathbf{q}\nu}} \sum_{\mathbf{k}, n, m} |g_{\mathbf{k}n, \mathbf{k}+\mathbf{q}m}^{\nu}|^2 \delta(\varepsilon_{\mathbf{k}n} - \varepsilon_F) \delta(\varepsilon_{\mathbf{k}+\mathbf{q}m} - \varepsilon_F),$$

where N_F is the electronic density of states at the Fermi level (ε_F), $\omega_{\mathbf{q}\nu}$ are the phonon frequencies, $g_{\mathbf{k}n, \mathbf{k}+\mathbf{q}m}^{\nu}$ the electron-phonon coupling matrix elements obtained from DFPT calculations, with n and m electronic band indices, and $\varepsilon_{\mathbf{k}n}$ ($\varepsilon_{\mathbf{k}+\mathbf{q}m}$) the electronic eigenvalue for band n (m) and electronic wave vector \mathbf{k} ($\mathbf{k}+\mathbf{q}$) obtained from DFT calculations.

III. RESULTS AND DISCUSSION

A. $2H$ -TaSe $_2$ and $2H$ -TaS $_2$

All single crystal alloys of $2H$ -TaSe $_2$ - x S $_x$ ($0 \leq x \leq 2$) crystallize into $P6_3/mmc$ crystal structure (Fig. 1) [9, 10]. Wyckoff positions of atoms and their contributions to the Γ -point phonons together with corresponding Raman tensors for the two end compounds are listed in Table I.

In total, the symmetry analysis predicts four Raman-active modes ($A_{1g} + E_{1g} + 2E_{2g}$) for the two end compounds of $2H$ -TaSe $_2$ - x S $_x$. According to the Raman tensors presented in Table I, A_{1g} modes can be observed only in parallel polarization configuration, whereas E_{2g} modes can be observed in spectra measured both in parallel and crossed polarization configurations. For our backscattering configuration, where laser beam is focused along the c -axis onto ab plane, the E_{1g} mode is unobservable.

Raman spectra of the two end compounds $2H$ -TaSe $_2$ and $2H$ -TaS $_2$ measured at $T = 150$ K are presented in Fig. 2 (a) and (c), respectively. This particular temperature has been chosen because it is significantly higher than the critical temperature of the CDW phase transitions. The Raman spectra of the pristine samples exhibit two prominent peaks, assigned as A_{1g} and E_{2g}^2 symmetry modes, accompanied by a broad structure and an additional peak in the case of $2H$ -TaS $_2$. For clarity, Raman-active modes observed in $2H$ -TaS $_2$ spectra are marked with asterisk. Theoretically predicted modes are first analyzed, after which we address the origin and nature of additional peak and broad structure.

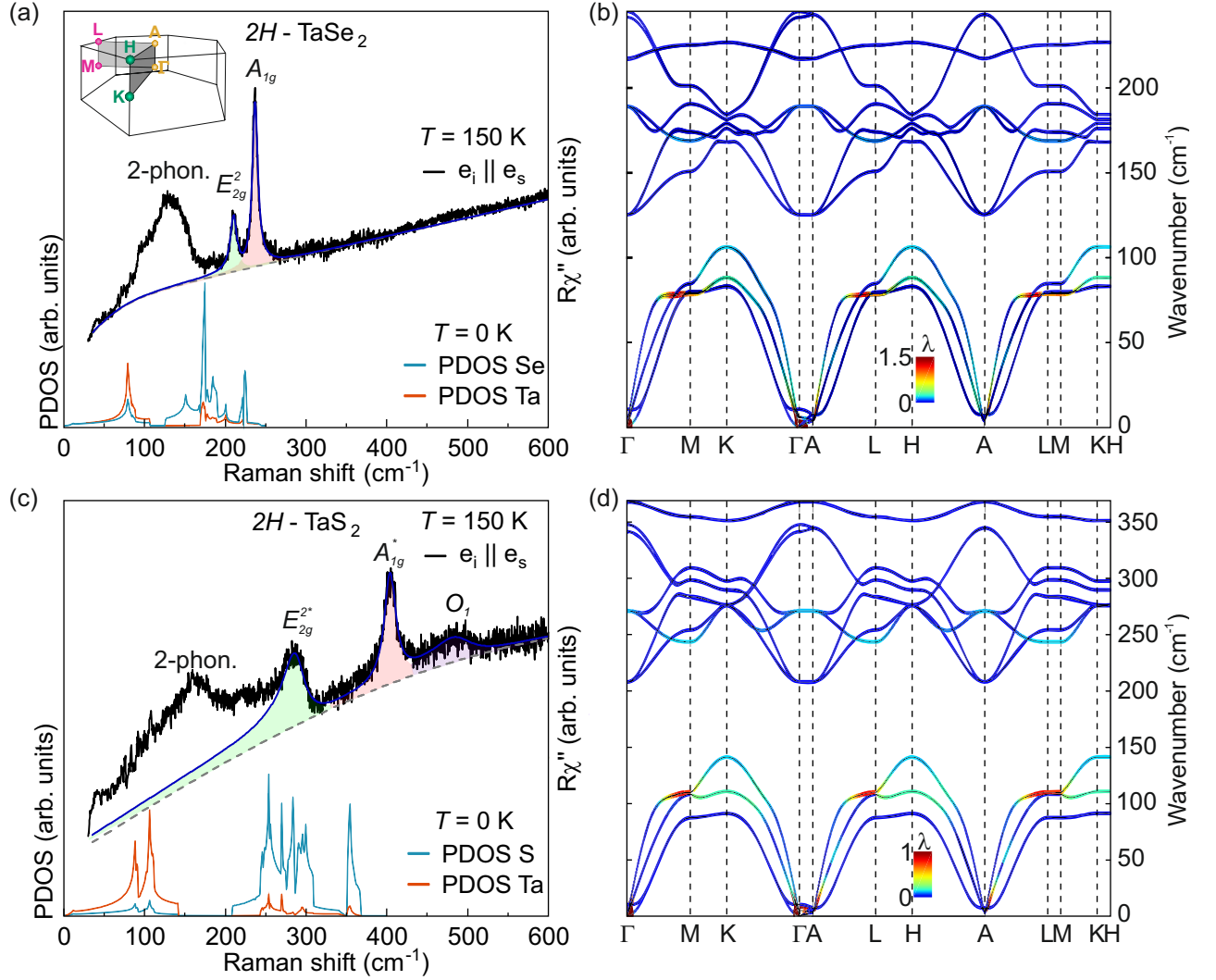


FIG. 2: (a) and (c) Raman response of $2H\text{-TaSe}_2$ and $2H\text{-TaS}_2$ for parallel polarization configuration at $T = 150\text{ K}$. The blue solid line represents the cumulative fit consisting of electronic continuum (dashed line) and phonon modes. The electronic continuum was modeled using simplified approach as described in [24], utilizing the following function: $R''_{\chi}(\omega, T) = a(T) \cdot \tanh\left(\frac{\omega}{c(T)}\right) + b(T) \cdot \frac{\omega}{c(T)}$. Here $a(T)$, $b(T)$ and $c(T)$ are temperature dependent parameters determined for each sample. Phonon modes were fitted using Fano line profile. Parts of spectra not covered by cumulative fit were identified to originate from two-phonon scattering. The contributions of the Ta (orange) and Se/S atoms (blue) to the calculated PDOS are shown as insets. First Brillouin zone with indicated points and lines of high symmetry is also shown in the inset of (a). (b) and (d) The calculated phonon dispersion of corresponding pure samples in the normal phase, with the value of electron-phonon coupling constant (λ) indicated through the color scale.

The Raman responses of both samples were analyzed with a cumulative fit to include electronic continuum and discrete single-phonon excitations. The Breit-Wigner-Fano profile was used to model single-phonon excitations, due to visible asymmetry of the E_{2g}^2 modes. For simplicity, the A_{1g} modes were fitted with constant Fano parameter $|q| = 50$, as they are highly symmetric. The phonon energies obtained in this way, alongside the calculated ones are listed in Table II. The lower frequency E_{2g}^1 modes are related to shear (S) movements of the layers, with frequencies of $\sim 5\text{ cm}^{-1}$ for both compounds,

whereas B_{1g}^1 modes represent the layer breathing (LB) and are expected at frequencies $\sim 10\text{ cm}^{-1}$.

As can be seen in Table II, the discrepancy in experimental and theoretical phonon energy is less than 10 % for all observed modes. The E_{2g}^1 modes were not observed in the investigated energy region in accordance to our numerical calculations and are not in the focus of this study. The phonon dispersions, obtained from the DFPT calculations, are shown in Fig. 2 (b) and (d), together with the calculated values of the EPC constant λ . These indicate the presence of non-zero EPC in E_{2g}^2

TABLE I: Wyckoff positions of atoms and their contributions to the Γ -point Raman active phonons for the $P6_3/mmc$ space group of $2H$ -TaSe₂ (and $2H$ -TaS₂) together with the corresponding Raman tensors.

Space group: $P6_3/mmc$ (194)	
Atoms	Irreducible representations
Ta ($2b$)	E_{2g}
Se/S ($4f$)	$A_{1g} + E_{1g} + E_{2g}$
Raman tensors	
$A_{1g} = \begin{pmatrix} a & 0 & 0 \\ 0 & a & 0 \\ 0 & 0 & b \end{pmatrix}$	
${}^1E_{1g} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & c \\ 0 & c & 0 \end{pmatrix} \quad {}^2E_{1g} = \begin{pmatrix} 0 & 0 & -c \\ 0 & 0 & 0 \\ -c & 0 & 0 \end{pmatrix}$	
${}^1E_{2g}^2 = \begin{pmatrix} d & 0 & 0 \\ 0 & -d & 0 \\ 0 & 0 & 0 \end{pmatrix} \quad {}^2E_{2g}^2 = \begin{pmatrix} 0 & -d & 0 \\ -d & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$	

TABLE II: Phonon symmetries and phonon energies (in units of cm^{-1}) at Γ and their degeneracies for the $P6_3/mmc$ structure of $2H$ -TaS₂ and $2H$ -TaSe₂. The experimental values were determined at 300 K with experimental uncertainty of 0.3 cm^{-1} . The DFPT calculations were performed at zero temperature. In case of the lowest two modes, ‘S’ stands for shear and ‘LB’ for layer breathing.

Symm.	Deg.	$2H$ -TaS ₂		$2H$ -TaSe ₂	
		Calc.	Exp.	Calc.	Exp.
E_{2g}^1 (S)	2	4.9		5.5	
B_{1g}^1 (LB)	1	10.4		10.7	
E_{2u}	2	208.0		125.4	
E_{1g}	2	208.1		125.4	
E_{2g}^2	2	271.3	288.8	189.1	207.7
E_{1u}	2	271.3		189.2	
A_{2u}	1	341.6		241.8	
B_{1g}^2	1	348.0		245.1	
B_{1u}	1	368.0		216.9	
A_{1g}	1	368.5	402.9	217.7	234.7

scattering channel, giving rise to asymmetric line profiles. On the other hand in the A_{1g} channel no EPC was found, thus they should have symmetric profiles, as we have observed.

In addition to the observed Raman-active modes, the spectra of $2H$ -TaS₂ in the parallel polarization configuration host the O_1 peak, with energy at about $\sim 490 \text{ cm}^{-1}$. This peak cannot be explained in terms of the first-order Raman scattering as its energy is well beyond

the phonon energy range (see Fig. 2 (d)). Despite this, we have included this peak in the cumulative fit in order to track its energy. Previously reported large two-phonon scattering cross section in this material [10, 13–16] indicates a second-order Raman scattering, which might occur due to defect and/or enhanced EPC. The absence of additional first-order peaks at energies with the highest phonon density of states (PDOS) values contradicts the expectations for defect-induced peaks [25]. Considering that one of the PDOS maxima ($\sim 250 \text{ cm}^{-1}$) is located near the energies that correspond to the half energy of O_1 peak (Fig. 2 (c)), we believe that this peak is an overtone in nature and is observable due to enhanced EPC. To determine the validity of this assumption, the phonon dispersion curves and electron-phonon coupling constant λ (Fig. 2 (d)) were further examined. If the O_1 peak is, indeed, observable due to electron-phonon coupling, we would expect that optical phonon branches around energies $\omega_{O_1}/2$ express high values of λ . As it is indicated in Fig. 2 (d), several optical branches along the lines of high symmetry, at energies just below 250 cm^{-1} , meet this condition thus supporting our assumption.

The broad structures in the spectra of $2H$ -TaSe₂ and $2H$ -TaS₂, with energies at about $\sim 130 \text{ cm}^{-1}$ and $\sim 160 \text{ cm}^{-1}$, respectively, are centered in the gap of the theoretical PDOS, thus not a result of the first-order Raman scattering. Considering previous discussion on the origin of the O_1 peak in $2H$ -TaS₂ spectra and applying similar analysis, these structures can likewise be explained as a two-phonon processes, predominantly overtones in nature but also with combinations contribution (as can be deduced from Fig. 5), also originating from the enhanced electron-phonon coupling. As shown in Fig. 2, peaks in PDOS values can be found in the $\omega_{2phon}/2$ energy ranges. By examining the phonon dispersion curves of $2H$ -TaS₂ and $2H$ -TaSe₂ (Fig. 2), several phonon branches with enhanced EPC are in the appropriate energy range for the two-phonon process. In the case of $2H$ -TaS₂, these branches fall within the energy range of $100 - 150 \text{ cm}^{-1}$, while for $2H$ -TaSe₂, they lie in the range of $50 - 100 \text{ cm}^{-1}$. It was reported in a previous study [16] that the origin of two-phonon structure in $2H$ -TaS₂ is in the vicinity of the CDW wave vector $q_{CDW} \cong \frac{2}{3}\Gamma M$, where strong EPC exists. We argue that the origin of large scattering cross section for the two-phonon process is enhanced EPC in a much larger phase space that is required to reproduce the high Raman cross-section of the observed structures.

The phonon branches along (and in the vicinity) the lines of high symmetry: $\Gamma - M$, $A - L$, and $L - M$ (illustrated in Fig. 2), demonstrate the highest values of the electron-phonon coupling constant, and we believe that these regions of phase space contribute to the two-phonon process. As anticipated, these branches closely coincide with energy levels that correspond to $\omega_{2phon}/2$, since the major contribution lies in the A_{1g} symmetry corresponding to the overtones (see Appendix A). This provides a strong argument to support our assumptions, regarding the na-

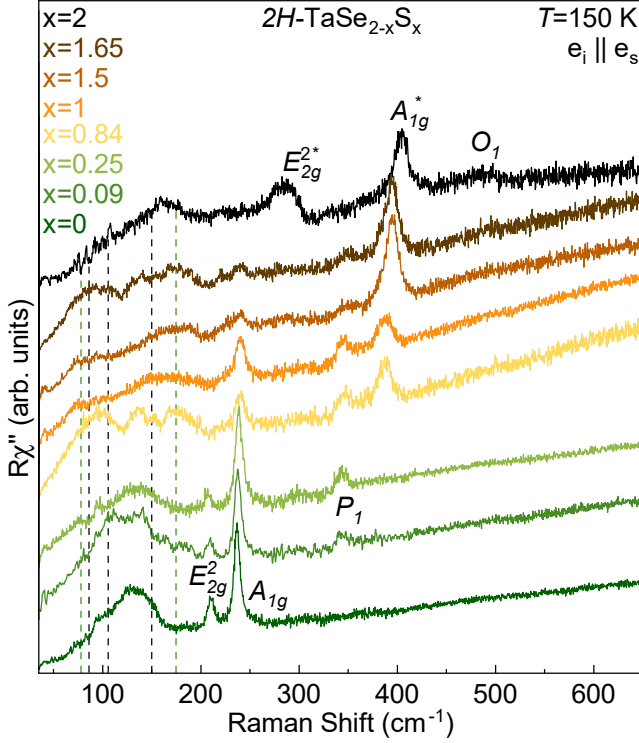


FIG. 3: Raman spectra of $2H\text{-TaSe}_{2-x}\text{S}_x$ measured at 150 K in parallel polarization configuration. Additionally to the A_{1g} , A_{1g}^* , E_{2g}^2 and E_{2g}^{2*} modes, one peak assigned as P_1 and PDOS projection onto two-phonon structure arise in the spectra of doped materials. With dashed lines are marked places of PDOS maxima in both pure samples. Spectra contain offset in y-direction for clarity.

ture and the origin of the two-phonon structures. Significantly higher intensity of the two-phonon structure in $2H\text{-TaS}_2$, compared to the O_1 peak, can be attributed to the stronger EPC observed in the corresponding phonon branches, as supported by our DFT calculations.

B. $2H\text{-TaSe}_{2-x}\text{S}_x$ ($0 \leq x \leq 2$)

Raman spectra of the $2H\text{-TaSe}_{2-x}\text{S}_x$ ($0 \leq x \leq 2$) single crystals measured in parallel polarization configuration at $T = 150$ K are presented in Fig. 3. The two modes that are present in the spectra of pristine $2H\text{-TaSe}_2$ - A_{1g} and E_{2g}^2 , can also be observed in the spectra of the doped samples. The A_{1g} symmetry mode persists in the spectra up to $2H\text{-TaSeS}$. As x increases further we cannot be certain that the existing feature in spectra is A_{1g} mode, because of low Se content and possible $2H\text{-TaS}_2$ PDOS contributions. Conversely, E_{2g}^2 vanishes for $x = 0.84$, while A_{1g}^* and E_{2g}^{2*} modes can last be found in the spectra of $2H\text{-TaSe}_{1.16}\text{S}_{0.84}$ and $2H\text{-TaSe}_{0.5}\text{S}_{1.5}$, respectively. Considering that E_{2g}^2 and E_{2g}^{2*} arise from the out-of-phase in-plane oscillations of

Ta and Se/S atoms, the dependence on sulfur content x exhibits a discontinuous behavior. For the A_{1g} mode (out-of-plane movement of Se/S atoms) we observe continuous change with doping alongside simultaneous coexistence of A_{1g} and A_{1g}^* peaks in samples where Se/S concentration ratio is around unity. In the specific case of $2H\text{-TaSeS}$, at the nanoscale level, localized regions may emerge where either Se or S atoms predominate. Although these regions possess a sufficient size to generate detectable signals, they are significantly smaller in comparison to the laser spot ($\sim 5 \mu\text{m}^2$), resulting in the inclusion of a considerable number of such clusters of atoms within the laser's spatial coverage. Consequently, the resulting Raman spectrum displays discernible contributions from both elements. The concentrations at which the E_{2g}^2 mode vanishes directly correlate with disorder reaching its maximum value. Given that doping modifies the bond lengths of Ta-S and Ta-Se atoms, which, in conjunction with the accompanying lattice disorder, has a detrimental effect on the E_{2g}^2 mode.

The gradual substitution of the selenium atoms with sulfur in the $2H\text{-TaSe}_2$ crystals introduces crystallographic disorder, giving rise to new scattering channels that result in additional peak observed in the Raman spectra of doped materials. This new peak with energy at about 342 cm^{-1} , assigned as P_1 is observed exclusively in doped spectra measured in parallel polarization configuration, therefore obeying A_{1g} symmetry rules. It is presumably an overtone mode, as its energy closely matches double that of the location where the highest PDOS value of the $2H\text{-TaSe}_2$ is situated.

To further investigate the effects of crystallographic disorder, we have inspected the evolution of phonon parameters with the sulfur content x , depicted in Fig. 4. As in the case of pure samples, all phonon lines were fitted using Fano profiles. No asymmetry was observed for the A_{1g} modes and they were fitted with the value of Fano parameter q being fixed at 50. All modes but E_{2g}^2 harden with the increase in the sulfur concentration in the measured crystals. Given the difference in the atomic mass of Se and S and a reduction of the unit cell volume [17], one would expect that the E_{2g}^2 mode also hardens with increasing x [26–28]. The unexpected behavior of E_{2g}^2 mode might be attributed to the enhanced EPC, which potentially overcompensates previously mentioned effects. All Raman modes broaden due to increased crystallographic disorder. Considering that the Fano profiles are used to describe the line shape originating from coupling between phonon and electronic continuum, and that the Fano parameter depends on the interaction strength between the phonon and the continuum, the inverse value of Fano parameter $1/|q|$ can be used as a direct measure of the strength of EPC [29–32]. The interference of two-phonon structure in the fitting procedure produces large uncertainty (see Figs. 4(c)), preventing a clear assertion about the EPC, other than it persists through growing disorder. This result falls within the range of the potential increase in disorder-induced EPC [17].

Crystallographic disorder also has a significant impact on the evolution of two-phonon structure. Changing the sulfur content to $x = 0.09$, the broad structure in pristine $2H$ -TaSe₂ spectra centered at around 130 cm^{-1} undergoes a further broadening accompanied by pronounced evolution of the shoulder at around $\sim 110 \text{ cm}^{-1}$, likely due to more pronounced evolution of the EPC in the related phonon branches. Raising the sulfur content to 0.25 and thus further increasing disorder restores the structure to a solitary broad peak. However, the most drastic change is observed in the spectra when $x = 0.84$, where four first order peaks are superimposed onto a broad two-phonon structure. As discussed previously, the additional peaks might become observable in Raman experiment due to defect scattering and are related to the regions where the PDOS reaches its maximum: at $\sim 90 \text{ cm}^{-1}$ and $\sim 110 \text{ cm}^{-1}$ for $2H$ -TaSe₂, at $\sim 80 \text{ cm}^{-1}$ and $\sim 175 \text{ cm}^{-1}$ for $2H$ -TaSe₂ (see Fig. 2). These correspond rather well with the energies of the newly observed phonon modes. Raising the concentration of sulfur to $x = 1$ results in a wholly diluted structure consisting of two broad peaks at around $\sim 80 \text{ cm}^{-1}$ and $\sim 160 \text{ cm}^{-1}$. In the spectra with sulfur content $x = 1.65$, structure resembles that of the $x = 0.84$, albeit of less clarity among peaks. From these observations, we can see that the primary impact of disorder on the two-phonon structure is manifested by the broadening of peaks and the projection of the PDOS. Furthermore, the persistence of the two-phonon structure across doped samples suggests that disorder does not hinder the electron-phonon coupling. This corollary aligns with previous observations regarding the $1/|q|$ parameter of the E_{2g}^2 mode.

Our results reveal the origin of two-phonon structures in the $2H$ -TaSe_{2-x}S_x compounds. It is also evident that, although disorder does not directly influence the strength of the EPC (in the range from $x = 0.25$ to $x = 1.65$), it affects the phenomena that share EPC as a seemingly common origin differently. While the CDW is destroyed, SC and the two-phonon structure survive [17] suggesting that strong EPC alone is insufficient to sustain the CDW in a disordered lattice. It is interesting to note that these materials simultaneously and independently experience the effects of disorder and EPC.

IV. CONCLUSIONS

In this study, we conducted a Raman scattering analysis of $2H$ -TaSe_{2-x}S_x ($0 \leq x \leq 2$) alloys. The Raman spectra of the end compounds host two out of the three symmetry-expected Raman active modes for backscattering configuration and an additional peak O_1 of A_{1g} symmetry, present only in the $2H$ -TaS₂ spectra. This O_1 peak is a result of second-order Raman scattering, overtone in nature, observable due to the prominent EPC, characteristic for CDW materials. The broad two-phonon structures observed in pristine samples were linked to enhanced EPC in the corresponding phonon

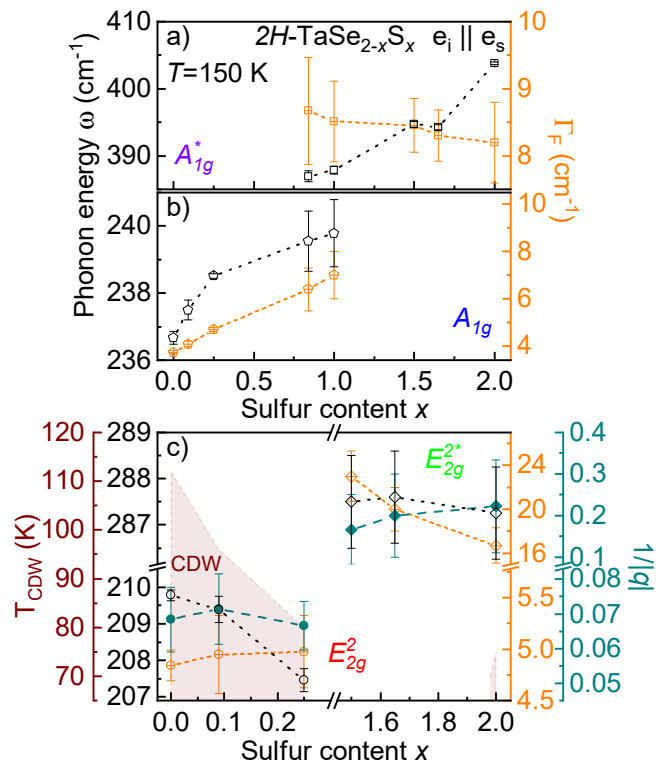


FIG. 4: The evolution of phonon energies (black) and line widths (orange) of the (a) A_{1g}^* and (b) A_{1g} Raman-active modes. (c) The evolution of phonon energies, line widths and inverse value of Fano parameter $1/|q|$ (teal) of the E_{2g}^{2*} and E_{2g}^2 modes with the sulfur content x . T_{CDW} (dark red) dependence on the sulfur content x (obtained from electrical resistivity measurements presented in Ref. [17]), together with the electronic phase diagram (shaded red) of $2H$ -TaSe_{2-x}S_x reflect the evolution of crystallographic disorder.

branches, as obtained from our first-principles calculations. The gradual substitution of Se atoms with S atoms results in crystallographic disorder, introducing new scattering channels – in the form of an additional peak P_1 of A_{1g} symmetry, as well as PDOS projection and broadening of all phonon modes. Dependence on sulfur content showed hardening of all modes, except for E_{2g}^2 mode. Softening of the E_{2g}^2 mode and its discontinuous dependence were attributed to strong EPC. Coexistence of A_{1g} peaks in intermediate doping levels arises probably due to nanoscale clusters comprised mainly of either Se or S atoms. In spectra where disorder reaches its maximum, four single-phonon peaks accompanied the background two-phonon structure, with energies corresponding to the region of PDOS maxima. The negligible influence of disorder on the EPC was supported by the continued presence of the two-phonon structure and the behavior of the $1/|q|$ parameter associated with the E_{2g}^2 mode. Thus, in the absence of changes in either electron-electron [17] or EPCs, significant rise of T_{SC} in doped alloys is only

due to disorder-induced CDW suppression. Our findings provide insights into the intricate relationship between disorder and EPC, shedding light on their combined influence in governing the vibronic and collective electronic behavior of $2H\text{-TaSe}_{2-x}\text{S}_x$ compounds.

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Appendix A: Mode assignment and nature of additional peaks

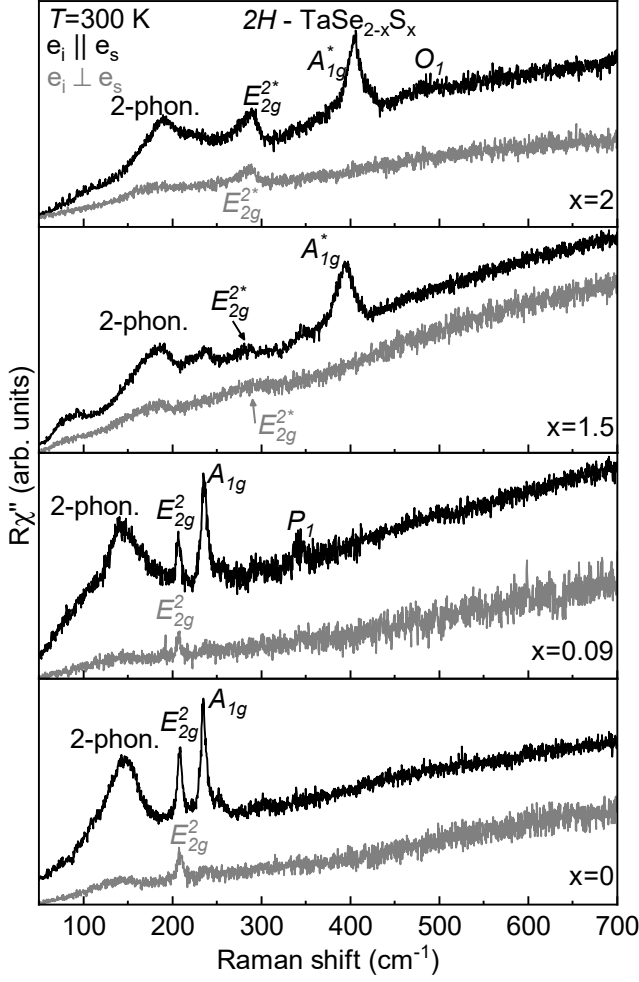


FIG. 5: Raman response of $2H\text{-TaSe}_2$, $2H\text{-TaSe}_{1.5}\text{S}_{0.5}$, $2H\text{-TaSe}_{1.91}\text{S}_{0.09}$ and $2H\text{-TaS}_2$ for parallel (black) and crossed (gray) polarization configurations at $T = 300$ K. The possible overtone modes O_1 and P_1 are only present in parallel polarization configuration.

Raman spectra of all samples were measured in parallel and crossed polarization configurations in order to correctly assign theoretically predicted modes as well as the unexpected and additional peaks and structures. From the Raman spectra presented in Figure 5 theoretically predicted A_{1g} and E_{2g}^2 modes were easily identified. Given that the O_1 and P_1 peaks are only present in parallel polarization configuration, they obey pure A_{1g} symmetry rules. Two-phonon structure is present in both polarization configurations, but with significantly higher intensity in A_{1g} channel. Additional features in two-phonon structure in the spectra of $2H\text{-TaSe}_{1.5}\text{S}_{0.5}$ are only present in parallel polarization configuration, thus they are possibly overtones in nature or single-phonon

excitations of A_{1g} symmetry.

Appendix B: Fitting details

All peaks were fitted using both asymmetric Fano profiles and symmetric Voigt profiles. Since the line widths of the analyzed phonon modes were much greater than the resolution of spectrometer σ , the real width of the peaks could be obtained without the need to use Fano profiles convoluted with a Gaussian function, where $\Gamma_L = \sigma$. The comparison between the obtained fits are presented in Figure 6. As it can be seen, peaks E_{2g}^2 and E_{2g}^{2*} show clear asymmetric line shape, whereas the A_{1g} symmetry peaks do not. Therefore, the latter were fitted with Fano parameter value $|q|$ being fixed at 50. Acquired phonon parameters as a function of sulfur content x are presented in Figures 4.

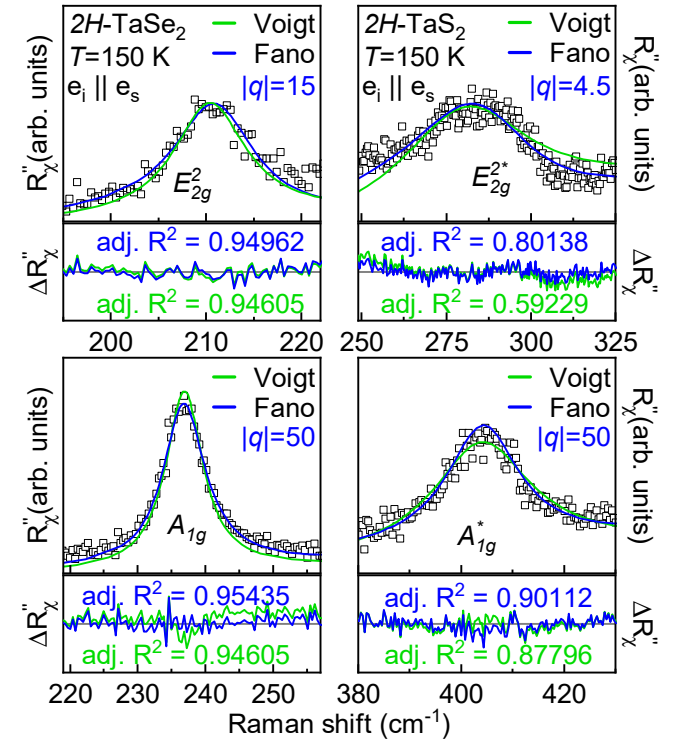


FIG. 6: Raman response as a function of the Raman shift. Quantitative analysis of the E_{2g}^2 , E_{2g}^{2*} , A_{1g} and A_{1g}^* modes at $T = 150$ K. The blue and green solid lines represent Fano and Voigt profiles fitted to the experimental data, respectively. Experimental data is represented by open squares.

The increase in doping leads to the significant decrease in the relative intensity of E_{2g}^2 Raman active modes, therefore the obtained phonon parameters have slightly higher error bar compared to the A_{1g} modes. Fits obtained using Fano profiles in the energy range of peaks E_{2g}^2 and E_{2g}^{2*} are presented in Figure 7. The two-phonon structure interferes with the lower energy side of the peaks in the spectra of all doped samples, contributing to the mentioned error.

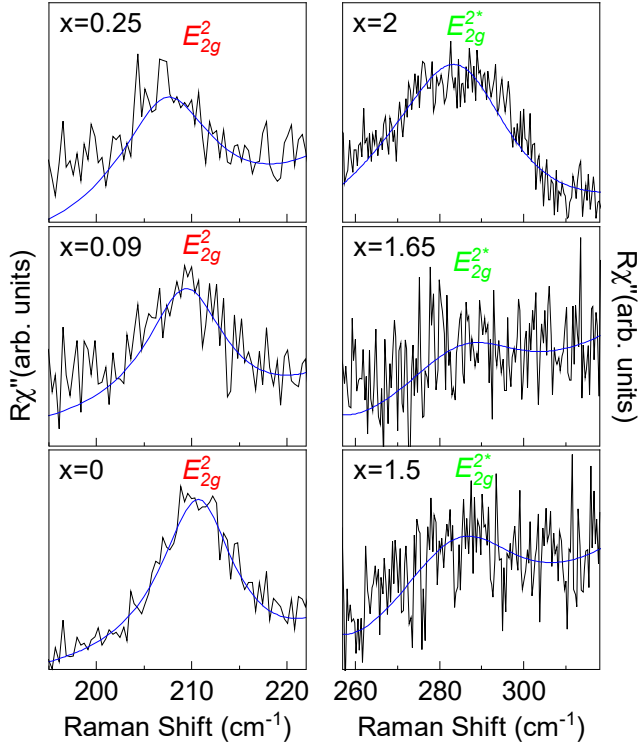


FIG. 7: Raman response as a function of the Raman shift. Quantitative analysis of the E_{2g}^2 and E_{2g}^{2*} modes for indicated sulfur content x in the measured crystals. The blue lines represent Fano profiles fitted to the experimental data.

6-7

Effect of disorder and electron-phonon interaction on $2H\text{-TaSe}_{2-x}\text{S}_x$ lattice dynamics probed by Raman spectroscopy

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Transitional metal dichalcogenides have attracted a lot of attention due to their rich phase diagrams, thickness-dependent transport, distinctive optical characteristics, and the emergence of collective electron phenomena (e. g. charge density waves - CDW and superconductivity) which can co-exist, contrary to what was predicted by previous theoretical studies. Given that both superconductivity and CDW phase have been experimentally confirmed in the crystal alloys of $2H\text{-TaSe}_{2-x}\text{S}_x$, these materials represent perfect candidates to investigate an intricate connection between these two phenomena. Additionally, it was recently shown that in the metallic single crystal alloys of $2H\text{-TaSe}_{2-x}\text{S}_x$ the crystalline disorder favours superconductivity while suppressing CDW phase. In this study, Raman spectroscopy was used to investigate the lattice dynamics of $2H\text{-TaSe}_{2-x}\text{S}_x$ ($0 \leq x \leq 2$) alloys. Experimental results were compared to density functional theory (DFT) and density functional perturbation theory (DFPT) calculations. In the Raman spectra of pristine samples two out of three symmetry predicted Raman active modes were observed, with the missing mode being unobservable in the used backscattering geometry. Experimental values of phonon energies are in good agreement with theoretical calculations. The temperature dependence of phonon energies and line widths directly reflects existing CDW transitions. The Raman spectra of doped materials were compared to those of pure samples in order to inspect how the electron-phonon interaction and crystallographic disorder affect the phonons. Additional peaks and a dramatic development of the two-phonon structure are detected in the Raman spectra of the doped samples. A signature of the crystallographic disorder can also be identified in the sulfur content dependence of phonon energies, line widths and Fano parameter.

S06-CMPSP-112 / Oral presentation

Effects of structural disorder on phonon spectra of $2H$ -TaSe_{2-x}S_x ($0 \leq x \leq 2$) single crystals

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Transition metal dichalcogenides, as a well studied family of quasi-2D materials, have attracted considerable attention in recent years due to rich phase diagrams, thickness-dependent transport, unique optical properties and collective electron phenomena which occur at experimentally accessible temperatures. Additionally, it was recently shown that in the metallic single crystal alloys of $2H$ -TaSe_{2-x}S_x the crystalline disorder promotes superconductivity, while suppressing charge density wave (CDW) order. In this work, the lattice dynamics of TaSe_{2-x}S_x ($0 \leq x \leq 2$) alloys was probed using Raman spectroscopy and results were compared to density functional theory (DFT) calculations. In order to investigate whether crystallographic disorder affects the phonons, spectra of doped materials were compared to the ones belonging to the end alloys. The Raman spectra of the end compounds ($x = 0$ and $x = 2$) host two out of three symmetry-expected Raman active modes for backscattering configuration. Calculated phonon energies agree well with the experimental ones. In Raman spectra of the doped samples additional peaks, though of low intensity, can be easily identified. These additional peaks most likely arise from the crystalline disorder. Dependence of phonon energies and linewidths on sulfur content x also reveals a clear fingerprint of crystallographic disorder.

S06-CMPSP-113 / Oral presentation

DFT study of AlF₃ intercalated in HOPG: a rechargeable battery application

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Effect of disorder and electron-phonon interaction on $2H\text{-TaSe}_{2-x}\text{S}_x$ lattice dynamics

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Abstract. Quasi-2D materials have gained a significant attention in the last few years because of their unique physical properties. The family of transition metal dichalcogenides is particularly intriguing due to their complex phase diagrams, characteristic optical properties and possibility to observe collective electron phenomena at higher temperatures. A strong correlation was observed between the electron-phonon interaction and the CDW phase in materials that display such states. Recent study has revealed that crystalline disorder promotes superconductivity while simultaneously suppresses CDW phase in metallic single crystal alloys of $2H\text{-TaSe}_{2-x}\text{S}_x$.

Raman spectroscopy was used to investigate the effect of defects on lattice dynamics and electron-phonon coupling in these materials, and the results were compared to theoretical calculations. In our scattering configuration two out of four Raman active modes predicted by symmetry for parent compounds are identified. Additionally, in the spectra of pure samples broad two-phonon structures are observed, emerging as a consequence of strong electron-phonon coupling in related phonon branches. By substituting Se with S atoms, extra peak obeying A_{1g} selection rules, overtone in nature, appears in spectra along with an intriguing evolution of two-phonon structure. The Raman spectra of the $x = 0.84$ sample shows single-phonon excitations that are superimposed on already existing structure. These excitations correspond to PDOS maxima projected due to significant crystallographic disorder. Symmetry predicted A_{1g} modes expectedly harden with doping as unit cell volume decreases, whereas E_{2g} modes exhibit anomalous behavior attributed to enhanced electron-phonon coupling. The analysis of E_{2g} mode Fano parameter indicates that disorder has a minor impact on electron-phonon interaction.

8-8

Vibrational properties of doped 2H-TaSe_{2-x}S_x samples investigated by Raman spectroscopy

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Recent research has highlighted transition metal dichalcogenides (TMDs) for their fascinating electronic and quantum properties. These materials are unique in that their phase transitions can be finely tuned through pressure, doping, or layer thickness, making them ideal for investigating phenomena such as charge ordering, spin-orbit coupling, and potential topological phases. This adaptability creates exciting opportunities for advancements in quantum materials and device technologies. Notably, experiments have demonstrated the coexistence of superconductivity (SC) and charge density waves (CDWs) with findings in 2H-TaSe_{2-x}S_x suggesting that disorder promotes SC while suppressing the CDW phase. In this study, we used Raman spectroscopy and density functional theory to explore the vibrational properties of 2H-TaSe_{2-x}S_x (0 ≤ x ≤ 2) single crystals. In undoped samples, two symmetry predicted Raman modes and a prominent two-phonon feature linked to strong electron-phonon coupling (EPC) were observed. Doping with sulfur atoms leads to the appearance of additional peaks due to crystallographic disorder. The evolution of E_{2g}² mode Fano parameter and the persistence of the two-phonon structure suggest that disorder has negligible effect on EPC. This study offers valuable insights into the lattice properties, the influence of crystallographic disorder on Raman spectra and its interplay with EPC in 2H-TaSe_{2-x}S_x compounds.

6-2

Study of crystal phases and temperature dependence of InSiTe₃

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In recent years, quasi-low-dimensional materials have attracted significant attention due to their distinctive properties and possible applications in nanoelectronics and spintronics. The material of a specific interest within this group is InSiTe₃. Unlike related compounds, such as CrSiTe₃ and CrGeTe₃, research results InSiTe₃ are limited, most likely due to the unclear nature of its crystal structure. Detailed experimental and theoretical investigation was conducted to determine the crystal structure of InSiTe₃. Inelastic light scattering experiment performed on the InSiTe₃ reveals presence of six ($3A_{1g} + 3E_g$) out of eight and seven ($5A_g + 2E_g$) out of ten Raman active modes for proposed $P\bar{3}1m$ and $P\bar{3}$ space groups, respectively. These findings suggest a coexistence of two trigonal crystal phases: a high symmetry one corresponding to $P\bar{3}1m$ and a lower symmetry one that corresponds to $P\bar{3}$ space group. Additional excitations were detected in parallel scattering configuration; two broad features in the gap of PDOS that can be a consequence of two-phonon processes and a third one, at about 500 cm⁻¹ that might indicate local symmetry breaking at nano scale. Temperature dependent measurements from 80 K to 300 K show monotonous decrease in energy and increase in linewidth up to 200 K at which point discontinuities appear across all analyzed modes. However, this anomaly overcomes the scope of this research and remains an open question.

P2	Interplay of disorder and electron-phonon coupling in 2H-TaSe_{2-x}S_x (0≤x≤2) investigated by Raman Spectroscopy <i>Jovan Blagojević, Institute of Physics Belgrade</i>
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The quasi-2D nature of layered transition metal dichalcogenides introduces novel phenomena not observed in bulk materials. Features such as complex phase diagrams, diverse electronic and optical properties, temperature-driven transitions, and strong spin-orbit and electron-phonon coupling have driven significant research interest in these materials. Recent studies reveal a correlation between electron-phonon coupling and the charge density wave (CDW) phase. In doped alloys such as 2H-TaSe_{2-x}S_x, where sulfur doping induces lattice disorder, the CDW phase is effectively suppressed, while superconductivity is enhanced.

In this study, we investigated the lattice dynamics of 2H-TaSe_{2-x}S_x (0≤x≤2) alloys using Raman spectroscopy, supported by density functional theory (DFT) calculations. The Raman spectra of undoped samples reveal two of the four symmetry-predicted Raman-active modes and a broad two-phonon structure, indicative of strong electron-phonon coupling. In doped samples, disorder introduces new scattering channels, resulting in additional peaks. Our analysis shows that disorder has a negligible effect on electron-phonon coupling, suggesting that the CDW phase is more susceptible to structural changes, while the robust superconducting order persists due to the enduring electron-phonon coupling. This research provides insights into the effects of crystallographic disorder and electron-phonon coupling on Raman spectra and lattice dynamics of 2H-TaSe_{2-x}S_x compounds.

Symmetry-Resolved Raman Study of Temperature-Induced Phonon Anomalies in ZrTe₅

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Abstract. Zirconium pentatelluride (ZrTe₅) has attracted considerable attention in the condensed matter community due to its temperature-dependent band gap variations. Additionally, it has been proposed that ZrTe₅ lies near a phase boundary between strong (STI) and weak topological insulator (WTI) phases, as well as undergoing an electronic topological (Lifshitz) transition. Depending on the sample and synthesis conditions, the Lifshitz transition temperature varies between 50 and 150 K [1–3]. In this study, we performed Raman spectroscopic analysis of ZrTe₅ single crystals over a temperature range of 76 to 300 K. The measurements were carried out in symmetry-resolved scattering geometries using parallel and cross polarization configurations along the principal crystallographic directions. In this setup, only phonon modes of A_g and B_{2g} symmetries are allowed in the respective configurations. The results reveal pronounced temperature-dependent behavior, including variations in phonon linewidths and peak positions. Notably, at lower temperatures, certain phonon modes exhibit asymmetric line shapes that are well described by the Fano profile. As the temperature increases, these features gradually evolve into symmetric peaks. This behavior indicates a strong coupling between lattice vibrations and electronic excitations in ZrTe₅. Our Raman scattering results provide valuable insight into the phonon dynamics and electron–phonon coupling in ZrTe₅, contributing to a deeper understanding of its fundamental properties.

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Anisotropic Strain Response in FeSe

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Abstract.

Unconventional superconductivity often arises in materials with complex interactions, where competing ordered states such as magnetism, nematicity, and superconductivity, interact and sometimes overlap, making their nature elusive. Among iron-based superconductors, the isostructural FeSe and FeS may appear similar but they differ significantly in their physical properties. While FeSe undergoes a nematic and structural phase transition, FeS shows no structural transition even at the lowest temperatures, with its critical temperature (T_c) halved compared to FeSe. Interestingly, substituting selenium with sulfur in FeSe suppresses the nematic transition temperature to zero near a quantum critical point (QCP), which coincides with a significant drop in T_c . It has been suggested that while spin-fluctuations dominate below the QCP and significantly affect electron-phonon interactions, nematic fluctuations become prominent above the QCP. Here, we present a detailed Raman scattering study of FeSe under uniaxial strain applied along two high-symmetry crystallographic directions, $\langle 110 \rangle$ and $\langle 100 \rangle$, to investigate how symmetry-breaking perturbations affect its lattice dynamics. Our results reveal a pronounced anisotropy in the phonon response to strain: orthorhombic distortion along the $\langle 110 \rangle$ direction leads to a moderate narrowing of the temperature window over which phonon anomalies occur, while strain along $\langle 100 \rangle$ which introduces rhombohedral distortion, results in a significant broadening of the temperature range over which phonon mode splitting, and energy and linewidth anomalies are observed. We find that the fully symmetric A_{1g} phonon mode is particularly sensitive to symmetry-breaking perturbations, while the B_{1g} phonon mode remains largely unaffected.

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O₂ plasma modification of MoS₂ nanotubes for photocatalytic degradation of organic water pollutants

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Abstract. The dye contaminated wastewater is adversely affecting human health and the environment around the world. The issue can be resolved by photocatalytic degradation, which converts dyes into non-toxic compounds without producing secondary waste. In this work, we investigate the ability of MoS₂ nanotubes (NT) to decompose the rhodamine B (RhB) dye. MoS₂ NT synthesized by chemical vapor transport have high specific surface area, numerous S vacancies and favourable energy gap for visible light decomposition [1-2]. However, the initial degradation tests yielded poor results. The inert surface of MoS₂ NT was etched with O₂/Ar plasma which increased a number of catalytically active sites and introduced Mo-O bonds. UV-Vis and Raman spectroscopy, imaging techniques such as SEM, TEM and STM as well as the KPFM work function measurements verified the alteration. Simple MoO₃-MoS₂ system is formed, in which the reducibility of electron in the conductive band of MoS₂ increases, while the oxidability of hole in the valence band of MoO₃ also increases (Figure 1). We report over 90 % degradation efficiency, with the majority decomposing in the first 15 min of LED irradiation.

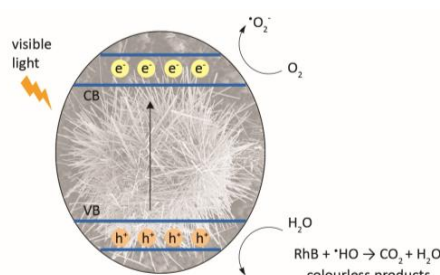


FIGURE 1. Schematic representation of photodegradation of RhB on MoS₂-MoO₃ NT.

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Raman Spectroscopy Analysis Of Disorder Effects In $2H\text{-TaSe}_{2-x}\text{S}_x$ Alloys

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Abstract. Layered transition metal dichalcogenides (TMDs) possess remarkable electronic and optical properties, largely due to their quasi-two-dimensional structures. These materials display complex phase diagrams and temperature-driven transitions [1]. In these compounds charge density wave (CDW) and superconductivity (SC) are shown to coexist at certain temperatures - illustrating the delicate balance and inherent competition between these two phases [2]. Isoelectronic substitution such as sulfur doping in $2H\text{-TaSe}_{2-x}\text{S}_x$, primarily introduces disorder, which disrupts the CDW phase and enhances superconductivity [3]. Tuning properties through doping and structural changes highlights the potential of TMDs for future quantum materials and devices.

In this work, we examined the lattice dynamics of $2H\text{-TaSe}_{2-x}\text{S}_x$ single crystals ($0 \leq x \leq 2$) using Raman spectroscopy alongside density functional theory (DFT) calculations. For the undoped samples, the Raman spectra show two out of the four symmetry-allowed modes and a broad two-phonon feature, which is correlated with strong electron-phonon coupling (EPC). With sulfur doping, the induced disorder opens new scattering channels, giving rise to extra peaks. Our results indicate that disorder minimally affects the EPC, suggesting that while the CDW phase is sensitive to structural changes, the SC state remains robust. Overall, our study sheds light on how crystallographic disorder and EPC shape the Raman response and lattice dynamics in these compounds.

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Raman Signatures Of Instabilities In InSiTe₃

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Abstract. Layered van der Waals materials have gained considerable interest for their unique physical properties, yet InSiTe₃ remains largely unexplored due to uncertainties surrounding its crystal structure. In this work, we present a comprehensive experimental and theoretical investigation of InSiTe₃, confirming a rhombohedral structure with $P\bar{3}$ space group symmetry via single-crystal X-ray diffraction. Polarization-resolved Raman scattering reveals nine out of ten Raman-active modes expected for this symmetry, further validating the structural assignment. Beyond conventional phonon behavior, we identify strong anharmonicity and the emergence of a self-organized coherent phonon state associated with a high-energy A_g mode near 500 cm⁻¹. Analysis of phonon-phonon coupling parameters indicates that A_g modes exhibit coupling strengths up to eight times greater than E_g modes. Temperature-dependent Raman measurements from 80 to 300 K reveal notable changes in A_g mode intensities around 200 K and the appearance of broad spectral features in the phonon gap region, attributed to overtone excitations. Our findings point to an intrinsic lattice instability in InSiTe₃, driven by strong anharmonic interactions. However, further studies are required to fully uncover the microscopic origin of these instabilities and their implications for the material's physical properties.

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