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EDITED BY
Muhammad Sohail Bashir,
University of Jinan, China

REVIEWED BY
Yee Sin Ang,
Singapore University of Technology and
Design, Singapore

*CORRESPONDENCE
Archit Dhingra,
✉ archit.dhingra@uv.es

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Layered GeI₂: A wide-bandgap semiconductor for thermoelectric applications—A perspective

Archit Dhingra^{1,2*}

¹Department of Physics and Astronomy, University of Nebraska—Lincoln, Jorgensen Hall, Lincoln, NE, United States, ²Carrer del Catedràtic José Beltrán Martínez, Institut de Ciència dels Materials de la Universitat de València (ICMUV), University of Valencia, Valencia, Spain

Layered GeI₂ is a two-dimensional wide-bandgap van der Waals semiconductor, which is theorized to be a promising material for thermoelectric applications. While the value of the experimentally extrapolated indirect optical bandgap of GeI₂ is found to be consistent with the existing theoretical calculations, its potential as a thermoelectric material still lacks experimental validation. In this Perspective, recent experimental efforts aimed towards investigating its dynamical properties and tuning its bandgap further, *via* intercalation, are discussed. A thorough understanding of its dynamical properties elucidates the extent of electron-phonon scattering in this system, knowledge of which is crucial in order to open pathways for future studies aiming to realize GeI₂-based thermoelectric devices.

KEYWORDS

2D materials, wide-bandgap, van der waals materials, thermoelectric, intercalation, x-ray photoemission spectroscopy, debye temperature

Introduction

Pristine layered germanium (II) iodide (GeI₂) is a two-dimensional (2D) van der Waals (vdW) material, which is predicted to be thermally stable at temperatures as high as ~ 600 K (Liu et al., 2018; Hu et al., 2020). Adding to its versatility, for potential nanodevice applications, are its: (i) low cleavage energy (~0.16 J/m²) (Liu et al., 2018), which is even lower than that of graphite (Zacharia et al., 2004); (ii) calculated appreciable charge carrier mobilities (Liu et al., 2018); (iii) a wide-bandgap (Liu et al., 2018; Hoat et al., 2019; De Andrade Deus and De Oliveira, 2020; Hu et al., 2020; Naseri et al., 2020; Ran et al., 2020; Opoku et al., 2022), which is now experimentally verified (Dhingra et al., 2022b); and (iv) the fact that it can exist without the undesirable edge disorders (Aivilov and Imamov, 1968; Urgiles et al., 1996; Liu et al., 2018) that have plagued some well-researched 2D materials (Wimmer et al., 2008; Banhart et al., 2010; Mucciolo and Lewenkopf, 2010; Wurm et al., 2011; Komsa et al., 2012; Dugaev and Katsnelson, 2013; Lo et al., 2014; Shi et al., 2014; Addou et al., 2015; Dong et al., 2015; Lin et al., 2016; Mlinar, 2017; Rosenberger et al., 2018; Blades et al., 2020; Debbarma et al., 2021). Besides being a great candidate for standalone nanodevice applications, its heterostructures have also shown some promise

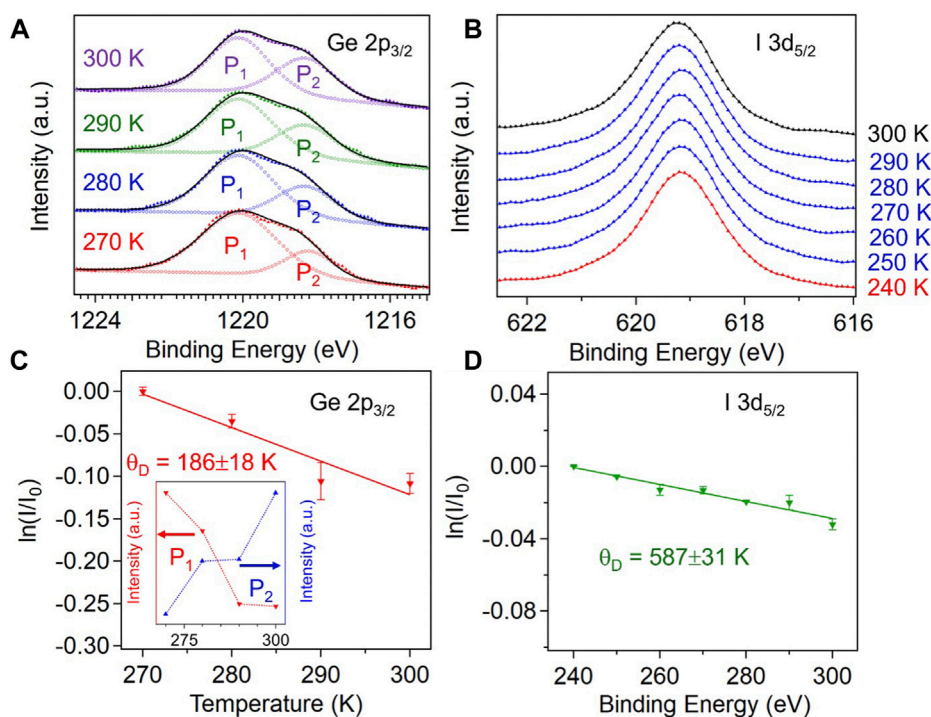


FIGURE 1

(A) Raw XPS of the Ge $2p_{3/2}$ core-level (solid markers) collected at temperatures ranging from 270 K to 300 K, along with the total fit (black line). The hollow markers show the P_1 and P_2 features. (B) XPS spectra of the I $3d_{5/2}$ core-level recorded at temperatures ranging from 240 K to 300 K. (C) Debye–Waller factor plot for the P_1 feature of the Ge $2p_{3/2}$ core-level, as well as the temperature dependence of the XPS intensities of the P_1 and P_2 components (inset). (D) Debye–Waller plot for the I $3d_{5/2}$ core-level (Dhingra et al., 2022b). © IOP Publishing. Reproduced with permission. All rights reserved.

for applications in development of low-power spintronic devices (Shao et al., 2021) as well as for enhancing photocatalytic hydrogen generation performance (Opoku et al., 2022).

In this Perspective, some recent efforts that were aimed towards experimental investigation of the understudied, but highly promising, pristine 2D layered GeI_2 are discussed. The main purpose of this article is to comment on the dynamical stability of the 2D layered GeI_2 , and the intercalation-induced tuning of its bandgap, in the context of its potential as a wide-bandgap thermoelectric material.

Dynamical behavior of the layered GeI_2 : What do temperature variations do to GeI_2 ?

A fundamental understanding of the effects of temperature variation on any material is indispensable if that material is to be used for thermoelectric applications. Therefore, in this subsection the dynamical behavior of the pristine 2D layered GeI_2 is described and discussed. The deterrent effects of the thermal motion of atoms on the physical and electronic

properties of materials are well-known (Körmann et al., 2017; Randle et al., 2018; Dhingra et al., 2020; 2021a; 2021b). One certain way to quantify the extent of electron-phonon scattering (or the effects of thermal motion of atoms) in a material is to ascertain its Debye (or effective Debye) temperature. In this regard, temperature dependent x-ray photoemission spectroscopy (XPS) is routinely exploited to determine a system's effective Debye temperature (Clarke, 1985; Borca et al., 2000; McHale et al., 2011; Evans et al., 2018; Dhingra et al., 2020; 2021a; 2021b).

Figures 1A, B show the temperature dependent XPS spectra of the Ge $2p_{3/2}$ core-level and the I $3d_{5/2}$ core-level, respectively. The effective Debye temperatures of the Ge $2p_{3/2}$ component closer to the surface (the P_1 feature of the Ge $2p_{3/2}$ core-level) and the I $3d_{5/2}$ core-level, as extracted from the linear fits to the Debye–Waller plots in Figures 1C, D, are found to be 186 ± 18 K and 587 ± 31 K, respectively (Dhingra et al., 2022b). It is worth mentioning that, unlike the observed temperature dependence of the photoemission intensities of the P_1 feature of the Ge $2p_{3/2}$ core-level and the I $3d_{5/2}$ core-level, the XPS intensity of the P_2 feature of the Ge $2p_{3/2}$ core-level is found to increase with increasing temperature (see inset of Figure 1C). Such a direct

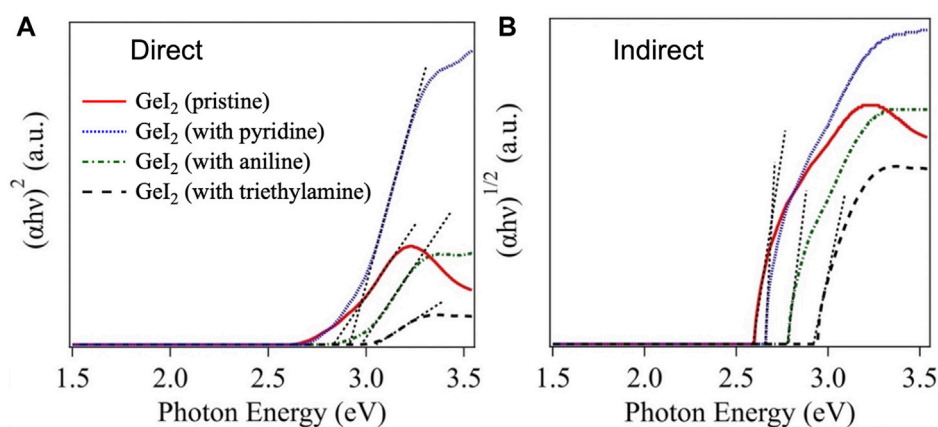


FIGURE 2

Tauc plots for extrapolation of (A) direct and (B) indirect band gaps of the pristine and intercalated GeI₂ systems. Reprinted by permission from Springer Nature (Dhingra et al., 2022a), 2022.

relationship between the photoemission intensity of the P₂ component of the Ge 2p_{3/2} core-level and temperature implies segregation of germanium in the subsurface (or selvedge) of GeI₂ (Dhingra et al., 2022b).

Effects of intercalation: can the bandgap of GeI₂ be widened further?

Over the years, intercalation of various 2D materials has resulted in widening of their bandgaps (Eknapakul et al., 2014; Song et al., 2015; Xiong et al., 2015; Wan et al., 2016; Feng et al., 2019). While the pristine GeI₂ already has a wide-bandgap, intercalating it with easily synthesizable organic molecules to widen its bandgap further is bound to amplify its utility for low-dimensional high-temperature thermoelectric and optoelectronic applications; and this is exactly what was demonstrated in a recent study (Dhingra et al., 2022a).

Figure 2 shows the Tauc plots through which the direct (Figure 2A) and indirect (Figure 2B) bandgaps of pristine and intercalated GeI₂ can be extrapolated (Tauc et al., 1966; Wood and Tauc, 1972). From Figure 2, it is clear that both the direct and indirect optical bandgaps of the pristine GeI₂ are in agreement with the ones reported elsewhere (Dhingra et al., 2022b), and the extrapolated direct (indirect) optical bandgaps upon intercalation with the organic molecules, namely: pyridine, aniline, and triethylamine, are found to lie somewhere between 2.9 (2.66)–3.1 (2.92) eV.

Discussion

Theoretically, layered GeI₂ is a low-dimensional material that is waiting to get exploited for thermoelectric applications (Hu et al., 2020; Lu and Guan, 2022); nevertheless, there are no experiments to confirm the same. The limited experimentally obtained information that we do have on this exciting nanomaterial suggests that the surface of the layered GeI₂ is not stable enough under ambient conditions, and is prone to a large amount of phonon scattering since the effective Debye temperature of the surface is way less than the room temperature (Dhingra et al., 2022b). However, if the complexities encountered at the surface of this material are addressed by depositing a thin layer of a stiff oxide, as has been done for some other quasi-1D materials (Lipatov et al., 2015; Dhingra, 2022), then it may well be possible to realize its potential as a thermoelectric nanomaterial. A stable GeI₂ surface with suppressed phonon scattering, when taken together with the ease with which the bandgap of GeI₂ can be widened further (Dhingra et al., 2022a), will augment its versatility for high-temperature thermoelectric applications. Finally, it must be noted that as is the case when it comes to forming contacts with most 2D materials (S. Schulman et al., 2018; Ang et al., 2021; Zheng et al., 2021; Dhingra et al., 2020a; Dhingra et al., 2021c; Dhingra et al., 2022c; Dhingra et al., 2022d), forming Ohmic contacts to GeI₂ may not be trivial; especially because the germanium rich surface could adversely affect the contact potentials for GeI₂-based nanodevices (Dhingra et al., 2022b). This Perspective, thus, acts as a guide for future studies dealing with the GeI₂-based thermoelectric nanodevices.

Data availability statement

The original contributions presented in the study are included in the article/supplementary materials, further inquiries can be directed to the corresponding author.

Author contributions

AD wrote the whole manuscript.

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Conflict of interest

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