Electrically tunable polarizer based on 2D orthorhombic ferrovalley materials

Xin-Wei Shen\textsuperscript{1,3}, Wen-Yi Tong\textsuperscript{1,3}, Shi-Jing Gong\textsuperscript{1} and Chun-Gang Duan\textsuperscript{1,2}

\textsuperscript{1} Key Laboratory of Polar Materials and Devices, Ministry of Education, Department of Electronic Engineering, East China Normal University, Shanghai 200241, People’s Republic of China
\textsuperscript{2} Collaborative Innovation Center of Extreme Optics, Shanxi University, Taiyuan, Shanxi 030006, People’s Republic of China
\textsuperscript{3} Cofirst authors.

E-mail: cgduan@clpm.ecnu.edu.cn

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Abstract
The concept of ferrovalley materials has been proposed very recently. The existence of spontaneous valley polarization, resulting from ferromagnetism, in such hexagonal 2D materials makes nonvolatile valleytronic applications realizable. Here, we introduce a new member of ferrovalley family with orthorhombic lattice, i.e. monolayer group-IV monochalcogenides (GIVMs), in which the intrinsic valley polarization originates from ferroelectricity, instead of ferromagnetism. Combining the group theory analysis and first-principles calculations, we demonstrate that, different from the valley-selective circular dichroism in hexagonal lattice, linearly polarized optical selectivity for valleys exists in the new type of ferrovalley materials. On account of the distinctive property, a prototype of electrically tunable polarizer is realized. In the ferrovalley-based polarizer, a laser beam can be optionally polarized in $x$- or $y$-direction, depending on the ferrovalley state controlled by external electric fields. Such a device can be further optimized to emit circularly polarized radiation with specific chirality and to realize the tunability for operating wavelength. Therefore, we show that 2D orthorhombic ferrovalley materials are the promising candidates to provide an advantageous platform to realize the polarizer driven by electric means, which is of great importance in extending the practical applications of valleytronics.

1. Introduction
Since the discovery of graphene [1], the interests in exploring novel two-dimensional (2D) materials rooted in hexagonal lattice have been intensively stimulated [2–9]. Similar to charge and spin, valley index [10, 11], as an emerging degree of freedom of electrons, constitutes the binary states in solids, leading to valleytronic devices on information functional applications [12–15]. Very recently, Tong et al [16] introduced the concept of ferrovalley material in transition metal dichalcogenides. Compared with valley degeneracy splitting through external means [17–22], the valley polarization in the proposed ferrovalley monolayers, originating from ferromagnetism, is spontaneous. It is, thus, of great importance in paving the way to the practical use of valleytronics in a nonvolatile way. Interestingly, due to the polarized optical selectivity for valleys, chirality-dependent optical band gap exists here, which indicates the possibility to judge the valley polarization utilizing noncontact and nondestructive optical ways, and offers the additional opportunities for ferrovalley materials as optical devices.

However, the valley polarization in previous study [16] originates from the inherent exchange interaction, making an external magnetic field necessary for the hexagonal ferrovalley monolayer to control its polarity. Compared with the control of degrees of freedom by the energy-intensive magnetic way, manipulation via purely electric fields is highly desirable due to its advantages of ultra-high speed and ultra-low power consumption. In this context, if we could connect ferrovalley with ferroelectricity, such an additional mechanism for intrinsic valley polarization would provide the opportunity to realize the attractive electrical control of ferrovalley states.

As we know, valley-related research mainly focused on pioneering 2D materials with hexagonal lattice [23–25]. However, multi-valley band structures are proved to exist in other crystallographic systems, such as the orthorhombic phases [26]. They provide a promising
and entirely new platform for the studies of the fundamental physics in valleytronics, and more importantly, make the realization of tuning valley degree of freedom through electric means possible. It is then naturally to raise the following questions: Could the robust ferrovalley states persist in new kinds of crystallographic systems? How to dynamically utilize the valley polarization on the modulation of ferroelectricity? Would the new member of valleytronic materials demonstrate distinctive valley-related optical properties, other than the valley-selective circular dichroism (CD) in hexagonal systems [13, 16, 23]?

In this Letter, we propose the monolayer group-IV monochalcogenides in ferroelectric state as an entirely new ferrovalley member, extending the concept of ferrovalley materials from hexagonal to orthorhombic systems. Here, the spontaneous valley polarization is induced by ferroelectricity, instead of ferromagnetism. Taking the monolayer GeSe as an example, our analytical research and first-principles calculations show that in this system, the optical transitions at valleys are coupled to linear dichroism (LD). Consequently, linear polarization-dependent optical band gap could be observed in these ferrovalley materials. The interesting non-degenerate optical excitation offers the possibility to realize a prototype of electrically tunable polarizer. In the polarizer, x- or y-polarized light can be optionally emitted, depending on the specific ferroelectric state manipulated by electric fields. Compared with traditional mechanically driven ones, the ferrovalley-based polarizer provides a more accurate and rapid way to electrically control the polarization state of linearly polarized radiation. Additionally, the proposed device based on 2D materials meets the requirement of next-generation electronic products towards miniaturization and multi-functionality. On account of the inverse piezoelectric effect [27, 28], continuous tunability of band gap in monolayer GeSe can be realized via external electric fields. Thus, the more advanced polarizer with advantage of wide operating wavelength range is emerging to further widen the valleytronic applications.

2. Methods

Our first-principles calculations are carried out by density-functional theory (DFT) using the projector augmented wave (PAW) [29] method which implemented in the Vienna ab initio simulation package (VASP) [30, 31]. The exchange correlation potential is described by generalized gradient approximation (GGA) of Perdew–Burke–Ernzerhof (PBE) [32] functions. The cutoff energy of 500 eV is applied to the plane wave basis set. All the structures are optimized until the forces tolerance below 1 meV Å$^{-1}$ and self-consistent convergence for electronic energy is $10^{-6}$ eV. For the optical property calculations, we take the own-developed code OPTICPACK [33–37]. The Brillouin zone (BZ) is sampled with a Γ-centered $12 \times 12 \times 1$ grid and increase to $24 \times 24 \times 1$ for optical calculations. A vacuum space of 15 Å is used to avoid interactions between periodically repeated layers.

3. Results and discussion

In analogy to black phosphorus [38], group-IV monochalcogenides [39–44] exhibit waved structures. Figures 1(a) and (b) display the primitive cell of pristine monolayer GeSe with inversion symmetry. There is no relative displacement between cations and anions along x or y axis, indicating an in-plane paraelectric phase, labelled as $P_{\infty}$. As plotted in figure 2(a), the band structure of the nonpolar GeSe obviously displays multi-valley characteristics, where two pairs of valleys locate in the X–Γ and Y–Γ high-symmetry paths instead of the symmetry corners of the BZ, as illustrated in figure 1(c).
They are related by the fourfold rotational symmetry, and energetically degenerate with the $C_{2v}$ point group symmetry. Note valleys $V_{x+y}$ and $V_{x-y}$ are related by time-reversal operation. Since the spin–orbit coupling (SOC) barely affects the band gaps at valleys [43] which relate to valley polarization, and spin degree of freedom of electrons is inessential to explore valley physics of the monolayer group-IV monochalcogenides, we just discuss the electronic and optical properties without the SOC effects at valleys. In spite of the distinct lattice site properties without the SOC effects at valleys, we just discuss the electronic and optical properties without the SOC effects at valleys. We notice that basis functions with linear combination of atomic orbitals are invariant through rotational symmetry. Then, the overall azimuthal quantum number $M_x$ is eventually calculable, 

$$\exp [iM_x(\varphi + \pi)] = \exp [iM_x\varphi] \cdot (4)$$

As a consequence, both the VBM and CBM hold the identical $M_x = 0$. For the optical transition at $V_x$ valley, the angular momentum selection rule indicates that $\Delta M_x = 0$, corresponding to the absorption of $x$-polarized photons with the symmetry of $\cos(\theta)$ in the case. Similarly, under the twofold rotation $C_{2v}$ at $V_y$ valley, we can deduce that $\Delta M_x = 0$, indicating the optical absorption at $V_y$ valley can only be excited by the $y$-polarized light. Therefore, based on conservation of overall angular momentum, the valley-dependent optical selection rule in paraelectric GeSe monolayer could be summarized as the following: at $V_x$ valley the optical transition from VBM to CBM could only be excited by $x$-polarized light, while at $V_y$ valley it couples to the radiation polarized along $y$-direction. It is interesting to point out that the valley-dependent optical selection rule in such orthorhombic material is distinct from that of the hexagonal systems. It
corresponds to linearly polarized light here rather than the circularly one in graphene and transition metal dichalcogenides. In analogy to circular dichroism [13], the anisotropy absorption of linearly x- and y-polarized light at valleys can be referred to as valley-selective linear dichroism.

Because of the specific linearly optical selection rule here, as shown in figure 2(b), the calculated optical curves related to x- and y-polarized light are completely overlapped. Their identical optical band gap with the magnitude of 0.83 eV is in accordance with the electronic one gained from figure 2(a). We note that the degeneracy of optical properties excited by x- and y-polarized radiation is due to the paravalley state in such monolayer group-IV monochalcogenides.

When the intrinsic ferroelectricity of monolayer GeSe is taken into account, as shown in figures 3, the relative displacement between Ge and Se atoms occurs, making its inherent inversion symmetry broken. The ferroelectric states with polarization direction along x and y axis are labelled as P_x (figure 3(a)) and P_y (figure 3(b)), accordingly. Both the theoretical and experimental investigations have confirmed that the ferroelectric phase of such orthorhombic systems is the ground state, which is dynamically and thermally stable at high transition temperature [46–49]. The spontaneous in-plane polarization (P) estimated in monolayer GeSe can reach to \(3.5 \times 10^{-10} \text{ C m}^{-1}\), which is well consistent with previous works [50–52].

Their band structures are plotted in figures 4(a) and (b). Compared with electronic distribution in paraelectric GeSe monolayer, the occupied states of VBM and CBM at valleys V_x and V_y are nearly unchanged. Nevertheless, degeneracy between two pairs of valleys is lifted. The ferroelectricity breaks the fourfold rotational symmetry, making the band gaps of V_x and V_y no longer identical, which is the critical sign of spontaneous valley polarization. Therefore, the intrinsic ferrovalley with nonvolatile polarization state has been induced by ferroelectricity in monolayer GeSe. As clearly shown in figure 4(a), for the ferroelectric GeSe monolayer under P_x case, the global band gap locates in V_x valley with the magnitude of 1.14 eV. While, V_y valley owns a larger band gap ~1.67 eV. Interestingly, although the V_x valley still belongs to C_2v point group with the x-polarized light selective excitation,
ferroelectric polarization along x direction makes the symmetry of \( V_x \) valley reduced. Now, it holds the \( C_{3v} \) point group, where the optical transition from VBM to CBM can be excited by both the x- and y-polarized radiation. As a result, the occurrence of energetically non-degenerate valleys implies decoupled optical band gaps of linearly polarized light.

When the ferroelectric polarization occurs along y direction, the polarity of valley polarization is reversed in the meantime. The global band gap with the same magnitude as \( P_x \) state now shifts to \( V_y \) valley (see figure 4(b)). Group theory analysis indicates that, in such case, the \( V_y \) valley reverts to its original symmetry with \( C_{2v} \) point group, whose interband transition between VBM to CBM can be merely excited by the y-polarized light. For the \( V_y \) valley with lower symmetry, x- and y-polarized radiation belong to the identical irreducible representation, making the optical transition couple to the double-degenerate in-plane linearly polarized radiations. The energetical non-degeneracy between valleys is obviously verified as unequal optical band gaps between \( xx \) and \( yy \) components in figure 5(b).

In a word, the intrinsic ferroelectricity in monolayer GeSe breaks the spatial symmetry and lifts the valley degeneracy. The valley polarization, then, naturally appears in the ferrovalley state, whose polarity is optionally controlled by the direction of the ferroelectric polarization. The ferrovalley states existing in such orthorhombic systems break through the restriction of conventional hexagonal lattice. The valley-related properties in the new member of ferrovalley family seem peculiar and quite different from previous ones based on hexagonal lattice. Table 1 lists their comparisons.

|        | VSe\textsubscript{2} | GeSe |
|--------|----------------------|------|
| Mechanism for valley polarization | Ferromagnetism | Ferroelectricity |
| Lattice structure | Hexagonal | Orthorhombic |
| Optical properties | Circular dichroism | Linear dichroism |
| Anomalous valley Hall effect | ✓ | × |

Figure 5. The imaginary parts of complex dielectric function \( \varepsilon_2 \) excited by linearly x-polarized light and y-polarized light of ferrovalley GeSe monolayer in (a) \( P_x \) and (b) \( P_y \) state.
to the point group symmetry, majority carriers bear zero Berry curvature and then pass through the Hall bar straightforwardly without transverse deflection. In addition, pairs of valleys protected by time-reversal symmetry, i.e. valleys $V_{+x(y)}$ and $V_{-x(y)}$, are energetically degenerated, which restricts the possibility of transverse charge Hall current. We note that the anomalous valley Hall effect is absent in both slightly n- and p-doped ferrovalley materials with orthorhombic lattice.

In consideration of linearly polarized optical selectivity for valleys, optical approach is an effective method with advantage of noncontact and nondestructiveness to determine the occurrence of valley polarization and its polarity reversal in such ferrovalley monolayers. In turn, the specific ferrovalley state, which depends on the orientation of electric dipole and is switchable via external electric fields [55], offers the possibility to tune the direction of linearly polarized light. An idea for the unique valleytronic device application is thus inspired. Figure 6 displays the prototype of electrically controllable polarizer based on the monolayer GeSe.

In the polarizer, a laser beam with the energy of 1.14 eV is incident on the pristine monolayer GeSe. The valley polarization of monolayer GeSe could be induced by an external electric fields. When the in-plane electric field is applied along the $x$ axis, the band gap locates in the $V_x$ valley, where electrons jump to the excited state by absorbing the laser beam. In consideration of the photoluminescence process, the excited electrons at $V_x$ valley finally re-emit radiations as they drop back to the ground state. Due to the optical selectivity for the $V_x$ valley, the excitation light is polarized in $x$ direction. If the double-throw switch applies the electric field along $y$ axis, monolayer GeSe undergoes a ferroelectric phase transition from $P_x$ to $P_y$ state. Polarity of its valley polarization simultaneously reverses. Now, the $V_y$ valley possesses the smallest band gap. Its symmetry indicates the emission of $y$-polarized radiation. Hence, through controlling the direction of external electric fields, ferroelectric phase, and then valley polarization of monolayer GeSe could be manipulated. Generally, the polarization switching in a ferroelectric behaves an ultrafast dynamics, of which the time scale can vary from microseconds to nanoseconds [56–58]. With the linearly polarized optical selectivity, the ferrovalley-based polarizer offers the possibility to dynamically polarize the laser beam in $x$- or $y$-direction in a more accurate and high speed electrical way.

Furthermore, if a set of quarter-wave plate is placed perpendicular to the propagation of linearly polarized light with its optical axis deviating $45^\circ$ anti-clockwise from the $x$ direction (or equivalently clockwise from the $y$ direction), then chirality of circularly polarized light could be electrically tunable in a similar way. The linearly polarized light in $x$ direction, which is emitted by applying the electric field along $x$ axis, transfers to the left-handed radiation. Converting the switch makes the monolayer GeSe be in $P_y$ state. Then, the generated circularly polarized light consequently becomes the right-handed one.

Previous analysis implies that utilizing the ferroelectricity in monolayer group-IV monochalcogenides, a laser beam with specific wavelength can be electrically polarized to linearly and even circularly polarized radiation. The polarization state of the light is optionally tuned via advantageous electric means. More interestingly, the giant inverse piezoelectric effects and strain-sensitive band gap [59–62] in these materials indicate the potential to realize an advanced polarizer with wide operating wavelength range.

To further study the influence of epitaxial strain on the band gap of ferrovalley GeSe, we change the lattice constant $a$ ($b$) in the ferroelectric polarization for $P_x$ ($P_y$) state within the range 4.18–4.35 Å, which is equivalent to the strain from $-2.0\%$ to $+2.0\%$. The
lattice constant \(b(a)\), as well as atomic coordinates, are optimized for each case. As shown in figure 7, when the internal generation of a mechanical strain, resulting from an applied electric field, varies from the compressive to the tensile one, the band gap of monolayer GeSe increases linearly.

For the \(P_x\) state, strain makes few effect on location of the direct band gap in \(k\)-space. It is always at \(V_x\) valley, corresponding to \(x\)-polarized radiation. Whereas, the value of the band gap greatly changes, from the maximum of 1.20 eV under tensile 2% strain to the lowest \(\sim 1.09\) eV under the same magnitude of compressive one, which equals to the enhancement of 105 nm in wavelength for the emission. Note that the band gap of monolayer GeSe in \(P_x\) state with respect to epitaxial strain is identical to the case in \(P_x\) state. However, it locates in the \(V_y\) valley with the \(y\)-polarized light selective excitation. As a result, electric-field-induced strain engineering demonstrates an effective way to change the direct or equivalently optical band gap of monolayer GeSe in ferrovalley state. The wavelength for emergent light could be controlled in a quite large range when the mechanical strain varies within a small scale. Such effect implies that through the external electric fields, we can not only generate either linearly polarized light or circularly polarized one can be acquired and optionally tuned through advantageous electric means. Furthermore, band gap in these systems is sensitive to epitaxial strain, indicating the possibility to continuously control the operating wavelength of the polarizer in a large scale. The new kind of 2D materials with spontaneous valley polarization widens the members of ferrovalley family, and is of great potentialities in valleytronic and optoelectronic applications.

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**Figure 7.** The band gap of monolayer GeSe in ferrovalley state with respect to epitaxial strain from \(-2.0\%\) to \(+2.0\%\).
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