Controllable optical and magneto-optical properties of magnetic CrI$_3$ nanoribbons

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ABSTRACT A monolayer of CrI$_3$ has an amazing ferromagnetic ground-state. In this work, we calculate band structures and magnetic moments of tensile-strained and bent zigzag CrI$_3$ nanoribbons with density functional theory. The edge iodine atoms form flat low-lying conduction bands and couple with chromium atoms ferromagnetically, while the non-edge iodine atoms weakly couple antiferromagnetically. CrI$_3$ nanoribbons have a nearly equal preference for the out-of-plane and in-plane magnetic moment configurations, slightly favoring the in-plane one. We also calculate optical absorption with many-body perturbation GW-BSE (Bethe-Salpeter equation) and investigate magneto-optical properties, including magnetic dichroism, Faraday and magneto-optical Kerr effects. The low-energy dark excitons are mainly from transitions between electrons and holes with unlike spins and are non-Frenkel-like, while the bright excitons have mixed spin configurations. Tensile strains and bending manifestly modulate the absorption spectra and magneto-optical properties of CrI$_3$ nanoribbons within a technologically important photon energy-range of ~1.0-2.0 eV, suggesting a potential application in tunable magnetic optoelectronics.

The discovery of the intrinsic ferromagnetism (FM) in one two-dimensional (2D) monolayer of the layered material chromium triiodide (CrI$_3$) is amazing,$^1$ since the Mermin-Wagner theorem$^2$ theoretically predicts that no magnetic order can exist in the 2D isotropic Heisenberg model at finite temperatures. However, one can realize the long-range FM order in 2D systems at finite temperatures by breaking the isotropic spin-rotational invariance$^3$. From the measurement of the zero wavevector spin-wave energy at low temperatures with neutron scattering, it was found that the out-of-plane FM phase transition in the 2D honeycomb lattice CrI$_3$ is controlled by spin-orbit coupling (SOC) induced magnetic anisotropy, instead of magnetic exchange coupling as in a conventional ferromagnet$^4$. More interestingly, the SOC induced magnetic anisotropy can be tuned by substituting some chloride atoms with bromine ones in the similar chromium halide CrCl$_{3-x}$Br$_x$ system, achieving a magnetization pointing toward any spatial direction, while the easy-axis of CrCl$_3$ is in-plane and the one of CrBr$_3$ is out-of-plane$^5$. This demonstrates the tuning of magnetization in chromium halides with mixed halide chemistry and provides more flexibility in design for compact 2D magnetic devices.

Huang et al.$^1$ conducted the measurements of the polar magneto-optical Kerr effect (PMOKE) with an applied external magnetic field perpendicular to the 2D plane for CrI$_3$ monolayer and multilayer samples and revealed the large Kerr rotation angles $\theta_K$ (of linearly polarized light) of about 10 mrad for a monolayer, and about 70 mrad for a trilayer, and the metamagnetic effect with a suppressed magnetization for a bilayer. Seyler et al.$^6$ revealed the spontaneous circularly polarized photoluminescence in monolayer CrI$_3$ under linearly polarized light excitation, with a magnetization direction dependent helicity, and attributed the

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phenomenon to the d-d transition of Cr\(^{3+}\) complexes and strong vibronic coupling induced linewidth broadening. These results suggest an attractive potential application of magnetic Cr\(_3\)I for atomically thin magnetic devices and novel magneto-optoelectronic devices. Electric fields can also induce magnetic effects in Cr\(_3\)I layers. Both Jiang et al.\(^7\) and Huang et al.\(^8\) observed a large linear magnetoelectric (ME) effect in the Cr\(_3\)I bilayer field-effect device, in which an externally applied out-of-plane electric field can reversibly switch the bilayer between FM and AFM (antiferromagnetic) states. Moreover, with CrI\(_3\)-graphene vertical heterostructures, Jiang et al.\(^9\) enabled the electrostatic electron or hole doping by the gate voltage to the Cr\(_3\)I layer and demonstrated the tunability of magnetic ordering strength for monolayer CrI\(_3\), and both the strong doping-dependent interlayer exchange coupling and the robust switching of magnetization (AFM vs. FM) in bilayer Cr\(_3\)I by the gate voltage, even in the absence of an external magnetic field. Those findings suggest the unique potential of the 2D magnets for electrically controlled spintronic and valleytronic device applications through proximity coupling in van der Waals homo- or heterostructures.

The optical absorption and magneto-optical properties of monolayers CrI\(_3\) and CrBr\(_3\) were extensively investigated by Wu et al.\(^10,11\) with the developed SOC enabled spinor GW\(^12,13\) and BSE\(^14\) many-body perturbation approach, which allows the accurate calculation of spin-splitting, SOC and self-energy corrections, electron-hole interactions, and both the diagonal and off-diagonal elements of the layer’s macroscopic dielectric tensor, and hence reasonably reproduces the experimentally measured optical absorption and MO Faraday angles for monolayer CrI\(_3\). It showed that the optical and MO responses of 2D magnetic semiconductors are strongly enhanced by the SOC, self-energy, and excitonic effects.

Nanoribbon is a practical form for applications of 2D materials in nanodevices, since strips or flakes of 2D layers are usually cut off and placed in the devices, especially in the spintronic devices. The further reduced dimensionality of nanoribbons usually leads to enhanced quantum confinements and provides unique edge states, which enable more functionality. Both Jiang et al.\(^15\) and Wang et al.\(^16\) calculated the magnetic properties of CrI\(_3\) nanoribbons and confirmed that FM is the stable state for CrI\(_3\) nanoribbons, and the edge states dominate the band structures around the Fermi level. By applying uniaxial strains or bending, one can create local strains on the nanoribbons and effectively modify the edge states and band structures\(^17-20\), leading to controllable electronic and optical properties. In this work, we investigate tensile strains and bending induced tunability of electronic structures of the magnetic CrI\(_3\) nanoribbons. Our DFT calculations\(^21-25\) show for narrow CrI\(_3\) nanoribbons, that the ferromagnetic state with spin moments perpendicular to the ribbon plane is almost equally stable with the ferromagnetic state with the in-plane spin moments along with the ribbon periodical direction, different from the monolayer CrI\(_3\). The recently developed mTASK metaGGA\(^25\) gives improved magnetic moments and edge band gaps for CrI\(_3\) nanoribbons, showing an improved description for p-orbital derived edge states. We also use the full spinor enabled GW-BSE many-body perturbation method to study the optical absorption and magneto-optical properties of the CrI\(_3\) magnetic nanoribbon. We found that there are more dark exciton states with low energy in the CrI\(_3\) nanoribbon, and the dark excitons are not Frenkel-like excitons, different from the monolayer CrI\(_3\) case. The exciton states, optical absorption and magneto-optical property all show manifest tunability with bending or strains. This suggests the application of the magnetic nanoribbon in controllable magnetic optoelectronic nanodevices.

**Band structures and gaps.** The nanoribbons are cut from the monolayer CrI\(_3\) with iodine (I) atom terminated edges (Figures 1(a)-(e)). The Cr atoms close to the edges form zigzag chains along the ribbon periodical direction, which is the supercell vector c (or axis z). The nanoribbon supercells with 4, 6, and 8 Cr atoms are studied and denoted as Z4CrI\(_3\), Z6CrI\(_3\) and Z8CrI\(_3\), respectively. The relaxed structures of nanoribbons under different bending curvatures \(\kappa (\kappa = 1/R, R\) is the bending curvature radius) or tensile
strains $\epsilon$ are shown Figures 1(f)-(h). The supercell vectors $a$, $b$ and $c$ are orientated along axes $x$, $y$ and $z$, respectively. The relaxed structures of narrow $Z4CrI_3$ (of ribbon width $\sim 12.4$ Å) are almost flat. Therefore, bending produces an effective compression along the ribbon’s width direction. $Z6CrI_3$ (width $\sim 18.5$ Å) and $Z8CrI_3$ (width $\sim 24.5$ Å) nanoribbons show relatively large deformations around the ribbon’s middle region, while the regions close to the edges are relatively flat. After relaxations, the length of vector $c$ of the supercell (LC) (Figure 1(m)), which measures the lattice constant along the periodic direction of nanoribbons, slightly increases from a larger tensile strain towards flat, and to larger bending curvatures. $Z6CrI_3$ has a larger LC than $Z4CrI_3$ and $Z8CrI_3$; at larger bending curvatures, due to its two asymmetric edges about the middle of the ribbon (Figure 1(d)). The band structures (Figures 1(i)-(l)) show almost flat conduction bands near the Fermi level. The partial density of states (pDOS) analysis (Figure 1(n)) shows a large contribution to the valence band edge (close to the Fermi level) from the p-orbital of both the edge and the non-edge I atoms, while the nearly degenerate two flat edge conduction bands right above the Fermi level are dominated by the p-orbital of the edge I atoms. The lower valence bands (energy $\sim 0.5$-1.0 eV) have both contributions from the p-orbital of I and d-orbital of Cr atoms. The conduction band continuum ($\sim 1.0$-1.5 eV) above the edge bands is derived from the Cr d-orbitals and I p-orbitals with minor contribution from the edge I atoms, while the higher conduction bands (over 2.5 eV) are mainly derived from the Cr d-orbitals.mTASK gives band structures of $Z4CrI_3$ similar to those from PBE-U around the Fermi level, while it pushes up the higher conduction bands (which begin about 2.5 eV above the Fermi level in PBE-U) derived mainly from the Cr d-orbitals by about 1.5 eV more than PBE-U does.

The band gaps are shown in Figures 2(a)-(c). While $Z4CrI_3$ shows a slight variation of gaps with bending and tensile strains, LSDA-U, mTASK, and G$_0$W$_0$ all show the highest gap at $\kappa = 0.125$/Å (or $R = 8$ Å). For the edge gaps (EG), mTASK gives higher values among the density functional approximations used here and is closer to the many-body perturbation method G$_0$W$_0$ evaluated with LSDA-U reference. For non-edge gaps (NEG), SCAN gives larger values than other DFTs, however, with a vibrating trend in the bending curvature region, while PBE, PBE-U, mTASK, and LSDA-U all give similar values for NEG. r$^2$SCAN results are similar to SCAN results for EG and are slightly lower than those of SCAN for NEG. G$_0$W$_0$ shows large values for both EG ($\sim 2.3$ eV) and NEG ($\sim 3.1$ eV), indicating the large self-energy correction to the quasiparticle energy for the nanoribbon systems. TASK, mTASK, SCAN and r$^2$SCAN are meta-GGA density functionals with ingredients related to the electron kinetic energy density with improved the description of band gaps. The p-orbital EG of $Z4CrI_3$ is better described by mTASK, consistent with reports for other p-orbital materials.

Magnetic moments. Table 1 shows the calculated magnetic moments for the $Z4CrI_3$ flat nanoribbon. All DFTs in Table 1 show approximately the same magnetic moment of 0.21-0.26 of the edge I atoms (atom indices 10-13), while LSDA-U, PBE, SCAN and r$^2$SCAN give similar values ($\sim 3.0$-3.2) for Cr atoms (atoms1-4), and both mTASK and TASK give the values ($\sim 3.5$-3.6) close to those of PBE-U ($\sim 3.3$). The mTASK value is very close to the DMC (diffusion Monte Carlo) value ($3.62$) for Cr in monolayer CrI$_3$. All DFTs here give the moments on Cr and edge I atoms pointed to the positive y direction and perpendicular to the ribbon plane, while the relatively small moments on other non-edge I atoms point to the negative y direction, indicating that those I atoms have a weak antiferromagnetic coupling with Cr and edge I atoms. It was suggested that Cr atoms are coupled with super-exchange via p-orbitals of the neighboring I atoms. The edge I atoms only connect with one Cr atom and have unsaturated dangling bonds, and hence ferromagnetically couple with Cr atoms. The calculations with PBE-U for the flat $Z4CrI_3$ nanoribbon with constrained magnetic moments show a small energy difference ($\sim 0.3$ meV/atom, Supporting table S1) between the y axis (out-of-plane) and z axis (in-plane), and a more stable in-plane configuration. This is different from the monolayer CrI$_3$ case, where the out-of-plane configuration is more
stable. We believe that this effect is due to the in-plane rotational symmetry breaking by the ribbon edges and the interaction between edge states. The average magnetic moment of Cr atoms in Z4CrI3 shows little variations with bending curvatures, except for the results of SCAN, where a similar vibrating trend is shown in line with that of NEG. This may relate to the numerical stability of SCAN previously reported. r²SCAN is based on the SCAN framework but improves the smoothness of the interpolation function and hence the numerical stability.

**Optical absorptions and exciton states.** The calculated absorption spectra of Z4CrI3 are shown in Figures 3(a)-(e). The three main peaks A, B, C below the quasiparticle gap indicate the strong excitonic effect in nanoribbons. From flat to R8, the three peaks move slightly to higher energies, maybe due to the slightly higher EG and NEG gaps of R8. From R8 to R5, both EG and NEG decrease, the three peaks move to low energies by ~0.1-0.2 eV. The peak positions barely change from flat to T2, due to a relatively small strain. Further increasing the tensile strain to 5%, the peaks move towards low energies. The height of peak B is more affected by the tensile strain and bending, showing an increase with tensile strains and a decrease with bending curvatures. In all tensile and bending cases, there are about 6-8 dark excitons within energy range of 0.5-1.1 eV lower than that of Peak A. Those dark excitons are mainly contributed by the transitions from valence bands V1 to V11 to the edge conduction band C1 and C2, see Figures 4(a) and (b) for plots of dark excitons at 0.94 and 0.97 eV. The edge C1/C2 are mainly from the p-orbitals of edge I atoms with a minority spin polarization (Figures 3(f)-(j)), while the valences V1-V11 have a mixed spin polarization. Analyzing the spin polarization of the involved valence and conduction states of the dark excitons (Supporting figures S1(a)-(d)) shows that the valence states are of minority spin and most of the hole states are of majority spin, indicating the formation of the dark excitons from electrons and holes of unlike spin polarizations, consistent with that of the dark excitons in the CrI3 monolayer. However, due to the presence of the edges, the dark excitons in nanoribbons have a more extended region in real space and can span the ribbon width (subplots I-III in Figures 4(a) and (b)), indicating non-Frenkel-like excitons, different from the Frenkel-like dark exciton in monolayer CrI3.

Due to strong SOC effects, the Bloch wave functions of the electron and hole states composing an exciton in the system have various spin polarization directions. This may give rich and complex excitonic spin configurations. Peak A (peak B) is composed of excitons with transitions mainly from V1-V5 to C1/C2 (from V1-V7 to C1/C2), see Figures 4(c) and (d) for the band transition decomposition of the bright excitons at 1.25 eV in peak A and at 1.58 eV in peak B, which have the highest oscillator strength in the peaks. The valence bands V3-V7 have mixed spin polarizations with both majority and minority ones, therefore, the spin configuration of excitons forming peaks A and B is generally complex. The exciton states with the highest oscillator strength in peaks A and B have contributions of transitions from both minority and minority spin valence states to the minority spin conduction edge states (Supporting figures S1(e) and (f)), indicating a rich and mixed spin configuration for those bright excitons, while in a CrI3 monolayer, the bright bound excitons are mainly of like-spin polarization of electron and hole states. The excitons of the highest oscillator strength in peak C and the smaller peak D all have a diverse contribution of transitions from both minority and minority spin valence states to the minority spin conduction edge states (Supporting figures S1(e) and (f)), indicating a rich and mixed spin configuration for those bright excitons, while in a CrI3 monolayer, the bright bound excitons are mainly of like-spin polarization of electron and hole states. The excitons at 1.25 eV in peak A and at 1.58 eV in peak B have a more extended spatial region along the two edges, with the former mainly localized around Γ in the momentum (k) space and the latter nodal in k space and slightly nodal in real space, while the excitons at 1.85 and 2.03 eV in peaks C and D are slightly nodal in the k space and very nodal in the real space with an obvious concentration of electron wavefunction around the vicinity of the hole, mainly due to the relatively wide range of conduction and valence bands involved in the formation of those excitons.
Magneto-optical properties. The zigzag Z4CrI₃ nanoribbon has a mirror symmetry about the plane (parallel to the yz-plane) through the middle of the ribbon. It leads to two elliptically polarized modes (EPM) propagating along the x direction. To demonstrate its magneto-optical (MO) properties, we calculated the magnetic dichroism (MDC), Faraday and MO Kerr effects for the Z4CrI₃ nanoribbon. The results are shown in Figures 5(a)-(f) for the flat nanoribbon. Figure 5(a) shows the very different absorption spectra of the two opposite (left or right) EPMs, denoted as σ⁺ and σ⁻. This leads to a large MDC contrast η, and η = (Abs(σ⁺) − Abs(σ⁻))/([Abs(σ⁺) + Abs(σ⁻)]), where Abs(σ±) denotes the absorbance of σ±, as shown in Figure 5(b), especially within the region of 1.0-2.0 eV, manifesting the significant effect of excitons to MDC. For the transmitted Faraday effect, the rotation angle θᵢ and ellipticity χᵢ (see Ref. 10 for definitions) can be about 2~3 mrad within the region of 1.0-2.0 eV with the GW-BSE results (Figure 5(c)), while the results without electron-hole (e-h) interactions (GW-RPA) (Figure 5(d)) show a zero ellipticity χᵢ and a slightly increasing rotation angle θᵢ. This demonstrates the inclusion of e-h interactions is important. The slightly increasing θᵢ from GW-RPA may be due to the normal optical anisotropy along the y and z directions. Similarly, for the reflective MO Kerr effect, the rotation θᵣ and ellipticity χᵣ can be as large as 600~700 mrad within the region of 1.0-2.0 eV from GW-BSE (Figure 5(e)), indicating a strong MO Kerr effect. The results without e-h interaction (Figure 5(f)) give a zero rotation θᵣ and an almost constant ~600 mrad ellipticity χᵣ, also due to the normal optical anisotropy along axes y and z. Bending and tensile strains can change the MDC, Faraday and MO Kerr signals manifestly, as shown in Figures 5(g)-(k). For example, within the region of 1.0 -1.2 eV, the region of mainly peak A, the case of bending R5 has a large difference from other ones. This demonstrates the tunability of those properties with bending and tensile strains, and can be harnessed for controllable optoelectronic devices.

In summary, from first principles, we have calculated the band structures, band gaps, and magnetic moments of zigzag CrI₃ nanoribbons under different tensile strains and bending curvatures with density functional approximations, and their optical absorption spectra with the many-body perturbation GW-BSE method, and investigated their magneto-optical properties, including magnetic dichroism, Faraday and magneto-optical Kerr effects. The flat conduction bands close to the Fermi level are dominated by the p-orbital of the edge iodine atoms, while the valence bands close to the Fermi level are mostly from the p-orbital of I atoms (both edge and non-edge) and to a lesser extent from Cr d-orbitals. The conduction band beyond the edge bands mostly consists of Cr d-orbitals. The edge I atoms ferromagnetically couple with Cr atoms, while the non-edge I atoms weakly couple antiferromagnetically. Unlike the more stable out-of-plane magnetic moments of a CrI₃ monolayer, CrI₃ nanoribbons have almost an equal stability (energy difference ~0.2 meV/atom) between out-of-plane and in-plane (along the ribbon axis), with a slightly more stable in-plane configuration. The band structures and characters are vital for the formations of the dark and bright bound excitons in the CrI₃ nanoribbons. The dark excitons with energies lower than that of the lowest-energy absorption peak are mainly formed from holes and edge electrons with unlike spin polarizations and are non-Frenkel-like excitons, while the bright excitons in the main absorption peaks are from holes and electrons with mixed spin polarizations. Tensile strains and bending can manifestly modulate the absorption spectrum, magnetic dichroism, Faraday and magneto-optical Kerr effects of the CrI₃ nanoribbons within a technologically important photo energy range of ~1.0-2.0 eV, suggesting a potential application in tunable magnetic optoelectronic nanodevices.

Computational details. Density functional theory (DFT) calculations were conducted in the Vienna Ab initio Software Package (VASP) with projector augmented-wave pseudopotentials. PBE, SCAN, TASK, and mTASK, approximations with spin-orbit coupling (SOC) were used to calculate the band structures of
nanoribbons. In the PBE-U calculation, a rotationally invariant approach with U=2.65 eV for Cr atoms was used. The vacuum layer of more than 15 Å is added along the direction of nanoribbon width and along the out-of-plane direction of the nanoribbon, to avoid the interactions with periodic images. The energy cutoff is 500 eV. The k-point mesh of $1 \times 1 \times 7$ was used for relaxation and $1 \times 1 \times 24$ was used for electronic calculations. All nanoribbons were fully structurally relaxed with PBE with all forces less than 0.02 eV/Å. During the relaxation, the x and y coordinates of the two outer most metal atoms close to the two edges were fixed, while their coordinates along the ribbon axis direction, and all the coordinates of other atoms were allowed to relax. The spinor $G_0W_0$ and $G_0W_0+BSE$ calculations were conducted in BerkeleyGW by pairing with Quantum ESPRESSO. The $G_0W_0$ is based on the LSDA-U calculations with U = 1.5 eV and J = 0.5 eV for Cr atoms. The wavefunction energy cutoff is 70 Ry (~950 eV). The energy cutoff for the epsilon matrix is 14 Ry (~190 eV). The k-point mesh of $1 \times 1 \times 64$ (interpolated from $1 \times 1 \times 16$) and the valence bands of 15 and conduction bands of 10 were set for optical absorption calculations. The band number for summation is 800. The correction of the exact static remainder and the wire Coulomb truncation for 1D systems were also used.

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Table 1. The magnetic moment (in Bohr magneton $\mu_B$) of atoms in the Z4CrI$_3$ flat nanoribbon calculated with different methods. Atom indices 1-4 represent Cr atoms, 10-13 for the four edge I atoms and “other” for the other non-edge I atoms.

| Atom index | 10, 12 | 3   | 1   | 2   | 4   | 11,13 | Others      |
|------------|--------|-----|-----|-----|-----|-------|-------------|
| LSDA-U     | 0.235  | 2.969 | 2.996 | 2.997 | 2.968 | 0.234  | -0.055±0.007|
| PBE        | 0.245  | 3.029 | 3.070 | 3.070 | 3.028 | 0.250  | -0.061±0.006|
| SCAN       | 0.262  | 3.049 | 3.097 | 3.097 | 3.049 | 0.266  | -0.065±0.006|
| PBE-U      | 0.224  | 3.323 | 3.364 | 3.365 | 3.323 | 0.228  | -0.110±0.006|
| mTASK      | 0.225  | 3.589 | 3.632 | 3.634 | 3.589 | 0.229  | -0.213±0.006|
| TASK       | 0.215  | 3.507 | 3.557 | 3.558 | 3.507 | 0.208  | -0.182±0.008|
| r$^2$SCAN  | 0.251  | 3.137 | 3.182 | 3.183 | 3.136 | 0.254  | -0.080±0.007|
Figure 1. The structures of CrI3 nanoribbons, calculated band structures, supercell lattice constant, and density of states. The supercell’s vectors a, b and c are orientated along axes x, y, and z, respectively. The structure of Z4CrI3 is in (a) (top view), in (b) (side view) and in (c) (side view). Blue balls represent Cr atoms and purple ones for I atoms. The structure of Z6CrI3 is in (d) and Z8CrI3 in (e). The boxes in (a), (d) and (e) indicate the periodic unit of the nanoribbon. (f)-(h) show the relaxed structures of Z4CrI3,
Z6CrI3, and Z8CrI3 nanoribbons under different bending or tensile conditions, where T5 (T2) represents the tensile strain of 5% (2%) along the width direction (vector a direction) with respect to the monolayer structure, and R8 represents the bending curvature radius R = 8 Å and so on for others. Note that after relaxation, all Z4CrI3 nanoribbons are almost flat. (i) shows the band structures of Z4CrI3 nanoribbons under different bending or tensile strains (from left to right: tensile 5%, tensile 2%, flat with no bending or tensile strain, bending R = 8 Å, and R = 5 Å) with the PBE-U calculation with U= 2.65 eV for the Cr atom. EG (edge gap) represents the energy gap between the edge band and the valence top at Γ, while NEG (non-edge gap) represents that between the lowest non-edge conduction band and the valence top at Γ, as depicted in one of the panels in (i). (j) shows the band structure of Z4CrI3 from the mTASK calculation. (k) and (l) show the PBE-U band structures of Z6CrI3 and Z8CrI3 nanoribbons, respectively. The changing trend of the length of the supercell’s vector c with bending or tensile strains for the three nanoribbons is in (m). The partial density of states (DOS) of flat Z4CrI3 is in (n).
Figure 2. The edge band gap (EG) and non-edge band gap (NEG) of CrI₃ nanoribbons and the average magnetic moment of Cr atoms. EG and NEG of the Z₄CrI₃ nanoribbon under different tensile or bending calculated with PBE, SCAN, r²SCAN, PBE-U (with U=2.65 eV for Cr atoms), mTASK, TASK, LSDA-U (with U = 1.5 eV and J = 0.5 eV for Cr atoms), and G₀W₀@LSDA-U are in (a) and (b), respectively. EGs of Z₄CrI₃, Z₆CrI₃, and Z₈CrI₃ with bending curvatures are in (c). The average magnetic moment of the four Cr atoms in the cell of Z₄CrI₃ nanoribbon with different tensile strains and bending curvatures is in (d). The schematic of the orientation of magnetic moments of atoms is in (e). Blue balls represent Cr atoms and purple ones for I atoms. The bending curvature is \( \kappa = 1/R \), with a unit of 1/Å, where R is the bending curvature radius. The tensile strain \( \epsilon \) along the ribbon width direction is represented as a negative number, with -0.05 (-0.02) representing 5% (2%) tensile strain.
Figure 3. The optical absorption spectra and $G_{0}W_{0}$ quasiparticle band structures of nanoribbon $Z4CrI_3$ under different tensile strains or bending curvatures. The optical spectra of tensile strain 5% (T5) is in (a), 2% (T2) is in (b), flat ribbon is in (c), bending curvature $\kappa = 0.125 /\text{Å}$ (or $R = 8 \text{ Å}$) is in (d), and $\kappa = 0.20 /\text{Å}$ (or $R = 5 \text{ Å}$) is in (e). All curves are calculated with a Gaussian broadening of 28 meV. “eh” and “noeh” in legends stand for with electron-hole interactions (GW-BSE method) and without electron-hole interactions (GW-RPA method), respectively. From (f)-(j), the spin resolved $G_{0}W_{0}@\text{LSDA-U}$ quasiparticle band structures are for T5, T2, flat, R8 and R5, respectively. (k) is the color bar for the spin polarization of the states. The spin polarization is calculated as $\langle \psi_{n,k} | \sigma_z | \psi_{n,k} \rangle$, where $\psi_{n,k}$ is the wavefunction and $\sigma_z$ the Pauli matrix.
Figure 4. The wavefunction and the electron-band and hole-band composition of excitons at energy of 0.94 eV (a), 0.97 eV (b), 1.25 eV (c), 1.58 eV (d), 1.85 eV (e), and 2.03 eV (f) of the Z4CrI3 flat nanoribbon. For each exciton, the subplots I, II and III are the squared modulus of exciton wavefunction in real space in its top and side views. The hole is located about the sixth Cr atom in the third Cr atom row in subplot I. Blue balls represent Cr atoms and purple ones for I atoms. Subplot IV is the band summed amplitude (arbitrary unit) of exciton, $\sum_{k} |A_{v,c}(k)|^2$, representing the distribution of exciton in k-space. The contributing hole (valence) and electron (conduction) bands for each exciton is in subplot V.
Figure 5. Magneto-optical properties of the Z4CrI3 nanoribbon. The absorbance spectra of the left ($\sigma^+$) and right ($\sigma^-$) elliptically polarized modes (EPMs) is in (a) and its magnetic dichroism (MDC) contrast $\eta$ is in (b). The transmitted Faraday rotation angle $\theta_F$ and ellipticity $\chi_F$ with e-h interaction (GW-BSE) versus photon energy is in (c). The same quantities as in (c) without e-h interaction (GW-RPA) is in (d). The reflective magneto-optical (MO) Kerr rotation $\theta_K$ and ellipticity $\chi_K$ with e-h interactions (GW-BSE) versus photon energy is in (e) and without e-h interactions (GW-RPA) in (f). The dashed vertical lines in (a)-(f) denote the quasiparticle band gap. The Faraday $\theta_F$ under different tensile strains and bending curvatures is in (g), and the Faraday $\chi_F$ under different tensile strains and bending curvatures is in (h). The MO Kerr $\theta_K$ and $\chi_K$ under different tensile strains and bending curvatures is in (i) and (j),
respectively. The MDC contrast under different tensile strains and bending curvatures is in (k). The GW-BSE results are shown in (g)-(k).