Optical activity in chiral stacks of 2D semiconductors

Abstract: We show that the stacks of two-dimensional semiconductor crystals with the chiral packing exhibit optical activity and circular dichroism. We develop a microscopic theory of these phenomena in the spectral range of exciton transitions that takes into account the spin-dependent hopping of excitons between the layers in the stack and the interlayer coupling of excitons via electromagnetic field. For the stacks of realistic two-dimensional semiconductors such as transition metal dichalcogenides, we calculate the rotation and ellipticity angles of radiation transmitted through such structures. The angles are resonantly enhanced at the frequencies of both bright and dark exciton modes in the stack. We also study the photoluminescence of chiral stacks and show that it is circularly polarized.

Keywords: optical activity; chiral structures; TMDs; van der Waals structure.

1 Introduction

Optical activity, the ability of certain media to rotate the plane of light polarization in the absence of a magnetic field, is a remarkable manifestation of polarization-dependent interaction between light and matter [1]. It stems from the different strengths of coupling to right-handed and left-handed circularly polarized light leading to circular birefringence and circular dichroism [2]. From the pioneering experiment by J.C. Bose in 1898 with left- and right-twisted jute elements [3], optical rotation is commonly associated with chirality. Optically active media attract much attention because they allow one to manipulate the polarization state of light. This is particularly important for superresolution imaging and the development of broadband optical components, such as filters and sensors with high polarization suppression ratio [4, 5]. Moreover, photon polarization is currently considered as the degree of freedom to code and carry quantum information [6].

Optical activity, also referred to as gyrotropy, can be of either intrinsic or extrinsic nature. The former occurs in macroscopically homogeneous systems with chiral molecule components or crystals of the gyrotropic symmetry classes [7]. A canonical example of optically active biological media is the tartaric acid, which was discovered by L. Pasteur and explained by the predominance of one of the two possible stereoisomers of the acid molecules [8]. Tellurium is an example of gyrotropic crystals [9]. This elemental semiconductor can exist in two enantiomorphic forms with the atoms, constituting the crystal lattice, bound into left-handed or right-handed chains. An example of gyrotropic but not chiral bulk crystals is silver gallium sulphide [10]. Microscopically, optical activity originates from the spatial dispersion of susceptibility [11]. It is increased in the vicinity of exciton resonances, as was observed in bulk wurtzite crystals [12] and II–VI quantum well structures [13]. The term “extrinsic optical activity” coined recently is primarily used for metamaterials, metasurfaces, and photonic crystals composed of the arrays of chiral or achiral elements such as gammadions [14–16], G-shape nanostructures [17], and split-ring resonators [18] or chiral stacks of metasurfaces [19, 20]. The efficiency of polarization conversion can be enhanced by plasmonic resonances in metal nanostructures [21, 22].

Apart from the above two distinct types of gyrotropic structures, we highlight the opportunity of an intermediate case of artificial materials with chiral stacking of atomic layers. Previously, they would be hypothetical. The emerging technology of van der Waals structures made of two-dimensional (2D) crystals of atomic thickness [23, 24] enables the formation of such materials. Recently, it has been experimentally demonstrated that a pair of graphene layers stacked with a twist exhibits optical activity [25]. The optical rotation angle normalized to the sample thickness was found to be several orders of magnitude.
larger than that in natural materials. The 2D crystals beyond the graphene, such as transition metal dichalcogenides (TMDC) MoS₂ and WS₂, are of particular interest for application in optics. TMDC layers have optical gaps and demonstrate strong light-matter coupling in the spectral range of exciton transitions [26–29]. Pronounced exciton resonances have been observed in the reflectance spectra of TMDC layers and thin films [30–32]. The properties of twisted stacks including interlayer coupling strength are also being widely studied [33–38]. At the same time, the optical polarization effects related to chirality slipped away from the focus of previous research.

In this paper, we study the optical properties of chiral stacks of 2D semiconductors. We show that properly arranged stacks (see Figure 1) exhibit pronounced optical activity while individual layers do not. We develop a microscopic theory of the optical activity and the accompanying phenomenon of circular dichroism. It is also predicted that the photoluminescence (PL) of chiral stacks is circularly polarized.

The paper is organized as follows. In Section 2, we develop a theoretical approach to describe the exciton states in the chiral stacks of TMDC layers and the polarization optical properties of such stacks. We start by considering chiral bilayers and then generalize the model to multilayer stacks. In Section 3, we describe the spectral behavior of light transmission, optical activity, and circular dichroism for the chiral bilayers, multilayers, and thick TMDC stacks. In Section 4, we discuss the circularly polarized PL of chiral stacks. Section 5 summarizes the results of the paper.

2 Model

The spatial symmetry of a single TMDC layer is described by the $D_{3h}$ point group. The layers are optically inactive and demonstrate isotropic linear response at the normal incident of radiation. We consider the optical response associated with bright direct excitons with the spin projections $s = \pm 1$ along the normal to the layer that are excited by the $\sigma^+$ circularly polarized light, respectively. The excitons are formed by electrons and holes located at the $K$ and $K'$ valleys of the 2D Brillouin zone. The exciton states with the spin projections $s = \pm 1$ at zero in-plane wave vector are degenerate in energy.

Arranging two identical TMDC layers into a twisted stack reduces the point-group symmetry of the system to $D_3$. The exciton states in such a bilayer are described by an effective Hamiltonian $H_{n,n'}$, where $n, n' = 1, 2$ stand for the monolayer index and $s, s' = \pm 1$ stand for the spin index. We note that in multilayer systems, there are also excitations consisting of electrons and holes localized in different layers [39, 40]. Those interlayer excitons are far separated in energy from the intralayer excitons because of the reduced Coulomb interaction and are not considered here.

The $D_3$ point group of the bilayer imposes restrictions on the form of the effective exciton Hamiltonian. The presence of the three-fold rotation axis along the bilayer normal eliminates the matrix elements of the Hamiltonian with $s \neq s'$, so that $H_{n,n',s,s'} = H_{n,n',s,s'}^{(s)}$. The two-fold rotation axes lying in the bilayer plane impose the constraints $H_{11}^{(s)} = H_{22}^{(s)}$ and $H_{12}^{(s)} = H_{21}^{(s)}$. Finally, the time-reversal symmetry requires $H_{n,n',s,s'}^{(s)} = H_{n,n',s,s'}^{(s)}$. Combining all the above symmetry constraints together with the requirement that the Hamiltonian is Hermitian, we arrive to the most general form of the exciton Hamiltonian in a chiral bilayer:

$$H^{(s)} = \begin{pmatrix} \omega_s & -J e^{i \phi} \\ -J e^{-i \phi} & \omega_s \end{pmatrix}$$

with real parameters $\omega_s$, $J$, and $\phi$. Here, $\omega_s$ is the exciton frequency in an isolated layer, $J$ describes the interlayer hopping of excitons due to, e.g., tunneling or Förster excitation transfer, and $\phi$ is the phase acquired at the interlayer hopping due to the chirality of the bilayer. The Planck constant is set to unity. The dependence of the phase $\phi$ on the parameters of particular TMDC bilayers can be obtained from atomistic calculations, which are beyond the scope of the present paper. Instead, we focus below on the general optical properties of chiral bilayers and multilayers. We only note that the symmetry analysis yields that the dependence of the phase $\phi$ and the interlayer hopping parameter $J$ on the twist angle is $2\pi/3$-periodic and the optical activity vanishes in achiral stacks with the twist angles $0$, $\pi/3$, $2\pi/3$, etc. It was found in experiments and confirmed by atomistic calculations that in energetically
favourable TMDC bilayers, the interlayer coupling varies with the twist angle [33–35].

The model above can be readily generalized to the chiral stack of $N$ identical layers. Assuming the hopping of excitons between the nearest-neighbor layers only, we shall use the exciton Hamiltonian with the matrix elements

$$H_{n,n'}^{(s)} = \omega_s \delta_{n,n'} - J e^{i\phi} \delta_{n+1,n'} - J e^{-i\phi} \delta_{n-1,n'}$$

with $n, n' = 1, \ldots, N$.

Now we consider the interaction of excitons with the electromagnetic field. Because of high symmetry of an individual layer, the strength of exciton-photon coupling in a layer is described by a single parameter $\Gamma_n$, which determines the exciton radiative decay rate. For the incident light propagating along the $z$ axis (Figure 1), the exciton polarizations in the layers $P_n^{(s)}$ are determined by the equation set (see Ref. [41])

$$(\omega + i\Gamma_n)P_n^{(s)} - \sum_{n'} \left( H_{n,n'}^{(s)} - i\Gamma_n e^{i\phi(n-n')} \right) P_{n'}^{(s)} = -\frac{\Gamma_n}{2\pi q} E_0^{(s)} e^{i\omega n d},$$

where $E_0^{(s)}$ is the electric field amplitude of the incident radiation with a certain helicity, $\omega = cq$ and $q$ are the photon frequency and wave vector, respectively, $nd$ is the position of the $n$-th layer, and $d$ is the distance between the centers of the neighboring TMDC layers. The last term in the left-hand side of Eq. (3) describes both the radiative decay of excitons (at $n=n'$) and the radiative coupling of excitons in different layers (at $n \neq n'$). Equation (3) also takes into account the nonradiative decay of excitons with the rate $\Gamma_n$ [42–44].

The amplitude of a circularly polarized electromagnetic wave transmitted through the stack is given by

$$E_n^{(s)} = E_0^{(s)} + 2\pi q \sum_n P_n^{(s)} e^{-i\omega n d}.$$

By solving Eq. (3), we calculate $E_n^{(s)}$ and the amplitude transmission coefficients for the right-handed and left-handed circularly polarized radiation $t_n^{(s)} = E_n^{(s)} / E_0^{(s)}$.

Consider now the transmission of linearly polarized light through the chiral stack. The spectrum of transmission is determined by

$$|t(\omega)|^2 = |t^{(1)}|^2 + |t^{(2)}|^2.$$

Because of the difference in the transmission coefficients $t^{(1)}$ and $t^{(2)}$, the linearly polarized light, when transmitted through the stack, rotates its polarization plane (optical activity) and acquires ellipticity (circular dichroism). The corresponding rotation angle $\alpha$ and ellipticity angle $\epsilon$ are defined by

$$\alpha(\omega) = \frac{1}{2} \arg \frac{t^{(1)}}{t^{(2)}}, \epsilon(\omega) = \arctan \frac{|t^{(1)}| - |t^{(2)}|}{|t^{(1)}| + |t^{(2)}|}.$$  

At $|t^{(1)} - t^{(2)}| \ll |t^{(1)}|$, the rotation and ellipticity angles are small, and Eq. (6) can be rewritten in the compact form

$$\alpha(\omega) + i\epsilon(\omega) = \frac{t^{(1)} - t^{(2)}}{t^{(1)} + t^{(2)}},$$

Below, we use these relations to analyze optical activity and circular dichroism in bilayers and multilayer stacks.

## 3 Results and discussion

### 3.1 Chiral bilayer

In a bilayer, the interlayer hopping of excitons leads to the formation of the symmetric and antisymmetric exciton modes denoted by the numbers $m=1$ and $m=2$, respectively [45]. Each of them is two-fold degenerate in the spin index $s$. Diagonalization of the Hamiltonian (1) yields the eigenfrequencies of the modes

$$\omega^{(1)} = \omega_s - J, \quad \omega^{(2)} = \omega_s + J$$

and the eigenfunctions

$$\Phi^{(1,2)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ e^{-i\phi} \end{pmatrix}, \quad \Phi^{(2,1)} = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ e^{i\phi} \end{pmatrix}.$$

The symmetric (bright) mode is efficiently coupled to the electromagnetic field while the coupling of the antisymmetric (dark) mode to the field is very weak [41, 42].

Figure 2A shows the transmission spectra of linearly polarized light for the chiral bilayers. Red and blue curves correspond to the cases of low and high nonradiative exciton decay rate $\Gamma$. The frequencies of the symmetric and antisymmetric exciton modes are indicated by vertical lines. The transmission spectra feature a single dip at the frequency of the symmetric mode with a barely noticeable trace of the antisymmetric mode.

At $J \gg \Gamma_n$, the symmetric and antisymmetric modes are well separated in frequency and do not interact with each other. It follows from Eq. (3) that the interaction of an individual exciton mode $(s, m)$ with the circularly polarized
light propagating along the $z$ axis ($\rightarrow$) or in the opposite direction ($\leftarrow$) is characterized by the rates
\begin{equation}
\gamma^{(s,m)} = \frac{\Gamma_0}{2} \sum_n \Phi_n^{(s,m),\gamma} e^{i\gamma n \phi} \Gamma_n^{(s,m),\gamma},
\end{equation}
where $\Phi_n^{(s,m)}$ are the components of the columns (9). In particular, the rates of exciton-photon interaction in a bilayer are given by
\begin{align}
\gamma^{(1,1)} &= \gamma^{(-1,-1)} = -\frac{\Gamma_0}{2} [1 + \cos(s\phi + qd)], \\
\gamma^{(1,-1)} &= \gamma^{(-1,1)} = \frac{\Gamma_0}{2} [1 - \cos(s\phi + qd)].
\end{align}

The total radiative decay rate of excitons in the mode $(s, m)$ is $\gamma^{(s,m)} = \gamma^{(s,m)} - \gamma^{(s,m)}$. The symmetric exciton mode is superradiant and is coupled to light twice stronger than the exciton mode in a single layer. The width of the dip in the transmission spectrum (Figure 2A) associated with the symmetric exciton mode is about $2\Gamma_0 + \Gamma$ [42, 43]. In contrast, the coupling of the antisymmetric mode to light is very weak being determined by the small parameters $qd$ and $\phi$.

Figure 2B and C show the frequency dependence of the rotation angle $\alpha(\omega)$ and the ellipticity angle $\epsilon(\omega)$, respectively. In contrast to the transmission spectra (Figure 2A), the spectra of optical rotation (Figure 2B) and circular dichroism (Figure 2C) comprise two resonances of the same strength at the frequencies of both exciton modes. Polarization conversion at the frequency of the dark mode is as effective as that at the frequency of the bright mode, whereas the bilayer transparency at the dark mode frequency is very high.

This striking result can be explained as follows. The efficiency of polarization conversion at the exciton mode $m$ is determined by the difference of the strengths of the exciton coupling to the right-handed and left-handed circularly polarized light propagating in the same direction:
\begin{equation}
\Delta\gamma^{(s,m)} = \gamma^{(1,m)} - \gamma^{(-1,m)},
\end{equation}
These values for the bright and dark exciton modes in the chiral bilayer have the form
\begin{equation}
\Delta\gamma^{(1)} = -\Delta\gamma^{(-1)} = -q_d \sin \phi,
\end{equation}
which follows from Eq. (11). As a result, the bright and dark exciton modes are revealed in the spectra of optical rotation and circular dichroism as the resonances of equal strengths and opposite signs.

Analytical solution of Eq. (3) shows that the rotation and ellipticity angles in the relevant case of $qd \ll 1$ are given by
\begin{equation}
\alpha(\omega) + i\epsilon(\omega) = \frac{2\Gamma_0 q_d \sin \phi}{(\omega - \omega_\chi - i\Gamma)(\omega - \omega_\chi + i\Gamma)}.
\end{equation}

The angles are proportional to the chiral hopping parameter $J \sin \phi$, which decays exponentially with the increase of the interlayer distance. Therefore, the interlayer distance should not exceed a few TMDC lattice constants. The widths of the resonance features in $\alpha(\omega)$ and $\epsilon(\omega)$ are determined by the nonradiative exciton decay rate $\Gamma$. In high-quality structures with $\Gamma \ll \Gamma_0$, the resonances are very sharp and the rotation and ellipticity angles reach $|\alpha|_{\max}, |\epsilon|_{\max} = (\Gamma_0 / \Gamma) q_d |\sin \phi|$. In the case of $\Gamma \gg J$, the resonances overlap and partly compensate each other leading to the decrease of the rotation angle and the ellipticity angle by the factor $J / \Gamma$. 
3.2 Multilayer stacks

The stack consisting of \( N \) layers supports \( N \) spatial exciton modes. Diagonalization of the Hamiltonian (2) yields the eigenfrequencies of the modes

\[
\omega^{(m)} = \omega - 2J \cos \frac{m\pi}{(N+1)}
\]

and the eigenfunctions \( \Phi^{(s,m)} \) with the elements

\[
\Phi^{(s,m)} = \frac{2}{\sqrt{N+1}} \sin \frac{m\pi}{N+1} e^{-i\phi}.
\]

where the index \( m \) enumerates the spatial modes and the index \( s \) stands for the spin projection. Neglecting the phase factor, the functions \( \Phi^{(s,m)} \) are either even or odd with respect to the center of the stack. The eigen exciton modes in the stack of \( N = 5 \) layers are sketched in the inset of Figure 3A.

The modes are coupled to the electromagnetic field differently. The rates of exciton-photon interaction for the modes \((s, m)\) calculated after Eq. (10) have the form

\[
\gamma^{(s,m)} = \frac{2\Gamma_0 \cot^2 \frac{m\pi}{2(N+1)}}{N+1} \quad (\text{odd } m).
\]

The radiative decay rates of excitons in the modes with even \( m \) is much lower:

\[
\gamma^{(s,m)} = \frac{\Gamma_0(N+1)\cot^2 \frac{m\pi}{2(N+1)}}{2} \left[ \phi + (qd)^2 \right] \quad (\text{even } m).
\]

In the case of noninteracting modes, \( \omega^{(m+1)} - \omega^{(m)} \gg \gamma^{(m)} + \gamma^{(m+1)} \), the amplitude transmission coefficient of circularly polarized radiation through the stack is the sum of resonant contributions stemming from individual exciton modes

\[
ed^{(s,m)}(\omega) = \frac{1}{N} \sum_{m=1}^{N} \frac{2\gamma^{(s,m)}}{\omega - \omega^{(m)} + i[\gamma^{(s,m)} + \Gamma]} + \sum_{m=1}^{N} \Delta \gamma^{(m)}.
\]

Using the definition Eq. (7), we obtain the expression for the rotation and ellipticity angles:

\[
\alpha(\omega) + i\epsilon(\omega) = \sum_{m=1}^{N} \frac{\Delta \gamma^{(m)}}{\omega - \omega^{(m)} + i\Gamma}.
\]

\[
\Delta \gamma^{(m)} = \frac{\Gamma_0 q \phi}{2(N+1)} \left[ \frac{\cot^2 \frac{m\pi}{2(N+1)}}{\sin^2 \frac{m\pi}{2(N+1)}} \right] \left[ 1 - (-1)^m (N+1)^2 \right].
\]

Figure 3: Optical properties of chiral stacks with \( N = 5 \) layers. 
(A) Transmittance \( |t(\omega)|^2 \), (B) polarization rotation angle \( c(\omega) \), and (C) ellipticity angle \( \epsilon(\omega) \). Vertical lines indicate the frequencies of eigen exciton modes. Inset in Figure 3A shows the distributions of the exciton polarization over the layers for the eigenmodes. Red and blue curves are calculated for the stacks with low and high nonradiative decay rates of excitons, respectively. Parameters are the same as for Figure 2.
Figure 3 shows the spectra of transmission, optical activity, and circular dichroism of the chiral stacks of \( N = 5 \) layers. Red and blue curves correspond to the stacks with low and high nonradiative exciton decay rates \( \Gamma \). The transmission spectrum at low \( \Gamma \) (red curve in Figure 3A) features strong dips at the frequencies of the bright exciton modes (with odd \( m \)) and weak dips at the frequencies of the dark modes (with even \( m \)). The strengths of the resonances of both kinds decrease with the increase of the mode index \( m \). At high \( \Gamma \), the resonances associated with individual exciton modes are widen, and the fine structure of the transmission spectrum is not resolved.

The spectral dependences of the rotation angle (Figure 3B) and the ellipticity angle (Figure 3C) consist of five resonant contributions at the frequencies of eigen exciton modes \( \omega^{(n)} \). The resonances are well resolved in the case of low nonradiative exciton decay rate \( \Gamma \). The strengths of the resonances are determined by \( \Delta \gamma^{(n)} \). As it is seen in Figure 3B and C and also follows from Eq. (22), the resonances at the frequencies of bright and dark exciton modes are of comparable strengths and of opposite signs. Interestingly, the strongest resonance occurs at the frequency of the (dark) mode with \( m = 2 \). At high nonradiative decay rate \( \Gamma \) (see blue curves in Figure 3B and C), the resonances corresponding to the neighboring modes overlap and tend to cancel each other because \( \sum_{m=1}^{N} \Delta \gamma^{(m)} = 0 \).

### 3.3 Thick stacks

Figure 4 shows the spectra of transmission, optical activity, and circular dichroism for the chiral stack of \( N = 10 \) layers. In stacks with large \( N \), the frequencies of eigen exciton modes, indicated by gray vertical lines in Figure 4, fill the miniband from \( \omega_x - 2J \) to \( \omega_x + 2J \). At large enough \( N \), the frequency separation between the neighboring modes \( \Delta \omega = 4J/N \) becomes smaller than the broadening \( \Gamma \), and the individual resonances are overlapped. The resulting angles of optical rotation and ellipticity are determined by the exciton modes with the frequencies close to the miniband bottom \( \omega_b = \omega_x - 2J \); see blue curves in Figure 4C and D.

To describe the optical properties of a thick stack, we replace the set of Eq. (3) for the exciton polarizations in individual layers \( P^{(i)}_s \) with the differential equation for the continuous function \( P_s(z) \)

\[
(\omega + i\Gamma)P^{(s)}(z) - \hat{H}^{(s)}P^{(s)}(z) = -\frac{\Gamma_s}{2\pi qd}E^{(s)}(z), \quad (23)
\]

where \( \hat{H}^{(s)} \) is the Hamiltonian of excitons with the spin projection \( s \) at the miniband bottom

\[
\hat{H}^{(s)} = \omega_b + \frac{k_s^2}{2M} + 2J\phi dk_z, \quad (24)
\]

\( k_z = -i(d/dz), \) \( M = 1/(2d^2) \) is the effective exciton mass, and \( E^{(s)}(z) \) is the amplitude of the electric field. The last term in the Hamiltonian (24) has the form of spin-orbit interaction linear in the exciton wave vector [47]. It is the term originating from the chirality of the stack that gives rise to the optical activity. We also note that the continuous description above is valid provided \( qdL_s \ll \Gamma \), which is well fulfilled in realistic structures.

Equation (23) together with the Maxwell equation

![Figure 4](image-url)

**Figure 4:** Optical properties of chiral stacks with \( N = 10 \) layers. (A) Transmittance \( |t(\omega)|^2 \), (B) polarization rotation angle \( \alpha(\omega) \), and (C) ellipticity angle \( \epsilon(\omega) \). Vertical lines indicate the frequencies of eigen exciton modes. Red and blue curves are calculated for the stacks with low and high nonradiative decay rates of excitons. Parameters are the same as in Figure 2. Dashed green curves in Figure 4B and C are plotted after analytical Eq. (31) and correspond to the limit of thick stacks or high nonradiative decay rates.
form the closed set of differential equations for the functions \( P^{(i)}(z) \) and \( E^{(i)}(z) \). These coupled equations describe the exciton-polaritons in the stack [11, 41].

The solution of Eqs. (23) and (25) in the bulk of the stack has the form \( P^{(i)}(z) \approx \exp(iQz) \) with the dispersion \( \omega(Q) \) given by

\[
Q^2(\omega) = q^2 \left( 1 - \frac{2\Gamma_0/\omega}{\omega - \omega_b + i\Gamma} \right),
\]

where \( Q \) is the exciton-polariton wave vector.

Neglecting the mass term in the exciton dispersion and considering the \( Q \)-linear spin-dependent term as a small correction, we obtain

\[
Q(\omega) = \pm Q_0(\omega) + s\delta Q(\omega),
\]

where

\[
Q_0(\omega) = q\sqrt{1 - \frac{2\Gamma_0/\omega}{\omega - \omega_b + i\Gamma}},
\]

\[
\delta Q(\omega) = -\frac{2\Gamma_0\phi}{(\omega - \omega_b + i\Gamma)^2}.
\]

Figure 5 sketches the dispersions of exciton-polaritons with the spin projections \( s = \pm 1 \) along the wave vector (solid red and blue curves, respectively) given by Eq. (26) for a lossless structure. The exciton-polariton dispersion is formed as a result of the avoided crossing of the light dispersion (dashed black line) and the exciton dispersion (dashed red and blue curves). The exciton spin-orbit interaction described by the last term in the Hamiltonian (24) leads to the spin-orbit splitting of the polariton branches. The splitting is particularly strong for the lower polariton branch at high wave vectors, i.e. at the frequencies close to \( \omega_0 \), where the original exciton splitting is strong and the exciton contribution to the polariton state is high. The spin-orbit splitting of polariton branches leads to a difference in the transmission coefficients for the right-handed and left-handed radiation.

To calculate the transmission coefficients, we solve Eqs. (23) and (25) with the polariton dispersion (27) and the boundary conditions of the continuity of the functions \( E^{(i)} \) and \( dE^{(i)}/dz \) at the front and back surfaces of the stack. Because the major contribution to the rotation and ellipticity angles is proportional to the stack thickness, small possible spin-dependent corrections to the boundary conditions can be neglected. The calculation yields

\[
t^{(i)}(\omega) = \frac{4Q_0g\exp[i(\delta\omega_Q-qL)]}{(Q_0 + q)^2 \exp[-i\theta_0] - (Q_0 - q)^2 \exp[i\theta_0]},
\]

where \( L = (N - 1)d \) is the stack thickness.

Finally, for the rotation and ellipticity angles in thick chiral stacks, we obtain

\[
\alpha(\omega) + i\epsilon(\omega) = \frac{2\Gamma_0qL\phi}{(\omega - \omega_b + i\Gamma)^2}.
\]

The rotation and ellipticity angles grow linearly with the stack thickness. The most pronounced conversion of the light polarization occurs for the light frequencies close to \( \omega_0 \), where the polariton spin-orbit splitting is strongest; see Figure 5. The dependences \( \alpha(\omega) \) and \( \epsilon(\omega) \) calculated after Eq. (31) for the stack of 10 layers are shown in Figure 4B and C, respectively, by green dashed curves. One can see that the analytical dependences agree well with the results of exact numerical calculations (blue curves). With the further increase of the number of layers \( N \) or the nonradiative decay rate of excitons \( \Gamma \), the agreement becomes even better. An estimation after Eq. (31) yields \( \alpha/L \sim 10^2 \) rad/\( \mu \)m and \( \epsilon/L \sim 0.3 \) rad/\( \mu \)m for stacks with low nonradiative decay rate \( \Gamma = 0.1 \) meV and high nonradiative decay rate \( \Gamma = 2 \) meV, respectively, and the other parameters \( \Gamma_0 = 0.3 \) meV [46], \( J = 2 \) meV, \( qd = 0.01 \), and \( \phi = 0.1 \).

4 Polarized PL of chiral stacks

Finally, we discuss the PL of chiral stacks and show that the PL is circularly polarized. To calculate the PL spectrum,
we apply the approach developed in Ref. [48] for multiple quantum well structures. This approach suggests that the PL is caused by random sources of excitons in the layers. The sources of exciton polarization in the layers are supposed to be identical and incoherent.

In this model, the exciton polarizations in the layers are described by Eq. (3) where the right-hand side is replaced with the rate of polarization generation in the layer:

\[
\langle \omega + i\Gamma \rangle P^{(s)}_n - \sum_{n'} \langle H_{n'n'} - i\Gamma e^{i\phi_{n'n'}} \rangle P^{(s)}_{n'} = S_n^{(s)}. \tag{32}
\]

The exciton generations in the layers are independent; therefore, the correlation function has the form

\[
\langle S_{n}^{(s)}S_{n'}^{(s')} \rangle = S^2(\omega)\delta_{n,n'}\delta_{s,s'}. \]

The amplitude of the electric field of the radiation emitted by the stack in the +z direction is given by Eq. (4) with \(E_0^{(s)} = 0\). This yields the expressions for the spectral density of the emitted radiation:

\[
I^{(s)}_+ (\omega) \propto q^2 \sum_n F_n^{(s)} e^{-\gamma_{nn}^0} \tag{33}
\]

and \(I^{(s)}_- (\omega) = I^{(-s)}_+ (\omega)\).

Following the procedure described in the previous section, we calculate the exciton polarizations [Eq. (32)] in the layers and then the PL spectrum [Eq. (33)]. Particularly, in the case of noninteracting exciton modes in the multilayer structure, the spectral density of the PL with the certain circular polarization \(s\) assumes the form

\[
I^{(s)}_+ (\omega) \propto q^2 S^2 \sum_{m} \frac{2\nu^{(s,m)}_+}{(\omega - \omega^{(m)})^2 + (\nu^{(s,m)}_+ + \Gamma)^2}. \tag{34}
\]

Figure 6A–C show the total PL spectra of chiral stacks consisting of \(N = 2, 5, \text{ and } 10\) monolayers. Parameters are the same as in Figure 2.
Figure 6D–F show the spectral dependence of the PL circular polarization degree defined by
\[ P_c(\omega) = \frac{I^{\uparrow}(\omega) - I^{\downarrow}(\omega)}{I^{\uparrow}(\omega) + I^{\downarrow}(\omega)}. \] (35)

The PL is circularly polarized because of the chiral stacking of the layers. In structures with low nonradiative decay rate \( \Gamma \) (red curves), the degree of circular polarization at the frequencies of dark exciton modes is much higher than that at the frequencies of bright modes. The reason is that \( P_c \) of the radiation emitted by the \( m \)-th exciton mode is determined by the ratio \( \Delta \gamma^{(m)} / \gamma^{(m)} \), as follows from Eqs. (34) and (35). While \( \Delta \gamma^{(m)} \) is of the same order for bright and dark modes [Eq. (22)], much smaller value of \( \gamma^{(m)} \) for dark modes than for bright modes [Eqs. (18) and (19)] leads to the higher degree of circular polarization. In stacks with high \( \Gamma \) (blue curves), the contributions of individual exciton modes overlap and partly compensate each other leading to the decrease of the PL circular polarization degree.

5 Summary

To summarize, we have developed a microscopic theory of optical activity and circular dichroism in chiral stacks of 2D crystals, such as the layers of transition metal dichalcogenides, in the spectral range of exciton transitions. The theory takes into account spin-dependent transport of excitons between the layers of the stack and coupling of excitons to an electromagnetic field. We have shown that the frequency dependence of the optical rotation angle and the ellipticity angle exhibits complex behavior reflecting the structure of eigen exciton modes in chiral stacks. In stacks with low nonradiative decay rate of excitons, the spectra of optical activity and circular dichroism comprise sharp resonances associated with individual exciton modes. In stacks with high nonradiative decay rate, the individual resonances overlap and partially compensate each other. The spectra of optical activity and circular dichroism in thick chiral stacks is well described by the developed analytical theory of exciton-polaritons with the effective spin-orbit coupling. We have also calculated the spectra of exciton PL of chiral stacks in the conditions of nonresonant pumping and shown that the PL is circularly polarized. While the consideration was focused on chiral stacks of identical layers, we expect similar polarization optical effects to occur in chiral van der Waals heterostructures in the spectral range of interlayer excitons.

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