Spin Hall Effect of Excitons

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Spin Hall effect for excitons in alkali halides and in Cu2O is investigated theoretically. In both systems, the spin Hall effect results from the Berry curvature in $k$ space, which becomes nonzero due to lifting of degeneracies of the exciton states by exchange coupling. The trajectory of the excitons can be directly seen as spatial dependence of the circularly polarized light emitted from the excitons. It enables us to observe the spin Hall effect directly in the real space-time.

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I. INTRODUCTION

Spin Hall effect (SHE) is attracting interest recently because it can produce spin current without magnetism or magnetic field. The research was triggered by the two theoretical proposals on the intrinsic mechanism on the SHE, and it has been intensively studied both theoretically and experimentally. There are various experiments on the SHE in doped semiconductors and in metals by optical and electrical methods. In these observations in electronic systems, the spin current is seen as an effect summed over many electrons, while the motion of the individual electrons cannot be seen. Therefore, comparison between theory and experiments is sometimes indirect and not straightforward. An experimental method to see directly the electron trajectory is highly desired. At first sight it seems impossible because condensed materials have a huge number of electrons, which cannot be distinguished from each other.

Apart from electronic systems, we have one example where one can observe directly the SHE as a trajectory of the particle: light. As the intrinsic SHE is induced by the Berry phase, it is not limited to electronic systems but also seen in other (even classical) wave phenomena such as light. In this SHE of light, the difference of the refractive indices at an interface of two different media is the first proposal of a real-space observation of the Berry-phase-driven SHE in electronic systems.

II. SPIN HALL EFFECT OF EXCITONS IN ALKALI HALIDES

Due to the spin-orbit coupling, exciton states in alkali halides with the lowest energy consists of an electron in the $\Gamma^+_{5}$ conduction band, and a hole in the $\Gamma^-_{7}$ valence band, and these states are further classified into pure spin-triplet states (total angular momentum $J = 2$) and spin singlet-triplet mixed states ($J = 1$). Exchange interaction and the spin-orbit coupling lifts the degeneracy among these states, and the energies of the $J = 2$ excitons are lower than those of the $J = 1$ due to the analytic exchange interaction. The $J = 1$ excitons are allowed for optical dipolar transition, and are suitable for real-space imaging of the SHE. Meanwhile, the $J = 2$ states are dipolar forbidden. Hence we restrict ourselves to the $J = 1$ excitons. The nonanalytic exchange Hamiltonian with the basis $\{|O_{y}\}, |O_{x}\rangle, |O_{z}\rangle$ within the $J = 1$ states is given by

$$H_{ex}(\vec{K}) = \frac{\Delta_{LT}}{K^2}(K^2 - (\vec{K} \cdot \vec{S})^2),$$

where $\vec{S}$ is the set of the spin-$1$ matrices. $\Delta_{LT}$ is the longitudinal-transverse (L-T) splitting, which can be experimentally determined e.g. from polarization beating of the emission. We neglect higher order terms in $\vec{K}$. In addition, for simplicity, we assume that the analytic exchange (the splitting between $J = 1$ and $J = 2$) is much larger than the nonanalytic one $\Delta_{LT}$. In the calculation of the Berry curvature, this assumption allows us to retain only the matrix elements within the $J = 1$ states among the various matrix elements in the $8 \times 8$ Hamiltonian in the space spanned by $J = 1$ and $J = 2$ states (see...
Longitudinal mode, whose energies differ by $\Delta_{LT}$ splitting gives rise to the Berry curvature for the mechanism is responsible for the SHE of electrons in p-spins move along opposite directions to each other. This Berry phase changes sign when the spin direction of light $\lambda$ is called anomalous velocity, which leads to the SHE. The Berry curvature for the $J=1$ excitons, leading to the SHE.

When the eigenstates are degenerate, a wavepacket follows the semiclassical equations of motion\
\[\dot{\tilde{R}}_c = \frac{1}{\hbar} \frac{\partial \varepsilon_n(\tilde{K}_c)}{\partial \tilde{K}_c} + \tilde{K}_c \times \eta \tilde{F}_n(\tilde{K}_c) \eta, \quad (2)\]
\[\hbar \dot{\tilde{K}}_c = -\frac{\partial V(\tilde{R}_c)}{\partial \tilde{R}_c}, \quad \dot{\eta} = -i \tilde{K}_c \cdot \tilde{A}_n(\tilde{K}_c) \eta, \quad (3)\]

where $\tilde{R}_c, \tilde{K}_c$ are the center position and the wavevector of the wavepacket, $\varepsilon_n(\tilde{K}_c)$ is the energy dispersion of the $n$-th band, $V(\tilde{R}_c)$ is an external potential, and $\eta = (\eta_1, \eta_2)$ is the internal degree of freedom of the two degenerate transverse exciton bands. $\tilde{A}_n(\tilde{K}_c)$ and $\tilde{F}_n(\tilde{K}_c)$ are Berry connection and Berry curvature, which are defined as

\[\begin{bmatrix} A_{\mu}^n(\tilde{K}_c) \end{bmatrix}_{ij} = -i \langle n_i(\tilde{K}_c) | \frac{\partial}{\partial K_\mu} | n_j(\tilde{K}_c) \rangle, \]
\[\begin{bmatrix} F_{\mu}^n(\tilde{K}_c) \end{bmatrix} \equiv \varepsilon_{\mu
u\rho} \left( \frac{\partial A_{\nu}^n(\tilde{K}_c)}{\partial K_\mu} + i A_{\lambda}^n(\tilde{K}_c) A_{\lambda}^\ast_{\mu}(\tilde{K}_c) \right), \quad (4)\]

where $| n_i(\tilde{K}_c) \rangle$ is an eigenstate of the $n$-th band and $i$ is the label for each eigenstate within the degenerate band. The term $\tilde{K}_c \times \eta \tilde{F}_n \eta$ in the equation of motion for $\tilde{R}_c$ is called anomalous velocity, which leads to the SHE. The Berry phase changes sign when the spin direction is reversed. Therefore, two wavepackets with opposite spins move along opposite directions to each other. This mechanism is responsible for the SHE of electrons in p-type semiconductors and that of light.

The Berry curvature for the $J=1$ exciton states can be calculated from $H_{ex}$ in the same way as that in the SHE of light, because the two cases share the same feature of L-T splitting in the spin-1 systems. Therefore the Berry curvature of the transverse states with helicity of $\lambda = \pm 1$ is then calculated as

\[\begin{align*}
\mathcal{F}_n(\tilde{K}_c) &= \lambda \frac{K_\mu}{K^3}.
\end{align*}\]

The longitudinal state ($\lambda = 0$) has a vanishing Berry curvature, and it does not undergo a shift due to the SHE.

We propose an experiment to detect the SHE in the real space and evaluate the Hall shift. The SHE requires a nonzero $\tilde{K}_c$ as seen from Eq. (3). Namely, one should apply an external force to the exciton to see a shift due to the SHE. For electrons an electric field is sufficient, whereas an exciton cannot be accelerated by an electric field. Instead, a local strain gives rise to a potential gradient and accelerates excitons, inducing the SHE. Thus we propose the following setup: we prepare an transverse exciton wavepacket with momentum along the $z$ direction, and apply a uniaxial local strain, so that the excitons feel a force along the $x$ direction, as shown in Fig. 1.

A strain-induced potential well has been developed for Cu$_2$O, but not for alkali halides to our knowledge. Therefore, we estimate the shift from existing data on alkali halides. From the data on the thin-film RbI for example, the effect of uniaxial strain is 25-45 meV for 1 kbar. Because the crystal is easily broken by high uniaxial pressure, we take a lower value for a trapping potential. The force acting on the exciton wavepacket is $3.2 \times 10^{-18} N$, and the corresponding rate of the wavevector change is $K_x \approx 3.0 \times 10^{16} \text{m}^{-1} \text{s}^{-1}$. When we take RbI for example, the typical wavenumber is $k_0 = 0.8 \times 10^5 \text{cm}^{-1}$. The magnitude of the Berry curvature is $F^z = k_0^2 \approx 1.6 \times 10^{-16} \text{m}^2$. Therefore the anomalous velocity is $v_a = K_x F^z \approx 4.8 \text{m/s}$ and the shift is $y_a = v_a \tau \approx 8 \text{nm}$, where $\tau = 1.7 \text{ns}$ is the lifetime of the exciton in RbI, which is governed by self-trapping process. We note that this self-trapping instability can be reduced or avoided by choosing other materials such as III-V or II-VI compounds, AgBr, and TlBr, where the free state of exciton is more stable than the self-trapped state. In these materials, the shift of the excitons could be much longer.

Because of the uncertainty principle, in order for the wavepacket to have a well-defined wavenumber, the size of the wavepacket in $k$ space should be much larger than the wavenumber. Hence the ratio between the size of the exciton wavepacket and the transverse shift is small, and the direct observation of the SHE might be difficult. Nevertheless, a wavepacket deflected to the transverse direction is spin-polarized and emits a circularly polarized light.
light. Therefore, one can observe the SHE by detecting the spatial dependence of the circular polarization from the two wavepackets deflected in the opposite direction.

III. SPIN HALL EFFECT OF ORTHOEXCITON IN Cu₂O

In Cu₂O, the exciton states with the lowest energy, composed of the Γ 2-valence band and the conduction band, is the 1S exciton. Because the valence band and the conduction band share the same parity, radiative recombination of the 1S exciton is dipolar forbidden, and therefore this state has a long radiative lifetime. The four states in the 1S yellow excitons are classified into three Γ 5 orthoexciton states and one Γ 7 paraexciton state. The orthoexcitons are singlet-triplet mixed states, while the paraexciton is purely spin-triplet. Therefore exchange interaction exists only in the singlet states, and affects only the energy of the orthoexcitons, while the paraexcitons remain intact. The energy splitting between ortho and paraexcitons due to the exchange interaction is about 12meV. Furthermore, the degeneracy of the three orthoexciton states is lifted by (nonanalytic) exchange splitting. The matrix form of the exchange interaction among the orthoexciton states \{ |Oyz⟩, |Ozx⟩, |Oxy⟩ \} is given as

\[
H_{ex}(\vec{K}) = \begin{bmatrix}
\Delta_0 (K_0^2 - K^2) + \Delta_3 (3K_x^2 - K^2) & (\Delta_0 (K_y^2 - K_z^2) + \Delta_3 K_x K_y) & (\Delta_0 (K_z^2 + \Delta_3) K_x K_y) & (\Delta_0 (K_z^2 + \Delta_3) K_x K_z) \\
(\Delta_0 (K_y^2 - K_z^2) + \Delta_3 K_x K_y) & \Delta_0 (K_x^2 - K_z^2) + \Delta_3 (3K_y^2 - K^2) & (\Delta_0 (K_z^2 + \Delta_3) K_y K_z) & (\Delta_0 (K_z^2 + \Delta_3) K_y K_x) \\
(\Delta_0 (K_z^2 + \Delta_3) K_x K_y) & (\Delta_0 (K_z^2 + \Delta_3) K_y K_z) & \Delta_0 (K_x^2 - K_z^2) + \Delta_3 (3K_y^2 - K^2) & (\Delta_0 (K_z^2 + \Delta_3) K_x K_z) \\
(\Delta_0 (K_z^2 + \Delta_3) K_x K_z) & (\Delta_0 (K_z^2 + \Delta_3) K_y K_x) & (\Delta_0 (K_z^2 + \Delta_3) K_x K_z) & \Delta_0 (K_x^2 - K_z^2) + \Delta_3 (3K_y^2 - K^2)
\end{bmatrix}.
\]

FIG. 2: (a) Energy dispersion of the exchange interaction and (b) distribution of the Berry curvature \(F_n^{\mu}(\vec{K})\) in Cu₂O. They are shown as a function of the polar angle \(\theta\) of \(\vec{K}\), with the azimuthal angle \(\phi = 45^\circ\). The strain \(\epsilon_{yz}\) is set to be zero.

The wave-vector dependence of the exchange interaction \(H_{ex}\) is illustrated in Fig. 2a). The eigen energies \(E_1(\vec{K})\) and \(E_2(\vec{K})\) are degenerate along the [001] direction and \(E_3(\vec{K})\) and \(E_5(\vec{K})\) are degenerate along the [111] direction. One possible experiment is to make a potential trap exert a force to the exciton, as we considered in alkali halides. In Cu₂O, however, the strain is typically of the order of meV, much larger than the exchange coupling (\(\sim\) \(\mu\)eV). Hence one cannot ignore the strain in the calculation of the Berry curvature. This local strain in general reduces considerably the Berry curvature stemming from the exchange coupling, because of its larger energy scale. To overcome this difficulty, we consider another type of strain: a shear strain its larger energy scale. To overcome this difficulty, we consider another type of strain: a shear strain its larger energy scale. To overcome this difficulty, we consider another type of strain: a shear strain its larger energy scale. To overcome this difficulty, we consider another type of strain: a shear strain its larger energy scale. To overcome this difficulty, we consider another type of strain: a shear strain its larger energy scale. To overcome this difficulty, we consider another type of strain: a shear strain its larger energy scale.

\[
\hat{R}_\mu = \frac{1}{\hbar} \frac{\partial E_n}{\partial K_\mu} - i \eta^\dagger \mathcal{F}_n^{\mu}(\vec{K}) \eta.
\]

with Berry connection and Berry curvature that are defined as

\[
[A_n^\mu(\vec{K})]_{ij} \equiv -i \langle n(\vec{K}) | \frac{\partial}{\partial \epsilon} | n_j(\vec{K}) \rangle,
\]

\[
\mathcal{F}_n^{\mu}(\vec{K}) \equiv \frac{\partial A_n^\mu}{\partial \epsilon} - \frac{\partial A_n^\mu}{\partial K_\mu} + i [A_n^\mu, A_n^\mu],
\]

Because the Hamiltonian matrix \(H_{ex} + H'\) is real, the eigenvectors can be chosen as real. The Berry curvature \(\mathcal{F}^{\mu}(\vec{K})\) is then pure imaginary and Hermitian. If the state considered is nondegenerate, the Berry curvature is scalar (1 \times 1 matrix), and therefore it vanishes. On the other hand, when the state is twofold degenerate, as in [001] or in [111] direction, the Berry curvature is a 2 \times 2 matrix. It is therefore proportional to the Pauli matrix \(\sigma_y\):

\[
\mathcal{F}^{\mu}(\vec{K}) = F^{\mu}(\vec{K}) \sigma_y.
\]

Thus to see the SHE, the exciton states should be degenerate, which occurs along the high-symmetry lines. For
concreteness, we hereafter focus on the twofold degeneracy along the [0,0,1] direction \((\vec{K}\parallel \hat{z})\) as the degenerate bands in the semiclassical equation of motion \(3\). Then the eigenstates \(|n_1(\vec{K})\rangle \) and \(|n_2(\vec{K})\rangle\) with eigenenergies \(E_1(\vec{K})\) and \(E_2(\vec{K})\) in Fig. 2(a) are considered as pseudospin states. Along the \([001]\) direction, these states become \(|O_{yz}\rangle \) and \(|O_{xz}\rangle\).

Figure 2(b) is the distribution of \(F^{\mu}\). We note that \(F^{\mu}\) depends on gauge, even though the anomalous velocity does not, and Fig. 2(b) is based on a particular gauge choice. The typical size of the Berry curvature is expected to be \(F \sim (\Lambda/\Delta_{\text{gap}})k_0\) from consideration of relevant energy scales, where \(\Delta_{\text{gap}}\) denotes the energy gap between the \(|n_1\rangle\) and \(|n_2\rangle\) states and the \(|n_3\rangle\) state. Because \(\Lambda\) and \(\Delta_{\text{gap}}\) are of the order of meV and \(\mu eV\), respectively, this estimate agrees with Fig. 2(b).

In fact, for \(\vec{K}\parallel \hat{z}\) the Berry curvature can be calculated analytically as \(F^{\mu\nu}(\vec{K}) = (\Delta x)/9\Delta_{\text{gap}} k^3\) = \(4.06 \times 10^{-5}\text{m}\), and the other components are zero: \(F^{\nu y}(\vec{K}) = 0\), \(F^{x y}(\vec{K}) = 0\). The reason for the vanishing \(y\) and \(z\) components is the mirror symmetry with respect to the \(yz\) plane, and the twofold rotational symmetry around the \(z\) axis, respectively. Therefore, for \(\vec{K}\) along the \([001]\) direction, the anomalous velocity is along the \(x\) direction. Because the SU(2) Berry curvature \(F^{\mu}(\vec{K})\) is proportional to \(\sigma_y\), we take the eigenvectors of \(\sigma_y\), i.e. \(\frac{1}{\sqrt{2}}(|\uparrow\rangle \pm |\downarrow\rangle)\) (in the \(|n_1\rangle\) \(-|n_2\rangle\) basis), and the semiclassical equations of motion \(3\) is diagonalized. In this basis, the spin \(\eta\) only acquires \(U(1)\) phase in time evolution, but does not change its direction. Therefore, for the wavenumber along the \([001]\) direction, the wavelike for \(|O_{xz}\rangle \pm i|O_{yz}\rangle\) \(10^2\) have opposite anomalous velocity, and their spins are along \(\pm z\), respectively. These excitons emit circularly polarized light depending on its spin state\(22\). This enables us to see this spin Hall shift directly by an optical method.

The anomalous velocity is proportional to \(\dot{\epsilon}\). Therefore, in order to induce the SHE, the strain should be varied externally. One may consider adding an oscillating strain with frequency \(\omega\). Then the typical size of the shift is \(\epsilon(\Lambda/\Delta_{\text{gap}})/k_0 \sim (E_{\text{str.}}/\Delta_{\text{gap}})/k_0\), where \(E_{\text{str.}}\) \((\sim \Lambda)\) is the energy shift of excitons by strain. Thus only the small strain of the order of \(\mu eV\) gives rise to the shift of the order of a wavenumber \(\sim 600\text{nm}\). Although the radiative lifetime is \(\tau_{\text{rad}} \sim 14\text{fs}\), the lifetime of the orthoexcitons is much shorter: \(\tau \sim 3\text{ns}\), due to a nonradia-
tive rapid conversion from orthoexcitons to paraexcitons. The oscillation of the strain \(\epsilon\) should be faster than \(1/\tau\), i.e. be as fast as gigahertz in frequency.

The light emission from the orthoexciton may be reduced by several reasons. First, among all the ortho-
excitons only the fraction of \(\tau/\tau_{\text{rad}} \sim 2 \times 10^{-4}\) emit light. The resolution to detect this emission is it to be well achievable, because the radiative decay rate of excitons has been measured in experiment\(22\). Furthermore, when the density of the orthoexcitons exceed a critical value \((\sim 10^{15}\text{cm}^{-3})\), the spin exchange process between two ortho-
excitons will be effectively convert orthoexcitons into paraexcitons in a short timescale \((\sim 100\text{ps})\). A typical density of excitons by continuous wave (CW) laser is \(10^{13}-10^{14}\text{cm}^{-3}\); it is well below the critical density, and it is not a problem for the proposed experiment. The interaction between orthoexcitons also leads to phase decoherence, but it does not affect the SHE, as Eqs. \(2\) \(3\) remains unaffected. This situation is similar to the electrons in solids, where the mean free path is much shorter than the excitons, but still shows the spin Hall effect. This is because the spin Hall effect is the accumulative effect of the transverse motion of the particles, which does not require the coherence of the process.

IV. SUMMARY AND DISCUSSIONS

In conclusion, we theoretically investigate the SHE of the excitons in allball halides and in \(\text{Cu}_2\text{O}\). The exchange coupling lifts the threefold degeneracy of the ortho-
excitons, while in some directions of the wavenumber double degeneracy remains. This remaining double degener-
gacy gives rise to nonzero SU(2) Berry curvature, leading to the SHE. This SHE can be observed as a position-
dependent circularly polarized light emitted from the ortho-
excitons.

Recently Yao and Niu\(26\) proposed SHE for excitons in GaAs quantum well. In their paper the main contribution to the Berry curvature comes from the heavy-hole light-hole mixing in the quantum well, whereas in the present paper the exchange coupling between the hole and electron spins is the main source of the Berry curvature. Because of the degeneracy of the energy spectrum, the Berry curvature is enhanced in our setup, thereby the SHE becomes prominent. Furthermore, we propose in this paper a realistic setup with target material specified. The proposed setup enables us to use modulation spectroscopy with high precision. This provides us with a space-time resolved measurement of the SHE.

As a closely related subject, an optical SHE has been observed in an exciton-polariton system\(22\), whose mechanism is totally different from the SHE in the present paper. The two different mechanisms for the intrinsic SHE are \(A\) precession due to the \(k\)-dependent (Zeeman-like) field acting on the spin, and \(B\) the anomalous velocity from the \(k\)-space Berry curvature. Although they are often confused with each other, they are dis-
tinct. The mechanism \(A\) is used in the optical SHE in exciton-polaritons\(27,28,29,30\), and in the SHE in the Rashba system\(2\). In these cases the spin-orbit coupling is linear in terms of the spin, which means that the spin-
orbit splitting can be regarded as a “Zeeman-like” field, although the external magnetic field is zero. In these systems the mechanism \(B\) is absent because the contribu-
tion of the Berry curvature cancels between the two bands involved. On the other hand, the mechanism \(B\) causes the SHE in excitons in the present paper, as well as the SHE in the Luttinger model\(1\). This mechanism works even when the Hamiltonian is not linear in the spin.
operator. This effect due to (B) is enhanced when band crossings exist near the Fermi energy, e.g. in the SHE in platinum\textsuperscript{31}, while it is not the case in (A). Moreover, (B) gives an additional spin-dependent (anomalous) velocity and deflects the exciton trajectory, while (A) does not. Thus the mechanisms (A) and (B) are distinct, and the experiments proposed in the present paper allows us a first real-space observation of the Berry-curvature mechanism (B) in electronic systems.

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