Spin Hall effect of excitons

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Spin Hall effect for excitons in alkali halides and in Cu_2O 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. This research was triggered by the two theoretical proposals on the intrinsic mechanism on the SHE,[1,2] and it has been intensively studied both theoretically and experimentally. There are various experiments on the SHE in doped semiconductors and in metals[3–6] 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.[7] As the intrinsic SHE is induced by the Berry phase, it is not only 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 plays the role of the "electric field" in the electronic SHE. The SHE of light at the interface is recently measured in a high accuracy of about 1 Å using weak measurement.[8]

In this paper we theoretically propose a way to optically observe the trajectory of an elementary excitation driven by the SHE. We consider two candidates: transverse excitons in alkali halides and orthoexcitons in Cu_2O. We propose an experimental setup and estimate the shift size due to the SHE, which turns out to be enough magnitude for observation. In both systems, an electron-hole exchange coupling lifts the degeneracy of the excitonic states, which gives rise to the Berry curvature in k space of the center-of-mass motion. It leads to the SHE, namely, spin-dependent trajectory of the excitons. After the radiative lifetime, these excitons emit light, whose circular polarization is determined by the exciton spins. Thus by spatially resolving the circular polarization of the emitted light, we can see how the excitons move in real space in a spin-dependent way. It 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 consist of an electron in the Γ_6^+ conduction band and a hole in the Γ_8^- 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,[9] 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 $\{|O_x\rangle, |O_y\rangle, |O_z\rangle\}$ within the $J=1$ states is given by[10]

$$H_{\mathrm{ex}}(\vec{K}) = \frac{\Delta_{\mathrm{LT}}}{K^2}[K^2 - (\vec{K}\cdot\vec{S})^2], \tag{1}$$

where $\vec{S}$ is the set of the spin-1 matrices. Δ_{LT} is the longitudinal-transverse (LT) splitting, which can be experimentally determined, e.g., from polarization beating of the emission.[11] We neglect higher-order terms in $\vec{K}$. In addition, for simplicity, we assume that the analytic exchange (the splitting between $J=1$ and 2) is much larger than the nonanalytic one Δ_{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 8×8 Hamiltonian in the space spanned by $J=1$ and 2 states (see Ref. 10 and Table VIII in Ref. 12). This Hamiltonian H_{ex} is diagonalized by eigenstates of the helicity $\lambda = (\vec{K}\cdot\vec{S})/K$ with eigenvalues of $\lambda = \pm 1, 0$. Hence, the eigenstates of $H_{\mathrm{ex}}(\vec{K})$ are twofold-degenerate transverse modes and a longitudinal mode, whose energies differ by Δ_{LT}. This LT splitting gives rise to the Berry curvature for the $J=1$ excitons, leading to the SHE.

When the eigenstates are degenerate, a wave packet follows the semiclassical equations of motion,[1,13–15]

$$\dot{\vec{R}}_c = \frac{1}{\hbar}\frac{\partial \varepsilon_n(\vec{K}_c)}{\partial \vec{K}_c} + \dot{\vec{K}}_c \times \eta^\dagger \vec{\mathcal{F}}_n(\vec{K}_c)\eta, \tag{2}$$

$$\hbar \dot{\vec{K}}_c = -\frac{\partial V(\vec{R}_c)}{\partial \vec{R}_c}, \quad \dot{\eta} = -i\dot{\vec{K}}_c \cdot \vec{\mathcal{A}}_n(\vec{K}_c)\eta, \tag{3}$$

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

$$\begin{cases} [\mathcal{A}_n^\mu(\vec{K})]_{ij} \equiv -i\left\langle n_i(\vec{K}) \left| \dfrac{\partial}{\partial K_\mu} \right| n_j(\vec{K}) \right\rangle, \\ \mathcal{F}_n^\rho(\vec{K}) \equiv \epsilon_{\mu\nu\rho}\left[\dfrac{\partial \mathcal{A}_n^\nu(\vec{K})}{\partial K_\mu} + i\mathcal{A}_n^\mu(\vec{K})\mathcal{A}_n^\nu(\vec{K}) \right], \end{cases} \tag{4}$$

where $|n_i(\vec{K})\rangle$ is an eigenstate of the nth band and i is the label for each eigenstate within the degenerate band. The term $\dot{\vec{K}} \times \eta^\dagger \vec{\mathcal{F}} \eta$ in the equation of motion for $\dot{R}_c$ is called anomalous velocity, which leads to the SHE. The Berry phase changes sign when the spin direction is reversed. Therefore, two wave packets with opposite spins move along opposite directions to each other. This mechanism is responsible for the SHE of electrons in p-type semiconductors[1] and that of light.[7]

The Berry curvature for the $J=1$ exciton states can be calculated from $H_{\rm ex}$ in the same way as that in the SHE of light[7] because the two cases share the same feature of LT splitting in the spin-1 systems. Therefore the Berry curvature of the transverse states with helicity of $\lambda=\pm 1$ is then calculated as

$$\mathcal{F}_n^\rho(\vec{K}) = \lambda \frac{K_\rho}{K^3}. \tag{5}$$

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 $\dot{\vec{K}}_c$ 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 a transverse exciton wave packet 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_2O (Ref. 16) but not for alkali halides, to our knowledge.

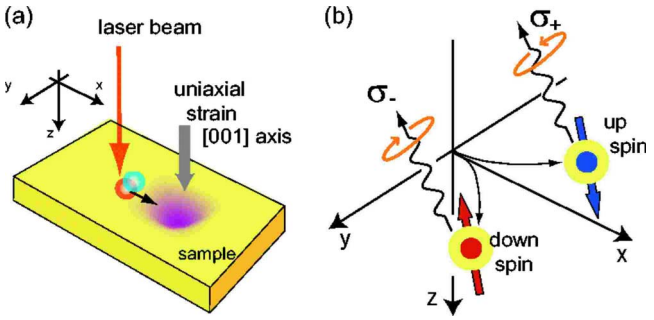

FIG. 1. (Color online) (a) Experimental setup for the detection of SHE in alkali halides. The twofold-degenerate wave packet of the transverse excitons moves toward the center of the uniaxial trapping potential along the x direction. (b) Schematic figure of the spin Hall effect of excitons. The up- and down-spin wave packets are deflected to the $\mp y$ directions, respectively, and they emit light with opposite circular polarizations.

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.[17–19] Because the crystal is easily broken by high uniaxial pressure, we take a lower value for a trapping potential such as 4 meV for 0.1 kbar as an example. We assume the size of the trap to be several hundred micrometers, as developed for Cu_2O.[16] Thus we consider a 200 μm-4 meV configuration of the trapping potential. The force acting on the exciton wave packet is 3.2×10^{-18} N, and the corresponding rate of the wave-vector change is $\dot{K}_x \simeq 3.0\times10^{16}$ m^{-1} s^{-1}. When we take RbI, for example, the typical wave number is $k_0=0.8\times10^6$ cm^{-1}. The magnitude of the Berry curvature is $F^z=k_0^{-2}\simeq 1.6\times10^{-16}$ m^2. Therefore the anomalous velocity is $v_a=\dot{K}_x F^z \simeq 4.8$ m/s and the shift is $y_a=v_a\tau\simeq 8$ nm, where $\tau=1.7$ ns is the lifetime of the exciton in RbI, which is governed by self-trapping process.[20] 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.[21]

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

III. SPIN HALL EFFECT OF ORTHOEXCITON IN Cu_2O

In Cu_2O, the exciton states with the lowest energy, composed of the Γ_7^+ 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 1*S* exciton is dipolar forbidden, and therefore, this state has a long radiative lifetime. The four states in the 1*S* yellow excitons are classified into three Γ_5^+ orthoexciton states and one Γ_2^+ 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 orthoexcitons and paraexcitons due to the exchange interaction is about 12 meV. 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 $\{|O_{yz}\rangle, |O_{zx}\rangle, |O_{xy}\rangle\}$ is given as

$$H_{\mathrm{ex}}(\vec{K}) = \begin{bmatrix} \Delta_Q \frac{K_y^2 K_z^2}{K^2} + \Delta_3(3K_x^2 - K^2) & \left(\Delta_Q \frac{K_z^2}{K^2} + \Delta_5\right) K_x K_y & \left(\Delta_Q \frac{K_y^2}{K^2} + \Delta_5\right) K_z K_x \\ \left(\Delta_Q \frac{K_z^2}{K^2} + \Delta_5\right) K_x K_y & \Delta_Q \frac{K_z^2 K_x^2}{K^2} + \Delta_3(3K_y^2 - K^2) & \left(\Delta_Q \frac{K_x^2}{K^2} + \Delta_5\right) K_y K_z \\ \left(\Delta_Q \frac{K_y^2}{K^2} + \Delta_5\right) K_z K_x & \left(\Delta_Q \frac{K_x^2}{K^2} + \Delta_5\right) K_y K_z & \Delta_Q \frac{K_x^2 K_y^2}{K^2} + \Delta_3(3K_z^2 - K^2) \end{bmatrix}, \tag{6}$$

where the parameters are $\Delta_Q k_0^2 = 5.0\ \mu\mathrm{eV}$, $\Delta_3 k_0^2 = -1.3\ \mu\mathrm{eV}$, $\Delta_5 k_0^2 = 2.0\ \mu\mathrm{eV}$ (Ref. 22) with the wave number $k_0 \equiv 2.62 \times 10^7\ \mathrm{m}^{-1}$, as obtained experimentally from the high-resolution spectroscopy of polaritons.[22]

The wave-vector dependence of the exchange interaction (6) is illustrated in Fig. 2(a). The eigenenergies $E_1(\vec{K})$ and $E_2(\vec{K})$ are degenerate along the [001] direction, and $E_2(\vec{K})$ and $E_3(\vec{K})$ 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 Cu_2O, 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 $\epsilon \equiv \epsilon_{yz}$. The shear strain brings about an additional term to the Hamiltonian as $H'_{ij} = \Lambda \epsilon_{yz}(\delta_{i2}\delta_{j3} + \delta_{i3}\delta_{j2})$. For simplicity we change the normalization of the dimensionless strain parameter $\epsilon \equiv \epsilon_{yz}$ by taking $\Lambda = 8.1$ meV which is the energy shift expected for 5 kbar shear strain that is calculated from the data in Ref. 16. Using this we consider a Berry curvature in the hyperspace of ϵ-$\vec{K}$, which follows[13]

$$\dot{R}_\mu = \frac{1}{\hbar}\frac{\partial E_n}{\partial K_\mu} - \dot{\epsilon}\eta^\dagger \mathcal{F}_n^{\epsilon\mu}(\vec{K})\eta, \tag{7}$$

with Berry connection and Berry curvature that are defined as

$$[\mathcal{A}_n^\epsilon(\vec{K})]_{ij} \equiv -i\left\langle n_i(\vec{K}) \left| \frac{\partial}{\partial \epsilon} \right| n_j(\vec{K}) \right\rangle, \tag{8}$$

$$\mathcal{F}_n^{\epsilon\mu}(\vec{K}) \equiv \frac{\partial \mathcal{A}_n^\mu}{\partial \epsilon} - \frac{\partial \mathcal{A}_n^\epsilon}{\partial K_\mu} + i[\mathcal{A}_n^\epsilon, \mathcal{A}_n^\mu]. \tag{9}$$

Because the Hamiltonian matrix $H_{\mathrm{ex}} + H'$ is real, the eigenvectors can be chosen as real. The Berry curvature $\mathcal{F}^{\epsilon\mu}(\vec{K})$ is then pure imaginary and Hermitian. If the state considered is nondegenerate, the Berry curvature is scalar (1×1 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 2×2 matrix. It is therefore proportional to the Pauli matrix σ_y,

$$\mathcal{F}^{\epsilon\mu}(\vec{K}) = F^{\epsilon\mu}(\vec{K})\sigma_y. \tag{10}$$

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 ($\vec{K} \| \hat{z}$) as the degenerate bands in the semi-

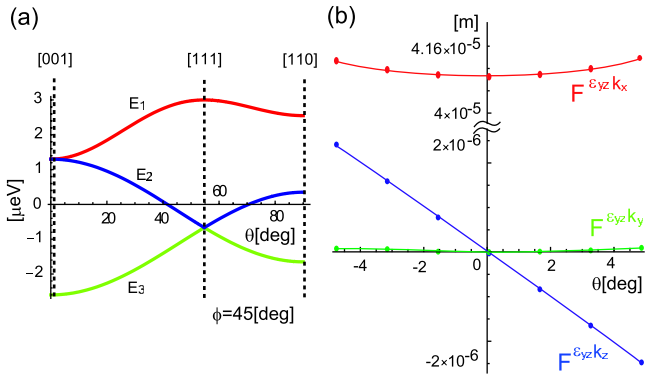

FIG. 2. (Color online) Energy dispersion of the (a) exchange interaction and (b) distribution of the Berry curvature $F_n^{\epsilon\mu}$ in Cu_2O. They are shown as a function of the polar angle θ of $\vec{K}$, with the azimuthal angle $\phi = 45°$. The strain ϵ_{yz} is set to be zero.

classical 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 $|O_{zx}\rangle$.

Figure 2(b) is the distribution of $F^{\epsilon\mu}$. We note that $F^{\epsilon\mu}$ 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 Δ_{gap} denotes the energy gap between the $(|n_1\rangle, |n_2\rangle)$ states and the $|n_3\rangle$ state. Because Λ and Δ_{gap} are of the order of meV and μeV, respectively, this estimate agrees with Fig. 2(b).

In fact, for $\vec{K} \| \hat{z}$ the Berry curvature can be calculated analytically as $\mathcal{F}^{\epsilon x}(\vec{K}) = (\Delta_5 \Lambda)/(9\Delta_3 k_0^3) = 4.06 \times 10^{-5}$ m, and the other components are zero: $\mathcal{F}^{\epsilon y}(\vec{K}) = 0$ and $\mathcal{F}^{\epsilon z}(\vec{K}) = 0$. 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 $\mathcal{F}^{\mu}(\vec{K})$ is proportional to σ_y, we take the eigenvectors of σ_y, i.e., $\frac{1}{\sqrt{2}}\binom{1}{\pm i}$ (in the $|n_1\rangle - |n_2\rangle$ basis), and the semiclassical equations of motion (3) is diagonalized. In this basis, the spin η only acquires U(1) phase in time evolution but does not change its direction. Therefore, for the wave number along the [001] direction, the wave packets for $(|O_{xz}\rangle \pm i|O_{yz}\rangle)/\sqrt{2}$ have opposite anomalous velocity, and their spins are along $\pm z$, respectively. These excitons emit circularly polarized light depending on its spin state.[23] 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 ω. 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 $E_{\text{str}}(\sim \Lambda\epsilon)$ is the energy shift of excitons by strain. Thus only the small strain of the order of μeV gives rise to the shift of the order of a wave number $\sim$600 nm. Although the radiative lifetime is $\tau_{\text{rad}} \sim 14$ μs,[24] the lifetime of the orthoexcitons is much shorter: $\tau \sim 3$ ns, due to a nonradiative rapid conversion from orthoexcitons to paraexcitons. The oscillation of the strain ϵ 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 orthoexcitons only the fraction of $\tau/\tau_{\text{rad}} \sim 2 \times 10^{-4}$ emit light. The resolution to detect this emission is well achievable because the radiative decay rate of excitons has been measured in experiments.[24] Furthermore, when the density of the orthoexcitons exceed a critical value ($\sim 1 \times 10^{15}$ cm^{-3}), the spin-exchange process between two orthoexcitons will effectively convert orthoexcitons into paraexcitons in a short timescale ($\sim$100 ps).[25] A typical density of excitons by continuous wave (CW) laser is 10^{13} cm^{-3} to 10^{14} 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) and (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 alkali halides and in Cu_2O. The exchange coupling lifts the threefold degeneracy of the orthoexcitons, while in some directions of the wave number double degeneracy remains. This remaining double degeneracy 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 orthoexcitons.

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,[27] whose mechanism is totally different from the SHE in the present paper. The two different mechanisms for the intrinsic SHE are (i) precession due to the **k**-dependent (Zeeman-type) field acting on the spin and (ii) the anomalous velocity from the **k**-space Berry curvature. Although they are often confused with each other, they are distinct. The mechanism (i) is used in the optical SHE in exciton polaritons[27–30] 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-type" field, although the external magnetic field is zero. In these systems the mechanism (ii) is absent because the contribution of the Berry curvature cancels between the two bands involved. On the other hand, the mechanism (ii) 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 (ii) is enhanced when band crossings exist near the Fermi energy, e.g., in the SHE in platinum,[31] while it is not the case in (i). Moreover, (ii) gives an additional spin-dependent (anomalous) velocity and deflects the exciton trajectory, while (i) does not. Thus the mechanisms (i) and (ii) are distinct, and the experiments proposed in the present paper allows us a real-space observation of the Berry-curvature mechanism (ii) in electronic systems.

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