

$E_{3/2}(\mathbf{k})$, leading to the optical polarization $(\mathbf{e}_1 + i\mathbf{e}_2)(d_{cv}/\sqrt{2})e^{-iE_{3/2}(\mathbf{k})(t-t_2)}dG_2$ at time t , where $E_{3/2}(\mathbf{k}) = k^2/(2m_e) + k^2/(2m_{3/2})$ is the transition energy of a pair of electron and hole with mass m_e and $m_{3/2}$, respectively. The radiation has the same circular polarization as the input because of the angular momentum conservation. Summation over all possible transitions and integration over time give the linear optical response to the field \mathbf{F}_2 as

$$\begin{aligned} \mathbf{P}^{(1)}(t) &= \frac{i}{3} |d_{cv}|^2 \int_{-\infty}^t e^{-iE_{3/2}(\mathbf{k})(t-t_2)} \\ &\times \sum_{\pm} (1 - f_{\pm}) (\mathbf{e}_1 \mp i\mathbf{e}_2) (\mathbf{e}_1 \mp i\mathbf{e}_2)^* \cdot \mathbf{F}_2 e^{-i\omega_2 t_2} dt_2. \end{aligned} \quad (3)$$

Thus $\mathbf{P}^{(1)} \propto \mathbf{s}_{\mathbf{k}} (\mathbf{e}_1 \mathbf{e}_2 - \mathbf{e}_2 \mathbf{e}_1) \cdot \mathbf{F}_2 = \mathbf{F}_2 \times \mathbf{s}_{\mathbf{k}}$, which has a transparent physical meaning: The linear polarization of the output field is related to that of the input one by a rotation about the spin, essentially a Faraday rotation due to the spin acting as a magnet. When the effect of the intra-band driving by \mathbf{F}_1 is included, the momentum k should be replaced with the accelerated one $\tilde{\mathbf{k}}_{\tau} \equiv \mathbf{k} - e\mathbf{F}_1 \int_{-\infty}^{\tau} \exp(-i\omega_1 t_1) dt_1$ at time τ , and the phase $E_{3/2}(\mathbf{k})(t - t_2)$ accumulated from the creation time t_2 to the recombination time t should be replaced with $\int_{t_2}^t E_{3/2}(\tilde{\mathbf{k}}_{\tau}) d\tau$. By expansion to the linear order of \mathbf{F}_1 , we have $\tilde{k}_{\tau}^2 \approx k^2 - 2e\mathbf{k} \cdot \mathbf{F}_1 \int_{-\infty}^{\tau} \exp(-i\omega_1 t_1) dt_1$, so the second-order optical response can be written as $\mathbf{P} \propto \mathbf{F}_2 \times \mathbf{s}_{\mathbf{k}} e\mathbf{v}_{\mathbf{k}} \cdot \mathbf{F}_1$, where $\mathbf{v}_{\mathbf{k}} \equiv \mathbf{k}/m_e$ is the velocity of the electron with momentum \mathbf{k} . The physical meaning of $e\mathbf{v}_{\mathbf{k}} \cdot \mathbf{F}_1$ is obviously the power done by the field to the electron. $e\mathbf{s}_{\mathbf{k}}\mathbf{v}_{\mathbf{k}}$ is just the spin current tensor contributed by the electron.

For a distribution of electrons, the summation over the momentum space gives the sum-frequency response as $\mathbf{P} = \zeta \mathbf{F}_2 \times (\mathbb{J} \cdot \mathbf{F}_1)$, with

$$\begin{aligned} \zeta &= \left(\frac{\varepsilon_r + 2}{3} \right)^3 \frac{(2/3) |d_{cv}|^2 (1 + m_e/m_{3/2})}{(\omega_1 + \omega_2 - E_{3/2})(\omega_2 - E_{3/2}) \omega_1} \\ &\quad - \left(E_{3/2}, m_{3/2} \rightarrow E_{1/2}, m_{1/2} \right), \end{aligned} \quad (4)$$

derived by Fourier transformation of Eq. (3) including the intra-band driving and the contribution of the SO band, where the factor containing the material dielectric constant ε_r takes into account the difference between the macroscopic external field and the microscopic local field [29], m_j denotes the mass of the spin- j hole band, and E_j is the transition energy from the spin- j band to the Fermi surface [see Fig. 2 (d)]. The constants in Eqns. (1) and (2) are such that $\alpha_1 = -z_2 = z_3 = \zeta$ and others = 0. With the HH-LH splitting neglected, the sum-frequency susceptibility has a compact form with only one independent parameter. This feature is due to the separation of the spin and motion degrees of freedom of the electrons and holes. When the HH-LH splitting