Room temperature spin thermoelectrics in metallic films

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Considering metallic films at room temperature, we present the first theoretical study of the spin Nernst and thermal Edelstein effects which takes into account dynamical spin-orbit coupling, i.e., direct spin-orbit coupling with the vibrating lattice (phonons) and impurities. This gives rise to two novel processes, namely a dynamical Elliott-Yafet spin relaxation and a dynamical side-jump mechanism. Both are the high-temperature counterparts of the well-known $T = 0$ Elliott-Yafet and side-jump, central to the current understanding of the spin Hall, spin Nernst and Edelstein effects at low $T$. We consider the experimentally relevant regime $T > T_D$, with $T_D$ the Debye temperature, as the latter is lower than room temperature in transition metals such as Pt, Au and Ta typically employed in spin injection/extraction experiments. We show that the interplay between intrinsic (Bychkov-Rashba type) and extrinsic (dynamical) spin-orbit coupling yields a nonlinear $T$-dependence of the spin Nernst and spin Hall conductivities.

I. INTRODUCTION

Efficient heat-to-spin conversion is the central goal of spin caloritronics.1 When considering metallic systems, two interesting phenomena stand out in this field: the spin Nernst2,3 and thermal Edelstein effects.4–7 They consist in the generation of, respectively, a spin current or a spin polarization transverse to an applied temperature gradient. That is, they are the thermal counterparts of the well known spin Hall effect and Edelstein effects.8–10 These phenomena are due to spin-orbit coupling and do not require the presence of magnetic textures or Zeeman fields, and are typically classified as intrinsic or extrinsic depending on their origin – respectively band and device structure or impurities.

Spin Hall measurements are typically performed in transition metals such as Au, Pt or Ta9,21–23 where such effects are orders of magnitude larger than in standard semiconductors13 and, very importantly, at room temperature. In this temperature regime the dominant momentum-degrading scattering mechanism in bulk is electron-phonon scattering. Therefore the latter will, through spin-orbit coupling, heavily affect the spin Hall signals. An identical reasoning applies to the Edelstein, thermal Edelstein and spin Nernst effects, though the last two have yet to be experimentally observed. Indeed, the spin-orbit interaction adds an interesting twist to the coupling between electrons and phonons: electrons in a disordered lattice at $T = 0$ move in a “frozen” electrostatic potential $U(r) = V_{\text{cryst}}(r)+V_{\text{imp}}(r)$ arising from the crystal lattice and the impurities, yielding in the Hamiltonian the terms

$$\mathbf{\sigma} \cdot \mathbf{p} + \frac{\lambda_0}{4\hbar} \mathbf{\sigma} \times \nabla U(r) \cdot \mathbf{p},$$

\begin{equation}
U(r),
\end{equation}

$\mathbf{\sigma}, \mathbf{p}$ and $\lambda_0$ being, respectively, the vector of Pauli matrices, the electron momentum and the Compton wavelength. The potential becomes, however, time-dependent at finite $T$, $U(r) \to U(r,t)$. Thus, the lattice (impurity) dynamics will not only give rise to standard electron-phonon (dynamical impurity) scattering through the term $U(r,t)$, but will also couple directly to the carrier spin through the dynamical spin-orbit interaction $\lambda_0 \mathbf{\sigma} \times \nabla U(r,t) \cdot \mathbf{p}/4\hbar$. Remarkably, such a direct “spin-phonon” (“spin-dynamical impurities”) coupling has not yet been studied, and even standard electron-phonon scattering has received minimal attention in the present context. To the best of our knowledge, the only theoretical work considering the impact of standard electron-phonon interaction on the spin Hall effect is that of Grimaldi et al.13 which is focused on a 2-Dimensional Electron Gas (2DEG) with Bychkov-Rashba8 spin-orbit coupling at $T \to 0$.

Our purpose is to start filling this gap, considering the spin Hall, spin Nernst, Edelstein and thermal Edelstein effects in a metallic thin film at room temperature. Moreover, we wish to identify the possible connections between the four phenomena. It is known, for example, that in a 2DEG at low $T$ the spin Hall and Edelstein effects are closely related17,18 and that such a relation can be extended to thin (quasi-2D) films as well.19 Whether this connection exists, possibly in a modified form, also at high $T$ or in 3D is an open question. Another important point concerns the $T$-dependence of the above cited effects. For example, whereas this is expected to be linear for a purely extrinsic spin Nernst effect21, it is not known how the interplay between extrinsic and intrinsic mechanism will modify such behavior. Similarly, for the spin Hall effect its $T$-dependence should allow to establish its specific intrinsic or extrinsic origin.21 The latter is still a somewhat controversial issue, in particular in Au and Pt.22,23

Our treatment relies on two central assumptions. The first one is based on the observation that the Debye temperature $T_D$ of bulk Au (165 K), Pt (240 K) or Ta (240 K) is lower than room temperature, and in this regime electron-phonon scattering is predominantly elastic.24 This leads to a remarkable simplification of the quantum kinetic equations we will employ, allowing to extend to the present case the analysis of the $T=0$ scenario.24 The second one concerns the type of spin-orbit interaction in a metallic film. There is yet no...
general theory capable of identifying its precise effective form, but experiments show that a strong Rashba-like spin-orbit interaction appears at the interface between transition metals and insulators/vacuum, where inversion symmetry is broken; density functional theory has been recently used to estimate its size in Ag, Au or Al on W(110) structures. In general, spin-orbit splittings of up to several hundreds of meV are reported — that is, considerably larger than in a standard GaAs 2DEG. We will thus assume the intrinsic spin-orbit mechanism to be described by a Rashba term in the Hamiltonian. The extrinsic one will be treated in analogy with the semiconductor case, where the spin-orbit interaction with the impurity potential is mediated by an effective Compton wavelength renormalized by the lattice.

Experimentally realized films explore the full 2D to 3D range, thicknesses ranging from one or few monolayers, up to few to tens of nanometers. We will start by considering a strictly 2D metallic layer, and later comment on its 3D counterpart. For the latter case our approach follows the spirit of Ref. [32], which and later comment on its 3D counterpart. For the latter case our approach follows the spirit of Ref. [32], which and later comment on its 3D counterpart. For the latter case our approach follows the spirit of Ref. [32], which

As customary, the static lattice potential $V_{\text{crys}}(\mathbf{r})$ does not appear explicitly anymore, its effects having been incorporated in the effective mass ($m_0 \rightarrow m$) and effective Compton wavelength ($\lambda_0 \rightarrow \lambda$). Above, $\mathbf{\hat{z}}$ is the unit vector pointing towards the metal-substrate interface, whereas $\mathbf{p}, \mathbf{r}$ can be either vectors in the $x$-$y$ plane for strictly 2D films, or also have a $z$-component for thicker, 3D systems. The second term on the r.h.s. is the Bychkov-Rashba intrinsic spin-orbit coupling due to structure symmetry breaking (metal-substrate interface), characterized by a coupling constant $\alpha$, whose strength can be measured by angle-resolved photoemission and estimated by ab-initio methods. $V_{\text{imp}}(\mathbf{r})$ is the random impurity potential, see Sec. III. Impurities give also rise to the fourth term, which represents extrinsic spin-orbit interaction. In the strictly 2D limit the Hamiltonian used to study the spin Hall and related effects. This last one is a nontrivial behavior.

For finite temperatures ($T \neq 0$) the now time-dependent potential $U(\mathbf{r}, t)$ is expanded around its static configuration:

$$U(\mathbf{r}, t) = V_{\text{imp}}(\mathbf{r}) + \delta V_{\text{crys}}(\mathbf{r}, t) + \delta V_{\text{imp}}(\mathbf{r}, t) + \ldots ,$$

where $\delta V_{\text{crys}}(\mathbf{r}, t)$, $\delta V_{\text{imp}}(\mathbf{r}, t)$ are linear in the small ion/impurity displacements. Note that the static lattice potential $V_{\text{crys}}(\mathbf{r})$ has already been effectively taken into account, and so it does not appear in Eq. (3) above. Neither does the phononic term, since we are not interested in the phonon dynamics; the phonons are assumed to be in equilibrium. The Hamiltonian thus becomes

$$H = H_0 + \delta V_{\text{crys}}(\mathbf{r}, t) + \delta V_{\text{imp}}(\mathbf{r}, t) - \frac{\lambda^2}{4\hbar} \mathbf{\sigma} \times \nabla [\delta V_{\text{crys}}(\mathbf{r}, t) + \delta V_{\text{imp}}(\mathbf{r}, t)] \cdot \mathbf{p} .$$ (4)

The second term on the r.h.s. gives rise to the electron-phonon interaction, the third to electron scattering with dynamical impurities, and the fourth describes dynamical spin-orbit coupling (see Fig. 1). This last one is novel and crucial for our purposes, as it yields the dynamical Elliott-Yafet spin relaxation and the dynamical side-jump mechanism. Neither of these two processes have been considered previously, even though their static counterparts are central in $T = 0$ treatments of the spin Hall and related effects. A third potentially relevant process is phonon skew scattering. This will be discussed elsewhere, since its treatment requires going beyond the Born approximation, which is beyond the scope of the present work.

In order to employ the $SU(2)$-covariant kinetic formulation mentioned in the Introduction, the intrinsic Bychkov-Rashba term is rewritten as a non-Abelian vector potential.

$$-\frac{\alpha}{\hbar} \mathbf{p}_i \varepsilon_{i\alpha\beta} \mathbf{a}^\alpha = \frac{p_i A_{\alpha}^i \mathbf{a}^\alpha}{2m} ,$$

II. THE MODEL AND THE ONSAGER FORMULATION

Let us start from the following effective (static) model Hamiltonian for conduction electrons in a parabolic band:

$$H_0 = \frac{\mathbf{p}^2}{2m} - \frac{\alpha}{\hbar} \mathbf{\sigma} \cdot \mathbf{\hat{z}} \cdot \mathbf{p} + V_{\text{imp}}(\mathbf{r}) - \frac{\lambda^2}{4\hbar} \mathbf{\sigma} \cdot \nabla V_{\text{imp}}(\mathbf{r}) \cdot \mathbf{p} .$$ (2)
with $A^x_A = -A^y_A = 2m\alpha/h$, all other components of $A^x$ being zero, whereas $\varepsilon_{x\alpha z}$ is the $z$-component of the antisymmetric tensor. Here and throughout the paper upper (lower) indices will indicate spin (real space) components, while repeated indices are summed over unless otherwise specified.

The final step is defining the relevant transport coefficients within linear response. Assuming homogeneous conditions and taking as driving fields an electric field $E_x$ and a temperature gradient $\nabla_x T$, we are interested in the generation of (i) a $y$-spin polarization $s^y$ (Edelstein\cite{22} and thermal Edelstein\cite{23} effects); (ii) a $z$-polarized spin current flowing along $y$, $j^z_y$ (spin Hall\cite{4} and spin Nernst\cite{23} effects). In the presence of spin-orbit coupling, i.e., when spin is not conserved, the spin current has a diffusion term even under homogeneous conditions\cite{35},

$$j^z_y = 2maDs^y + j^z_y\text{drift},$$

with $D$ the diffusion constant. Extending the standard Onsager formulation of thermoelectric transport to the present spin-thermoelectric context, we then write

$$s^y = P_{SE} E_x + P_{ST} \nabla_x T,$$

$$j^z_y\text{drift} = \sigma_{SE}\text{drift} E_x + \sigma_{ST}\text{drift} \nabla_x T.$$  

The conductivities $\sigma_{SE}\text{drift}, \sigma_{ST}\text{drift}$ correspond, in Kubo diagrammatic, to “bare” response bubbles. For the full spin current $j^z_y$ one has

$$j^z_y = \sigma_{SE} E_x + \sigma_{ST} \nabla_x T,$$

where $\sigma_{SE}, \sigma_{ST}$ are bubbles with “dressed” vertices, the same holding for $P_{SE}, P_{ST}$. The spin Hall conductivity $\sigma^{3H} \equiv \sigma_{SE}$, whereas the spin Nernst one is defined under open circuit conditions, $\sigma^{SN} \equiv S\sigma_{SE} + \sigma_{ST}$, with $S$ the Seebeck coefficient. Similarly, the Edelstein effect is directly given by the spin polarization response to the electric field, $P \equiv P_{SE}$, while for its thermal counterpart $P^t \equiv SP_{SE} + P_{ST}$.

Our goal is the computation of the transport coefficients $P_{SE}, P_{ST}, \sigma_{SE}, \sigma_{ST}$ defined above. For the sake of clarity we have introduced them within a drift-diffusion picture, however Eqs. (7) and (9) are general, and our treatment works in the ballistic limit as well. Finally, Onsager reciprocity is duly respected\cite{24,25} and is here between $j^z_y \leftrightarrow j_x$ (spin Hall $\leftrightarrow$ inverse spin Hall effect) and $s^y \leftrightarrow j^z_y$ (Edelstein $\leftrightarrow$ inverse Edelstein or spin-galvanic effect\cite{22}.

**III. THE KINETIC EQUATIONS**

The kinetic (Boltzmann-like) equation for the $2 \times 2$ distribution function $f_p = f^0 + \mathbf{v} \cdot \mathbf{f}$, where $f^0$ and $\mathbf{f}$ are the charge and spin distribution functions, respectively\cite{25} reads

$$\partial_t f_p + \nabla \cdot \left[ \frac{P}{m} f_p + \Delta j_{ij} \right] + \frac{1}{2} \{ \mathbf{F} \cdot \nabla f_p, f_p \} = I_0 + I_{ij} + I_{EY},$$

where we introduced the covariant spatial derivative and the SU(2) Lorentz force due to the Rashba spin-orbit coupling:

$$\nabla = \nabla + \frac{i}{\hbar} \left[ A^x, \frac{\sigma^x}{2} \right],$$

$$\mathbf{F} = -\frac{\mathbf{P}}{m} \times \mathbf{B} \frac{\sigma^x}{2},$$

$$B^y_j = -\frac{1}{2\hbar} \varepsilon_{ijk} \sigma^{abc} A^i_j A^k.$$  

A summation over identical indices is implied unless stated otherwise. Note that an external magnetic field is not included in these equations (since it is not needed for the present purpose). The term $\Delta j_{ij}$ in Eq. (10) is a correction to the current due to side-jumps.

Next we consider the collision operators on the r.h.s. of Eq. (10), where $I_0$ describes scattering with dynamical impurities and phonons, $I_{ij}$ the contribution due to side-jumps, and $I_{EY}$ Elliott-Yafet spin relaxation due to spin-flip processes. At zero temperature the collision operators are obtained from the impurity averaged self-energies within the self-consistent Born approximation (see Fig. 1). For isotropic scattering, the impurity correlations are given by

$$V_{imp}(\mathbf{r})V_{imp}(\mathbf{r}') = n_{imp} v_0^2 \delta(\mathbf{r} - \mathbf{r}') = \frac{\hbar}{2\pi N_0 \tau_{imp}} \delta(\mathbf{r} - \mathbf{r}'),$$

with $n_{imp}$ the impurity concentration, $v_0$ the scattering amplitude, and $1/\tau_{imp}$ the momentum relaxation rate due to impurities; $N_0$ is the density of states per area (volume) and spin in two (three) dimensions. More generally, $v_0^n \rightarrow \langle |\mathbf{v}(\mathbf{q})|^2 \rangle$, where $\langle \ldots \rangle$ denotes the angular average, and $\mathbf{q}^2 = (\mathbf{p} - \mathbf{p}')^2/\hbar^2 = 2m^2 (1 - \cos \theta)$, since $|\mathbf{p}| = |\mathbf{p}'| = p_F$.

In order to include the impurities’ thermal fluctuations, we consider small time-dependent displacements $\delta r_i(t)$ of the $i$-th impurity, which leads to

$$\delta V_{imp}(\mathbf{r}, t) = -\nabla \cdot \sum_i \delta r_i(t) v(\mathbf{r} - \mathbf{r}_i),$$

where $v$ is the single-impurity potential. We further assume that the displacement fluctuations of different impurities are independent, and can be approximated by the classical harmonic oscillator expression, i.e.,

$$\frac{\delta r_i^a(t) \delta r_j^b(t')}{M \omega_D^2} \approx \delta_{ij} \delta_{ab} \frac{k_B T}{M \omega_D^2},$$

where $M$ and $\omega_D$ are the typical mass and frequency; we also considered short times, $\omega_D |t - t'| \ll 1$. Then we obtain

$$\frac{\delta V_{imp}(\mathbf{r}, t) \delta V_{imp}(\mathbf{r}', t')}{2\pi N_0 \tau_{dyn}} \approx \frac{\hbar}{2\pi N_0 \tau_{dyn}} \delta(\mathbf{r} - \mathbf{r}')$$

with

$$\frac{1}{\tau_{dyn}} = 2\pi n_{imp} v_0^2 N_0 \frac{2k_B T p_F^2}{\hbar^2 M \omega_D^2}.$$
FIG. 1. Shown are the self-energies which determine the collision operators in the Boltzmann equation. The arrowed line represents the Green’s function in Keldysh space, a cross (dot) the potential due to an impurity (a crystal displacement). The dashed line depicts the impurity correlation either for static (straight line) or for dynamical impurities (wavy line). The wavy solid line illustrates the phonon propagator and a box around a vertex the spin-orbit coupling due to the boxed potential.

More precisely, as follows from the corresponding self-energy expression (Fig. 1), \( v_0^2 \rightarrow \langle (1 - \cos \theta) |v(q)|^2 \rangle \) in [18]. In order of magnitude, \( \gamma_{\text{imp}}/\gamma_{\text{dyn}} \simeq k_B T/\epsilon_F \) since \( \hbar \omega_{D}^2 \simeq (m/M)^2 \epsilon_F^2 \). Note that the \( \delta \)-function in Eq. (17) has to be interpreted in connection with the corresponding self-energy diagram. A detailed analysis shows that the result given in Eq. (18) applies for high temperatures, \( k_B T \gg \hbar \omega_D \), where scattering processes essentially are elastic.

A similar reasoning can be employed for electron-phonon scattering at high \( T \), which leads to

\[
\delta V_{\text{cryst}}(r, t) \delta V_{\text{cryst}}(r', t') \simeq \frac{\hbar}{2\pi N_0 \tau_{\text{ph}}} \delta(r - r'), \tag{19}
\]

where \( 1/\tau_{\text{ph}} = 2\pi N_0 g^2 k_B T/\hbar \) is the standard (high \( T \)) momentum relaxation rate. Based on the Keldysh technique, the collision operators can be derived as usual. The result corresponds, in the classical limit, to

\[
\delta V_{\text{cryst}}(r, t) \delta V_{\text{cryst}}(r', t') = \frac{ig^2}{2} D^K (r - r', t - t'), \tag{20}
\]

where \( g \) is the electron-phonon coupling constant and \( D^K \) denotes the Keldysh component of the phonon Green’s function in equilibrium.

Since \( 1/\tau_{\text{ph}} \) can be several orders of magnitude larger than \( 1/\tau_{\text{imp} \text{ or } \text{dyn}} \), the total momentum relaxation rate \( 1/\tau = 1/\tau_{\text{imp}} + 1/\tau_{\text{dyn}} + 1/\tau_{\text{ph}} \) is typically dominated by electron-phonon scattering, \( 1/\tau \simeq 1/\tau_{\text{ph}} \), in the high-temperature regime.

The above discussion shows that one may use the results for the collision operators and the side-jump correction given in Refs. [18] and [34],

\[
I_0 = -\frac{1}{\tau} \langle f_p - \langle f_p \rangle \rangle, \tag{21}
\]

\[
I_{\sigma_j} = \frac{\lambda^2}{8\hbar^2} \varepsilon_{abc} \left\{ \left( \nabla_a \sigma^b \right) p_c f_p - \langle p_c f_p \rangle \right\}, \tag{22}
\]

\[
I_{\text{EY}} = -\frac{1}{\tau} \left( d - 1 \right) \left( \frac{\lambda p}{2\hbar} \right)^4 \times \sum_{a=x,y,z} \left( \frac{1}{3} g^2 - f^a + \langle f^a \rangle \right) \sigma^a \tag{23}
\]

\[
\Delta j_{\sigma_j} = \frac{\lambda^2}{8\hbar^2} \langle \left\langle (p' - p) \times \sigma, f_{p'} \right\rangle \rangle_{p'}, \tag{24}
\]

where \( 1/\tau \) is now the total scattering rate. The wavy brackets represent the anti-commutator and \( d = 2, 3 \) the dimensionality. Formally, the diagrams in Fig. 1 together with Eqs. (21), (23), show that the phenomenological substitution \( 1/\tau_{\text{imp}} \rightarrow 1/\tau \) for \( T = 0 \rightarrow T \neq 0 \) is fully justified for all spin-dependent processes at the Born approximation level of accuracy.

Finally, the \( y \) spin polarization \( s^y \) and the \( z \)-polarized spin current flowing along \( y, j^z_y \), are defined according to Ref. [18].

\[
s^y = \int \frac{dp}{(2\pi \hbar)^d} f^y \int d\epsilon_p N_0 \langle f^y \rangle, \tag{25}
\]

and

\[
j^z_y = \frac{\text{Tr}}{2} \int \frac{d\epsilon_p}{(2\pi \hbar)^d} \left[ \frac{p_y}{m} f_p + \frac{\lambda}{8\hbar} \left\langle \left\langle (p \times \sigma) \right\rangle \right\rangle_{p_y} f_p \right]. \tag{26}
\]

### IV. SPIN NERNST AND THERMAL EDELSTEIN EFFECTS

In this section we present and discuss our results, i.e., the spin transport coefficients \( P_{\text{SC}}, P_{\text{DT}}, \sigma_{\text{SC}}, \) and \( \sigma_{\text{DT}} \). We find that the competition between intrinsic and extrinsic spin-orbit mechanisms can lead the former to have a non-linear temperature dependence. Notice that when only extrinsic mechanisms are considered, the spin Nernst conductivity was instead predicted to be simply linear in \( T \) Though the spin Nernst nonlinearity will prove to be relevant for certain experimentally realized systems. Indeed, spin diffusion takes place as long as the spin-orbit splitting is smaller than the lifetime broadening, which
in a Rashba-like system means $2 \alpha p_F / h < h / \tau$, $p_F$ being the Fermi surface momentum. At room temperature $h / \tau \approx 10^{-2}$ eV, whereas $2 \alpha p_F / h$ can vary substantially in metallic films, $10^{-3}$ eV $\lesssim 2 \alpha p_F / h \lesssim 10^{-1}$ eV. Thus, the full diffusive-to-ballistic spectrum can in principle be explored.

In the diffusive regime the Boltzmann equation (10) for $\langle f^y \rangle$ can be solved within the $p$-wave approximation $(f_p \approx \langle f^y \rangle + \mathbf{p} \cdot \mathbf{\partial f_p})$, in terms of the $x$-spatial derivative of the local equilibrium charge distribution function,

$$\nabla_x f^{eq} = \left( \frac{\epsilon_p - \epsilon_F}{T} \nabla_x T + \nabla_x \mu \right) \left( - \frac{\partial f^{eq}}{\partial \epsilon_p} \right).$$  \hspace{1cm} (27)

Here $\epsilon_p(\epsilon_F)$ is the particle (Fermi) energy. The chemical potential gradient is identified with the electric field, $eE_x \equiv \nabla_x \mu$ with $e = |e|$. The temperature gradient and the electric field act as driving terms in the charge sector of the Boltzmann equation, which is easily solved. Via Eqs. (22) and (24), the charge distribution enters the spin sector, from which we determine $\langle f^y \rangle$ and hence the spin polarization linear in $\nabla_x T$ according to Eq. (25). In the last step, integrating the $y$ spin component of Eq. (19) we obtain

$$\partial_t s^y + \frac{2m\alpha}{h^2} j_z^y = - \int d\epsilon_p N_0 \tau_s (f^y).$$  \hspace{1cm} (28)

From this relation, we then calculate $j_z^y$. Note that no spatial gradients (beyond $\nabla_x T$ and $\nabla_x \mu$) are considered. In Eq. (28), the (weakly energy-dependent) Elliott-Yafet relaxation rate is proportional to the momentum relaxation rate, and given by

$$\frac{1}{\tau_s} = \frac{1}{\tau} \left( \frac{\lambda p}{2h^2} \right)^4.$$  \hspace{1cm} (29)

Specifically, in order to obtain $\langle f^y \rangle$ we perform a Fourier transformation in time, $t \to \omega$, multiply the $z$ spin component of the Boltzmann equation by $p_y$, the charge component by $p_x$, and perform the momentum angular average of these two equations as well as of the $y$ spin component of the Boltzmann equation. The result is

$$\langle f^y \rangle = - F_\omega \cdot \nabla_x f^{eq}$$  \hspace{1cm} (30)

with

$$F_\omega = p^2 \frac{\alpha}{h^3} \frac{\tau_s}{1 - i \omega \tau} \left[ 2 \left( \frac{\alpha \tau}{h} \right)^2 + \frac{\lambda^2}{2} (1 - i \omega \tau) \right]$$

$$\times \left[ 2 \left( \frac{4 \alpha \tau}{\lambda^2 p} \right)^2 + (1 - i \omega \tau_s)(1 - i \omega \tau) \right]^{-1}.$$  \hspace{1cm} (31)

From this expression, we are now able to determine the transport coefficients, similar to Mott’s formula in thermoelectrics. We find

$$P_{SE}(\omega) = - e \int d\epsilon_p N_0 F_\omega \left( - \frac{\partial f^{eq}}{\partial \epsilon_p} \right),$$  \hspace{1cm} (32)

$$P_{ST}(\omega) = - \int d\epsilon_p N_0 F_\omega \left( - \frac{\epsilon_F - \epsilon_F}{T} \right) \left( - \frac{\partial f^{eq}}{\partial \epsilon_p} \right),$$  \hspace{1cm} (33)

$$\sigma_{SE}(\omega) = \frac{e^2}{2 \alpha \tau_s} \int d\epsilon_p N_0 \tau_s (1 - i \omega \tau_s) F_\omega \left( - \frac{\partial f^{eq}}{\partial \epsilon_p} \right),$$  \hspace{1cm} (34)

$$\sigma_{ST}(\omega) = \frac{\hbar^2}{2 \alpha \tau_s} \int d\epsilon_p N_0 \tau_s (1 - i \omega \tau_s) F_\omega \left( - \frac{\partial f^{eq}}{\partial \epsilon_p} \right).$$  \hspace{1cm} (35)

In the following we consider the first non-vanishing order of the Sommerfeld expansion of Eqs. (32)–(35). Also, all energy-dependent quantities are given at the Fermi energy unless mentioned otherwise. The following Mott-like formulas are obtained:

$$P_{ST} = - S_0 e \alpha P'_{SE},$$  \hspace{1cm} (36)

$$\sigma_{ST} = - S_0 e \alpha \sigma'_{SE},$$  \hspace{1cm} (37)

with $S_0 = - \pi^2 k_f^2 T / (3e \alpha \epsilon_F)$, $P'_{SE} \equiv \partial_{\epsilon_p} P_{SE} \mid_{\epsilon_F}$, and $\sigma'_{SE} \equiv \partial_{\epsilon_p} \sigma_{SE} \mid_{\epsilon_F}$.

First we discuss the simple case of a 2DEG with an energy-independent relaxation rate $1 / \tau$ in the static case ($\omega = 0$). We refer to App. A for more general formulas. Concerning the spin polarization, we find

$$P_{SE} = - \frac{2m\alpha}{h^2} \frac{\tau_s}{\tau_s / \tau_{DP} + 1} \left( \sigma_{int}^{sH} + \sigma_{sE}^{sH} \right),$$  \hspace{1cm} (38)

$$P_{ST} = - S_0 \frac{2m\alpha}{h^2} \frac{\tau_s}{\tau_s / \tau_{DP} + 1} \left( \sigma_{int}^{sH} + \sigma_{sE}^{sH} \right).$$  \hspace{1cm} (39)

Here, $1 / \tau_{DP} = (2m \alpha / h^2)^2 D$ is the Dyakonov-Perel relaxation rate in the diffusive regime, with $D = v_F^2 / \tau$ the diffusion constant, whereas $\sigma_{int}^{sH} = (N_0 e h / 4m) / (2 \tau / \tau_{DP})$ and $\sigma_{sE}^{sH} = e \lambda^2 / (4 \hbar)$ are the intrinsic and side-jump spin Hall conductivity, respectively. Note that for a 2DEG we have $N_0 e h / 4m = e / 8 \tau_{DP}$ giving the “universal” intrinsic spin Hall conductivity. Clearly, $P_{ST}$ is in general nonlinear in temperature due to the $T$-dependence of the spin relaxation rates,

$$\frac{1}{\tau_{DP}} \sim \tau \sim \frac{1}{T}, \quad \frac{1}{\tau_s} \sim \frac{1}{\tau} \sim T.$$  \hspace{1cm} (40)

An experimental relevant setup would be an open circuit along $x$, i.e., along the direction where the thermal gradient is applied. Then, the electric field can be expressed by the thermal gradient as $E_x = S \nabla_x T$, where $S$ is the Seebeck coefficient. For a 2DEG with an energy-independent relaxation rate $S = S_0$. With the open circuit condition the thermal Edelstein polarization coefficient is given as a sum of electrical and thermal contributions, and reads

$$\mathcal{P} \equiv S_0 P_{SE} + P_{ST},$$  \hspace{1cm} (41)

which is shown in Fig. 2 for $\tau / \tau_s = 0.01$. The parameter $\tau_{s,r} / \tau_{DP,r}$, the subscript $r$ indicating that the value of a
The interplay of intrinsic and extrinsic spin-orbit coupling leads to a nonlinear temperature dependence, provided the intrinsic spin-orbit coupling dominates [Fig. 3b]). On the other hand, when intrinsic and extrinsic spin relaxation times are comparable [Fig. 3a]), the spin Nernst conductivity is small since the thermal and the electrical contribution cancel each other. Indeed, for vanishing intrinsic-spin-orbit coupling, the spin Nernst conductivity is zero for a 2DEG.

Finally, we comment on the three-dimensional case. As can be seen in App. A the quantitative change is rather small since only $\tau_s$ changes by a numerical prefactor of 8/9, while the other relevant quantities remain unchanged. We remark, however, that in 3D we encounter an energy-dependent density of states. In addition, the momentum relaxation rate is in general also energy-dependent. This manifests itself directly in the thermal part of the spin transport coefficients where we encounter the factors $\eta = \epsilon_F N_0/N$ and $\beta = \epsilon_F \tau'/\tau$, namely the change in energy of the density of states and the momentum relaxation rate at the Fermi energy. Therefore, the relative weight between thermal and elec-
trical contribution can be modified. Note that in case of an open circuit along the thermal gradient, the electrical contribution is also modified by $\eta$ and $\beta$ since the Seebeck coefficient is then given by $S = S_0(1 + \eta + \beta)$, as follows from the charge component of the Boltzmann equation.

A. The “clean” limit

At room temperature, one enters the “clean” regime for $2eV_F/h > 10^{-2} eV$. Under homogeneous conditions, Eq. (10) can be solved in this limit as well – note that Eq. (28) is valid irrespective of the regime considered. The procedure is straightforward, yet lengthy and not particularly illuminating; therefore we simply give the results for the 2D case when $2eV_F/h^2 \gg 1$.

The transport coefficients read

$$ P_{SE} = -\frac{2m_0}{h^2} 2\tau \left( \sigma^{SH}_{int} + \sigma^{SH}_{sj} \right) / 2 \, ,$$

$$ P_{ST} = S_0 \frac{2m_0}{h^2} 2\tau \sigma^{SH}_{sj} \, ,$$

$$ \sigma_{SE} = \frac{2\tau}{\tau_s} \left( \sigma^{SH}_{int} + \sigma^{SH}_{sj} \right) \, ,$$

$$ \sigma_{ST} = -S_0 \frac{2\tau}{\tau_s} \sigma^{SH}_{sj} \, ,$$

where now $\sigma^{SH}_{int} = e/8\pi h$. From Eqs. (41) and (44) it is immediate to see that the thermal Edelstein effect is constant in $T$, whereas the spin Nernst is linear. This overall simpler behavior is expected, as in the “clean” limit $1/\tau_{DP} \rightarrow 1/2\tau$, i.e., both the Dyakonov-Perel and the Elliott-Yafet relaxation rates are proportional to $T$.

V. CONCLUSIONS

We have explicitly considered the dynamical spin-orbit interaction of conduction electrons with phonons, which gives rise to dynamical Elliott-Yafet spin relaxation and side-jump mechanism. The focus has been on the high-temperature regime $T > T_D$. Symmetric, Mott-like formulas for the (thermal) Edelstein and spin Hall and Nernst coefficients have been derived. The temperature-dependence of the spin transport coefficients was shown to be nontrivially affected by the competition between extrinsic and intrinsic spin-orbit coupling mechanisms, the origin lying in the mixing of the spin relaxation times $\tau_{DP}$ and $\tau_s$. In the diffusive regime the latter have different temperature dependences, which ultimately causes the thermal Edelstein and spin Nernst effect to exhibit a nonlinear $T$-behavior. The nonlinearity is in general stronger for the thermal Edelstein effect, and, especially in the spin Nernst case, it becomes weaker with decreasing intrinsic spin-orbit coupling strength.

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Appendix A: General Expressions for the spin polarization/current

This appendix shows more general expressions for $P_{SE}, P_{ST}, \sigma_{SE},$ and $\sigma_{ST}$, valid at finite frequency for both 2D and 3D systems. The transport coefficients are obtained by the Sommerfeld expansion of Eqs. (32–35). This implies that all quantities appearing below are evaluated at the Fermi energy unless otherwise specified.

The dynamical Edelstein coefficient and the spin Hall conductivity are given as follows:

$$ P_{SE} = -\frac{2m_0}{h^2} \left[ \frac{\tau_s}{\tau_{DP}} + d(1-i\omega\tau_s)(1-i\omega\tau) \right] \times \left[ \frac{2\sigma^{SH}_{int} + d\sigma^{SH}_{sj}(1-i\omega\tau)}{1-i\omega\tau} \right] \, ,$$

$$ \sigma_{SE} = \frac{1-i\omega\tau_s}{\tau_s} \left[ \frac{\tau_s}{\tau_{DP}} + d(1-i\omega\tau_s)(1-i\omega\tau) \right] \times \left[ \frac{2\sigma^{SH}_{int} + d\sigma^{SH}_{sj}(1-i\omega\tau)}{1-i\omega\tau} \right] \, .$$

Here, the form of $\sigma^{SH}_{int}$ and $\sigma^{SH}_{sj}$ [see Eq. (39)] remains unchanged in 3D and the Dyakonov-Perel relaxation rate remains exactly as it is in 2D, i.e., the diffusion constant which there appears is the 2D one. Only the Elliott-Yafet relaxation rate exhibits a pre-factor of $8/9$ compared to $\tau_s$ in 2D. A plot of the spin Hall conductivity $\sigma_{SE}(\omega)$ is shown in Fig. 4.

FIG. 4. The spin Hall conductivity in 3D in units of $N_0 e\hbar/4m$ for various ratios of $\tau_s/\tau_{DP}$ vs. $\omega\tau$, separated into its real part (solid lines) and its imaginary part (dashed lines). The extrinsic spin-orbit strength is chosen such that $\tau/\tau_s = 0.01$. 

$\tau_s/\tau_{DP} = 1$
$\tau_s/\tau_{DP} = 20$
$\tau_s/\tau_{DP} = 100$
The thermal contribution is now obtained by Eqs. (36) and (37). Since the resulting equations are rather cumbersome, we just show formulas for the static case, \( \omega = 0 \). We find

\[
P_T = -S_0 \frac{2\alpha_0}{\hbar^2} \frac{\tau_s}{(2\tau_s/\tau_{DP} + d)^2} \times \left\{ 2\sigma_{\text{int}}^H \left[ d - \eta \left( \frac{2\tau_s}{\tau_{DP} + d} \right) - \beta \left( \frac{2\tau_s}{\tau_{DP} + 3d} \right) \right] + d\sigma_{\text{int}}^{\text{H}} \left[ d - \eta \left( \frac{2\tau_s}{\tau_{DP} + d} \right) + \beta \left( \frac{2\tau_s}{\tau_{DP} - d} \right) \right] \right\},
\]

\[
\sigma_{\text{ST}} = -S_0 \frac{1}{(2\tau_s/\tau_{DP} + d)^2} \times \left\{ 2\sigma_{\text{int}}^H \left[ \frac{4\tau_s}{\tau_{DP}} + d + \eta \left( \frac{2\tau_s}{\tau_{DP} + d} + 2d\beta \right) \right] + d\sigma_{\text{int}}^{\text{H}} \left[ \frac{4\tau_s}{\tau_{DP}} + d + \eta \left( \frac{2\tau_s}{\tau_{DP} + d} \right) \right] \right\}. \tag{A3}
\]

Note that here the energy derivative of the density of states (momentum relaxation rate) at the Fermi energy comes into play by \( \eta = \epsilon_F N_0 / N_0 (\beta = \epsilon_F \tau / \tau) \) which does have an influence on the thermal contribution to the spin Nernst conductivity and the spin polarization in case of an open circuit. But we remark that also the electrical contribution, \( S_0 \sigma_{\text{SE}} \), is affected by \( \eta \) and \( \beta \) since the Seebeck coefficient then reads \( S = S_0 (1 + \eta + \beta) \).

**Appendix B: On the ratio \( \tau_s / \tau_{DP} \)**

We estimate the size of the ratio \( \tau_s / \tau_{DP} \), defining the relative importance of extrinsic and intrinsic spin-orbit coupling. The general form of the Dyakonov-Perel relaxation rate, valid from the “clean” to the “dirty” regime, reads

\[
\frac{1}{\tau_{DP}} = \frac{1}{2} \left( \frac{2\alpha_{PF} \tau / h^2}{\tau_{DP}} \right)^2 + 1.
\]

where \( 2\alpha_{PF} / h \) is the spin-orbit splitting. Therefore

\[
\frac{\tau_s}{\tau_{DP}} = \frac{1}{2} \left( \frac{2\alpha_{PF} \tau / h^2}{\tau_{DP}} \right)^2 + 1 \left( \frac{2h}{\lambda_{PF}} \right)^4. \tag{B2}
\]

In doped semiconductors one typically finds \( 10^{-2} \lesssim \lambda / \lambda_{PF} \lesssim 1 \). Though there is yet no theory capable of estimating \( \lambda \) in a metal one can argue that, since the spin-orbit energy is small compared to the Fermi one, the relation \( \lambda / \lambda_{PF} \ll 1 \) will hold in a metallic film. Taking \( \lambda / \lambda_{PF} \approx 10^{-1} \) yields \( 1 \lesssim \tau_s / \tau_{DP} \lesssim 10^2 \). The lower value is valid in “dirty” (or with weak intrinsic spin-orbit) samples, \( 2\alpha_{PF} / h \lesssim 10^{-2} \), the upper one in “clean” (or with strong intrinsic spin-orbit) ones, \( 2\alpha_{PF} / h > 1 \).

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We remark that in three dimensions Eq. (23) is not the complete collision operator as obtained from the self-energy depicted in Fig. 1: we have dropped terms where the momentum is not parallel to the Pauli vector. These contributions are negligible when investigating the spin transport quantities.

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