

Large spin Hall angle in MnPt-based antiferromagnetic alloys

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Antiferromagnets (AFMs) have emerged as crucial materials for spintronic technologies for their ability to host spin-dependent transport phenomena, despite their zero net magnetization. Their robustness against external magnetic fields and ultrafast spin dynamics make them ideal for efficient spin-charge interconversion. However, unlike ferromagnets, many details of spin transport mechanisms in AFMs remain unexplored. In this study, we theoretically investigate the enhancement of the spin Hall effect in AFM materials, with a focus on Mn-based binary alloys. By alloying MnPt with Ir ($\text{MnPt}_x\text{Ir}_{1-x}$) or Pd ($\text{MnPt}_x\text{Pd}_{1-x}$), we aim to elucidate the impact of tuning the Fermi energy and of changing the spin-orbit coupling strength on the spin Hall conductivity, and ultimately on the spin Hall angle (SHA). We show that lightly doping MnPt with Ir enhances the SHA from approximately 3% in pristine MnPt, as experimentally reported at room temperature, to nearly 12% under the same conditions. Building on these findings, we employ density-of-states and band-structure analyses to unravel the microscopic origin of this enhancement. Overall, our results highlight the potential of antiferromagnetic alloys for efficient spin-current generation and underscore their promise for advancing spintronic applications.

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I. INTRODUCTION

Ferromagnets and nonmagnetic heavy elements such as Pt, W, and Ta have been utilized as platforms for spin current generation [1–7]. More recently, it was shown that antiferromagnets (AFMs) can generate large spin currents as well [8,9]. Depending on the crystal symmetry, typically in cases of noncollinear spin structures [10,11], AFMs may also show anomalous Hall currents. Here, in particular, the class of magnetic materials known as altermagnets [12,13] have garnered substantial interest. These materials share properties with both AFMs by hosting a total net zero magnetization and with ferromagnets through their nonrelativistic band splitting, a characteristic indicative of materials with broken parity time-reversal ($\mathcal{PT}$) symmetry.

For practical application in spintronics devices, AFMs have to generate spin currents efficiently. This has led to extensive studies [8,9,14,15] into antiferromagnetic materials such as MnPt, MnPd, and MnIr. These investigations revealed that AFMs can exhibit a significant intrinsic contribution to the spin Hall conductivity. For MnPt, it has been demonstrated [8,9] that spin currents are generated as efficiently as in Pt, the archetype heavy metal known for its large spin-orbit coupling (SOC).

As SOC is considered the primary driver behind the spin Hall effect (SHE) [16,17] in heavy metals, this has motivated extensive studies focusing on materials including heavy elements [1–7]. In this case, electrons driven by a longitudinal charge current will be deflected to opposite edges of the sample depending on their respective spins, thereby generating a transverse spin current. This spin current is subsequently utilized to exert a torque on the magnetization of an adjacent ferromagnet [18,19].

For practical applications, it becomes crucial to consider the conversion efficiency, defined as the spin Hall angle (SHA). The SHA is given by the ratio between the transverse spin current and the longitudinal charge current [20,21]. The objective is to identify materials with a significant SHA, thereby optimizing the efficiency of spin current generation.

In this paper, we demonstrate that high SHA and efficient spin currents can be achieved when alloying MnPt with Ir ($\text{MnPt}_x\text{Ir}_{1-x}$), while in the case of Pd alloying ($\text{MnPt}_x\text{Pd}_{1-x}$) the SHA remains stable and does not decrease as a function of temperature. This approach provides a greater understanding of the impact of altering the electronic structure in the case of Ir alloying, and separately of tuning the SOC strength when Pd is introduced. More specifically, we will examine the evolution of the total spin Hall conductivity (SHC), incorporating the intrinsic contribution, side-jump, and skew scattering processes [22–24]. In all cases, we will analyze the SHA as a function of the impurity concentration within these alloys. Furthermore, for $\text{MnPt}_{0.85}\text{Ir}_{0.15}$, we will investigate the effect of temperature on the SHA with a special focus on room temperatures most relevant for applications.

After a brief introduction of the alloy systems, we will discuss the key aspects of the numerical method employed here. We confirm the latter by characterizing the intrinsic

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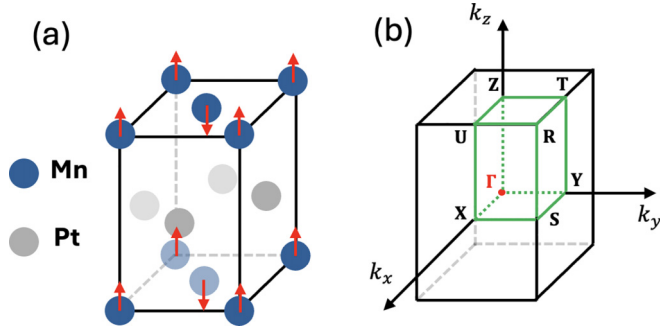


FIG. 1. (a) Lattice structure of MnPt, blue circles represent Mn atoms and grey circles the Pt atoms. The red arrows represent the magnetizations of Mn atoms. (b) Brillouin zone of MnPt with high-symmetry points (Γ , X, S, Y, Z, U, R, T).

properties of clean MnPt. This will be followed by the results sections on $\text{MnPt}_x\text{Ir}_{1-x}$ and $\text{MnPt}_x\text{Pd}_{1-x}$ incorporating our findings at room temperature for $\text{MnPt}_{0.85}\text{Ir}_{0.15}$. Finally, we will highlight our main findings and give a brief outlook on the next steps and future research into antiferromagnetic alloy systems.

II. LATTICE STRUCTURE

MnPt, MnPd, and MnIr are collinear antiferromagnetic materials with CuAu-I ($L1_0$) ordering [8,25], where the magnetizations of Mn atoms antialign in the z direction due to $3d$ electrons, as shown in Fig. 1. They all belong to the space group $4/mmm$ with international No. 123. The corresponding magnetic Laue group where inversion symmetry $\mathcal{P}$ is included is $4'/mm'm$. The lattice constants for our alloys $\text{MnPt}_x\text{Ir}_{1-x}$ and $\text{MnPt}_x\text{Pd}_{1-x}$ are taken to be constant across the concentration range x : $a = b = 4 \text{ \AA}$ and $c = 3.67 \text{ \AA}$ [8], since there is no considerable difference between the lattice constants among the original materials (MnPt, MnPd, and MnIr). The magnetic order is maintained up to very high temperatures, as the Néel temperature is approximately $T_N = 970 \text{ K}$ [25,26]. Additionally, $\mathcal{PT}$ is present in our system, which implies that anomalous Hall currents are not allowed and only spin Hall currents are present. Below are the shapes of the conductivity and spin conductivity tensors [27], where all the components of the σ^y tensor can be expressed via elements of the σ^x tensor:

$$\sigma = \begin{pmatrix} \sigma_{xx} & 0 & 0 \\ 0 & \sigma_{xx} & 0 \\ 0 & 0 & \sigma_{zz} \end{pmatrix}, \quad \sigma^x = \begin{pmatrix} 0 & 0 & \sigma_{xz}^x \\ 0 & 0 & \sigma_{yz}^x \\ \sigma_{zx}^x & \sigma_{zy}^x & 0 \end{pmatrix},$$

$$\sigma^y = \begin{pmatrix} 0 & 0 & -\sigma_{yz}^x \\ 0 & 0 & -\sigma_{xz}^x \\ -\sigma_{zy}^x & -\sigma_{zx}^x & 0 \end{pmatrix}, \quad \sigma^z = \begin{pmatrix} \sigma_{xx}^z & \sigma_{xy}^z & 0 \\ -\sigma_{xy}^z & -\sigma_{xx}^z & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$

It is important to note that the spin conductivity tensors present six independent variables: σ_{xz}^x , σ_{yz}^x , σ_{zx}^x , σ_{zy}^x , σ_{xx}^z , and σ_{xy}^z . This number reduces to only 3 when considering the conventional SHE, where the directions of the spin Hall conductivity tensor element are orthogonal; namely, σ_{yz}^x , σ_{zx}^x , and σ_{xy}^z . In Sec. V, we will present the SHAs associated with the

two spin Hall conductivities; $\sigma_{xz}^y = -\sigma_{yz}^x$ for $\text{MnPt}_x\text{Ir}_{1-x}$ and σ_{zy}^x for the alloy $\text{MnPt}_x\text{Pd}_{1-x}$. These components are chosen deliberately because they correspond to the maximal value for each alloy. The rest of the SHA components are shown in Appendixes C and D.

III. METHODS

We employ the Korringa-Kohn-Rostoker coherent potential approximation (KKR-CPA) [28–32] Green's function method based on density functional theory to calculate the charge conductivity, spin Hall conductivity, electronic band structures, and Bloch spectral functions for $\text{MnPt}_x\text{Ir}_{1-x}$ and $\text{MnPt}_x\text{Pd}_{1-x}$ alloys.

Both the longitudinal charge and spin Hall conductivities are evaluated within the Kubo–Bastin (Kubo–Středa) formalism, which naturally incorporates the effects of substitutional disorder through the CPA. The full expressions used are given in Appendix A.

In our approach, the spin-dependent transport is analyzed in terms of both the intrinsic spin Hall effect, which arises from the electronic structure of the clean system (excluding vertex corrections), and the extrinsic contributions that originate from impurity scattering, captured through the inclusion of vertex corrections [33,34]. It should be noted that in the KKR framework, even the no-vertex-correction limit contains a portion of the side-jump contribution as discussed in Ref. [35].

Ultimately, we are interested in maximizing the spin Hall angle, defined as the ratio of the spin Hall conductivity to the charge conductivity. For an electric field applied along the y direction and spin current flowing along the x direction with spin polarization along the z direction, the SHA is computed as

$$\Theta_{xy}^z = \frac{\sigma_{xy}^z}{\sigma_{yy}}. \quad (1)$$

The SHA is often used as a figure of merit to quantify the conversion efficiency of charge current to spin current.

As an additional check for pristine MnPt, we also calculate the intrinsic SHC using the Kubo–Greenwood formula as implemented in PAOFLOW [36]. We have previously compared spin Hall conductivity predictions between this approach and KKR techniques for $\beta - W$ and found good agreement [5]. Details of the computational parameters used for both KKR and PAOFLOW methodologies are provided in Appendix B.

Finally, it is crucial to mention that we only account for thermal lattice displacements when incorporating the effect of temperature; the Fermi–Dirac occupation is not part of the present formalism, which is expected to have a negligible impact at room temperature ($k_B T \approx 26 \text{ meV}$) [37]. We employed the KKR-CPA method as described in Ref. [38]. In this alloy analogy model, the coherent potential approximation is used to capture the effects of temperature dependent quasistatic random displacements through a configurational average. Additionally, we assume that the magnetization does not change in direction or magnitude as a function of temperature, and that thermal spin fluctuations are negligible due to the high Néel temperature of the system, thus playing no role in the transport properties of the alloys.

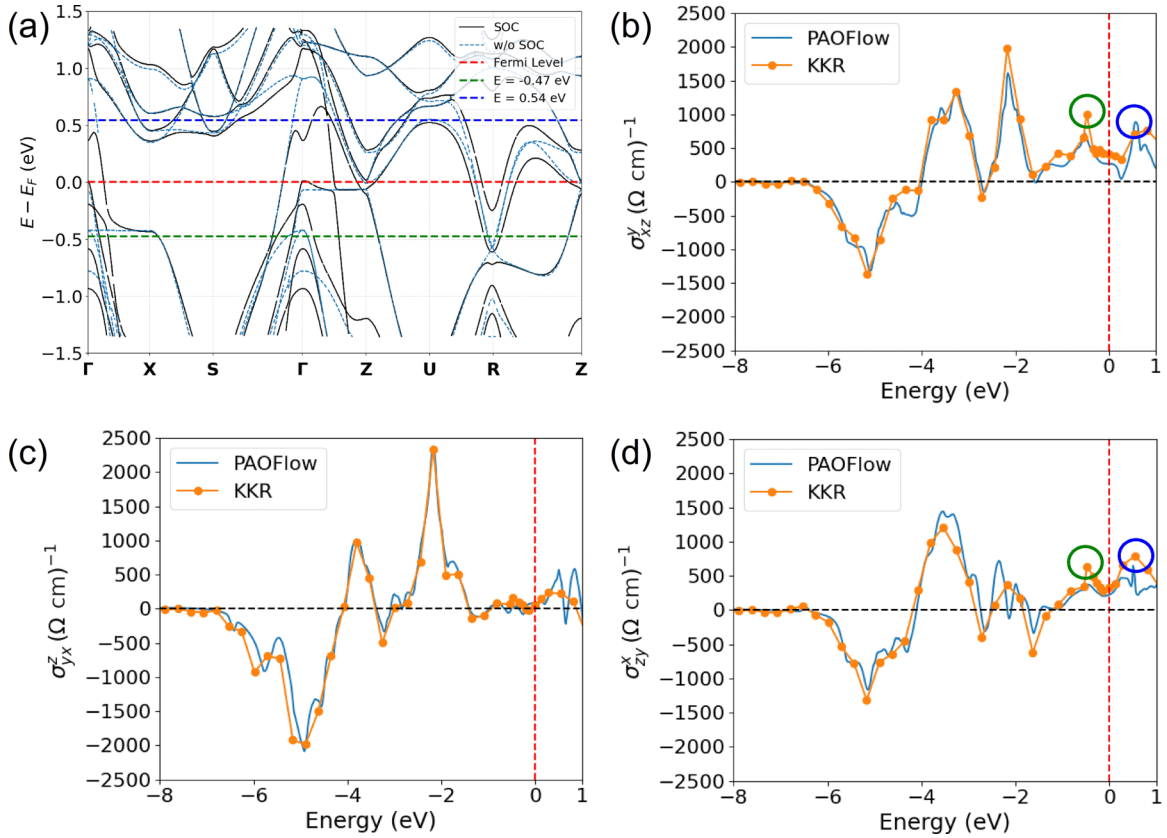


FIG. 2. (a) Band structure of MnPt along the high-symmetry path (Γ , X, S, Γ , Z, U, R, Z) using KKR method. The black curve represents the band structure including SOC, while the light blue curve shows the band structure without SOC. The horizontal red dotted line indicates the Fermi energy. The energy level $E = -0.47$ eV is also marked in the figure and is illustrated by a green dotted horizontal line. The energy level $E = 0.54$ eV is represented by a dark blue dotted horizontal line. (b) SHC component σ_{xz}^y [$(\Omega \text{ cm})^{-1}$] as a function of energy E (eV). The light blue curve corresponds to the PAOFlow method, while the orange curve corresponds to the KKR method. The vertical red dotted line marks the Fermi energy, the green circle highlights the energy $E = -0.47$ eV, and the dark blue circle corresponds to the energy $E = 0.54$ eV. (c) σ_{yx}^z [$(\Omega \text{ cm})^{-1}$] as a function of energy E (eV). (d) σ_{zy}^x [$(\Omega \text{ cm})^{-1}$] as a function of energy E (eV).

IV. MNPT: INTRINSIC CHARACTERIZATION

We have characterized the intrinsic spin Hall conductivity in the antiferromagnet MnPt using the plane wave Kubo-Greenwood approach and the KKR-CPA technique, employing the computational settings detailed in Appendix B.

As shown in Fig. 2, both methods yield consistent results regarding the SHC evolution as a function of energy for all three components σ_{xz}^y , σ_{yx}^z , and σ_{zy}^x , especially near the Fermi energy. Table I summarizes the computed SHC values at the Fermi energy for both KKR and PAOFlow, alongside the data by Zhang *et al.* [8]. Notably, the σ_{xz}^y and σ_{zy}^x components

exhibit particularly large magnitudes just below and above the Fermi energy, with σ_{xz}^y reaching $1000 (\Omega \text{ cm})^{-1}$ near $E = -0.47$ eV and σ_{zy}^x attaining $750 (\Omega \text{ cm})^{-1}$ near $E = 0.54$ eV. We aim to exploit these high values of the SHC near the Fermi energy by following two strategies. The first approach involves tuning the Fermi level by varying the total number of electrons, thereby shifting it to positions where avoided band crossings maximize the SHC. Figure 2 indicates that this is possible either by adding or removing the total number of electrons. Specifically, as shown in Fig. 2(a), the energies $E = -0.47$ eV (green dotted line) and $E = 0.54$ eV (dark blue dotted line) each align with avoided crossings around the Γ and Z points, respectively. In the following section, we demonstrate that reducing the number of electrons in our system by substituting Pt with Ir can push the Fermi level to the desired energy ($E = -0.47$ eV). In the second approach, we modify the SOC strength by replacing Pt with Pd. As illustrated in Fig. 2(a), an appropriately tuned SOC can also align the Fermi level with avoided crossings at high-symmetry points (e.g., Γ , Z, and R). This strategy provides an additional handle for maximizing the SHC, without altering the position of the Fermi level.

TABLE I. SHCs σ_{zy}^x , σ_{xz}^y , and σ_{yx}^z [$(\Omega \text{ cm})^{-1}$] for clean MnPt at the Fermi energy are shown for current KKR and PAOFlow calculations, and those reported by Zhang *et al.* [8].

Method	σ_{zy}^x	σ_{xz}^y	σ_{yx}^z
KKR	327.4	410.9	53.1
PAOFlow	221.7	268.8	81.0
Zhang <i>et al.</i> [8]	219.9	303.9	60.3

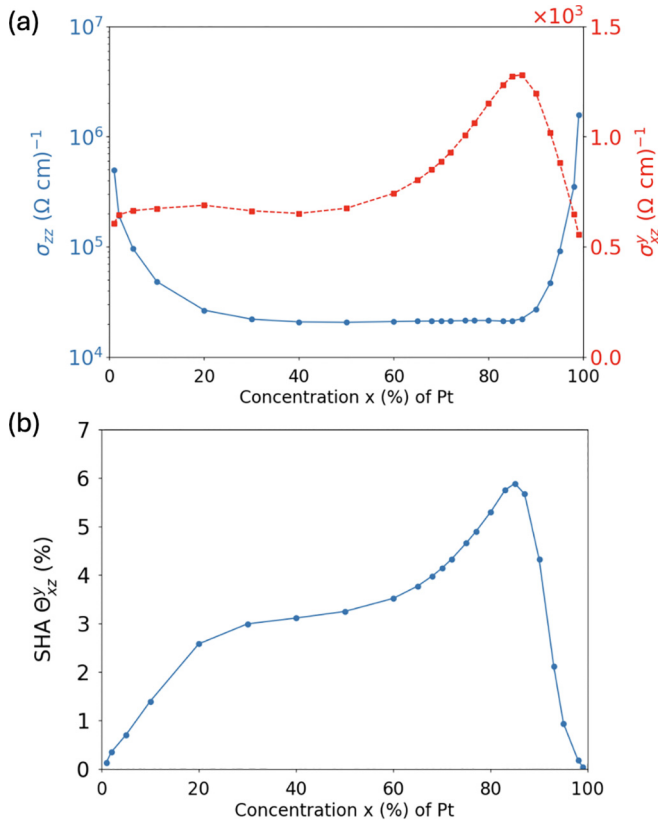


FIG. 3. (a) σ_{zz} in log scale and σ_{xz}^y versus the concentration x of Pt for the alloy $\text{MnPt}_x\text{Ir}_{1-x}$ (ranging from 1% to 99%, i.e., excluding the pristine materials). (b) SHA Θ_{xz}^y as function of the same Pt concentration x in $\text{MnPt}_x\text{Ir}_{1-x}$.

V. RESULTS

A. $\text{MnPt}_x\text{Ir}_{1-x}$

In this section, the antiferromagnetic material MnPt is doped with Ir, which has one less valence electron compared to Pt. We anticipate that this doping will significantly alter the band structure, particularly by shifting the Fermi energy. Figure 3 presents a typical trend in charge conductivity, σ_{zz} , which exhibits high values near the clean limit, where the impurity concentration is minimal. As the alloy becomes more disordered (around $x = 50\%$), σ_{zz} reaches a minimum due to increased scattering events experienced by charge carriers in the system. The SHC σ_{xz}^y displays conventional behavior, showing a maximum near the clean limit of MnPt , specifically at an impurity concentration of $x = 85\%$ [39]. Consequently, the SHA Θ_{xz}^y reaches a peak value of 6% at about 15% doping with Ir, corresponding to an SHC of $\sigma_{xz}^y = 1250$ ($\Omega \text{ cm}^{-1}$). Notably, this value is on the same order of magnitude to the high SHC of approximately 2000 ($\Omega \text{ cm}^{-1}$) reported for heavy-metal Pt, which is known for its strong SOC [17,40–42]. Within the Pt concentration range of [20%, 60%], the SHC exhibits a steady variation, reaching a local minimum at $x = 40\%$. This behavior leads to a quasilinear increase in the SHA within this regime. Notably, the introduction of Pt impurities does not significantly modify the SHC as observed with Ir impurities. This behavior is characteristic of the SHC and has also been observed in non-magnetic Pt-based binary

alloys, where the SHC maximum tends to be at higher Pt concentrations [39].

To gain deeper insight into the maximum SHC observed for the $\text{MnPt}_{0.85}\text{Ir}_{0.15}$ alloy, in Fig. 4 we plot the Bloch spectral function (BSF) $\mathcal{A}(\vec{k}, E)$ for various alloys near the Pt concentration of $x = 85\%$. It is defined as

$$\mathcal{A}(\vec{k}, E) = \sum_n \delta(E - E_n(\vec{k})), \quad (2)$$

measuring the number of states per energy E and wave vector $\vec{k}$. The sum in Eq. (2) runs over the band indices n , where E is the energy and $E_n(\vec{k})$ is the energy level for band n with wave vector $\vec{k}$.

The primary effect of doping with Ir is to lower the Fermi level deeper into the valence bands. The maximum SHC is achieved at a Ir concentration of 15% [Fig. 4(c)]. This concentration corresponds to the energy shift of $E = -0.47$ eV that was previously discussed and shown in Fig. 2, where we obtained a similar shape of the band structure along with a SHC of $\sigma_{xz}^y = 1000$ ($\Omega \text{ cm}^{-1}$). In this particular case, where $E = -0.47$ eV, the energy level lies exactly midway between the conduction and valence bands around the Γ point, suggesting that these bands dominate the SHC contribution. Interestingly, the SHC increases only slightly due to doping with Ir, going from 1000 ($\Omega \text{ cm}^{-1}$) in clean system MnPt and shifting energy by $E = -0.47$ eV to 1250 ($\Omega \text{ cm}^{-1}$) by actually doping MnPt with 15% of Ir, indicating that changes to the band structure beyond the energy shift are minimal. This implies that the primary mechanism driving the SHC maximum is the optimal alignment of the Fermi level between the conduction and valence bands, which enhances carrier contributions near the Γ point.

Another reason for the increased value of SHC at $x = 85\%$ is further elucidated in Fig. 5, which depicts the projection of the BSF at Fermi energy $\mathcal{A}(\vec{k} = (k_x, k_y, 0), E_F)$ in the plane $k_z = 0$.

Doping MnPt with Ir atoms broadens the electronic states around the Γ point for concentrations of $x = 75\%$ and $x = 80\%$, reflecting more scattering events. The same states around the Γ point become sharper for higher concentrations of $x = 85\%$ and $x = 90\%$. Additionally, the arced states connecting $X = (\pi/a, 0, 0)$ to $Y = (0, \pi/a, 0)$ also exhibit significant scattering as the doping concentration of Ir increases. The slight increase of the SHC observed at $x = 85\%$ is likely attributed to these arced states, which display a pronounced discrepancy between spin-up and spin-down electrons. This difference, evident in the “Diff” BSF, reduces spin-mixing processes, thereby favoring higher SHC. Indeed, it is well established in the literature that spin-flip processes have an influence on the extrinsic contribution to the SHC, as demonstrated by Gradhand *et al.* [29], using first principles calculations. In contrast, at $x = 75\%$, the spin-mixing process is highly prevalent. Spin-up and spin-down electrons can interchange their spins readily, leading to a reduction in SHC.

Importantly, we can predict the optimal concentration x^{opt} of Pt impurities in $\text{MnPt}_x\text{Ir}_{1-x}$ alloys at which the maximum of SHC occurs solely by calculating the DOS for clean system MnPt , shown in Fig. 6. By assuming that doping MnPt with Ir only shifts the Fermi level without disrupting the electronic

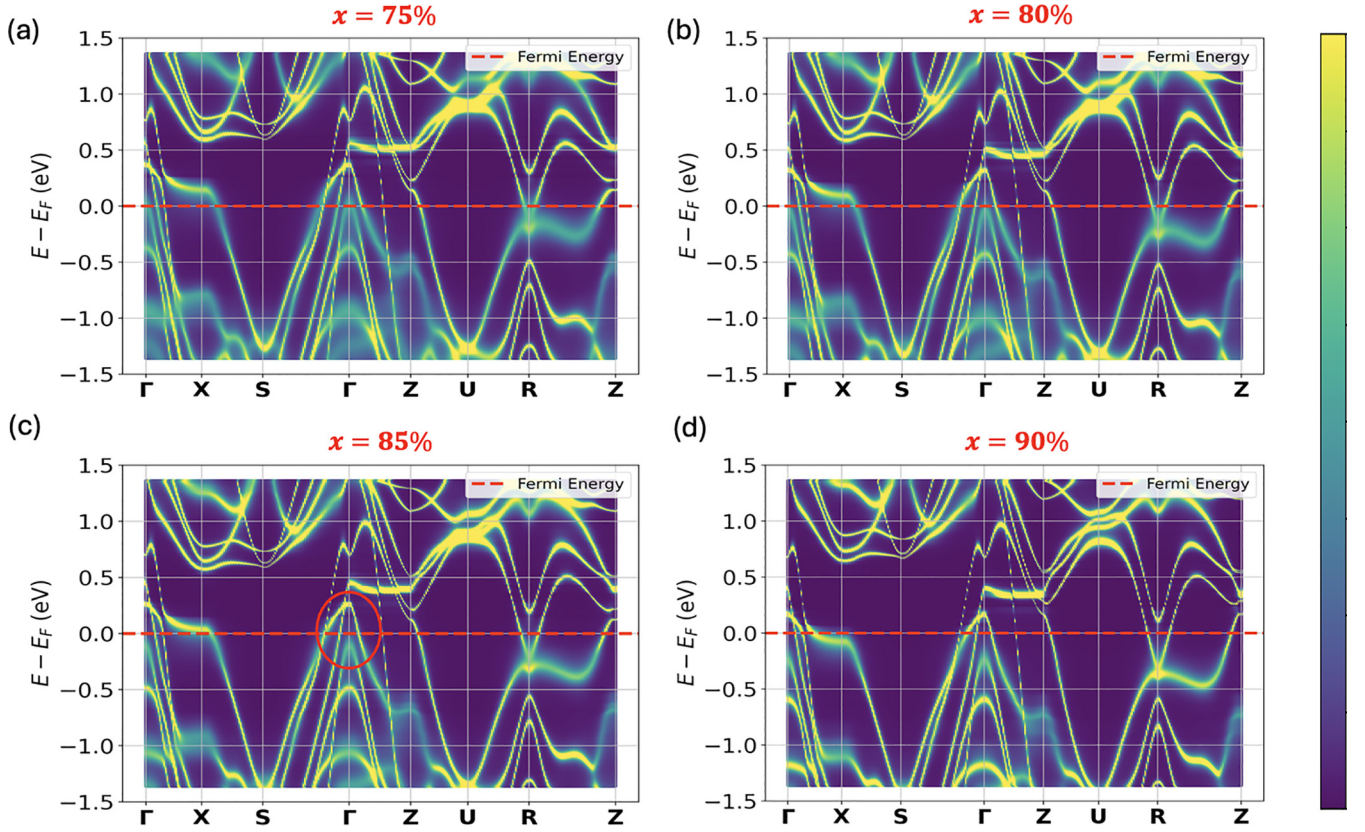


FIG. 4. Bloch spectral function $\mathcal{A}(\vec{k}, E)$ of $\text{MnPt}_x\text{Ir}_{1-x}$ along the high-symmetry path (Γ , X, S, Y, Z, U, R, Z) for different concentrations x : (a) $x = 75\%$, (b) $x = 80\%$, (c) $x = 85\%$. The red circle around the Γ point illustrates the band anticrossing. (d) $x = 90\%$. The 85% Pt concentration corresponds to the energy shift of $E = -0.47$ eV illustrated in Fig. 2 with the horizontal green dotted line. The color bar represents the magnitude of the Bloch spectral function $\mathcal{A}(\vec{k}, E)$, expressed in arbitrary units, where brighter colors correspond to higher spectral weight (i.e., higher density of states at a given $\vec{k}$ and energy E).

structure, and that the maximum of SHC occurs at energy $E = -0.47$ eV as mentioned before in Fig. 2(b), we numerically compute the total number of electrons up to that energy and compare it to the value obtained by integrating the DOS up to the Fermi level. Assuming a rigid band shift, the total number of electrons for the $\text{MnPt}_x\text{Ir}_{1-x}$ alloyed system in the unit cell can be defined as $2 \times (10 + 7x + 6(1 - x)) = 2 \times (16 + x)$, where x is the doping concentration of Pt:

$$N(E_F) = \int_{-\infty}^{E_F} \frac{dN}{dE} dE = 34.0$$

$$N(E) = \int_{-\infty}^{-0.47 \text{ eV}} \frac{dN}{dE} dE = 33.6.$$

By dividing the two quantities and comparing it to the rigid band model, we can extract the optimal doping concentration x^{opt} of Pt impurities:

$$\frac{N(E)}{N(E_F)} = \frac{16 + x^{\text{opt}}}{17}.$$

We finally get that

$$x^{\text{opt}} = 17 * \frac{N(E)}{N(E_F)} - 16 = 80.0\%.$$

This finding is particularly significant because, when doping MnPt with Ir merely shifts the Fermi level, the information

obtained from the clean-limit calculations (Fig. 2) can provide sufficient information to estimate both the optimal concentration x^{opt} and the corresponding SHC. In such a case, performing a complete CPA calculation may not be strictly necessary. Nevertheless, a full CPA-based concentration scan remains essential for assessing whether impurities induce more significant band-structure modifications or scattering effects beyond a simple Fermi-level shift.

For the $\text{MnPt}_{0.85}\text{Ir}_{0.15}$ alloy to be functional in real spintronics devices, we study the effect of varying the temperature on the SHC and SHA, as illustrated in Fig. 7. The effect of temperature is to decrease both the charge conductivity σ_{yy} and SHC σ_{xz}^y , but not at the same rate.

In fact, the SHC only experiences a slight drop, going from a value of $1250 (\Omega \text{ cm})^{-1}$ at $T = 0$ K to $1050 (\Omega \text{ cm})^{-1}$ at room temperature $T = 300$ K, leading the SHA to increase from around 6% at $T = 0$ K to 8% at $T = 300$ K. These high values of the SHC and SHA show that the AFM alloy $\text{MnPt}_{0.85}\text{Ir}_{0.15}$ can be a promising candidate for spintronics applications.

B. $\text{MnPt}_x\text{Pd}_{1-x}$

Modifying the strength of SOC can be achieved by substituting Pt with atoms of similar chemical structures, specifically those in the same column of the periodic table,

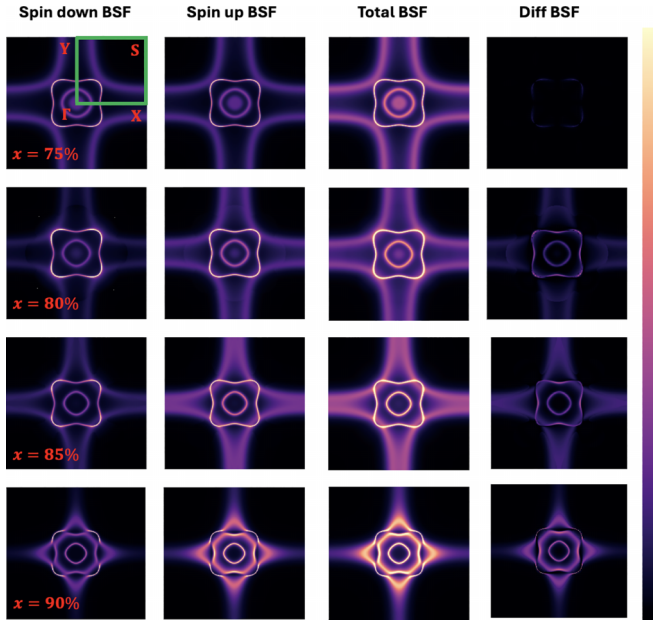


FIG. 5. BSF $\mathcal{A}(\vec{k}, E)$ at Fermi energy E_F in the reciprocal plane $k_z = 0$ for $\text{MnPt}_x\text{Ir}_{1-x}$. This plane includes the four high-symmetry points (Γ , X , S , Y). The images show the BSF at different impurity concentrations: $x = 75\%$, $x = 80\%$, $x = 85\%$ and $x = 90\%$ from top to bottom, and for spin-down, spin-up, total BSF and difference between up BSF and down BSF, from left to right.

such as Pd. Given its higher atomic mass, Pt exhibits a stronger SOC compared to Pd. We expect the maximal value of SHA to occur in Pt rich alloys.

Figure 8 illustrates the total spin Hall conductivity $\sigma_{zy}^{x,\text{tot}}$ calculated with vertex corrections together with the corresponding values without vertex corrections ($\sigma_{zy}^{x,\text{nvc}}$), which encompass both the intrinsic and side-jump contributions [35] as a function of Pt concentration (x) in the $\text{MnPt}_x\text{Pd}_{1-x}$ alloy. Notably, the SHA reaches its maximum near the MnPd limit at $x = 20\%$, which might be surprising as Pd exhibits the smaller SOC. In contrast, earlier studies of non-magnetic $\text{Pt}_x\text{Pd}_{1-x}$

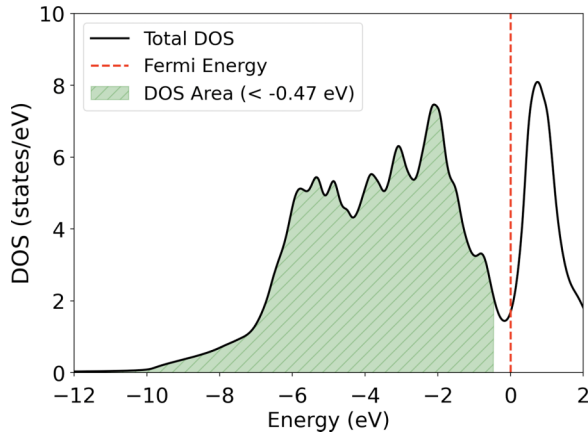


FIG. 6. Total DOS of clean MnPt. The green dashed area represents the integration area up to $E = -0.47$ eV, and the vertical red dotted line illustrates the Fermi energy.

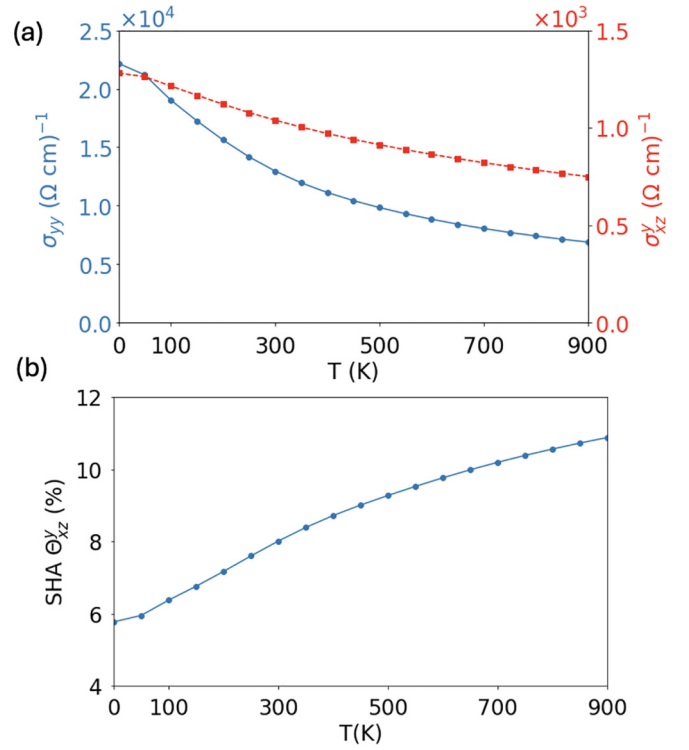


FIG. 7. (a) σ_{zz} and σ_{xz}^y versus temperature $T(K)$ for the alloy $\text{MnPt}_{0.85}\text{Ir}_{0.15}$. (b) Spin Hall angle Θ_{xz}^y as a function of temperature $T(K)$ for $\text{MnPt}_{0.85}\text{Ir}_{0.15}$.

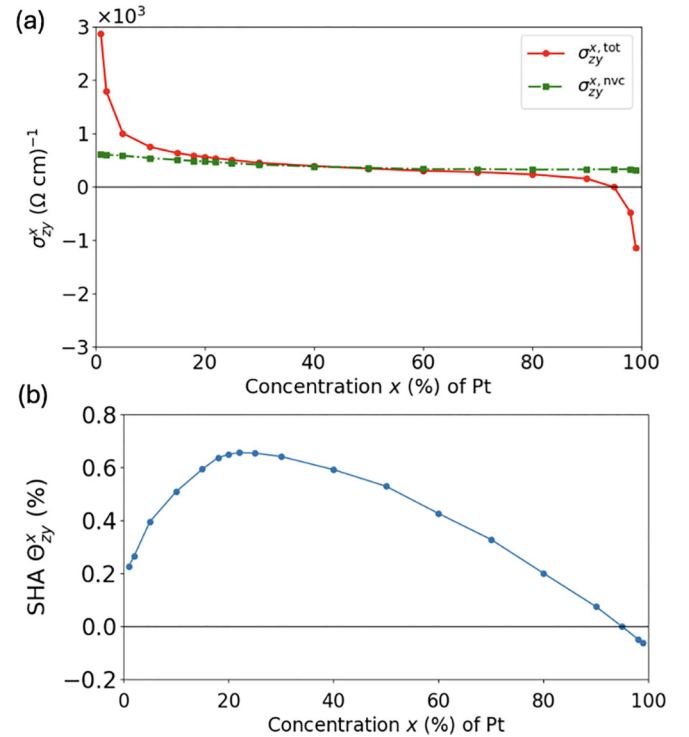


FIG. 8. (a) $\sigma_{zy}^{x,\text{tot}}$ and $\sigma_{zy}^{x,\text{nvc}}$ ($\Omega \text{ cm}$) $^{-1}$ versus the Pt concentration x (ranging from 1% to 99%, i.e., excluding the pristine materials) for the alloy $\text{MnPt}_x\text{Pd}_{1-x}$. (b) SHA Θ_{zy}^x as a function of the same Pt concentration in $\text{MnPt}_x\text{Pd}_{1-x}$. The black line represents $y = 0$.

TABLE II. SHA at $T = 0$ K and $T = 300$ K, and SHA^{poly} at $T = 300$ K for both alloys $\text{MnPt}_{0.85}\text{Ir}_{0.15}$ and $\text{MnPt}_{0.2}\text{Pd}_{0.8}$.

Alloy	SHA at $T = 0$ K	SHA at $T = 300$ K	SHA^{poly} at $T = 300$ K
$\text{MnPt}_{0.85}\text{Ir}_{0.15}$	6%	8%	12%
$\text{MnPt}_{0.2}\text{Pd}_{0.8}$	0.65%	0.9%	4%

alloys showed that the SHA peaks at $x = 50\%$, corresponding to minimal charge conductivity [39].

The no-vertex contribution decreases from about $600 (\Omega \text{ cm})^{-1}$ on the MnPd-rich side to approximately $300 (\Omega \text{ cm})^{-1}$ on the MnPt-rich side; a counterintuitive trend, since the stronger SOC of Pt would normally suggest larger intrinsic values. Appendix E confirms this expectation by showing the intrinsic SHC of pure MnPd and MnPt across a range of energies, and specifically at the Fermi level, where all components of the SHC are indeed higher for MnPt. Appendix F further supports this interpretation: Using the scaling method of the total SHC as a function of the corresponding longitudinal conductivity, the extrapolated value for MnPd is about $528 (\Omega \text{ cm})^{-1}$ and for MnPt about $313 (\Omega \text{ cm})^{-1}$, suggesting a much stronger side-jump contribution for MnPd and explaining the unusually high no-vertex value on the MnPd-rich side. This trend was also observed for ferromagnetic metals, where FePd exhibits a pronounced side-jump component of the AHC compared to FePt [43,44].

This competition between the vertex-corrected and no-vertex curves produces the sign change of the total SHC at around $x = 95\%$ and the associated suppression of the SHA at this concentration, where we go from a value of around 0.22% near the clean limit of MnPd to a value of approximately -0.06% near the other clean limit of MnPt. While these values of SHA remain small compared to those obtained for $\text{MnPt}_x\text{Ir}_{1-x}$, this specific case shows, as a counterexample, that tuning SOC alone does not necessarily lead to a significant enhancement of the SHA. Instead, this system highlights that a combination of mechanisms; including intrinsic, side-jump, and skew scattering effects, can strongly influence the resulting SHA, even when the constituent elements have similar chemical structures and numbers of valence electrons.

Finally, we show in Appendix G and Table II that temperature does not drastically affect the SHA in the case of the specific alloy $\text{MnPt}_{0.2}\text{Pd}_{0.8}$ (the alloy for which we get the maximum of SHA at zero temperature). In fact, the SHA steadily increases from 0.65% at 0 K to 0.9% at 300 K.

VI. DISCUSSION

We obtain a high SHA in the case of $\text{MnPt}_x\text{Ir}_{1-x}$ even at room temperature. However, in our theoretical work, we have only considered substitutional disorder for the calculation of charge resistivity. This resistivity is expected to be higher when other types of disorder, such as structural disorder or grain boundaries, are considered. Since our theoretical resistivity values are much lower compared to experimental values, which have been reported in Ref. [8] to be $\rho_{\text{MnPt}}^{\text{exp}} = 164.0 \mu\Omega \text{ cm}$ for MnPt, $\rho_{\text{MnPd}}^{\text{exp}} = 223.0 \mu\Omega \text{ cm}$ for MnPd, and $\rho_{\text{MnIr}}^{\text{exp}} = 269.3 \mu\Omega \text{ cm}$ for MnIr, we use a linear interpolation between the two base materials MnPt and MnM to estimate

the total resistivity of the alloy

$$\rho_{\text{MnPt}_x\text{M}_{1-x}}^{\text{tot}} = x \rho_{\text{MnPt}}^{\text{exp}} + (1-x) \rho_{\text{MnM}}^{\text{exp}},$$

where M represents in this case either the metal Ir or Pd. The experimental resistivities mentioned here correspond to polycrystalline materials. Accordingly, SHA values are evaluated only at $T = 300$ K in this section, since the reported longitudinal conductivities were measured on polycrystalline samples at room temperature. To define the SHA in the context of incorporating these experimental polycrystalline resistivities, we should solely account for the intrinsic contribution to the SHC. The side-jump and skew-scattering contributions to the SHC scale linearly and quadratically, respectively, with the theoretical charge conductivity, near the clean limits. Additionally, to account for the polycrystalline nature of the alloys, we consider the averaged intrinsic SHC for all components

$$\bar{\sigma}^{\text{int}} = \frac{1}{3} (\sigma_{yz}^{\text{int}} + \sigma_{xz}^{\text{int}} + \sigma_{yx}^{\text{int}}).$$

We now define the polycrystalline SHA for the alloy $\text{MnPt}_x\text{M}_{1-x}$ as follows:

$$\bar{\Theta}_{\text{MnPt}_x\text{M}_{1-x}} = \frac{\bar{\sigma}^{\text{int}}}{\sigma_{\text{MnPt}_x\text{M}_{1-x}}^{\text{tot}}},$$

or, more explicitly,

$$\bar{\Theta}_{\text{MnPt}_x\text{M}_{1-x}} = \frac{1}{3} (\sigma_{yz}^{\text{int}} + \sigma_{xz}^{\text{int}} + \sigma_{yx}^{\text{int}}) \times (x \rho_{\text{MnPt}}^{\text{exp}} + (1-x) \rho_{\text{MnM}}^{\text{exp}}).$$

This formulation is applicable only in the highly disordered regime, where the side-jump and skew-scattering contributions make a negligible impact on the SHC. In Fig. 9, under the assumption of a polycrystalline alloy and accounting for experimental resistivity values, we reach a very high polycrystalline SHA simply by lightly doping MnPt with 15%

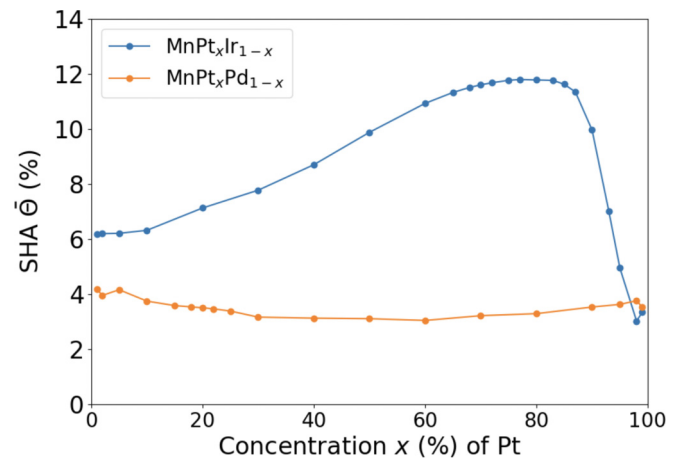


FIG. 9. SHA $\bar{\Theta}$ for both alloys as function of the doping concentration x . The blue curve represents $\text{MnPt}_x\text{Ir}_{1-x}$, while the orange curve corresponds to the $\text{MnPt}_x\text{Pd}_{1-x}$ alloy.

Ir, yielding $\bar{\Theta} = 12\%$ at $T = 300$ K. For comparison, pristine MnPt exhibits a polycrystalline SHA of about 3% at room temperature in experiment (Ref. [8]), and 4.4% using our KKR values reported in Table I, indicating a roughly fourfold enhancement achieved through Ir doping. Under the same conditions, the polycrystalline SHA of the Pd-rich alloy MnPt_{0.2}Pd_{0.8} remains comparatively modest at $\bar{\Theta} = 4\%$.

VII. CONCLUSION

Our investigation into the SHA in AFM MnPt-based binary alloys, specifically MnPt_xIr_{1-x} and MnPt_xPd_{1-x}, has demonstrated significant potential for advancing spintronic applications. By strategically alloying MnPt with either Ir or Pd, we have effectively manipulated the electronic structure and SOC to achieve substantial increases in the SHA. Moreover, we have considered the system's anisotropy by focusing on the largest component of the SHA, which can help guide crystal growth and thin film orientation for optimal device development. We have also treated the case where we have polycrystalline alloys, and shown that we get even higher values of SHA at room temperature. In this case, light doping of MnPt with 15% Ir yields a polycrystalline SHA of about 12% at $T = 300$ K, compared with about 3% reported experimentally for pristine MnPt, representing roughly a fourfold enhancement.

Although our present analysis focuses on doping MnPt with Ir or Pd, the methodology we employ is readily generalizable to other dopants. For instance, substituting Pt with Cu atoms, which add more valence electrons to the system, may enable large SHAs by aligning the Fermi level with the additional local maximum of the SHC located at $E = 0.54$ eV. Hence, the approach discussed here can be utilized to systematically investigate doping strategies aimed at further enhancing spin Hall effects in this class of alloys.

Overall, our study strongly supports the viability of antiferromagnetic alloys as efficient generators of spin currents, offering substantial promise for spintronic applications.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available because they contain commercially sensitive information. The data are available from the authors upon reasonable request.

APPENDIX A: GREEN'S FUNCTIONS AND THE KUBO-BASTIN FORMALISM FOR CHARGE AND SPIN CONDUCTIVITIES

In this appendix, we clarify the Green's function formalism used to calculate the electronic density and the transport coefficients in disordered alloys [46,47]. The single-particle

Green's function reads

$$G(\vec{r}, \vec{r}', E) = \langle \vec{r} | \hat{G}(E) | \vec{r}' \rangle, \quad (\text{A1})$$

and the fully relativistic electron density follows from the imaginary part of the site-diagonal Green's function integrated up to the Fermi level

$$\rho(\vec{r}) = -\frac{1}{\pi} \int_{-\infty}^{E_F} \text{Im} G(\vec{r}, \vec{r}, E) dE. \quad (\text{A2})$$

1. Kubo-Bastin formula for spin Hall conductivity

Following Refs. [22,33,35,48,49], the SHC at zero temperature is described by the Kubo-Bastin (or Kubo-Středa) formalism. At $T = 0$ K the total SHC splits into a Fermi-surface term (I) and a Fermi-sea term (II):

$$\sigma_{ij}^k = \sigma_{ij}^{k,\text{I}} + \sigma_{ij}^{k,\text{II}}, \quad (\text{A3})$$

with

$$\sigma_{ij}^{k,\text{I}} = \frac{\hbar}{4\pi N\Omega} \text{Tr} \left\langle \hat{\mathcal{J}}_i^k (G^+ - G^-) \hat{j}_j G^- - \hat{\mathcal{J}}_i^k G^+ \hat{j}_j (G^+ - G^-) \right\rangle_c, \quad (\text{A4})$$

$$\sigma_{ij}^{k,\text{II}} = \frac{\hbar}{4\pi N\Omega} \int_{-\infty}^{E_F} dE \text{Tr} \left\langle \hat{\mathcal{J}}_i^k G^+ \hat{j}_j \frac{dG^+}{dE} - \hat{\mathcal{J}}_i^k \frac{dG^+}{dE} \hat{j}_j G^+ - \hat{\mathcal{J}}_i^k G^- \hat{j}_j \frac{dG^-}{dE} + \hat{\mathcal{J}}_i^k \frac{dG^-}{dE} \hat{j}_j G^- \right\rangle_c. \quad (\text{A5})$$

Here G^+ and G^- denote the retarded and advanced Green's functions, Ω is the unit-cell volume, N the number of sites, and $\langle \dots \rangle_c$ denotes the configuration average. The indices i, j, k correspond respectively to the current direction, the applied electric field direction, and the spin-polarization direction. The operators are

$$\hat{j}_j = e\hat{v}_j, \quad \hat{\mathcal{J}}_i^k = \frac{1}{2}\{\hat{s}_k, \hat{v}_i\},$$

with $\hat{v}_i$ the velocity operator and $\hat{s}_k$ the spin operator along k .

2. Charge conductivity

The charge conductivity is obtained from Eqs. (A4) and (A5) by replacing the spin-current operator $\hat{\mathcal{J}}_i^k$ with the charge-current operator $\hat{j}_i = e\hat{v}_i$ and by dropping the spin index k . For the longitudinal charge conductivity in a normal metal at zero magnetic field, the Fermi-sea term vanishes and the expression reduces to

$$\sigma_{ij} = \frac{\hbar}{2\pi N\Omega} \text{Tr} \langle \hat{j}_i G^+ \hat{j}_j G^- \rangle_c. \quad (\text{A6})$$

APPENDIX B: COMPUTATIONAL DETAILS

For the self-consistent KKR-CPA calculations, we considered an angular momentum cutoff of $l_{\text{max}} = 3$. The range of energies included is $[-0.2, E_F]$ Ry with 32 energy points on a semicircular contour in the complex plane. For the sampling of the Brillouin zone, the number of $\vec{k}$ points increases as we approach the Fermi energy. The convergence of the self-consistent solution in terms of the number of $\vec{k}$ points was also checked using Lloyd's formula as reported in Refs. [50–52], increasing the accuracy of the position of the Fermi energy.

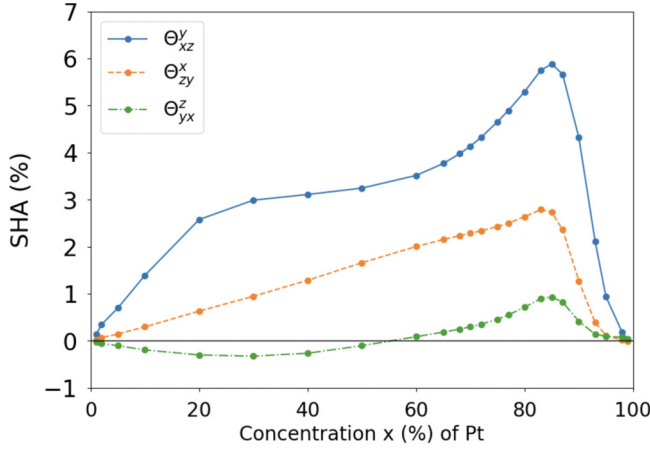


FIG. 10. SHAs Θ_{xz}^y , Θ_{zy}^x , and Θ_{yx}^z as a function of x concentration of Pt in $\text{MnPt}_x\text{Ir}_{1-x}$ at $T = 0$ K.

For calculating transport quantities, a similar approach is adopted, but using more energy points $NE = 80$ and $\vec{k}$ points in the Brillouin zone (up to 10^7). For the KKR calculations of the clean MnPt system, we use an imaginary part of the energy $\text{Im}(E) = 10^{-6}$, strictly greater than zero to avoid divergences in the SHC, as we consider the system to be clean, without any impurities. When calculating the SHC for alloyed systems, the imaginary part is set to zero since the broadening is already accounted for through doping. The convergence of the values of the charge and spin Hall conductivities has been rigorously tested.

For the intrinsic SHC calculated via PAOFLOW, a self-consistent calculation is first performed using the plane wave code QUANTUM ESPRESSO [53]. Plane wave cutoffs of 80 $\mathcal{Ry}$ and 800 $\mathcal{Ry}$ are used for the wave functions and charge density, respectively. A Monkhorst-Pack grid of $24 \times 24 \times 24$ and Methfessel-Paxton smearing of 0.02 $\mathcal{Ry}$ are employed in the self-consistent calculations. The exchange and correlation energies are based on the local density approximation. Mn and Pt are described using projector augmented wave pseudopotentials. Once the electronic structure is converged, the wave functions are mapped to PAOFLOW's tight-binding

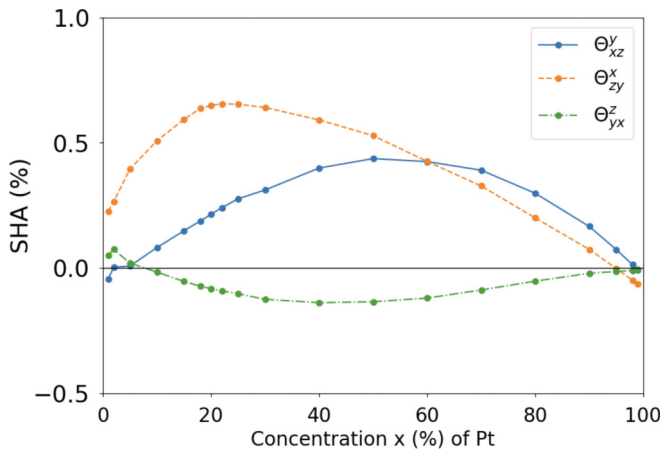


FIG. 11. SHAs Θ_{xz}^y , Θ_{zy}^x , and Θ_{yx}^z as a function of x concentration of Pt in $\text{MnPt}_x\text{Pd}_{1-x}$ at $T = 0$ K.

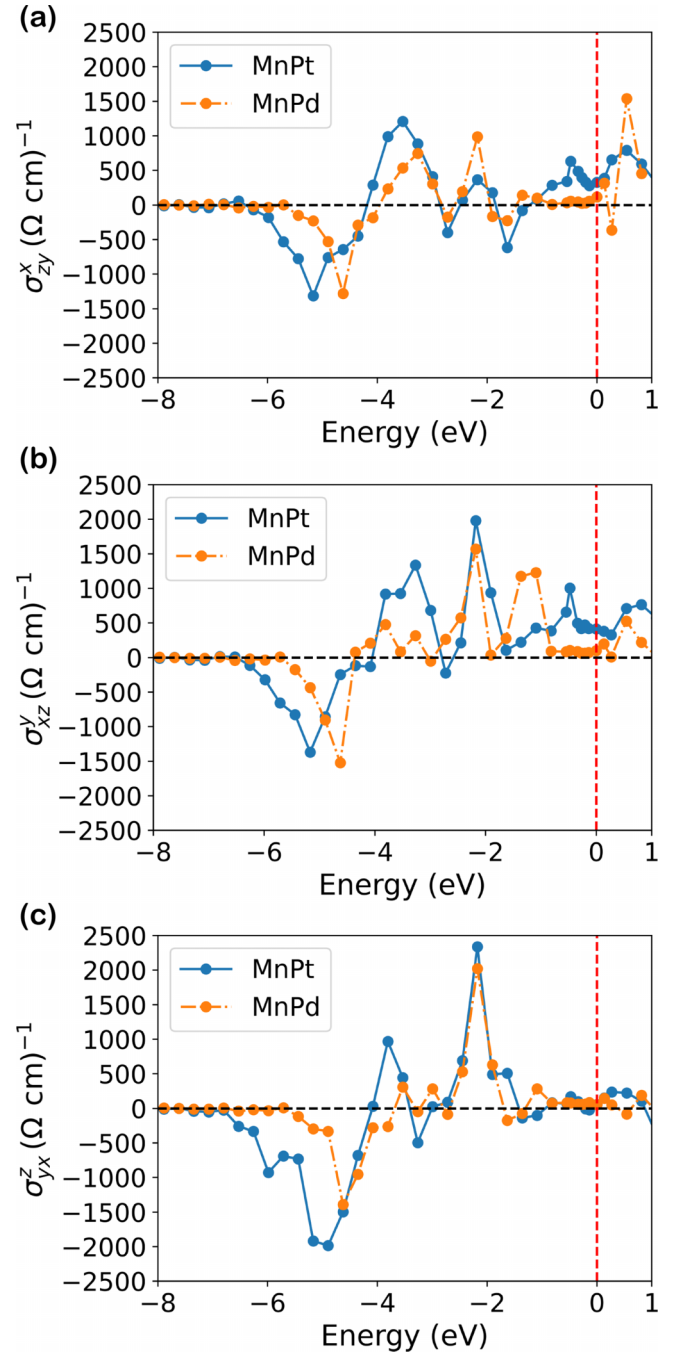


FIG. 12. SHCs as a function of energy E for pristine MnPt and MnPd using KKR method. (a) σ_{zy}^x , (b) σ_{xz}^y and (c) σ_{yx}^z .

basis set of pseudoatomic orbitals for efficient Brillouin zone sampling with an adaptive smearing integration scheme [54]. Ultimately, the lattice parameters used in PAOFLOW are identical to those employed in the KKR-CPA calculations to ensure consistency.

APPENDIX C: SHAS FOR $\text{MnPt}_x\text{Ir}_{1-x}$ AT $T = 0$ K

Figure 10 shows the variation of the SHAs Θ_{xz}^y , Θ_{zy}^x and Θ_{yx}^z at $T = 0$ K, as a function of dopant concentration, x , for

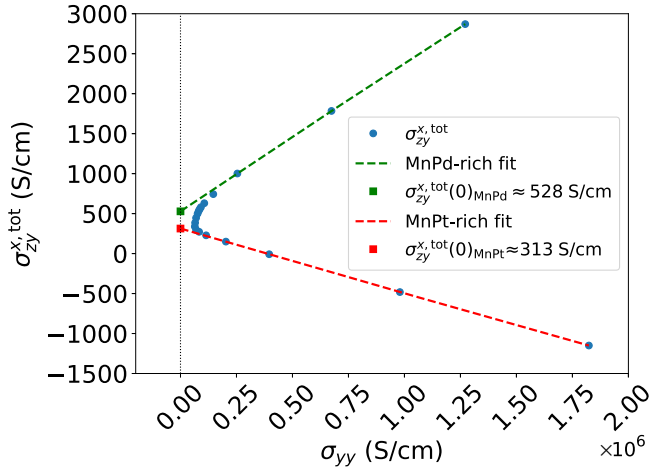


FIG. 13. Linear extrapolation of the SHC $\sigma_{zy}^{x,tot}$ as a function of the longitudinal conductivity σ_{yy} for $\text{MnPt}_x\text{Pd}_{1-x}$. The square markers at $\sigma_{yy} = 0$ show the extrapolated SHC values for MnPd (~ 528 S/cm) and MnPt (~ 313 S/cm).

$\text{MnPt}_x\text{Ir}_{1-x}$, where configurational averages are described via application of the CPA.

APPENDIX D: SHAS FOR $\text{MnPt}_x\text{Pd}_{1-x}$ AT $T = 0$ K

Figure 11 demonstrates the SHAs Θ_{xz}^y , Θ_{zy}^x and Θ_{yx}^z at $T = 0$ K, as a function of Pt concentration x in $\text{MnPt}_x\text{Pd}_{1-x}$, where configurational averages are described via application of the CPA.

APPENDIX E: SHCS OF MnPt AND MnPd AS FUNCTION OF ENERGY E AT $T = 0$ K

Figure 12 illustrates the SHCs σ_{zy}^x , σ_{xz}^y and σ_{yx}^z of both pristine materials MnPt and MnPd at $T = 0$ K as a function of energy E , calculated within the KKR formalism. For all tensor components, the SHC of MnPt exceeds that of MnPd, indicating a stronger spin-orbit-coupling-driven contribution to spin transport in MnPt.

APPENDIX F: LINEAR EXTRAPOLATION OF THE SHC FOR $\text{MnPt}_x\text{Pd}_{1-x}$ AT $T = 0$ K

We show below that the large jump in the no-vertex-corrected SHC, illustrated in Fig. 8(a) is indeed due to the side-jump process. In the dilute limit, skew scattering scales inversely with impurity concentration and thus with the longitudinal charge conductivity σ_{yy} . Following this, the total SHC

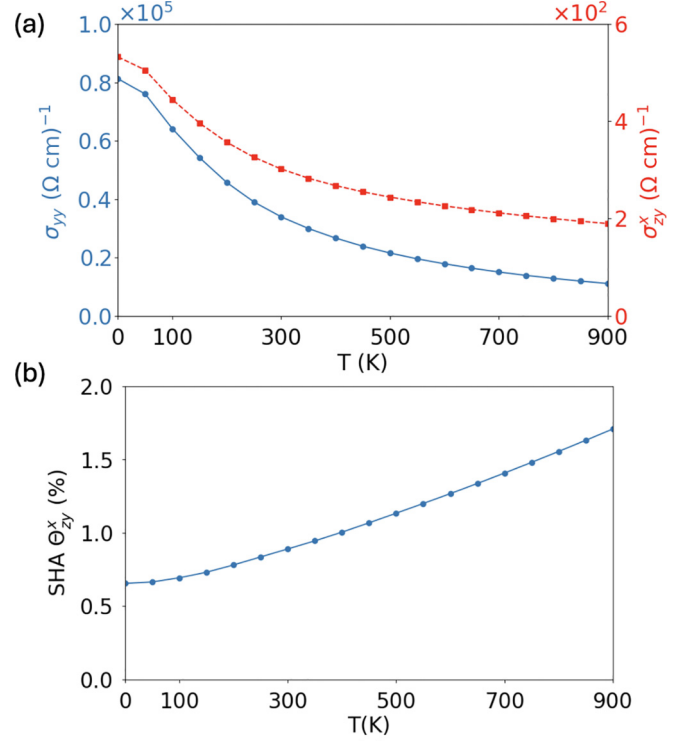


FIG. 14. (a) σ_{yy} and σ_{zy}^x versus temperature T for the alloy $\text{MnPt}_{0.2}\text{Pd}_{0.8}$. (b) Spin Hall angle Θ_{zy}^x as a function of temperature T for $\text{MnPt}_{0.2}\text{Pd}_{0.8}$.

can be expressed as

$$\sigma_{zy}^{x,tot}(\sigma_{yy}) = \sigma_{zy}^{x,int} + \sigma_{zy}^{x,sj} + \Theta_{zy}^{x,skew}\sigma_{yy}. \quad (\text{F1})$$

Extrapolating to $\sigma_{yy} \rightarrow 0$ yields the combined intrinsic + side-jump contribution ($\sigma_{zy}^{x,int} + \sigma_{zy}^{x,sj}$) [35], as illustrated in Fig. 13 for the $\text{MnPt}_x\text{Pd}_{1-x}$ alloy at both end points MnPd and MnPt.

In the clean limit of MnPd, the combined SHC reaches ~ 528 S/cm, implying a side-jump contribution of about 400 S/cm. By contrast in the MnPt limit the side-jump term is comparatively small.

APPENDIX G: SHA Θ_{zy}^x FOR $\text{MnPt}_{0.2}\text{Pd}_{0.8}$ AS A FUNCTION OF T

Figure 14 shows the temperature dependence of the longitudinal conductivity σ_{yy} and the SHC σ_{zy}^x (panel a), together with the corresponding SHA Θ_{zy}^x (panel b), for $\text{MnPt}_{0.2}\text{Pd}_{0.8}$. Both σ_{yy} and σ_{zy}^x decrease monotonically with increasing T , leading to an increase in the SHA Θ_{zy}^x as a function of temperature.

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