

This is the accepted manuscript made available via CHORUS. The article has been published as:

Sign Change of the Spin Hall Effect due to Electron Correlation in Nonmagnetic CuIr Alloys

Zhuo Xu, Bo Gu, Michiyasu Mori, Timothy Ziman, and Sadamichi Maekawa

Phys. Rev. Lett. **114**, 017202 — Published 6 January 2015

DOI: [10.1103/PhysRevLett.114.017202](https://doi.org/10.1103/PhysRevLett.114.017202)

Sign change of the spin Hall effect due to electron correlation in nonmagnetic CuIr alloys

Zhuo Xu^{1,2}, Bo Gu^{1,2}, Michiyasu Mori^{1,2}, Timothy Ziman^{3,4}, and Sadamichi Maekawa^{1,2}

¹Advanced Science Research Center, Japan Atomic Energy Agency, Tokai 319-1195, Japan

²CREST, Japan Science and Technology Agency, Sanbancho, Tokyo 102-0075, Japan

³Institut Laue Langevin, Boîte Postale 156, F-38042 Grenoble Cedex 9, France

⁴LPMCC (UMR 5493), Université Grenoble 1 and CNRS, 38042 Grenoble, France

(Dated: August 25, 2014)

Recently a *positive* spin Hall angle (SHA) of 0.021, was observed experimentally in nonmagnetic CuIr alloys [Niimi *et al.*, Phys. Rev. Lett. **106**, 126601 (2011)] and attributed predominantly to an extrinsic skew scattering mechanism, while a *negative* SHA was obtained from *ab initio* calculations [Fedorov *et al.*, Phys. Rev. B **88**, 085116 (2013)], using consistent definitions of the SHA. We reconsider the SHA in CuIr alloys, with the effects of the local electron correlation U in $5d$ orbitals of Ir impurities, included by the quantum Monte Carlo method. We found that the SHA is negative if we ignore such local electron correlation, but becomes positive once U approaches a realistic value. This may open up a way to control the sign of the SHA by manipulating the occupation number of impurities.

PACS numbers: 71.70.Ej, 72.25.Ba, 85.75.-d

The spin Hall effect, which converts charge current into spin current via the spin-orbit interaction (SOI), is one of the key phenomena for the further development of spintronics devices [1, 2]. The spin Hall angle (SHA) describes the conversion efficiency from the injected longitudinal charge current into the scattered transverse spin current. Recently a *positive* SHA of 2.1% was measured in nonmagnetic CuIr alloys [3] and argued to be predominantly due to extrinsic skew scattering, while *negative* SHA of -0.035 and -0.029 were calculated for CuIr alloys, from the Boltzmann equation and the Kubo-Streda formula respectively [4], with consistent definitions [5, 6]. The spin Hall effect in CuIr alloys in experiment is mainly due to skew scattering [3], which is well described by the phase-shift model of Fert and Levy [7, 8]. According to this model, the SHA is proportional to the phase shift parameter δ_1 , which was originally taken to be $|\delta_1| \simeq 0.1$ [7, 8]. It is given as $\delta_1 = \pi(N_p^{Ir} - N_p^{Cu})/6$, where N_p^{Ir} and N_p^{Cu} are the occupation numbers for the $6p$ orbitals around an Ir impurity and the $4p$ orbitals of Cu host, respectively. In the atomic limit $N_p^{Ir} = N_p^{Cu} = 0$, while in the CuIr alloys N_p^{Ir} and N_p^{Cu} become finite, but small, due to the mixing with the other orbitals. To predict the sign of the SHA in CuIr alloys, we therefore need to calculate precisely the sign of δ_1 [7, 8].

In this paper, we argue that the question of the *sign* of the phase shift parameter δ_1 and the SHA in CuIr alloys is highly non-trivial. Our idea is shown schematically in Fig. 1. The nonmagnetic CuIr alloys can be described by an Anderson impurity model [9], where Ir is an impurity. In a simplified Anderson model within the Hartree-Fock approximation, it is clear that due to the on-site Coulomb correlation U the impurity level increases, while the impurity occupation number decreases [9].

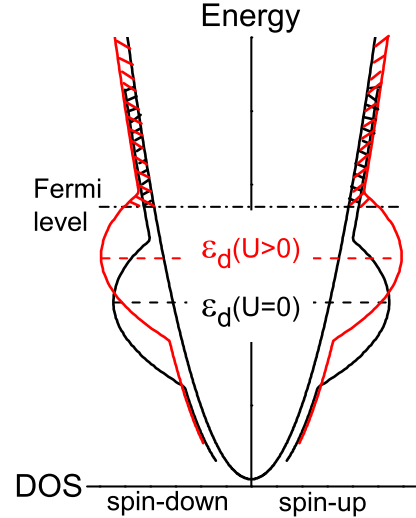


FIG. 1. (color online). Schematic picture of the density of states (DOS) of spin-up and spin-down electrons, in the nonmagnetic CuIr alloys. The DOS of Ir $5d$ orbitals are shown by the outer curves for the on-site Coulomb repulsion $U=0$ (black) and $U>0$ (red), respectively. ϵ_d is the impurity level in the Hartree-Fock approximation. The occupation number of Ir $5d$ decreases as the correlation U increases.

For Ir impurities in Cu, we define the occupation numbers of the $5d$ (N_d^{Ir}), $6s$ (N_s^{Ir}) and $6p$ (N_p^{Ir}) states to be projections of the occupied electronic states onto the impurity states of corresponding symmetry centered on the substitutional atomic site:

$$N_{s,p,d}^{Ir} = \int_{-\infty}^{E_f} \rho_{s,p,d}^{Ir}(\epsilon) d\epsilon. \quad (1)$$

E_f is the Fermi level. This projection is *not* simply onto the atomic states of the Ir atom but onto the Wannier states centered at the Ir sites and extended in the whole

supercell. In a metal the net charge of the impurity must vanish so that the total occupation number of such extended orbitals, is equal to the number of atomic valence electrons,

$$N_s^{Ir} + N_p^{Ir} + N_d^{Ir} = 9. \quad (2)$$

This constraint is respected by our density functional theory (DFT) calculation (see later).

According the Friedel sum rule, the phase shifts δ can be calculated by the occupation numbers of the corresponding orbitals as [10, 11]

$$\begin{aligned} \delta_1 &= \delta_p^{Ir} - \delta_p^{Cu} = \frac{\pi(N_p^{Ir} - N_p^{Cu})}{6}, \\ \delta_2^+ &= \delta_{d+}^{Ir} - \delta_{d+}^{Cu} = \frac{\pi(N_{d+}^{Ir} - N_{d+}^{Cu})}{6}, \\ \delta_2^- &= \delta_{d-}^{Ir} - \delta_{d-}^{Cu} = \frac{\pi(N_{d-}^{Ir} - N_{d-}^{Cu})}{4}. \end{aligned} \quad (3)$$

When the correlation U is included into the $5d$ states of the Ir impurity, the decreased N_d^{Ir} is accompanied by increased N_s^{Ir} and N_p^{Ir} , according to Eq. (2). By Eq. (3), a negative δ_1 ($\sim N_p^{Ir} - N_p^{Cu}$) may change sign and become positive, due to the increased N_p^{Ir} . This will be confirmed by the calculations in the following sections.

Spin Hall Angle due to impurities of d orbitals— For an electron scattered by a potential with the SOI, the amplitudes of the scattered wave are given by [5, 12]

$$\begin{aligned} f_\uparrow(\theta) &= f_1(\theta)|\uparrow\rangle + e^{i\varphi}f_2(\theta)|\downarrow\rangle, \\ f_\downarrow(\theta) &= f_1(\theta)|\downarrow\rangle - e^{-i\varphi}f_2(\theta)|\uparrow\rangle, \end{aligned} \quad (4)$$

for incoming spin-up and spin-down electrons. $f_1(\theta)$ and $f_2(\theta)$ represent the spin non-flip and spin flip scattering amplitudes, respectively. θ and φ are the polar and azimuthal angles of the scattered wave vector. The scattering amplitudes can be expressed in terms of the phase shifts δ_l of the orbitals l as

$$\begin{aligned} f_1(\theta) &= \sum_l \frac{P_l(\cos\theta)}{2ik} [(l+1)(e^{2i\delta_l^+} - 1) + l(e^{2i\delta_l^-} - 1)], \\ f_2(\theta) &= \sum_l \frac{-\sin\theta}{2ik} (e^{2i\delta_l^+} - e^{2i\delta_l^-}) \frac{d}{d\cos\theta} P_l(\cos\theta), \end{aligned} \quad (5)$$

with the two phase shifts $\delta_{l\pm}^\pm = \delta_{l\pm\frac{1}{2}}$ differing because of spin-orbit terms; without them $\delta_l^+ = \delta_l^- = \delta_l$.

The spin-independent part $I(\theta)$ and skewness $S(\theta)$ of the scattering cross section are represented by

$$\begin{aligned} I(\theta) &= |f_1(\theta)|^2 + |f_2(\theta)|^2, \\ S(\theta) &= \frac{2\text{Im}[f_1^*(\theta)f_2(\theta)]}{|f_1(\theta)|^2 + |f_2(\theta)|^2}. \end{aligned} \quad (6)$$

And the transport skewness γ_k is defined as [13]

$$\gamma_k = \frac{\int d\Omega I(\theta) S(\theta) \sin\theta}{\int d\Omega I(\theta) (1 - \cos\theta)}. \quad (7)$$

The SHA α can be defined either by conductivity σ or by resistivity ρ as [5, 13]

$$\alpha(\sigma) = \sigma_{yx}^{(+)} / \sigma_{xx}^{(+)} = \frac{\gamma_{k_F}}{2}, \quad (8)$$

$$\alpha(\rho) = \rho_{yx}^{(+)} / \rho_{xx}^{(+)} = -\alpha(\sigma). \quad (9)$$

We note that it was $\alpha(\rho)$ that was measured in the experiment of Ir doped Cu [3], while $\alpha(\sigma)$ was calculated in the previous theory [4]. Using consistent definitions, the SHA in Ref. [4] is opposite in sign to the experimental value of Ref. [3].

For the nonmagnetic impurity Ir as an extrinsic scatterer in Cu, the skew scattering arises from the interference between the antisymmetric scattering of $l=2$ and the symmetric scattering of $l=1$ channels [8]. It is assumed that only the d -wave scattering is the resonant channel with appreciable SOI, while the p -wave is taken spin independent. Then substituting Eq. (5) with parameters of δ_1 , δ_2^+ and δ_2^- into Eq. (9), the SHA is obtained as

$$\alpha(\rho) = -\frac{6 \sin \delta_1 [\sin(\delta_2^- - \delta_1) \sin \delta_2^- - \sin(\delta_2^+ - \delta_1) \sin \delta_2^+]}{5 (3 \sin^2 \delta_2^+ + 2 \sin^2 \delta_2^-)}. \quad (10)$$

We note that a factor $1/2$ was missing from the equation for the SHA due to skew scattering of d orbitals in Refs. [14–16], and the quoted numerical results for that angle should have been divided by 2.

LDA+SOI results.— We now calculate the phase shift parameters δ_1 , δ_2^+ and δ_2^- for the Ir impurity doped in Cu host based on DFT with the local density approximation (LDA) plus SOI. We use the code of Quantum Espresso (QE) [17] with Hubbard $U = 0$. A supercell of Cu_{26}Ir was used to calculate the occupation numbers of the orbitals around the Ir impurity, while the primitive cell of a single Cu atom gave the occupation numbers of the orbitals of the Cu host. The cutoff energy is 50 Ry for ultrasoft pseudopotentials with the Perdew-Burke-Ernzerhof (PBE) type of exchange-correlation functionals [18], and the energy convergence limit is 10^{-8} Ry, with a k lattice of $8 \times 8 \times 8$.

From these calculations for $U=0$, the occupation numbers, phase shifts and SHA are listed in Table I. The total occupation number of Ir with $5d$, $6s$ and $6p$ states was $N_s^{Ir} + N_p^{Ir} + N_d^{Ir} = 9.0$, in agreement with Eq. (2). The negative sign of the obtained SHA is opposite to the positive value of $+2.1\%$ in experiment [3]. However, the small value of δ_1 is consistent with previous estimation of $|\delta_1| \simeq 0.1$ [7, 8]. Motivated by the idea in Fig. 1, we will check the effect of local electron correlation U in the following section.

U (eV)	N_d^{Ir}	δ_2^+	δ_2^-	N_s^{Ir}	N_p^{Ir}	δ_1	SHA
0	7.82	-0.73	-0.38	0.32	0.86	-0.05	-1.1%
0.1	7.50	-0.76	-0.57	0.41	1.09	0.07	+0.8%
0.2	7.20	-0.89	-0.61	0.49	1.31	0.19	+2.5%
0.3	6.82	-1.06	-0.65	0.59	1.59	0.33	+4.6%
0.4	6.38	-1.26	-0.70	0.70	1.92	0.50	+5.4%
0.5	5.96	-1.45	-0.75	0.82	2.22	0.66	+3.4%

TABLE I. Occupation numbers (N), phase shifts (δ), and calculated SHA for an Ir impurity in a Cu host, with local electron correlation U on the $5d$ orbitals of Ir varying from 0 to 0.5 eV. For $U=0$ (first line, in boldface), the occupation numbers are calculated with LDA+SOI by DFT, which also gave $N_p^{Cu}=0.96$ and $N_d^{Cu}=9.68$ for the Cu host. For $U > 0$, N_d^{Ir} , δ_2^+ and δ_2^- are calculated by the QMC method. N_s^{Ir} and N_p^{Ir} are then estimated from Eq. (2), keeping the ratio of N_p^{Ir}/N_s^{Ir} as for $U=0$. For each value of U the phase shift δ_1 follows from the Friedel sum rule, Eq. (3) and the SHA from Eq. (10) in the text.

Quantum Monte Carlo results.— Due to the experimental observation that the SHA of CuIr is independent of the concentration of Ir impurities [3], we employ a single-impurity multi-orbital Anderson model with SOI [9, 15, 16]

$$\begin{aligned}
H = & \sum_{\mathbf{k}, \alpha, \sigma} \epsilon_{\mathbf{k}\alpha} c_{\mathbf{k}\alpha\sigma}^\dagger c_{\mathbf{k}\alpha\sigma} + \sum_{\mathbf{k}, \alpha, \xi, \sigma} (V_{\xi\mathbf{k}\alpha} d_{\xi\sigma}^\dagger c_{\mathbf{k}\alpha\sigma} + H.c.) \\
& + \sum_{\xi, \sigma} \epsilon_\xi n_{\xi\sigma} + U \sum_{\xi} n_{\xi\uparrow} n_{\xi\downarrow} + \frac{U'}{2} \sum_{\xi \neq \xi', \sigma, \sigma'} n_{\xi\sigma} n_{\xi'\sigma'} \\
& - \frac{J}{2} \sum_{\xi \neq \xi', \sigma} n_{\xi\sigma} n_{\xi'\sigma} + \frac{\lambda}{2} \sum_{\xi, \sigma} d_{\xi\sigma}^\dagger (\ell)_{\xi\xi}^z (\sigma)_{\sigma\sigma}^z d_{\xi\sigma}, \quad (11)
\end{aligned}$$

where $\epsilon_{\mathbf{k}\alpha}$ is the energy band α of host Cu, ϵ_ξ is the energy level of the $5d$ orbital ξ of impurity Ir, $V_{\xi\mathbf{k}\alpha}$ is the hybridization between the $5d$ orbital ξ of Ir and the band α of Cu, and λ is the strength of SOI. U (U') is the on-site Coulomb repulsion within (between) the $5d$ orbitals of Ir, and J is Hund coupling between the $5d$ orbitals of Ir. The $\epsilon_{\mathbf{k}\alpha}$, ϵ_ξ and $V_{\xi\mathbf{k}\alpha}$ can be obtained from the codes of QE [17] and Wannier90 [19], respectively [20].

We apply the quantum Monte Carlo (QMC) method [20–22], which can correctly include the local electron correlations, to calculate the occupation numbers n_ξ of $5d$ orbitals of Ir. The parameters of correlation and SOI of Ir are given to be $U=0.5$ eV and $\lambda=0.5$ eV [23, 24]. The relations of $U = U' + 2J$ and $J/U = 0.3$ [25] give $J=0.15$ eV and $U'=0.2$ eV.

In order to allow convergence down to room temperature in a tractable calculation, of the five $5d$ orbitals of Ir only the three t_{2g} orbitals were retained, and only the diagonal component of the SOI is conserved. It is reasonable to neglect the e_g orbitals because there are no spin-orbit matrix elements. It is convenient to transform the three t_{2g} orbitals into t_{-1} , t_0 and t_1 : $t_{-1} \equiv -\frac{1}{\sqrt{2}}(xz - iyz) = -Y_{2,-1}$, $t_0 \equiv -ixy = -\frac{1}{\sqrt{2}}(Y_{2,2} - Y_{2,-2})$

and $t_1 \equiv -\frac{1}{\sqrt{2}}(xz + iyz) = Y_{2,1}$, which are expressed by spherical harmonics with the orbital angular momentum $\ell^z = -1, 0$ and 1 of $l=2$, respectively. The QMC calculation is performed with more than 10^5 Monte Carlo sweeps, the Matsubara time step $\Delta\tau=0.25$, and temperature $T=360$ K.

The QMC calculation [15, 16] can give the occupation number $\langle n_{-1} \rangle$, $\langle n_0 \rangle$ and $\langle n_1 \rangle$ for each t_{2g} orbital. The average value of the z component of spin-orbit correlation function $\langle \ell^z \sigma^z \rangle \equiv \langle -n_{-1\uparrow} + n_{-1\downarrow} + n_{1\uparrow} - n_{1\downarrow} \rangle$, can also be obtained. The occupation numbers of parallel and antiparallel states can be written as $n_P = \langle n_{-1\downarrow} \rangle + \langle n_{1\uparrow} \rangle$, $n_{AP} = \langle n_{-1\uparrow} \rangle + \langle n_{1\downarrow} \rangle$, which can be obtained from the relations of $\langle \ell^z \sigma^z \rangle = n_P - n_{AP}$ and $\langle n_1 \rangle + \langle n_{-1} \rangle = n_P + n_{AP}$. These occupation numbers are related to the the phase shift of the parallel (antiparallel) state of the t_{2g} orbitals of Ir as $\delta_P = \pi n_P / 2$ ($\delta_{AP} = \pi n_{AP} / 2$). We assume that δ_P (resp. δ_{AP}) from the simplified model can be taken as the phase shift δ_{d+}^{Ir} (δ_{d-}^{Ir}) of the full spin-orbit split $5d$ orbital of Ir.

With correlation $U=0.5$ eV on the $5d$ states of Ir impurity, the QMC calculation gives $\delta_{d+}^{Ir} = 1.59$ and $\delta_{d-}^{Ir} = 2.30$. The occupation numbers can be calculated as $N_{d+}^{Ir} = 6\delta_{d+}^{Ir} / \pi = 3.03$ and $N_{d-}^{Ir} = 4\delta_{d-}^{Ir} / \pi = 2.93$, and the phase shifts of δ_2^+ and δ_2^- then obtained by Eq. (3) to be -1.45 and -0.75 , respectively, as listed in the case of $U=0.5$ eV in Table I. Comparing the cases with U of 0.0 and 0.5 eV in Table I, it is clear that the splitting of the phase shifts, $|\delta_2^+ - \delta_2^-|$, is enlarged from 0.35 to 0.70 by the correlation $U=0.5$ eV. Moreover, the occupation number of $5d$ states is $N_d^{Ir} = N_{d+}^{Ir} + N_{d-}^{Ir} = 5.96$, which is smaller than the $N_d^{Ir} = 7.82$ without U , in agreement with the schema in Fig. 1.

The decreased electron number in $5d$ states, which is 1.86, will be transferred to the $6s$ and $6p$ states of Ir impurity as discussed by Eq.(2). We do not know precisely how many electrons are transferred to the $6s$ states, and how many to the $6p$ states. As an approximate estimation [26], we fix the ratio $N_p^{Ir}/N_s^{Ir}=2.7$ which is obtained by LDA with $U=0$. This gives the increased $N_p^{Ir}=2.22$, as shown in the case of $U=0.5$ eV in Table I. Since the occupation numbers for pure Cu (N_p^{Cu} , N_d^{Cu}) do not change, the data of $N_p^{Cu}=0.96$ and $N_d^{Cu}=9.68$ obtained from the $U=0$ case are employed for calculating δ_1 and SHA for all positive U in Table I. Finally, positive δ_1 of 0.66 and SHA of +3.4% follow from Eqs. (3) and (10), respectively, for $U=0.5$ eV. The SHA agrees in sign, but overestimates the experimental of +2.1% in magnitude [3]. Our theory indicates, then, that the correlation U on the $5d$ states of Ir impurity is the crucial factor to give a positive phase shift δ_1 and a positive SHA as in experiment.

To show the change of SHA between cases of $U=0$ and $U=0.5$ eV, intermediate values of U were calculated. Keeping the condition in Eq.(2) and fixing the ra-

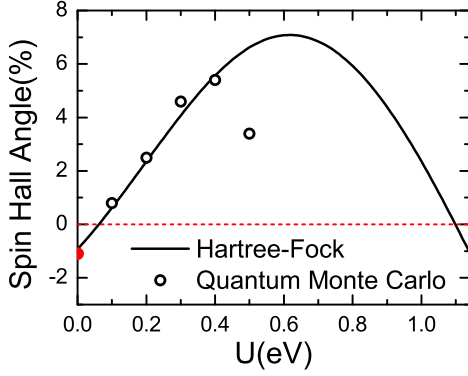


FIG. 2. (color online). Calculated SHA by LDA under $U=0$ (red dot), by QMC under $U > 0$ (black circles) from Table I, and by the Hartree-Fock approximation for spin-orbit-split $5d$ orbitals (black line).

tio $N_p^{Ir}/N_s^{Ir}=2.7$, the resulting SHA by Eq. (10) ranges from -1.1% to $+5.4\%$, as listed in Table I and plotted in Fig. 2. An U as small as 0.1 eV would be enough to change the sign of SHA. As for the discrepancy in magnitude compared to experiment for the more realistic value of $U=0.5$ eV, the energies of the $5d$ states of Ir without correlation were determined by LDA, which tends to overestimate the level of the states under the Fermi level. The level of the $5d$ states of Ir is then too close to the Fermi level. This then overestimates the decrease of N_d^{Ir} for a given correlation U . Errors in the predicted magnitude of the SHA may also come from the simplification from five $5d$ orbitals to three t_{2g} orbitals of Ir in the Hamiltonian in Eq. (11), as well as uncertainty in the parameters of U and N_p^{Ir}/N_s^{Ir} . Accurate U and N_p^{Ir} might be measured by the X-ray spectroscopy [27].

For comparison, we also show in Fig. 2 the SHA using a Hartree-Fock calculation of the occupation numbers of the full set of spin-orbit split d orbitals, using the same values of U' and J as in the QMC and obtaining the width Δ and levels for $E_{0,d+}$ and $E_{0,d-}$ from LDA, but neglecting crystal field splitting:

$$\begin{aligned} \Delta \cot(\pi n_{d+}) &= E_{0,d+} \\ &+ U n_{d+} + U'(4n_{d+} + 4n_{d-}) - J(2n_{d+} + 2n_{d-}) + \lambda, \\ \Delta \cot(\pi n_{d-}) &= E_{0,d-} \\ &+ U n_{d-} + U'(6n_{d+} + 2n_{d-}) - J(3n_{d+} + n_{d-}) - \frac{3}{2}\lambda. \end{aligned} \quad (12)$$

We note that for the entire range of U shown in Fig. 2, the non-magnetic solution is stable following Ref. [9]. This shows that the change of sign and non-monotonic behavior of the SHA as a function of U are *not* a result of the projection onto t_{2g} states but more general.

Discussion— Since the sign of SHA is sensitive to the sign of δ_1 [Eq. (10)] and the corresponding small change

of N_p^{Ir} [Eq. (3)], the sign of SHA might be controlled as long as the occupation number of the $6p$ states of the impurity were properly manipulated. For instance, a laser pulse [28] can decrease the occupation number of the impurity by excitation. An improved combination of noble metal hosts and $5d$ metal impurities with a long lifetime of the excited states may be imagined as a means to control the sign of the SHA.

In summary, we reconsider the theory of spin Hall effect in CuIr alloys by the QMC method, where the local Coulomb correlation U in $5d$ states of Ir impurities is included. Taking U to be 0.5 eV, we obtain a positive SHA, consistent with experiment, in contrast to the negative angle predicted without correlation U . Our result reveals the key physics determining the spin Hall effect in CuIr alloys, explaining the positive sign of the SHA in experiment. This may also open up a way to control the sign of the spin Hall effect by manipulating the occupation number of the impurities.

We are very grateful to A. Fert, P. Levy, M. Chshiev, H. X. Yang, Y. Otani, and Y. Niimi for many stimulating discussions. T. Ziman would like to thank the KITP, UCSB for hospitality during the Spintronics Program 2013, with support in part by the National Science Foundation under Grant No. NSF PHY11-25915. The work has been supported by a REIMEI project of JAEA.

-
- [1] M. I. Dyakonov and V. I. Perel, Sov. Phys. JETP Lett. **13**, 467 (1971).
 - [2] J. E. Hirsch, Phys. Rev. Lett. **83**, 1834 (1999).
 - [3] Y. Niimi, M. Morota, D. H. Wei, C. Deranlot, M. Basletic, A. Hamzic, A. Fert, and Y. Otani, Phys. Rev. Lett. **106**, 126601 (2011).
 - [4] D. V. Fedorov, C. Herschbach, A. Johansson, S. Ostanin, I. Mertig, M. Gradhand, K. Chadova, D. Kodderitzsch, and H. Ebert, Phys. Rev. B **88**, 085116 (2013).
 - [5] B. Gu, Z. Xu, M. Mori, T. Ziman, and S. Maekawa, arXiv:1402.3012 (unpublished).
 - [6] The minus sign comes from a consistent definition of the SHA, see below, after Eq. [9] and the previous reference.
 - [7] A. Fert and P. M. Levy, Phys. Rev. Lett. **106**, 157208 (2011); Phys. Rev. Lett. **111**, 199904 (2013).
 - [8] A. Fert, A. Friederich, and A. Hamzic, J. Magn. Magn. Mater. **24**, 231 (1981).
 - [9] P. W. Anderson, Phys. Rev. **124**, 41 (1961).
 - [10] Y. Niimi, Y. Kawanishi, D. H. Wei, C. Deranlot, H. X. Yang, M. Chshiev, T. Valet, A. Fert, and Y. Otani, Phys. Rev. Lett. **109**, 156602 (2012).
 - [11] D. C. Langreth, Phys. Rev. **150**, 516 (1966).
 - [12] H. A. Bethe, and R. Jackiw, *Intermediate Quantum Mechanics* (The Benjamin/Cummings Publishing Company, Inc. Menlo Park, CA, 1986), Chap. 16.
 - [13] H. A. Engel, B. I. Halperin, and E. I. Rashba, Phys. Rev. Lett. **95**, 166605 (2005).
 - [14] G. Y. Guo, S. Maekawa, and N. Nagaosa, Phys. Rev. Lett. **102**, 036401 (2009).
 - [15] B. Gu, J.-Y. Gan, N. Bulut, T. Ziman, G.-Y. Guo, N.

- Nagaosa, and S. Maekawa, Phys. Rev. Lett. **105**, 086401 (2010).
- [16] B. Gu, I. Sugai, T. Ziman, G. Y. Guo, N. Nagaosa, T. Seki, K. Takanashi, and S. Maekawa, Phys. Rev. Lett. **105**, 216401 (2010).
- [17] P. Giannozzi et al., <http://www.quantum-espresso.org>.
- [18] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).
- [19] A. A. Mostofi et al., wannier90: A Tool for Obtaining Maximally-Localised Wannier Functions, Comput. Phys. Commun., **178**, 685 (2008).
- [20] B. Gu, J.-Y. Gan, N. Bulut, G.-Y. Guo, N. Nagaosa, and S. Maekawa, J. Phys. Conf. Ser. **200**, 062007 (2010).
- [21] J. E. Hirsch and R. M. Fye, Phys. Rev. Lett. **56**, 2521 (1986).
- [22] W. M. C. Foulkes, L. Mitas, R. J. Needs, and G. Rajagopal, Rev. Mod. Phys. **73**, 33 (2001).
- [23] S. J. Moon, H. Jin, K.W. Kim, W. S. Choi, Y. S. Lee, J. Yu, G. Cao, A. Sumi, H. Funakubo, C. Bernhard, and T.W. Noh, Phys. Rev. Lett. **101**, 226402 (2008).
- [24] B. J. Kim, H. Jin, S. J. Moon, J.-Y. Kim, B.-G. Park, C. S. Leem, J. Yu, T. W. Noh, C. Kim, S.-J. Oh, J.-H. Park, V. Durairaj, G. Cao, and E. Rotenberg, Phys. Rev. Lett. **101**, 076402 (2008).
- [25] S. Maekawa, T. Tohyama, S. E. Barnes, S. Ishihara, W. Koshibae and G. Khaliullin, *Physics of Transition Metal Oxides*, Springer, 2004.
- [26] Due to the fact that the s and p waves of Ir are quite extended compared to the d waves, we assume the same ratio of N_p^{Ir}/N_s^{Ir} in the model with finite U on 5d around 0.5 eV. The sign of SHA with U=0.5 eV is not sensitive to the ratio, which is confirmed by varying the ratio around 2.7 by 50%, from 1.4 to 4.1. For the ratio of 1.4, it gives $N_p^{Ir}=1.78$ and SHA=+4.1%; for the ratio of 4.1, it gives $N_p^{Ir}=2.45$ and SHA=+2.2%.
- [27] J. Ghijsen, L. H. Tjeng, J. van Elp, H. Eskes, J. Westerkamp, G. A. Sawatzky, and M. T. Czyzyk, Phys. Rev. B **38**, 11322 (1988).
- [28] V. P. Zhukov, O. Andreyev, D. Hoffmann, M. Bauer, M. Aeschlimann, E. V. Chulkov, and P. M. Echenique, Phys. Rev. B **70**, 233106 (2004).