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What determines the sign of the spin Hall effects in Cu alloys doped with 5d elements?

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ABSTRACT

We perform a systematical analysis of the spin Hall effect (SHE) in the Cu alloys doped with a series of 5*d* elements, by the combined approach of density functional theory and Hartree–Fock approximation. We find that not only the spin orbit interactions (SOI) in both the 5*d* and 6*p* orbitals, but also the local correlations in the 5*d* orbitals of the impurities, are decisive on the sign of the spin Hall angle (SHA). Including all of these three factors properly, we predict the SHA for each alloy in the series. The signs of CuIr and CuPt are sensitive to perturbation of the local correlations. This observation is favorable for controlling the sign of the transverse spin Hall voltage.

1. Introduction

Spintronics is a rich research field, not only for the wide applications of low energy consumption and energy transformation devices, but also for the new physics with the interplays among charge, spin, orbital, heat and so on [1]. The spin Hall effect (SHE), which converts the injected longitudinal charge current into the transverse spin current via the spin–orbit interaction (SOI), is crucial for the development of spintronic devices. The SHE is characterized by the ratio between the transverse and the longitudinal resistivities (or conductivities), called spin Hall angle (SHA). The magnitude of SHA describes conversion efficiency between the charge current and the spin current, while the sign distinguishes the scattering direction of electrons, i.e., clockwise or anticlockwise into the transverse direction. It is well known that to raise the magnitude of SHA is the key to enhance the whole efficiency of the devices based on the SHE [2,3]. However, the sign of SHA is not yet utilized as a degree of freedom in spintronic devices.

In the experiment of the dilute CuIr alloys, the dominant contribution to the SHE was verified to be an extrinsic skew scattering mechanism, and the SHA was measured to be positive 2.1% [4]. We have found out in theory that including the local correlation effects of the 5*d* orbitals of Ir is decisive to obtain a positive sign of SHA consistent with experiment, and a small change of the 5*d* electron number of Ir with the local correlations may change the sign [5,6].

On the other hand, among the previous theoretical works about the SHE of the Cu alloys doped with 5*d* elements, Fert and Levy estimated the SHA by the atomic electron numbers of 5*d* orbitals with SOI, obtaining a sign change of SHA in the middle of the series of 5*d* impurities [7]. In contrary, based on the *ab initio* calculations, including the SOI in both 5*d* and 6*p* orbitals, Fedorov et al. obtained an uniform sign of SHA contributed by the skew scattering among the series of 5*d* impurities from Lu to Pt [8].

In the present work, we analyze the SHE among the dilute Cu alloys with a series of 5*d* elements as impurities, and find the key factors which are decisive on the sign of SHA. Furthermore, we screen out the alloys whose signs of SHA are sensitive to perturbation of the local correlations as CuIr, which will be favorable for the sign control of SHE.

2. Theoretical approach

The extrinsic SHE is caused by the spin–orbit interactions (SOI) in the orbitals of the impurities. As one of the mechanisms of extrinsic SHE, the skew scattering generates the spin Hall resistivity linear with the impurity concentration in the dilute alloys. Based on the Anderson model [9], the contribution of skew scattering on the extrinsic SHE in the nonmagnetic Cu alloys with dilute 5*d* impurities can be described by a single-impurity multi-

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orbital model, including the SOI in both the 6*p* orbitals ζ and the 5*d* orbitals ξ of the impurity with the parameters λ_p and λ_d, respectively, together with the local correlations of the on-site Coulomb repulsion U (U') within (between) the 5*d* orbitals, and the Hund coupling J between the 5*d* orbitals of the impurity [6]:

$$\begin{aligned} H_0 = & \sum_{\mathbf{k},\alpha,\sigma} \epsilon_{\alpha\mathbf{k}} c^{\dagger}_{\mathbf{k}\alpha\sigma} c_{\mathbf{k}\alpha\sigma}, \\ & + \sum_{\mathbf{k},\alpha,\beta,\sigma} (V_{\beta\mathbf{k}\alpha} d^{\dagger}_{\beta\sigma} c_{\mathbf{k}\alpha\sigma} + \text{H. c.}) + \sum_{\beta,\sigma} \epsilon_{\beta} d^{\dagger}_{\beta\sigma} d_{\beta\sigma}, \\ & + \frac{\lambda_p}{2} \sum_{\zeta\sigma,\zeta'\sigma'} d^{\dagger}_{\zeta\sigma}(\mathbf{l})_{\zeta\zeta'}(\boldsymbol{\sigma})_{\sigma\sigma'} d_{\zeta'\sigma'} \\ & + \frac{\lambda_d}{2} \sum_{\xi\sigma,\xi'\sigma'} d^{\dagger}_{\xi\sigma}(\mathbf{l})_{\xi\xi'}(\boldsymbol{\sigma})_{\sigma\sigma'} d_{\xi'\sigma'}, \end{aligned} \tag{1}$$

$$\begin{aligned} H = & H_0 + U \sum_{\xi} n_{\xi\uparrow} n_{\xi\downarrow} \\ & + \frac{U'}{2} \sum_{\xi\neq\xi',\sigma,\sigma'} n_{\xi\sigma} n_{\xi'\sigma'} - \frac{J}{2} \sum_{\xi\neq\xi',\sigma} n_{\xi\sigma} n_{\xi'\sigma}. \end{aligned} \tag{2}$$

In Eq. (1), $\epsilon_{\alpha\mathbf{k}}$ is the energy band α of the Cu host, ϵ_{β} is the energy level of the orbital β of the 5*d* impurity, and $V_{\beta,\alpha}(\mathbf{k})$ is the hybridization between the orbital β of the impurity and the band α of the host. In Eq. (2), the local correlations are included only within the localized 5*d* orbitals ξ of the impurity, employing the relations of $U = U' + 2J$ [10] and $J/U = 0.3$.

In the orbital with the orbital angular momentum l, the SOI split the states of the orbital into two groups of degenerated states with the total angular momentum $j = l \pm \frac{1}{2}$ and the degeneracy $D_{l\pm} = 2j + 1$. In the Cu alloys with the 5*d* impurities including SOI, according to the Friedel sum rule [11,12], the phase shifts $\delta_l^{\pm}$ can be calculated from the occupation numbers $N_{l\pm}$ of the spin–orbit split states of the impurity and the Cu host,

$$\delta_l^{\pm} = \pi (N_{l\pm}^{imp} - N_{l\pm}^{Cu})/D_{l\pm}, \tag{3}$$

where $l=1$ corresponds to the 6*p* states of the impurity and the 4*p* states of Cu, $l=2$ corresponds to 5*d* states of the impurity and the 3*d* states of Cu, respectively. The total occupation number of the 5*d* states of the impurity N_d^{imp} can be obtained by

$$N_d^{imp} = N_{2+}^{imp} + N_{2-}^{imp}. \tag{4}$$

The occupation number of each of the degenerate 5*d* states of the impurity $n_{d\pm}$ can be obtained by

$$n_{d\pm} = N_{2\pm}^{imp}/D_{2\pm}, \tag{5}$$

where $n_{d\pm}$ will be between 0 and 1. The occupation numbers of the impurities are defined via projections of the occupied states onto the Wannier states centered at the impurities as point defects and extended in the whole supercell. In the dilute alloys, a point defect cannot be charged in metal. Thus the total occupation numbers of the 6*s*, 6*p* and 5*d* states of the impurities are conserved to the atomic valence electron numbers [5,6],

$$N_s^{imp} + N_p^{imp} + N_d^{imp} = C^{imp}, \tag{6}$$

where the constant C^{imp}=4, 5, 6, 7, 8, 9, 10, 11 and 12, for the impurities of Hf, Ta, W, Re, Os, Ir, Pt, Au and Hg, respectively.

Based on the model in Eq. (1), by the Hartree–Fock (HF) approximation, the 5*d* states of the impurities can be considered as virtual bound states with a width parameter Δ, and the $5d\pm$ states correspond to the energy levels of $\epsilon_{0,d\pm}$, respectively. It has the relations of [6,9]

$$\Delta \cot(\pi n_{d\pm}) = \epsilon_{0,d\pm} - \epsilon_F = E_{0,d\pm}, \tag{7}$$

where the ϵ_F is the Fermi level and $E_{0,d\pm}$ are the energy levels of the $5d\pm$ states relative to the Fermi level. While the local correlations are included in the 5*d* states of the impurity as in Eq. (2), it has the self-consistent relations of [6]

$$\begin{aligned} E_{d\pm} = & \Delta \cot(\pi n_{d\pm}) \\ = & E_{0,d\pm} + U\left(\frac{3}{5}n_{d+} + \frac{2}{5}n_{d-}\right) + U'\left(\frac{24}{5}n_{d+} + \frac{16}{5}n_{d-}\right) \\ & - J\left(\frac{12}{5}n_{d+} + \frac{8}{5}n_{d-}\right), \end{aligned} \tag{8}$$

which will raise the 5*d* energy level and lower the 5*d* occupation numbers of the impurity [5,6,9]. The decrease of N_d^{imp} will induce the increase of N_p^{imp} according to Eq. (6), which can be approximately estimated by fixing the ratios of N_p^{imp}/N_s^{imp} and $N_{1+}^{imp}/N_{1-}^{imp}$. The corresponding phase shifts can be obtained by Eq. (3).

For the SHE contributed by the skew scattering in Cu alloys with 5*d* impurities, the SHA defined in terms of resistivity ρ [5,13] can be calculated from the phase shifts $\delta_1^{\pm}$ of the $p\pm$ channels and $\delta_2^{\pm}$ of the $d\pm$ channels. Including the SOI in both the *p* and *d* channels, the SHA Θ is obtained by [6]

$$\begin{aligned} & \Theta(\delta_1^+, \delta_1^-, \delta_2^+, \delta_2^-) = A/B, \\ & A = -2[9\sin(\delta_1^+ - \delta_2^+)\sin\delta_1^+\sin\delta_2^+ \\ & \quad - 4\sin(\delta_1^+ - \delta_2^-)\sin\delta_1^+\sin\delta_2^- \\ & \quad - 5\sin(\delta_1^- - \delta_2^-)\sin\delta_1^-\sin\delta_2^-], \\ & B = 45\sin^2\delta_2^+ + 30\sin^2\delta_2^- + 50\sin^2\delta_1^+ + 25\sin^2\delta_1^- \\ & \quad + 6\sin\delta_1^+\sin(2\delta_2^+ - \delta_1^+) + 12\sin\delta_1^-\sin(2\delta_2^+ - \delta_1^-) \\ & \quad + 14\sin\delta_1^+\sin(2\delta_2^- - \delta_1^+) - 2\sin\delta_1^-\sin(2\delta_2^- - \delta_1^-). \end{aligned} \tag{9}$$

When the SOI in the *p* (or *d*) channels are omitted, with $\delta_1 = \delta_1^+ = \delta_1^-$ (or $\delta_2 = \delta_2^+ = \delta_2^-$), the simplified formula for SHA $\Theta_d(\delta_1, \delta_1, \delta_2^+, \delta_2^-)$ (or $\Theta_p(\delta_1^+, \delta_1^-, \delta_2, \delta_2)$) can be obtained from Eq. (9).

Based on the model in Eq. (2) and the formula in Eq. (9), to calculate the SHA for the Cu alloys with a series of 5*d* impurities, we employ the combined method of the density functional theory (DFT) and HF approximation, as we have done before for the CuIr alloys [6].

3. Results and discussions

For the dilute alloys of Cu with a series of 5*d* impurities, including the SOI in both the 5*d* and 6*p* orbitals, the 5*d* occupation numbers N_d and the SHA are calculated by the DFT+HF method following Eqs. (8) and (9), without the local correlations (U=0) or with the local correlations of U=0.5 eV in the 5*d* orbitals of the impurities, respectively, as shown in Fig. 1. Estimates from experiments give the parameter of the on-site Coulomb repulsion U of 5*d* orbitals in the range of 0–1.0 eV for pure metals [14], 0–1.5 eV for insulating compounds [15], less than 0.4 eV for the ferromagnetic metal of osmates [15], about 0.5 eV for iridates [16], and 0.5–0.6 eV for Pt alloys [14]. Below we take U=0.5 eV to compare the whole series of 5*d* impurities in Cu alloys.

As U switches from 0 to 0.5 eV, according to Eq. (2) based on the Anderson model [5,6,9], the 5*d* state will be shifted up, accompanied by a decrease of N_d. The amount of the shift varies among the 5*d* impurities, as intuitively shown in Fig. 2. The N_d