Difference between magnetotransport properties of **doped alloys** and **doped crystals** via ab-initio calculations

Demise of a traditional paradigm

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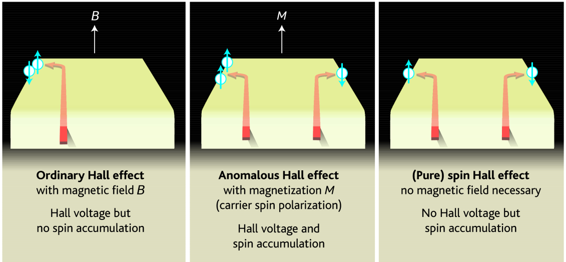
Outline

- Anomalous Hall effect (AHE), spin Hall effect (SHE): what to look for
- Substitutional alloy: ways of dealing with disorder
- Results on AHE and SHE conductivities σ_{xy} and σ_{xy}^{z} for an $Fe_{19}Ni_{81}$ alloy doped with V, Co, Pt, Au.
- Relativity principle of scattering
- Bonus: Temperature-dependence of AHE and SHE

Off-diagonal conductivity

Focusing on current along x if applied electric field is along y.

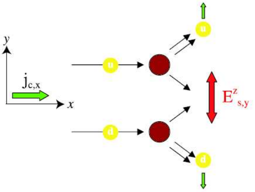


Inoue & Ohno, Science **309**, 2004 (2005)

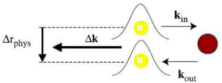
AHE	**SHE**
$\boldsymbol{\sigma}_{xy}$	$\boldsymbol{\sigma}^{z}_{xy}$

"Normal" diagonal conductivity σ_{xx}: current along x, field along x.

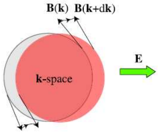
Useful intuitive concepts based on semiclassical approach



skew scattering



side jump



intrinsic

Vignale, J. Supercond. Nov. Magn. **23**, 3 (2010)

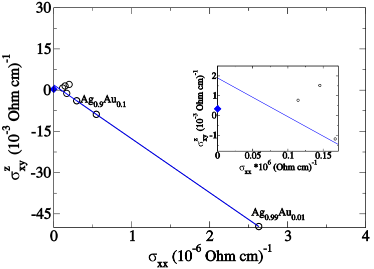
Dependence of AHE on the concentration c of impurities:

$$\sigma_{xy}^{\text{skew}} \sim c^{-1}\,, \qquad \sigma_{xy}^{\text{s-j}} \sim c^{0}\,, \qquad \sigma_{xy}^{\text{intr}} \sim c^{0}$$

$$\rho_{xy}^{\text{skew}} \sim c\,, \qquad \rho_{xy}^{\text{s-j}} \sim c^{2}\,, \qquad \rho_{xy}^{\text{intr}} \sim c^{2}$$

Dilute limit: dependence of σ_{xy} and of σ^z_{xy} on σ_{xx}

Impurities in crystal: low c means high σ_{xx}.

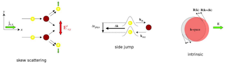


In the dilute limit skew scattering dominates and is proportional to σ_{xx}.

$$\sigma^{\text{skew}}_{xy} = S\,\sigma_{xx}\ .$$

Lowitzer, PhD thesis (2010)

Contributions due to $\sigma^{\text{skew}}_{xy}$, $\sigma^{\text{s-j}}_{xy}$, and $\sigma^{\text{intr}}_{xy}$ to σ_{xy} can be disentangled by extrapolating in the dilute limit the linear behaviour ($\sigma_{xy} \sim \sigma_{xx}$) down to $\sigma_{xx} = 0$.



Ab-initio: Kubo-Bastin equation

Generalized conductivity $\mathcal{C}_{\mu\nu}$, generalized current operator $\hat{O}_{\mu}$

$$
\begin{aligned}
\mathcal{C}_{\mu\nu} &= \mathcal{C}^{I}_{\mu\nu} + \mathcal{C}^{II}_{\mu\nu} \ , \\
\mathcal{C}^{I}_{\mu\nu} &= \frac{\hbar}{4\pi\Omega} \operatorname{Tr} \left\langle \hat{O}_{\mu}(\hat{G}^{+} - \hat{G}^{-})\hat{j}_{\nu}\hat{G}^{-} - \hat{O}_{\mu}\hat{G}^{+}\hat{j}_{\nu}(\hat{G}^{+} - \hat{G}^{-}) \right\rangle_{c} \ , \\
\mathcal{C}^{II}_{\mu\nu} &= \frac{\hbar}{4\pi\Omega} \int_{-\infty}^{E_{\mathrm{F}}} \operatorname{Tr} \left\langle \left(\hat{O}_{\mu}\hat{G}^{+}\hat{j}_{\nu}\frac{d\hat{G}^{+}}{dE} - \hat{O}_{\mu}\frac{d\hat{G}^{+}}{dE}\hat{j}_{\nu}\hat{G}^{+} \right) - \right. \\
&\qquad \left. \left(\hat{O}_{\mu}\hat{G}^{-}j_{\nu}\frac{d\hat{G}^{-}}{dE} - \hat{O}_{\mu}\frac{d\hat{G}^{-}}{dE}\hat{j}_{\nu}\hat{G}^{-} \right) \right\rangle_{c} \mathrm{d}E \ .
\end{aligned}
$$

Relativistic electric current operator: $\hat{\mathbf{j}} = -|e|c\boldsymbol{\alpha}$.

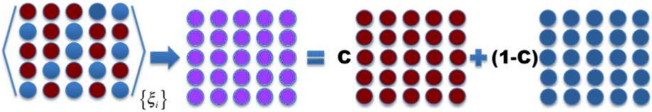
Relativistic spin current density operator:

$$\hat{J}^{z}_{\mu} = \left(\beta\Sigma_{z} - \frac{\gamma_{5}\hat{p}_{z}}{mc} \right) |e|c\alpha_{\mu} \ .$$

Treatment of disorder

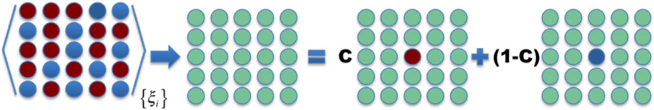
virtual crystal approximation (VCA)

system treated as a *crystal* of "intermediate" atomic type



coherent potential approximation (CPA)

disorder is included



Investigated system, used methods

Permalloy (Py) $Fe_{19}Ni_{81}$ doped with V, Co, Au, and Pt atoms.

Host is not a crystal but an alloy.

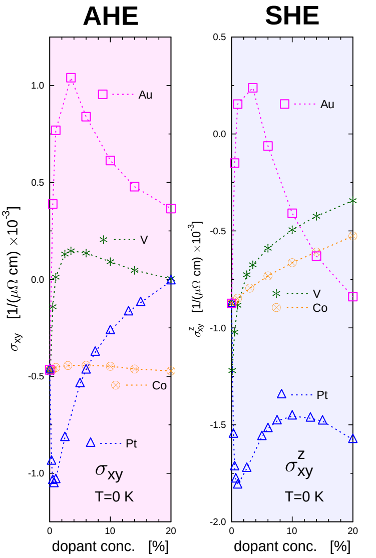
Fully relativistic spin-polarized KKR-Green function formalism, implemented in the SPRKKR code.

Generalized gradient approximation using PBE functional.

What to do:

Investigate how anomalous and spin Hall conductivities σ_{xy} and σ^{z}_{xy} depend on the concentration of V, Co, Au, and Pt impurities.

Dependence of σ_{xy} and σ_{xy}^z on dopant concentration



Anomalous Hall conductivity σ_{xy} (AHE).

Spin Hall conductivity σ_{xy}^z (SHE).

- Highly nonmonotonic dependence on the dopant concentration.
- Different dopants give rise to quite different dependencies.
- Even the sign of σ_{xy} or σ_{xy}^z can be reverted by doping.

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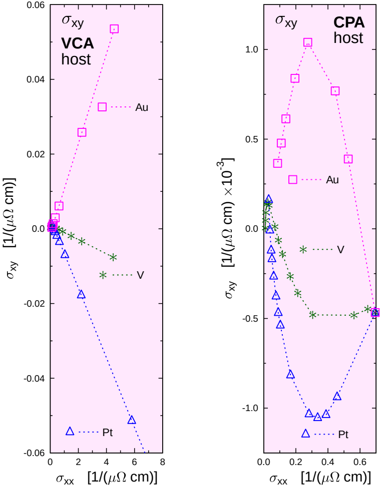
Dependence of AHE σ_{xy} on σ_{xx} *(disorder in the host)*

host formally treated as

crystal (VCA) alloy (CPA)



If host is treated as a crystal (via VCA, *which is wrong*), σ_{xy} is proportional to σ_{xx} for low dopant concentrations.

If host is treated as alloy (via CPA), off-diagonal conductivities σ_{xy} is not proportional to σ_{xx} for low dopant concentrations.

$Fe_{19}Ni_{81}$ doped with V, Au, and Pt.

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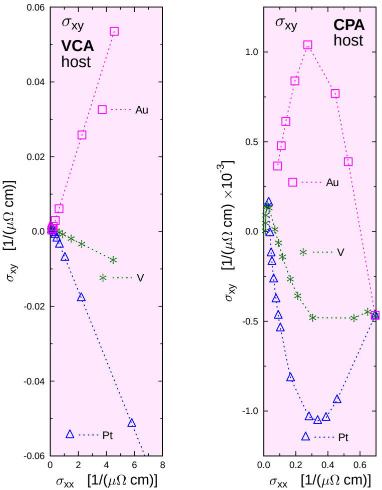


If host is treated as a crystal (via VCA, *which is wrong*), σ_{xy} is proportional to σ_{xx} for low dopant concentrations.

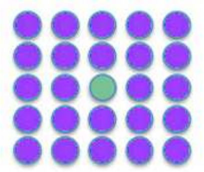
If host is treated as alloy (via CPA), off-diagonal conductivities σ_{xy} is not proportional to σ_{xx} for low dopant concentrations.

If the host is an alloy, concepts of skew scattering and side-jump scattering can be misleading.

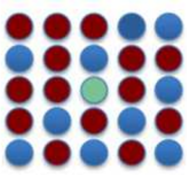
$Fe_{19}Ni_{81}$ doped with V, Au, and Pt.

Watch out: Scattering in a crystal and in an alloy

Definitions of $\sigma_{xy}^{\text{skew}}$, $\sigma_{xy}^{\text{s-j}}$, $\sigma_{xy}^{\text{intr}}$ are related to scattering.



impurity in crystal

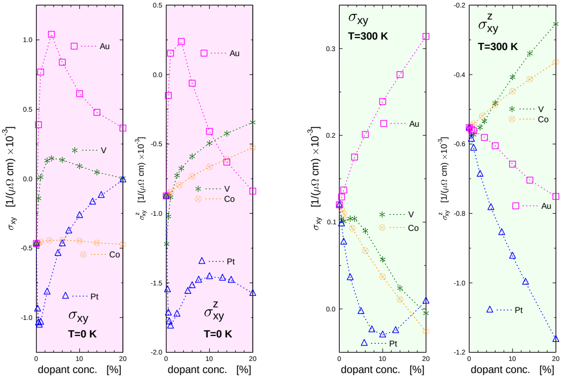


impurity in alloy

The very concept of scattering relies on a well-defined background: scattering with respect to what?

Dilute limit for alloys is not the same as clean limit, which can be achieved only for crystals.

AHE σ_{xy} and SHE σ^z_{xy} for $T = 0$ K and for $T = 300$ K

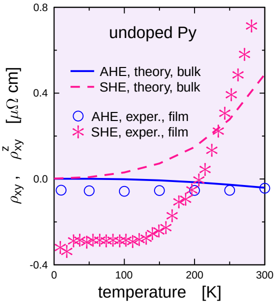


Finite temperature effects via alloy analogy model: atomic vibrations and spin fluctuations are treated as localized and uncorrelated.

Šipr *et al.*
PRB **101**, 085109 (2020)

Dependence of σ_{xy} and σ^z_{xy} on the dopants concentration is quite different for T=0 K and for T=300 K.

Comparing theory and experiment for AHE and SHE



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Temperature-dependence of AHE resistivity ρ_{xy} and of SHE resistivity ρ^{z}_{xy} for undoped Py ($Fe_{19}Ni_{81}$) obtained from our calculations and from the experiment [Omori *et al.* PRB **99**, 014403 (2019)].

Disclaimer: comparing a bit different systems — calculations done for bulk Py whereas experiment done for a thin Py film.

The agreement between theory and experiment is satisfactory enough to make the analysis based on ab-initio calculations trustworthy.

Transport in a doped alloy: Conclusions

- Having host an alloy instead of a crystal has profound influence on how off-diagonal AHE and SHE conductivities σ_{xy} and σ_{xy}^{z} depend on the dopant concentration.
  - σ_{xy} and σ_{xy}^{z} are **not** proportional to σ_{xx} for low dopant concentrations.
  - Concepts of skew scattering, side-jump scattering, or intrinsic contributions are of limited use.

- Dependence of σ_{xy} and σ_{xy}^{z} on the concentration of the dopants is non-monotonic.

- The temperature substantially changes the way the σ_{xy} and σ_{xy}^{z} depend on the concentration of impurities.

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