Ab-initio calculations of transport properties of doped permalloy

Exploring the effect of the host disorder

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Outline

Theoretical framework

Longitudinal conductivity

Anomalous Hall effect, spin Hall effect

Dependence of AHE and SHE on the temperature

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Transport: semiclassical Boltzman equation

Rate of change of the distribution function $f_{\boldsymbol{k}}$

$$-\left.\frac{\partial f_{\boldsymbol{k}}}{\partial t}\right|_{\text{scatt.}} + \left.\frac{\partial f_{\boldsymbol{k}}}{\partial t}\right|_{\text{field}} = 0$$

Relaxation time:	$\tau_{\boldsymbol{k}}$
Transition probability :	$P_{\boldsymbol{k}\boldsymbol{k}'} \sim \left|\langle\psi_{\boldsymbol{k}}\vert V_{\text{imp}}\vert\psi_{\boldsymbol{k}'}\rangle\right|^2$
Group velocity:	$\boldsymbol{v} = \frac{1}{\hbar}\frac{\partial E_{\boldsymbol{k}}}{\partial \boldsymbol{k}}$
Vector mean free path:	$\boldsymbol{\Lambda}_{\boldsymbol{k}} = \tau_{\boldsymbol{k}}\left(\boldsymbol{v}_{\boldsymbol{k}} + \sum_{\boldsymbol{k}'} P_{\boldsymbol{k}\boldsymbol{k}'}\boldsymbol{\Lambda}_{\boldsymbol{k}'}\right)$

Conductivity tensor

$$\sigma_{\mu\nu} = \frac{e^2}{(2\pi)^3}\sum_n \int_{E_{\boldsymbol{k}}=E_F} \mathrm{d}S_{\boldsymbol{k}}\,\frac{1}{v^n_{\boldsymbol{k}}}\,v^{n,\mu}_{\boldsymbol{k}}\,\Lambda^{n,\nu}_{\boldsymbol{k}}$$

Transport: Kubo-Bastin equation

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

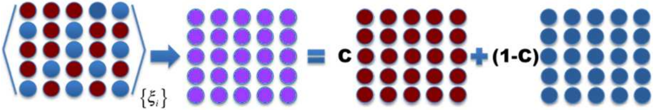
$$
\begin{aligned}
\mathcal{C}_{\mu\nu} &= \mathcal{C}^{I}_{\mu\nu} + \mathcal{C}^{II}_{\mu\nu} \ , \\
\mathcal{C}^{I}_{\mu\nu} &= \frac{\hbar}{4\pi\Omega} \mathrm{Tr} \left\langle \hat{O}_\mu (\hat{G}^+ - \hat{G}^-) \hat{j}_\nu \hat{G}^- - \hat{O}_\mu \hat{G}^+ \hat{j}_\nu (\hat{G}^+ - \hat{G}^-) \right\rangle_c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

Electric current operator $\hat{\boldsymbol{j}} = -|e|c\boldsymbol{\alpha}$.

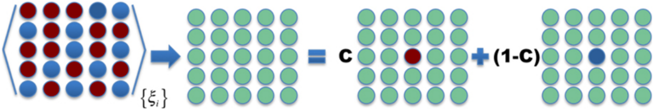
Spin current density operator $\hat{J}^z_\mu = \left(\beta\Sigma_z - \frac{\gamma_5 \hat{p}_z}{mc} \right) |e| c\alpha_\mu$.

Treatment of disorder

virtual crystal approximation (VCA)



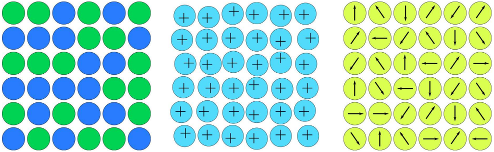
coherent potential approximation (CPA)



Vertex corrections: difference between configurational average of product and product of configurational averages

$$\left\langle \hat{O}_\mu \hat{G}^+ \hat{j}_\nu \hat{G}^- \right\rangle_c - \left\langle \hat{O}_\mu \hat{G}^+ \right\rangle_c \left\langle \hat{j}_\nu \hat{G}^- \right\rangle_c .$$

Finite temperature: alloy analogy model



Chadova, PhD thesis (2017)

Square deviation from the equilibrium position is

$$\langle u^2 \rangle_T \;=\; \frac{3\hbar^2}{m k_B \Theta_D} \left[\frac{\Phi(\Theta_D/T)}{(\Theta_D/T)} + \frac{1}{4} \right] ,$$

where $\Phi(\Theta_D/T)$ is Debye function.

Probability of spin orientation along $\hat{\boldsymbol{e}}_f$ is

$$x_f \;=\; \frac{\sin\theta_f \, \mathrm{e}^{w(T)\,\hat{\boldsymbol{z}}\cdot\hat{\boldsymbol{e}}_f\,/\,(k_B T)}}{\sum_{f'} \sin\theta_{f'} \, \mathrm{e}^{w(T)\,\hat{\boldsymbol{z}}\cdot\hat{\boldsymbol{e}}_{f'}\,/\,(k_B T)}} ,$$

where $w(T)$ is Weiss field parameter obtained from $M(T)$.

Calculation: Technical details

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

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

Generalized gradient approximation using PBE functional.

Angular momentum cutoff $\ell_{\rm max}$=3.

Potentials subject to the atomic sphere approximation (ASA).

Energy integration on a semicircle in a complex plane.

Lattice constant by minimizing the total energy (except for Co doping).

Evaluating Kubo-Bastin formula requires very dense $\boldsymbol{k}$-mesh for energies close to E_F.

Usually 576^3 $\boldsymbol{k}$-points in the full BZ used at E_F and 288^3 $\boldsymbol{k}$-points at the energy next-nearest to E_F.

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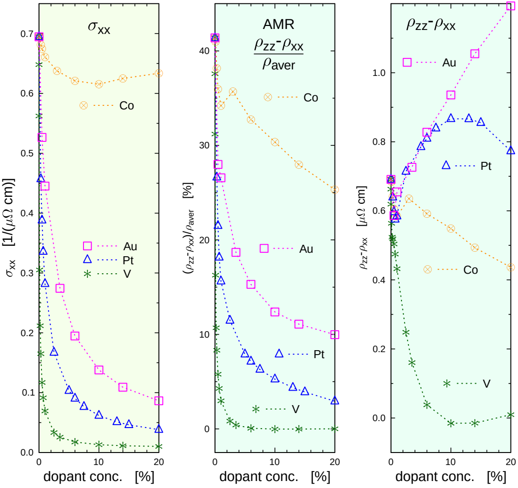
Dependence on dopant type and concentration



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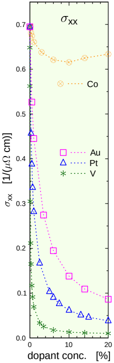
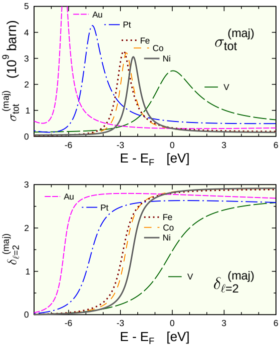
- Decrease of conductivity σ_{xx} with dopant concentration follows the sequence Co-Au-Pt-V.
- AMR defined as $(\rho_{zz} - \rho_{xx})/\rho_{\text{aver}}$ follows same pattern as σ_{xx}, because ρ_{aver} dominates.

  $\rho_{zz} - \rho_{xx}$ provides a better insight.

Why is V so harmful for conductivity of Py?



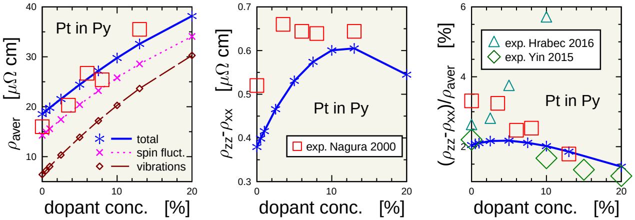


What matters is the situation at E_F.

Cross-section of majority-spin electrons $\sigma_{tot}^{(maj)}(E_F)$ decreases in order V-Pt-Au.

Note: Efficiency of V in reducing conductivity of Py is *not* linked to the antiparallel orientation of magnetic moments of V and of Fe and Ni in Py.

Comparison with experiment



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PRB **101**, 085109 (2020)

Nagura *et al.* JMMM **212**, 53 (2000)
Yin *et al.* PRB **92**, 024427 (2015)
Hrabec *et al.* PRB **93**, 014432 (2016)

Matthiessen rule (additivity of vibrations and of spin fluctuations)

$$[\rho_{\mathrm{aver}}^{(\mathrm{sfluct})}(T) - \rho_{\mathrm{aver}}(0)] + [\rho_{\mathrm{aver}}^{(\mathrm{vibr})}(T) - \rho_{\mathrm{aver}}(0)] = \rho_{\mathrm{aver}}^{(\mathrm{combi})}(T) - \rho_{\mathrm{aver}}(0)$$

satisfied with accuracy better than 5 % (typically about 1 %).

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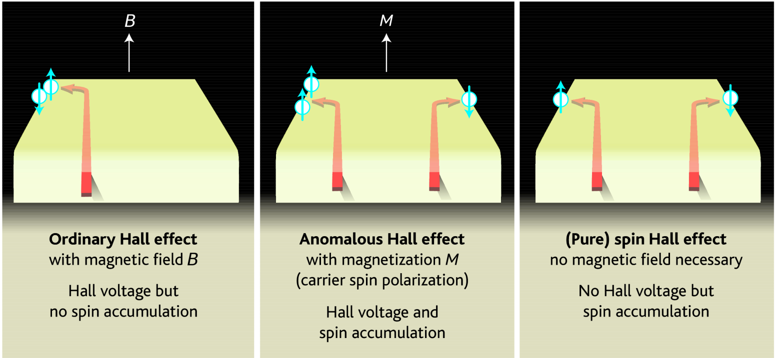
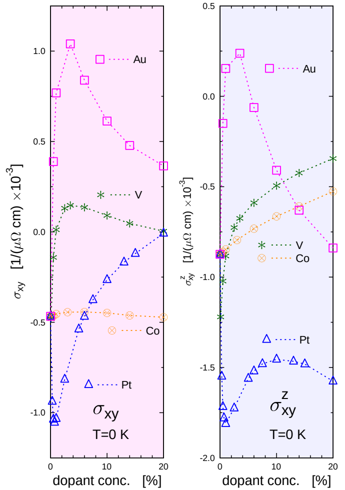
Offdiagonal conductivity



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

AHE **SHE**

Dependence of σ_{xy} and σ^z_{xy} on dopant concentration



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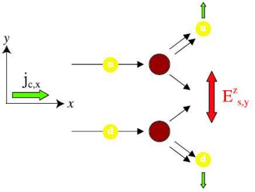
Anomalous Hall conductivity σ_{xy}.
Spin Hall conductivity σ^z_{xy}.

- Highly nonmonotonic dependence.
- Different dopants give rise to quite different dependencies.
- Sign can be reverted.
- σ_{xy} and σ^z_{xy} smoothly approach values for undoped Py (even though it does not look like it).

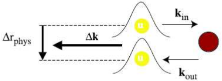
σ_{xy} and σ^z_{xy} diverge in the clean limit.

However, for Py host we are not in the clean limit even for zero dopant concentration.

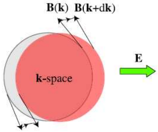
Useful intuitive concepts based on semiclassical approach



skew scattering



side jump



intrinsic

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

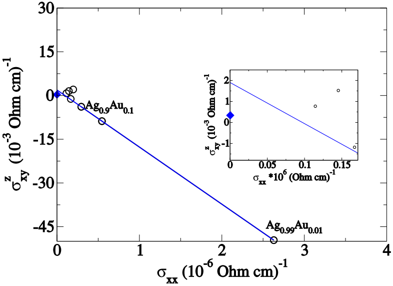
Dependence on the concentration of impurities c

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

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

Dilute limit

Au impurity in Ag crystal



Lowitzer, PhD thesis (2010)

In the dilute limit skew scattering dominates.

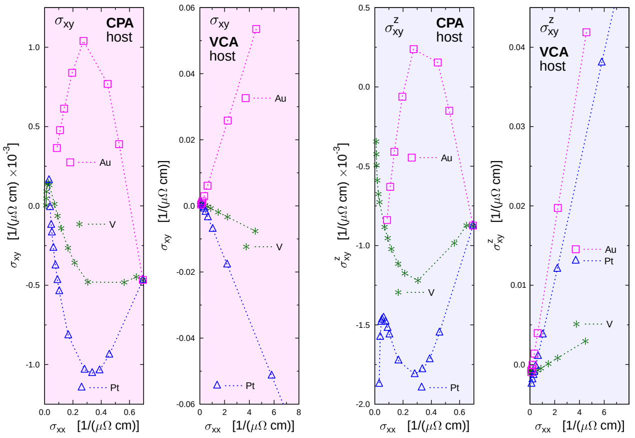
One can write [Nagaosa *et al.* RMP **82**, 1539 (2010)]

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

S is skewness.

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

Treating the host within CPA and within VCA



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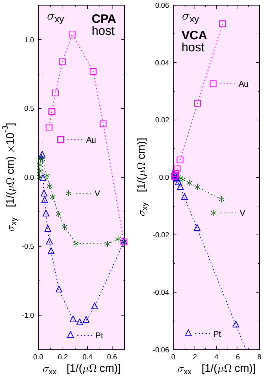
If the host is alloy, off-diagonal conductivities σ_{xy} and σ^{z}_{xy} are not proportional to the longitudinal conductivity σ_{xx} for low dopant concentrations (unlike for crystalline hosts).

Disorder in the host



If host is treated within VCA (i.e., as a crystal), σ_{xy} and σ^z_{xy} are proportional to σ_{xx} for low dopant concentrations.

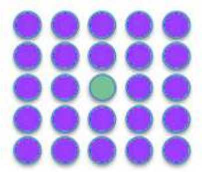
For disordered host, the dependence of AHE and SHE on the dopant concentration cannot be described in terms of skew scattering, side-jump scattering, or intrinsic contribution as for crystalline host.

This scheme, namely, assumes that for zero dopant concentration the electron participating in the transport is not scattered.

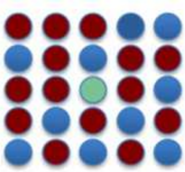
If the host material is an alloy, the concepts of skew scattering and side-jump scattering can be misleading.

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.

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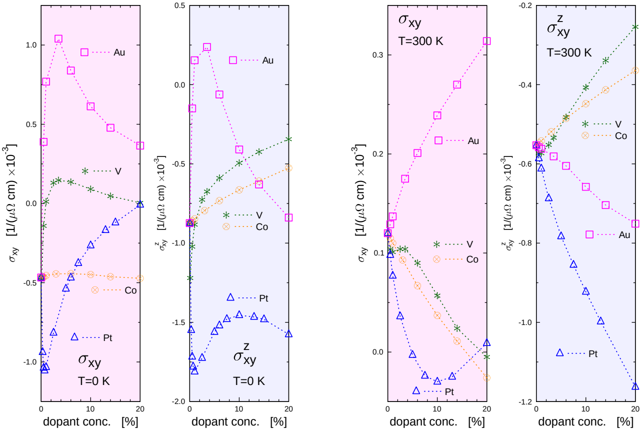
σ_{xy} and σ_{xy}^z for $T = 0$ K and $T = 300$ K



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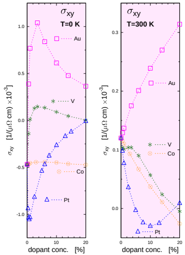
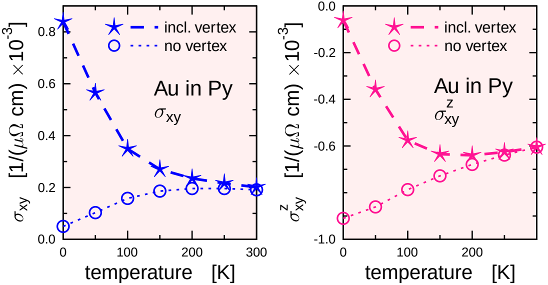
Dependence of σ_{xy} and σ_{xy}^z on the dopants concentration is quite different for T=0 K and for T=300 K.

Vertex corrections and temperature



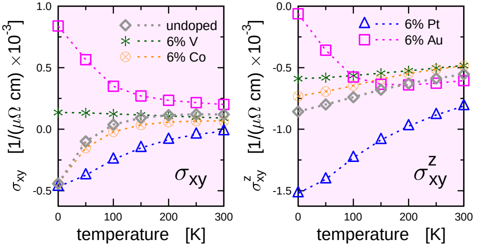


Vertex corrections are less important for high temperature.

For high temperature the electron undergoes many scattering events, there is more disorder, electron states loose their crystal-like character, the differences between various trajectories decrease.

All electrons undergo same scattering events in the end, albeit in a different sequence, and the vertex corrections become unimportant.

High-temperature limit

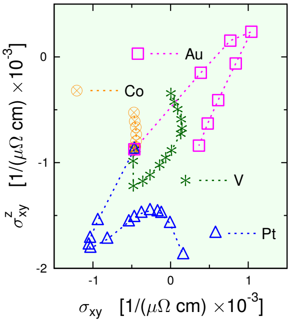


Šipr *et al.* PRB **101**, 085109 (2020)

If the temperature increases, the thermal effects dominate and, consequently, the differences between various dopants decrease.

Relation between AHE and SHE (1)

Both AHE and SHE are spin-dependent transport phenomena related to spin-orbit coupling.



Tsukahara *et al.* PRB **89**, 235317 (2014), Omori *et al.* PRB **99**, 014403 (2019):
skew scattering contributions to AHE and SHE conductivities are proportional,

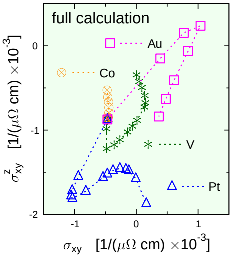
$$\sigma_{xy}^{\text{skew}} = p\, \sigma_{xy}^{z,\text{skew}} \,,$$

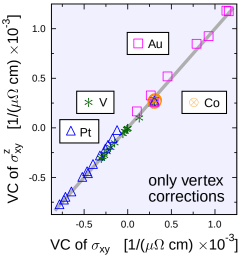
where p is the spin polarization of the current,

$$p = \frac{\sigma_{xx}^{(\text{maj})} - \sigma_{xx}^{(\text{min})}}{\sigma_{xx}^{(\text{maj})} + \sigma_{xx}^{(\text{min})}} \,.$$

Clearly, the linear relation does not hold for σ_{xy} and σ_{xy}^{z}.

Relation between AHE and SHE (2)





If one focuses just on the vertex corrections to σ_{xy} and σ^z_{xy}:

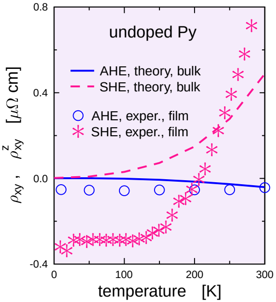
$$\sigma_{xy}(\mathrm{VC}) = \sigma^z_{xy}(\mathrm{VC}) .$$

Concept of skew scattering is of limited use when dealing with disordered hosts. However, vertex corrections (a.k.a. incoherent contributions) are well-defined.

For an ordered host, vertex corrections represent within our approach the skew scattering [Onoda *et al.* PRB **77**, 165103 (2008)].

What makes the AHE conductivity σ_{xy} and SHE conductivity σ^z_{xy} different is the coherent or intrinsic contribution.

Comparing theory and experiment for AHE and SHE



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

The calculations concern bulk Py while the experiment was done for a thin film.

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

Transport in doped Py: Conclusions

- The rate of the decrease of σ_{xx} on dopant concentration follows the sequence Co–Au–Pt–V, in accordance with scattering cross-sections at E_F.
- Dependence of σ_{xy} and σ_{xy}^z on the dopant concentration is non-monotonic and strongly depends on temperature.
- Having host an alloy instead of a crystal has profound influence on how 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.
- Vertex corrections to σ_{xy}^z are approximately equal to the vertex corrections to σ_{xy}.

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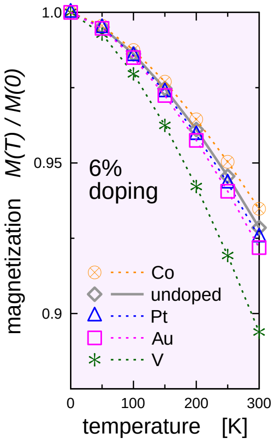
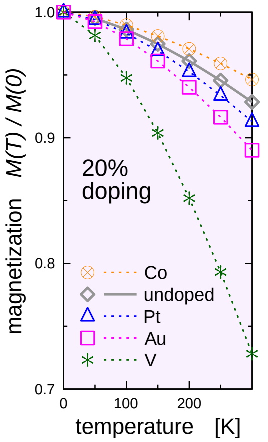


magnetization M(T) / M(0)
1.0
0.95
0.9
6%
doping
Co
undoped
Pt
Au
V
0
100
200
300
temperature [K]



magnetization M(T) / M(0)
1.0
0.9
0.8
0.7
20%
doping
Co
undoped
Pt
Au
V
0
100
200
temperature [K]