The spin Hall effect as a probe of non-linear spin fluctuations

D. H. Wei and Y. Niimi

Institute for Solid State Physics, University of Tokyo,

5-1-5 Kashiwa-no-ha, Kashiwa, Chiba 277-8581, Japan

B. Gu and S. Maekawa

Advanced Science Research Center,

Japan Atomic Energy Agency, Tokai 319-1195, Japan and

CREST, Japan Science and Technology Agency,

Sanbancho, Tokyo 102-0075, Japan

T. Ziman

Institut Laue Langevin, Boîte Postale 156,

F-38042 Grenoble Cedex 9, France and

Laboratoire de Physique et Modélisation des Milieux Condensés (UMR 5493),

Université Joseph Fourier and CNRS,

Maison des Magistères, BP 166, 38042 Grenoble, France

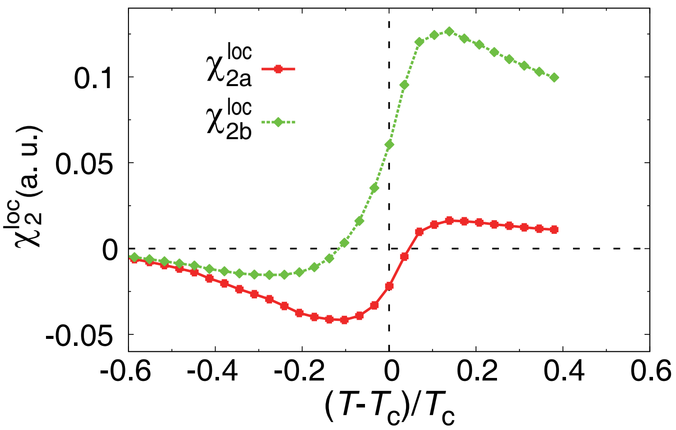
Y. Otani

Institute for Solid State Physics, University of Tokyo,

5-1-5 Kashiwa-no-ha, Kashiwa, Chiba 277-8581, Japan and

RIKEN-ASI, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

Supplementary Figures



Supplementary Figure S1: **Monte Carlo simulations**. We present Monte Carlo simulations of the fourth order spin correlations appearing in the expansion of $\chi_{2a}^{\rm loc}$ and $\chi_{2b}^{\rm loc}$ for nearest neighbors in a three-dimensional classical Heisenberg model. The correlations for further neighbors are similar in shape but successively smaller in amplitude. A small magnetic field is applied to stabilize the symmetry breaking and a lattice size of 20^3 sites is used. Further neighbor correlations have similar temperature dependence but decrease with increasing distance.

Supplementary Discussion

Magnetic susceptibilities are defined in the expansion of the total magnetization M_{tot} in terms of the uniform magnetic field H:

$$M_{\mathrm{tot}} = M_{\mathrm{tot},0}(T) + \chi_0^{\mathrm{uni}} H + \chi_1^{\mathrm{uni}} H^2 + \chi_2^{\mathrm{uni}} H^3 + \cdots , \tag{S1}$$

where χ_i^{uni} is the uniform magnetic susceptibility of the i-th order. In terms of cumulants we have

$$\chi_0^{\mathrm{uni}} = \beta \langle (M_{\mathrm{tot}} - \langle M_{\mathrm{tot}} \rangle)^2 \rangle \tag{S2}$$

$$\chi_1^{\mathrm{uni}} = \beta^2 \langle (M_{\mathrm{tot}} - \langle M_{\mathrm{tot}} \rangle)^3 \rangle \tag{S3}$$

$$\chi_2^{\mathrm{uni}} \approx \beta^3 \langle (M_{\mathrm{tot}} - \langle M_{\mathrm{tot}} \rangle)^4 \rangle \tag{S4}$$

where $\beta = 1/(k_{\mathrm{B}}T)$, k_{B} is the Boltzmann constant, and T is the temperature.

Based on the simple molecular field theory for an Ising spin $S = 1/2$, the total magnetization M_{tot} is given as [23]

$$\frac{M_{\mathrm{tot}}}{N\mu_{\mathrm{B}}} = \tanh\left(\frac{T_{\mathrm{C}} M_{\mathrm{tot}}}{T N \mu_{\mathrm{B}}} + \frac{\mu_{\mathrm{B}} H}{k_{\mathrm{B}} T}\right), \tag{S5}$$

where μ_{B} is the Bohr magneton. Just below T_{C} we can expand in powers of M

$$\chi_0^{\mathrm{uni}} \approx N\mu_{\mathrm{B}}^2 \left(\frac{1}{k_{\mathrm{B}}T_{\mathrm{C}}}\right)\left(\frac{1}{2}|\varepsilon|^{-1}\right), \tag{S6}$$

$$\chi_1^{\mathrm{uni}} \approx N\mu_{\mathrm{B}}^3 \left(\frac{1}{k_{\mathrm{B}}T_{\mathrm{C}}}\right)^2\left(-\frac{\sqrt{3}}{12}|\varepsilon|^{-\frac{5}{2}}\right), \tag{S7}$$

$$\chi_2^{\mathrm{uni}} \approx N\mu_{\mathrm{B}}^4 \left(\frac{1}{k_{\mathrm{B}}T_{\mathrm{C}}}\right)^3\left(\frac{5}{72}|\varepsilon|^{-4}\right), \tag{S8}$$

where $\varepsilon = (T - T_{\mathrm{C}})/T_{\mathrm{C}}$. Just above the Curie Temperature, $T \gtrsim T_{\mathrm{C}}$ we obtain

$$\chi_0^{\mathrm{uni}} \approx N\mu_{\mathrm{B}}^2 \left(\frac{1}{k_{\mathrm{B}}T_{\mathrm{C}}}\right)|\varepsilon|^{-1}, \tag{S9}$$

$$\chi_1^{\mathrm{uni}} = 0, \tag{S10}$$

$$\chi_2^{\mathrm{uni}} \approx N\mu_{\mathrm{B}}^4 \left(\frac{1}{k_{\mathrm{B}}T_{\mathrm{C}}}\right)^3\left(-\frac{1}{3}|\varepsilon|^{-4}\right). \tag{S11}$$

The divergent behavior near, but not right at, T_{C} in Fig 4b can be reproduced with Supplementary Equations (S8) and (S11) (to obtain χ_2^{uni} in Fig. 4b, one has to multiply by $-(k_{\mathrm{B}}T_{\mathrm{C}})^3$ as we shall explain later).

In order to explain the smearing of the divergence near T_C, we have performed Monte Carlo simulations. First we extend Kondo's theory [13] for the AHE in ferromagnetic metals by including the short-range spin-spin correlations since they can be dominant as we approach the critical point. The short-range spin-spin correlations, which are neglected in Kondo's theory, can dramatically change the behavior of the longitudinal resistance near T_C in ferromagnetic metal as discussed by Fisher and Langer [24].

Using the second Born approximation, but including the spin-spin correlations between different sites, the magnetic susceptibilities χ_i can be generalized to quantities we shall denote χ_i^{loc}. The superscript "loc" indicates that the magnetic susceptibilities depend not just on on-site correlations, but on all local correlations of spins. After integration over the Fermi surface only those up to a finite separation will contribute to the transport coefficients as below:

$$\chi_0^{\mathrm{loc}} = \sum_{n=1}^{N} e^{i(\mathbf{k}'-\mathbf{k})(\mathbf{R}_n-\mathbf{R}_1)} \langle (M_n - \langle M_n \rangle)(M_1 - \langle M_1 \rangle) \rangle, \tag{S12}$$

$$\chi_1^{\mathrm{loc}} = \sum_{n=1}^{N} e^{i(\mathbf{k}'-\mathbf{k})(\mathbf{R}_n-\mathbf{R}_1)} \langle (M_n - \langle M_n \rangle)^2 (M_1 - \langle M_1 \rangle) \rangle, \tag{S13}$$

$$\chi_{2a}^{\mathrm{loc}} = 2c_2 \sum_{n=1}^{N} e^{i(\mathbf{k}'-\mathbf{k})(\mathbf{R}_n-\mathbf{R}_1)} \langle (M_n - \langle M_n \rangle)^2 (M_1^2 - \langle M_1^2 \rangle) \rangle, \tag{S14}$$

$$\chi_{2b}^{\mathrm{loc}} = 2c_2 \sum_{n=1}^{N} e^{i(\mathbf{k}'-\mathbf{k})(\mathbf{R}_n-\mathbf{R}_1)} \langle (M_n - \langle M_n \rangle)(M_n^2 - \langle M_n^2 \rangle)(M_1 - \langle M_1 \rangle) \rangle. \tag{S15}$$

Here $\mathbf{k}$ and $\mathbf{k}'$ are wave vectors of the conduction electron, and $\mathbf{R}_n$ is the position of the n-th site. M_n is the magnetization of the localized electron of the n-th site, $\langle M_n \rangle$ is its average value thermally averaged. The subscript of M_1 is to emphasize that the expectation value is for the moment on a single site 1. c_2 is the material parameter, $c_2 = -1/2$ for Ni. As for χ_2^{loc}, there are two terms, i.e., χ_{2a}^{loc} and χ_{2b}^{loc}. The difference between the two terms originate from different pairings of linear and quadratic operators on the different sites, but preserving the total order of 4. In the strictly local theory of Kondo [13], only one site was involved, so there was just one term denoted r_2.

For zero momentum transfer $\mathbf{k} - \mathbf{k}' = 0$, χ_{2a}^{loc} in Supplementary Equation (S14) and χ_{2b}^{loc} in Supplementary Equation (S15) near T_C can be approximated as follows;

$$\chi_{2a}^{\mathrm{loc}} \approx \chi_{2b}^{\mathrm{loc}} \approx 2c_2 (k_\mathrm{B} T_\mathrm{C})^3 \chi_2^{\mathrm{uni}} \equiv \chi_2'. \tag{S16}$$

It is this quantity χ_2' that is plotted in Fig. 4b in the main text.

Note however that there is a finite momentum transfer $(\mathbf{k} - \mathbf{k}')$ in $\chi_{2a}^{\rm loc}$ and $\chi_{2b}^{\rm loc}$, which effectively reduces the summation over more than a few neighbors once they are integrated over. This should cut off any divergence near $T_{\rm C}$ in integrated quantities such as transport coefficients, as it does for the longitudinal resistance near $T_{\rm C}$ in ferromagnets, as discussed by Fisher and Langer [24]. In Figure S1, we present Monte Carlo simulations of the fourth order spin correlations appearing in the expansion of $\chi_{2a}^{\rm loc}$ and $\chi_{2b}^{\rm loc}$ for nearest neighbors in a three-dimensional classical Heisenberg model. It is apparent that this result is in contrast to the uniform nonlinear susceptibility $\chi_2^{\rm uni}$ in Fig. 4b which is summed to all distances and should diverge in the limit of zero field. However, we note that it is very difficult to fully reproduce the experimental results with our Monte Carlo simulations where we put the cut-off distance by hand.

Supplementary References

[23] T. Sato, and Y. Miyako, Nonlinear susceptibility and specific heat of $(Pd_{0.9966}Fe_{0.0034})Mn_{0.05}$., J. Phys. Soc. Jpn. **51**, 1394-1400 (1981).

[24] M. E. Fisher and J. S. Langer, Resistive anomalies at magnetic critical points., Phys. Rev. Lett. **20**, 665-668 (1968).