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Reply to: “Extracting Kondo temperature of strongly-correlated systems from the inverse local magnetic susceptibility”

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REPLYING TO A. A. Katanin *Nature Communications* https://doi.org/10.1038/s41467-021-21641-2 (2021).

In his comment[1], Katanin reanalyzes our LDA + DMFT results[2] for the temperature-dependent static local spin susceptibility of Sr_2RuO_4 and V_2O_3 fitting them to a Curie–Weiss (CW) form, $\chi(T) \simeq a/(T+\theta)$. Invoking Wilson's analysis[3] of the impurity susceptibility of the spin-½ one-channel Kondo model (1CKM) in the wide-band limit, he extracts spin Kondo temperatures using $T_K = \theta/\sqrt{2}$, obtaining $T_K = 350$ K and 100 K for Sr_2RuO_4 and V_2O_3, respectively. Noting that these are significantly smaller than the scales $T_{sp}^{onset} = 2300$ K and 1000 K reported in ref. [2], he argues that our T_{sp}^{onset} scales “do not characterize the screening process”.

We welcome Katanin's use of our data. However, his implication that our T_{sp}^{onset} was intended to fully characterize the screening process is misleading. Our work uses the full susceptibility vs. temperature curve to describe properly spin screening, not just a single number. Furthermore, our T_{sp}^{onset} was defined to characterize the high-temperature onset of spin screening, whereas his T_K characterizes the CW regime found at intermediate (i.e., lower) temperatures. The fact that T_K is much smaller than T_{sp}^{onset} is therefore not surprising but natural.

We agree with Katanin that, for Hund metals in general and Sr_2RuO_4 in particular, it is reasonable to approximate $\chi(T)$, using results of a Kondo impurity which features a CW law at intermediate temperatures. (In the Supplementary material we analyze $\chi(T)$ taken data from DMFT studies of the model Hund system used in ref. [2].) However, this was already well known. For Sr_2RuO_4, a comparison to the exact solution of a (fully screened) spin-1 Kondo model impurity model was carried out in the inset of Fig. 3a of ref. [4] (ref. 17 of ref. [2]), reproduced as Fig. 1(left) below, and a CW fit of that data was published in Fig. 2a of ref. [5] (cited as ref. 5 of ref. [2]). We reproduce it as Fig. 1(right) below. Since Sr_2RuO_4 and V_2O_3 have an atomic ground state configuration spin closer to 1 than ½, the use of a (fully screened) spin-1 Kondo model is more reasonable. Furthermore, when interpreting LDA+DMFT results, it is preferable to use definitions of the Kondo scale that rely on the low-temperature portion of the susceptibility curve, as was done in refs. [4,10], as opposed to the high-temperature portion as in Katanin's proposal to characterize spin screening. We elaborate on these points and propose a simple way to characterize spin crossovers of Hund metals below.

Since Katanin's comment invokes the 1CKM, we start by summarizing some of its well-established properties[3,6,7]. $\chi(T)$ exhibits a very broad crossover, from Curie-like high-temperature behavior governed by a local-moment fixed point describing a free spin, to Pauli-like low-temperature behavior governed by a Fermi-liquid fixed point describing a fully screened spin. A proper description of this crossover requires a crossover scaling function, $F(T/T_K)$ and a crossover scale, the Kondo scale T_K, with $\chi(T) = F(T/T_K)/T$. Wilson showed that $F(x)$ is universal under the assumptions of very weak impurity-bath coupling and infinite bandwidth, and computed it numerically. There are multiple ways of defining T_K, evoking the behavior of $F(x)$ for either $x \gg 1$, $x \simeq 1$, or $x \ll 1$, yielding T_K values differing only by factors of order unity. Wilson's definition of T_K (adopted by Katanin), denoted T_W here, evokes the $x \gg 1$ limit. For high temperatures, $T \gtrsim 16T_W$, he found $\chi(T) \simeq 1/(4T)\left[1 - 1/\ln(T/T_W) + O\left(1/\ln^3(T/T_W)\right)\right]$, with T_W defined such that the coefficient of $1/\ln^2(T/T_W)$ vanishes. For intermediate temperatures, $0.5T_W < T < 16T_W$, his numerical results are well approximated by a CW form, with $a = 0.17$ and $\theta \sim \sqrt{2}T_W$[3,6] (as used by Katanin). At zero temperature, Wilson found $\chi(0) \sim 0.103/T_W$ (Eq. (IX.91) of ref. [3]). Subsequent Bethe-Ansatz (BA) calculations of the scaling function[6,7] matched Wilson's numerical results. Analogous results have been obtained for fully screened Kondo models with higher spins[8,9].The BA works showed that the curve $\chi(T)$ vs. T/T_K depends on the spin S, with $\chi(T) \propto S(S+1)]/(3T)$ for $T/T_K \gg 1$ and $\chi(T) \propto S$ for $T/T_K \ll 1$. The Kondo scales defined in these BA works are

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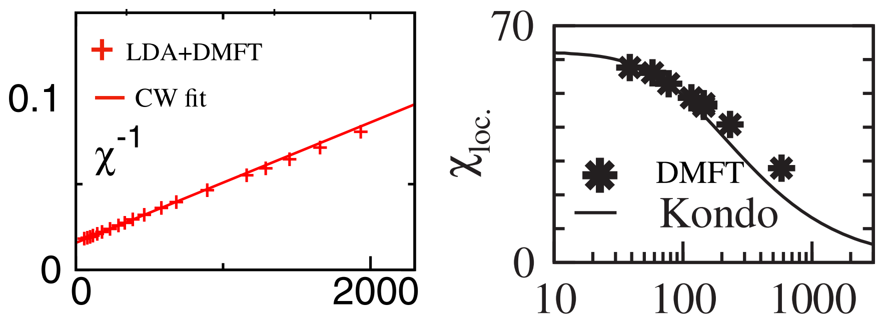

Fig. 1 Earlier work comparing Kondo impurity model with LDA+DMFT results for Sr_2RuO_4. Left: $1/\chi(T)$ versus T, with LDA+DMFT results for Sr_2RuO_4 (red symbols) and a Curie–Weiss fit (straight red line) reproduced from the inset of Fig. 2a of ref. [5]. Right: Bethe-Ansatz results for the spin-1,2-channel Kondo model $\chi(T)$ vs. T with $T_{BA} = 240$ K (solid line) in good agreement with the LDA + DMFT results for Sr_2RuO_4 (black symbols) reproduced from inset of Fig. 3a of ref. [4].

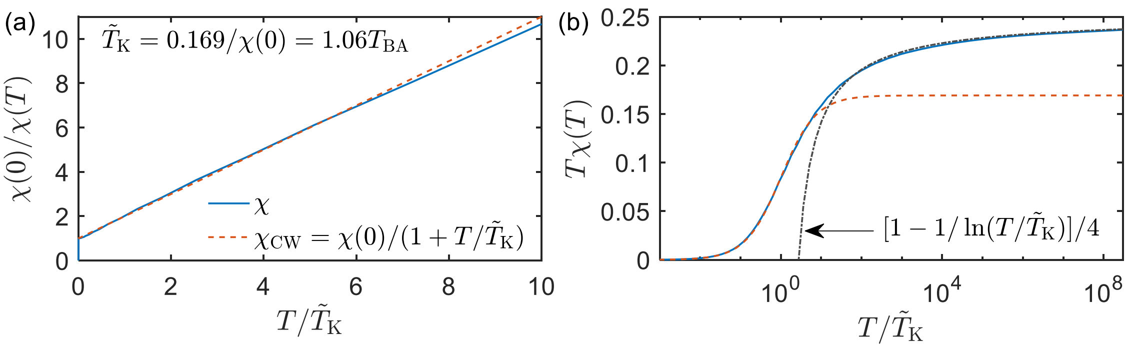

Fig. 2 Two representations of the impurity susceptibility $\chi(T)$ as defined by Wilson[3] (blue lines), for the 1CKM in the wide-band limit, computed using the numerical renormalization group (NRG). a The Curie–Weiss form (red dashed line) works reasonably well for intermediate temperatures, but (**b**) not at all for large temperatures, $T/\tilde{T}_K \gg 1$, where logarithmic corrections are large (black dash-dotted line).

independent of spin as in Eq. (21) of ref. [9]: $T_{BA} = S/[\pi\chi(0)]$, with $T_{BA}/T_W = 1.55$ for $S = 1/2$.

In ref. [2], we used a strategy similar to Wilson's: we identified the regions where the behavior of $\chi_{spin}(T)$ and $\chi_{orb}(T)$ is governed by atomic physics or Fermi-liquid theory and numerically computed the crossover function bridging them. We defined two scales for the onset and completion of spin screening, T^{onset}_{sp} and T^{cmp}_{sp} as the temperatures above or below which $\chi_{spin}(T)$ shows pure Curie behavior ($\sim 1/T$) or pure Pauli behavior ($\sim$ const), respectively, and similarly T^{onset}_{orb} and T^{cmp}_{orb} for orbital screening. Our T^{onset}_{sp} and T^{cmp}_{sp} scales are similar in spirit to Wilson's $16T_W$ and $0.5T_W$. So even within the 1CKM framework, an extraction of T_W from our results, using $T_W \simeq T^{onset}_{sp}/16$, would yield $2300\,\mathrm{K}/16 \simeq 140$ K for Sr_2RuO_4 and $1000\,\mathrm{K}/16 \simeq 60$ K, and the order of magnitude discrepancy claimed by Katanin disappears.

Contrary to this crude estimate, in ref. [2] we did not assume T^{onset}_{sp} to be proportional to a single Kondo scale since even for an impurity model without DMFT self-consistency, T^{onset}_{sp} is known to be affected by energy scales not present in the wide-band 1CKM (e.g., a finite bandwidth or a finite charging energy), since such scales cut off high-temperature logarithmic corrections [cf. ref. [10], Fig. 2b, c]. This is even more important for Mott systems, where the emergence of a quasi-particle resonance with decreasing temperatures affects the bath bandwidth via DMFT self-consistency.

In ref. [2], we supplemented our LDA+DMFT study of actual materials by DMFT studies of a multi-orbital model Hamiltonian, again computing $\chi(T)$ numerically. We found signatures distinguishing Mottness and Hundness (such as $T^{onset}_{spin} \simeq T^{onset}_{orb}$ for the former but $T^{onset}_{spin} < T^{onset}_{orb}$ for the latter) similar to those found in the materials. We defined a Kondo scale $T^{dyn}_{K,spin}$ (denoted T_K in ref. [2]) through the imaginary part of the $T = 0$ dynamical spin susceptibility, $\chi''\left(\omega = T^{dyn}_{K,spin}\right) = \text{maximal}$. $T^{dyn}_{K,spin}$ characterizes the intermediate region, with $T^{cmp}_{spin} < T^{dyn}_{K,spin} < T^{onset}_{spin}$. It is shown as a red line in Fig. 5b of ref. [2], yielding $T^{dyn}_{K,spin} = 0.12t = 600$ K for our Hund system H1 mimicking Sr_2RuO_4, and $T^{dyn}_{K,spin} = 0.04t = 200$ K for our Mott system M1 mimicking V_2O_3 (using the conversion factor $t = 5000$ K stated in Fig. 1).

We take Katanin's comment as an incentive to propose a standardized scheme for extracting a Kondo scale, $\tilde{T}_K$, from a computed $\chi(T)$ curve. Our scheme (i) does not involve a fit to predictions of a specific impurity model, since in general it is unclear which impurity model to compare to, and (ii) uses the $x \le 1$ part of the crossover scaling function, since it is more universal than the $x \gg 1$ part[8–10]; and (iii) reduces to impurity-model results when these are applicable. We propose to define $\tilde{T}_K$ through the relation $\chi(\tilde{T}_K)/\chi(0) = 1/2$. (If $\chi(0)$ is not known but $\chi(T)$ shows CW-type behavior at intermediate temperatures, $\chi(0)$ can be estimated by linear extrapolation of $1/\chi(T)$ vs. T to zero temperature.) This definition ensures that $T^{comp}_{sp} < \tilde{T}_K < T^{onset}_{sp}$, as it should. For the CW form it yields $\tilde{T}_K = \theta$. For the 1CKM, NRG computations (Fig. 2) show that $\tilde{T}_K = 0.169/\chi(0) = 1.06T_{BA} = 1.64T_W$. For the materials Sr_2RuO_4 and V_2O_3 studied in ref. [2], Katanin's CW extraction of θ-values implies $\tilde{T}_K = 574$ K or 164 K, respectively. This illustrates, yet again, the main point of this reply: the Kondo scale is generically much smaller than T^{onset}_{sp}, and it is misleading to conflate these two scales.

Data availability

The authors declare that the data supporting the findings of this study are available from the authors.

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Author contributions

All the authors X.D., K.M., K.H., S.S.B.L., A.W., J.v.D., and G.K. discussed the comment and participated in the drafting of the response.

Competing interests

The authors declare no competing interests.

Additional information

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