Generic Seebeck effect from spin entropy

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Graphical abstract

Public summary
- Magnetism can offer a significant contribution to thermoelectricity
- A generic Seebeck effect exists in magnetic conductors as a result of transport spin entropy of delocalized d electrons
- The magnetocaloric effect and the Seebeck effect are thermodynamically correlated with each other
Generic Seebeck effect from spin entropy

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1D). Note that the Peltier effect, i.e., the reverse phenomenon of the Seebeck effect, is demonstrated in Figures 1A, 1B, and 1D because it provides a better comparison with the magnetocaloric effect given their common features in solid-state cooling.

INTRODUCTION

The Seebeck coefficient (\( \alpha \)) is a key parameter determining the efficiency of useful thermoelectric devices. It measures temperature-difference-induced electric voltage in a conducting solid, being essentially a non-equilibrium thermal transport phenomenon. Nevertheless, in special circumstances this effect can be well described thermodynamically by employing thermodynamic state variables. One such example is the scaling at low-temperature limit, where electron diffusive transport is suppressed, between \( \alpha \) and the electronic specific heat \( C_{el} \)—a fundamental thermodynamic property—in a wide range of materials spanning from simple to correlated metals.1 Reflecting electron transport kinetics, diffusive characteristics such as the energy-dependent charge mobility can generate a large Seebeck effect too,2 as was also revealed by charge-scattering engineered thermoelectricity in nanoscaled materials.3

Recent years have witnessed increasing efforts to pursue large values of \( \alpha \) in magnetic materials.4–6 A general understanding gained so far is that a spin-dependent Seebeck effect (SdSE) might be a material-specific property related to magnon drag, spin fluctuation, or spin-dependent scattering of conduction electrons. No generic SdSE has been known. Here, we demonstrate for a large number of \( d \)-electron-based magnetic conductors with ferromagnetic (FM), weakly ferromagnetic (WF), or antiferromagnetic (AF) transition that a sizable SdSE takes place over a wide temperature range from slightly below to well above the ordering temperature. This additional Seebeck contribution to the Seebeck effect over an extended temperature range from slightly below to well above the magnetic transition temperature. This contribution is interpreted as resulting from transport spin entropy of (partially) delocalized conducting \( d \) electrons with strong thermal spin fluctuations, even semiquantitatively in a single-band case, in addition to the conventional diffusion part arising from their kinetic degrees of freedom. As a highly generic effect, the spin-dependent Seebeck effect might pave a feasible way toward efficient "magnetic thermoelectrics."

Keywords: Seebeck effect; magnetocaloric effect; spin entropy; thermoelectric material

How magnetism affects the Seebeck effect is an important issue of wide concern in the thermoelectric community but remains elusive. Based on a thermodynamic analysis of spin degrees of freedom on varied \( d \)-electron-based ferromagnets and antiferromagnets, we demonstrate that in itinerant or partially itinerant magnetic compounds there exists a generic spin contribution to the Seebeck effect over an extended temperature range from slightly below to well above the magnetic transition temperature. This contribution is interpreted as resulting from transport spin entropy of (partially) delocalized conducting \( d \) electrons with strong thermal spin fluctuations, even semiquantitatively in a single-band case, in addition to the conventional diffusion part arising from their kinetic degrees of freedom. As a highly generic effect, the spin-dependent Seebeck effect might pave a feasible way toward efficient “magnetic thermoelectrics.”

Thermodynamic considerations

In a conducting solid exposed to a temperature difference \( dT \), both chemical \( (\mu) \) and electrical potential \( \psi \) change thermodynamically at the two ends. In a standard setting of thermoelectric measurements, the \( dT \)-induced voltage measures the difference of electrochemical potential \( \mu = \mu + \psi \). Approximately, the Seebeck effect in response to \( dT \) arises in two parts:5,6

\[
\frac{1}{\alpha} = \frac{d\psi}{dT} + \frac{d\mu}{dT}.
\]  

(Equation 1)

Here \( e \) is the free electron charge. The first term is purely thermodynamic and is known as the Kelvin formula.5 The second includes kinetic information arising from charge relaxation processes. It is sometimes referred to as theoretical or effective Seebeck effect,7 as compared with the experimental one detecting both.

From thermodynamic consideration of an electronic system, the chemical potential \( \mu \) is related to the derivative of the total electronic entropy \( S \) with respect to the carrier number \( N \) of the system,\(^6\)

\[
\frac{d\mu}{dT} = - \alpha \frac{dS}{dT}.
\]  

(Equation 2)

Equations 1 and 2 show that \( \alpha \) probes entropy \( S \) per charge carrier as long as the details of electron kinetics can be ignored or is of minor importance. In magnetic conductors, the relevant spin entropy \( S_m \) is of interest and \( S_m = k_B \ln g \), with \( k_B \) being the Boltzmann constant and \( g \) the total number of spin configurations. Hence, the well-known Helike formula,\(^8\)

\[
\alpha_m = \frac{- (k_B e \psi \ln g)}{e^2/k_B T}
\]

can naturally be obtained. This is applicable to systems of interacting localized electrons and valid at high enough temperatures where all the spin degrees of freedom are active.

Next we consider the spin entropy of an FM compound on a more general basis within the mean-field approximation,\(^11\) whereas AFM state can be represented by two antiferromagnetically coupled FM sublattices:

\[
S_m(T, H) = R \ln \left[ \frac{\sinh (JX/2k_B T)}{\sinh (X)} \right] - X B_J(X).
\]  

(Equation 3)

Here \( R \) is the molar gas constant, \( B_J(X) \) the Brillouin function with \( X = g_\mu B H_{eff}/k_B T, J \) the total angular momentum, \( \mu_B \) the Bohr magneton, and \( g \) the Lande factor. The effective field \( H_{eff} \) reads

\[
H_{eff} = H + \frac{3k_B T c B_J(X)}{\mu_B g L(J + 1)}.
\]  

(Equation 4)

where the first and second terms at the right-hand side represent external and molecular field, respectively. Equations 3 and 4 are
frequently employed to estimate the magnetocaloric effect near a magnetic transition.\textsuperscript{11}

**RESULTS AND DISCUSSION**

The results of mean-field consideration are summarized in Figures 2A–2D. Figure 2A shows the magnetization per electron \( m = g_\Sigma J_{\text{HF}}(X) \) as a function of \( T \) for zero and a finite magnetic field \( h = 0.2 \). Figure 2B displays their corresponding \( S_m(T) \). It saturates to \( \text{Rdn} = 5.76 \text{J/mol K} \), the magnetic entropy associated with the ground-state doublet, immediately at \( T_C \) for \( h = 0 \), whereas the saturation trend is slowed down in finite field. The spin-entropy difference \( \Delta S_m \) (dashed line) is the magnetocaloric effect and assumes a peak at \( T_C \). Figure 2C shows the magnetic specific heat, which usually is the directly measured quantity for estimating \( S_m \). Figure 2D the isothermal field suppression of \( S_m \) at selected temperatures. As depicted by Equation 2, the step-like profile of \( S_m(T) \) (Figure 2B) characteristic of the magnetic ordering indicates a step-like change of \( \alpha(T) \) at \( T_C \) if the magnetic electrons are itinerant or partially itinerant, and \( \alpha_m(H) \) at constant temperature will decrease analogous to \( S_m \) (see Figure 2D). The additional contribution \( \alpha_{\text{mag}} \) to the Seebeck effect marks the difference of the Peltier cooling shown in Figures 1A and 1B.

To substantiate the spin-entropy contribution to \( \alpha(T) \) in Figure 3 we compile currently available literature data of various \( d \)-electron-based magnets, which are either FM, WFM, or AFM (see also Table 1). A generic, step-like increase of \( \alpha(T) \) at \( T = T_C \) or \( T_\text{S} \) is observed for all of them. To avoid complex transport properties caused by spin-density-wave gap in the AFM cases, sample choice is made for those with a simple resistivity drop below \( T_\text{S} \). Application of a magnetic field suppresses the spin-entropy \( \alpha(T) \) at constant temperature reduced to well below \( T_\text{S} \). This hints at a thermodynamic relationship between the two quantities. In-depth investigations into the 5d5SSE of WFM compounds \( \text{Fe}_2\text{V}_{1-x}\text{Cr}_x\text{Al}_1, \text{Sb}_2 \) (Tsujii et al.\textsuperscript{5}) and \( \text{CaFe}_4\text{Sb}_{12} \) (Takabatake et al.\textsuperscript{18}), whose cooperative magnetism has been known to be due to itinerant 3d bands, have shown that applying a magnetic field can suppress \( \alpha(T) \), evidencing the involvement of strong spin fluctuations. The spin-entropy description can readily capture the saturation of \( \alpha(T) \) above \( T_C/T_\text{S} \) because
The electronic structure of this compound is determined by the combined effect of an exchange splitting of Mn-3d electrons to the Seebeck effect by transferring spin entropy in addition to their kinetic one (see Figure 1B). To verify this proposition, field-tuned $\alpha_m(T)$ will be discussed semiquantitatively in terms of the spin entropy $S_m(T)$ (Figure 2D) by focusing on MnSi (Figures 4A and 4B). By contrast, the steplike $\alpha_m(T)$ in Li-doped MnTe has recently been interpreted based on local-moment picture and paramagnon-electron drag.6 The difficulties arising in the local-moment explanation will be explained below. Here, we note that the Seebeck effect of var-

Figure 3. Temperature-dependent Seebeck effect of varied d-electron-based FM, WFM, and AFM conductors For all these magnets, a steplike magnetic contribution $\alpha_m(T)$ emerges on top of a sublinear background $\alpha_S(T)$ (dashed line) that is derived from conventional charge-diffusion effect. The sublinear background is drawn for only a part of the samples for the sake of clarity. Note that $\alpha_m(T)$ and $\alpha_S(T)$ have same signs in the materials shown in (A) and opposite signs in (B) (see text). The vertical arrows mark the positions of $T_C$ or $T_M$ (for WFM compounds, the position of the maximum in magnetic susceptibility).

Table 1. Characterization of the steplike SdSE in some typical d-electron-based FM, WFM, and AFM materials

| Materials | Magnetic ordering | $T_C/T_N$ (K) | $\alpha_{step}$ (uVK) | $M_D$ (µB) | Refs. |
|-----------|------------------|---------------|---------------------|------------|-------|
| Co$_2$TiSn | FM               | 355           | $-40$              | 1.97       | Balke et al.12 Barth et al.13 |
| MnSi      | FM               | 29            | 8                   | 0.45       | Lamago et al.14 Hirokane et al.15 |
| Ni        | FM               | 627           | 10                  | $-0.6$     | Tang et al.16 Abadila et al.17 |
| CaFe$_2$Sb$_{12}$ | WFM | 50            | $-15$              | 0.5        | Takabatake et al.18 Schneitl et al.19 |
| Fe$_{0.5}$V$_{0.5}$Co$_{1.1}$ | WFM | 160           | 7                   | 0.4        | Tsujii et al.6 |
| MnTe (5.3% Li) | AFM | 307           | 150                 | 4.55       | Zheng et al.5 |
| YbMn$_2$Sb$_2$ | AFM | 120           | $-15$              | 3.6        | Nikiforov et al.20 Morozkin et al.21 |

Among these, Co$_2$TiSn and CaFe$_2$Sb$_{12}$ are representatives of a large group of magnetic Heusler and filled skutterudite compounds, respectively, with similar $\alpha(T)$ profiles, see Balke et al.12 and Takabatake et al.18.

*While a clear indication of ferromagnetic ordering is absent, CaFe$_2$Sb$_{12}$ shows a shoulder at $T = 50$ K in magnetic susceptibility due to incipient ferromagnetism.19

In YbMn$_2$Sb$_2$, magnetic moment originates from Mn d bands and divalent Yb ion is nonmagnetic.

While not included in Figure 3, a prominent case where spin entropy has been invoked to interpret the SdSE is NaCo$_2$O$_4$ (Terasaki et al.27), a paramagnet with Curie-Weiss behavior. Field-induced reduction of $\alpha(H)$ scaling to $H/T$, a feature derivable from Equation 3 assuming $T_C = 0$, has been considered evidence of spin-entropy contribution.28 This argument was rationalized in terms of hopping conduction of local 3d electrons.29 Electronic-structure calculation performed later on found that this compound locates at an itinerant magnetism instability with large electronic density of states derived from 3d orbitals.30 Within the itinerant picture, the $\alpha(H)$ profile of NaCo$_2$O$_4$ is also approachable by considering magnetic-field tuning to spin polarization at the Fermi level.31 Here, spin entropy is involved in shaping the electronic structure, as concluded from recent angle-resolved photoemission spectroscopy experiments32 in this sense, NaCo$_2$O$_4$ appears to be a special case of considerable d-electron-derived SdSE with a vanishing $T_C$. To quantitatively verify the correlation between spin entropy and SdSE, below we scrutinize $\alpha(T, H)$ of MnSi. MnSi is a prototype of itinerant ferromagnet with long-wavelength helical modulation of spin structure. Applying magnetic field reduces the spin entropy near $T_C$, as quantified by the static magnetocaloric effect $\Delta S_m$ (see Figure 2B and Arora et al.24). Consequently, the transport spin entropy detected by $\alpha_m$ is expected to diminish as well (Equation 2 and Figure 1A). Correspondence between the two quantities obtained experimentally is demonstrated in Figure 4A: $\Delta S_m$ (left axis) reveals a negative peak close to $T_C \approx 29$ K; see also Figure 4B for the measured $\alpha(T)$ at $H = 0$ and 5 T (Hirokane et al.15), from which $\Delta S_m$ is obtained as their difference. An apparent scaling between $\Delta S_m$ and $\Delta S_m$ ($H = 5$ T, right axis, Arora et al.24) can be observed. Hall-effect measurements reveal a simple one-band Hall resistivity with carrier concentration $\approx 0.99$ d-hole/MnSi, along with an anomalous contribution derived from ferromagnetism.7 On this basis, $\Delta S_m$ reported in unit of $\mu$K/kg C can be readily converted in accordance to the unit of $\alpha(T)\mu$V/K, by normalizing by the carrier density of d holes. The hence obtained spin-entropy-derived Seebeck effect, denoted as $\Delta S_{MCE}(T)$ (blue empty squares in Figure 4A), reveals a reasonable agreement with the measured $\Delta S_{MCE}(T)$ (blue solid squares in Figure 4A) within a factor of 2. This yields compelling evidence that the thermodynamics of delocalized d-electron spin degrees of freedom contribute substantially to the SdSE near and above the magnetic ordering temperature. The SdSE herein identified is generic to magnetic materials that are FM, WFM, AFM, or even paramagnetic near a magnetic instability with at least partially itinerant magnetism. It is a thermodynamic consequence originating from the spin degrees of freedom pertinent to magnetic d electrons. A large number of candidate compounds with significant SdSE can be found in the...
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magnon-drag effect appears only well below paramagnetic state. One is the sign problem as discussed for Ni: here, the transport and static spin entropy may generally to be negative due to the conduction electrons. Unlike MnSi, the transport and static spin entropy may generally to be negative due to the conduction electrons. 36,37

enhancement recently found in some magnetic ion-doped semiconductors, for example, Fe3GeTe2 (May et al.33), the mother compounds of iron-based superconductors14 and various manganese oxides. 35 Thermoelectricity derived Seebeck effect, we focus on some representative d-electron-based magnets to spin entropy of (partially) delocalized d electrons with strong thermal spin fluctuations. The fundamental correlation between the Seebeck effect and spin entropy appears highly instructive for future exploration of useful magnetic thermoelectrics. Furthermore, identification of the physical origin of the SdSE also helps us to understand the nature of collective magnetism in d-electron-based systems, which is often not straightforward because of the localized-itinerant duality and its influence on transport properties.

**Conclusion**

In conclusion, based on a thermodynamic analysis we have attributed the excess SdSE near and above T_C/T_N in a wide range of d-electron-based magnets to spin entropy of (partially) delocalized d electrons with strong thermal spin fluctuations. The fundamental correlation between the Seebeck effect and spin entropy appears highly instructive for future exploration of useful magnetic thermoelectrics. Furthermore, identification of the physical origin of the SdSE also helps us to understand the nature of collective magnetism in d-electron-based systems, which is often not straightforward because of the localized-itinerant duality and its influence on transport properties.

**MATERIALS AND METHODS**

The calculations of magnetization, spin entropy, and specific heat shown in Figures 2A–2D are based on the mean-field approximation assuming the Brillouin function description of the spin system. To better verify our proposal on the spin-entropy-derived Seebeck effect, we focus on some representative d-electron-based ferromagnetic, weakly ferromagnetic, and antiferromagnetic conductors whereby a nearly T-linear background, i.e., the conventional diffusion contribution to the Seebeck effect in metal, can be confirmed. These materials show at least partially delocalized features in their collective magnetism and have been experimentally investigated previously regarding the Seebeck effect. All experimental data on the Seebeck effect can be found in literature.

**REFERENCES**

1. Behnia, K., Jaccard, D., and Flouquet, J. (2004). On the thermoelectricity of correlated electrons in the zero-temperature limit. J. Phys. Condens. Matter 16, 5187.
2. Sun, P., Wei, B., Zhang, J., et al. (2015). Large Seebeck effect by charge-mobility engineering. Nat. Commun. 6, 7475.
3. Martin, J., Wang, L., Chen, L., and Nolas, G.S. (2009). Enhanced Seebeck coefficient through energy-barrier scattering in PtTe nanocomposites. Phys. Rev. B 79, 115311.
4. Zhao, W., Liu, Z., Sun, Z., et al. (2017). Superparamagnetic enhancement of thermoelectric performance. Nature 549, 247.
5. Tsuji, N., Nishide, A., Hayakawa, J., and Mori, T. (2019). Observation of enhanced thermopower due to spin fluctuation in weak itinerant ferromagnet. Sci. Adv. 5, eaat5935.
6. Zheng, Y., Lu, T., Polash, M.D.M., et al. (2019). Paramagnon drag in high thermoelectric figure of merit Li-doped MnTe. Sci. Adv. 5, eaat9461.
7. Cai, J., and Mahan, G.D. (2006). Effective Seebeck coefficient for semiconductors. Phys. Rev. B 74, 075201.
8. Apritz, Y., Ouerdane, H., Goupil, C., and Lecoeur, Ph. (2016). A note on the electrochemical nature of the thermoelectric power. Eur. Phys. J. Plus 131, 76.
9. Peterson, M.R., and Shastri, B.S. (2010). Kelvin formula for thermopower. Phys. Rev. B 82, 195105.
10. Chakkin, P.M., and Beni, G. (1976). Thermopower in the correlated hopping regime. Phys. Rev. B 13, 647.
The Innovation

11. Tishin, A.M. (1990). Magnetocaloric effect in strong magnetic fields. Cryogenics 30, 127.

12. Balke, B., Guardi, S., Graf, T., et al. (2010). Seebeck coefficients of half-metallic ferromagnets. Solid State Commun. 150, 529.

13. Barth, J., Fecher, G.H., Balke, B., et al. (2010). Itinerant half-metallic ferromagnets Co2TiZ (Z=Si, Ge, Sn): ab initio calculations and measurement of the electronic structure and transport properties. Phys. Rev. B 81, 064404.

14. Lamago, D., Georgii, R., and Boni, P. (2003). Magnetic susceptibility and specific heat of the itinerant ferromagnet MnSi. Phys. B 359-361, 1171.

15. Hirokane, Y., Tomioka, Y., Imai, Y., et al. (2016). Longitudinal and transverse thermoelectric transport in MnSi. Phys. Rev. B 93, 014436.

16. Tang, S.H., Craig, P.P., and Kitchens, T.A. (1971). Seebeck coefficient at the Curie temperature: specific heat of charge carriers in ferromagnets. Phys. Rev. Lett. 27, 593.

17. Abadilla, L., Gasser, F., Khalouk, K., et al. (2014). New experimental methodology, setup and LabView program for accurate absolute thermoelectric power and electrical resistivity measurements between 25 and 1600 K. Application to pure copper, platinum, tungsten, and nickel at very high temperatures. Rev. Sci. Instrum. 85, 095121.

18. Takabatake, T., Matsuoka, E., Narazu, S., et al. (2006). Roles of spin fluctuations and rattling in magnetic and thermoelectric properties of AT4Sb12 (A = Ca, Sr, Ba, La; T = Fe, Ru, Os). Phys. B 383, 93.

19. Schnelle, W., Leithe-Jasper, A., Schmidt, M., et al. (2005). Itinerant iron magnetism in filed skutterudites CaFe6Sb12 and YbFe6Sb12. stable divalent state of ytterbium. Phys. Rev. B 72, 020402(R).

20. Nikiforov, V.N., Piyadun, V.V., Morozkin, A.V., and Irkhin, V.Yu. (2014). Anomalies of transport properties in antiferromagnetic YbMn2Sb2 compound. Phys. Lett. A 378, 1488.

21. Masek, J., Velicky, B., and Janis, V. (1987). A tight binding study of the electronic structure of MnTe. J. Phys. C: Solid State Phys. 20, 34.

22. May, A.F., Calder, S., Cantoni, C., et al. (2016). Magnetic structure and phase stability of the van der Waals bonded ferromagnet Fe52GeTe5. Phys. Rev. B 93, 014411.

23. Pallecchi, I., Caglieris, F., and Putti, M. (2016). Thermoelectric properties of iron-based superconductors and parent compounds. Supercond. Sci. Technol. 29, 073002.

24. Asamitsu, A., Morimoto, Y., and Tokura, Y. (1996). Thermoelectric effect in La1-x SrMnO3. Phys. Rev. B 53, R2952.

25. Aharya, S., Anwar, S., Mori, T., and Soni, A. (2018). Coupling of charge carriers with magnetic entropy for power factor enhancement in Mn doped Sn0.3Te for thermoelectric applications. J. Mater. Chem. C 6, 6489.

26. Watzman, S.J., Duine, R.A., Tserkovnyak, Y., et al. (2016). Magnon-drag thermopower and Nernst coefficient in Fe, Co, and Ni. Phys. Rev. B 94, 144407.

27. Geishendorf, K., Vir, P., Shekhar, C., et al. (2020). Signatures of the magnetic entropy in the thermopower signals in nanoribbons of the magnetic Weyl semimetal Co2Sn2S2. Nano Lett. 20, 300.

28. Allen, J.W., Lucovsky, A., and Mikkelsen, J.C., Jr. (1977). Optical properties and electronic structure of crossroads material MnTe. Solid State Commun. 24, 367.

29. Okabe, T. (1994). Itinerant ferromagnetism in nickel. J. Phys. Soc. Jpn. 63, 4155.

30. Masek, J., Velicky, B., and Janis, V. (1987). A tight binding study of the electronic structure of MnTe. J. Phys. C: Solid State Phys. 20, 34.

31. Wang, Y.Y., Rogado, N.S., Cava, R.J., and Ong, N.P. (2003). Spin entropy as the likely source of enhanced thermopower in NaCo2O4. Nature 423, 425.

32. Chen, S.D. (2017). Large thermopower from dressed quasiparticles in the layered cobaltates and rhodates. Phys. Rev. B 96, 081109(R).

33. May, A.F., Calder, S., Cantoni, C., et al. (2016). Magnetic structure and phase stability of the van der Waals bonded ferromagnet Fe52GeTe5. Phys. Rev. B 93, 014411.

34. Pallecchi, I., Caglieris, F., and Putti, M. (2016). Thermoelectric properties of iron-based superconductors and parent compounds. Supercond. Sci. Technol. 29, 073002.

35. Asamitsu, A., Morimoto, Y., and Tokura, Y. (1996). Thermoelectric effect in La1-x SrMnO3. Phys. Rev. B 53, R2952.

36. Ahmed, F., Tsujii, N., and Mori, T. (2017). Thermoelectric properties of CuGa1-xMnxTe2: power factor enhancement by incorporation of magnetic ions. J. Mater. Chem. A 5, 7545.

37. Arora, P., Chattopadhyay, M.K., and Roy, S.B. (2007). Magnetocaloric effect in MnSi. Phys. B 409, 1425.

38. Schnelle, W., Leithe-Jasper, A., Schmidt, M., et al. (2005). Itinerant iron magnetism in filed skutterudites CaFe6Sb12 and YbFe6Sb12. stable divalent state of ytterbium. Phys. Rev. B 72, 020402(R).

39. Schnelle, W., Leithe-Jasper, A., Schmidt, M., et al. (2005). Itinerant iron magnetism in filed skutterudites CaFe6Sb12 and YbFe6Sb12. stable divalent state of ytterbium. Phys. Rev. B 72, 020402(R).

40. Nikiforov, V.N., Piyadun, V.V., Morozkin, A.V., and Irkhin, V.Yu. (2014). Anomalies of transport properties in antiferromagnetic YbMn2Sb2 compound. Phys. Lett. A 378, 1488.

41. Morozkin, A.V., Isnard, O., Henry, P., et al. (2006). Synthesis and magnetic structure of the YMn3Sb21 compound. J. Alloys Compd. 420, 34.

42. Allen, J.W., Lucovsky, A., and Mikkelsen, J.C., Jr. (1977). Optical properties and electronic structure of crossroads material MnTe. Solid State Commun. 24, 367.

43. Arora, P., Chattopadhyay, M.K., and Roy, S.B. (2007). Magnetocaloric effect in MnSi. Phys. B 409, 1425.

44. Gehl, M., and Nørskov, J.K. (2006). Magnetocaloric effect in the Kondo systems CePd3 and CeIn3. Appl. Phys. Lett. 92, 101909.

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AUTHOR CONTRIBUTIONS

P.S. conceived the project. P.S., K.R.K., M.L., Z.W., and J.X. discussed the thermodynamic description of the Seebeck effect and performed data mining from the literature. P.S. and W.Z. did the mean-field calculations and wrote the manuscript. All authors reviewed and approved the manuscript.

DECLARATION OF INTERESTS

The authors declare that they have no competing interests.

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