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Ahmad Us Saleheen
Louisiana State University

Tapas Samanta
Louisiana State University

Mojammel Khan
Louisiana State University

Philip W. Adams
Louisiana State University

David P. Young
Louisiana State University

See next page for additional authors

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Authors

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SPECIAL TOPICS

The influence of hydrostatic pressure on the magnetic and magnetocaloric properties of DyRu₂Si₂

Ahmad Us Saleheen,^{1,a)} Tapas Samanta,¹ Mojammel Khan,¹ Philip W. Adams,¹ David P. Young,¹ Igor Dubenko,² Naushad Ali,² and Shane Stadler¹

¹Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803, USA

²Department of Physics, Southern Illinois University, Carbondale, Illinois 62901, USA

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We report the magnetic and magnetocaloric properties of the tetragonal rare-earth compound DyRu₂Si₂ under applied hydrostatic pressure. The isothermal entropy change (ΔS) and the adiabatic temperature change (ΔT_{ad}) were calculated from magnetization data collected at different applied pressures and from heat capacity measurements conducted at atmospheric pressure, respectively. The application of hydrostatic pressure significantly modified the multi-step magnetization curve and the saturation magnetization. A suppression of the magnetization was observed for $P = 0.588$ GPa and $P = 0.654$ GPa whereas, at about $P \approx 1$ GPa, the saturation magnetization increased and the magnetization isotherms again resembled the curves measured at atmospheric pressure. A small thermal hysteresis was observed between the heating and cooling $M(T)$ curves at $T_1 = 3.4$ K, with an applied magnetic field of $H = 0.1$ T. This thermal hysteresis indicates a first-order like transition which was also supported by the Arrott plot analysis. The volume magnetostriction was estimated from the pressure-dependent magnetization measurements using a Maxwell relation. *Published by AIP Publishing.* [<http://dx.doi.org/10.1063/1.4974466>]

I. INTRODUCTION

Magnetic cooling technology based on the magnetocaloric effect (MCE) has evolved as a viable technological alternative to conventional cooling technology based on the vapor compression cycle.^{1,2} The MCE is the isothermal entropy change or adiabatic temperature change of a magnetic material when subjected to an external magnetic field. Extensive research has revealed classes of materials, such as Gd₅Si₂Ge₂,³ MnAs-based compounds,^{4,5} La(Fe_{1-x}Si_x)₁₃,⁶ Mn(Co, Ni)Ge-based compounds, and⁷⁻¹⁰ Ni₂MnGa and Ni₂MnIn-based Heusler alloys,^{11,12} all of which show large MCEs near room temperature. A large MCE at low temperature is also useful for specific technological purposes, such as space science and, most prominently, in gas liquefaction.¹³⁻¹⁵ Antiferromagnetic (AFM) systems can be good candidates in the cryogenic temperature range since they may show large MCE values due to magnetic-field-induced metamagnetic transitions from antiferromagnetic (AFM) to ferromagnetic (FM) states.¹³ In this study, we report our findings on the magnetocaloric and magnetic properties of DyRu₂Si₂. This compound belongs to a class of materials with the general formula RT₂X₂, where R is a rare earth element, T is a transition metal, and X is silicon or germanium. These materials show a wide variety of properties such as heavy fermionic behavior, superconductivity, and complex and multiple magnetic transitions.¹⁶

DyRu₂Si₂ crystallizes in the body centered tetragonal ThCr₂Si₂-type structure with space group $I4/mmm$, where only the Dy atoms possess a magnetic moment.¹⁷⁻²⁰ In the temperature range 2–400 K, DyRu₂Si₂ shows two magnetic

transitions (M vs. T), and multi-step metamagnetic transitions (M vs. H) at low temperature. We observed a maximum total isothermal entropy change (ΔS_T) of -6.6 J/kg K at $T = 29.2$ K for a 7 T magnetic field change, calculated from magnetization isotherms. From field-dependent heat capacity data, we calculated an adiabatic temperature change (ΔT_{ad}) of 8.2 K at $T = 5.1$ K for a 5 T magnetic field change. For comparison, other materials in this class, GdCr₂Si₂,²¹ HoRu₂Si₂,¹⁴ and ErRu₂Si₂,¹³ were reported to show the entropy changes (ΔS) of 14.1 J/kg K, 9.1 J/kg K, and 19.3 J/kg K at $T = 8$ K, 19 K, and 5.5 K, respectively, for a field change of 5 T. In the case of ErRu₂Si₂, an adiabatic temperature change (ΔT_{ad}) of 12.9 K was found at $T = 5.5$ K for a 5 T magnetic field change. DyRu₂Si₂ showed magnetostrictive behavior, and the volume magnetostriction ($\Delta V/V$) was estimated from the pressure-dependent magnetization measurements. Hydrostatic pressure significantly altered the saturation magnetization and magnetic interactions, as well as the magnetocaloric properties of this compound.

II. EXPERIMENTAL METHODS

Polycrystalline DyRu₂Si₂ buttons (approximately 2 g) were fabricated from high-purity elements (better than 99.9%) Dy, Ru, and Si by conventional arc-melting in an argon atmosphere. The weight loss after melting was found to be less than 0.3%. The samples were placed inside an evacuated quartz tube and annealed at 750 °C in a tube furnace for 5 days and slowly cooled to room temperature. X-ray diffraction (XRD) measurements were done with a room-temperature X-ray diffractometer using CuK α radiation and θ - θ geometry. Magnetization measurements within a temperature interval of 2–400 K and up to 7 T magnetic fields were done with a

^{a)}Electronic mail: aussal1@lsu.edu

superconducting quantum interference device magnetometer [SQUID, Quantum Design magnetic property measurement system (MPMS)]. Irregularly shaped samples of about 3 mg were used in these measurements. Magnetic measurements under hydrostatic pressure were performed using a commercial BeCu cylindrical pressure cell manufactured by Quantum Design. Daphne 7373 oil was used as the pressure transmitting medium. The magnitude of the applied pressure was calibrated by measuring the shift of the superconducting transition temperature of Pb, which was placed in the cell with the sample as a reference manometer. Heat capacity measurements were done on a Quantum Design Physical Properties Measurement System (PPMS). For this purpose, a $4.4 \times 1 \times 0.6$ mm sample of around 18 mg was cut using a spark cutter.

III. RESULTS AND DISCUSSION

From the room temperature XRD pattern for DyRu_2Si_2 (Fig. 1), the ThCr_2Si_2 -type tetragonal structure was verified. The lattice parameters were calculated as $a = 4.147(8)$ Å and $c = 9.522(8)$ Å, consistent with the previously reported values.²² The magnetization as a function of temperature (M vs. T) with $H = 0.1$ T at atmospheric pressure is shown in Fig. 2. Two magnetic transitions, one at $T_N = 29.2$ K and another at $T_t = 3.4$ K, are clearly visible. A small but distinct hysteresis of about 0.3 K was observed between the zero-field-cooled (ZFC) warming and field-cooled-cooling (FCC) curves in the temperature range of 8 K to around $T_t = 3.4$ K. Previous studies reported another transition at $T'_t = 1.5$ K,^{17,18} (and references therein) but this was beyond the temperature range of our instrument. DyRu_2Si_2 has three temperature-dependent phases, namely: a low temperature phase (LTP) below T'_t , an intermediate temperature phase (ITP) at $T'_t < T < T_t$, and a high temperature phase (HTP) at $T_t < T < T_N$.^{17,18}

According to the model proposed by Kawano *et al.*,^{17,18} the ITP has a two dimensional spin configuration with a magnetic unit cell of $9a \times 9a \times c$, where a and c represent the lattice constants of the chemical unit cell. In the same study, the spin configuration for the HTP had been shown to exhibit a one dimensional magnetic structure. In light of this model, at T_t the system transforms from a one dimensional

FIG. 2. Magnetization (M) vs. temperature (T) for DyRu_2Si_2 at $H = 0.1$ T applied field at atmospheric pressure. (Inset) Zoomed-in view of the same data to show the thermal hysteresis.

magnetic structure to a two dimensional structure upon cooling, giving rise to a high magnetization phase that is evident from the peak in the magnetization around T_t . The origin of the thermal hysteresis in the M vs. T data may be the order-order transition between the two-dimensional and one-dimensional magnetic configurations.

Fig. 3 shows the Arrott plot analysis of the magnetization isotherms at atmospheric pressure for some selected temperatures around T_t and T_N . It is known (Banerjee criterion)²³ that a negative slope in the Arrott plot is an indication of a first-order magnetic transition, whereas a positive slope indicates a second-order transition. The curves around T_t formed “S” like shapes with negative slopes at lower field values and positive slopes at higher fields. This occurrence of a negative curvature in Arrott plots is usually associated with a first-order magnetic transition.^{21,24–28} The small temperature hysteresis in the M vs. T data also points towards the first-order nature of the transition at T_t . Curves showing negative slopes continue up to the Néel temperature (T_N) and, starting from 30 K, they start to have positive slopes even at low fields. At higher temperatures, no negative

FIG. 1. Room temperature powder XRD pattern for DyRu_2Si_2 .

FIG. 3. Arrott plots of the magnetization data for DyRu_2Si_2 (M^2 vs. H/M) at selected temperatures and atmospheric pressure.

slopes were observed, implying a second-order transition to a paramagnetic (PM) state. We also attempted to construct a universal curve for the entropy change at an atmospheric pressure (not shown), in light of the works by Franco and Conde.²⁹ For materials showing second-order magnetic transitions, the ΔS vs. T curves measured at different fields should collapse into a single curve upon rescaling of the temperature axis, whereas failure to collapse is considered to be an indication of a first-order transition.^{29,30} In our case, the curves collapsed into a single universal curve around T_N . However, below T_N , and around T_I , the curves failed to collapse. This could be construed as an indication of a first-order transition, however, this view was challenged by Smith *et al.*,³¹ arguing that there was no one-to-one connection between the behavior of the scaling procedure and the order of the phase transition. Based on these findings, and especially the observed thermal hysteresis, it is plausible that the magnetic phase transition at T_I is first-order like.

Fig. 4 shows the magnetization isotherms at 2 K at various pressures. In increasing field, three distinct transitions were observed with critical fields of $H_1 = 0.2$ T, $H_2 = 1.2$ T, and $H_3 = 2.2$ T, consistent with previous reports.¹⁹ For pressures of $P = 0.588$ and 0.654 GPa, the transitions at $H > 0.2$ T were suppressed (barely discernible in the data). At a pressure of 0.934 GPa, these phases return and, at 1.24 GPa, they have nearly the same critical fields as they had had at atmospheric pressure. Moreover, at atmospheric pressure, although the M vs. H curve does not fully saturate even at 7 T, a saturation tendency could be seen around $H = 3$ T. For pressures of $P = 0.588$ and 0.654 GPa, this saturation tendency was not observed, and the curves follow a nearly linear dependence on increasing field, pointing towards a weaker magnetic ordering. Also, the magnetization values at $H = 7$ T for $P = 0.588$ and 0.654 GPa are considerably smaller than those at atmospheric pressure. This saturation tendency and non-linearity return at 0.934 GPa, and continue up to 1.24 GPa, but the magnetization starts to saturate at a higher field value than that observed at atmospheric pressure. Moreover, the magnetization at $H = 7$ T is larger for $P = 1.24$ GPa than at atmospheric pressure.

FIG. 4. Magnetization (M) vs. magnetic field (H) for DyRu_2Si_2 at $T = 2$ K for various applied hydrostatic pressures.

Heavy rare-earth elements, such as pure Dy and materials consisting of rare-earth elements often exhibit magnetostriction,^{32–34} which lead us to explore the magnetostrictive properties of this compound. Using the Maxwell relation

$$\left(\frac{\partial M}{\partial P}\right)_{H,T} = -\left(\frac{\partial V}{\partial H}\right)_{P,T}, \quad (1)$$

we calculated the volume magnetostriction, $\Delta V/V$, where V is the volume of the unit cell calculated from the lattice parameters obtained from the XRD measurements. In Eq. (1), M is the magnetization, P is the applied pressure, V is the volume, and H is the applied magnetic field. This method was also applied by Nikitin *et al.*^{33,34} to estimate the volume magnetostriction in single crystals of Dy and Y_2Fe_{17} . Fig. 5 shows the temperature dependence of the volume magnetostriction at various pressures. At low fields, a peak in the volume magnetostriction was observed around T_N . This is consistent with the observation that the onset of magnetic ordering gives rise to magnetostriction in heavy rare earth metals as reported by Nikitin *et al.*³⁵ As the magnetic field increases, the magnitude of the volume magnetostriction increases, and the peak in the $\Delta V/V$ vs. T curves shifts towards lower temperatures. The highest value of the volume magnetostriction was found to be 0.35% at $H = 7$ T and an applied pressure of $P = 0.588$ GPa. A volume magnetostriction of 0.7×10^{-3} at a field of 1.2 T was reported for a pure Dy single crystal,³³ whereas we calculated a value of 0.1×10^{-3} for the same magnetic field. For pressures of $P > 0.588$ GPa, the volume magnetostriction decreased systematically, as shown in Fig. 6(a).

The presence of magnetostriction in this compound can be attributed to the variation of magnetic properties due to applied hydrostatic pressure. This kind of pressure dependent behavior had been observed in invar alloys, heavy rare-earth elements such as Tb, Gd, Dy, etc., Tb-Gd alloys, and others.^{36,37} According to Eq. (1), sharp changes in magnetization with pressure result in volume changes. We observed large changes in the magnetic properties of DyRu_2Si_2 at pressures of $P = 0.588$ and 0.654 GPa which were translated into volume changes as expected from the Maxwell relation (Eq. (1)). As pointed out by Doerr *et al.*, crystal field and exchange striction are the two most important mechanisms for magnetostriction in rare-earth systems.³² Tomala *et al.*³⁸ studied the crystal field properties of DyRu_2Si_2 by Mössbauer spectroscopy and calculated the crystal electric field (CEF) parameters, B_2^0 with an anomalously large value of -4.94 K and B_4^0 with a value of 0.0050 K. With the application of pressure, both the crystal electric field environment and the exchange interaction can change. However, identifying the mechanism that plays the dominant role in the magnetostriction requires further investigation involving single crystals.

The total entropy change was calculated from the magnetization isotherms using the integrated Maxwell relation

$$\Delta S_T(T) = \int_0^H \left(\frac{\partial M}{\partial T}\right)_H dH. \quad (2)$$

Fig. 7 shows the temperature dependence of ΔS_T for fields up to 7 T at various pressures. At atmospheric pressure,

FIG. 5. (a)–(f) Temperature (T) dependence of the volume magnetostriction ($\Delta V/V$) of DyRu_2Si_2 at magnetic fields up to 7 T for various pressures (P).

peaks were observed around T_N , and the highest value of ΔS_T was -6.6 J/kg K at $T = 29 \text{ K}$ for $\Delta H = 7 \text{ T}$. An inverse MCE ($\Delta S_T = 2.2 \text{ J/kg K}$) was also observed at $T = 3 \text{ K}$ for $\Delta H = 7 \text{ T}$, at this pressure. As the temperature increases beyond 3 K, the inverse MCE vanishes and a normal MCE emerges. For the 1 T curve, the inverse MCE starts at 5 K and continues up to 27 K. This inverse MCE could be

associated with the low-temperature AFM phase. Subtle but clear signs of the transition at T_I were also evident, especially at lower fields. The value of ΔS_T decreased at an applied pressure of 0.588 GPa and, in this case, two distinct peaks were observed, one at 5 K (around T_I) and the other at 31 K (around T_N) with ΔS_T values of -3.6 and -2.4 J/kg K , respectively. It was noted that, at this pressure, the largest value of ΔS_T was found near T_I rather than at T_N . This scenario prevails at $P = 0.654 \text{ GPa}$ but, at $P = 0.710 \text{ GPa}$, the largest value of ΔS_T was again observed at T_N . This trend continues up to the highest applied pressure of 1.24 GPa. After the initial decrease of the overall values of ΔS_T for applied pressures of 0.588 GPa and 0.654 GPa, the values start to increase at pressures exceeding 0.710 GPa. At $P = 1.24 \text{ GPa}$, the peak value of -6.3 J/kg K at $H = 7 \text{ T}$ is very close to the atmospheric pressure value of -6.6 J/kg K (Fig. 6(b)). Moreover, the overall shape and features of the curves at $P = 1.24 \text{ GPa}$ are similar to those at atmospheric pressure. This tendency was also observed in the M vs. H curves at different pressures (Fig. 4).

The origin of this pressure-dependent behavior may lie with the change of the a/c ratio with pressure. As pointed out by Szytula *et al.*,¹⁶ in RT_2X_2 systems, the magnetic interaction is governed by the Ruderman–Kittel–Kasuya–Yosida (RKKY) model and the a/c ratio plays a crucial role in defining the nature and strength of the interaction. In RT_2X_2

FIG. 6. (a) The maximum values of $\Delta V/V$ at $H = 7 \text{ T}$ for DyRu_2Si_2 at various applied pressures. (b) The maximum values of ΔS_T at $H = 7 \text{ T}$ for various applied pressures.

FIG. 7. (a)–(g) Temperature (T) dependence of the total entropy change (ΔS) of DyRu_2Si_2 for various changes in applied magnetic field (H) and pressure (P).

systems containing heavy rare earths ($R = \text{Tb-Tm}$), a simple collinear AFM structure is observed when $a/c < 0.415$ and oscillatory magnetic structures emerge when $a/c > 0.415$. At atmospheric pressure, DyRu_2Si_2 has an a/c ratio of 0.435, well above the critical value of 0.415. At pressures of 0.588 and 0.654 GPa, the M vs. H (Fig. 4) data at $T = 2$ K point toward a simpler AFM structure than that which occurs at atmospheric pressure. But for $P > 0.710$ GPa, the curves again resemble those measured at atmospheric pressure. From this fact, and from Fig. 4, it is plausible that, at pressures of 0.588 and 0.654 GPa, the system favors a simpler AFM-like interaction, while at atmospheric pressure and at pressures greater than 0.654 GPa, the system tends toward more complicated interactions. Also, the exchange integral depends on the interatomic distances.³⁶ The change in volume, and hence the interatomic distances, due to applied pressure can affect the exchange integral, resulting in a variation of magnetic interactions. The observed variation in magnetic properties with pressure can possibly be attributed to the oscillatory nature of the RKKY interaction.

As there is a volume anomaly due to magnetostriction, the total field-induced entropy change is a sum of the magnetic and structural entropy changes,^{39,40} i.e.,

$$\Delta S_T = \Delta S_{\text{mag}} + \Delta S_{\text{str}}. \quad (3)$$

Gschneidner *et al.*³⁹ compiled a collection of ΔS_{str} (i.e., structural contribution to the total entropy change) for different materials undergoing magnetostructural transitions, as well as volume anomalies, and found a linear relationship between ΔS_{str} and $\Delta V/V$. In light of this, we can use the relationship^{41,42} $\delta[\Delta V/V(\%)]/\delta[\Delta S_{\text{str}}] = 0.08 \text{ (J/kg K)}^{-1}$ to estimate the structural contribution to the total entropy change. According to this relation, the observed volume magnetostriction of 0.35% at $H = 7$ T and $P = 0.588$ GPa corresponds to a structural entropy change of $\Delta S_{\text{str}} = 4.4 \text{ J/kg K}$. In Fig. 6, it can be seen that, as the pressure increases, the volume magnetostriction $\Delta V/V$, and therefore ΔS_{str} , decrease, whereas the value of the total entropy change (ΔS_T) increases. This may indicate that the structural entropy change (ΔS_{str}) opposes the magnetic entropy change (ΔS_{mag}), thereby reducing ΔS_T .

From the zero-field heat capacity measurements at atmospheric pressure (Fig. 8), a well defined anomaly was observed starting at $T = 28.3$ K and ending at $T = 29.3$ K with a drop of about 11.8 J/kg K during heating, which is a characteristic of magnetic ordering. No clear signature of an anomaly was observed in the $C(T)$ data around $T_1 = 3.4$ K, probably due to low resolution in that temperature range. However, a sharp spike in the C/T vs. T plot as shown in Fig. 8(b) was observed around $T = 2$ K and 29 K.

By integrating the C/T data for $H = 0$ T and 5 T, the entropy at these two fields, $S(0 \text{ T})$ and $S(5 \text{ T})$, were calculated

FIG. 8. Temperature (T) dependence of the heat capacity (C_p) of DyRu_2Si_2 measured at atmospheric pressure and zero applied magnetic field. (a) Heat capacity (C_p) measured at $H=0$ T and 5 T magnetic field. (b) C_p/T vs. T at $H=0$ T and 5 T magnetic field.

FIG. 9. (a) Temperature (T) dependence of the adiabatic temperature change (ΔT_{ad}) of DyRu_2Si_2 . (b) Temperature (T) dependence of the total entropy change (ΔS) calculated from the heat capacity data.

and subtracted to get $\Delta S = S(5\text{ T}) - S(0\text{ T})$, as shown in Fig. 9(b). A peak value of -5.5 J/kg K was found at $T=29.3\text{ K}$ which is the Néel temperature, while a value of -4.6 J/kg K was found from the atmospheric pressure magnetization data for an applied field of 5 T. These numbers are in reasonable agreement. To calculate the adiabatic temperature change (ΔT_{ad}), $S(0\text{ T})$ and $S(5\text{ T})$ were first interpolated from the S vs. T plots. T vs. S plots were then plotted with the interpolated S data for 0 and 5 T magnetic fields and, after interpolating $T(0\text{ T})$ and $T(5\text{ T})$, ΔT_{ad} was calculated using the relation $\Delta T_{ad} = T(5\text{ T}) - T(0\text{ T})$. Fig. 9(a) shows the adiabatic temperature change ΔT_{ad} , and a peak value of 8.2 K was observed at $T=5.1\text{ K}$, which is near the transition temperature T_1 . A second peak of magnitude 4.2 K was observed at $T=29.2\text{ K}$ (T_N).

IV. CONCLUSIONS

Pressure-dependent magnetization measurements have been carried out on DyRu_2Si_2 , from which we have estimated the volume magnetostriction and entropy changes (ΔS_T) as a function of hydrostatic pressure. We observed the

multi-step metamagnetic transitions in the $M(H)$ curves at atmospheric pressure, the features of which diminished at intermediate pressures ($P=0.588\text{ GPa}$ and 0.654 GPa) and reappeared at higher pressures ($P \approx 1\text{ GPa}$). Moreover, the saturation magnetization decreased at intermediate pressures, following a near-linear dependence on increasing field, indicating a weaker magnetic ordering. At higher pressures, the saturation magnetization increased again and the $M(H)$ curves resembled those measured at atmospheric pressure. This trend of pressure dependence was also observed in the (ΔS_T) vs. T curves measured at various pressures. At intermediate pressures, the entropy changes (ΔS_T) decreased, whereas these values gradually increased with higher applied pressures. The variation of the magnetic properties with pressure is likely connected to the modulation of the exchange integral (RKKY-type behavior), caused by a change in inter-atomic distances due to applied hydrostatic pressure.

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- ¹K. A. Gschneidner, Jr., V. K. Pecharsky, and A. O. Tsokol, *Rep. Prog. Phys.* **68**, 1479 (2005).
- ²A. M. Tishin and Y. I. Spichkin, *The Magnetocaloric Effect and Its Applications* (Institute of Physics, London, 2003).
- ³V. K. Pecharsky and K. A. Gschneidner, Jr., *Phys. Rev. Lett.* **78**, 4494 (1997).
- ⁴H. Wada and Y. Tanabe, *Appl. Phys. Lett.* **79**, 3302 (2001).
- ⁵S. Gama, A. A. Coelho, A. de Campos, A. M. G. Carvalho, F. C. G. Gandra, P. J. von Ranke, and N. A. de Oliveira, *Phys. Rev. Lett.* **93**, 237202 (2004).
- ⁶A. Fujita, S. Fujieda, Y. Hasegawa, and K. Fukamichi, *Phys. Rev. B* **67**, 104416 (2003).
- ⁷T. Samanta, I. Dubenko, A. Quetz, S. Stadler, and N. Ali, *Appl. Phys. Lett.* **101**, 242405 (2012).
- ⁸N. T. Trung, L. Zhang, L. Caron, K. H. J. Buschow, and E. Brück, *Appl. Phys. Lett.* **96**, 172504 (2010).
- ⁹T. Samanta, I. Dubenko, A. Quetz, S. Temple, S. Stadler, and N. Ali, *Appl. Phys. Lett.* **100**, 052404 (2012).
- ¹⁰E. Liu, W. Wang, L. Feng, W. Zhu, G. Li, J. Chen, H. Zhang, G. Wu, C. Jiang, H. Xu, and F. de Boer, *Nat. Commun.* **3**, 873 (2012).
- ¹¹S. Stadler, M. Khan, J. Mitchell, N. Ali, A. M. Gomes, I. Dubenko, A. Y. Takeuchi, and A. P. Guimares, *Appl. Phys. Lett.* **88**, 192511 (2006).
- ¹²J. Liu, T. Gottschall, K. P. Skokov, J. D. Moore, and O. Gutfleisch, *Nat. Mater.* **11**, 620 (2012).
- ¹³T. Samanta, I. Das, and S. Banarjee, *Appl. Phys. Lett.* **91**, 152506 (2007).
- ¹⁴T. Paramanik, K. Das, T. Samanta, and I. Das, *J. Appl. Phys.* **115**, 083914 (2014).
- ¹⁵H. Zhang, B. G. Shen, Z. Y. Xu, J. Shen, F. X. Hu, J. R. Sun, and Y. Long, *Appl. Phys. Lett.* **102**, 092401 (2013).
- ¹⁶Andrzej Szytuła and Janusz Leciejewicz, *Handbook on the Physics and Chemistry of Rare Earths* (Elsevier Science Publishers B.V., 1989), Chap. 83, pp. 133–211.
- ¹⁷S. Kawano, M. Takahashi, T. Shigeoka, N. Iwata, and T. Kosugi, *Appl. Phys. A* **74**, S646 (2002).
- ¹⁸S. Kawano, M. Takahashi, T. Shigeoka, N. Iwata, and M. Bull, *Physica B* **346–347**, 99 (2004).

- ¹⁹B. Andreani, G. L. F. Fraga, A. Garnier, D. Gignoux, D. Maurin, D. Schmitt, and T. Shigeoka, *J. Phys.: Condens. Matter* **7**, 1889 (1995).
- ²⁰N. Iwata, K. Hattori, M. Kadota, M. Takahashi, S. Kawano, and T. Shigeoka, *J. Magn. Magn. Mater.* **256**, 189 (2003).
- ²¹L. Li, G. Hu, I. Umehara, D. Huo, W. D. Hutchison, T. Namiki, and K. Nishimura, *J. Alloys Compd.* **575**, 1 (2013).
- ²²K. Hiebl, C. Horvath, P. Rogl, and M. Sienko, *J. Magn. Magn. Mater.* **37**, 287 (1983).
- ²³B. Banerjee, *Phys. Lett.* **12**, 16 (1964).
- ²⁴S. Pramanick, P. Dutta, S. Chatterjee, S. Giri, and S. Majumdar, *J. Magn. Magn. Mater.* **405**, 270 (2016).
- ²⁵B. Maji, M. K. Ray, K. G. Suresh, and S. Banerjee, *J. Appl. Phys.* **116**, 213913 (2014).
- ²⁶W. Dunhui, T. Shaolong, H. Songling, Z. Jianrong, and D. Youwei, *J. Magn. Magn. Mater.* **268**, 70 (2004).
- ²⁷X. Chen, Y. Zhuang, J. Yan, and F. Fei, *J. Alloys Compd.* **479**, 35 (2009).
- ²⁸N. T. Trung, Z. Q. Ou, T. J. Gortenmulder, O. Tegus, K. H. J. Buschow, and E. Brück, *Appl. Phys. Lett.* **94**, 102513 (2009).
- ²⁹V. Franco and A. Conde, *Int. J. Refrig.* **33**, 465 (2010).
- ³⁰C. M. Bonilla, J. Herrero-Albillos, F. Bartolomé, L. M. García, M. Parra-Borderías, and V. Franco, *Phys. Rev. B* **81**, 224424 (2010).
- ³¹A. Smith, K. K. Nielsen, and C. R. H. Bahl, *Phys. Rev. B* **90**, 104422 (2014).
- ³²M. Doerr, M. Rotter, and A. Lindbaum, *Adv. Phys.* **54**, 1 (2005).
- ³³S. Nikitin, A. Tishin, and P. Leontiev, *J. Magn. Magn. Mater.* **92**, 405 (1991).
- ³⁴S. Nikitin, A. Tishin, M. Kuz'min, and Y. Spichkin, *Phys. Lett. A* **153**, 155 (1991).
- ³⁵S. Nikitin, D. Kim, and O. Chistyakov, *Zh. Eksp. Teor. Fiz.* **71**, 1610 (1976).
- ³⁶S. A. Nikitin, *Moscow Univ. Phys. Bull.* **66**, 519 (2011).
- ³⁷S. A. Nikitin and R. V. Bezduzhnyi, *Sov. Phys. JETP* **66**, 1058 (1987).
- ³⁸K. Tomala, J. P. Sanchez, and R. Kmiec, *J. Phys.: Condens. Matter* **1**, 4415 (1989).
- ³⁹J. K. A. Gschneidner, Y. Mudryk, and V. Pecharsky, *Scr. Mater.* **67**, 572 (2012).
- ⁴⁰V. Pecharsky, A. Holm, J. K. A. Gschneidner, and R. Rink, *Phys. Rev. Lett.* **91**, 197204 (2003).
- ⁴¹T. Samanta, D. L. Lepkowski, A. U. Saleheen, A. Shankar, J. Prestigiacomo, I. Dubenko, A. Quetz, I. W. H. Oswald, G. T. McCandless, J. Y. Chan, P. W. Adams, D. P. Young, N. Ali, and S. Stadler, *Phys. Rev. B* **91**, 020401(R) (2015).
- ⁴²J. L. Wang, L. Caron, S. J. Campbell, S. J. Kennedy, M. Hofmann, Z. X. Cheng, M. F. Md Din, A. J. Studer, E. Brück, and S. X. Dou, *Phys. Rev. Lett.* **110**, 217211 (2013).