Extraordinary induction heating effect near the first order Curie transition

M. R. Barati,[1] C. Selomulya,[2] K. G. Sandeman,[3,4] and K. Suzuki[1,a)]
[1]*Department of Materials Engineering, Monash University, Clayton, Victoria 3800, Australia*
[2]*Department of Chemical Engineering, Monash University, Clayton, Victoria 3800, Australia*
[3]*Department of Physics, Blackett Laboratory, Imperial College London, London SW7 2AZ, United Kingdom*
[4]*Department of Physics, Brooklyn College, CUNY, 2900 Bedford Avenue, Brooklyn, New York 11210, USA*

(Received 7 September 2014; accepted 16 October 2014; published online 24 October 2014)

While materials with a 1st order Curie transition (T_C) are known for the magnetic cooling effect due to the reversibility of their large entropy change, they also have a great potential as a candidate material for induction heating where a large loss power is required under a limited alternating magnetic field. We have carried out a proof-of-concept study on the induction heating effect in 1st order ferromagnetic materials where the temperature is self-regulated at T_C. $LaFe_{11.57}Si_{1.43}H_{1.75}$, a well-known magnetocaloric material, was employed in this study because T_C of this compound (319 K) resides in the ideal temperature range for hyperthermia treatment of cancerous cells. It is found that the hysteresis loss of $LaFe_{11.57}Si_{1.43}H_{1.75}$ increases dramatically near T_C due to the magnetic phase coexistence associated with the 1st order magnetic transition. The spontaneous magnetization (M_s) shows a very abrupt decrease from 110 Am^2kg^{-1} at 316 K to zero at 319 K. This large M_s immediately below T_C along with the enhanced irreversibility of the hysteresis curve result in a specific absorption rate as large as 0.5 kWg^{-1} under a field of 8.8 kAm^{-1} at 279 kHz. This value is nearly an order of magnitude larger than that observed under the same condition for conventional iron oxide-based materials. Moreover, the large heating effect is self-regulated at the 1st order T_C (319 K). This proof-of-concept study shows that the extraordinary heating effect near the 1st order Curie point opens up a novel alloy design strategy for large, self-regulated induction heating. [http://dx.doi.org/10.1063/1.4900557]

The idea of utilizing the power losses of magnetic materials for remote heating[1–4] was introduced as early as in the 1950s for hyperthermia treatment of cancerous cells.[1] This approach could promise to be minimally invasive, selective, and inexpensive if successfully applied for the intra-cellular hyperthermia treatment. However, there has been one major problem unresolved to date; namely that the current specific heating power of magnetic particles available for treatment is far from satisfactory.[5] As a result, the clinical applications of magnetic particles for hyperthermia therapy[6,7] usually require either: (a) overwhelmingly large applied magnetic fields at very high frequencies or (b) a massive dose of magnetic materials to ensure sufficient heating effects to destroy the tumour tissue.

Given the attainable concentration of magnetic particles in the cells and the required therapeutic temperatures (315 to 319 K) for the effective intra-cellular hyperthermia therapy, the heating power of the magnetic material needs to be at least ~ 1 kWg^{-1}(Ref. 8) with a small radio-frequency (RF) power where healthy cells are unaffected. Although such a biocompatible RF power remains undefined universally, some authors have proposed[8] that the product of the magnetic field and frequency ($H{\cdot}f$) needs to be less than $\sim 10^9$ $A(m{\cdot}s)^{-1}$. Under this biocompatible condition, the specific absorption rate (SAR)[9] of traditional superparamagnetic particles[10] remains typically two orders of magnitude smaller than what is required. Recently, a novel approach based on exchange-coupled core-shell particles has been employed by Lee *et al.*[11] They reported a large heating power above 1 kWg^{-1} for the core-shell structure with two distinct local magnetocrystalline anisotropy constants. However, the $H{\cdot}f$ value used was well above 10^{10} $A(m{\cdot}s)^{-1}$ where an animal body could be affected.[12]

It is well known that the loss power of magnetic particles reflects the out-of-phase susceptibility (χ'').[2] Jeun *et al.*[13] have investigated the induction heating effect in Fe_3O_4 nanoparticles in both superparamagnetic and ferrimagnetic states and concluded that the attainable power loss in superparamagnetic state is far lower than that in the ferrimagnetic state because of the limited χ'' in a pure superparamagnetic state. Hence, a higher induction heating effect is expected for a magnetically ordered state through the hysteresis loss. Kita *et al.*[8] have demonstrated that their Co-doped iron oxide nanoparticles in the ferromagnetic state exhibit a high specific loss power of 420 Wg^{-1} at $H{\cdot}f = 6 \times 10^9$ $A(m{\cdot}s)^{-1}$, reflecting the advantage of the magnetically ordered state.

Utilizing the hysteresis-loss heating effect in ferro- and ferri-magnetic materials has another advantage. Since the hysteresis loss in these magnetically order materials diminishes at the Curie temperature (T_C), the temperature rise induced by induction heating is self-regulated at T_C.[14–17] Consequently, magnetic hyperthermia treatment could be free from external temperature control when the T_C is tuned to the therapeutic temperature range. Brezovich *et al.*[14] for ferromagnetic Ni-Cu alloys and Shimizu *et al.*[16] for their biocompatible ferrimagnetic (Mg,Ti)-ferrite demonstrated that the temperature rise is self-regulated by the Curie point in these ferro- and ferri-magnetic materials. While the self-regulated induction heating effect is ideal for biomedical

a)Author to whom correspondence should be addressed. Electronic mail: kiyonori.suzuki@monash.edu

applications, the spontaneous magnetization (M_s) of T_C-tuned magnetic materials with the conventional 2nd order Curie transition is very limited[18] at the human body temperature because the latter (310 K) is only about 5 K below the Curie point when T_C is tuned to match the therapeutic temperature (315 K). Hence, the advantage in utilizing the large hysteresis loss of the magnetically ordered state is lost. Nevertheless, we propose that this fundamental limitation due to the 2nd order Curie transition can be avoided in ferromagnetic materials with the 1st order Curie transition. The spontaneous magnetization of such a 1st order material disobeys the Brillouin function and M_s diminishes abruptly at T_C, resulting in large M_s values at temperatures immediately below T_C. Hence, a large loss power is expected for the 1st order ferromagnetic materials at the human body temperature even if T_C is tuned to the therapeutic temperature. Furthermore, an extra irreversible effect on the magnetic polarization process is expected owing to thermomagnetic hysteresis associated with the coexistence of magnetic phases near the 1st order Curie point. This irreversibility may lead to a dramatic increase in the loss power near T_C. Consequently, 1st order ferromagnetic materials have a great potential as a candidate of magnetic materials for self-regulated induction heating.

In this short report, we have carried out a proof-of-concept study on utilizing the 1st order Curie transition for self-regulated large induction heating. La(Fe, Si)$_{13}$-based materials[19–27] have been chosen in this study because the 1st order Curie point of this compound is tuneable by incorporation of hydrogen over a wide temperature range[20,22–24] which covers the ideal therapeutic temperatures.

$LaFe_{11.57}Si_{1.43}$ ingots were prepared in an arc melting furnace in a purified argon atmosphere using elemental La, Fe, and Si. Melt-spinning was carried out using a Cu roller at a tangential wheel speed of 40 ms^{-1}. The melt-spun ribbons were encapsulated in an evacuated quartz tube and annealed at 1323 K for 7.2 ks followed by water quenching. A fully hydrogenated $LaFe_{11.57}Si_{1.43}H_{2.27}$ compound was prepared from the annealed ribbons by subsequent annealing at 523 K for 7.2 ks under a H_2 atmosphere with a pressure of 5 MPa. The hydrogen content of $LaFe_{11.57}Si_{1.43}H_{2.27}$ was adjusted by annealing under a reduced pressure (10^{-2} Pa) for 3.6 ks at temperatures between 473 K and 553 K. The hydrogen content was estimated from the weight loss upon dehydrogenation on a Setaram SenSys Evo thermo-gravimetric analyser. The hysteresis loops were acquired on a vibrating-sample magnetometer (VSM). The heat generation ability of magnetic suspensions in water was investigated using an induction heating system (Ambrell EASYHEAT 0224). The changes in temperature of the magnetic suspensions were monitored as a function of time using an optical thermometer (FLIR Thermacam SC2000) with an accuracy of $\pm$ 1 K. Details of the preparation of magnetic suspensions are given in the supplemental material.[28]

In Fig. 1, we show the temperature dependence of magnetization (M) at 0.4 MA/m for $LaFe_{11.57}Si_{1.43}H_{2.27}$ annealed isochronally for 3.6 ks under a reduced pressure of 10^{-2} Pa. Results for fully hydrogenated $LaFe_{11.57}Si_{1.43}H_{2.27}$ are also shown for comparison. These M-T curves were acquired at a heating rate of 1×10^{-2} K/s. The hydrogen contents of

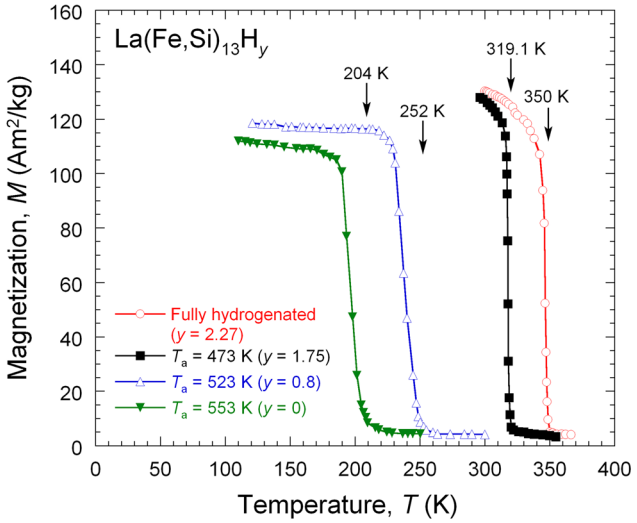

FIG. 1. Temperature dependence of magnetization for $La(Fe,Si)_{13}H_{2.27}$ annealed isochronally for 3.6 ks at annealing temperature (T_a).

annealed samples are shown in this figure. A sharp and abrupt drop of magnetization, characteristic of the 1st order Curie transition, is evident on each of the four M-T curves. The 1st order Curie point estimated from the M-T curves for the $LaFe_{11.57}Si_{1.43}H_y$ ($y = 0$, 0.8, 1.75 and 2.27) alloys varies between 204 K and 350 K, where the ideal therapeutic temperatures (315 to 319 K) for hyperthermia treatments are fully covered. These T_C values agree exceptionally well with those of Lyubina *et al.*[24] for the same alloy system. It is clear that T_C of the La(Fe, Si)$_{13}$ compound is tuneable and an ideal T_C value of 319 K is confirmed for the $LaFe_{11.57}Si_{1.43}H_{1.75}$ compound.

In Fig. 2, we show the magnetization curves of $LaFe_{11.57}Si_{1.43}H_{1.75}$ which were acquired upon heating at a range of temperatures between room temperature and 326 K. The effect of temperature on the M-H curves is limited up to 315 K and the spontaneous magnetization (M_s) remains well above 100 Am^{2}kg^{-1}. However, the magnetization curve at 319 K exhibits a linear M-T relationship, characteristic of a paramagnetic state. This abrupt change in M_s from above

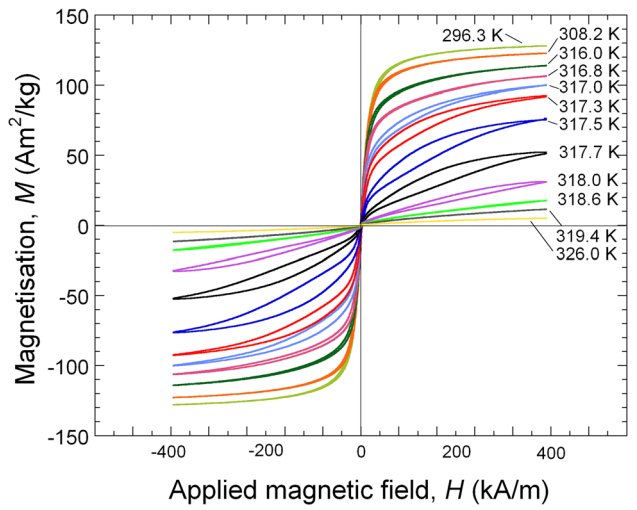

FIG. 2. Magnetization curves of $LaFe_{11.57}Si_{1.43}H_{1.75}$ acquired upon heating at a rate of 1.6×10^{-3} K/s. The Curie point on heating was 319 K.

$100\,Am^2kg^{-1}$ to zero over a very narrow temperature window (3 K) reflects the 1st order magnetic transition. Unlike the 2nd order Curie transition where the ferromagnetic order of spins diminishes cooperatively at a critical temperature, both ferromagnetic and paramagnetic phases may coexist in $La(Fe, Si)_{13}$ near the 1st order Curie point, which has been confirmed experimentally by magneto-optic Kerr microscopy,[26] and the abrupt decrease of M_s is governed by the nucleation-and-growth process of the paramagnetic phase rather than the molecular field in the ferromagnetic phase.

The 1st order magnetic transition in $LaFe_{11.57}Si_{1.43}H_{1.75}$ is also reflected in the shape of the *M-H* curve. The *M-H* curves well below T_C show little irreversibility, whereas the irreversibility is pronounced considerably between 315 and 319 K, immediately below T_C. However, no appreciable increase in the coercivity is evident on the *M-H* curves at these temperatures, indicating that this hysteretic behavior is unrelated to conventional magnetic hardening caused by the magnetic anisotropy. Rather, this enhanced irreversibility near T_C is attributable to the phase coexistence where the ferromagnetic phase is induced in a matrix of the paramagnetic phase by an external magnetic field. It is also known that the ferromagnetic to paramagnetic phase transition in $La(Fe, Si)_{13}$ is accompanied by a large volume change of approximately 1%.[21] Consequently, the nucleation-and-growth process associated with the magnetic phase transition requires stress accommodation, adding an extra irreversibility to the Curie transition. The role of interface strain in determining magnetic hysteresis in a 1st order system has been discussed by Moore *et al.*[29] Although this irreversibility on the *M-H* curve is often problematic when this compound is applied for magnetic refrigeration, this very effect due to the 1st order transition is ideal for enhancing the loss power in the vicinity of T_C.

The static hysteresis loss of $LaFe_{11.57}Si_{1.43}H_{1.75}$ was estimated by integrating the area inside the *M-H* curve. The results are shown in Fig. 3 where the loss is plotted as a function of temperature. A dramatic enhancement of the hysteresis loss from $10 \pm 0.6\,kJm^{-3}$ at 315 K to $80 \pm 5.7\,kJm^{-3}$ at 317.7 K is seen owing to the phase coexistence around T_C.

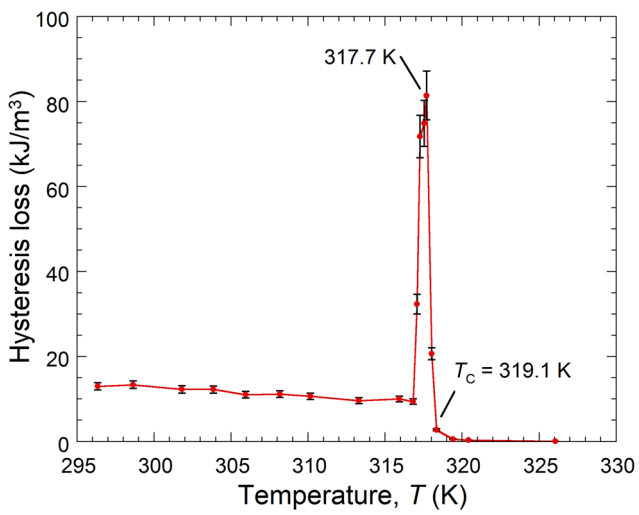

FIG. 3. Change in static hysteresis loss as a function of temperature for $LaFe_{11.57}Si_{1.43}H_{1.75}$.

Moreover, this large hysteresis loss diminishes almost completely at 319 K where the sample becomes paramagnetic. Our hysteresis loss analysis indicates that the energy loss of the $LaFe_{11.57}Si_{1.43}H_{1.75}$ compound is highly dependent on temperature and the expected loss power is maximized just below T_C followed by immediate attenuation of the heating effect at T_C. This unique behavior associated with the 1st order Curie transition is ideal for self-regulated induction heating by an external ac magnetic field.

An increase in the induction heating effect immediately below T_C is expected also for the rotational relaxation process in magnetic fluids. Ondeck *et al.*[30] have modeled the effect of temperature on the rotational relaxation of single-domain particles by incorporating the temperature dependence of intrinsic magnetic properties into a model proposed by Rosensweig.[2] They showed that the heating effect could be enhanced near the Curie temperature because of an increase in susceptibility due to the Hopkinson effect. However, the Hopkinson effect is limited to the conventional 2nd order Curie transition and the anomalous increase in the hysteresis loss observed for $LaFe_{11.57}Si_{1.43}H_{1.75}$ is unrelated to this effect.

The induction heating effect of magnetic particles is often examined by measuring the temperature of water where the magnetic particles are suspended. In the present study, 1 ml of distilled water containing 10 mg of magnetic particles was used. The specific adsorption rate (*SAR*) of magnetic materials is commonly evaluated by

$$SAR = C_s \frac{m_s}{m_m} \left(\frac{dT}{dt}\right)_{t=0}, \tag{1}$$

where C_S is the specific heat capacity of suspension ($4.18\,Jg^{-1}K^{-1}$), dT/dt at $t = 0$ is the initial gradient of the heating curve, m_s and m_m are the specific masses of the suspension and magnetic particles, respectively. The initial gradient in the present study was obtained based on the solution of the relevant heat conduction equation, i.e.,

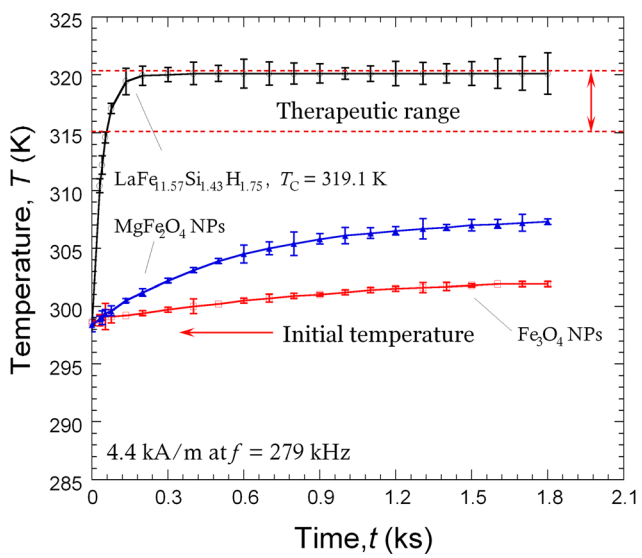

FIG. 4. Change in temperature as a function of induction heating time under $H = 4.4$ kA/m at $f = 279$ kHz for $LaFe_{11.57}Si_{1.43}H_{1.75}$, $MgFe_2O_4$ and Fe_3O_4 suspensions.

TABLE I. Specific absorption rate (*SAR*) and heating properties for $LaFe_{11.57}Si_{1.43}H_{1.75}$, Fe_3O_4 and $MgFe_2O_4$ at $f = 279$ kHz.

Material	H (kAm^{-1})	$H{\cdot}f$ ($\times 10^9$ Am^{-1}s^{-1})	*SAR* (Wg^{-1})	dT/dt (Ks^{-1})	τ (s)
$LaFe_{11.57}Si_{1.43}H_{1.75}$	4.4	1.25	232 ± 10	0.550	39
$LaFe_{11.57}Si_{1.43}H_{1.75}$	8.8	2.5	522 ± 20	1.235	18
$MgFe_2O_4$	4.4	1.25	6.7 ± 0.3	0.015	584
$MgFe_2O_4$	8.8	2.5	19.6 ± 1	0.046	480
Fe_3O_4	4.4	1.25	1.3 ± 0.1	0.003	1045
Fe_3O_4	8.8	2.5	10.7 ± 0.2	0.025	835

$$T(t) = T_0 + \Delta T_{max}[1 - e^{-\frac{t}{\tau}}], \quad (2)$$

where T_0 is the initial temperature of the suspension, $\Delta T_{\max}$ ($T_{\max} - T_0$) is the saturation temperature increase, and τ is the time constant of heating which corresponds to the time when the temperature reaches approximately 63% of $T_{\max}$. The initial gradient dT/dt is defined by the latter two parameters and is given by $\Delta T_{\max}/\tau$.

The heating effect was examined for $LaFe_{11.57}Si_{1.43}H_{1.75}$ ($T_C = 319$ K) along with other conventional materials with the 2nd order Curie transition for comparison. The suspension was kept initially at room temperature and it was then exposed to a constant applied field of 4.4 kAm^{-1} at 279 kHz. Figure 4 shows the relationship between the temperature of the suspension and the exposure time for $LaFe_{11.57}Si_{1.43}H_{1.75}$, commercial Fe_3O_4 and $MgFe_2O_4$ prepared in our laboratory.[31] The temperature of the $LaFe_{11.57}Si_{1.43}H_{1.75}$ suspension shows a dramatic increase with time and reaches the ideal therapeutic range for hyperthermia treatment in less than 60 s. However, despite such a massive heating effect of $LaFe_{11.57}Si_{1.43}H_{1.75}$, the temperature is well regulated at 320 K because of the abrupt attenuation of the loss power at T_C. This is realized by applying a constant ac field, demonstrating the advantage of the abrupt drop of the spontaneous magnetization at the T_C of the 1st order magnetic transition on heating. The magnetic heating effect by the two conventional 2nd order materials is simply insufficient presumably because of the limited spontaneous magnetization. Thus, the temperature of the suspension does not reach the therapeutic range. The anomalous hysteresis loss observed for $LaFe_{11.57}Si_{1.43}H_{1.75}$ appears to be quite effective in generating heat through alternating magnetic fields.

The specific adsorption rate (*SAR*) for $LaFe_{11.57}Si_{1.43}H_{1.75}$, Fe_3O_4 and $MgFe_2O_4$ was estimated from the measured T-t curves through the relationship in Eq. (1). The results are summarized in Table I. In addition to the plots shown in Fig. 4, similar analysis was also carried out at a higher magnetic field of 8.8 kA/m at 279 kHz and the *SAR* values under this heating condition are also included in this table. We limited the highest $H{\cdot}f$ value to 2.5×10^9 Am^{-1}s^{-1} (8.8 kA/m at 279 kHz) in the present study because it has been reported that an animal body could be seriously affected by a $H{\cdot}f$ value of 1.2×10^{10} Am^{-1}s^{-1}.[12] The *SAR* values of $LaFe_{11.57}Si_{1.43}H_{1.75}$ are as high as 200–500 Wg^{-1}, more than an order of magnitude higher than those of Fe_3O_4 and $MgFe_2O_4$. The high *SAR* value of $LaFe_{11.57}Si_{1.43}H_{1.75}$ also stands out when it is compared with literature values for superparamagnetic and ferromagnetic materials. A comprehensive report by Kallumadil *et al.*[10] has shown that the highest intrinsic loss power (*ILP*) among 16 different commercial superparamagnetic materials is 3.12×10^{-12} Hm^{2}g^{-1}, equivalent to a *SAR* value of 67 Wg^{-1} at $H{\cdot}f = 2.5 \times 10^9$ Am^{-1}s^{-1}. Kita *et al.*[8] has realized high *SAR* values up to 420 Wg^{-1} by employing ferromagnetic Co-doped iron oxide nanoparticles, a 2nd order ferromagnet with T_C at about 800 K. However, the *SAR* value at $H{\cdot}f = 2.5 \times 10^9$ Am^{-1}s^{-1} was approximately 180 Wg^{-1}. Hence, the *SAR* value for $LaFe_{11.57}Si_{1.43}H_{1.75}$ (522 ± 20 Wg^{-1}) appears to be exceptional. Furthermore, unlike $LaFe_{11.57}Si_{1.43}H_{1.75}$, none of these conventional superparamagnetic or ferromagnetic materials with relatively large *SAR* exhibits a self-regulated heating effect within the therapeutic temperature range. The *SAR* value confirmed under a biocompatible magnetic field for the 1st order $LaFe_{11.57}Si_{1.43}H_{1.75}$ alloy is close to the required minimum heating effect ($\sim$1 kWg^{-1}) for *in-vivo* hyperthermia applications, demonstrating the effectiveness of utilizing the 1st order Curie transition for magnetic induction heating.

Warburg[32] is widely credited with discovering the heat dissipation associated with magnetic hysteresis. However, many articles in the magnetocaloric literature have incorrectly credited him with having also first observed the magnetocaloric effect,[33] i.e., an isothermal entropy change or adiabatic temperature change induced by a pseudo-static magnetic field. In this article, we have brought the research fields of magnetocaloric materials and hysteretic dissipation together, utilizing Warburg's hysteresis in the region of phase coexistence around the Curie temperature of a first order magnetocaloric material. Our study confirms that the extraordinary loss power diminishes very abruptly at the Curie point on heating. This leads to a unique self-regulated heating effect with a massive loss power which is unmatched by other conventional ferromagnetic materials. This proof-of-concept study concludes that the extraordinary heating effect near the first-order Curie point opens a novel alloy design strategy for realizing *in-vivo* hyperthermia therapy.

[1]R. K. Gilchrist, R. Medal, W. D. Shorey, R. C. Hanselman, J. C. Parrott, and C. B. Taylor, Ann. Surg. **146**, 596 (1957).
[2]R. E. Rosensweig, J. Magn. Magn. Mater. **252**, 370 (2002).
[3]Q. A. Pankhurst, J. Connolly, S. K. Jones, and J. Dobson, J. Phys. D **36**, R167 (2003).
[4]A. H. Habib, C. L. Ondeck, P. Chaudhary, M. R. Bockstaller, and M. E. McHenry, J. Appl. Phys. **103**, 07A307 (2008).
[5]R. Hergt and S. Dutz, J. Magn. Magn. Mater. **311**, 187 (2007).
[6]S. Wada, K. Tazawa, I. Furuta, and H. Nagae, Oral Dis. **9**, 218 (2003).
[7]A. Jordan, R. Scholz, K. Maier-Hauff, F. K. H. van Landeghem, N. Waldoefner, U. Teichgraeber, J. Pinkernelle, H. Bruhn, F. Neumann, B. Thiesen, A. von Deimling, and R. Felix, J. Neuro-Oncol. **078**, 7 (2006).
[8]E. Kita1, T. Oda, T. Kayano, S. Sato, M. Minagawa, H. Yanagihara, M. Kishimoto, C. Mitsumata, S. Hashimoto, K. Yamada, and N. Ohkohchi, J. Phys. D: Appl. Phys. **43**, 474011 (2010).

[9]See for example, E. A. Périgo, F. A. Sampaio, and M. F. de Campos, Appl. Phys. Lett. **103**, 264107 (2013).
[10]M. Kallumadil, M. Tada, T. Nakagawa, M. Abe, P. Southern, and Q. A. Pankhurst, J. Magn. Magn. Mater. **321**, 1509 (2009).
[11]J. H. Lee, J. T. Jang, J. S. Choi, S. H. Moon, S. H. Noh, J. W. Kim, J. G. Kim, I. S. Kim, K. I. Park, and J. Cheon, Nat. Nanotechnol. **6**, 418 (2011).
[12]R. Ivkov, S. J. DeNardo, W. Daum, A. R. Foreman, R. C. Goldstein, V. S. Nemkov, and G. L. DeNardo, Clin. Cancer Res. **11**, 7093S (2005).
[13]M. Jeun, S. Lee, J. K. Kang, A. Tomitaka, K. W. Kang, Y. I. Kim, Y. Takemura, K. W. Chung, J. Kwak, and S. Bae, Appl. Phys. Lett. **100**, 092406 (2012).
[14]I. Brezovich, W. J. Atkinson, and D. P. Chakraborty, Med. Phys. **11**, 145 (1984).
[15]H. G. Bagaria, J. L. Phillips, D. E. Nikles, and D. T. Johnson, in *Proceedings* of the *AICHE Annual Meeting Conference* (AICHE, 2005), p. 14336.
[16]T. Shimizu, H. Asano, and M. Matsui, J. Magn. Magn. Mater. **310**, 1835 (2007).
[17]E. Pollert, K. Knížek, M. Maryško, P. Kašpar, S. Vasseur, and E. Duguet, J. Magn. Magn. Mater. **316**, 122 (2007).
[18]Provided that the spontaneous magnetization simply follows the Brillouin function, $S = 1/2$ and $T_C = 315$ K leads to a normalized spontaneous magnetization $[M_s/M_s(0)]$ of 0.22 at 310 K.
[19]F. X. Hu, B. G. Shen, J. R. Sun, Z. H. Cheng, G. H. Rao, and X. X. Zhang, Appl. Phys. Lett. **78**, 3675 (2001).
[20]S. Fujieda, A. Fujita, K. Fukamichi, Y. Yamazaki, and Y. Iijima, Appl. Phys. Lett. **79**, 653 (2001).
[21]A. Fujita, S. Fujieda, K. Fukamichi, H. Mitamura, and T. Goto, Phys. Rev. B **65**, 014410 (2001).
[22]S. Fujieda, A. Fujita, and K. Fukamichi, Appl. Phys. Lett. **81**, 1276 (2002).
[23]A. Fujita, S. Fujieda, Y. Hasegawa, and K. Fukamichi, Phys. Rev. B **67**, 104416 (2003).
[24]J. Lyubina, O. Gutfleisch, M. D. Kuz'min, and M. Richter, J. Magn. Magn. Mater. **320**, 2252 (2008).
[25]J. D. Moore, K. Morrison, K. G. Sandeman, M. Katter, and L. F. Cohen, Appl. Phys. Lett. **95**, 252504 (2009).
[26]J. Lyubina, R. Schäfer, N. Martin, L. Schultz, and O. Gutfleisch, Adv. Mater. **22**, 3735 (2010).
[27]L. F. Bao, F. X. Hu, L. Chen, J. Wang, J. R. Sun, and B. G. Shen, Appl. Phys. Lett. **101**, 162406 (2012).
[28]See supplementary material at http://dx.doi.org/10.1063/1.4900557 for preparation of magnetic suspensions.
[29]J. D. Moore, G. K. Perkins, Y. Bugoslavsky, M. K. Chattopadhyay, S. B. Roy, P. Chaddah, V. K. Pecharsky, K. A. Gschneidner, Jr., and L. F. Cohen, Appl. Phys. Lett. **88**, 072501 (2006).
[30]C. L. Ondeck, A. H. Habib, P. Ohodnicki, K. Miller, C. A. Sawyer, P. Chaudhary, and M. E. McHenry, J. Appl. Phys. **105**, 07B324 (2009).
[31]M. R. Barati, C. Selomulya, and K. Suzuki, J. Appl. Phys. **115**, 17B522 (2014).
[32]E. Warburg, Ann. Phys. **249**, 141 (1881).
[33]A. Smith, Eur. Phys. J. **H38**, 507 (2013).