Contribution of energy-gap in the ferromagnetic spin–wave spectrum on magnetocaloric parameters of $CeRu_2Ge_2$

2009 J. Phys.: Condens. Matter 21 026010

(http://iopscience.iop.org/0953-8984/21/2/026010)

J. Phys.: Condens. Matter **21** (2009) 026010 (4pp) doi:10.1088/0953-8984/21/2/026010

Contribution of energy-gap in the ferromagnetic spin–wave spectrum on magnetocaloric parameters of $CeRu_2Ge_2$

Tapas Samanta, I Das and S Banerjee

Saha Institute of Nuclear Physics, 1/AF, Bidhannagar, Kolkata 700 064, India

E-mail: tapas.samanta@saha.ac.in and indranil.das@saha.ac.in

Received 1 September 2008
Published 9 December 2008
Online at stacks.iop.org/JPhysCM/21/026010

Abstract

A study of the magnetocaloric effect has been performed on a polycrystalline $CeRu_2Ge_2$ compound, which exhibits an antiferromagnetic ordering below $T_N = 8.3$ K and enters into a ferromagnetic ground state at $T_C = 7.4$ K. The origins of the magnetocaloric parameters (the isothermal entropy change: $-\Delta S$ and the adiabatic temperature change: ΔT_{ad}) of the $CeRu_2Ge_2$ compound below T_C have been analyzed. A sharp decrease in $-\Delta S$ has been observed below T_C. However, the ΔT_{ad} does not fall as sharply as $-\Delta S$ with decreasing temperature in the corresponding temperature region. This behavior results in an additional value of ΔT_{ad} at low temperature, which originates from the exponential decrease of the magnetic contribution of specific heat associated with an increase of energy-gap in the ferromagnetic spin–wave spectrum with the application of magnetic field.

(Some figures in this article are in colour only in the electronic version)

The magnetocaloric effect (MCE) is associated with the change of temperature of magnetic materials with the application of external magnetic field under adiabatic conditions. Since the discovery of giant MCE near room temperature [1], the study of MCE is an intriguing field of research due to its potential application in energy-efficient environment-friendly cooling technology. The main focus in such studies is to find out new materials with large MCE, which can be applicable in different temperature regions from room temperature to liquid He temperature [1–10]. Besides its technological importance, the MCE can provide valuable information about magnetic materials, for example the nature of magnetic ordering [9], magnetic field-induced transitions [6, 7], major contribution to magnetoresistance [11] etc.

The adiabatic temperature change (ΔT_{ad}) and the isothermal entropy change ($-\Delta S$) are the two parameters which describe the MCE for given magnetic materials. From the physics point of view, the comprehensive analysis of both $-\Delta S$ and ΔT_{ad} is important. The total entropy of a magnetic material with well localized magnetic moments consists of magnetic, lattice and electronic contributions. Usually the changes of lattice and electronic entropies are independent of magnetic field. In that situation, with the application of magnetic field, the magnetic moments of ferromagnetic materials become oriented along the magnetic field direction and consequently the total entropy decreases due to a decrease of magnetic entropy. According to the laws of thermodynamics, the total entropy of the system remains constant under the condition of adiabatic changes of magnetic field. To fulfil the above-mentioned condition, the decrease of magnetic entropy is compensated by the increase of combined lattice and electronic entropies, which results in the increase of sample temperature. Lots of studies on MCE based upon ferromagnetic intermetallic compounds have been highlighted in the literature. However, there are hardly any studies on MCE of magnetic materials associated with an energy-gap (Δ) in the ferromagnetic spin–wave spectrum, such as $CeRu_2Ge_2$, where the value of Δ has been reported to be modified with the application of magnetic field below T_C [12, 13]. This change of Δ and consequently the change of magnetic contribution of specific heat in the presence of magnetic field may have some influence both on $-\Delta S$ and ΔT_{ad}. Keeping this idea in mind, we have studied the magnetocaloric properties of the $CeRu_2Ge_2$ compound highlighting the ferromagnetic region. In this present work, the main objective is to find out the contribution of energy-gap in the ferromagnetic spin–wave

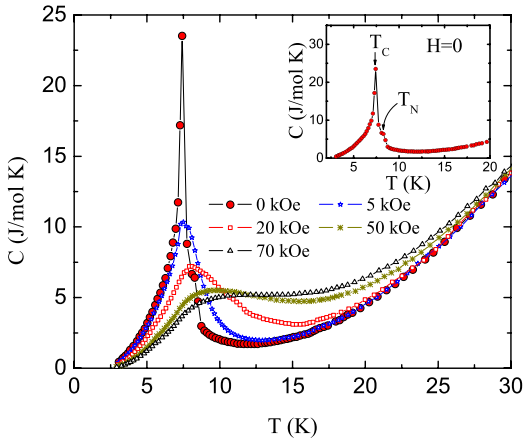

Figure 1. Heat capacity (C) as a function of temperature for $CeRu_2Ge_2$ in the presence of different constant magnetic fields. Inset: zero-field heat capacity as a function of temperature. Arrows indicate the transition temperatures.

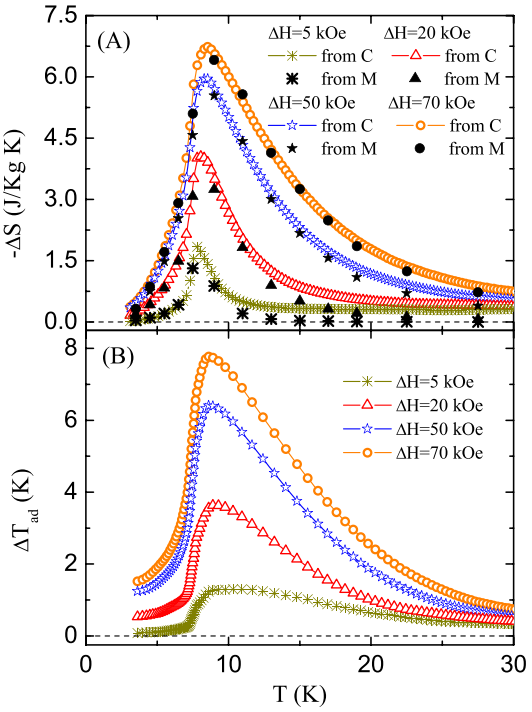

Figure 2. (A) Isothermal entropy change ($-\Delta S$) of $CeRu_2Ge_2$ as a function of temperature for different ΔH derived both from heat capacity (C) and magnetization (M) data. (B) Temperature dependence of the adiabatic temperature change (ΔT_{ad}) of $CeRu_2Ge_2$ for different ΔH calculated from C data.

spectrum on the magnetocaloric parameters ($-\Delta S$ and ΔT_{ad}) in $CeRu_2Ge_2$.

We prepared the ternary polycrystalline samples by arc melting of constituent elements of purity better than 99.9% in argon atmosphere followed by annealing at 800 °C for one week in an evacuated sealed quartz tube. The crystallographic phase purity of the $CeRu_2Ge_2$ compound has been confirmed by x-ray diffraction patterns, which crystallize in the body-centered tetragonal $ThCr_2Si_2$ structure as reported earlier [14]. The magnetization measurements were carried out employing a superconducting interference device (SQUID) magnetometer within the temperature range 3–40 K and for applied magnetic field up to 70 kOe. The specific heat (C) measurements were performed within the temperature interval of 3–30 K in the absence as well as in the presence of 5, 20, 50 and 70 kOe magnetic fields using the semiadiabatic heat-pulse method.

The heat capacity (C) of $CeRu_2Ge_2$ as a function of temperature for various constant magnetic fields is plotted in figure 1. Two closely spaced magnetic phase transitions at $T_N = 8.3$ K and $T_C = 7.4$ K are clearly visible in the zero-field $C(T)$ data (shown in the inset of figure 1 by arrows) and are in good agreement with the previous results [14, 15]. The sharp peak at T_C in the zero-field $C(T)$ curve is due to a first order transition from antiferromagnetic to ferromagnetic phase with decreasing temperature. The signature of the transition point at the antiferromagnetic to ferromagnetic state vanishes in the presence of higher (even for 5 kOe) external magnetic field, only the peak corresponding to the ferromagnetic transition is visible with lower amplitude. Earlier reports suggest that the low temperature specific heat of $CeRu_2Ge_2$ is dominated by the ferromagnetic spin–wave spectrum with an energy-gap (Δ/k_B) of order 10 K [12–15]. Because of the presence of an energy-gap, the magnetic contribution of specific heat (C_M) falls off exponentially at low temperature below T_C. The expression of C at low temperature can be described as [12]

$$C = C_E + C_M = \gamma T + \beta T^{3/2} e^{-(\frac{\Delta}{k_B T})} \quad (1)$$

where C_E ($= \gamma T$) is the electronic heat capacity. From the fitting of low temperature $C(T)$ data by keeping the same γ value as mentioned in [12] for the corresponding H (the γ value for 40 kOe magnetic field as mentioned in [12] was considered as the same for 50 kOe data in our case), it was found that the values of Δ are enhanced with increasing H. Simultaneously, the coefficient of the magnetic contribution (β) of C decreases with increasing H.

The thermal variation of isothermal magnetic entropy change ($-\Delta S$) for different magnetic field changes (ΔH) is shown in figure 2(A), which has been deduced from both magnetization and specific heat data. From magnetization isotherms, the values of $-\Delta S$ have been estimated using Maxwell's thermodynamic relation $(\partial S/\partial H)_T = (\partial M/\partial T)_H$ whereas the values of $-\Delta S$ have been calculated from specific heat data (shown in figure 1) taking the difference isothermally between the two entropy curves measured in the presence as well as in the absence of magnetic field. The temperature dependence of the entropy (S) curve was obtained from $C(T)$ data at the corresponding magnetic field using the relation $S = \int_0^T \frac{C}{T} dT$. The values of $-\Delta S$ for different ΔH evaluated from both magnetization and specific heat data are similar to each other, signifying that the isothermal entropy change is purely magnetic in origin. The plot of $-\Delta S$ (T) shows a positive caret-like shape with maxima around the ferromagnetic ordering temperature (T_C) for all ΔH. However, no anomaly at T_N has been observed in the $-\Delta S(T)$ curve because of the fact that the antiferromagnetic ordering vanishes with the application of less than 5 kOe magnetic field [14].

The above observation indicates that the $CeRu_2Ge_2$ compound exhibits only ferromagnetic ordering for the field values $H \geqslant 5$ kOe [16], there is no signature of antiferromagnetic ordering. A fast reduction in the values of $-\Delta S(T)$ have been observed below T_C with decreasing temperature and the decreases are sharper for higher ΔH. This feature can be explained considering the modification in the value of Δ in the presence of magnetic field. The value of the energy-gap ($\Delta/k_B \sim 10$ K) at the ferromagnetic spin–wave spectrum is larger than that of the T_C (~7.4 K) of $CeRu_2Ge_2$ and the magnitude of Δ increases with the enhancement of magnetic field. This implies that the thermal fluctuation effect on the orientation of magnetic moments (or thermal effect on spin–wave excitation) is very much less at low temperature below T_C for $CeRu_2Ge_2$, which also reduces further with increasing magnetic field due to enhancement in the magnitude of Δ. As a result, the change of entropy (or relative orientation of magnetic moments) with the application of magnetic field is very small in comparison to the zero-field as the temperature reduces further below T_C due to quenching of ferromagnetic spin–wave excitation (or freezing of magnetic moments) by the application of magnetic field. Therefore, the change of $-\Delta S(T)$ is substantial in a narrow temperature region just below T_C.

The adiabatic temperature change (ΔT_{ad}) for different ΔH is plotted in figure 2(B), which has been derived from specific heat data (shown in figure 1) taking the difference between the two entropy curves from zero-field to in-field isentropically. The plot of $\Delta T_{ad}(T)$ shows a maxima around T_C and it is positive in the entire temperature range under investigation for all ΔH. The above-mentioned feature is an indication of ferromagnetic ordering in $CeRu_2Ge_2$ for the field values $H \geqslant 5$ kOe [16]. The peak values of ΔT_{ad} are 1.3, 3.6, 6.4 and 7.8 K for $\Delta H = 5$, 20, 50 and 70 kOe respectively. The observed ΔT_{ad} values of $CeRu_2Ge_2$ are reasonably large, although the moment value of the Ce^{3+}-ion is the lowest among the rare-earth family. There is a distinct difference between $\Delta T_{ad}(T)$ and $-\Delta S(T)$ in the case of $CeRu_2Ge_2$ at low temperatures below T_C. Apart from a sharp reduction as $-\Delta S(T)$ at low temperature, the decrease of $\Delta T_{ad}(T)$ slows down with decreasing temperature, resulting in an extra value of ΔT_{ad}, whenever compared with $-\Delta S$. A possible explanation of the above-mentioned dissimilarity has been discussed as follows.

According to Pecharsky *et al* [17], the temperature dependence of adiabatic temperature change [ΔT_{ad}] is related to the isothermal magnetic entropy change [ΔS] as

$$\Delta T_{ad}(T, \Delta H) = -\frac{T}{C_{H2}(H, T)} \Delta S(T, \Delta H) \qquad (2)$$

where C_{H2} is the total heat capacity for the final applied magnetic field. This is quite clear from equation (2) that for given $-\Delta S(T)$, the value of $C_{H2}(T)$ plays a crucial role in obtaining the value of $\Delta T_{ad}(T)$. The total heat capacity of magnetic materials is expressed as $C_{tot} = C_E + C_L + C_M$, where C_E ($= \gamma T$), C_L($= \delta T^3$) and C_M (expression is shown previously) are electronic, lattice and magnetic contribution of heat capacity respectively. Previously, it has been discussed that the value of C_M in the case of $CeRu_2Ge_2$

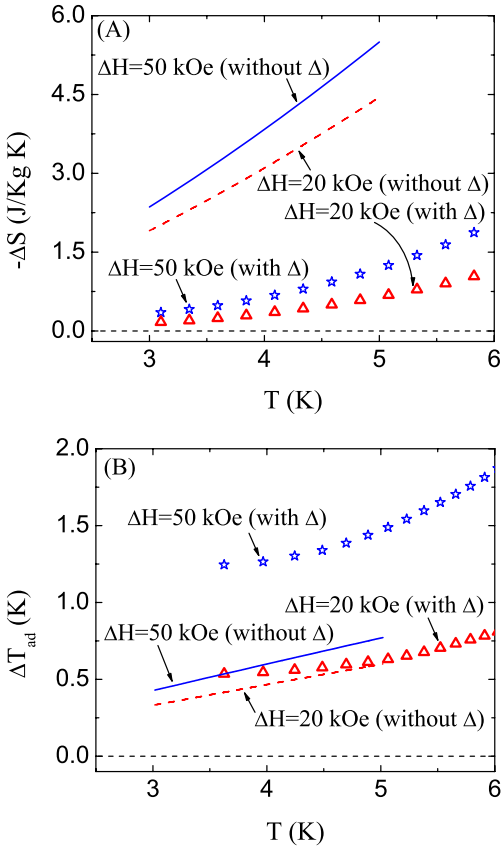

Figure 3. (A) A plot of $-\Delta S(T)$ including energy-gap Δ in a ferromagnetic spin–wave spectrum (shown by symbols) and excluding Δ (shown by lines) for $\Delta H = 20$ and 50 kOe. (B) Temperature dependence of ΔT_{ad} including Δ (shown by symbols) and excluding Δ (shown by lines) for $\Delta H = 20$ and 50 kOe.

decreases exponentially with Δ at low temperature below T_C due to enhancement of Δ with the increase of magnetic field. However, the change of C_E with the application of magnetic field is not substantial in comparison with C_M and the lattice heat capacity (C_L) is also very small as well as being independent of magnetic field at low temperature. Therefore it appears that the domination of exponential decrease in C_M at low temperature, due to the increase of energy-gap in the ferromagnetic spin–wave spectrum with the application of higher magnetic field, is the cause of the observed difference between $\Delta T_{ad}(T)$ and $-\Delta S(T)$ in the case of $CeRu_2Ge_2$.

A further analysis has been made for better clarification of the observed difference between $\Delta T_{ad}(T)$ and $-\Delta S(T)$ at low temperature below T_C. The experimental C data were fitted using equation (1) within the temperature range 3–5 K for 0, 20 and 50 kOe magnetic fields. Previously, it has been mentioned that the exponential factor in C_M is due to the presence of an energy-gap in the ferromagnetic spin–wave spectrum. To obtain a spin–wave energy-gap independent total heat capacity (denoted as $C_{-\Delta}$), we have regenerated the $C_{-\Delta}$ excluding the exponential factor in the C_M term of equation (1). During the evaluation of $C_{-\Delta}$, the values of γ and β of the corresponding magnetic fields obtained from the fitting of experimental C data have been kept constant. After that, the

spin–wave energy-gap independent magnetocaloric parameters have been deduced by taking $C_{-\Delta}$ data in a similar way to that described previously. It appears that the enhanced value of ΔT_{ad} at low temperature is reduced by eliminating the spin–wave energy-gap (shown in figure 3(B)), which establishes our idea mentioned earlier. Most interestingly, the value of $-\Delta S$ is enhanced without taking into account the contribution of the energy-gap in the ferromagnetic spin–wave spectrum (shown in figure 3(A)), which is in contradiction to the behavior of ΔT_{ad} at low temperature.

The influence of energy-gap in the ferromagnetic spin–wave spectrum on magnetocaloric parameters in the $CeRu_2Ge_2$ compound has been investigated below T_C. An enhancement (reduction) in the value of ΔT_{ad} $(-\Delta S)$, associated with the increase of Δ in the presence of magnetic field, has been observed at low temperature below T_C. Our observation indicates that the modification of the energy-gap in the ferromagnetic spin–wave spectrum with the application of external magnetic field plays a significant role at low temperature below T_C concerning the values of the magnetocaloric parameters.

References

[1] Pecharsky V K and Gschneidner K A Jr 1997 *Phys. Rev. Lett.* **78** 4494

[2] Gschneidner K A Jr, Pecharsky V K and Tsokol A O 2005 *Rep. Prog. Phys.* **68** 1479

[3] Tishin A M 1999 *Handbook of Magnetic Materials* vol 12, ed K H J Buschow (Amsterdam: North-Holland) p 395

[4] Pecharsky V K and Gschneidner K A 1999 *J. Magn. Magn. Mater.* **200** 44

[5] Phan M H and Yu S C 2007 *J. Magn. Magn. Mater.* **308** 325

[6] Samanta T, Das I and Banerjee S 2007 *Appl. Phys. Lett.* **91** 082511

[7] Samanta T, Das I and Banerjee S 2007 *Appl. Phys. Lett.* **91** 152506

[8] Biswas A, Samanta T, Banerjee S and Das I 2008 *J. Appl. Phys.* **103** 013912

[9] Biswas A, Samanta T, Banerjee S and Das I 2008 *Appl. Phys. Lett.* **92** 212502

[10] Biswas A, Samanta T, Banerjee S and Das I 2008 *Appl. Phys. Lett.* **92** 012502

[11] Samanta T and Das I 2006 *Phys. Rev.* B **74** 132405

[12] Bohm A, Caspary R, Habel U, Pawlak L, Zuber A, Steglich F and Loidl A 1988 *J. Magn. Magn. Mater.* **76/77** 150

[13] Fontes M B, Bud'ko S L, Continentino M A and Baggio-Saitovitch E M 1999 *Physica* B **270** 255

[14] Wilhelm H, Alami-Yadri K, Revaz B and Jaccard D 1999 *Phys. Rev.* B **59** 356

[15] Raymond S, Haen P, Calemczuk R, Kambe S, Fak B, Lejay P, Fukuhara T and Flouquet J 1999 *J. Phys.: Condens. Matter* **11** 5547

[16] Tishin A M, Gschneidner K A Jr and Pecharsky V K 1999 *Phys. Rev.* B **59** 503

[17] Pecharsky V K, Gschneidner K A Jr, Pecharsky A O and Tishin A M 2001 *Phys. Rev.* B **64** 144406