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Magnetocaloric effect and critical behavior in Pr0.5Sr0.5MnO3: An analysis on the validity of Maxwell relation and nature of phase transitions.

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Magnetocaloric effect (MCE) is an intrinsic property of magnetic materials. It is characterized by an isothermal change in magnetic entropy (ΔS_M) or an adiabatic change in temperature (ΔT_{ad}) of the material subject to a dc magnetic field. The MCE forms the basis for development of solid-state magnetic refrigeration (MR) technology, which attracts global interest owing to its energy-efficient and environment-friendly advantages over existing gas compression-expansion refrigeration techniques [1]. The majority of MR is to find suitable magnetocaloric materials that are cost-effective and exhibit large ΔSM spanning over a wide temperature range, namely, the large refrigerant capacity (RC) [2]. It has been reported that large MCE can be achieved in both first- and second-order magnetic transition materials [1,2]. While the first-order magnetic transition (FOMT) often induces a larger ΔS_M, it is restricted to a narrower temperature range resulting in a smaller RC [3]. The second-order magnetic transition (SOMT) induces a smaller ΔS_M but with a distribution over a broader temperature range, thus resulting in a larger RC. Research in this field is therefore focused on exploring magnetocaloric composite materials that possess large ΔS_M and undergo multiple successive SOMTs.
To evaluate MCE in a magnetocaloric material, one usually calculates ΔS_M from magnetic field-dependent magnetization (M-H) curves using the Maxwell relation, due to its simplicity and convenience. While this method has been validated over the years for determining ΔS_M of SOMT materials [1,2], its applicability to FOMT materials still remains under debate [4-6]. While previous studies were focused mainly on magnetocaloric materials with a first-order PM-FM transition, an interesting question emerges regarding the validity of using the Maxwell relation for calculation of ΔS_M in magnetocaloric materials with a first-order ferromagnetic to antiferromagnetic (FM-AFM) transition. A recent study has suggested that there exists a correlation between the MCE and critical exponents near the SOMT in magnetocaloric materials [7]. However, no effort has been devoted to studying the effect of phase coexistence on MCE and critical exponents in mixed-phase materials.
To address these outstanding issues, we have systematically characterized the MCE in phase-separated manganites of $Pr_{0.5}Sr_{0.5}MnO_3$ (PSMO) by means of the Maxwell relation and the Clausius-Clapeyron equation. PSMO serves as an excellent model system for the purpose of our study, as it undergoes a high temperature second-order paramagnetic to ferromagnetic (PM-FM) transition at ~245 K followed by a low temperature first-order ferromagnetic to antiferromagnetic (FM-AFM) transition ~165 K. In this study, magnetic measurements were performed using a commercial Physical Property Measurement System (PPMS) from Quantum Design in the temperature range of 5 to 300 K at applied fields up to 7 T. The magnetization isotherms (M-H) were measured with a field step of 0.05 mT in the range of 0-5 T and with a temperature interval of 3 K over a temperature range of 5 to 300 K. Our experimental and theoretical analyses reveal that around the second-order PM-FM transition the ΔS_M can be precisely determined from isothermal M-H curves using the Maxwell relationship, but around the first-order FM-AFM transition values of ΔS_M calculated by the Maxwell relation largely differ from those calculated by the Clausius-Clapeyron equation at the magnetic field range where a conversion between the AFM and FM phases occurs. A systematic analysis of the critical behavior near the SOMT using a non-iterative method has suggested the existence of some ferromagnetic correlations (e.g. the possible presence of ferromagnetic clusters) at temperatures above the T_C and its influence on the critical exponents and MCE. Our studies provide important insights into the validity of the Maxwell relation for calculating MCE from magnetization measurements in magnetocaloric materials with a first-order magnetic transition and a correlation between the MCE and critical exponents near the second-order magnetic transition in phase-separated materials.

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[1] V.K. Pecharsky, K.A. Gschneidner, A.O. Tsokol, Rep. Prog. Phys. 68, 1479 (2005).
[2] M.H. Phan, S.C. Yu, J. Magn. Magn. Mater. 308, 325 (2007).
[3] N.S. Bingham, M.H. Phan, H. Srikanth, M.A. Torija, and C. Leighton, J. Appl. Phys. 106, 023909 (2009).
[4] G.J. Liu, J.R. Sun, J. Shen, B. Gao, H.W. Zhang, F.X. Hu, and B. G. Shen, Appl. Phys. Lett. 90, 032507 (2007).
[5] J.S. Amaral and V.S. Amaral, Appl. Phys. Lett. 94, 042506 (2009).
[6] A. Magnus G. Carvalho, A.A. Coelho, P.J. von Ranke, and C.S. Alves, J. All. Compd. 509, 3452 (2011).
[7] M.H. Phan, V. Franco, A. Chaturvedi, S. Stefanoski, G.S. Nolas, and H. Srikanth, Physics Review B 84, 054436 (2011).