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Magnetoelastic coupling influence on the magnetocaloric effect in ferromagnetic materials

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Abstract

The Landau theory for phase transitions framework is applied to describe the magnetocaloric effect in ferromagnetic systems with magnetoelastic and magnetoelectronic couplings. The M^4 coefficient in the energy expansion, whose signal determines the order of the phase transition, and its temperature dependence, are shown to be of crucial importance. The existence of a broad magnetic entropy peak above T_c is related to these couplings.

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The interest on the research of magnetocaloric cooling systems has raised considerably in recent years due to its potential impact on environmental concerns and energy saving. An increased research effort on magnetocaloric studies has revealed new materials with large magnetic entropy changes under an applied magnetic field near magnetic phase transitions, such as $Gd_5(Si_{2-x}Ge_{2+x})$ [1], $MnFeP_{1-x}As_x$ [2], $LaFe_{13-x}Si_x$ [3] and manganites [4]. One of the most interesting features of these new materials is the possibility of tuning the operation range (i.e., the Curie temperature) by suitable chemical composition adjustment.

In previous studies [5–7], the magnetic properties of manganites, with particular emphasis on the paramagnetic phase, were interpreted using the Landau theory of phase transitions. The anomalous upturns of the $M(H)$ curves observed above T_c (inset in Fig. 1) were related to the additional contribution of magnetoelastic couplings and electron condensation energy (metal–insulator transition), shown to directly affect the coefficient B in the Landau theory magnetic energy expansion [8]:

$$G(T,M) = G_0 + \tfrac{1}{2}AM^2 + \tfrac{1}{4}BM^4 + \tfrac{1}{6}CM^6 - M.H. \quad (1)$$

To account for first (for $B<0$) and second order (for $B>0$) phase transitions the expansion includes up to the sixth power in M. The coefficients A, B and C depend on temperature. Those additional interactions decrease B from the regular positive values found in normal ferromagnets to negative values, changing the order of the phase transition. A similar approach was also proposed for the spin fluctuation model applied to the itinerant electron metamagnetism of RCo_2 intermetallic compounds [9].

From energy minimization, a magnetic equation of state is derived within this theory

$$\frac{H}{M} = A + BM^2 + CM^4 \quad (2)$$

and the corresponding magnetic entropy is obtained from differentiation of the magnetic part of the free energy with respect to temperature

$$S_M(T,H) = -\tfrac{1}{2}A'(T)M^2 - \tfrac{1}{4}B'(T)M^4 - \tfrac{1}{6}C'(T)M^6. \quad (3)$$

$A'(T)$, $B'(T)$ and $C'(T)$ are the temperature derivatives of the expansion coefficients. The same result is obtained using the equation of state and integraton of Maxwell relations. For a simple ferromagnet, with B and C constant and positive, $S_M(T,H)$ presents a narrow peak at T_c. The additional magnetoelastic and electron

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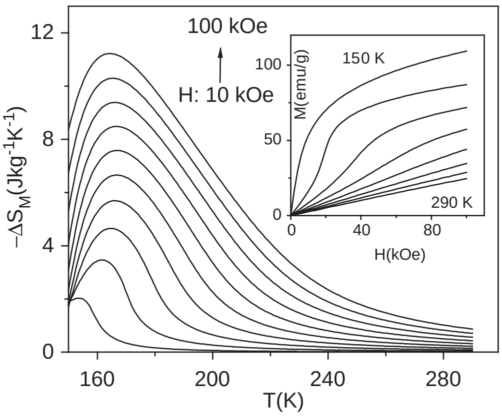

Fig. 1. Magnetic entropy change for $La_{0.60}Y_{0.07}Ca_{0.33}MnO_3$ manganite. Inset: Field dependence of magnetization.

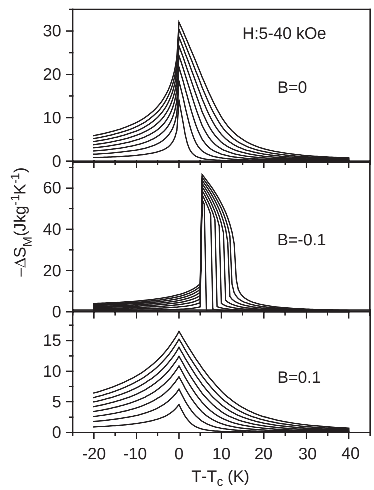

Fig. 2. ΔS_M temperature dependence for constant B coefficient.

interactions contribute directly to the magnetic entropy and its temperature dependence: if B is an increasing function of temperature ($B' > 0$), the magnetic entropy decrease under an applied magnetic field will be larger, and the peak is broadened. This effect is illustrated in Fig. 1 for the sample $La_{0.60}Y_{0.07}Ca_{0.33}MnO_3$ with $T_c \sim 150\,K$. Using the temperature dependence of the expansion coefficients determined by fitting the measured magnetization to the equation of state (Eq. (2)) [5–7], the magnetic entropy change $\Delta S_M(T,H) = S_M(T,H) - S_M(T,0)$ is calculated. An interesting additional effect is the shift of the maximum entropy change to temperatures above T_c, in close connection with the magnetization upturns.

The effect of the additional interactions on the magnetic entropy through the B coefficient was modeled using the Landau expansion. Representative results are shown in Figs. 2 and 3 respectively, for constant B or constant B' (with $B = 0$ at T_c). $A(T)$ was taken as linear

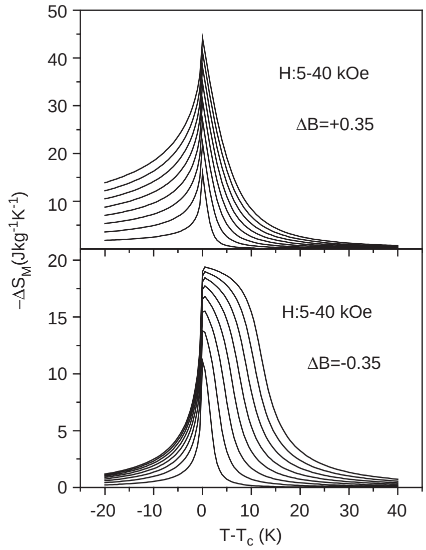

Fig. 3. ΔS_M temperature dependence for positive and negative B temperature derivative. ΔB is the change in the temperature interval displayed.

vs. T–T_c with slope 67 gOe/emu. C was made constant: $5 \times 10^{-6}\,g^5Oe/emu^5$. B values are in g^3Oe/emu^3. The direct effect of B' on ΔS_M is evidenced in Fig. 3: Positive B' leads to higher ΔS_M peak values. These dependences (particularly when B is negative above T_c), are in close agreement with experimental results in many systems, where a broad region of fairly high values is found: RCo_2 [10] and LaFeSi metamagnets [3,11], MnAs based systems [2] or manganites. Finally, we mention that the Landau equation of state (2) provides a convenient numerical interpolation scheme to treat magnetization data in order to determine the entropy change.

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