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Barocaloric and magnetocaloric effects in La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$

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In this paper we calculate the barocaloric and the magnetocaloric effects under applied pressure in the compound La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$. We use a microscopical model in the framework of the band theory of magnetism, where the Coulomb interaction between itinerant electrons is treated in the mean field approach. Our theoretical calculations point out that both the barocaloric potentials $[\Delta S]_B$ and $[\Delta T_{ad}]_B$ at fixed magnetic field, and the magnetocaloric potentials ΔS and ΔT_{ad} under simultaneous variation of magnetic field and pressure, reach large values in a wide range of temperatures. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2938841]

I. INTRODUCTION

The magnetocaloric effect[1,2] occurs basically due to the entropy change upon variation of the applied magnetic field. This effect is characterized by the entropy change (ΔS) in an isothermal process and by the temperature change (ΔT_{ad}) in an adiabatic process. Experimental works[3–8] show that the isothermal entropy changes in metallic compounds undergoing a first order phase transition, such as Gd$_5$Si$_2$Ge$_2$, MnAs, MnFeP$_{0.45}$As$_{0.55}$, and La(Fe$_x$Si$_{1-x}$)$_{13}$, reach large values around the respective magnetic ordering temperature. Since the discovery of the giant magnetocaloric effect[3] in Gd$_5$Si$_2$Ge$_2$, the search for new magnetic materials with giant values of the magnetocaloric potentials is one of the big challenges to develop magnetic refrigeration. It has also been shown[9–12] that applied pressure is a good trigger to change the magnitudes of the peaks of the magnetocaloric potentials and select the temperature range of their maximum values. Besides, applied pressure itself can also be used to change the entropy of a solid without the need for magnetic field variation. The characteristic quantities of this process, known as barocaloric effect, are the isothermal entropy change $[\Delta S]_B$ and the adiabatic temperature change $[\Delta T_{ad}]_B$ upon pressure variation. A few years ago, the barocaloric effect was measured[13] in the compound Pr$_{1-x}$La$_x$NiO$_3$. In such a compound the temperature change upon pressure variation, which was attributed to the modification of the magnetic state due to the pressure effect on the crystalline electrical field interaction, is very small and is not suitable for practical application. Very recently, it was proposed[14] that the barocaloric potentials $[\Delta S]_B$ and $[\Delta T_{ad}]_B$ can be very large in metallic compounds, in which applied pressure yields big changes in the magnetic ordering temperature.

Experimental data[8] show that the peaks of the isothermal entropy changes in the compounds La(Fe$_x$Si$_{1-x}$)$_{13}$ doped with hydrogen are pushed toward higher temperatures as the hydrogen concentration is increased. However, the magnitudes of the magnetocaloric potentials ΔS and ΔT_{ad} are almost the same. On the other hand, it has been experimentally shown[12] that an applied pressure as large as 8 kbar makes significant changes in the magnetic ordering temperature in the doped compound La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$ and yields large values of the magnetocaloric potential ΔS. These experimental evidences indicate that the series of compounds La(Fe$_x$Si$_{1-x}$)$_{13}$ has an enormous potential to be used as magnetic refrigerants in magnetic refrigerators in a wide range of temperatures. Moreover, experimental data upon applied pressure make the compound La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$ an interesting candidate to study the barocaloric effect.

In this work, we theoretically study the barocaloric effect and the magnetocaloric effect under external pressure, in the doped compound La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$. We also discuss the magnetocaloric effect when both magnetic field and pressure are changed. Our theoretical calculations show that the magnetocaloric potential ΔS is in very good agreement with the available experimental data at ambient pressure and low applied pressure. The theoretical calculations show that the barocaloric potentials $[\Delta S]_B$ and $[\Delta T_{ad}]_B$ reach sizeable values in a wider range of temperatures, as compared with the magnetocaloric counterparts, calculated at fixed pressure. Our calculations show that when both magnetic field and applied pressure are changed, the magnetocaloric potentials ΔS and ΔT_{ad} also get large values in a wide temperature range. These theoretical results indicate that the barocaloric effect as well as the magnetocaloric effect under simultaneous variation of magnetic field and applied pressure can be an important tool to develop magnetic refrigeration.

II. MODEL

The theoretical calculations of the magnetocaloric effect in the compound La(Fe$_{0.89}$Si$_{0.11}$)$_{13}$ may be done using a model Hamiltonian with two sublattices, in the framework of the band theory of magnetism. However, such a calculation is much more complex and is not in the scope of the present work. So, for the sake of simplicity, we consider here an effective density of states containing mainly the outer electrons from Fe and Si ions. This is a reasonable approach, since the magnetization in this doped compound is entirely

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dominated by Fe atoms, once La and Si are nonmagnetic. So, we start with the following model Hamiltonian, in the mean field approximation:

$$\mathcal{H} = \sum_{i\sigma} (\varepsilon_i + U^d\langle n_{-\sigma}\rangle - \mu_B B) a_{i\sigma}^{+} a_{i\sigma} + \sum_{ij\sigma} T_{ij\sigma} a_{i\sigma}^{+} a_{j\sigma}. \quad (1)$$

Here ε_i is an effective energy level, U^d is the Coulomb interaction parameter between itinerant electrons, and B is the applied magnetic field. The term $T_{ij\sigma}=\Sigma_{ij}\widetilde{\varepsilon}_{k\sigma}e^{ik(R_i-R_j)}$ represents the electron hopping energy between two different sites, where $\widetilde{\varepsilon}_{k\sigma}=\alpha_p(1-\gamma^{el}M^2)$ is the renormalized electron dispersion relation.[15] Here γ^{el} is the electronic magnetoelastic coupling parameter, M is the magnetization, and the parameter α_p accounts for the effects of an external pressure. The local Green function associated with the previous Hamiltonian is given by

$$g_{00\sigma}(\omega) = \int \frac{\rho_0(\varepsilon')d\varepsilon'}{[\omega - U^d\langle n_{-\sigma}\rangle - \mu_B B - \alpha_p(1-\gamma^{el}M^2)\varepsilon']}, \quad (2)$$

where $\omega=\varepsilon+i0$ and $\rho_0(\varepsilon')$ is a standard paramagnetic density of states. The effective density of states per spin direction is given by

$$\rho_\sigma(\varepsilon) = -\frac{1}{\pi}\mathrm{Im}[g_{00\sigma}(\omega)]. \quad (3)$$

The electron occupation number is calculated from $n_\sigma=\int\rho_\sigma(\varepsilon)f(\varepsilon)d\varepsilon$, where $f(\varepsilon)$ is the Fermi distribution function. Notice that the applied magnetic field and the electron-electron interaction introduce a shift in the energy bands of opposite spins favoring the appearance of the ordered magnetic state. On the other hand, the applied pressure increases the width of the effective band and opposes the onset of the ordered magnetic state, as assured by the Stoner criterion. The magnetoelastic coupling, described in the model by the parameter γ^{el}, controls the nature of the magnetic phase transition. In fact, the magnetic phase transition changes from second to first order when the parameter γ^{el} becomes larger than a given critical value.[15]

The total entropy is given by $S(T,B,p)=S_{lat}(T,B,p)+S_{mag}(T,B,p)+S_{el}(T)$. Here $S_{el}(T)=\gamma T$ is the entropy from the *sp*-conduction electrons, where γ is the Sommerfeld coefficient. $S_{mag}(T,B,p)$ is the contribution from the itinerant 3*d*-electrons given by[15,16]

$$S_{mag}(T,B,p) = N_m R\left[\sum_\sigma \int_{-\infty}^{\mu} \ln(1+e^{-\beta(\varepsilon-\mu)})\rho_\sigma(\varepsilon)d\varepsilon + \frac{1}{k_B T}\sum_\sigma \int_{-\infty}^{\mu} (\varepsilon-\mu)\rho_\sigma(\varepsilon)f(\varepsilon)d\varepsilon\right], \quad (4)$$

where N_m is the number of magnetic ions per unit formula; μ is the chemical potential of the itinerant electrons; R is the gas constant; and $\beta=1/k_BT$, k_B being the Boltzmann constant. The contribution from the crystalline lattice to the total entropy is given in the Debye approximation by

$$S_{lat}(T,B,p) = N_i\left[-3R\ln(1-e^{-\Theta_D/T}) + 12R\left(\frac{T}{\Theta_D}\right)^3 \int_0^{\Theta_D/T} \frac{x^3}{e^x-1}dx\right], \quad (5)$$

where N_i is the number of ions per unit formula. Here $\Theta_D=\Theta_0(1-\gamma^{lat}M^2)$ is a renormalized Debye temperature,[15] where Θ_0 is the bare value of the Debye temperature, M is the total magnetization, and γ^{lat} is the lattice magnetoelastic coupling parameter. The magnetocaloric potentials ΔS and ΔT_{ad} upon magnetic field variation from B_0 to B_1 are respectively calculated by $\Delta S=S(T,B_1,p)-S(T,B_0,p)$ and $\Delta T_{ad}=T_2-T_1$ under the adiabatic condition $S(T_2,B_1,p)=S(T_1,B_0,p)$. Similarly, the barocaloric potentials $[\Delta S]_B$ and $[\Delta T_{ad}]_B$ upon pressure variation from p_0 to p_1 are respectively calculated by $[\Delta S]_B=S(T,B,p_1)-S(T,B,p_0)$ and $\Delta T_{ad}=T_2-T_1$ under the adiabatic condition $S(T_2,B,p_1)=S(T_1,B,p_0)$.

III. RESULTS AND DISCUSSION

In order to calculate the barocaloric and the magnetocaloric effects in the doped compound $\mathrm{La(Fe_{0.89}Si_{0.11})_{13}}$, we adopt a standard paramagnetic density of states[17] $\rho_0(\varepsilon')$ with a renormalized bandwidth. The Coulomb interaction parameter (U^d), the energy bandwidth (W), and the magnetoelastic coupling parameter (γ^{el}) were properly chosen to reproduce the experimental value of the saturation magnetization[8] at ambient pressure. Here we use the following values: $U^d/W=0.302$ and $\gamma^{el}=1.35$. The temperature dependence of the Debye temperature was self-consistently calculated from $\Theta_D=\Theta_0(1-\gamma^{lat}M^2)$ with $\Theta_0=315$ K and $\gamma^{lat}=-0.003$. The value of Θ_0 was taken in order to adjust the available experimental data[8] of the total entropy in the compound $\mathrm{La(Fe_{0.89}Si_{0.11})_{13}}$ at ambient pressure. When an external pressure is applied, the lattice parameters are reduced and the energy bandwidth is increased. As a result, the magnetic ordering temperature, which depends on the energy bandwidth, will be a function of the applied pressure. Generally speaking, increasing applied pressure reduces the magnetic ordering temperature in compounds with itinerant electron magnetism. The effect of pressure on the energy bandwidth at $T=0$ K can be described by first principles band calculations. However, such calculations are very complex and are not in the scope of the present paper. Moreover, at finite temperature, the pressure effects cannot be properly incorporated in such energy band calculations. Therefore, in order to discuss the physics involved in the magnetocaloric effect in the compound $\mathrm{La(Fe_{0.89}Si_{0.11})_{13}}$, under applied pressure, we phenomenologically describe the increase of the bandwidth and the pressure dependence of the magnetic ordering temperature by the model parameter α_p. Therefore, this parameter may be taken proportional either to the lattice parameters or to the magnetic ordering temperature. In this work, for the sake of simplicity, we consider the following relationship, $\alpha_p\simeq\{1+[T_C(0)-T_C(p)]/T_C(0)\}^{1/4}$, where $T_C(0)$ and

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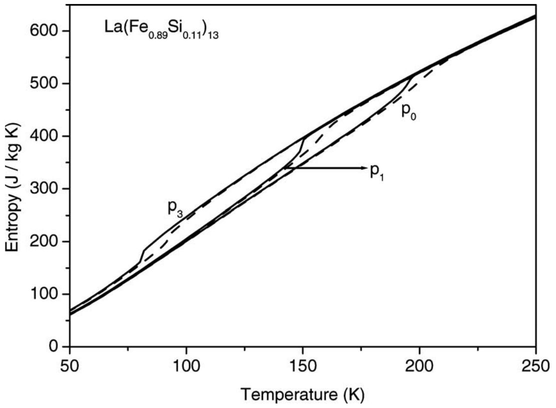

FIG. 1. Total entropy in La$(Fe_{0.89}Si_{0.11})_{13}$ at ambient pressure (p_0) and for applied pressures of 4.5 kbar (p_1) and 8.3 kbar (p_3). Solid (dashed) lines represent the calculations for $B=0(5\ \text{T})$.

$T_C(p)$ are, respectively, the magnetic ordering temperature at ambient pressure and at a given applied pressure. Thus, using the available experimental values[12] of T_C, we get $\alpha_p=1.0$ for ambient pressure and $\alpha_p=1.050$, $\alpha_p=1.090$, and $\alpha_p=1.123$ for applied pressures of 4.5 kbar (p_1), 6.5 kbar (p_2), and 8.3 kbar (p_3), respectively.

In Fig. 1, we plot the total entropy for $B=0$ (solid lines) and $B=5$ T (dashed lines) at ambient pressure (p_0) and for 4.5 kbar (p_1) and 8.3 kbar (p_3). In Figs. 2 and 3, we plot the isothermal entropy changes and the adiabatic temperature changes in La$(Fe_{0.89}Si_{0.11})_{13}$ under applied pressure and for a magnetic field variation from 0 to 5 T. From Fig. 2, we can observe that the agreement between the theoretical calculations and the available experimental data[12] at ambient pressure (p_0) and for an applied pressure of 4.5 kbar (p_1) is very reasonable. At an applied pressure of 8.3 kbar (p_3) the calculations and the experimental data of ΔS are in agreement only outside the temperature range of the first order magnetic phase transition. Around the magnetic ordering temperature, the theoretically calculated values of ΔS are much smaller than the available experimental data.[12] In our present calculations, the magnetic entropy change alone is not able to account for the large peak observed in ΔS at an applied pressure of 8.3 kbar. In order to account for such a great value of the isothermal entropy change, it would be necessary to include in the calculations of ΔS a very large contribution from the crystalline lattice entropy. However, there is no physical evidence that the crystalline lattice could contribute with such a large amount of entropy change. We believe that the experimental data of ΔS in this range of temperature may not be so reliable. This is because ΔS was indirectly obtained using magnetization data through the Maxwell relation $(\partial S/\partial B)_T=(\partial M/\partial T)_B$, whose validity in the case of first order phase transition is not assured.[18,19] As a matter of fact, in the derivation of the Maxwell relation it is supposed that both the magnetization and entropy are derivable functions in all ranges of temperatures and magnetic fields. This condition is not fulfilled around the first order phase transition, where there is a jump in the magnetization and entropy curves. Therefore, it is extremely important to obtain further experimental data for ΔS, mainly extracted from heat capacity measurements, to remove this puzzle between theoretical calculations and experimental data. The calculated adiabatic temperature changes for some values of the applied pressure, shown in Fig. 3, are our theoretical predictions and need experimental data to be confirmed.

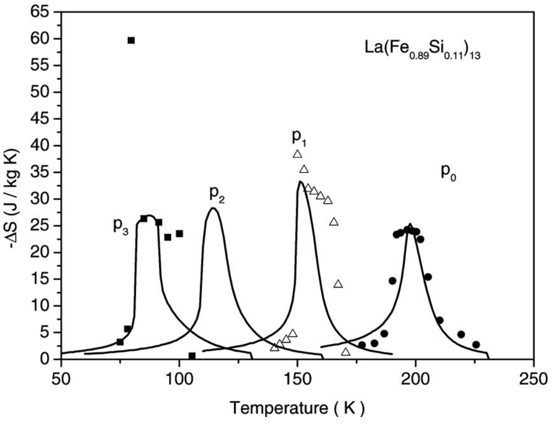

FIG. 2. Isothermal entropy changes for La$(Fe_{0.89}Si_{0.11})_{13}$ upon magnetic field variation from 0 to 5 T at ambient pressure (p_0) and for applied pressures of 4.5 kbar (p_1), 6.5 kbar (p_2), and 8.3 kbar (p_3). Solid lines are our calculations whereas symbols represent the available experimental data (Ref. 12).

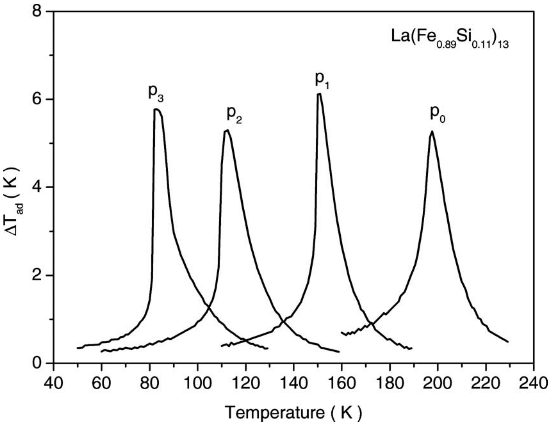

FIG. 3. Calculated adiabatic temperature changes in La$(Fe_{0.89}Si_{0.11})_{13}$ upon magnetic field variation from 0 to 5 T at ambient pressure (p_0) and for applied pressures of 4.5 kbar (p_1), 6.5 kbar (p_2), and 8.3 kbar (p_3).

In order to further discuss the effect of applied pressure on the magnetocaloric properties of the doped compound La$(Fe_{0.89}Si_{0.11})_{13}$, we calculate the magnetocaloric potentials ΔS and ΔT_{ad} when both the magnetic field and pressure are changed. For the sake of simplicity, we take a magnetic field variation from 0 to 5 T and consider a pressure variation from 0 at 197 K to 8.3 kbar at 85 K. The magnetocaloric potentials ΔS and ΔT_{ad} calculated with this particular variation of applied pressure and magnetic field are shown by the solid lines in Figs. 4 and 5, respectively. For the sake of comparison, we also plot in these figures the magnetocaloric potentials at constant pressure $p_0=0$ and $p_3=8.3$ kbar for the same magnetic field variation (dashed lines). From these figures, it can be observed that the magnetocaloric potentials, when both magnetic field and pressure are changed, get large values in a wider range of temperatures, as compared with the ones calculated at constant pressure. This kind of behavior of the magnetocaloric potentials is very useful for im-

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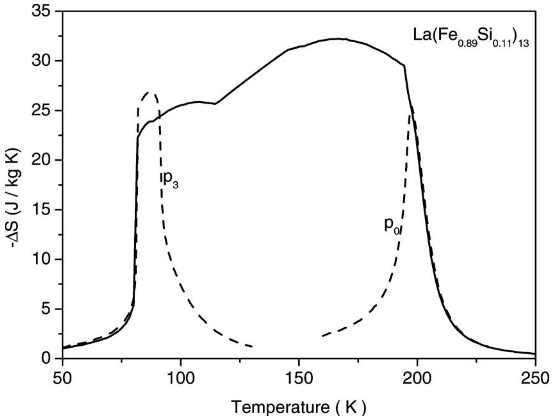

FIG. 4. Calculated isothermal entropy changes for $\mathrm{La(Fe_{0.89}Si_{0.11})_{13}}$ upon magnetic field variation from 0 to 5 T and pressure variation from 0 at 197 K to 8.3 kbar (p_3) at 85 K (solid line). The dashed lines represent the calculation upon magnetic field variation from 0 to 5 T at constant pressure $p_0=0$ and $p_3=8.3$ kbar.

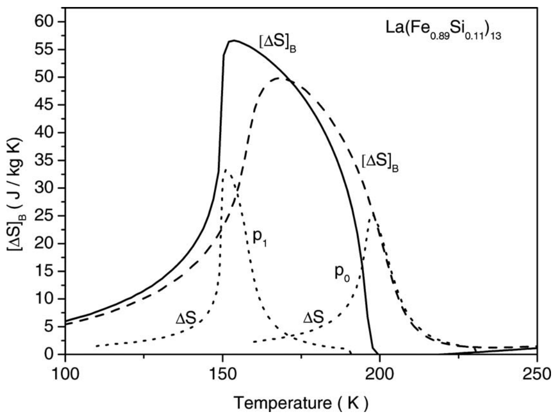

FIG. 6. Barocaloric potential $[\Delta S]_B$ in $\mathrm{La(Fe_{0.89}Si_{0.11})_{13}}$ calculated upon pressure variation from 4.5 kbar (p_1) to 0 at constant magnetic field of $B=0$ (solid line) and $B=5$ T (dashed line). The dotted lines represent the magnetocaloric potential ΔS calculated upon magnetic field variation from 0 to 5 T at constant pressure $p_0=0$ and $p_1=4.5$ kbar.

proving the efficiency of magnetic refrigerators, operating in the Ericsson cycle. Similar effects are expected to occur in any compound whose magnetic ordering temperature suffers big changes as a function of the applied pressure.

According to experimental data[12] the magnetic ordering temperature in the compound $\mathrm{La(Fe_{0.89}Si_{0.11})_{13}}$ is greatly reduced with increasing applied pressure. Thus, it is expected that the barocaloric quantities $[\Delta S]_B$ and $[\Delta T_{ad}]_B$ reach sizeable values upon variation of the applied pressure. In Figs. 6 and 7, we plot the barocaloric potentials $[\Delta S]_B$ and $[\Delta T_{ad}]_B$ in the compound $\mathrm{La(Fe_{0.89}Si_{0.11})_{13}}$ calculated for a fixed magnetic field of $B=0$ (solid line) and $B=5$ T (dashed line) upon pressure variation from 4.5 kbar to 0. For the sake of comparison, we also plot in these figures the magnetocaloric potentials ΔS and ΔT_{ad} calculated at fixed applied pressures ($p_0=0$ and $p_1=4.5$ kbar) and magnetic field variation from 0 to 5 T (dotted lines). From these figures, we can observe that the barocaloric potentials $[\Delta S]_B$ and $[\Delta T_{ad}]_B$ reach values larger than the magnetocaloric potentials ΔS and ΔT_{ad}. This result, which needs experimental observation to be confirmed, also indicates that applied pressure can be a very important tool for developing magnetic refrigeration.

In conclusion, in this paper we have calculated the barocaloric effect at fixed magnetic field and the magnetocaloric effect under applied pressure in the compound $\mathrm{La(Fe_{0.89}Si_{0.19})_{13}}$. Our theoretical calculations point out the following: (i) The barocaloric potentials $[\Delta S]_B$ and $[\Delta T_{ad}]_B$ in $\mathrm{La(Fe_{0.89}Si_{0.19})_{13}}$ can reach large values in a wide temperature range. (ii) The magnetocaloric potentials ΔS and ΔT_{ad} when both magnetic field and pressure are changed can also reach sizeable values in a wide range of temperatures. (iii) Our theoretical calculations somehow point out that around the first order phase transition, the isothermal entropy change obtained using magnetization measurements through the Maxwell relation can be overestimated. Whether or not the procedures discussed in this work prove their practical application, our theoretical results indicate that it is very important to study the combined effect of magnetic field and applied pressure on the caloric properties of transition-like metallic compounds, because it can reveal important aspects of the underlying physics behind them.

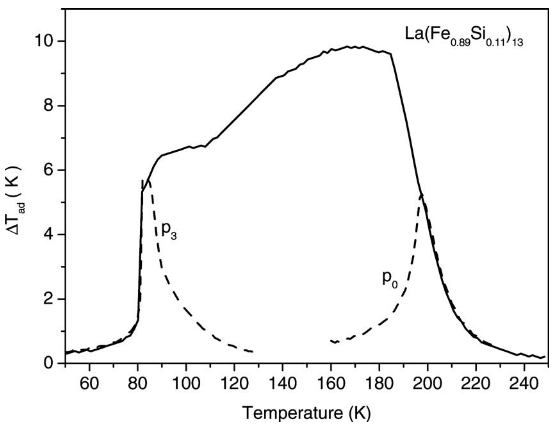

FIG. 5. Calculated adiabatic temperature change in $\mathrm{La(Fe_{0.89}Si_{0.11})_{13}}$ upon magnetic field variation from 0 to 5 T and pressure variation from 0 at 197 K to 8.3 kbar (p_3) at 85 K (solid line). The dashed lines represent the calculation upon magnetic field variation from 0 to 5 T at constant pressure $p_0=0$ and $p_3=8.3$ kbar.

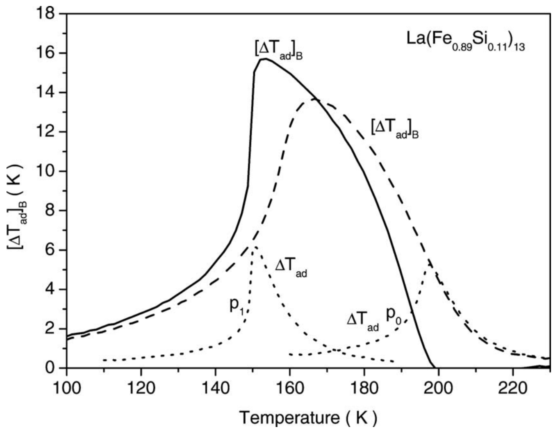

FIG. 7. Barocaloric potential $[\Delta T_{ad}]_B$ in $\mathrm{La(Fe_{0.89}Si_{0.11})_{13}}$ calculated upon pressure variation from 4.5 kbar (p_1) to 0 at constant magnetic field of $B=0$ (solid line) and $B=5$ T (dashed line). The dotted lines represent the magnetocaloric potential ΔT_{ad} calculated upon magnetic field variation from 0 to 5 T at constant pressure $p_0=0$ and $p_1=4.5$ kbar.

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