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Magnetocaloric effect in transition metals based compounds: a theoretical approach

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Abstract. In this paper we study the magnetocaloric effect in transition metals based compounds. For this purpose, we use a microscopical model, based on the band theory of magnetism, where the magnetic lattice is coupled with the crystalline lattice and with the external magnetic field. We apply the model to calculate the magnetocaloric effect in the compound MnAs, which undergoes a first order magnetic phase transition. The theoretically calculated isothermal entropy changes and the adiabatic temperature changes upon magnetic field variation exhibit a good agreement with the available experimental data.

PACS. 75.30.Sg Magnetocaloric effect, magnetic cooling – 75.10.Lp Band and itinerant models – 75.20.En Metals and alloys

1 Introduction

The magnetocaloric effect [1–10] is intrinsic to all magnetic materials and is due to the coupling between the magnetic lattice and the external magnetic field. The magnetocaloric effect [1–4] is measured by the isothermal entropy change ΔS upon magnetic field variations, i.e., $\Delta S(T) = S(T, h^{ext} \neq 0) - S(T, h^{ext} = 0)$ and by the temperature change $\Delta T_{ad}(T) = T_2 - T_1$ determined under the adiabatic condition $S(T_2, h^{ext} \neq 0) = S(T_1, h^{ext} = 0)$. Experimental data [1–4] show that the magnetocaloric effect in heavy lanthanide metals, and their compounds is larger than that observed in transition metals based compounds. The largest values of the magnetocaloric effect in rare earth compounds, are usually associated with large saturation magnetization and small magnetic ordering temperature as compared with the corresponding values of the transition metals compounds. How to get magnetic compounds exhibiting large values for the magnetocaloric effect has been a long standing question for many years. In the later nineties, Pecharsky and Gschneidner [5,6] have discovered that the compound $Gd_5Si_2Ge_2$, which has saturation magnetization around 7.0 μ_B and undergoes a magnetic phase transition around room temperature, exhibits a giant magnetocaloric effect. For instance, around 300 K and for a magnetic field variation from 0 to 5 T the isothermal entropy change in this compound is about 19 J/K Kg. It has been pointed out that this large value of the isothermal entropy change is due to a first order magnetic phase transition combined with a crystallographic transformation. Recent experiments [11–13] reveal that at room temperature and for a magnetic field variation from 0 to 5 T, the isothermal entropy change in the compound MnAs, is as large as 40 J/K Kg. The large value of the isothermal entropy change in this compound is also due to a first order magnetic phase transition. Although the saturation magnetization in the compound MnAs, typically of the order of 3.4 μ_B, is smaller than that of rare earth compounds, its isothermal entropy change is much greater than that observed in $Gd_5Si_2Ge_2$. This fact indicates that not only the magnitude of the magnetic moment but also the nature of the magnetic phase transition is important to the establishment of the magnetocaloric effect.

More recently, Tegus et al. [14] have shown that the compound $MnFeP_{0.45}As_{0.55}$ also exhibits at room temperature an isothermal entropy change as large as that observed in $Gd_5Si_2Ge_2$. The giant isothermal entropy change in the compound $MnFeP_{0.45}As_{0.55}$ is also due to the first order magnetic phase transition. These experimental findings put the compounds MnAs and $MnFeP_{0.45}As_{0.55}$ together with $Gd_5Si_2Ge_2$ as interesting candidates with a potential to be used as active magnetic refrigerants at room temperature. Independently, whether or not these compounds will prove their technological applications, they constitute important systems to investigate the magnetocaloric effect associated with the first order magnetic phase transition. Although, the existence of a giant value of the isothermal magnetic entropy change associated with a first order magnetic phase transition can be easily understood via the macroscopic Maxwell thermodynamic relation, i.e., $\Delta S = \int_0^{h^{ext}} (\partial M/\partial T) dh^{ext}$, the microscopical physical mechanism involved in the isothermal magnetic entropy change in this class of compounds, is not yet well

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understood. Recently, Pecharsky et al. [15] have experimentally estimated that the isothermal entropy change in Gd_5Ge_4 has a large contribution from the crystallographic transformation induced in this compound by the applied magnetic field.

The magnetocaloric effect of rare-earth compounds undergoing a second order magnetic phase transition has been explained by using a model Hamiltonian of localized spins [16–18]. The magnetocaloric effect in the compounds RCo_2 (R= Er, Ho and Dy) which undergo a first order magnetic phase transition, has also been explained on the basis of the localized model including a coupling with itinerant electrons of the Co ions [19,20]. Last year, it has been reported [21] a phenomenological calculation of itinerant electrons metamagnetism and its implication on the giant magnetocaloric effect in compounds with first order magnetic phase transition. In that paper, the authors started with a phenomenological equation of state based on the Landau theory of phase transitions and use the Maxwell thermodynamic relation to calculate the isothermal magnetic entropy, for some values of the model parameters. Very recently, we calculate the magnetocaloric effect in the compound $Gd_5(Si_xGe_{1-x})_4$ by using a Heisenberg-like model including the magnetoelastic effect [22] on the basis of the Bean and Rodbel model [23]. Using a similar model [24], we also calculate the magnetocaloric effect in the compound $MnAs_{1-x}Sb_x$. However, despite the good agreement with experimental data obtained in that paper [24], the magnetocaloric effect in transition metal compounds such as $MnAs_{1-x}Sb_x$ and $MnFeP_{0.45}As_{0.55}$ should be better described by a model Hamiltonian based on the band theory of magnetism which incorporates itinerant electrons.

To the best of my knowledge, all theoretical works that calculate the magnetocaloric effect, via the entropy versus temperature diagram, consider that the contribution from a crystalline lattice to the total entropy does not depend on the magnetization and on the external magnetic field. However, in compounds exhibiting first order magnetic phase transition, which is usually accompanied by a unit cell deformation, the interactions between the electrons and the lattice vibrations, should be considered in the calculations of both the magnetic and the crystalline lattice entropies. In this paper, we report for the first time on a microscopic model to describe the magnetocaloric effect in transition metals based compounds. In the model, the contributions from the magnetic and the crystalline lattices to the total entropy depend on the applied magnetic field. Our main results show that the magnetoelastic effect plays an important role in the physical mechanism of the magnetocaloric effect, in compounds with first order phase transition, as it has already been experimentally [15] observed in Gd_5Ge_4.

2 Model

In order to describe the magnetocaloric effect in transition metal based compounds, we start with a model Hamiltonian including Coulomb correlated itinerant electrons and phonons. Besides, it is considered that the magnetic and the crystalline lattices are coupled via the electron-phonon interactions. For the sake of simplicity, we treat the Coulomb correlations between itinerant electrons in the mean field approximation. We also take, for simplicity, a semi-phenomenological approximation to incorporate the electron-phonon interactions, which renormalize both the electron and the phonon energies. In fact, it is observed that the magnetic ordering temperature of magnetic materials exhibiting a first order phase transition, depends on the unit cell deformation due to the magnetoelastic coupling [23]. Bean and Rodbel [23] assumed in their paper, based on the model of localized magnetism, that the Curie temperature change with the lattice deformation as $T_c = T_0(1 - \lambda \Omega)$, where $\Omega = (V - V_0)/V_0$ and λ parametrizes the variation of T_c with the cell deformation. T_0 and V_0 would be the Curie temperature and the volume in the absence of the crystalline lattice deformation. Following a similar procedure used in the Bean and Rodbel's paper [23], the effect of the unit cell deformation on the magnetic ordering temperature in the model of itinerant electrons can be taken into account by a renormalization of the electron energy as $\tilde{\varepsilon}_{k\sigma} = \varepsilon_{k\sigma}(1 - \gamma_{el}\Omega)$. We also consider that the unit cell deformation renormalizes the phonon dispersion relation in an analogous form, i.e., $\tilde{\omega}_k = \omega_k(1 - \gamma_{ph}\Omega)$. So, within these considerations, we start with the following effective model Hamiltonian describing itinerant $3d$-electrons and phonons:

$$H = \sum_{i_\sigma} \left(\varepsilon_0 + U \langle n_{-\sigma}\rangle - \sigma\mu_B h^{ext}\right) d^+_{i\sigma} d_{i\sigma} + \sum_{ij\sigma} T_{ij\sigma} d^+_{i\sigma} d_{j\sigma} + \sum_{k\sigma} \hbar\tilde{\omega} a^+_{k\sigma} a_{k\sigma}. \quad (1)$$

The last term in this Hamiltonian describes the phonons, whereas the remaining terms describe a system of itinerant electrons, in the single band approximation. Here $T_{ij\sigma} = \sum \tilde{\varepsilon}_{k\sigma} e^{ik(r_i - r_j)}$ represents the electron hopping energy between two different sites, where the renormalized energy $\tilde{\varepsilon}_{k\sigma}$ takes into account the lattice deformation. ε_0 is an atomic energy level and U is the Coulomb interaction parameter between itinerant electrons. In the mean field approximation, where the cell deformation Ω, associated with the magnetoelastic coupling, can be taken proportional to the total magnetization, i.e., $\Omega \sim M^2$, the local Green function for the $3d$ electrons is given by:

$$g^{el}_{00\sigma}(z) = \int \frac{\rho^{el}_0(\varepsilon')d\varepsilon'}{z - (1 - \gamma_{el}M^2)\varepsilon' - \varepsilon_0 - U\langle n_{-\sigma}\rangle + \sigma\mu_B h^{ext}} \quad (2)$$

where $z = \varepsilon + i0$ and $\rho^{el}_0(\varepsilon')$ is a standard paramagnetic density of states. The spin dependent density of states for $3d$ electrons, calculated by $\rho^{el}_\sigma(\varepsilon) = -\frac{1}{\pi}\,\mathrm{Im}\, g^{el}_{00\sigma}(z)$, should be self consistently determined under the condition that the total occupation number of electrons remains constant. The magnetization is calculated by $M = 5(\langle n^{el}_\uparrow\rangle - \langle n^{el}_\downarrow\rangle)$, where the factor 5 accounts for the degeneracy of the $3d$ states and the electrons occupation number per spin direction is given by $\langle n_\sigma^{el} \rangle = \int \rho_\sigma^{el}(\varepsilon) f(\varepsilon) d\varepsilon$, $f(\varepsilon)$ being the Fermi distribution function. The onset of the magnetization,essentially depends on the width of the density of states, as is assured by the Stoner criterion. From equation (2) it can be observed that the width of the electrons density of states changes as a function of temperature due to the magnetoelastic coupling. If, for a large magnetoelastic coupling parameter, a sudden increase of the volume of the unit cell at the magnetic ordering temperature occurs, when the temperature is decreased from the paramagnetic phase, the width of the electrons density of states will be reduced. As a result, the magnetization will be large and the magnetic phase transition will be of first order. On the contrary, if there is no a sudden variation of the volume of the unit cell at the magnetic ordering temperature, the width of the electron density of states is almost temperature independent and the magnetic phase transition will be of second order. Hence the strength of the magnetoelastic coupling combined with the width of the electron density of states, control the order of the magnetic phase transition.

The magnetic entropy associated with the electronic part of the Hamiltonian is given by:

$$S_{mag}(T, h^{ext}) = R \left[\sum_\sigma \int_{-\infty}^{\mu} \ln\left(1 + e^{-\beta(\varepsilon-\mu)}\right) \rho_\sigma^{el}(\varepsilon) d\varepsilon \right.$$
$$\left. + \frac{1}{kT} \sum_\sigma \int_{-\infty}^{\mu} (\varepsilon - \mu)\, \rho_\sigma^{el}(\varepsilon) f(\varepsilon) d\varepsilon \right] \quad (3)$$

where μ is the chemical potential of the electron gas; R is the universal gas constant and $\beta = 1/kT$, k being the Boltzmann constant. Notice that the above expression contains both the magnetic and the electronic entropies. It should be stressed that in transition metals based compounds, it is very difficult to separate the magnetic and the electronic entropies, once in these compounds the magnetism is due to the itinerant electrons. It is also important to mention that in the present calculations of the magnetocaloric effect, we consider in S_{mag} only the contribution from the "*d*" electrons. The contribution from the "*s*" and "*p*" electrons to the magnetic entropy is neglected because it does not affect the magnetocaloric quantities. This is because it is supposed that the contribution from the "*s*" and "*p*"-electrons does not depend on the external magnetic field, once the "*s*" and "*p*" electrons densities of states are very broad. Notice also, that the magnetic entropy depends on the cell deformation through the parameter γ_{el} which modifies the width of the spin dependent electrons density of states. So, in the case of first order magnetic phase transition, where the $3d$ electrons density of states is suddenly reduced at T_C, there will be a jump in the magnetic entropy.

According to statistical mechanics, the lattice entropy related to the phonon part of the Hamiltonian is given by

$$S_{lat} = R \left[- \int \ln\left(1 - e^{-\beta\hbar\tilde{\omega}}\right) \rho^{ph}(\tilde{\omega}) d\tilde{\omega} \right.$$
$$\left. + \frac{1}{kT} \int \frac{\hbar\tilde{\omega}}{(e^{\beta\hbar\tilde{\omega}} - 1)} \rho^{ph}(\tilde{\omega}) d\tilde{\omega} \right] \quad (4)$$

where $\rho^{ph}(\tilde{\omega})$ is a renormalized density of phonons frequency. Using the Green function technique, $\rho^{ph}(\tilde{\omega})$ can be written as $\rho^{ph}(\tilde{\omega}) = -\frac{1}{\pi} \operatorname{Im} g^{ph}(z)$; where $g^{ph}(z)$ is given by:

$$g^{ph}(z) = \int \frac{\rho_0^{ph}(\omega')d\omega'}{z - (1 - \gamma_{ph} M^2)\omega'} \quad (5)$$

where $\rho_0^{ph}(\omega')$ is a standard density of phonons frequency. Notice that the density of phonons frequency and consequently the crystalline lattice entropy, depend on the external magnetic field via the magnetization. In the particular case of the Debye approximation, where $\rho_0^{ph}(\omega) = \left(3N/\omega_D^3\right)\omega^2$, with N and ω_D being respectively the number of ions and the Debye cutoff frequency, the equation (4) for the crystalline lattice entropy reduces to the well known Debye interpolation formula:

$$S_{lat}(T, h^{ext}) = \left[-3R \ln\left(1 - \exp\left(-\frac{\tilde{\Theta}_D}{T}\right)\right) \right.$$
$$\left. + 12R \left(\frac{T}{\tilde{\Theta}_D}\right)^3 \int_0^{\tilde{\Theta}_D/T} \frac{x^3}{\exp(x) - 1} dx \right]. \quad (6)$$

Here $\tilde{\Theta}_D = \Theta_D(1 - \gamma_{ph} M^2)$ is the renormalized Debye temperature, Θ_D being the bare value of the Debye temperature. The total entropy of the compound is the sum of the contributions from the crystalline (S_{lat}) and the magnetic (S_{mag}) lattices, i.e., $S = S_{lat} + S_{mag}$. The isothermal entropy change upon magnetic field variation is calculated by:

$$\Delta S(T) = S(T, h^{ext} \neq 0) - S(T, h^{ext} = 0). \quad (7)$$

Notice that the previous equation for the isothermal entropy change takes explicitly into account the contribution from lattice entropy. If the dependence of the lattice entropy with the external magnetic field is neglected, the change in the total entropy reduces to the calculation of the change in the magnetic part of the entropy, as it is usually made in the literature. The present model is quite general and can be applied to calculate the magnetocaloric effect of many transition metals based compounds, undergoing both first and second order magnetic phase transitions. In our model, what determines the nature of the magnetic phase transition, it is the magnitude of the coupling between the magnetic and the crystalline lattices combined with the width of the $3d$ electrons density of states. On the one hand, for a wide electrons density of states or weak coupling between the magnetic and crystalline lattices, the magnetization around T_C changes smoothly, and a large magnetocaloric effect is not expected to occur. On the other hand, for a narrow electrons density of states, combined with a strong magnetoelastic coupling, a large variation of the magnetization around T_C will be brought about and a giant magnetocaloric effect is likely to occur, even if the magnetic phase transition is of second order.

3 Application to MnAs

In this section, we illustrate the present model with the calculation of the magnetocaloric effect in the compound MnAs. It has been experimentally shown [25–30] that this compound crystallizes in the NiAs-type structure and undergoes a first order transition from ferromagnetic to paramagnetic phase together with a crystallographic phase transformation from the NiAs-type structure to MnP-type structure, accompanied by a decrease of the volume of the unit cell. The decrease of the volume of the unit cell modifies the phonons frequency and the electronic structure of the compound, i.e., the density of phonons frequency and the electrons density of states become narrower as the temperature goes to zero. In our model, these effects are taken into account by the parameters γ_{el} and γ_{ph}, which should also be evaluated by self consistent calculations. However, this kind of calculations is not in the scope of the present paper. For the sake of simplicity and in order to reproduce the experimental data of the magnetization around the magnetic ordering temperature, we take the parameter γ_{el}, which should be less than one, proportional to the variation of the volume of the unit cell with temperature [25–27]. We also consider a small linear dependence of this parameter with the external magnetic field which is taken from the slope of the curve $T_C \times h^{ext}$ inferred from the magnetization measurements [12]. The parameter γ_{ph} was taken as one tenth of the parameter γ_{el}. With this particular choice of the γ_{el} and γ_{ph} parameters, it is supposed that the magnetoelastic coupling affects more the electrons density of states rather than the phonons frequency spectrum. We consider an initial paramagnetic $3d$ electrons density of sates, whose width is approximately 5 eV, extracted from first principles calculations [31]. The Coulomb interaction parameter was chosen to reproduce the experimental value of the saturation magnetization [12]. Here we take $U = 0.477$ in units of the electron bandwidth. It is very important to mention that in the mean field approximation for a model of correlated itinerant electrons, the magnetic ordering temperature (T_C) is overestimated. To obtain a more accurate value of T_C is necessary to go beyond the mean field approximation to treat better the Coulomb correlations. However, this kind of approach is far beyond the scope of the present paper. So, in order to compare our theoretical calculations with the available experimental data, we make plots using the renormalized temperature T/T_C.

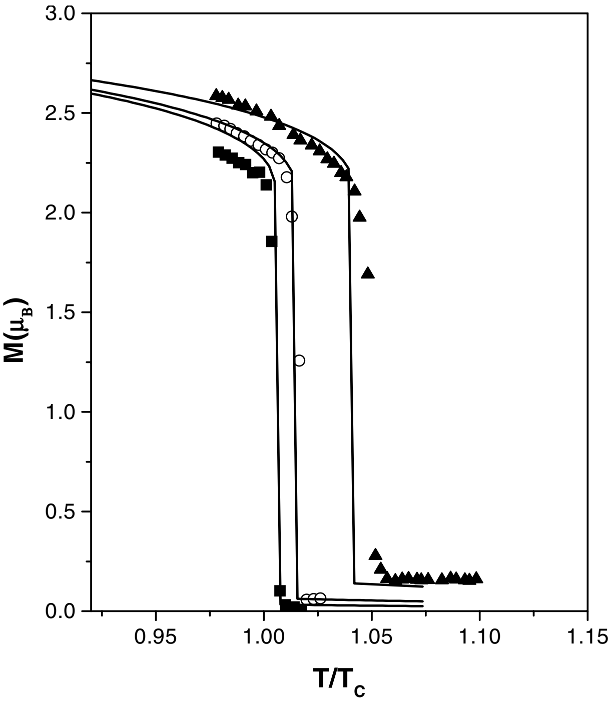

Fig. 1. Temperature dependence of the magnetization for MnAs. The solid lines correspond to our calculations whereas, squares, circles and triangles represent experimental data [12] for magnetic fields of 1 T, 2 T and 5 T respectively .

As far as the calculations of the crystalline lattice entropy are concerned, we use, for the sake of simplicity, a parabolic density of phonons frequency, as in the Debye approximation, and take the Debye temperature as $\Theta_D = 400$ K. Bearing in mind the previous discussion, we self consistently calculate the temperature dependence of the magnetization and the temperature dependence of the total entropy of the compound. For the set of the model parameters discussed previously, our calculations show that, as we go from low temperature to high temperature, the $3d$ electrons density of states, suddenly becomes broader around the magnetic ordering temperature. As a result, the magnetization goes abruptly to zero yielding a first order magnetic phase transition. The magnetizations calculated for low values of the applied magnetic field, depicted in Figure 1, are in good agreement with experimental data [12]. In Figure 2, we plot the temperature dependence of the total entropy for $h^{ext} = 0$ T (solid line); for $h^{ext} = 1$ T: (dashed line); for $h^{ext} = 2$ T (dotted line) and for $h^{ext} = 5$ T (dashed dotted line). Notice that there is a jump in the total entropy which is pushed to higher temperature as the magnetic field is increased. It is worth mentioning that both the magnetic and lattice entropies contribute to the jump in the total entropy, although the main contribution to it, comes from the magnetic part of the entropy. Our estimate is that the contribution from the crystalline lattice to the jump in the total entropy is less than 10%. Perchasky et al. [15] have experimentally estimated that the contribution from the crystallographic transformation to the isothermal entropy

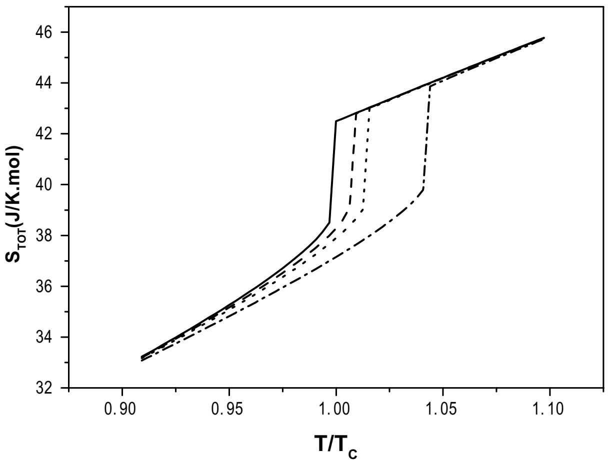

Fig. 2. Temperature dependence of the total entropy for MnAs compound. The solid line represents the calculation in the absence of an external magnetic field, whereas the dashed, dotted and the dashed-dotted lines represent the calculations for 1 T; 2 T and 5 T respectively.

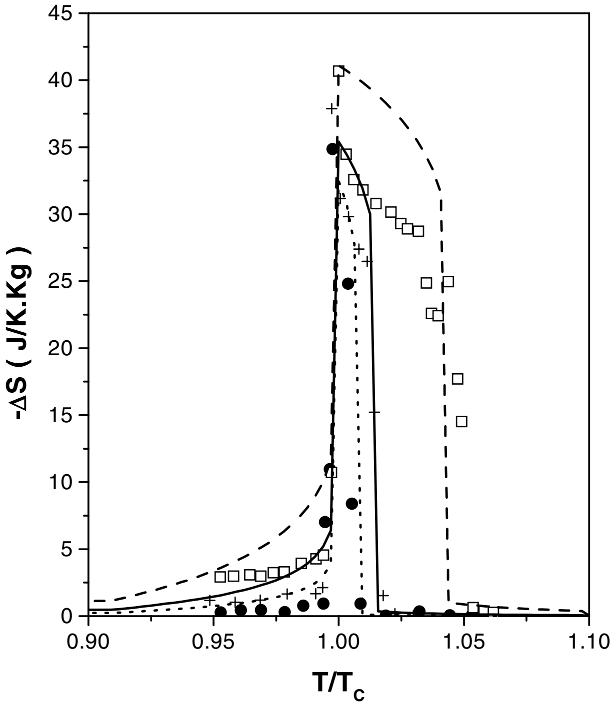

Fig. 3. Isothermal entropy change for MnAs. The dotted, solid and dashed lines correspond to our calculations, whereas circles, crosses and squares represent experimental data [12] for a magnetic field variations from 0 to 1 T; from 0 to 2 T and from 0 to 5 T respectively.

change in Gd_5Ge_4 is more than one half of the total effect in magnetic field below 2 T. Further theoretical and experimental studies are necessary to better understand the role of the crystalline lattice in the isothermal entropy change in compounds with first order phase transition.

In Figure 3, we show the corresponding isothermal entropy changes, which have also a small contribution from the lattice entropy. From this figure, we observe a better agreement with experimental data [12] for small magnetic field variations. However, for the magnetic field variation from 0 to 5 T, there are some deviations between our calculations and experiments. These discrepancies occur because, the effect of the magnetoelastic coupling extends to a larger range of temperature around T_C with increasing magnetic field. In order to obtain a better description of this effect it is necessary to take into account a more rigorous treatment of the electronic structure and of the magnetoelastic coupling. In Figure 4 we plot our theoretical calculations for the adiabatic temperature change (ΔT_{ad}) in MnAs. In this figure, we observe some deviations between the calculations and experiments, although the theoretical curves show the good trend of the experimental data [11]. Again, in order to obtain a better agreement between our theoretically calculated ΔT_{ad} and experiments it is necessary to include a more rigorous treatment of the electronic structure and of the magnetoelastic coupling.

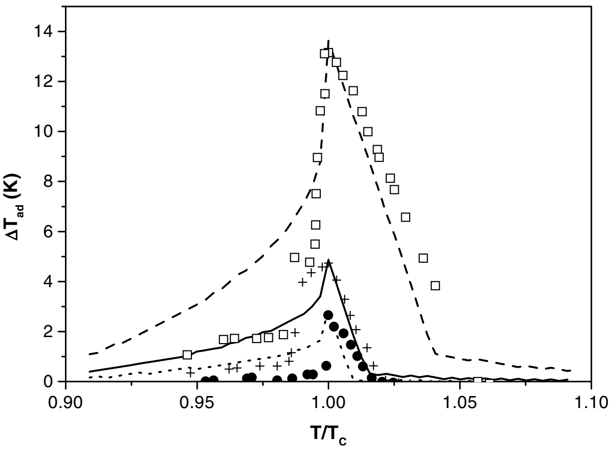

Fig. 4. Adiabatic temperature change for MnAs. The dotted, solid and dashed lines correspond to our calculations whereas, circles, crosses and squares represent experimental data [11] for a magnetic field variations from 0 to 1 T; from 0 to 2 T and from 0 to 5 T respectively.

4 Concluding remarks

In conclusion, in this paper we have proposed a microscopical model, based on the band theory of magnetism, to calculate the magnetocaloric effect in transition metals based compounds. The main points of the present model are: i) the coupling between the magnetic and the crystalline lattices, together with a narrow $3d$ electrons density of states, may lead to a first order magnetic phase transition and consequently to a giant magnetocaloric effect. ii) The inclusion of the magnetic field dependence of the crystalline lattice entropy opens a new horizon in the study of the magnetocaloric effect, mainly in the discussion of the physical mechanism involved in the isothermal entropy change.

Despite the approximations used in the model Hamiltonian, the agreement between the available experimental data and the theoretically calculated isothermal entropy change and the adiabatic temperature change, suggests that the calculations contain the correct physics involved in the process of the magnetocaloric effect, observed in the compound MnAs. The model is easily applied to calculate the magnetocaloric effect in many others transition metals based compounds. Besides it can be straightforwardly extended to calculate the barocaloric effect as well as the combined effect of the external pressure and applied magnetic field on the cooling power of transition metals based compounds. The results obtained in this paper indicate that a large magnetocaloric effect can be achieved in other metallic compounds based on Mn, by introducing interstitial or substitutional disorder in such a way to reduce the $3d$ electrons bandwidth associated with Mn ions. This is a suggestion for an experimental procedure to search novel magnetic materials based on transition metals exhibiting large magnetocaloric effect. It is also worth mentioning here, that the present model may be improved by using an approach beyond the mean field approximation, which certainly will provide a better value for T_C; by incorporating a more detailed calculations of the electronic structure and a more rigorous microscopic treatment of the electron-phonon interaction, which may provide better values for the γ_{el} and γ_{ph} parameters. However, it is expected that all of these improvements, which make the numerical calculations much more complex, do not change the main conclusions of this work.

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