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Magnetothermal Properties and Magnetocaloric Effect in the 3d Ferromagnetic Elements: Fe, Co, and Ni

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Abstract

We present a calculation of magnetothermal properties and magnetocaloric effect (MCE) for the ferromagnetic elements: Fe, Co, and Ni. In particular, we calculated the temperature and field dependences of magnetization, heat capacity, entropy, isothermal entropy change ΔS_m, adiabatic temperature change ΔT_{ad}, and the two figures-of-merit: the relative cooling powers RCP(S) and RCP(T). We have used the mean-field theory in calculating the magnetization, magnetic heat capacity, and magnetic entropy. The lattice and electronic contributions to the total heat capacity and entropy were calculated using standard relations to subsequently calculate ΔT_{ad}. Those contributions depend on the Debye temperature Θ_D and the coefficient of the electronic heat capacity γ_e respectively. The Maxwell relation is used to calculate ΔS_m and ΔT_{ad}. As an example of our results, the maximum ΔS_m for the three elements, in 6 T, is between 0.17 to 0.36 J/mol K and the maximum ΔT_{ad} is between 0.46 to 1.5 K/T for the same field change. The relative cooling power RCP(S) is in the 15–36 J/mol range for the three elements in a 6 T field. Also, the relative cooling power, RCP (T), is in the 162–1044 K^2 range for the same field. For Fe and Co the RCP (T) per Tesla values, i.e., 139 and 174 K^2/T respectively are comparable to that of Gd and other Gd-based magnetocaloric materials. The behavior of the magnetization, magnetic heat capacity, and magnetic entropy shows that the phase transition in these three elements is of the second order. The universal curve and Arrott plots further support this conclusion.

Keywords Magnetocaloric effect · 3d ferromagnetic elements · Mean-field theory

1 Introduction

The interest in the physical properties of the 3d elements is an old endeavor [1 and the references therein]. For example, studies on the magnetic, electronic, and elastic properties have been reported [2–10]. Works on the phase transition in Fe under pressure [2] and the dependence of the magnetization on temperature for iron nanoparticles [3] have been done.

Classical models, e.g., a model based on the premises of classical statistical mechanics, have been used to investigate the anisotropic magnetic properties of the 3d elements and their compounds [4, 10] and the size-dependent magnetic properties of elements, e.g., Fe and Gd [5, 6]. Recently, the anisotropic magnetocaloric effect (AMCE) in Fe has been reported [6]. Both of the mean-field model and a Hubbard-like model Hamiltonian, where electron–electron interaction is taken into account within the mean-field theory, were reported [7, 8]. The effect of high magnetic field on the magnetocaloric effect was reported by Tishin [9].

In the present paper, we present a detailed calculation, for Fe, Ni, and Co, of the magnetic, magnetothermal, e.g., heat capacity and entropy, and the magnetocaloric properties: the isothermal change in entropy and the adiabatic change in temperature. The relative cooling powers are also calculated and compared with bench-mark Gd and other Gd-based compounds. The nature of the magnetic phase transition in these elements is also investigated, in the light of the temperature and field dependences of the aforementioned properties, together with the Arrott plots and universal curves. At the end, we report on the temperature dependence of the magnetization and the isothermal change in entropy, for amorphous Fe, and compare these properties with its counterparts in crystalline Fe.

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2 Model and Analysis

In the mean-field theory (MFT) [11–17], the interaction between the magnetic moments is taken into consideration and is represented as a uniform internal (mean) field. The origin of the internal field, as described by Heisenberg, is the scalar product of spin operators. The internal field could be evaluated, from the magnetization, through a self-consistent calculation, unlike systems of non-interacting moments, e.g., paramagnetic systems where no internal field exists. The effective field is the sum of the external applied field and the internal field. There are some limitations, however, of the mean-field model at very low temperatures due to spin waves excitation and around the Curie temperature due to critical fluctuations. Albeit this limitation, the mean-field theory proved to be suitable in handling different crystalline and amorphous systems with up to three sublattices.

The effective magnetic field H_{eff} of any of the elements: Fe, Ni, and Co can be expressed as follows:

$$H_{eff}(T) = H + d\left[n_{tt}M(T)\right] \tag{1}$$

where H is the external applied magnetic field and $M(T)$ is the magnetic moment at temperature T. The factor $d = N_A \rho \mu_B / A$ converts the atomic moment from μ_B to Gauss, where A is the atomic mass in g per mole, ρ is the density in g/cm^3, N_A is Avogadro's number and the molecular field coefficient n_{tt} is dimensionless, and the symbol t stands for either Fe, Co, or Ni.

The magnetic moment $M(T, H)$ of any of the three elements is given by the equation:

$$M(T, H) = M_J B_J(x) \tag{2}$$

where $B_J(x)$ is the Brillouin function:

$$B_J(x) = \frac{2J+1}{2J}\coth\left(\frac{2J+1}{2J}x\right) - \frac{1}{2J}\coth\left(\frac{x}{2J}\right) \tag{3}$$

$$x = \frac{M_J H}{K_B T} \tag{4}$$

J is the total angular momentum quantum number.

$M_J = g\mu_B J$ the magnetic moment per atom.

From the following Maxwell relation, the magnetic entropy change is calculated [11]:

$$\left(\frac{\partial S(T)}{\partial H}\right)_T = \left(\frac{\partial M(T)}{\partial T}\right)_H$$

$$\Delta S_m(T) = \int_{H_0}^{H_f} \frac{\partial M(T)}{\partial T} dH \tag{5}$$

The above integral could be cast into a summation by using the well-known trapezoidal rule [18, 19]:

$$\Delta S_m(T) = \sum_i^{i+1} \frac{M_{i+1}(H_{i+1}, T_{i+1}) - M_i(H_i, T_i)}{T_{i+1} - T_i} \Delta H_i \tag{6}$$

The total heat capacity C_{tot} includes three contributions: the magnetic heat capacity C_m, the lattice heat capacity C_l, and the electronic heat capacity C_e [20].

$$C_{tot} = C_m + C_l + C_e \tag{7}$$

First, from the temperature derivative of the magnetic energy, we can calculate the magnetic contribution to heat capacity:

$$U_m = -\frac{1}{2}n_{tt}M^2(T) \tag{8}$$

$$C_m(T) = \frac{\partial U_m}{\partial T} \tag{9}$$

From the integration of the magnetic heat capacity, the magnetic entropy may be calculated as follows:

$$S_m(T) = \int_0^T \frac{C_m(T)}{T} dT \tag{10}$$

The theoretical maximum of the magnetic entropy is given by [11]

$$S_m(max) = R \, Ln(2J + 1) \tag{11}$$

where R is the gas constant.

Second, the lattice contribution to heat capacity is calculated from the Debye model [21, 22]:

$$C_l = 9N_a \, k\left(\frac{T}{\theta_D}\right)^3 \int_0^{\frac{\theta_D}{T}} \frac{e^y \, y^4}{(e^y - 1)^2} dy \tag{12}$$

where $y = \theta_D/T$ and θ_D is Debye temperature.

Third, the electronic heat capacity is proportional to temperature and is given by [23]:

$$C_e = \gamma_e T = (\pi^2/3)k^2 N_a \, N(E_f)T \tag{13}$$

where γ_e is the electronic heat capacity coefficient and $N(E_f)$ is the density-of-states at Fermi energy.

From Gibb's free energy, the Landau–Ginsburg theory is expressed as follows [24, 25]:

$$F = \frac{1}{2} A(T)M^2 + \frac{1}{4}B(T)M^4 + \frac{1}{6}C(T)M^6 - M.H \tag{14}$$

The magnetic entropy change is:

$$\Delta S_m(T) = -\left(\frac{dF}{dT}\right)_H = -\frac{1}{2}\frac{dA(T)}{dT}M^2 - \frac{1}{4}\frac{dB(T)}{dT}M^4 + \dots$$

(15)

From the equilibrium condition at T_c, $\frac{\partial F}{\partial M} = 0$ the magnetic equation of state is:

$$H = A(T)M + B(T)M^3 + \dots$$

(16)

$$\frac{H}{M} = A(T) + B(T)M^2 + \dots$$

(17)

where $A(T)$ and $B(T)$ are Landau's coefficients.

The adiabatic temperature change [20, 26] can be calculated from the following:

$$\Delta T_{ad=} -\int_{H_0}^{H_f} \frac{T}{C_{tot}(T,H)}\frac{\partial M(T,H)}{\partial T}dH$$

(18)

By using the Arrott–Belov–Kouvel (ABK) [27–29], the Arrott plots, i.e., the M^2 vs H/M plot, in the ferromagnetic region at different temperatures close to T_c, can be used to estimate the spontaneous magnetization and the Curie temperature. The Arrott plots are also used to determine the order of the phase transition involved, i.e., a second-order (SOPT) or a first-order phase transition (FOPT), from the sign of the plots slopes. Namely, positive slopes indicate SOPT, whereas negative slopes or s-shaped slopes indicate FOPT [30, 31].

The relation between $\Delta S_m / \Delta S_m^{peak}$ and θ is well known as the universal curve [32], one may choose the reference temperature T_r such that:

$$\Delta S_m(T_r) = 0.7\Delta S_m^{peak}$$

(19)

where θ is defined by:

$$\theta = (T - T_c)/(T_r - T_c)$$

(20)

The relative cooling power RCP(S) [17, 33] is defined as follows:

$$RCP(S) = \Delta S_{max}(T) \times \delta T_{FWHM}$$

(21)

where δT_{FWHM} is the full width at half maximum of the magnetic entropy change curve and $\Delta S_{max}(T)$ is the maximum magnetic entropy change.

Another figure-of-merit for the magnetocaloric materials is the RCP (T) defined as [33, 34]:

$$RCP(T) = \Delta T_{max} \times \delta T_{FWHM}$$

(22)

where δT_{FWHM} is the full width at half maximum of the ΔT_{ad} vs. T plot.

3 Results and Discussion

3.1 Magnetization

Figure 1a–c display the calculated magnetic moment as function of temperature for Fe, Ni, and Co respectively in fields of 0 and 5 T. The magnetic moment at very low
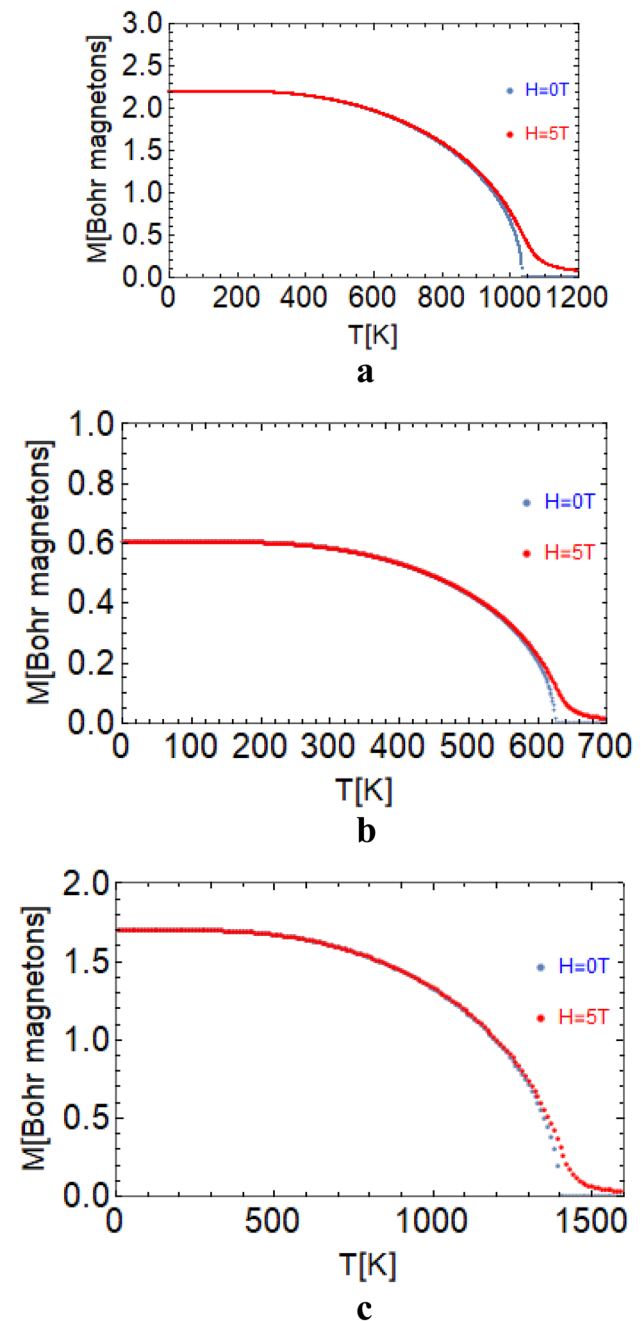

Fig. 1 **a** Magnetic moment dependence on temperature, in 0 and 5 T, for Fe. **b** Magnetic moment dependence on temperature, in 0 and 5 T, for Ni. **c** Magnetic moment dependence on temperature in 0 and 5 T, for Co
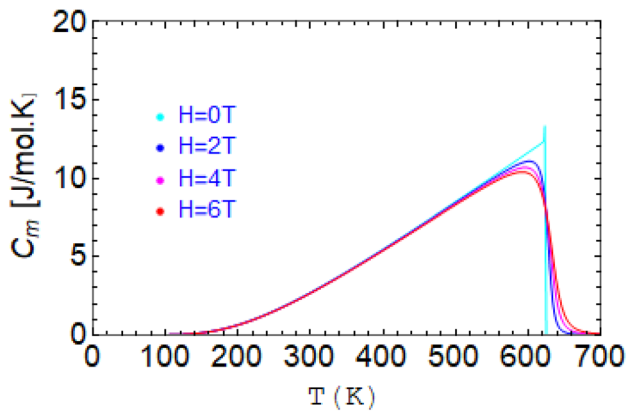
Fig. 2 Magnetic specific heat of Ni vs. temperature, in different fields

Table 1 Maximum S_m, in zero field, using Maxwell relation and the shown equation

Element	S_m (J/mol K) Maxwell method	S_m (J/mol K) = R Ln(2 J + 1)
Fe	9.044	9.118
Ni	5.738	5.753
Co	9.079	9.118

maximum values of the magnetic entropy, at and above T_c, as calculated from the mean-field theory are shown together with those calculated from the Eq. 11 e.g. [36]. The agreement is excellent as shown in Table 1.

3.3 The Isothermal Change in Entropy

The temperature dependence of the isothermal change in entropy for field changes 2, 4, 6, and 8 T, calculated from Maxwell relation via the trapezoidal method (Eqs. 5 and 6), is shown in Fig. 4a–c.

Table 2 lists both ΔS and RCP values in a 6 T field change. Moreover, the features of the temperature and field dependences of ΔS are those of SOPT materials [35].

We listed in Table 2 the isothermal change in entropy [18] and the corresponding (Eq. 21) RCP values in J/mol, for the three elements, in a 6 T field.

de Oliveira [37] used an itinerant electron model to study MCE in Fe, Co, Ni, and YFe$_3$. For the three elements, he reported ΔS_{max} of nearly: 8, 4, and 4 J/kg K in a 2.16 T field. These are 0.447, 0.235, and 0.236 J/mol K respectively. These values are about 2–3 times larger than our mean-field values.

3.4 The Adiabatic Change in Temperature

The adiabatic change in temperature for the three elements is shown in Fig. 5a–c in different magnetic fields.

The values of ΔT_{max} (K) for the three elements, as reported by De Oliveira [37], are in the range 2.5–5 K for fields in the range 2.16–3 T. Our values are in the range 1–7 K for fields in the range 2–4 T.

The RCP/ΔH values shown in Table 3 are to be compared with those of known materials, in the same units of course, e.g., 161.2 for Gd in 6 T, 96.0 for Gd$_5$ Si $_{2.06}$ Ge$_{1.94}$ in 5 T, 109.2 for Gd$_5$Si$_4$ in 5 T, and 145.1 for Gd$_4$Bi$_3$ in 10 T [33, 38].

3.5 High Field Effects

Figure 6a shows ΔS_m for Fe in fields up to 300 T. Two features of this figure are firstly, the peak temperature T_{peak} coincides with T_c even for these high fields using the present mean-field
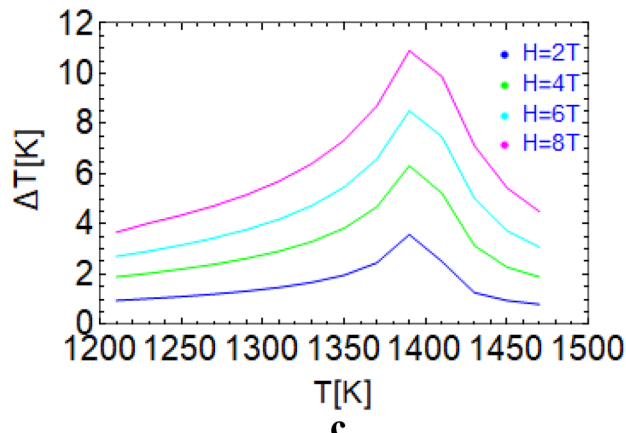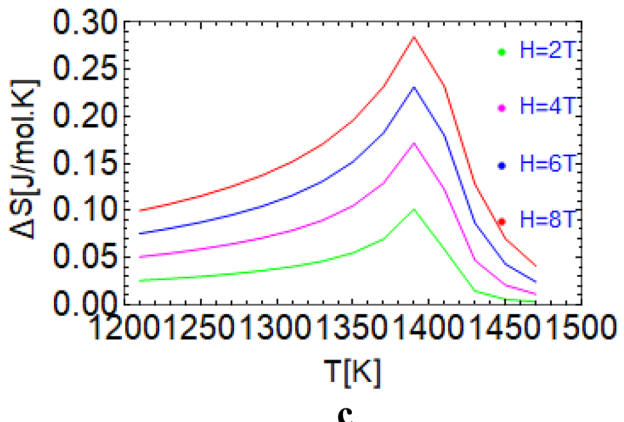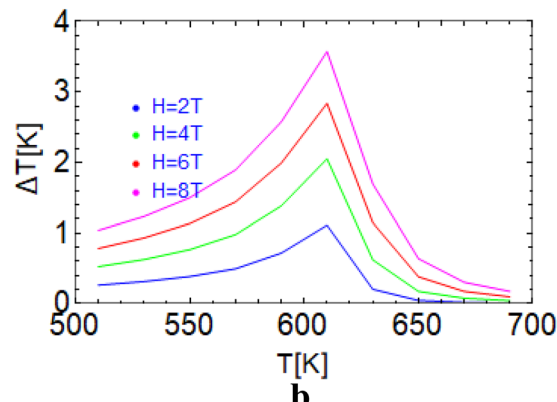
3.2 Magnetic Heat Capacity and Entropy

temperatures and the Curie temperatures of these three elements agree very well with experimental data, e.g., the magnetic moments are 2.2, 0.6, and 1.7 μ_B and T_c values are 1040, 630, and 1400 K respectively. Although the data in Fig. 1 are well known, we have found it necessary to show in order to demonstrate the fair success of the MFT theory. It is well known that both ΔS_m and ΔT_{ad} do depend on the temperature derivative of the magnetization at constant field and therefore on the MFT-calculated magnetization.

3.2 Magnetic Heat Capacity and Entropy

The magnetic contribution to heat capacity has been calculated from the magnetic energy (Eqs. 8 and 9). The magnetic specific heat of Ni is shown in Fig. 2 in a temperature range up to 700 K. The field dependence, at T_c, is clearly that of materials with SOPT [35]. The magnetic specific heat of Fe and Co show similar behavior.

The temperature dependence of the magnetic entropy of the three elements, in zero field, is shown in Fig. 3. The

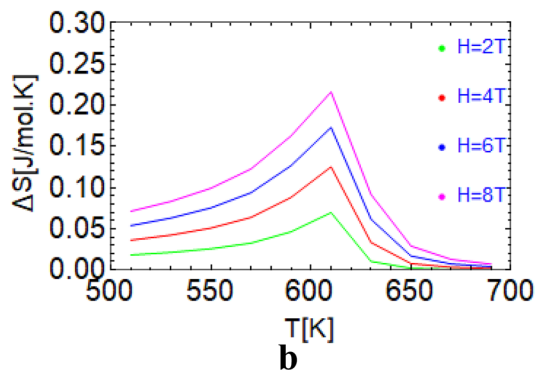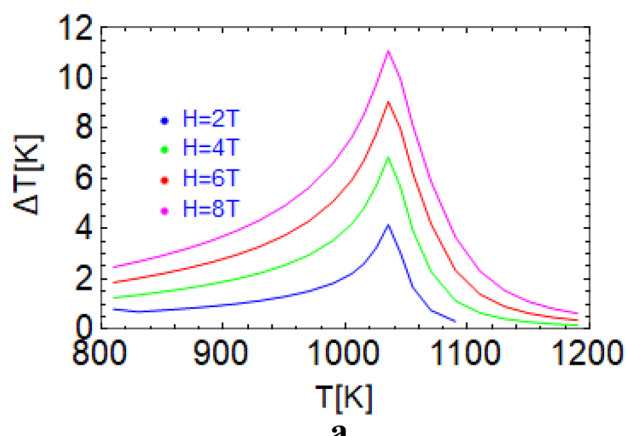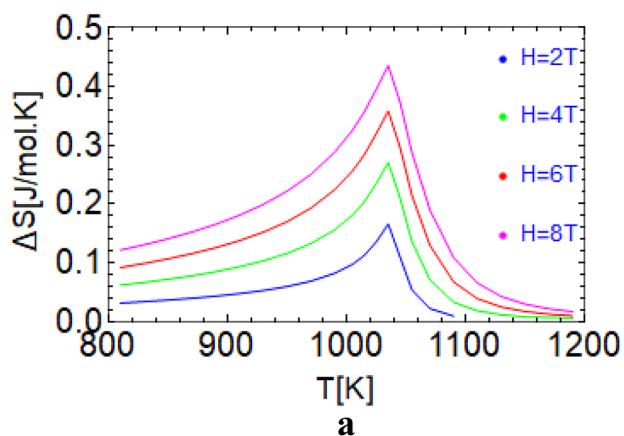**Fig. 3** Magnetic entropy of Fe, Ni, and Co, in zero field

Fig. 4 **a** Isothermal change in entropy for Fe in different fields. **b** Isothermal change in entropy for Ni in different fields. **c** Isothermal change in entropy for cobalt in different fields

calculation. This has been reported by [39], using numerical calculation, but for much lower fields (≤ 1.5 T). However, Franco

Table 2 Maximum isothermal entropy change and RCP(S), in 6 T, for the three elements

Element	δ_{FWHM} (K)	ΔS_{max} [J/mol.K]	RCP (J/mol)
Fe	101.2	0.3572	36.15
Ni	89.9	0.1738	15.62
Co	113	0.2309	26.09

Fig. 5 **a** Adiabatic change in temperature vs. temperature for Fe in different fields. **b** Adiabatic change in temperature vs. temperature for Ni in different fields. **c** Adiabatic change in temperature vs. temperature for cobalt in different fields

et al. showed, by using the Heisenberg model, that $T_{peak} > T_c$. Secondly, the curves become more flat, i.e. the FWHM of any given curve, in case of using high fields, becomes larger.

The adiabatic change in temperature, in high fields, has been studied by Tishin [9]. Figure 6b shows our results for Fe in fields of 20, 40, 60, and 100 T. It is clear from that the

Table 3 Adiabatic change in temperature, RCP (T) in 6 T field and RCP (T) per tesla

Element	δ_{FWHM} (K)	ΔT_{max} (K)	RCP (K^2)	RCP/ΔH (K^2/T)
Fe	92	9.09	836.28	139.38
Ni	57.1	2.844	162.39	27.065
Co	121	8.63	1044.23	174.038

Fig. 7 Arrott plots for Fe in field up to 1 T

maximum of ΔT_{ad} shifts as the field increases. The same trend is found for Co.

3.6 Arrott Plots and the Universal Curves

The Arrott plots are used to study the nature of the phase transition [28, 29] and the itinerant nature [9] of the electrons in Fe, Co, and Ni. Weak itinerant nature was found for Fe. The straight lines of the same slopes and the values of M^2 in zero field (Fig. 7) are indicative of weak itinerant nature according to Eq. 23 [40]. We have found that the percentage error between M^2 values in zero field is in the
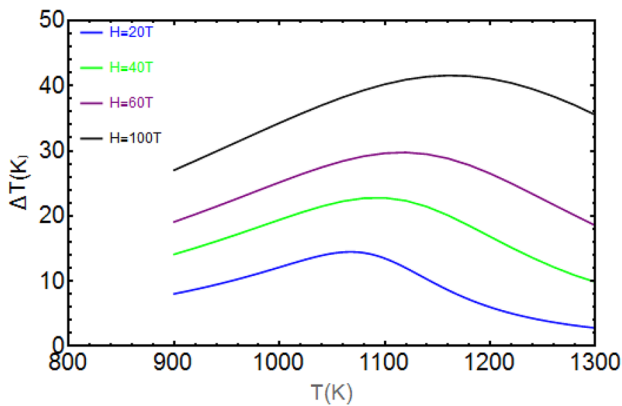
3.4–19% as calculated from the mean-field theory and from the following equation:

$$M(H,T)^2 = M(0.0)^2 \left[\left(1 - \frac{T^2}{T_c^2} \right) + \frac{2x_0 H}{M(H,T)} \right] \tag{23}$$

Figure 8a, b display the universal curves for Fe and Ni respectively. The features are clearly those of SOPT. In
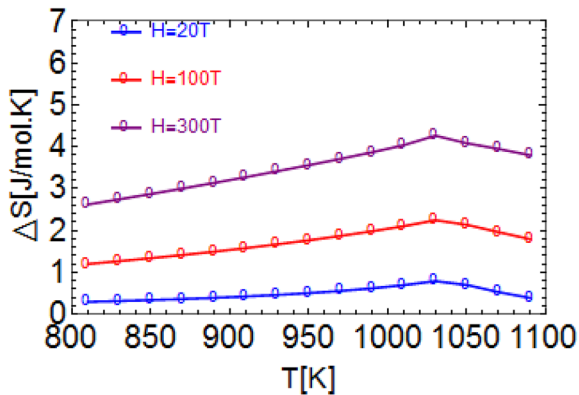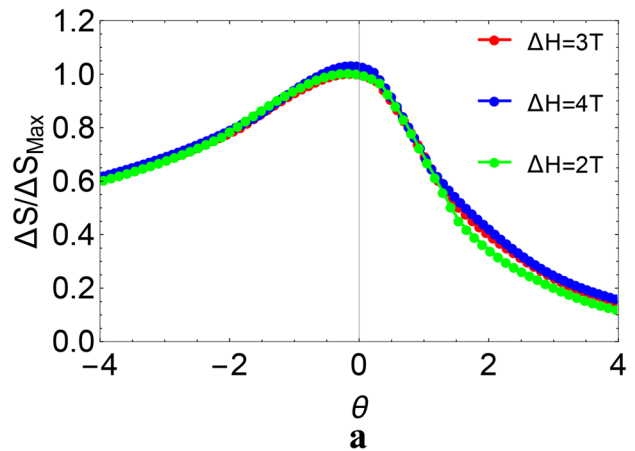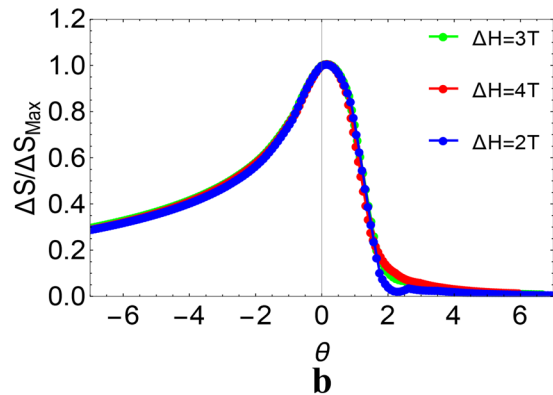

Fig. 6 **a** Isothermal entropy change vs. temperature, for Fe, in fields of: 20, 100, and 300 T. **b** Adiabatic change in temperature vs. temperature, for Fe, in fields of: 20, 40, 60, and 100 T

Fig. 8 **a** The universal curve for Fe. **b** The universal curve for Ni
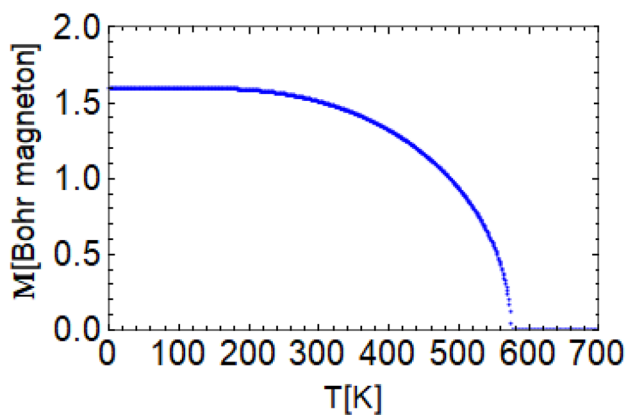
Fig. 9 The magnetization dependence on temperature, in zero field, for amorphous Fe

materials with FOPT, the low temperature ($\Theta < 0$) curves do not collapse but clearly diverge as the field changes [41].

3.7 Amorphous Fe

The amorphous alloys are known to have advantages in the field of magnetic refrigeration due to several factors [42]. Because of these advantages, we have calculated the temperature dependence of the magnetic moment in amorphous Fe using the mean-field model as well. Figure 9 displays our calculation. The exchange coefficient between Fe atoms in the amorphous case is much less (~ 30%) than its crystalline counterpart as reported by Grinstaff [43]. Both of the magnetic moment, at very low temperatures, and the Curie temperature are significantly reduced relative to crystalline Fe. In particular, T_c is about 580 K and the magnetic moment is about 1.6 μ_B.
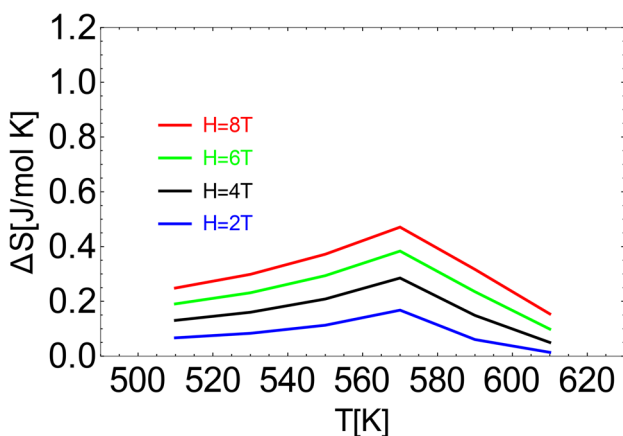

Fig. 10 The isothermal change in entropy for amorphous Fe

We have calculated the isothermal entropy change for a-Fe. The results are shown in Fig. 10. The curves also have their maximum at T_c, but their maxima ΔS are close to those of crystalline Fe, in the same field.

The absence of data on the total heat capacity of a-Fe, up to our knowledge, does not enable us to calculate the adiabatic change in temperature and compare it with that of crystalline Fe.

4 Conclusions

We have calculated the thermomagnetic and the magneto-caloric properties for Fe, Co and Ni using the mean-field theory. The isothermal entropy change ΔS_m has been calculated using Maxwell's relation. The highest ordinary ΔS_m for Fe, Co, and Ni, respectively, is 0.3, 0.23, and 0.17 J/molK for a magnetic field change of 6 T. The adiabatic temperature change ΔT_{ad} for Fe, Co, and Ni respectively, is 9, 8.6, and 2.8 K for a magnetic field change of 6 T. The relative cooling power RCP (T) is fairly comparable to those of Gd and some Gd-based MCE materials. Amorphous Fe has much less Curie temperature and a smaller spontaneous magnetic moment than its crystalline counterpart; however, its isothermal entropy change is comparable to crystalline Fe. The temperature and field dependences of the magnetization, magnetic entropy, magnetic specific heat, Arrott plots, and universal curves showed that the phase transition in these three elements is of the second order. The mean-field theory proved to be appropriate for calculating the abovementioned properties.

Declarations

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