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Thermodynamic properties of quasi-equilibrium magnons in crystalline bulk materials and thin films

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Abstract

The temperature dependence of spontaneous magnetization and specific heat for a system of magnons are investigated systematically. The correspondence of this system with the Bose-Einstein condensation phenomenon is used. Calculations are performed for both crystalline bulk materials and thin films. The calculated results are consistent with the available experimental data, indicating that our suggested approach is a suitable one for describing the thermodynamic properties for the magnon system. The thermodynamical parameters for magnons system have many similarities with the thermodynamical parameters for boson atomic gas. Results for crystalline bulk material can be used as a zeroth order approximation in any perturbative treatment of thin film. In contrast to a previous study, our method involves only analytic calculations.

Key Words: Bose-Einstein condensation of magnons, spontaneous magnetization, specific heat for magnetic materials.

1. Introduction

Recently, the phenomenon of Bose-Einstein condensation (BEC) has been experimentally realized in some quantum antiferromagnetic (AF) compounds and thin films [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. In this sense, the idea of BEC of magnons has been used for describing the phase transition in antiferromagnets from their nonmagnetic state to a magnetically ordered state under the influence of an external magnetic field h. The most important point is that the induced appearance of magnetization in finite magnetic fields at room temperature can formally described and interpreted in the language of condensation of magnetic excitations. In this case, BEC is not a collection of atoms but rather is a collection of an accumulation of a quasi-particles excitation of multi-particle systems with integer spin in one of the quantum states [12, 13].

In fact, for magnon system, equilibrium is not required, only the existence of quasi-equilibrium with non-zero chemical potential is sufficient [14, 15]. This system is created by increasing the number of magnons above the thermodynamic equilibrium level. Indeed, experimentally this scenario is realized, for example, by applying an external energy flow [12, 13]. When $h > h_c$, where h_c is the critical value of the applied magnetic filed, all spins align parallel to each other and excitations of the system are gapped ferromagnetic (FM) magnons, the number of magnons would be infinite in this case. As the field becomes lower than h_c, the gap closes and long-range AF ordering appears in the plane perpendicular to the field that corresponds to condensation of magnons with momentum equal to the AF vector. The magnetization (density of condensed magnons) is proportional to the value $h_c - h$. This value plays the same role played by the chemical potential in the condensation of atoms. When $h_c - h << h_c$ one can use the well-known results for dilute ideal Bose gas. The point $h_c = h$ is a quantum critical point that belongs to BEC universality class [16] and corresponds to the freezing of the chemical potential at zero value for a homogeneous atomic system. Furthermore, in spin systems, no two magnons can occupy the same site; thus there is a hard-core type interaction between them. This interaction finitizes the number of magnons.

The present paper is meant to be a new theoretical account for some measurable thermodynamical parameters for a system of magnons in crystalline bulk materials and thin films [17, 18]. Generally, an efficient method for describing these systems is the semiclassical approximation, density of states approach. This approach has been employed in considering the properties of condensed ideal gases trapped in power-law potentials, finite size effect [20, 21, 22, 23, 24, 25, 26, 27, 28], and for investigating interacting Bose gas confined in a 3D harmonic trap [29, 30, 31, 32]. Our results show that spontaneous magnetization falls monotonically to zero as temperature T approaches the critical temperature T_c and exhibits a tail for $T/T_c > 1$ for both geometries. Full agreement between theoretical calculation and experimental data was obtained. The specific heat exhibits a maximum at the same temperature at which BEC occurs, and drops rapidly with increasing temperature above the critical temperature. The specific heat is perfectly continuous and smooth at its maximum. The transition point, based on the maximum of the specific heat, is the most useful general criterion for BEC in this system [33]. Finally, results for the thermodynamic parameters of the crystalline bulk materials survive as the thermodynamic limit for the thin films versions of the materials.

The paper is organized as follows: the model and general relations are outlined in Section 2. Section 3 is devoted to the thermodynamic parameters for crystalline bulk materials. Thermodynamic parameters for parameters for thin films is given in Section 4. Conclusion is given in Section 5.

2. Model and general relation

In the following we introduce the necessary theoretical basis for this study. Generally in discussing the magnetic properties, both the excited states, as well as the ground state of the magnetic system might be considered. However, in this case one can be only able to calculate the thermodynamic properties of this system.

In the following we introduce the necessary theoretical basis for this study. In discussing the magnetic properties, both the excited states as well as the ground state of the magnetic system are generally considered. However, in this case one can be only able to calculate the thermodynamic properties of this system.

We start from the Hamiltonian of spin **s** quantum antiferromagnets on a 3-dimensional lattice with N

sites and lattice spacing a in a uniform magnetic field in z-direction:

$$\mathcal{H} = \frac{1}{2}\sum_{ij} \mathbf{J}_{ij}\mathbf{s}_i \cdot \mathbf{s}_j - h\sum_{i} \mathbf{s}_i^z, \tag{1}$$

where the sums are taken over all sites of the lattice, and $\mathbf{s}_i$ and $\mathbf{s}_j$ are spin operators. We assume nearest neighbor, and $\mathbf{J}_{ij} = 0$ otherwise. For sufficiently large h, the ground state of equation (1) is a saturated ferromagnetic with magnetization parallel to the field.

Within the second quantization approach the Hamiltonian can be written as [34]

$$\mathcal{H} = \sum_{\mathbf{q}} E_{\mathbf{q}}(\mathbf{q}) n_{\mathbf{q}}, \tag{2}$$

where $E_{\mathbf{q}}(\mathbf{q})$ is the dispersion energy, for cubic lattices $E_{\mathbf{q}} \propto \mathbf{q}^2$ for small $\mathbf{q}$. In equation (2) $n_{\mathbf{q}}$ is interpreted as the number of spin waves of wave vector $\mathbf{q}$, with quanta referred to as magnons. To show how the spontaneous magnetization and specific heat varies with temperature for these systems, used is the fact that magnons are bosons. Neglecting the interactions between spin waves, the main thermodynamical parameters can be calculated from the average number of magnons $< n_{\mathbf{q}} >$.

With N being the number of spins per unit volume, $M_0 = N\mu$ being the total magnetization per unit volume at $T = 0$, and μ as the magnetic moment per particle, the spontaneous magnetization is given by

$$M = M_0 - \mu \sum_{\mathbf{q}} < n_{\mathbf{q}} > . \tag{3}$$

Unfortunately it is difficult to find a reliable analytical approximation for the specific heat which allows us to study whether it has a maximum; and if it does, at which temperature it occurs. Instead one must treat the specific heat for magnons by differentiation the relevant sum for the internal energy U with respect to temperature, i.e.

$$C(T) = \frac{\partial U}{\partial T} = \frac{\partial}{\partial T}\sum_{\mathbf{q}} (\hbar\omega_{\mathbf{q}}) < n_{\mathbf{q}} > \tag{4}$$

The sum in equations (3) and (4) cannot evaluated analytically in a closed form. Another possible way to do this analysis is to approximate the sum directly by integrals. A crucial feature in obtaining a reliable semiclassical approximation is to use an appropriate density of states $\rho(E)$ [19, 20, 21, 29, 30, 31, 32].

3. Thermodynamical parameters for crystalline bulk materials

For crystalline bulk materials, the energy distribution of the magnon in a state of wave vector $\mathbf{q}$ is given by [34]

$$< n_{\mathbf{q}} > = \frac{1}{e^{\beta(\epsilon_{\mathbf{q}} - \mu_m)} - 1} = \sum_{j=1}^{\infty} z^j e^{-j\beta\epsilon_{\mathbf{q}}}, \tag{5}$$

where $\beta = (1/K_{\mathrm{B}}T), K_{\mathrm{B}}$ is the Boltzmann constant, and μ_m is the chemical potential. The fugacity z is defined as $z = e^{\beta\mu_m}$. Substitution from equation (5) in equation (3) and (4), and using the semiclassical approximation, leads to

$$\begin{aligned} M &= M_0 - \mu \sum_{j=1}^{\infty} z^j \sum_{\mathbf{q}} e^{-j\beta\epsilon_{\mathbf{q}}} \\ &= M_0 - \mu \sum_{j=1}^{\infty} z^j \int_0^{\infty} \rho(E) e^{-j\beta E} dE \end{aligned} \tag{6}$$

for spontaneous magnetization, and

$$\begin{aligned} C_{\mathrm{v}}(T) &= \left(\frac{\partial U}{\partial T}\right)_{\mathrm{v}} = \frac{\partial}{\partial T} \sum_{\mathbf{q}} (\hbar\omega_{\mathbf{q}}) < n_{\mathbf{q}} > \\ &= \frac{\partial}{\partial T} \sum_{j=1}^{\infty} z^j \int_0^{\infty} E\rho(E) e^{-j\beta E} dE \end{aligned} \tag{7}$$

for the specific heat at constant volume. Restricting ourselves to low temperatures, only the occupation numbers of the lowest excited states will be significant; and thus one must use the quadratic form of ω_q for small $\mathbf{q}$. For cubic lattices $E_{\mathbf{q}} \propto q^2$.

The approximation most often applied to the density of states $\rho(E)$ is such that $\rho(E)$ is proportional to the square root of the energy, which corresponds to the quadratic dispersion law. The density of states for the general 3D case has the form of $\rho(E) \propto \epsilon^{1/2}$, so the integral $\int \frac{\rho(E)dE}{z^{-1}e^{\beta\epsilon_q}-1}$ converges, giving rise to a finite number of magnons in the excited states. However, the accurate density of states in this case is given by

$$\rho(E) = \frac{1}{\Gamma(3/2)} \frac{E^{1/2}}{(\hbar\omega_{\mathbf{q}})^{3/2}}. \tag{8}$$

Now it is straightforward to calculate the spontaneous magnetization, and the specific heat.

Spontaneous magnetization for magnons is obtained from equation (6) and equation (8) and is given by

$$M = M_0 - \mu g_{3/2}(z) \left(\frac{K_{\mathrm{B}}T}{\hbar\omega_{\mathbf{q}}}\right)^{3/2}, \tag{9}$$

where $g_s(z) = \sum_j \frac{z^j}{j^s}$ is the Bose function. Equation (9) shows that $M_0 - M \propto T^{3/2}$, the famous law first obtained by Bloch, has been confirmed experimentally. At temperatures less than or equal to T_{c} equation (9) can be written as

$$\frac{M}{M_0} = 1 - \left(\frac{T}{T_{\mathrm{c}}}\right)^{3/2}, \tag{10}$$

where T_{c} is given by setting $\frac{M}{M_0} = 0$ and $z = 1$ (i.e. $h = h_c$) in equation (9) [27, 28]:

$$T_{\mathrm{c}} = \frac{\hbar\omega_{\mathbf{q}}}{K_{\mathrm{B}}} \left(\frac{M_0}{\zeta(3/2)}\right)^{2/3}. \tag{11}$$

This critical temperature is the temperature of the antiferromagnetic transition; it is a quantum critical point that belongs to BEC universality class [16]. This temperature corresponds to the critical temperature for BEC in an atomic gas with the number density of the atomic gas replaced by the magnetization density. So T_c distinguishes between two antiferromagnets material phases: between their nonmagnetic and magnetically ordered state.

The above approximated result for spontaneous magnetization precisely reproduces the experimental results. A comparison between calculated results from equation (10) and the experimental data of Takayama et al. [17] for $Sr_8CaRe_3Cu_4O_{24}$ ferrimagnetic compound is given in Figure 1. As shown in this figure, increasing

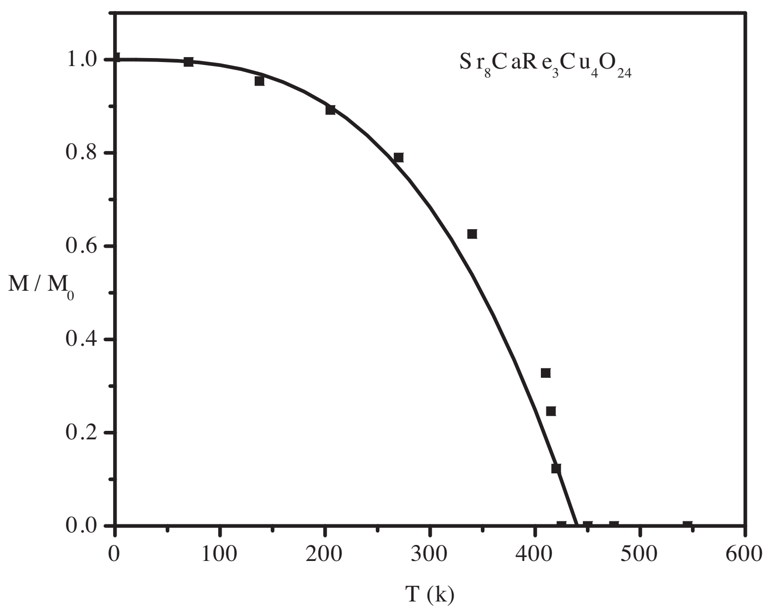

Figure 1. Spontaneous magnetization fraction as a function temperature T. The solid circles are the measured data reported in Reference [17] for $Sr_8CaRe_3Cu_4O_{24}$ material. The critical temperature for this material is given by $T_c = 440\,K$. The solid line is the results calculated from equation (10).

temperature T leads to a smooth decrease to zero in the spontaneous magnetization at $T = T_c$. This behavior classifies the transition in AF compounds from their nonmagnetic to magnetic states as a first-order phase transition. This compound is a magnetic insulator which has spontaneous magnetization at room temperature and, in some respects, resembles parent materials of high T_c superconductors.

For magnon systems, the specific heat is typically given at constant volume, and is thus an important state variable. When crossing a phase transition, its temperature dependence measures the degree of the changes in the system above and below the critical temperature. For $T < T_c$, i.e. $z = 1$, only the condensed component of the magnon gas contributes to the specific heat. For $T > T_c$, the magnetization $M = 0$. Substitution of equation (8) into equation (7) leads to

$$\frac{C_v(T)}{NK_B} = \frac{15}{4}\frac{\zeta(5/2)}{\zeta(3/2)}\left(\frac{T}{T_c}\right)^{3/2}, \qquad \text{for } T \leq T_c \tag{12}$$

$$= \left[\frac{15}{4}\frac{g_{5/2}(z)}{g_{3/2}(z)} - \frac{9}{4}\frac{g_{3/2}(z)}{g_{1/2}(z)}\right], \quad \text{for } T > T_c \tag{13}$$

In the limit $z \to 1$, the second term in equation (13) vanishes because of the divergence of $g_{1/2}(z)$, while the first term gives exactly the results appearing in equation (12). So, the specific heat is continuous at the critical temperature. The approximation formula for the specific heat above T_c is given by

$$\frac{C_v(T)}{NK_B} = 1.496 + 0.341\left(\frac{T_c}{T}\right)^{3/2} + 0.089\left(\frac{T_c}{T}\right)^{3}, \text{ for } T > T_c, \tag{14}$$

where the expansion properties of the Bose function $g_s(z) = \sum_j \frac{z^j}{j^s}$, and the values of zeta function, $\zeta(5/2) = 1.34149$ and $\zeta(3/2) = 2.61238$, are used here [35]. Equation (14) yields the exact values of C_v and $\frac{\partial C_v}{\partial T}$ at critical temperature T_c. Equation (7) enabled us to calculate the discontinuity of the specific heat at T_c as

$$\left(\frac{\partial C_v(T)}{\partial T}\right)_{T_c^-} - \left(\frac{\partial C_v(T)}{\partial T}\right)_{T_c^+} = 27\frac{\zeta(3/2)^2}{16\pi}\frac{NK_B}{T_c}.$$

The specific heat as a function of temperature for $Sr_8CaRe_3Cu_4O_{24}$ is shown in Figure 2. The specific heat

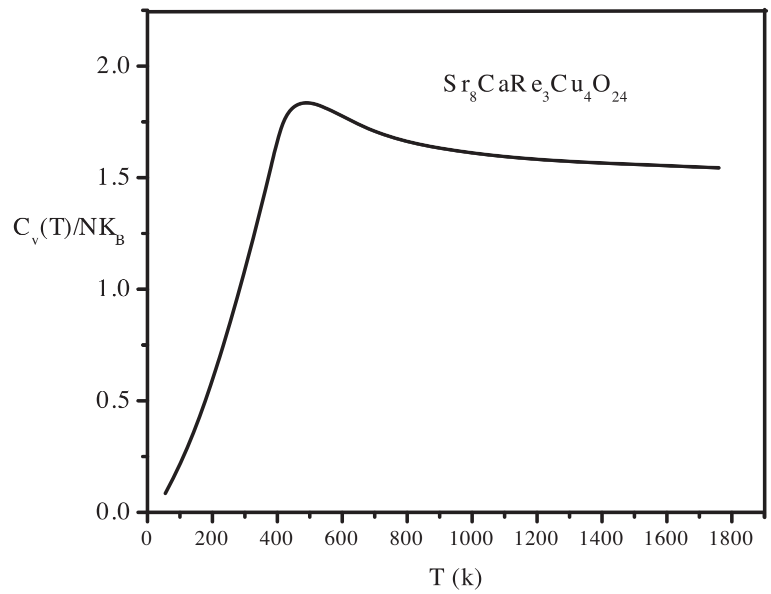

Figure 2. Temperature dependence of the specific heat for $Sr_8CaRe_3Cu_4O_{24}$ material. The critical temperature for this material is $T_c = 440$ K [17].

have a maximum at $T = T_c = 440$ K, and its behavior is consistent with the scaling property of the 3D Heisenberg model: $C \propto (\Delta T)^{-\alpha}, \alpha = -0.1162 < 0$ [36].

4. Thermodynamical parameters for thin films

Spontaneous magnetization of thin films can be calculated by using the density of states approach, if it is considered as a collective property of the whole film. The average number of magnons in this case is a function of the number of sites N and the film thickness n. The above assumption gives the average number of magnons, which leads to convergent results for spontaneous magnetization in the form [37]

$$\begin{aligned} < n_{\mathbf{q}} > &= \frac{1}{e^{\beta(\epsilon_{\mathbf{q}}-\mu_m)}-1} - \frac{1}{e^{\beta(N^2n+1)(\epsilon_{\mathbf{q}}-\mu_m)}-1}, \\ &= \sum_{j=1}^{\infty} z^j e^{-j\beta\epsilon_{\mathbf{q}}} - \sum_{j=1}^{\infty} z^j e^{-j\beta(N^2n+1)\epsilon_{\mathbf{q}}}. \end{aligned} \tag{15}$$

For sufficiently thick films the formula for $\rho(E)$ given in equation (8) can be used, except that $\omega_{\mathbf{q}}$ is replaced by $\Omega_{\mathbf{q}} = (\omega_{q_x}\omega_{q_y}\omega_{q_z})^{1/3}$ is the geometrical average of the magnons frequencies.

As in the case of crystalline bulk materials, the spontaneous magnetization for thin films is given by

$$M = M_0 - \mu g_{3/2}(z) \left(\frac{K_{\rm B}T}{\hbar\Omega_{\mathbf{q}}}\right)^{3/2} \left[1 - \frac{1}{(N^2n+1)^{3/2}}\right]. \tag{16}$$

The spontaneous magnetization at low temperature shows an oscillating character which depends on the film thickness (second term in equation (16)) and can be interpreted as a finite size effect such as the finite size effect obtained in the case of the dilute atomic gases [19]. At temperature equal to or less than the critical temperature $T_{\rm c}$, equation (16) can be written as

$$\frac{M}{M_0} = 1 - \left(\frac{T}{T_{\rm c}}\right)^{3/2} \left[1 - \frac{1}{(N^2n+1)^{3/2}}\right], \tag{17}$$

where $T_{\rm c}$, in this case, is given by

$$T_{\rm c} = \frac{\hbar\Omega_{\mathbf{q}}}{K_{\rm B}} \left(\frac{M_0}{\zeta(3/2)}\right)^{2/3} \left[1 - \frac{1}{(N^2n+1)^{3/2}}\right]. \tag{18}$$

Thus $T_{\rm c}$ shifts to a lower value due to finite size effects (i.e., thickness of the film). Decrease in critical temperature is proportional to $\left[\frac{1}{N^2n+1}\right]$. The specific heat is given by

$$\frac{C_{\rm v}(T)}{NK_{\rm B}} = \left[\frac{15}{4}\frac{\zeta(5/2)}{\zeta(3/2)}\left(\frac{T}{T_{\rm c}}\right)^{3/2}\right] \left[1 - \frac{1}{(N^2n+1)^{3/2}}\right], \tag{19}$$

for $T \leq T_{\rm c}$, and

$$\begin{aligned} \frac{C_{\rm v}(T)}{NK_{\rm B}} &= \left[1.496 + 0.341\left(\frac{T_{\rm c}}{T}\right)^{3/2} + 0.089\left(\frac{T_{\rm c}}{T}\right)^{3}\right] \\ &\quad \left[1 - \frac{1}{(N^2n+1)^{3/2}}\right], \end{aligned} \tag{20}$$

for $T \geq T_{\rm c}$.

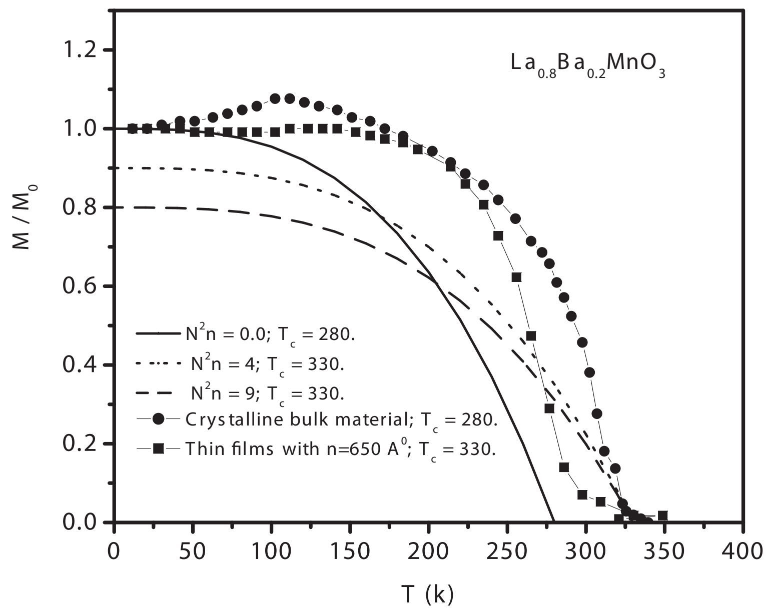

Figure 3. Spontaneous magnetization fraction as a function temperature T. The solid circles and squares are the measured data reported in Reference [18] for $La_{0.8}Ba_{0.2}MnO_3$ epitaxial thin films.

Results for the spontaneous magnetization, and the specific heat are basically identical to that found in crystalline bulk materials with a correction term due to finite size effects. There is no need to repeat the analysis here.

In Figure 3 the spontaneous magnetization for $La_{0.8}Ba_{0.2}MnO_3$ thin films is given. Solid square and circles are the experimental data of Kanki et al. [18]. Solid, dashed, and dotted lines are the calculated results

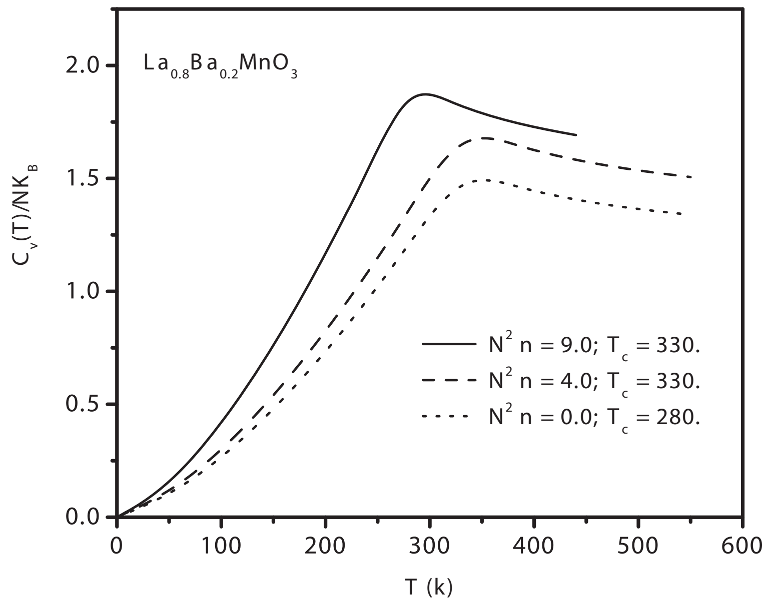

Figure 4. Temperature dependence of the specific heat for $La_{0.8}Ba_{0.2}MnO_3$ epitaxial thin films. The critical temperature for this material is $T_c = 330k$ and $n = 650A^o$ [17].

from equation (18). Temperature dependent specific heat for different thickness are given in Figure 4 for $N^2n =$ 0.0, 4 and 9. Increasing the film thickness leads to shifting the maximum value of the specific heat to lower values.

For films thicker than a critical thickness, a condensation of magnons occurs at the same temperature as that for crystalline materials ($N \gg 1$). This behavior suggests that the crystalline materials can be considered in the thermodynamic limit ($N \to \infty$) for thin films.

5. Conclusion

Based on a realistic quantum spin model in the hardcore Boson representation, the standard semiclassical approximation is used to calculate the thermodynamic parameters of magnons. As a first step toward a systematic study, the sums for the thermodynamic quantities are converted directly into ordinary integrals with an appropriate density of states. The spontaneous magnetization, and the specific heat, are investigated for both crystalline bulk materials and thin films. The spontaneous magnetization decreases monotonically toward zero with increasing temperature. The specific heat have a maximum value at a particular critical temperature T_c. This behavior has been attributed to the similarity between the system of magnons and the BEC of atoms. Our results provide a good agreement with experimental data for $Sr_8CaRe_3Cu_4O_{24}$ and $La_{0.8}Ba_{0.2}MnO_3$ [17, 18]. Finally, it would be prudent to use the results for the crystalline bulk materials as the zeroth order approximation in any perturbative treatment for the thin films studies. This concluding remark is similar to the finite size effects known in a gas of atoms [19].

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