Journal of Magnetism and Magnetic Materials 350 (2014) 30–34

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Magnon contribution to the thermodynamic properties of the iron based pnictides $SrFe_2As_2$

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ARTICLE INFO

Article history:
Received 15 April 2013
Received in revised form
27 July 2013
Available online 28 August 2013

Keywords:
Spin wave
Specific heat
Heat capacity pnictide

ABSTRACT

We investigate some of the thermodynamic properties of iron based pnictides, using an effective J_1–J_2–$J_\perp$–K spin-model containing only the local Fe moments. Both the multi-layered structure of the material, as well as the single ion anisotropy, are taken into account through the $J_\perp$ and K couplings, respectively. We derive a fully analytic expression for the magnetic contribution to the specific heat when $J_\perp = 0$; the case when $J_\perp \neq 0$ is evaluated numerically. Both solutions (with no adjustable parameters) are found to compare favorably with the reported experimental data for $SrFe_2As_2$. An improvement is, however, noticed for the $J_\perp \neq 0$ case, consistent with the layered character of these weakly coupled magnetic structures. We discuss the applicability of our formalism to other oxy/pnictides, or related magnetic materials.

1. Introduction

The huge amount of research activity devoted to the new iron based pnictides can be understood not only as a result of their ability to sustain high temperature superconductivity upon doping [1–3], or yet for their interesting magnetic properties, but also from the interplay between superconductivity and magnetism though these are usually considered antagonistic [4,5]. It is remarkable that both superconductivity and magnetism are correlated with the structural properties. The later correlation may be behind the often observed structural and magnetic phase transitions manifested in, e.g. the undoped, so-called 1111 (e.g. R $FeAsO_{1-x}F_x$; R=rare earth [see e.g. Ref. [4] and references cited therein]) or 122 (e.g. A Fe_2As_2; A=Ba, Sr, Ca) systems (see e.g. Refs. [6–9] and references cited therein). It has been a matter of debate in the literature whether or not these transitions are of the first or second order, or if they occur simultaneously or independently, or whether or not they are connected to the unusual magnetic and quasiparticle spectrum at low energies. Recently, we have argued that it is exactly these magnetic phase transitions that allow for the emergence of Dirac quasiparticles with a linear dispersion relation in the electronic spectrum [10]. Also, it is shown that, due to the antiferromagnetic structure with two Néel sublattices, there are two magnon gaps at the Γ point [10]. We have demonstrated that both results are consistent with ARPES and neutron scattering analyses, but it still remains to be compared to thermodynamic experiments.

In this work we consider the contribution of such multi-branched magnetic spectrum to the thermodynamic properties of $SrFe_2As_2$ which is expected to exhibit, as compared to other AFe_2As_2 (A=Ca, Ba, Eu), the highest magnetic contribution: as can be seen in Table 1, it has the highest Néel point and the strongest magnetic moment. We calculate the magnetic contribution to the specific heat and we show that, after subtracting lattice and electronic contributions, the agreement with experiments is remarkable considering that no adjustable parameters are used. We furthermore notice that the specific heat is nearly two-dimensional in this magnetic material, revealing the weakly coupled layered nature of the magnetic structure.

2. Theoretical background

2.1. The magnetic model

The metallic, antiferromagnetic Fe–arsenides are characterized by Fe_2As_2 layers that are, in most cases, sandwiched by diamagnetic sheets [6,7]. The magnetism is related to Fe ions which have a positive on-site single-ion anisotropy K (moment polarized within the plane) and are strongly coupled within each layer and weakly coupled among the different layers. Recently, Da Conceição et al. [10] showed that the magnetism of such a configuration (for simplicity, assumed to consist of coupled two-dimensional square layers) can be captured by a J_1–J_2–$J_\perp$–K model ($S > 1/2$):

$$\hat{H} = J_1 \sum_{\langle i,j\rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + J_2 \sum_{\langle\langle i,j\rangle\rangle} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_j + J_\perp \sum_{\langle \ell,m\rangle} \hat{\mathbf{S}}_\ell \cdot \hat{\mathbf{S}}_m - K \sum_i (\hat{S}_i^z)^2, \quad (1)$$

where $J_1 > 0$, $J_2 > 0$ and $J_\perp > 0$ are, respectively, the antiferromagnetic superexchanges between the intralayer nearest-neighbors $\langle i,j\rangle$, intralayer next-to-nearest neighbors $\langle\langle i,j\rangle\rangle$, and neighboring-interlayer

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http://dx.doi.org/10.1016/j.jmmm.2013.08.017

Table 1
Some relevant magnetic properties of AFe_2As_2 (A=Ca, Sr, Ba). μ_{ND} is the neutron-diffraction-determined magnetic moment of Fe which orders with a wave vector (1,0,1) (along an orthorhombic unit-cell). T_S and T_S denote, respectively, the temperature of the structural and magnetic transitions. KFe_2As_2 [11,12] manifests no T_S transition but its $\gamma = 60$ mJ/mol K^2 and $\theta_D = 201$ K. $EuFe_2As_2$ has T_S=195 K but the magnetic contribution of Eu sublattice [13] impedes an unambiguous determination of γ and θ_D. Finally, at present, no high-temperature caloric information is available on AFe_2As_2 (A=Cs [14] and Rb [15]).

x	$\vec{\mu}_{ND}$ (μ_B)	Δ (meV)	Structural T_s (K)	Transition order	Magnetic T_m (K)	Transition order	γ (J/mol K^2)[a]	θ_D (K)[b]	Refs.
$CaFe_2As_2$	$0.8\vec{a}$	6.9	173	First	173	First	4.7	258	[16–18]
$SrFe_2As_2$	$1\vec{a}$	6.5	201.5	First[c]	201.5	First[c]	6.5	245	[12,19–25]
$BaFe_2As_2$	$0.87\vec{a}$	9.8	134.5	Second[c]	133.75	First[c]	6.1	186	[8,9,25–34]

[a] γ are taken from the reported values (see Ref. [6] and references therein).
[b] θ_D (used in Figs. 1–3) are slightly different from the ones quoted in Ref. [6].
[c] The thermodynamic nature of both T_s and T_m transitions is highly controversial (see references). The quoted values and identification of $SrFe_2As_2$ and $BaFe_2As_2$ are taken from Refs. [19] and [8], respectively.

$\langle \ell, m\rangle$ spins $\hat{\mathbf{S}}_i$. The average intralayer spacing is a while the interlayer spacing among consecutive layers is d.

At lower temperatures, well below the magnetic transition but still with no magnons interaction, the magnetic Hamiltonian can be diagonalized exactly as [10]

$$\hat{H} = \sum_{\mathbf{k},k_\perp} \hbar\omega(\mathbf{k},k_\perp) a^\dagger_{\mathbf{k},k_\perp} a_{\mathbf{k},k_\perp} + \sum_{\mathbf{k},k_\perp} \hbar\Omega(\mathbf{k},k_\perp) A^\dagger_{\mathbf{k},k_\perp} A_{\mathbf{k},k_\perp}, \tag{2}$$

where $a^\dagger_{\mathbf{k},k_\perp}$ ($a_{\mathbf{k},k_\perp}$) and $A^\dagger_{\mathbf{k},k_\perp}$ ($A_{\mathbf{k},k_\perp}$) are the creation (annihilation) magnon operators for the acoustic and optical branches, respectively. The dispersions corresponding to these two magnetic modes will depend on whether or not $J_\perp$ and/or K are included. For the layered case wherein $J_\perp \neq 0$ and single-ion anisotropy $K \neq 0$, the spectrum is

$$\hbar\omega_{ac}(\mathbf{k},k_\perp) = \hbar c_- \sqrt{F_+(\mathbf{k}) + \Delta^2}, \tag{3}$$

$$\hbar\Omega_{op}(\mathbf{k},k_\perp) = \hbar c_+ \sqrt{F_-(\mathbf{k}) + \Delta^2 + \eta}, \tag{4}$$

where

$$\xi = \frac{2J_1}{J_2}, \tag{5}$$

$$\upsilon = \xi\left[1 + \frac{1}{4}\left(\frac{J_1^2}{4J_2^2 - J_1^2}\right)\right], \tag{6}$$

$$\Delta^2 = \frac{(S-\frac{1}{2})K}{Sa^2J_2}\left[1 + \frac{1}{8}\left(\frac{J_1^2}{4J_2^2 - J_1^2}\right)\right], \tag{7}$$

$$\eta = \frac{\xi}{a^2}\left(\frac{J_1^2}{4J_2^2 - J_1^2}\right), \tag{8}$$

$$F_\pm(\mathbf{k}) = \mathbf{k}^2 + 2\zeta(1 - \cos k_\perp d) \mp \upsilon k_x k_y, \tag{9}$$

$$\zeta = 2J_\perp J_2, \tag{10}$$

$$1/c_\pm^2 = \frac{\hbar^2}{8S^2a^2\left(4J_2^2 - J_1^2\right)}\left(1 \pm \frac{\sigma^2 J_1^2}{2J_2^2}\right), \tag{11}$$

where c_+ (c_-) is the optical (acoustic) spin-wave velocity, and σ is the spin projection. The contribution of each magnon mode to the total energy can be straightforwardly calculated once it is recalled that

$$\langle a^\dagger_{\mathbf{k},k_\perp} a_{\mathbf{k},k_\perp}\rangle = n_B(\hbar\omega(\mathbf{k},k_\perp)),$$
$$\langle A^\dagger_{\mathbf{k},k_\perp} A_{\mathbf{k},k_\perp}\rangle = n_B(\hbar\Omega(\mathbf{k},k_\perp)), \tag{12}$$

where, as usual, $n_B(\hbar\omega)$ is the Bose–Einstein distribution function. The corresponding energy contributions are now

$$E_{ac} = V \sum_{k_\perp = -\pi/d}^{\pi/d} \int \frac{\mathrm{d}^2\mathbf{k}}{(2\pi)^2} \hbar\omega(\mathbf{k},k_\perp)\, n_B(\mathbf{k},k_\perp), \tag{13}$$

$$E_{op} = V \sum_{k_\perp = -\pi/d}^{\pi/d} \int \frac{\mathrm{d}^2\mathbf{k}}{(2\pi)^2} \hbar\Omega(\mathbf{k},k_\perp)\, n_B(\mathbf{k},k_\perp), \tag{14}$$

where V is the unit-cell volume. All relevant thermodynamic properties can be calculated from these two equations.

2.2. 2D magnetic specific heat: an analytical expression

Let us now derive an exact and analytical expression for the purely 2D magnetic contribution to the specific heat. For this purpose, we temporarily switch off the interlayer coupling, $J_\perp = 0$, in which case no discrete sum over $k_\perp$ is to be performed in the energies (Eqs. (13) and (14). Once the 2D expression is obtained, the discrete sum over $k_\perp$ will be performed numerically.

The total magnetic energy of all excitations is

$$\overline{E} = E_{ac} + E_{op}, \tag{15}$$

where E_{ac} and E_{op} are given by Eqs. (13) and (14). The magnetic specific heat at constant volume is given by

$$C_M = \frac{1}{V}\frac{\mathrm{d}\overline{E}}{\mathrm{d}T}. \tag{16}$$

It is recalled that there are two gaps at $\mathbf{k}$=0: one for the softer (acoustic) mode, see Eqs. (3) and (13), which is given by $\Delta_{ac} = \Delta$, and another for the harder (optical) mode see Eqs. (4) and (14), which is given by $\Delta_{op} = \sqrt{\Delta^2 + \eta}$. Then on assuming magnon ideal gas conditions in Eqs. (3) and (4), one arrives at an expression for the 2D magnetic specific heat

$$C_{2D}(T) = C_{2D}^{ac}(T,\Delta_{ac}) + C_{2D}^{op}(T,\Delta_{op}), \tag{17}$$

which is a sum over the two types of magnons, each one giving a contribution to the specific heat of the form

$$C_{2D}(T,\Delta) = \frac{a^2 N_A k_B^3 \Delta^3}{f\pi\hbar^2 c_\pm^2 \sqrt{4-\upsilon^2}}\left(\frac{1}{T}\right) \times\left\{\ln(Z) + 3\frac{T}{\Delta}L_1(Z) + 6\left(\frac{T}{\Delta}\right)^2 L_2(Z) + 6\left(\frac{T}{\Delta}\right)^3 L_3(Z)\right\}, \tag{18}$$

here $L_i(Z)$ is the usual polylogarithm function with an argument $Z = \exp(-\Delta/T)$, f is the number of chemical formula per unit square-cell (see above), N_A is the Avogadro number and υ is given in Eq. (6).

For $T \ll \Delta$, Eq. (18) reduces to the exponentially decaying expression

$$C_{2D}^{LT}(T) = \frac{a^2 N_A k_B^3}{f\pi\hbar^2 c_\pm^2 \sqrt{4-\upsilon^2}}\frac{\Delta^3}{T}\exp\left(-\frac{\Delta}{T}\right), \tag{19}$$