Communications in Physics, Vol. 23, No. 3 (2013), pp. 235-240

INFLUENCE OF SPIN FLUCTUATION ON THE MAGNETIC PROPERTIES OF EUO ULTRA-THIN FILM

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Received 25 June 2013
Accepted for publication 15 September 2013

Abstract. *The Gaussian spin fluctuation theory is applied to study magnetic properties (Curie temperature, magnetization) of EuO ultra-thin films within nearest neighbor and next nearest neighbor exchange approximation. The dependence of Curie temperature on the thickness of the free EuO ultrathin film is calculated and compared with the results of the other mean field method-constant coupling approximation. The effect of nearly independence of saturation magnetization on the EuO films thickness measured in [8] can be explained by controlling the next nearest neighbor exchange in presence of substrates.*

I. INTRODUCTION

In the recent years, magnetic properties of thin films which have been intensively investigated by both theoretical and experimental methods expose a great potential for applications [1]. The important reasons of magnetic size effects in the thin films, such as the reduction of the Curie temperature T_C, are the finite thickness and the broken translational symmetry along the direction perpendicular to the film surfaces. Many theoretical methods are used to study the magnetic properties of thin films such as, the standard mean field (MF) methods [2] including the Weiss mean field, the Oguchi's cluster and the constant coupling approximations (CCA), spin wave method [3], and recently, the integral functional integral method [4]- [5]. Our paper is aimed to apply the Gaussian spin fluctuation (SF) theory developed by us [4] for a real spin system of EuO thin film. Here we consider the next nearest neighbor (NNN) exchange parallel with the nearest neighbor (NN) exchange interactions between spins for explanation of some special effects in EuO. EuO has low magnetic anisotropy, the spin of the Eu^{2+} is localized with value $S = 7/2$. Ultrathin EuO film is used as tunnel barriers in semiconductor spintronic devices and its large exchange splitting is investigated in [6]. Europium monoxide is nearly an ideal Heisenberg ferromagnet [6], so it is a good case for testing theories for thin films. In the

second part we give some essential details of the model and the functional integral method (FIM) applied for ultrathin films.

II. BRIEF THEORY

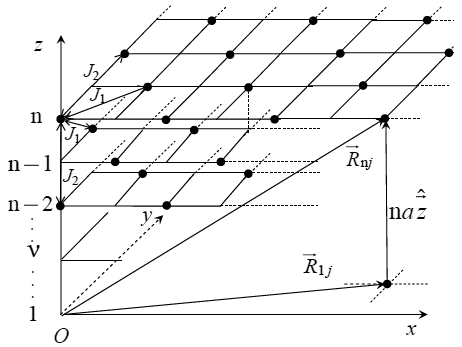

Fig. 1. A ferromagnetic thin film of the face centered cubic lattice. The spin position is defined by index νj, where ν is a plane index $\nu = 1, 2, ..., n$ and j stands for a two-dimensional lattice vector $\vec{R}_j$ in xOy plane parallel to the film surfaces . J_1 (J_2) is the NN (NNN) exchange integral.

We consider a thin film which has a face centered cubic (fcc) spin lattice comprising from n layers and there are N spins in every layer (Fig. 1). The Heisenberg Hamiltonian for the spin films, when the mean field is included in non-interacting Hamiltonian, is written as:

$$\begin{aligned} H &= H_0 + H_{int} \\ H &= -g\mu \sum_{\nu j} h_\nu S^z_{\nu j} - \frac{1}{2} \sum_{\nu j, \nu' j'} J_{\nu\nu'}(\vec{R}_j - \vec{R}_{j'}) \delta\vec{S}_{\nu j} \delta\vec{S}_{\nu' j'} \end{aligned} \tag{1}$$

with

$$h_\nu = h + (g\mu)^{-1} \sum_{\nu'} J_{\nu\nu'}(0) \langle S^z_{\nu'} \rangle \tag{2}$$

$$\delta S^z_{\nu j} = S^z_{\nu j} - \langle S^z_{\nu'} \rangle; \delta S^x_{\nu j} = S^x_{\nu j}; \delta S^y_{\nu j} = S^y_{\nu j}; \alpha = x, y, z \tag{3}$$

$$J_{\nu\nu'}(\vec{k}) = \sum_{j - j'} J_{\nu\nu'}(\vec{R}_j - \vec{R}_{j'}) exp[i\vec{k}(\vec{R}_j - \vec{R}_{j'})] \tag{4}$$

where the first term of (1) is the spin energy in the layer-dependent field h_ν consisting of the external field h and molecular (Weiss) field. The second term of (1) is the usual Heisenberg exchange interaction between spin fluctuations $\delta\vec{S}_{\nu j}$ and $\delta\vec{S}_{\nu' j'}$, which have three components described by (3). $J_{\nu\nu'}(\vec{R}_j - \vec{R}_{j'})$ denotes the exchange integral between spin fluctuations, which is considered as the matrix element of the matrix $\hat{J}(\vec{R}_j - \vec{R}_{j'})$. $J_{\nu\nu'}(0)$ is Fourier image of exchange integral taken at $\vec{k} = 0$ (see (4)).

According to [4], the free energy of the magnetic film in the Gaussian approximation has the following form:

$$F = \frac{N}{2}\sum_{\nu,\nu'} J_{\nu\nu'}(0)\langle S^z_\nu\rangle\langle S^z_{\nu'}\rangle - \frac{N}{\beta}\sum_{\nu} ln\frac{sh(S+1/2)y_\nu}{sh\frac{y_\nu}{2}}$$
$$+\frac{1}{2\beta}\sum_{\vec{k}} lndet\left|\hat{1}-\hat{A}(\vec{k})\right| + \frac{1}{\beta}\sum_{\vec{k},\omega} lndet\left|\hat{1}-\hat{B}(\vec{k},\omega)\right| \quad (5)$$

Here $\hat{A}(\vec{k})$ and $\hat{B}(\vec{k},\omega)$ are n×n matrices, which have the following form when the NN and NNN exchange interactions between spin fluctuation are taken into account:

$$\hat{A}(\vec{k}) = \begin{pmatrix} \beta b'(y_1)J_s(\vec{k}) & \beta b'(y_1)J(\vec{k}) & \beta b'(y_1)J_0(\vec{k}) & 0 & \dots & 0 \\ \beta b'(y_2)J(\vec{k}) & \beta b'(y_2)J_s(\vec{k}) & \beta b'(y_2)J(\vec{k}) & \beta b'(y_2)J_0(\vec{k}) & \dots & 0 \\ \beta b'(y_3)J_0(\vec{k}) & \beta b'(y_3)J(\vec{k}) & \beta b'(y_3)J_s(\vec{k}) & \beta b'(y_3)J(\vec{k}) & \dots & 0 \\ 0 & \beta b'(y_4)J_0(\vec{k}) & \beta b'(y_4)J(\vec{k}) & \beta b'(y_4)J_s(\vec{k}) & \dots & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & \dots & \beta b'(y_n)J_s(\vec{k}) \end{pmatrix} \quad (6)$$

and

$$\hat{B}(\vec{k},\omega) = \begin{pmatrix} \frac{\beta b(y_1)J_s(\vec{k})}{y_1-i\beta\omega} & \frac{\beta b(y_1)J(\vec{k})}{y_1-i\beta\omega} & \frac{\beta b(y_1)J_0(\vec{k})}{y_1-i\beta\omega} & 0 & \dots & 0 \\ \frac{\beta b(y_2)J(\vec{k})}{y_2-i\beta\omega} & \frac{\beta b(y_2)J_s(\vec{k})}{y_2-i\beta\omega} & \frac{\beta b(y_2)J(\vec{k})}{y_2-i\beta\omega} & \frac{\beta b(y_2)J_0(\vec{k})}{y_2-i\beta\omega} & \dots & 0 \\ \frac{\beta b(y_3)J_0(\vec{k})}{y_3-i\beta\omega} & \frac{\beta b(y_3)J(\vec{k})}{y_3-i\beta\omega} & \frac{\beta b(y_3)J_s(\vec{k})}{y_3-i\beta\omega} & \frac{\beta b(y_3)J(\vec{k})}{y_3-i\beta\omega} & \dots & 0 \\ 0 & \frac{\beta b(y_4)J_0(\vec{k})}{y_4-i\beta\omega} & \frac{\beta b(y_4)J(\vec{k})}{y_4-i\beta\omega} & \frac{\beta b(y_4)J_s(\vec{k})}{y_4-i\beta\omega} & \dots & 0 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & \dots & \frac{\beta b(y_n)J_s(\vec{k})}{y_n-i\beta\omega} \end{pmatrix} \quad (7)$$

with

$$y_\nu = \beta g\mu h_\nu \quad (8)$$

$$\omega = 2\pi l/\beta; l = 0, \pm1, \pm2, ... \quad (9)$$

Longitudinal and transversal spin fluctuation amplitudes can be calculated through the correlation functions of the spin fluctuation components (or time-ordered spin Green functions) as follows:

$$\begin{aligned} \chi^z_{\nu_1 j_1,\nu_2 j_2}(\tau_1,\tau_2) &= \chi^z_{\nu_1\nu_2}\left(\vec{R}_{j_1}-\vec{R}_{j_2},\tau_1-\tau_2\right) \\ &= \langle\hat{T}\left(S^z_{\nu_1 j_1}(\tau_1)-\langle S^z_{\nu_1}\rangle\right)\left(S^z_{\nu_2 j_2}(\tau_2)-\langle S^z_{\nu_2}\rangle\right)\rangle \end{aligned}$$

$$\chi^x_{\nu_1 j_1,\nu_2 j_2}(\tau_1,\tau_2) = \chi^x_{\nu_1\nu_2}\left(\vec{R}_{j_1}-\vec{R}_{j_2},\tau_1-\tau_2\right) = \langle\hat{T}S^x_{\nu_1 j_1}(\tau_1)S^x_{\nu_2 j_2}(\tau_2)\rangle \quad (10)$$
$$\chi^y_{\nu_1 j_1,\nu_2 j_2}(\tau_1,\tau_2) = \chi^y_{\nu_1\nu_2}\left(\vec{R}_{j_1}-\vec{R}_{j_2},\tau_1-\tau_2\right) = \langle\hat{T}S^y_{\nu_1 j_1}(\tau_1)S^y_{\nu_2 j_2}(\tau_2)\rangle$$

Within the Gaussian approximation the Fourier image of the spin correlation function (10) becomes :

$$\tilde{\chi}(\vec{k},\omega) = -\frac{1}{\beta}\left[J^{-1}_{\nu_1\nu_2}(\vec{k}) - \sum_{\nu} J^{-1}_{\nu_1\nu}(\vec{k}) C^{\alpha}_{\nu_1\nu_2}(\vec{k},\omega)\right] \tag{11}$$

where $C^{\alpha}_{\nu\nu'}(\vec{k},\omega)$ is the elements of the matrix $\hat{C}(\vec{k},\omega)$ and

$$\hat{C}^{z}(\vec{k},\omega) = \hat{C}^{z}(\vec{k}) = \left(\hat{1} - \hat{A}(\vec{k})\right)^{-1}; \hat{C}^{x(y)}(\vec{k},\omega) = \left(\hat{1} - \hat{B}(\vec{k},\omega)\right)^{-1}; \tag{12}$$

The relative magnetic moment per site of the ν^{th} spin layer in the mean field approximation (MFA) and spin fluctuation approximation (SFA) has the following forms:

$$m^{MF}_{\nu}(\beta) = \langle S^{z}_{\nu}\rangle_0 = b(y_\nu) \tag{13}$$

$$m^{SF}_{\nu}(\beta) = \langle S^{z}_{\nu}\rangle = \left\{ S(S+1) - \frac{1}{N}\sum_{\alpha,\vec{k},\omega}\left(\chi^{\alpha\alpha}_{\nu\nu}(\vec{k},\omega)\right)\right\}^{1/2} \tag{14}$$

In order to compare with experiments and other methods, an average relative (or dimensionless) magnetic moment per thin film site is given as

$$m(\beta) = \frac{M}{\mu N n} = \frac{1}{n}\sum_{\nu=1} n m_{\nu}(\beta) \tag{15}$$

Average magnetizations in the MF and SF approximations are denoted by m^{MF} and m^{SF}, respectively.

III. NUMERICAL RESULTS AND DISCUSSION

The Fourier components of exchange integrals are easily derived from the geometrical structure of fcc thin film spin lattice as:

$$J_s(\vec{k}) = J_{s_1}(\vec{k}) + J_{s_2}(\vec{k}) \tag{16}$$

with $J_{s_1}(\vec{k}) = 4J_1 \cos\left(\frac{k_x a}{2}\right)\cos\left(\frac{k_y a}{2}\right)$ and $J_{s_1}(\vec{k}) = 2J_2\left[\cos(k_x a) + \cos(k_y a)\right]$.

$$J(\vec{k}) = 2J_2\left[\cos\left(\frac{k_x a}{2}\right) + \cos\left(\frac{k_y a}{2}\right)\right] \tag{17}$$

$$J_0(\vec{k}) = J_2 \tag{18}$$

where a is a lattice constant of the thin film. In the following parts, a relative dimensionless temperature $\tau = \frac{T}{T^b_C}$ is used. T^b_C is the Curie temperature of the corresponding bulk spin lattice, in which $T^b_C = \frac{S(S+1)ZJ}{3k_B}$ (Z is the number of NN spins, for fcc spin lattice, $Z = 12$). The Curie temperature within the MF (SF) theory is obtained from a solution of an equation $m^{MF}(\tau_C) = 0$ ($m^{SF}(\tau_C) = 0$), and expressed by $\tau_C = \frac{T_C}{T^b_C}$.

The numerical calculations for the fcc EuO free thin film spin lattice at zero external magnetic field ($h = 0$) have been performed with following parameter values [2]: $S = 7/2$,

$J_1/k_B = 0.606K$, $J_2/k_B = 0.119K$. Table 1 presents the comparison of our results for T_C of EuO free thin film within the MFA and the SFA with ones of R. Rausch and W. Nolting in the CCA [2]. Since EuO has a low anisotropy, it can obey the Mermin-Wagner theorem [7]. Our SFA values for are lower than ones of MFA and CCA and agree better with the Mermin-Wagner theorem. Moreover, CCA gave unclear values of T_C when the thickness of the thin film larger 5 atomic layers (these values larger the Curie temperature for bulk EuO, $T_C^b \approx 69K$).

Table 1. Comparison of values of the Curie temperature T_C(n) which are calculated for EuO within MFA, SFA with ones within the CCA [2].

T_C(n) (K)			
n	MFA	SF	CCA
1	17.12	3.40	22.70
2	31.54	10.12	50.00
3	36.15	13.57	61.10
4	40.27	16.33	66.60
5	43.89	19.13	69.60
8	52.25	–	73.60

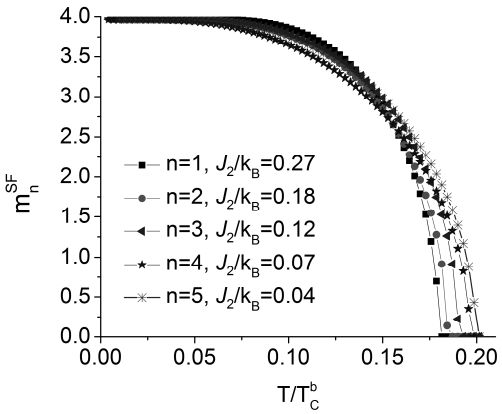

Fig. 2. The magnetization of EuO ultra-thin films with different thicknesses and NNN exchange integral as a function of temperature in SFA.

Our results given in the SF theory for free EuO films are much smaller than the experimental ones for EuO films on substrates [8]. The reason can be the influence of the substrates on the exchange interaction between the spins of EuO thin films. The substrates can make the Curie temperature of the thin films increase (or decrease). According to Ezana Negusse et al. [8], when EuO films were grown as a stepped wedge on Si/SiO2 /Cr(20$\mathring{A}$)/Cu(90$\mathring{A}$) and capped with Y(20$\mathring{A}$)/Al(80$\mathring{A}$), the film high quality and consistent magnetic properties were confirmed by superconducting quantum interference device magnetometry, *which revealed a constant saturation moment independent of film*

thickness. This phenomenon may be explained by increasing the role of NNN exchange J_2, which is in longer range than J_1, with reduction of the film thicknesses. Therefore, we fix the NN exchange integral and modify the NNN exchange integral J_2 (see Fig. 2).

From Fig. 2, one can see that the Curie temperature change only about 12% and maximum change of saturation magnetization below Curie temperature is about 3% when the film thicknesses reduce from 5 atomic layer to monolayer. This result gives the reasonable explanation for the experimental observation [8] that the saturation magnetization of EuO thin film is nearly independent on the reduction of the film's thickness.

IV. CONCLUSION

The FIM for solving Heisenberg model in quasi two dimensional case is applied to find the magnetization, Curie temperature as a function of temperature, thickness and exchange interaction of the EuO ultrathin films with and without the substrate. Our results for free EuO films given in SFA are more appropriate to Mermin-Wagner theorem than ones of CCA. In addition, we have shown that the effect of the nearly independence of the saturation magnetization on the films thickness observed experimentally for EuO thin films may be explained by enhancing the influence of NNN exchange interaction.

ACKNOWLEDGMENTS

This research is funded by Vietnam National Foundation for Science and Technology Development (NAFOSTED) under grant number 103.02.2012.37.

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