THE EXCITATION SPECTRUM OF THE UNI-AXIAL NON-HEISENBERG ANTIFERROMAGNET

M.B. GUSEINOV, V.S.TAGIEV, A.M. SULEYMANOV

Institute of Physics of Azerbaijan Republic Academy of Sciences
370143, Baku, H. Javid str., 33

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Using the standard-basis operator method in the Green function technique in random-phase-approximation anisotropic uniaxial antiferromagnets $S=3/2$ with bilinear (J), biquadratic (B) and octupolar (F) exchanges with account of atomic states multiplicity are studied. Considered the case of easy-axial antiferromagnet when uniaxial single-ion anisotropy $V>0$ and situation when $J>B,F$ that correspond to antiferromagnetic ordering. Account of multiplicity generates additional branches in spectrum of spin excitations of type $|\Delta s^z|=1,2,3$. The spectrum was analysed in various limit cases. It was established that dispersion of branches of type $|\Delta s^z|=m$ $(m=1,2,...,2S)$ cause by exchanges of type $(\vec{S}_i\vec{S}_j)^m, (\vec{S}_i\vec{S}_j)^{m+1}, ..., (\vec{S}_i\vec{S}_j)^{2S}$.

1. INTRODUCTION

There exist magnetic materials which are correctly described in the framework of the non-Heisenberg Hamiltonian in which besides of bilinear (Heisenberg) exchange the Higher order exchange terms must be taken into account [1-3].

In this case the exchange Hamiltonian for spin S is given by

$$H=\sum_{ij}\sum_{n=0}^{2S}J_n(i-j)\,(\vec{S}_i\vec{S}_j)^n \tag{1}$$

which coincides for $S=1/2$ with the Heisenberg Hamiltonian.

In [3] we have studied a ferromagnet for $S=3/2$ on the basis of the Hamiltonian (1).

In the present paper within the framework of the method of standard-basis operator (SBO) in the Green function technique [4] in the random-phase-approximation (RPA) anisotropic uni-axial non-Heisenberg antiferromagnet with the spin $S=3/2$ with the account for multiplicity of the atomic states (of all $2S+1$ states of the atom with the spin S) on the basis of exchange Hamiltonian (1) are considered.

The Hamiltonian of the system has a form

$$H=\sum_{ij}J(i-j)\,(\vec{S}_i\vec{S}_j)-\sum_{ij}B(i-j)\,(\vec{S}_i\vec{S}_j)^2+\sum_{ij}F(i-j)\,(\vec{S}_i\vec{S}_j)^3-V\sum_{r=i,j}\left(S_r^z\right)^2 \tag{2}$$

where J,B,F are the parameters of exchange (bilinear, biquadratic, octupolar) interaction (according to (1) $J=J_1$, $B=J_2$, $F=J_3$), $V>0$ is a parameter of uniaxial single-ion anisotropy and $\vec{S}_r$ is a spin operator of the site r, $J>0$, $B>0$ (model TB [5,6]), $F>0$ and $J>B,F$.

In order to take into account multiplicity of atomic states we express the components of the spin operator through the SBO [4].

For $S=3/2$ we have

$$\begin{aligned}
S_r^+ &= \sqrt{3}L_{12}^r+2L_{23}^r+\sqrt{3}L_{34}^r\ ,\\
S_r^- &= \sqrt{3}L_{21}^r+2L_{32}^r+\sqrt{3}L_{43}^r\ ,\\
S_r^z &= \frac{1}{2}\left(3L_{11}^r+L_{22}^r-L_{33}^r-3L_{44}^r\right)\ ,
\end{aligned} \tag{3}$$

where $L_{\alpha\beta}^r$ is a SBO, which describes the transition between the states α and β.

Let us note that for $S=3/2$ there are spin excitations of the types $|\Delta s^z|=1,2,3$.

2. EXCITATION SPECTRUM OF THE TYPE $|\Delta s^z|=1$.

Expressing the Hamiltonian (2) through SBO (3) and applying the properties of SBO [4] and the equation of motion

$$i\frac{dL_{\alpha\alpha'}^r(t)}{dt}=\left[L_{\alpha\alpha'}^r(t),H\right]\ , \tag{4}$$

we obtain the following equation for the Greens function in RPA

$$\begin{vmatrix}
\omega-\alpha_1-2V & 0 & 0 & -a_1D_{12} & -a_2D_{12} & -a_3D_{12}\\
0 & \omega-\alpha_2 & 0 & -a_2D_{23} & -a_4D_{23} & -a_2D_{23}\\
0 & 0 & \omega-\alpha_3+2V & -a_3D_{34} & -a_2D_{34} & -a_1D_{34}\\
a_1D_{34} & a_2D_{34} & a_3D_{34} & \omega+\alpha_3-2V & 0 & 0\\
a_2D_{23} & a_4D_{23} & a_2D_{23} & 0 & \omega+\alpha_2 & 0\\
a_3D_{12} & a_2D_{12} & a_1D_{12} & 0 & 0 & \omega+\alpha_1+2V
\end{vmatrix}
\begin{vmatrix}
\langle\langle L_{12}^1|L_{\gamma\gamma'}^n\rangle\rangle_{k,\omega}\\
\langle\langle L_{23}^1|L_{\gamma\gamma'}^n\rangle\rangle_{k,\omega}\\
\langle\langle L_{34}^1|L_{\gamma\gamma'}^n\rangle\rangle_{k,\omega}\\
\langle\langle L_{12}^2|L_{\gamma\gamma'}^n\rangle\rangle_{k,\omega}\\
\langle\langle L_{23}^2|L_{\gamma\gamma'}^n\rangle\rangle_{k,\omega}\\
\langle\langle L_{34}^2|L_{\gamma\gamma'}^n\rangle\rangle_{k,\omega}
\end{vmatrix}
=\frac{1}{2\pi}
\begin{vmatrix}
D_{12}^n\delta_{\gamma 2}\delta_{\gamma' 1}\delta_{n1}\\
D_{23}^n\delta_{\gamma 3}\delta_{\gamma' 2}\delta_{n1}\\
D_{34}^n\delta_{\gamma 4}\delta_{\gamma' 3}\delta_{n1}\\
D_{12}^n\delta_{\gamma 2}\delta_{\gamma' 1}\delta_{n2}\\
D_{23}^n\delta_{\gamma 3}\delta_{\gamma' 2}\delta_{n2}\\
D_{34}^n\delta_{\gamma 4}\delta_{\gamma' 3}\delta_{n2}
\end{vmatrix} \tag{5}$$

where

$$\alpha_1=2J(0)\sigma+B(0)d_1+F(0)d_2\ ,\quad \alpha_2=2J(0)\sigma+B(0)\sigma+F(0)d_3\ ,$$

$$\alpha_3=2J(0)\sigma+B(0)d_4+F(0)d_5\ ,\quad a_1=3/2[2J(\bar{k})-3B(\bar{k})+\frac{63}{8}F(\bar{k})]\ ,$$

$$a_2=\sqrt{3}\,[2J(\bar{k})+B(\bar{k})+\frac{103}{8}F(\bar{k})]\ ,\quad a_3=3/2[2J(\bar{k})+5B(\bar{k})+\frac{191}{8}F(\bar{k})]\ ,$$

$$a_4=2[2J(\bar{k})+B(\bar{k})+\frac{278}{16}F(\bar{k})]\ ,\quad d_1=1/2(15D_{12}+4D_{23}-9D_{34})\ ,$$

$$d_2=1/16(573D_{12}+412D_{23}+189D_{34})\ ,\quad d_3=1/16(309D_{12}+556D_{23}+309D_{34})\ ,\quad \sigma=\langle S^z\rangle$$

Eq. (5) defines an energy spectrum of spin excitations with Hamiltonian (2). A dispersion of these frequencies is conditioned by all exchange parameters $J(i-j)$, $B(i-j)$, $F(i-j)$.

For $T=0$ only one frequency shows a dispersion

$$\omega_1(\bar{k})=\{[a_3(0)+2V]^2-a_3^2(\bar{k})\}^{1/2} \tag{6}$$

while the remaining frequencies are $\bar{k}$ - independent.

The frequency gap (6) is equal to

$$\omega_1(0)=\{4V[a_3(0)+V]\}^{1/2} \tag{7}$$

In Fig.1 we show dependence of energy gaps $(h/2\pi=1)$ on parameters B, F, V of branches type $|\Delta s^z|=1$ for $T=0$.

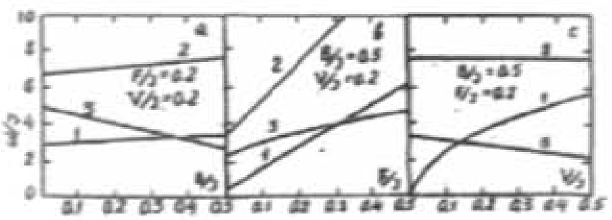

Fig.1. The dependence of energy gap on parameters: B - (a), F - (b), V - (c) of excitation $|\Delta s^z|=1$ for $T=0$ (for s.c.c.).

In the case considered in the present paper (i.e. $S=3/2$) we have three order parameters (dipolar σ, quadrupolar z, octupolar τ), connected with exchange interactions of $(\bar{S}_i\bar{S}_j)$, $(\bar{S}_i\bar{S}_j)^2$ and $(\bar{S}_i\bar{S}_j)^3$ types, respectively [3,7], for which we obtain the following expressions

$$\sigma=\langle S^z\rangle=1/2(3D_{14}+D_{23})=\frac{1}{2T_1}[3(1+\Phi_3)(1+\Phi_1+\Phi_2)+\Phi_1(1+3\Phi_2+\Phi_3)]\ , \tag{8}$$

$$z=3\langle(S^z)^2\rangle-S(S+1)=3(D_{12}-D_{34})=\frac{3}{T_1}[(1+\Phi_2)(1+\Phi_3)-\Phi_1\Phi_2]\ , \tag{9}$$

$$\tau=5\langle(S^z)^3\rangle-(3S^2+3S-1)\langle(S^z)\rangle=3/2(D_{14}-3D_{23})=\frac{3}{2T_1}[1-2\Phi_1(1+\Phi_3)+\Phi_2(1+\Phi_1)+\Phi_3(1+\Phi_2)]\ , \tag{10}$$

where

$$\Phi_1=\frac{1}{N}\sum_{\bar{k}}\sum_{n=1}^{6}A_n f(\omega_n),\ \Phi_2=\frac{1}{N}\sum_{\bar{k}}\sum_{n=1}^{6}B_n f(\omega_n),$$

$$\Phi_3=\frac{1}{N}\sum_{\bar{k}}\sum_{n=1}^{6}C_n f(\omega_n)$$

A_n, B_n, C_n are expansion coefficients of the Green functions (5) on the $(\omega-\omega_n)^{-1}$ and $f(\omega_n)=\left[\exp\left(\frac{\omega_n}{k_B T}\right)-1\right]^{-1}$ is Bose-Einstein distribution function, $T_1=(1+2\Phi_1)\times(1+\Phi_2)(1+\Phi_3)+\Phi_1\Phi_2(1+2\Phi_3)$ and the parameters D_α are expressed through Φ_1, Φ_2 and Φ_3 as follows

$$D_1=\frac{1}{T_1}(1+\Phi_1)(1+\Phi_2)(1+\Phi_3)\ , \tag{11}$$

$$D_2=\frac{1}{T_1}\Phi_1(1+\Phi_2)(1+\Phi_3)\ , \tag{12}$$

$$D_3=\frac{1}{T_1}\Phi_1\Phi_2(1+\Phi_3)\ , \tag{13}$$

$$D_4=\frac{1}{T_1}\Phi_1\Phi_2\Phi_3\ , \tag{14}$$

In the MFA

$$D_{\alpha+1}/D_\alpha=e^{-\frac{E_{\alpha+1}-E_\alpha}{\theta}} \tag{15}$$

or

$$D_2/D_1=e^{-\frac{a_{31}+2V}{k_B T}}=\psi_1 \tag{16}$$

$$D_3/D_2=e^{-\frac{a_{32}}{k_B T}}=\psi_2 \tag{17}$$

$$D_4/D_3=e^{-\frac{a_{33}-2V}{k_B T}}=\psi_3 \tag{18}$$

3. EXCITATION SPECTRUM OF THE TYPE $|\Delta s^z|=2$.

In this case a set of equations for Green functions has a form

$$\begin{vmatrix} \omega-\beta_1 & 0 & b_1 D_{13} & b_2 D_{13} \\ 0 & \omega-\beta_2 & b_2 D_{24} & b_1 D_{24} \\ -b_1 D_{24} & -b_2 D_{24} & \omega+\beta_2 & 0 \\ -b_2 D_{13} & -b_1 D_{13} & 0 & \omega+\beta_1 \end{vmatrix} \begin{vmatrix} \langle\langle L_{13}^1 | L_{\gamma\gamma'}^n \rangle\rangle_{\bar{k},\omega} \\ \langle\langle L_{24}^1 | L_{\gamma\gamma'}^n \rangle\rangle_{\bar{k},\omega} \\ \langle\langle L_{13}^2 | L_{\gamma\gamma'}^n \rangle\rangle_{\bar{k},\omega} \\ \langle\langle L_{24}^2 | L_{\gamma\gamma'}^n \rangle\rangle_{\bar{k},\omega} \end{vmatrix} = \frac{1}{2\pi} \begin{vmatrix} D_{13}^n \delta_{n1}\delta_{\gamma 3}\delta_{\gamma' 1} \\ D_{24}^n \delta_{n1}\delta_{\gamma 4}\delta_{\gamma' 2} \\ D_{13}^n \delta_{n2}\delta_{\gamma 3}\delta_{\gamma' 1} \\ D_{24}^n \delta_{n2}\delta_{\gamma 4}\delta_{\gamma' 2} \end{vmatrix} \tag{19}$$

where

$$\beta_1 = 4J(0)\sigma + B(0)(9D_{12}+4D_{23}-3D_{34}) + \frac{F(0)}{8}(441D_{12}+484D_{23}-249D_{34}) + 2V,$$

$$\beta_2 = 4J(0)\sigma - B(0)(3D_{12}-4D_{23}-9D_{34}) + \frac{F(0)}{8}(249D_{12}+484D_{23}+441D_{34}) - 2V,$$

$$b_1(\bar{k}) = 3/2[4B(\bar{k})+5F(\bar{k})], \quad b_2(\bar{k}) = 3/2[4B(\bar{k})+11F(\bar{k})]$$

For $T=0$ we have

$$\omega_1^2(\bar{k}) = \beta_1^2(0) - b_2^2(\bar{k}) \tag{20}$$

$$\omega_2^2(0) = \beta_2^2(0) \tag{21}$$

The dispersion of $\omega_1(\bar{k})$ is connected with the parameters $B(i-j)$ and $F(i-j)$.

It should be mentioned that with the large values of an uniaxial anisotropy (comparing to exchange integral) there is the inversion of energy levels and $|\Delta s^z| \geq 2$ type excitations becomes more preferable. Therefore, the experimental discovery of this type excitations is more probable in magnets with strong anisotropy. Indeed, the same type of excitation ($|\Delta s^z|=2$) with frequency $\nu=119$ cm^{-1} had been observed in the antiferromagnet $NiWO_4$ [8], which is characterised by the strong uniaxial anisotropy. Spin excitations of the type $|\Delta s^z| \geq 2$ energy gap is determined mainly by the parameters of exchange interactions and hence the frequencies of this excitations in the strong magnets lie in the optical region (~10^2 cm^{-1}).

Dependence of gaps of type $|\Delta s^z|=2$ on B, F, V for $T=0$ are ploted in Fig.2.

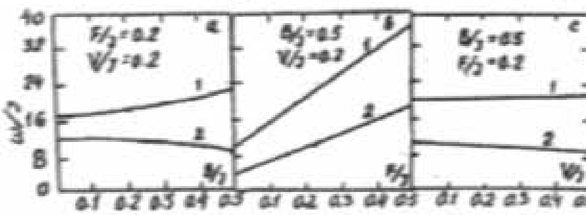

Fig.2. The dependence of energy gap on parameters: B - (a), F - (b), V - (c) of excitation $|\Delta s^z|=2$ for $T=0$ (for s.c.c.).

4. EXCITATION SPECTRUM OF THE TYPE $|\Delta s^z|=3$.

The frequencies of such excitations (which are obtained here for the antiferromagnets for the first time) derivate from equation

$$\begin{vmatrix} \omega-q & -t \\ t & \omega+q \end{vmatrix} \begin{vmatrix} \langle\langle L_{14}^1 | L_{\gamma\gamma'}^n \rangle\rangle_{\bar{k},\omega} \\ \langle\langle L_{14}^2 | L_{\gamma\gamma'}^n \rangle\rangle_{\bar{k},\omega} \end{vmatrix} = \frac{1}{2\pi} \begin{vmatrix} D_{14}^n \delta_{n1}\delta_{\gamma 4}\delta_{\gamma' 1} \\ D_{14}^n \delta_{n2}\delta_{\gamma 4}\delta_{\gamma' 1} \end{vmatrix} \tag{22}$$

where $q=6J(0)\sigma+3B(0)\sigma+F(0)/16(1071D_{14}+309D_{23})$, $t=9F(\bar{k})D_{14}$

From (22) we have

$$\omega^2(\bar{k}) = q^2 - t^2(\bar{k}) \tag{23}$$

As it is seen, the dispersion of this branch arises only via $F(i-j)$. Dependence of gaps of type $|\Delta s^z|=3$ on B, F is ploted in Fig.3 for $T=0$.

By comparison of excitations of the types $|\Delta s^z|=1,2,3$ we obtain that, as in the case of ferromagnet [3], a dispersive branch of the type $|\Delta s^z|=m$ $(m=1,2,\ldots,2S)$ is related with the presence of exchange interactions $(S_i S_j)^m, (S_i S_j)^{m+1}, \ldots, (S_i S_j)^{2S}$.

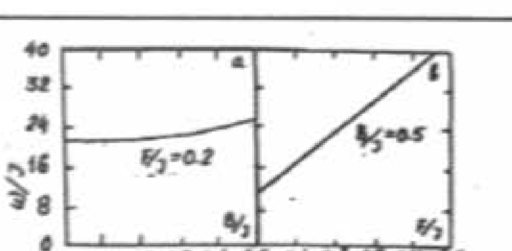

Fig.3. The dependence of energy gap on parameters: B - (a), F - (b), V - (c) of excitation $|\Delta s^z|=3$ for $T=0$ (for s.c.c.).

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M.B. Hüseynov, V.S. Tağıyev, A.M. Suleymanov

BİROXLU QEYRİ-HEYZENBERQ ANTİFERROMAQNİT HƏYƏCANLANMA SPEKTRİ

Qrin funksiyası texnikasında standart-bazis operatorlar metodu vasitəsilə qeyri-Heyzenberq antiferromaqnitlərin (bu zaman bixətti mübadilə $J\left(\vec{S}_i\vec{S}_j\right)$ ilə bərabər bikvadratik $B\left(\vec{S}_i\vec{S}_j\right)^2$ və oktopolyar $F\left(\vec{S}_i\vec{S}_j\right)^3$ mübadilələr də nəzərə alınır) enerji spektri tədqiq edilmişdir. $|\Delta S^z| = 1, 2, 3$ tipli spin həyəcanlanmalarının spektrində dispersiyalı əlavə budaqlar alınmışdır. Dispersiyanın mübadilənin tipindən asılılığı müəyyən edilmişdir.

М.Б.Гусейнов, В.С. Тагиев, А.М. Сулейманов

СПЕКТР ВОЗБУЖДЕНИЯ ОДНООСНОГО НЕГЕЙЗЕНБЕРГОВСКОГО АНТИФЕРРОМАГНЕТИКА

Методом стандартных - базисных операторов в технике функции Грина исследован энергетический спектр негейзенберговского антиферромагнетика (в этом случае, наряду с билинейным обменом $J\left(\vec{S}_i\vec{S}_j\right)$, учитываются также биквадратные $B\left(\vec{S}_i\vec{S}_j\right)^2$ и октуполярные $F\left(\vec{S}_i\vec{S}_j\right)^3$ обмены). Получены дополнительные ветви с дисперсией для спиновых возбуждений типа $|\Delta S^z| = 1, 2, 3$. Установлена зависимость дисперсии от типа обмена.

Редактор: Р.Р. Гусейнов