Spin excitations and electron paramagnetic resonance in two-layer antiferromagnetic system $\text{Y}_{1-x}\text{Yb}_x\text{Ba}_2\text{Cu}_3\text{O}_{6+y}$

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Abstract. An experimental and theoretical study of the EPR spectrum of $\text{Y}_{0.98}\text{Yb}_{0.02}\text{Ba}_2\text{Cu}_3\text{O}_6$ is reported. A strong anisotropy of the EPR linewidth is revealed for the parallel and perpendicular orientations of the external magnetic field relative to the tetragonal symmetry axis. This anisotropy is related to the indirect spin-spin interaction between the ytterbium ions via antiferromagnetic spin-waves. The explicit expressions for the coupling of ytterbium ions with the spin waves and the corresponding indirect spin-spin interactions are derived. An estimate of the exchange coupling between the ytterbium and copper ions is found from the comparison of the theory with the experiment.

1. Introduction

Antiferromagnetic properties and spin fluctuations of quasi-two-dimensional copper oxides remain a subject of intensive investigations due to their transformation into high-temperature superconductors by doping of CuO$_2$ planes with electronic holes. Soon after the discovery of high-$T_c$ superconductivity in La$_{2-x}$Ba$_x$CuO$_4$ [1] the attention was attracted to the YBCO family. Detailed inelastic neutron scattering experiments in the bilayer compound $\text{YBa}_2\text{Cu}_3\text{O}_6$ reveal a very large superexchange integral between nearest-neighbor Cu sites within the CuO$_2$ layers ($J_1 \simeq 2000$ K [2], $J_2 \simeq 1400$ K [3]), a rather strong coupling between the nearest layers $J_1$ ($\delta = J_1 / J \simeq 4 \cdot 10^{-2}$ [2], $\delta \simeq 7 \cdot 10^{-3}$ [3]), and an extremely small coupling to the next-nearest-layer with $J_2 / J \simeq 2 \cdot 10^{-5} - 3 \cdot 10^{-4}$ [2,3]. These results were obtained on the basis of the standard spin-wave theory, which for the bilayer system predicts two acoustical and two optical modes of spin waves. The observed energy gap for the modes involving spin deviations out of the CuO$_2$ planes was modeled by taking into account a week planar anisotropy $\alpha_y = \Delta J / J \simeq 2 \cdot 10^{-4} - 7 \cdot 10^{-4}$[2,3]. The Neel temperature $T_N = 415$ K at $y < 0.15$ could be explained by taking into account the quantum effect and a small anisotropy of the exchange integral taking the value the Cu-Cu coupling $J = 1700$ K [4]. Recently the analytic expression for $T_N$ was obtained by the Green’s function method in the Tyablikov approximation as a function of both exchange integrals between the CuO$_2$ planes for $J_1, J_2 = J$ and $\alpha_y = 0$ [5]. NMR and EPR were
found to be very powerful techniques for probing the local magnetic susceptibility and studying the spin kinetics of high-Tc oxides [6]. The EPR method is usually based on the EPR signal from the magnetic probes using ions having local d- or f-electrons. In particular, the use of Mn$^{2+}$ as an EPR probe in the CuO$_2$ plane allowed revealing a very fast spin relaxation rate of the Cu-ions due to their spin-phonon interaction, which prevents the observation of the EPR signal directly from the Cu-ions [7]. A detailed EPR study of the Yb$^{3+}$ spin relaxation as a function of temperature in YBa$_2$Cu$_3$O$_{6+y}$ ($y = 0.1, 0.4, 0.5, 0.6, 0.98$) shows that at high temperatures the main contribution to the relaxation rate is given by the Raman two-phonon process due to the modulation of the crystal electric field by optical modes of the oxygen ions in the CuO$_2$ planes [8]. In the superconducting samples ($y > 0.4$) an electronic contribution of the Korringa-type relaxation was found. However, the nature of the temperature independent relaxation rate remains unclear.

In this work we report our theoretical and experimental study of spin excitations and relaxation due to the interaction of Yb$^{3+}$ ions with antiferromagnetic (AF) spin waves in YBa$_2$Cu$_3$O$_{6+y}$. At low oxygen doping ($y < 0.15$) the CuO$_2$ planes are not doped with electronic holes. We find that indirect spin-spin interactions of Yb$^{3+}$ ions via AF magnons (Suhl-Nakamura interaction) are very long-range and give an important contribution to the EPR linewidth and position of the resonance line. From the comparison of our calculations with the experiment we estimate the exchange coupling between the Yb$^{3+}$ and Cu$^{2+}$ ions to be $|A| \approx 120$ K.

The paper is organized as follows. In Sec. 2 we find the energy gaps of AF spin waves in YBa$_2$Cu$_3$O$_{6+y}$ due to an external magnetic field and dipole-dipole interactions. The derivation of the Yb-Cu interaction and an indirect interaction between Yb$^{3+}$ ions via magnons is given in Sec. 3. The experimental results on the EPR spectra and a comparison with experiment follow in Sec. 4.

2. The energy gaps in the spin wave dispersion due to the external magnetic field and dipole-dipole interactions

2.1. The external magnetic field

In the YBa$_2$Cu$_3$O$_{6+y}$ the CuO$_2$ plains are grouped in bilayers with a small exchange interaction between different bilayers. In this study we neglect this coupling. The exchange spin Hamiltonian can be written in the form:

$$\frac{H_{ex}}{J} = \frac{1}{2} \sum_{nm} \left[ S^a_n S^b_m + S^b_n S^a_m - \alpha_{xy} \left( S^a_n S^b_m + S^b_n S^a_m \right) \right] + \delta \sum_n S^a_n S^b_n + \delta \sum_m S^b_m S^a_m. \quad (1)$$

Here $a$ and $b$ indicate two sub-lattices in every plane. In the layer 1 the sites $n$ and $m$ are related to the $a$ and $b$ sub-lattices correspondingly, while vise versa in the layer 2. Hereafter we take into account the nearest neighbors only. $J$ is the exchange constant within the plane, $\delta$ and $\alpha_{xy}$ correspond to the exchange coupling between the two planes and an anisotropic exchange term both measured in units $J$, respectively. We choose the direction of the spontaneous magnetizations along the $x$-axis, since due to the axial symmetry it can be chosen arbitrary.

We shall now take into account the external magnetic field applied to the system within the plane and directed along the $y$ axis:

$$H_{ZeCu} = B_{Cu} \sum_n \left( S^y_n + S^y_n \right) + B_{Cu} \sum_m \left( S^y_m + S^y_m \right), \quad B_{Cu} = g_{Cu} \mu_B H_0. \quad (2)$$

Here $g_{Cu}$ is the $g$-factor for the Cu ion, $\mu_B$ is the Bohr magneton, and $H_0$ – the external magnetic field.
We assume the field rotates the magnetizations of both sub-lattices slightly toward the $y$ direction by the angle $\phi$. It is convenient then to turn the axes so that the new $x$-axis is directed along the corresponding magnetizations of the sub-lattices:

$$S_x^a = S_x^y \cos \phi - S_y^y \sin \phi,$$

$$S_y^a = -S_y^x \sin \phi + S_y^y \cos \phi,$$

$$S_x^b = S_x^y \cos \phi + S_y^y \sin \phi,$$

$$S_y^b = -S_y^x \sin \phi + S_y^y \cos \phi.$$  \hspace{1cm} (3)

Using the standard Holstein-Primakoff formalism we make the following transformation for the layer:

$$S_{n1}^a = \frac{1}{2}a_n^+ a_n + \frac{1}{2}S_n^a,$$

$$S_{m1}^a = \frac{1}{2}b_m^+ b_m + \frac{1}{2}S_m^a,$$

$$S_{n1}^b = \frac{1}{2}a_n^+ a_n + \frac{1}{2}S_n^b,$$

$$S_{m1}^b = \frac{1}{2}b_m^+ b_m + \frac{1}{2}S_m^b,$$  \hspace{1cm} (4)

with $S^+ = -\mathcal{S}^{\dagger}iS^x$. For the layer $j = 2$ the transformation can be obtained by interchanging indices $n$ and $m$. Here $a^+$, $a$, $b^+$, $b$ are the boson creation and annihilation operators for the two sub-lattices.

Then we perform the Fourier transformation to the reciprocal lattice:

$$a_q = N^{-1/2} \sum_n e^{iq_n} a_n,$$

$$b_q = N^{-1/2} \sum_n e^{iq_n} b_n,$$  \hspace{1cm} (5)

where $N$ is the number of unit cells in the bilayer. From the energy minima condition for the spin system ground state in the external magnetic field we obtain the value of the rotation angle

$$\sin \phi = -\frac{B_{Cu}}{J(4 + \delta)} = -b_{Cu}.$$  \hspace{1cm} (6)

As a result of these steps the total spin Hamiltonian $H_{tot} = H_{ex} + H_{Ze}$ takes the form:

$$H_{tot} = \left(1 + \frac{\delta}{4}\right) \sum_q \left( a_{q1}^+ a_{q2} + a_{q1}^+ a_{q2}^+ a_{q1}^+ a_{q2}^+ + b_{q1}^+ b_{q2} + b_{q1}^+ b_{q2}^+ \right) +$$

$$+ \left(1 - b_{Cu}^2 - \frac{\alpha_{xy}}{2}\right) \sum_q \gamma_q \left( a_{q1}^+ b_{q2} + a_{q1}^+ b_{q2}^+ a_{q1}^+ b_{q2} + a_{q2}^+ b_{q2}^+ \right) +$$

$$+ \left(b_{Cu}^2 - \frac{\alpha_{xy}}{2}\right) \sum_q \gamma_q \left( a_{q1}^+ b_{q2}^+ + a_{q1}^+ b_{q2} + a_{q2}^+ b_{q2} + a_{q2}^+ b_{q2}^+ \right) +$$

$$+ \left(1 - b_{Cu}^2\right) \frac{\delta}{4} \sum_q \left( a_{q1}^+ b_{q2}^+ a_{q1}^+ b_{q2} + a_{q2}^+ b_{q2} + a_{q2}^+ b_{q2} + a_{q2}^+ b_{q2} \right) +$$

$$+ \frac{\delta}{4} b_{Cu}^2 \sum_q \left( a_{q1}^+ b_{q2} + a_{q1}^+ b_{q2}^+ + a_{q2}^+ b_{q2} + a_{q2}^+ b_{q2} \right).$$  \hspace{1cm} (7)

Here

$$\gamma_q = 1/2 \left[ \cos(q, a) + \cos(q, a) \right]$$  \hspace{1cm} (8)
To diagonalize this Hamiltonian we perform the Bogoliubov transformation to new creation and annihilation boson operators $\alpha_q^\ast$, $\beta_q^\ast$, $\eta_q$, $\kappa_q$, $\alpha_q$, $\beta_q$, $\eta_q$, $\kappa_q$, which must satisfy the commutation relations of the following type:

$$[\alpha_q^\ast, H_{\text{tot}}] = E_{\text{eq}} \alpha_q$$

(9)

Using these equations we obtain the energy eigenvalues and explicit expressions for the eigenoperators for two acoustical and two optical modes of spin waves.

For the acoustical modes we have:

$$\left(\frac{E_{\text{eq}}}{2J}\right)^2 = \left[1 - \gamma_q + 2b_{Cu}^2 \left(\gamma_q + \frac{\delta}{4}\right)\right] \left[1 + \gamma_q + \frac{\delta}{2} - \alpha_q \gamma_q\right],$$

(10)

$$\left(\frac{E_{\beta q}}{2J}\right)^2 = \left[1 + \gamma_q + \alpha_q \gamma_q\right] \left[1 + \gamma_q + \frac{\delta}{2} - 2b_{Cu}^2 \left(\gamma_q + \frac{\delta}{4}\right)\right].$$

One can see that at the Brillouin zone center $q = (0, 0)$ the gapless mode $E_{\text{eq}}$ has now a gap at the value close to the Zeeman energy $E_{\text{eq}} = 4b_{Cu}^2 = g_{Cu} \mu_B H_0$, while the other mode $E_{\beta q}$ maintains almost the same gap due to anisotropy of the exchange interaction. The eigenoperators of these modes are:

$$\alpha_q = \frac{1}{2} \left\{ u_{\alpha q} (a_{i q} + a_{2 q} + b_{i q} + b_{2 q}) - v_{\alpha q} (a_{i q}^\ast + a_{2 q}^\ast + b_{i q}^\ast + b_{2 q}^\ast) \right\},$$

$$\beta_q = \frac{1}{2} \left\{ u_{\beta q} (a_{i q} + a_{2 q} - b_{i q} - b_{2 q}) - v_{\beta q} (a_{i q}^\ast + a_{2 q}^\ast - b_{i q}^\ast - b_{2 q}^\ast) \right\},$$

(11)

The coefficients of the transformation in (11) are given by:

$$u_{\alpha,\beta}^2 = \frac{J}{E_{\alpha,\beta}} \left[ 1 + \frac{\delta}{4} m \alpha_q \gamma_q \pm b_{Cu}^2 \left(\gamma_q + \frac{\delta}{4}\right) + \frac{E_{\alpha,\beta}}{2J} \right],$$

$$v_{\alpha,\beta}^2 = \frac{J}{E_{\alpha,\beta}} \left[ 1 + \frac{\delta}{4} m \alpha_q \gamma_q \pm b_{Cu}^2 \left(\gamma_q + \frac{\delta}{4}\right) + \frac{E_{\alpha,\beta}}{2J} \right].$$

(12)

For the optical modes the eigenvalues of energy are:

$$\left(\frac{E_{\eta q}}{2J}\right)^2 = \left[1 - \gamma_q - \alpha_q \gamma_q\right] \left[1 - \gamma_q + \frac{\delta}{2} + 2b_{Cu}^2 \left(\gamma_q - \frac{\delta}{4}\right)\right],$$

$$\left(\frac{E_{\kappa q}}{2J}\right)^2 = \left[1 + \gamma_q - 2b_{Cu}^2 \left(\gamma_q - \frac{\delta}{4}\right)\right] \left[1 - \gamma_q + \frac{\delta}{2} + \alpha_q \gamma_q\right].$$

(13)

At the zone boundary for $q = (\pi/a, \pi/a)$ the parameter $\gamma_{\pi,\pi} = -1$, and the mode $E_{\eta q}$ now has a gap of about the Zeeman energy, while the other mode has about the same gap as it had before. The eigenoperators for the optical modes are the following:

$$\eta_q = \frac{1}{2} \left\{ u_{\eta q} (a_{i q} - a_{2 q} + b_{i q} - b_{2 q}) - v_{\eta q} (a_{i q}^\ast - a_{2 q}^\ast + b_{i q}^\ast - b_{2 q}^\ast) \right\},$$

$$\kappa_q = \frac{1}{2} \left\{ u_{\kappa q} (a_{i q} - a_{2 q} - b_{i q} + b_{2 q}) - v_{\kappa q} (a_{i q}^\ast - a_{2 q}^\ast - b_{i q}^\ast + b_{2 q}^\ast) \right\}.$$

(14)

The coefficients of the transformation in (13) are given by
The obtained expressions are valid for the external magnetic field below the critical value, at which the two sub-lattices collapse into one with magnetic moments directed along the magnetic field.

2.2. Dipole-dipole interactions

The internal field due to magnetic dipole-dipole interactions between the Cu ions may be more important than the external magnetic field below the critical value. The spin Hamiltonian of the dipole-dipole interactions has the form:

\[
H_{dd} = g^2 \mu_B^2 \sum_q \left( S_q S_r \right) \frac{R_q^2}{R_{ij}^5} - 3 \left( R_{ij} S_q \right) \left( R_{ij} S_r \right) \tag{16}
\]

We take into account the second term only, since the first term is isotropic and much smaller than the isotropic exchange interaction. We have found that the nearest neighbors (nn) do not contribute to the energy gap, so we have to take into account the next nearest neighbors (nnn) also. To simplify the expressions we set the external magnetic field to \( H = 0 \).

The following calculations are similar to the ones performed above: we make the Holstein-Primakoff and the Fourier transformations for \( H_{tot} = H_{ex} + H_{dd} \). The minima condition for the ground state is realized for the magnetization directed along the diagonal of the square lattice of the layers. The result of these calculations for the total Hamiltonian \( H_{tot} = H_{ex} + H_{dd} \) is the following:

\[
\frac{H_{tot}}{2J} = \left( 1 + \frac{\delta}{4} - \frac{d_1}{2} + 2d_3 + \frac{d_4}{2} \right) \sum_q \left( a_{q_1}^* a_{q_1} + a_{q_2}^* a_{q_2} + b_{q_1}^* b_{q_1} + b_{q_2}^* b_{q_2} \right) + \right.
\]

\[\left. + \sum_q \left( \gamma_q - \frac{\alpha_{xy} \gamma_q}{2} - \frac{d_1}{4} \gamma_{q_1} \right) \left( a_{q_1}^* b_{q_1} + a_{q_2}^* b_{q_2} + a_{q_1}^* b_{q_2} + a_{q_2}^* b_{q_1} \right) + \right.\]

\[\left. + \sum_q \left( -\frac{\alpha_{xy} \gamma_q}{2} + \frac{d_1}{4} \gamma_{q_1} \right) \left( a_{q_1}^* b_{q_1} + a_{q_2}^* b_{q_2} + a_{q_1}^* b_{q_2} + a_{q_2}^* b_{q_1} \right) + \right.\]

\[\left. + \left( \frac{\delta}{4} - \frac{d_1}{8} - \frac{d_4}{8} \right) \sum_q \left( a_{q_1}^* b_{q_2}^* - a_{q_2}^* b_{q_1}^* + a_{q_1}^* b_{q_2} + a_{q_2}^* b_{q_1} \right) + \right.\]

\[\left. - \frac{d_2}{8} \sum_{q_1} \left( a_{q_1}^* b_{q_1}^* + a_{q_1}^* b_{q_1} + a_{q_1}^* b_{q_1} + a_{q_1}^* b_{q_1} \right) + \right.\]

\[\left. + \frac{d_2}{8} \sum_{q_1} \gamma_{q_1} \left( a_{q_1}^* a_{q_1} + a_{q_1}^* a_{q_1} + b_{q_1}^* b_{q_1} + b_{q_1}^* b_{q_1} \right) - \right.\]

\[\left. - \frac{d_4}{8} \sum_{q_1} \gamma_{q_1} \left( a_{q_1}^* a_{q_1} + a_{q_1}^* a_{q_1} + b_{q_1}^* b_{q_1} + b_{q_1}^* b_{q_1} \right) \right),
\]

Here \( d_1 = 3g^2 \mu_B^2 / Ja^3 \), \( d_2 = 3g^2 \mu_B^2 / Jc^3 \), \( d_3 = 3g^2 \mu_B^2 / 4\sqrt{2}Ja^3 \), \( d_4 = 3g^2 \mu_B^2 / JR^5 \) with \( R = \sqrt{a^2 + c^2} \), \( \gamma_{q_1} = \cos(q, a) \); \( c \) is the distance between the layers. Performing then the Bogoliubov-transformation we obtain the eigenvalues for the spin wave energy.
The acoustical modes:

\[
\left( \frac{E_{\alpha\alpha}}{2J} \right)^2 = \left( 1 - \gamma_q + \Delta_\alpha \right) \left( 1 + \gamma_q + \frac{\delta}{2} - \alpha_{\nu\gamma} \gamma_q \right),
\]

\[
\left( \frac{E_{\beta\beta}}{2J} \right)^2 = \left( 1 - \gamma_q + \alpha_{\nu\gamma} \gamma_q + \Delta_\beta \right) \left( 1 + \gamma_q + \frac{\delta}{2} \right);
\]

\[
\Delta_\alpha = 2d_3, \quad \Delta_\beta = -\frac{d_1}{2} + \frac{d_2}{4} + 2d_3 + \frac{d_4}{2}.
\]

Here were neglected the dipole-dipole terms, which do not contribute to the energy gap at the zone-center \( q = 0 \). Since all the dipole-dipole terms \( d_1, d_2, d_3, d_4 <\delta \) (exchange coupling between the layers), we have omitted them in the corresponding sum. One can see that the first mode has now the energy gap \( E_{\alpha\alpha} = 4J\sqrt{d_3} >> g_{Cu}\mu_B H_0 \).

The optical modes:

\[
\left( \frac{E_{\eta\eta}}{2J} \right)^2 = \left( 1 + \gamma_q - \alpha_{\nu\gamma} \gamma_q + \Delta_\eta \right) \left( 1 - \gamma_q + \frac{\delta}{2} \right),
\]

\[
\left( \frac{E_{\alpha\alpha}}{2J} \right)^2 = \left( 1 + \gamma_q + \Delta_x \right) \left( 1 - \gamma_q + \frac{\delta}{2} + \alpha_{\nu\gamma} \gamma_q \right);
\]

\[
\Delta_\eta = -\frac{d_1}{2} + \frac{d_2}{4} + 2d_3 + \frac{d_4}{2}, \quad \Delta_x = 2d_3.
\]

Here were neglected the dipole-dipole terms, which do not contribute to the energy gap at the zone-boundary \( q = (\pi/a, \pi/a) \). The dipole-dipole terms in the sum with \( \delta / 2 \) were also omitted. It is evident that both energy gaps at the zone boundary are also much larger than the Zeeman energy.

3. Interactions of the Yb\(^{3+}\)-ions with the AF spin-waves and the Suhl-Nakamura interaction

3.1. The Hamiltonian of Yb-spin-wave interactions

An exchange interaction between the particular \( j \)-th Yb\(^{3+}\)-ion of the crystal lattice and the nearest eight Cu-ions laying in the two parallel CuO\(_2\) planes can be written in the form:

\[
H_{YbCu} = A Y_j \left\{ \sum_n \left( S_{n1}^a S_{n2}^a \right) + \sum_m \left( S_{m1}^b S_{m2}^b \right) \right\}.
\]

Here \( A \) is the coupling constant, \( Y_j \) is the spin of the \( j \)-th Yb\(^{3+}\)-ion. For the Cu-spins again the axes-rotation (3), the Holstein-Primakoff transformation (4), and the Fourier transformation (both for the magnon and ytterbium operators) are performed. Taking into account the Zeeman interaction we obtain after the Bogoliubov transformation (9) the following result for the Hamiltonian

\[
H_{YbCu} = H_{YbCu}^{(0)} + H_{YbCu}^{(1)} + H_{YbCu}^{(2)}
\]

for the magnetic field oriented along the \( y \)-axis:
Here $Y_{q}^{x,y,z}$ is the Fourier-transform of the site operators similar to (5); $F_{q}^{ac}$ and $F_{q}^{op}$ are form factors for the acoustical and the optical modes:

$$F_{q}^{ac} = \frac{q_{a}q_{a}}{2} \cos \frac{q_{a}q_{a}}{2}, \quad F_{q}^{op} = \frac{q_{a}q_{a}}{2} \sin \frac{q_{a}q_{a}}{2}. \quad (22)$$

In the case of the magnetic field oriented along the $z$-axis we obtain:

$$H^{(0)}_{YbCu} = 4A_{Cu}Y_{0}^{y}N^{1/2} - A_{Cu}Y_{0}^{y}N^{-1/2} \sum_{q} \left\{ \nu_{q}^{2} + \nu_{q}^{2} + \nu_{q}^{2} + \nu_{q}^{2} \right\},$$

$$H^{(1)}_{YbCu} = 2A \sum_{q} \left\{ F_{q}^{ac} \left[ Y_{q}^{z} \left( u_{q} + v_{q} \right) \right] \left( \alpha_{q} + \alpha_{q}^{*} \right) + ib_{Cu}Y_{q}^{x} \left( u_{q} + v_{q} \right) \left( \alpha_{q} - \alpha_{q}^{*} \right) \right\} +$$

$$+ F_{q}^{op} \left[ Y_{q}^{z} \left( u_{q} + v_{q} \right) \left( \kappa_{q} - \kappa_{q}^{*} \right) + ib_{Cu}Y_{q}^{x} \left( u_{q} + v_{q} \right) \left( \kappa_{q} - \kappa_{q}^{*} \right) \right] +$$

$$+ iY_{q}^{z} \left( 1 - b_{Cu}^{2} \right) \left\{ F_{q}^{ac} \left( u_{q} + v_{q} \right) \left( \beta_{q} + \beta_{q}^{*} \right) + F_{q}^{op} \left( u_{q} + v_{q} \right) \left( \kappa_{q} + \kappa_{q}^{*} \right) \right\}. \quad (21)$$

The term $H^{(2)}_{YbCu}$ quadratic in the boson operators is responsible for two-magnon processes, which are not considered here. The total Hamiltonian now takes the form:

$$H = H_{Z_{2b}} + \sum_{q} \left( E_{q}^{y}r_{q}^{y}r_{q}^{y} + E_{q}^{y}r_{q}^{y}r_{q}^{y} + E_{q}^{y}r_{q}^{y}r_{q}^{y} + E_{q}^{y}r_{q}^{y}r_{q}^{y} + \frac{1}{2} \left[ S, H_{0} + H_{YbCu} \right] + H_{YbCu} \right), \quad (23)$$

Here $H_{Z_{2b}}$ is the Zeeman energy of the ytterbium ions which has to be renormalized by the $H_{YbCu}$ interaction.

3.2. The Suhl-Nakamura interaction

An exchange between the ytterbium ions by the AF magnons creates an indirect spin-spin coupling known as the Suhl-Nakamura (SN) interaction. An effective spin-spin Hamiltonian can be obtained by the unitary transformation with the following elimination of the magnon operators. Using the Hamiltonian (23) we make the following transformation:

$$\hat{H} = e^{S}H_{E}e^{-S} = H_{0} + H_{YbCu} + \left[ S, H_{0} + H_{YbCu} \right] + \frac{1}{2} \left[ S, S, H_{0} + H_{YbCu} \right] + \ldots \quad (24)$$

Here $S$ is some yet unknown operator linear in the Yb-Cu coupling. To eliminate the linear terms we put

$$H_{YbCu} + \left[ S, H_{0} \right] = 0; \quad \langle m | S | n \rangle = \frac{\langle m | H_{YbCu} | n \rangle}{E_{m}^{0} - E_{n}^{0}}. \quad (25)$$

Here we have used the eigen states of the unperturbed Hamiltonian. The SN interaction can be obtained from the (21, 24, 25) in the second order coupling of Yb-Cu:
\[
H_{SN} = -2A^2 \sum_q \left( \frac{\left( u_{qq} + v_{qq} \right)^2}{E_{qq}} Y_q^z Y_{-q}^z + \frac{\left( u_{q\beta} - v_{q\beta} \right)^2}{E_{q\beta}} Y_q^1 Y_{-q}^1 \right) + \left( F_{q,q}^{op} \right)^2 \left( \frac{u_{qq} - v_{qq}}{E_{qq}} Y_q^y Y_{-q}^y + \frac{u_{q\gamma} + v_{q\gamma}}{E_{q\gamma}} Y_q^y Y_{-q}^y \right)
\]

(26)

Taking into account the explicit expressions for the Bogoliubov coefficients (12,15) we obtain the final result

\[
H_{SN} = -2A^2 \sum_q f_q \left( \frac{V_{xy}}{2} Y_q^{xx} Y_{-q}^{xx} + \frac{V_{yy}}{2} Y_q^{yy} Y_{-q}^{yy} \right), \quad f_q = \frac{\left( F_{q,q}^{ac} \right)^2}{\delta + \gamma_q} + \frac{\left( F_{q,q}^{op} \right)^2}{\delta - \gamma_q}.
\]

(27)

In the denominators the terms which are small compared to \( \delta \) were omitted. In the coordinate representation the dependence of the SN interaction on the distance between the ytterbium ions \( R \) can be roughly approximated as \( f \left( R \right) \propto J_1 \left( R \right) / R \), where \( J_1 \left( R \right) \) is the Bessel function. It means that this interaction is more long-range than the dipole-dipole one.

4. The Electron Paramagnetic Resonance of Yb\(^{3+}\)-ions in antiferromagnetic YBCO compound

4.1. Experimental results for the ytterbium EPR spectra

The EPR spectra were measured in the sample \( \text{Yb}_{0.98}\text{Ba}_{0.02}\text{Cu}_2\text{O}_6 \). We expect that at this level of oxygen doping \( y = 0.1 \) electronic holes are not yet present in the CuO\(_2\) planes. Figure 1 shows typical Yb\(^{3+}\) EPR spectra taken at \( T = 40 \text{ K} \) with the external magnetic field along and perpendicular to the crystal \( c\)-axis and with an alternating field within the \( xy\)-plane. It is well known that the ground-state multiplet \( \frac{3}{2} F \) of the Yb\(^{3+}\) ions \( 4f^{13} \) is expected to be split by the crystal electric field of tetragonal symmetry into four Kramers doublets \([9]\). Inelastic neutron-scattering measurements showed that in \( \text{YbBa}_2\text{Cu}_2\text{O}_7 \) the first-excited doublet lies 1000 K above the ground state doublet \([10]\). Our measurements reveal a very strong anisotropy of the EPR linewidth in the antiferromagnetic sample \( \text{Yb}_{0.98}\text{Ba}_{0.02}\text{Cu}_2\text{O}_6 \). The \( g \)-factors \( g_\perp = 3.54 \), \( g_p = 3.23 \) show an anisotropy similar to the one in the previous measurements for oxygen doping \( y = 0.4 \) (where the AF state is suppressed, while the symmetry remains tetragonal, and \( g_{\perp}^0 = 3.49 \), \( g_p^0 = 3.13 \)) with a shift of the resonance line to lower magnetic fields \([8]\). We suggest that these features of the Yb\(^{3+}\) EPR signal may be explained by the Yb-Cu exchange interaction and the corresponding coupling of Yb\(^{3+}\) ions with the AF spin-waves.
Figure 1. EPR spectra of Yb$^{3+}$ in grain-oriented Y$_{0.98}$Yb$_{0.02}$Ba$_2$Cu$_3$O$_{6.1}$ at $T = 40$ K. The solid line corresponds to the external magnetic field perpendicular to the crystal $c$-axis, while the dotted line corresponds to the external magnetic field along the $c$-axis.

4.2. Analysis of the EPR linewidth

One can expect that at relatively low temperatures the broadening of the EPR signal is caused by the spin-spin interactions: the usual magnetic dipole-dipole interactions and the SN interactions between the Yb$^{3+}$ ions. Their contribution can be calculated by the standard method of moments for the EPR line [9]. A contribution of the dipole-dipole interactions for the Yb$^{3+}$ ion concentration $x = 0.02$ was estimated by calculations of the second and fourth moments using the experimental $g$-factors. For the external magnetic field perpendicular to the crystal $c$-axis the result is $\Delta B^\perp_{dd} = 8$ mT, while for the parallel orientation it is slightly larger ($\Delta B^\parallel_{dd} = 8.2$ mT). The experimental values are significantly different: the calculated dipole-dipole contribution for the perpendicular orientation practically coincides with the experimental value $\Delta B^\perp_{exp} = 7.8$ mT, while for the parallel orientation the experimental linewidth is five times larger ($\Delta B^\parallel_{exp} = 41.3$ mT). Such behavior can be related to the anisotropy of the SN interaction. For the external field below the critical value the AF magnetization is almost perpendicular to external field. It means that if $\mathbf{H}_0$ is directed along $y$-axis, the alternating field is directed along the $x$-axis and a contribution to the second moment is defined by a commutator of the total spin $Y^z_0$ with $H_{SN}$. However, one can see from Eq. (27) that the operator structure of $H_{SN}$ gives $[Y^z_0, H_{SN}] = 0$, which means that for this orientation there is no contribution from the SN-interaction to the EPR linewidth.

The situation is different for the external magnetic field oriented along the $c$-axis. In this case the alternating field can be directed arbitrarily in the $xy$-plane. In a high-temperature approximation the second moment can be calculated by the formula:

$$M_2 = -\frac{\text{Sp}\left(\left[Y^z_0, H_{SN}\right]\left[Y^z_0, H_{SN}\right]\right)}{\text{Sp}\left(Y^z_0 Y^z_0\right)} = Y^z_0 = Y^x_0 \pm iY^y_0$$

The result is the following:
Similar calculations for the fourth moment give
\[ M_4 = \frac{8A^8}{J^4} \int dq_x \int dq_y f_q^2 = 0.051 \frac{A^8}{J^4} \]
(30)

For the diluted ytterbium spin-system the peak-to-peak EPR linewidth for the parallel orientation can be calculated by the formula [9]:
\[ \Delta B_{SN}^p = \frac{\pi}{\sqrt{3}} x \left( \frac{M_2^3}{M_4} \right)^{1/2} = 0.42 x \left( \frac{A}{J} \right) \]
(31)

This result allows to estimate the exchange coupling \( A \) between the ytterbium and copper ions if we suppose that the main contribution for the parallel orientation comes from the SN interaction. Taking the experimental value \( \Delta B_{exp}^p = 41.3 \) mT and extracting the dipole-dipole contribution of 8.2 mT, we relate the rest to the SN interaction. Using \( J = 1700 \) K [4] and the Yb concentration \( x = 0.02 \) we find \( |A| = 120 \) K. The sign remains unknown.

4.3. Analysis of the EPR g-factors
According to the Eqs. (21, 6) the renormalized Zeeman energy can be written in the form:
\[ \tilde{B}_{yb} = \mu_B H_0 \left[ \hat{g}_{yb}^0 - \hat{g}_{Cu}^0 \frac{A}{J} \left( 1 - \frac{1}{4} v_{tot}^2 \right) \right]; \quad v_{tot} = \frac{1}{N} \sum_q \left\{ v_{qx}^2 + v_{qy}^2 + v_{qz}^2 \right\} = 0.614 \]
(32)

We can now estimate the renormalized g-factors of the ytterbium ions due to their coupling with the AF spin waves. Following the above discussion we take the initial values of the Yb\(^{3+}\) g-factors for the experimental value of the paramagnetic state \( \hat{g}_{yb}^0 = \hat{g}_p^0 = 3.13 \) for the parallel orientation and \( \hat{g}_{yb}^0 = \hat{g}_p^0 = 3.49 \) for the perpendicular orientation [8]. The typical g-factors of the copper ion in the tetragonal field with the orbital ground state of the type \( (x^2 - y^2) \) are \( \hat{g}_Cu^p = 2.4 \) and \( \hat{g}_{Cu}^\perp = 2.1 \) for the parallel and perpendicular orientations of the external magnetic field, respectively [9]. If we adopt a negative sign of the exchange coupling found above \( A = -120 \) K and take again \( J = 1700 \) K, we obtain for the AF state the theoretical values \( \hat{g}_{yb}^p = 3.27 \) and \( \hat{g}_{yb}^\perp = 3.61 \), which are quite close to the experimental values \( \hat{g}_p = 3.23 \) and \( \hat{g}_{\perp} = 3.54 \). Some corrections to the g-factors can appear due to the SN-interaction and the terms \( \hat{H}_{ybCu}^{(2)} \) quadratic in the boson operators. Our preliminary evaluations show that they are very small, especially for our system with low concentration of ytterbium ions.

In conclusion, the present study of spin excitations and EPR in the bilayer antiferromagnetic system reveals a strong anisotropy of the EPR linewidth due to the anisotropic SN interaction between the ytterbium ions via antiferromagnetic spin waves. A comparison of the theoretical and experimental results allows to estimate the exchange coupling between the Yb\(^{3+}\) and Cu\(^{2+}\) ions. The energy gaps in the spin-wave spectra due to the external magnetic field and internal magnetic dipole-dipole interactions were found as well.

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