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COMPETING MAGNETIC INTERACTIONS AND LOW-DIMENSIONAL EFFECTS IN EMR SPECTRA OF $(R_xY_{1-x})_2Cu_2O_5$ SOLID SOLUTIONS

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Copper spin dynamics in $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ and $(Er_{0.5}Y_{0.5})_2Cu_2O_5$ solid solutions has been investigated by electron magnetic resonance (EMR) technique. The temperature dependence of the EMR integrated intensity of the resonance line showed a pronounced maximum at low temperatures and vanished at the transition to the antiferromagnetic phase. The value of temperature at which the EMR integrated intensity reaches maximum is different for the processes of heating and cooling. Study of the product of integrated intensity and temperature allowed determination of the dominating interaction in a particular temperature range. A model used previously to describe the AFM modes in the antiferromagnetic state of $Y_2Cu_2O_5$ was applied to explain the observed changes in the EMR spectra. For $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ the values of appropriate exchange constants connected with a hierarchy of the exchange interactions have been calculated. A dimensional crossover from 2D to 3D magnetic behaviour was observed and interpreted in terms of the spin correlation length.

INTRODUCTION

Compounds from $R_2Cu_2O_5$ group (R=rare-earth ion smaller than Gd, i.e. Tb, Dy, Ho, Yb, Er, Tm, Yb, Lu, as well as Y, Sc, In) crystallize in the orthorhombic $Pna2_1$ space group. The most characteristic feature of $R_2Cu_2O_5$ crystallographic structure is the presence of zig-zag copper chains along the *a*-axis (see Fig. 1). A distorted square planar arrangement of four oxygen atoms around copper exists, with fifth oxygen making a sort of pyramid. These copper-oxygen pyramids are joined at the common edges forming Cu_2O_8 dimers. These dimers, through bridging oxygen, form an infinite zig-zag Cu_2O_5 copper-oxygen chains. Furthermore, each copper ion is coupled to four other Cu ions along the *b*-axis forming *ab*-pseudoplanes. The rare-earth ions are octahedrally coordinated and resulting distorted RO_6 octahedra are linked in a three-dimensional network occupying the space between copper-oxygen planes (see Fig. 2).

All $R_2Cu_2O_5$ compounds order antiferromagnetically at low temperatures, ranging from 11 K (Dy) to 30 K (In), and most of them exhibit metamagnetic behavior below Neel temperature (Matsuoka *et al.*, 1998). The magnetic structure in an ordered state of $R_2Cu_2O_5$ could be viewed as consisting of ferromagnetic CuO layers parallel to the *ab*-plane coupled antiferromagnetically with the copper magnetic moments aligned along the *b*-axis. To explain the appearance of metamagnetic transitions in the antiferromagnetic phase of $R_2Cu_2O_5$ compounds a model has been proposed assuming pairing of two neighbouring Cu^{2+} spins in the copper chain along the *a*-axis (Kimura *et al.*, 1996). This way chains of copper dimers S=1 are formed, which are aligned parallel and form ferromagnetic *ab*-planes. Another parallel ferromagnetic plane of dimers is present in the unit cell of $R_2Cu_2O_5$ structure, and these two planes are coupled antiferromagnetically.

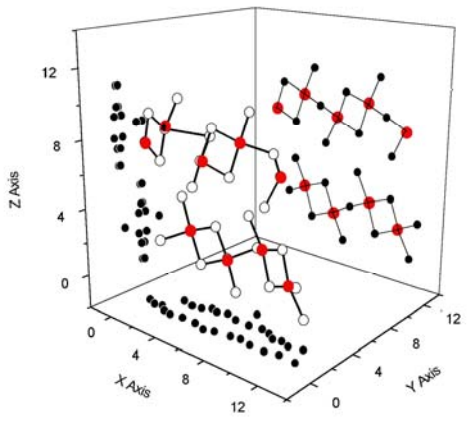

Fig.1. Zig-zag copper chains along the x-axis (two chains in unit cell). Copper atoms are presented as red balls, oxygen atoms as white balls. Rare-earth ions are not shown to make the copper chains more visible.

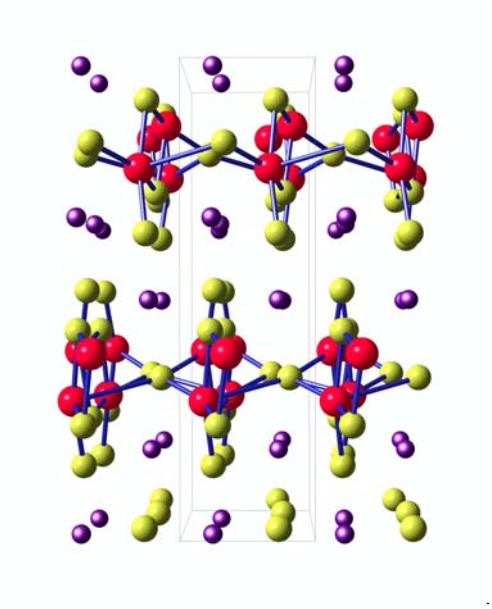

Fig.2. Crystal structure of $R_2Cu_2O_5$ viewed along *a*-axis. Two horizontal CuO *ab*-planes are visible. Six copper (red balls) chains are seen head-on. The rare-earth ions (blue balls) are situated between CuO planes.

An important feature of the $R_2Cu_2O_5$ structure as its magnetic behaviour is concerned is the existence of a hierarchy of exchange pathways: (a) intradimer Cu-O-Cu; (b) interdimer Cu-O-Cu; (c) intraplane and intrachain Cu-O-Cu; (d) interplane Cu-O-R-O-Cu and Cu-O-O-Cu. The coupling of two neighbouring copper ions through two Cu-O-Cu bridges lead to a strong ferromagnetic exchange J_0 within dimeric unit (Fig.3). The exchange between dimers J_1 is probably also ferromagnetic and, as a result, ferromagnetic chains along the *a*-axis are formed at low temperatures. The interaction J_2 between chains within CuO *ab*-plane is assumed also to be ferromagnetic. CuO layers are coupled by a weaker J_{AF} antiferromagnetic exchange.

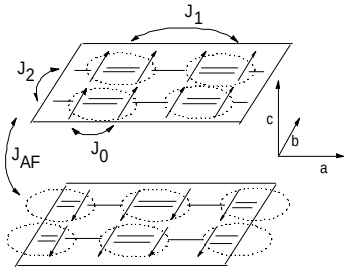

Fig.3. Schematic representation of the exchange interactions between different low-dimensional structural units in the $R_2Cu_2O_5$ structure.

Electron magnetic resonance (EMR) study of $(R_xR'_{1-x})_2Cu_2O_5$ solid solutions (where R and R' are different rare-earth ions) could help to resolve the problems connected with magnetic interactions in the $R_2Cu_2O_5$ structure. Already the following solid solutions have been investigated: $(Er_xY_{1-x})_2Cu_2O_5$, $(Dy_xY_{1-x})_2Cu_2O_5$, and $(Tb_xY_{1-x})_2Cu_2O_5$ (Typek *et al*., 2003; Typek *et al*., 2004; Typek *et al*., 2005). Only signal from the copper(II) ions has been registered by conventional X-band EMR spectroscopy. It was found that the relative EMR signal intensity varies with the magnetic rare-earth ion concentration index *x* according to a simple power law function.

In this paper the EMR study of $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ and $(Er_{0.5}Y_{0.5})_2Cu_2O_5$ solid solutions will be reviewed. The EMR results will be interpreted in frame of a model used previously to explain the metamagnetic transitions in the antiferromagnetic phase of $R_2Cu_2O_5$ compounds. The effects of magnetic low dimensionality and competition of magnetic interactions will be emphasized.

EXPERIMENTAL

Powder samples of $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ and $(Er_{0.5}Y_{0.5})_2Cu_2O_5$ solid solutions have been prepared by heating in air appropriate stoichiometric amounts of metal oxides. X-ray characterization showed that sample were single phase with the following orthorhombic lattice parameters: a=1.0817 nm, b=0.3502 nm, c=1.2462 nm for $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ (Typek *et al*., 2004) and a=1.0878 nm, b=0.3477 nm, c=1.2446 nm for $(Er_{0.5}Y_{0.5})_2Cu_2O_5$ sample (Typek *et al*., 2003).

EMR variable temperature experiments were performed on Bruker E 500 spectrometer operating at X-band microwave frequency equipped with TE_{102} cavity and 100 kHz field modulation. The investigated sample was in form of loose powders. Samples were cooled by flowing He gas and the temperature was controlled within 1% by using a digital temperature controller. Decomposition of the observed EMR spectrum on constituent components has been done by using the SIMPOW computer program.

RESULTS AND DISCUSSION

A. $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$

Integrated intensity defined as the area under the EMR absorption curve is proportional to the static susceptibility χ_{EMR} of spins participating in the resonance. Figure 4 (top panel) presents the temperature dependence of χ_{EMR} in the whole temperature range. Clear maximum of χ_{EMR} is observed at T_{max}=19 K (Typek & Guskos, 2006a, Typek, 2007a). The appearance of maximum of the integrated intensity at

temperature different than T_N=11 K is a clear manifestation of a low dimensional magnetic system. An empirical criterion for determination of the magnetic dimensionality is offered by the ratio T_N/T_{max}, where T_{max} is the temperature of maximum susceptibility (Jacobs *et al*., 1976). For 1D magnetic system T_N/T_{max} <0.1, for the 2D system 0.25<T_N/T_{max} <0.5, and T_N/T_{max} >0.9 for 3D magnet. Applying this simple criterion to our spin system (T_N/T_{max}=0.58) it could be concluded that $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ is 2D magnetic system.

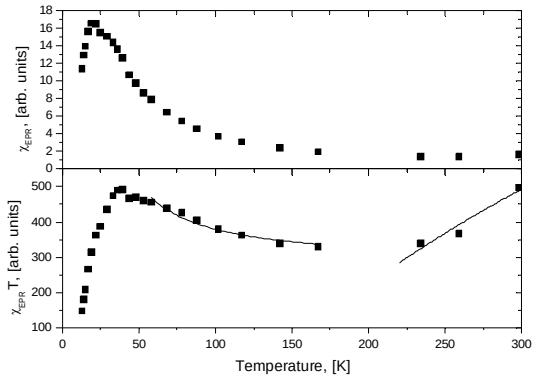

Fig.4. Temperature dependence of the EMR integrated intensity, χ_{EMR} (top panel) and the product of the integrated intensity and temperature, χ_{EMR}·T (lower panel) for $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$. The solid lines are the fittings to the Bleaney-Bowers expression (T>220 K) and the Baker expression (55 K<T<170 K).

The lower panel in Fig. 4 presents the thermal behaviour of the product of the integrated intensity and temperature which is proportional to the square of an effective magnetic moment of the investigated sample. Closer inspection of that dependence reveals three different temperature ranges: the high temperature range, T>200 K, where the antiferromagnetic interaction prevails, the intermediate temperature range, 40<T<200 K, where ferromagnetic interaction dominate, and the low temperature range, T<40 K, where once more the antiferromagnetic interaction takes over.

For intermediate temperature range, the ferromagnetic interactions govern the magnetic behaviour. If a simplifying assumption is made that the S=1 copper dimers form a simple square lattice within *ab* layer, with $J_1=J_2=J$ and the interaction between layers is ignored, than one can use only the isotropic exchange interaction and a nearest-neighbour Heisenberg model. The high-temperature series expansion for the magnetic susceptibility for this model gives (Baker *et al*., 1967):

$$\chi \cdot T / C = 1 + 2J / kT) + 2(J / kT)^2 + \frac{4}{3}(J / kT)^3 + \frac{13}{12}(J / kT)^4 + \frac{71}{60}(J / kT)^5 + \frac{367}{720}(J / kT)^6 \quad (1)$$

where C is the Curie constant, and the terms containing applied magnetic field have been omitted. Fitting of this expression to the ferromagnetic temperature range of the $\chi \cdot T$ product for $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ (i.e.55-170 K) allowed to calculate the exchange constant J/k=14.7(1.2) K. For additional evaluation of J, one may expand the simple Curie-Weiss law and compare the first two terms of the high-temperature series expansion. This comparison gives the following simple relationship: J=0.5 k·Θ. For $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ the Curie-Weiss constant, determined in the 70-170 K temperature range, is Θ=26 K (Typek & Guskos, 2006a), and thus J/k=13 K.

Above 200 K the product $\chi_{EMR} \cdot T$ starts to increase rapidly with the temperature increase. This behaviour apparently has noting to do with the copper-oxygen chains and we propose that it is connected with the presence of a small number of defect magnetic centres in the $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ structure. An obvious candidate for such a defect will be the appearance of a copper dimer coupled by antiferromagnetic exchange interaction. This leads to a singlet ground state and a thermally populated triplet excited state with the temperature dependence of susceptibility, $\chi(T)$ being modelled by the Bleaney-Bowers expression (Bleaney & Bowers, 1952):

$$\chi = \frac{N_A g^2 \mu_B^2}{3kT}\left[1 + \frac{1}{3}\exp\left(-\frac{2J_d}{kT}\right)\right]^{-1} \quad (2)$$

for hamiltonian $H=-2J_dS_aS_b$, where J_d is the antiferromagnetic intra-dimer coupling. The fitting of the high-temperature dependence of the product $\chi \cdot T$ gave J_d/k=-285 K. The Cu-Cu dimer could appear in the $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ crystal structure if the oxygen ion bridging two neighboring coppers is missing.

In the antiferromagnetic temperature range of the $\chi_{EMR} \cdot T$ product (i.e. below 38 K) the effective resonance field increases sharply with lowering temperature (Fig.5). This increase must be attributed to the appearance of local fields created by antiferromagnetically correlated spins between ferromagnetic *ab* layers. The effective resonance field could be approximated by the equation (Rabu *et al*. 2001):

$$B_r = \frac{C_1}{T \cdot \exp\left(\frac{4\pi J_{AF} S^2}{kT}\right)} \quad (3)$$

where C_1 is a constant, and J_{AF} is the antiferromagnetic exchange coupling between the ferromagnetic planes. Assuming that S=1 dimers form ferromagnetic planes, the least-square fitting produced J_{AF}/k=-3.5(2) K.

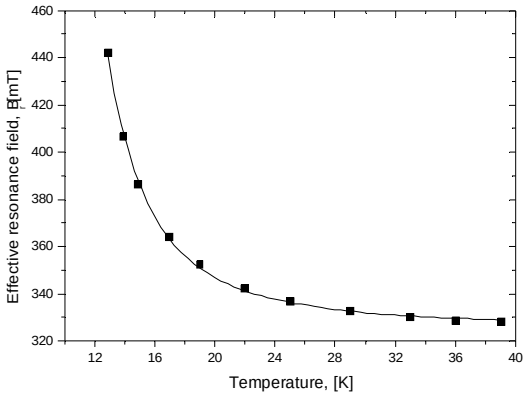

Fig.5. Temperature dependence of the effective resonance field, B_r, in the lowest temperature range. The solid line is the fitting to equation (3).

Another way of estimating the value of J_{AF} will be to use an expression obtained by Schultz (Schultz, 1996). He has considered a model consisting of parallel chains forming a square lattice with the longitudinal ($J_{||}$>0) and transverse ($J_{\perp}$<0) exchange constants. Due to the non-zero interchain coupling, the three-dimensional magnetic long-range order can appear below Neel temperature T_N. Treating the interchain coupling in the mean-field approximation the following equation could been obtained:

$$J_{\perp} = \frac{T_N}{1.28 \cdot \sqrt{\ln\left(\frac{5.8 J_{||}}{T_N}\right)}} \qquad (4)$$

As for $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ T_N=11 K, and using the above calculated value of the exchange coupling along ferromagnetic chain, $J/k=J_{||}/k$=14.7 K, from the equation (4) $J_{\perp}/k$=–4.2 K is calculated. It compares reasonably with the value J_{AF}/k=–3.5 K obtained from the temperature dependence of the effective resonance field. The obtained ratio of $|J/J_{AF}|$~4.3 signifies the importance of low-dimensional effects in the investigated material.

Drastic broadening of the resonance line below 70 K can be attributed to the rapid development of spin correlations with a subsequent growth of 3D correlations due to vicinity of Neel's temperature. When the temperature is lowered toward the critical temperature, the following power law $\Delta B_{eff} \propto \left(\frac{T - T_N}{T_N}\right)^{-p}$ approximately describes the thermal evolution of the effective linewidth. Figure 6 presents the dependence of linewidth as a function of the reduced temperature $\frac{(T - T_N)}{T_N}$ in log-log scale.

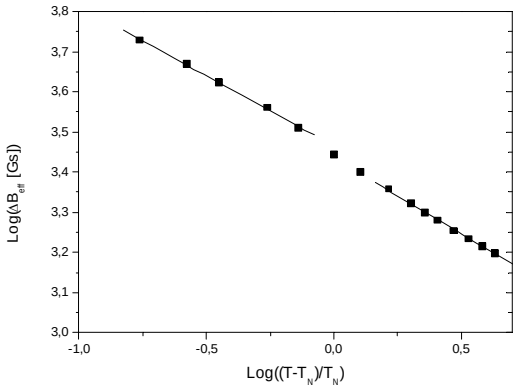

Fig.6. Experimental temperature dependence of the effective linewidth (squares) and fitting by power law (lines).

Experimental points are reasonable well fitted by the power low dependence over a wide temperature range (13 K<T<60 K) with T_N=11 K, but with two different values of the critical exponent p. In the high temperature range (T>30 K) p=0.376(7), in a low temperature range (T<20 K) p=0.349(7). The obtained values reflect the low-dimensional character of the EMR line when it is compared with the critical behavior of linewidth of typical antiferromagnets in the 1D case, such as $CuCl_2 \cdot 2HC_5H_5$ (p=0.5), or in the 3D case, such as GdB_6 (p=1.5). On the other hand, the observed change of value of the critical exponent could be associated with dimensional crossover that the magnetic system undergoes in that temperature range. Below 20 K the linewidth reflects 3D spin correlations, involving different CuO ferromagnetic planes, while above 30 K only the 2D correlations limited to a single CuO plane are involved.

The values of various exchange couplings J_i reported for $Y_2Cu_2O_5$ shows rather broad distribution. In papers (Ramakrishna & Ong, 1988) and (Janicki & Troc, 1992) the magnetic susceptibility was analyzed assuming a model of 1D chain with uniform ferromagnetic intrachain interaction, and the values of 36 K and 88 K were obtained for J_0/k, respectively. In (Paillaud *at al*., 1991) the magnetic susceptibility was modeled by alternating ferromagnetic intrachain interactions. The calculated values of J_0/k and J_1/k were 100 K and 82 K,

respectively. On the other hand, analysis of the antiferromagnetic modes below T_N in submillimetre-wave EMR technique gave for the same exchange couplings the values 340 K and -0.33 K (Kimura *et al*., 1996). As for the value of the sum of interchain interactions zJ' (where z is the number of copper magnetic neighbors) it ranges from -1.6 K (Janicki & Troc, 1992) to -12.4 K (Paillaud *at al*., 1991).

In conclusion, low-dimensional effects in the EMR spectrum of $(Dy_{0.375}Y_{0.625})_2Cu_2O_5$ solid solution have been studied. The model assuming existence of S=1 dimers was employed and the obtained values of exchange interaction constants seems reasonable. The temperature ranges of domination of 2D and 3D magnetic behaviour have been established and the dimensional crossover recorded.

B. $(Er_{0.5}Y_{0.5})_2Cu_2O_5$

Figure 7 (top panel) presents the temperature dependence of χ_{EMR} for the $(Er_{0.5}Y_{0.5})_2Cu_2O_5$ compound for cooling and heating runs (Typek & Guskos, 2006b). Clear maximum of χ_{EMR} is observed during the cooling run at 38 K. The appearance of maximum of the integrated intensity at temperature different than T_N is a clear manifestation of low magnetic dimensionality. Sample was first cooled down to temperature below T_N (EMR signal vanished) and further was heated up. This time the EMR signal started to grow in intensity with the temperature increase, but it was significantly weaker than during the cooling run (Fig. 7, top panel). The lower panel in Fig. 7 presents the thermal behaviour of the product of the integrated intensity and temperature. Thermal behaviour of $\chi_{EMR}\cdot T$ reveals the dominating magnetic interactions in the studied spin system Closer inspection of Fig. 7 reveals the presence of three different temperature regimes with different dominating interactions. For the cooling run: the high temperature range, T>100 K, the intermediate temperature range, 55<T<100 K, where ferromagnetic interactions dominate, and the low temperature range, T<55 K, where the antiferromagnetic interaction takes over. For the heating run there are only two ranges discernable: low temperature range, T<20 K, with antiferromagnetic interaction, and paramagnetic regime, T>20 K, where magnetic moment (and $\chi_{EMR}\cdot T$) do not change.

In the high temperature range, for both runs, the Curie-Weiss law, $\chi=C/(T-\Theta)$, holds, with Θ=26(3) K for the cooling run, and Θ=28.0(6) K for the heating run. Positive value of the Curie-Weiss temperature indicates that between isolated paramagnetic centres the ferromagnetic interaction exists.

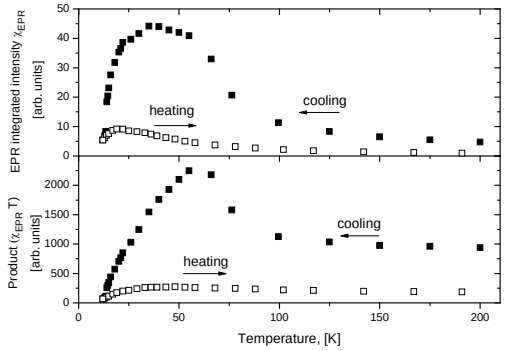

Fig.7. Temperature dependence of the EMR integrated intensity χ_{EMR} (upper panel) and the product of $\chi_{EMR}\cdot T$ (lower panel) for a cooling run (full squares) and heating run (empty squares).

For the cooling run, below 100 K, the magnetic moment starts to increase significantly on further cooling, indicative of strong ferromagnetic interactions. This situation could be described by a model used previously to explain the appearance of metamagnetic transitions in the antiferromagnetic phase of $R_2Cu_2O_5$. It assumes pairing of two neighbouring Cu^{2+} spins in the copper chain along the *a*-axis. Chains of copper dimers S=1 are formed, which are aligned parallel. These chains form ferromagnetic *ab*-planes. Another parallel ferromagnetic plane of dimers is present in the unit cell of $R_2Cu_2O_5$ structure, and these two planes are coupled antiferromagnetically. Thus in this model there is a hierarchy of the exchange interaction couplings: intra-dimer J_0>0, inter-dimer in the chain along the *a*-axis J_1, and inter-chain J_2. It has been estimated that J_0/k~260 K, $|J_1/J_0|$~1/380, $|J_2/J_0|$~1/1100 (Suzuki *et al*., 1994).

In the intermediate temperature range the ferromagnetic interaction governs the magnetic behaviour. If a simplifying assumption is made that the S=1 copper dimers form a simple square lattice within *ab*-layer, with $J_1=J_2=J$ and the interaction between layers is ignored, than one can use the isotropic exchange interaction and a nearest-neighbour Heisenberg model. In this temperature range the spins of a ferromagnetic layer are only correlated on a finite distance ξ, which for isotropic spins is correlated with the exchange constant J by the relationship (Suzuki *et al*., 1994):

$$\xi^2 = \left(\frac{J}{kT}\right)\exp\left(\frac{4\pi JS^2}{kT}\right) \tag{5}$$

For a 2D Heisenberg ferromagnet the magnetic susceptibility is given by (Takahashi, 1987):

$$\chi_{2D} = \frac{1}{3\pi JS}\exp\left(\frac{4\pi JS^2}{kT}\right) \qquad (6)$$

The least-squares fitting to Eq. (6) in the $55<T<100$ K range allowed to calculate the value of the exchange constant J/k=16.9(9) K. From Eq. (5) and (6) follows that the correlation length $\xi \sim \sqrt{\frac{\chi}{T}}$. Thus it could be calculated that between 100 K and 55 K the correlation length increased 2.1 times. This number seems reasonable as the specific dimensions in the crystal structure of $R_2Cu_2O_5$ are considered. The distance between copper ions in the chains running along the *a*-axis is ~3 Å, and the distance between chains along the *b*-axis is ~3.5 Å, while the distance between *ab*-planes is ~6.5 Å. Above 100 K the spins are uncorrelated so the correlation length probably does not exceed 3 Å. Between 100 K and 55 K the correlation length should be large enough as to enable interaction of neighbouring spins, because the spins form a 2D magnetic structure – a ferromagnetic plane (Fig. 8). Below 55 K the antiferromagnetic interaction between *ab*-planes begins to dominate (Fig. 7), what indicates that the correlation length starts to exceed the distance between *ab*-planes. Thus in the low temperature range we have to do with the domination of 3D magnetic behaviour, because the antiferromagnetic interaction between planes takes over the 2D ferromagnetic intraplane interaction. In terms of dimensional crossovers it could be recapitulated that the EMR results of the cooling run show 0D behaviour (separate spins) above 100 K, 2D behaviour in $100<T<55$ K range, and 3D conduct below 55 K (Typek, 2007b).

The heating run is characterised by much smaller EMR signal intensity and it seems to lack the ferromagnetic stage characteristic for the cooling run. Those two features are closely connected and the lack of the 2D magnetism could be explained by too small number of spins in the correlation length to form an interactive group of centres. The EMR active spins are too far away from each other, exceeding the correlation length, to develop ferromagnetic plane structure. The question why in a heating run the are less copper centres visible than during a cooling run is linked with the problem of how a particular rare-earth ion switches out the copper spins from being observed by the X–band EMR spectrometer and how many copper spins is one Er^{3+} ion able to turned out. The latter problem is easier to solve: copper and rare-earth magnetic systems are so strongly connected that the spin-lattice relaxation time τ is very short and the energy absorbed from the microwave field by copper ions is quickly transferred to the lattice through the rare-earths ions. As the EMR linewidth is determined by the relaxation rate τ^{-1}, the line is therefore so broad as to exceed the magnetic field swept by the spectrometer.

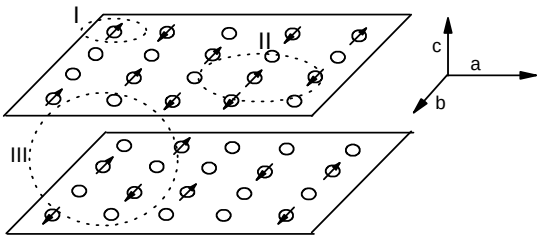

Fig.8. Schematic diagram of copper spins in $R_2Cu_2O_5$. Two CuO ab-planes are presented with only copper dimers S=1 shown. Only those copper dimers with arrows are visible in the EMR spectrum, the rest is EMR silent due to strong interactions with rare-earth ions (not shown). Circles designated I, II, III have the radius of the correlation lengths in the high-, intermediate-, and low-temperature ranges, respectively.

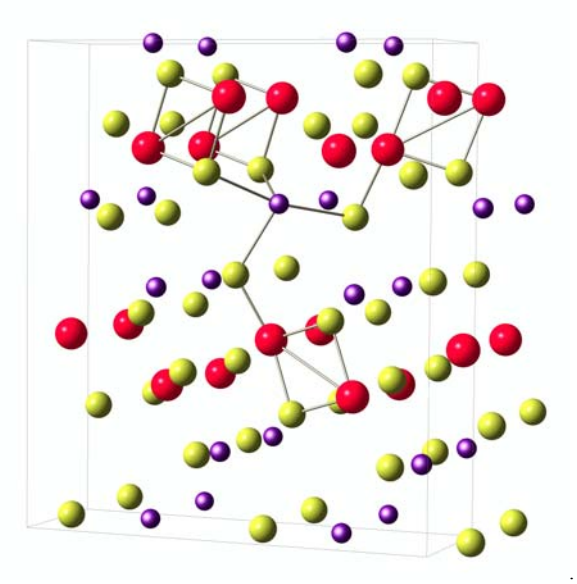

Fig.9. Crystal structure of $R_2Cu_2O_5$ viewed along *b*-axis with marked exchange interaction paths between R (blue balls) and Cu (red balls) ions.

The number of copper spins switched out from EMR by a single rare-earth ion may be determined by inspection of $R_2Cu_2O_5$ crystal structure with marked R-O-Cu exchange interaction paths (there are no direct R-Cu paths) (Fig. 9). As the Fig. 9 shows a single R ion could switch out seven Cu ions (six are in one *ab*-plane and one in another *ab*-plane in a unit cell). We believe this is indeed what happens in a heating run, when the sample is brought to the antiferromagnetic phase and

warmed up. After the sample is kept at room temperature for prolong time, a few of the exchange paths could be broken (e.g. by diffusion of the intervening oxygens) what results in an increase of the EMR signal from the sample. That temporal change of the signal was indeed observed for many $R_2Cu_2O_5$ samples.

In conclusion, from temperature dependence of χ_{EMR} and $\chi_{EMR} \cdot T$, the dominating interactions in a specific temperature range have been established and the dimensional crossover point specified. The value of the interplane exchange interaction constant J/k=16.9 K has been calculated. The role of oxygen ions participating in the R-O-Cu exchange interaction paths has been emphasized in change of magnetic dimensionality.

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