MAGNETIC STATE AND SPIN DYNAMICS OF $Cu_3B_2O_6$ SINGLE CRYSTAL

G.Petrakovskii, [1]B.Roessli, [1]J.Shefer, L.Bezmaternikh, O.Bayukov, [1]C.Neidermayer,
Institute of Physics SB RAS, Krasnoyarsk 660036, Russia
1) Paul-Sherrer Institute, Villigen, Switzerland

1.Introduction

Low-dimensional compounds attract intensive attention due to the variety of the ground magnetic states and the types of the magnetic excitations in this systems. $Cu_3B_2O_6$ is a copper oxide compound 2D structure. The magnetic susceptibility shows a broad maximum around 40K and sharp decrease below 11K [1]. In this paper was suggested that the kink in the susceptibility at 11 K corresponds probably to a collective phase transition from the paramagnetic state to a singlet ground state. It is appropriate to note that it was found theoretically [2] that such phase transition in 2D spin system may take place by the magnetoelastic coupling similar to spin-Peierls phase transition in 1D spin systems. The following investigations [3] showed that the magnetic structure of $Cu_3B_2O_6$ is complicated – probably it is the mixture of the magnetic ordering and the singlet state. So, in our opinion, the neutron scattering investigation is desirable for certain determination of the magnetic structure of $Cu_3B_2O_6$.

2. Experimental details

A. Sample preparation

Single crystal of $Cu_3B_2O_6$ with volume about 1 cm^3 was grown by spontaneous crystallization. The $Cu_3B_2O_6$ powder was prepared by standard ceramic method. The average Cu-O distance in *bc*-plane is 1.95 A°, while that between the neighboring planes is 2.90 A°. Thus, this compound is expected to belong to the 2D spin system [1].

B. Magnetic measurements

The power and single crystal magnetic susceptibility were performed with PPMS device (Quantum Design) Fig.1.The magnetic susceptibility shows the broad maximum near 39 K, decreases sharply at the temperature below 10K and grows when the temperature is decreased below 5K. We note that the magnitude of the magnetization is so small as ~0.05 μ_B/Cu at 30T for 1.5K[4].

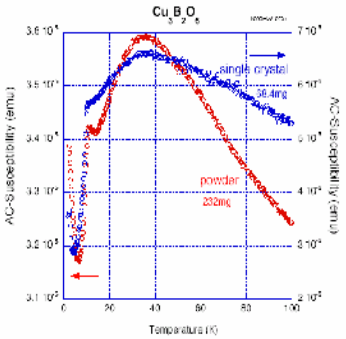

Fig.1

C. .Specific heat measurements

The specific heat measurements were performed with PPMS device in the temperature range 1.8≤T≤27K. We used a small mass 4.5 mg single crystal. Fig.2 shows the temperature dependence of the specific heat with the anomaly at the temperature T_c =9.8K. The shape of the anomaly indicates that phase transition is of second order. Because nothing the structure anomaly have found and because a strong dependence of the position of the anomaly at T_c on magnetic field take place it is possible to think that the temperature 9.8K is the phase transition in spin system. The similar specific heat anomaly was observed in spin-Peierls compound $CuGeO_3$ [5] and this fact supports the notion of a singlet state phase transition [6]. The temperature dependence of the specific heat for temperature range 3 – 9 K can be described by $C= \beta T^3 + \gamma \exp(-$

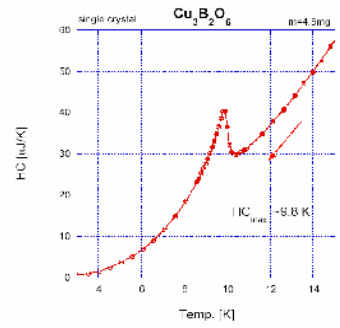

Fig.2

ΔE/T). The exponential T-dependence of the magnetic contribution with ΔE≈38 K indicated the opening of a gap in the magnetic excitations spectrum.

D. Neutron scattering measuremants

The elastic and inelastic neutron scattering measurements were performed with the single crystal diffractometer TriCS, powder diffractometer DMC and single crystal inelastic neutron scattering spectrometer TASP (SINQ, PSI). Fig.3 shows the elastic neutron scattering patterns for powder of $Cu_3B_2O_6$ at the temperatures 1.5K and 15K. There is nothing difference between two patterns for these two temperatures. The same result was found in case measuring of the elastic neutron scattering on single crystal sample. So, nothing magnetic long range ordering down to 1.5K found in this compound.

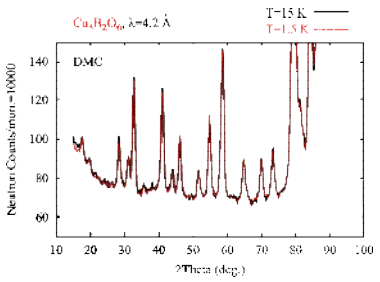

Fig.3

The inelastic neutron scattering investigation was performed using a horizontal focusing analyzer and no collimation in the beam, λ=1.97A°. The results of measuring for temperatures 1.5K and 100K are shown on Fig.4,5. The energy gap about 3.4 meV it can see clearly for the temperature 1.5K.

3. DISCUSSION

The elementary cell of $Cu_3B_2O_6$ contains 30 Cu^{2+} cations: 6 Cu^{2+} are localized in squares, 4 – in distorted pyramids and 20 – in distorted octahedrons of oxygen ions. There are 12 nonequivalent sites of Cu^{2+}ions in lattice. Crystal structure is 2D, so it is possible to take into account the exchange interactions only inside plane. We have here: six Cu^{2+} single ions, which connected with neighbours by 135° exchange bonds, eight pairs Cu^{2+}ions, which connected each other by 90° exchange bonds and every ion of pair connected with neighbour Cu^{2+} ions by two of 135° exchange bonds; two quartets with four ions connected each other by 90° exchange bonds and two inside ions connected by 135° exchange bonds with neighbour Cu^{2+} ions. Using the simple model of the exchange bonds we received exchange parameters: $J_{135°}$=-21K; $J_{90°}$=+9K for usual case, when all holes on Cu^{2+} ions are on d_{x2-y2} orbitals; and $J_{135°}$=21K; $J'_{135°}$=-7K; $J''_{135°}$=-4K; $J^{o.o}{}_{90°}$=-36K for case of the orbital ordering. Here we used exchange spin Hamiltonian in form H==-2J $\mathbf{S_1S_2}$. In case of orbital ordering there is the singletization of pairs Cu^{2+} ions, when the pairs and quartets of Cu^{2+}ions are characterized by the total spin equals zero. The pairs and fours form 2D lattice by the antiferromagnet bonds each other. The part of these exchange interactions is frustrated. The exchange interaction of the Cu^{2+} single ions is very week and ones are the paramagnetic down to low temperature because the spin system is 2D and the magnetic anisotropy is small. So, it can think, that spin system of $Cu_3B_2O_6$ compound is the superposition of the pairs and fours of spins and paramagnetic single spins. Because the ground state of the antiferromagnetically coupled spins of pairs and fours is singlet and the first excited state is triplet we can call this spin subsystem as singlet-triplet spin subsystem.

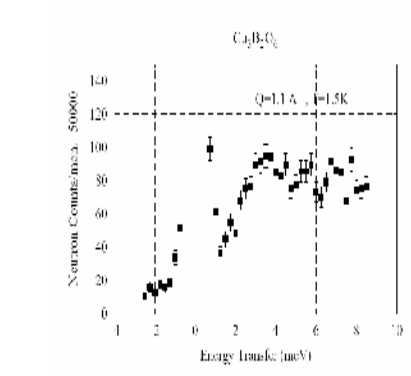

Fig.4

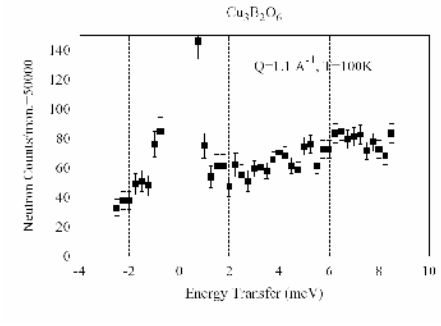

Fig.5

The totality of the experimental data permits to

suggest the following picture for the magnetic state of the spin subsystem $Cu_3B_2O_6$. There are three spin subsystems: the single ions Cu^{2+} (6 spins per unit cell) connected by week exchange interactions; the pairs of spins Cu^{2+} (8 pairs per unit cell); four of spins Cu^{2+} (2 fours per unit cell). The unit spins are remained as paramagnetic down to low temperatures. Their magnetic susceptibility $\chi(s)$ follows to Curie low. The isolated pairs and fours form the spin subsystem in which the ground state is singlet and the first excited state is triplet separated from ground state by the energy gap ~2J. The magnetic susceptibility of this subsystem $\chi(st)$ shows the maximums at the temperatures near J_{pair} and J_{four} and after this goes exponentially to zero. For interacted pairs and fours we must replace 2J to energy gap ΔE.The total magnetic susceptibility $\chi(\Sigma)=\chi(s)+\chi(st)$ is $\chi(\Sigma)==(0.00074/T)\ \exp(-11/T) + (0.00328/T)\ \exp(-56.55/T) + 0.00008/T$. This susceptibility was found by fitting $\chi(\Sigma)$ to experimental data (Fig.1). The exchange interaction between pairs and fours spin subsystems together with magnetic anisotropy in (*bc*) plane trends to order the singlet-triplet subsystem in competition with the aspiration of one to singlet state. As result, the phase transition to singlet state is at the temperature T_c=9.8K. Such picture of the crystal spin subsystem organization explains quite the neutron scattering data. There is not the long range magnetic ordering in accordance with fact, that the elastic neutron scattering don't shows the magnetic reflexes both for single crystal and powder. The singlet-triplet spin subsystem is characterized by the energy gap presence and this fact is supported by the inelastic neutron scattering data on the energy gap ~35K.

The reason of the specific heat anomaly is the magnetic phase transition to the singlet spin state. Near temperature T_c=9.8K the spin system shows the magnetic phase transition to singlet spin state for part of spin system of magnet. This phase transition is the result of the competition singlet spin state and long range ordered magnetic state. Strong anisotropy of the T_c dependence on magnetic field for different directions of magnetic field [5] is result of 2D character forming of the magnetic ordering , namely the temperature of magnetic ordering is determined by the anisotropy λ in (*bc*) plane: $T_c \approx \pi J/\ln(J/\lambda)$ [7]. So, for magnetic field in plane take place the competition between the anisotropy λ and the magnetic field, but for the magnetic field directed perpendicular plane take place the competition between the exchange interaction J and the magnetic field. It because the value of the magnetic anisotropy is much smaller than the exchange interaction, the field dependence of T_c for field in plane much stronger than one for field directed perpendicular plane. Near T_c a critical slowing down of the fluctuations of electronic spins is not observed [5]. This fact is in agreement with Buzdin, Bulaevskii paper [6], they found that in case of the phase transition to singlet state the critical region is very small.

As conclusion we found, that spin system of $Cu_3B_2O_6$ shows the phase transition to singlet state at the temperature near 10K. The magnetic system of this compound is the superposition of the paramagnetic and spin singlet subsystems.

Acknowledgements

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