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Slow Magnetic Relaxation in a S=1/2 copper-based mononuclear complexe

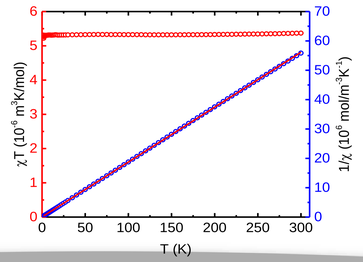
***A. Kliuikov*[1]**, O. Bukrynov[3], I. Potocnák[2], S. Vitushkina[3], L. Váhovská[4], and E. Cižmár[1]

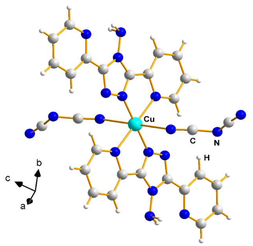
Motivation

- Low dimensional magnetic systems use in quantum computing as an implementation of qubits, memory with high recording density [1-3].
- Design and synthesis of new molecular magnets with a higher "working" temperature and with record values of the magnetization reversal barrier, for successful use in practical application.
- Obtaining molecular systems with significant magnetic relaxation times, a key task for creating quantum information processing devices [4].

Introduction

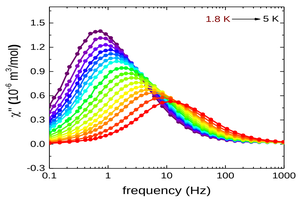
- The crystal structure of $Cu(abpt)_2(dca)_2$ consists of isolated molecules with Cu(II) ions carrying the spin S = ½.
- The temperature dependence of χT displays a decrease at very low temperatures (below 5 K), suggesting the presence of a weak antiferromagnetic (AF) exchange coupling.
- The room temperature value of χT of 5.37×10^{-6} m^{3}K/mol is typical for Cu(II) ions.
- A weak AF exchange coupling is manifested by a small value of the Weiss temperature of -0.24 K
- The presence of a very small exchange interaction supported the expected model of well-isolated molecules.

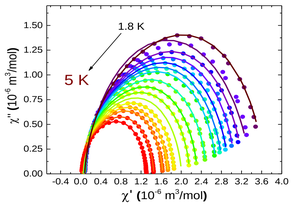


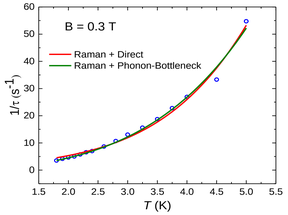


AC Measurements

Polycrystals

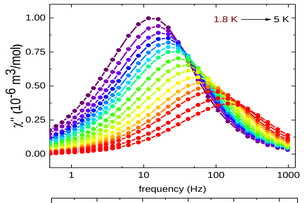


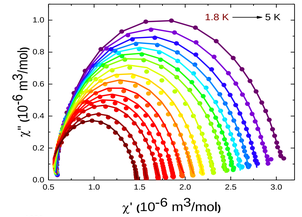


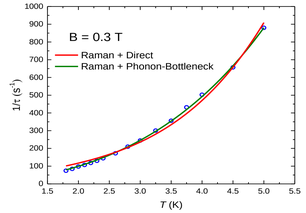


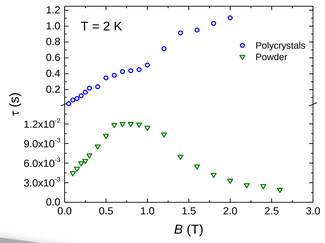
- and the relaxation is acceleratedIncrease of the relaxation time polycrystals sample was observed with the increasing magnetic field up to 2 T at 2 K
- Such slowing down of the relaxation up to magnetic fields even 3 T was observed in V(IV)-based complexed [1], above which a direct process dominates .

Powder









- The Cole-Cole diagrams indicate the presence of a single relaxation channel
- Used two models for analysis of the spin dynamics of the system to evaluate the relaxation mechanism:

Direct and Raman relaxation processes

$$\tau^{-1} = AB^m T + CT^n$$

Phonon Bottleneck effect and Raman relaxation process

$$\tau^{-1} = A_{PB}T^2 + CT^n$$

For polycrystals sample fit parameters
- Direct + Raman, A = 2.126 s^{-1}, C = 0.068 s^{-1},
- PB + Raman, A_{PB} = 0.947 s^{-1}, C = 0.046 s^{-1}.

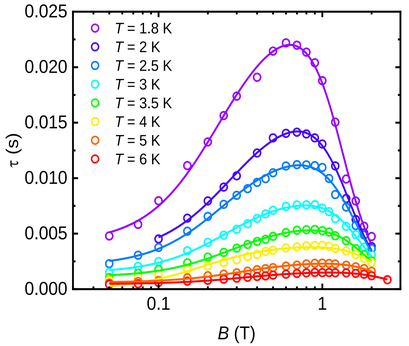
For powder sample fit parameters
- Direct + Raman, A = 50.145 s^{-1}, C = 1.058 s^{-1},
- PB + Raman, A_{PB} = 22.849 s^{-1}, C = 0.494 s^{-1}.

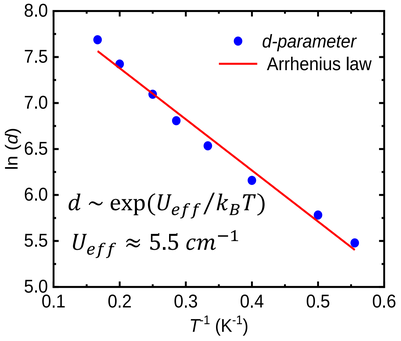
- Two types PB effect: the transfer of energy in space (***spatial bottleneck***) or in frequency (***spectral bottleneck***).

spatial bottleneck - when the heat generated at the spin and lattice coupled modes cannot be evacuated to the bath because of weak thermal contact between the sample and the bath.
spectral bottleneck - the heat generated by the excitation of the spin system by the ac field cannot be transferred because there are not sufficient phonons in the lattice vibration dispersion to absorb it.

- After pulverizing the nascent polycrystalline sample a typical behavior proposed for the combination of the field-dependent Raman process, described by Brons-Van Vleck model, and the direct process was observed.
- The behavior at fields B > 0.8 T is related to the more efficient direct mechanism of relaxation
- At the low fields, the Raman process will dominate the slow magnetic relaxation.

The low energy vibrations in Raman process





Brons–van Vleck model:

$$\tau^{-1} = cB^4 + d\frac{1+eB^2}{1+fB^2}$$

parameter ***d*** – activated behavior related to the energy of phonons participating in the Raman relaxation process [2]

- How the energy values of the vibrational modes correlates with the extracted activation energies of the relaxation mechanism?
- Low-energy phonons vibrational modes – anharmonic phonons, intermolecular vibrations – can be detected in far-infrared spectroscopy.

Conclusion

- The compound $Cu(abpt)_2(dca)_2$ consists of isolated molecules with spin S = ½.
- The magnetic relaxation characteristics obtained in the compound under study indicate an interesting example of slow magnetic relaxation, where the temperature dependence of the one-phonon direct relaxation process is renormalized under the influence of the PB effect.
- A possibility to suppress the observed PB effect allows extracting also the typical energy of the phonons participating in the Raman process from the field-dependence of the relaxation time estimated in the wide temperature and magnetic-field range using the Brons-van Vleck model.
- The extracted energy of low-lying phonon modes of 11 cm^{-1} is suggested to be related to the local molecular rotation and torsion modes of the molecular units in the crystal structure of $Cu(abpt)_2(dca)_2$.

References
1. Tesi et al., Dalton Trans. 42 (2016), 16635
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Acknowledgement
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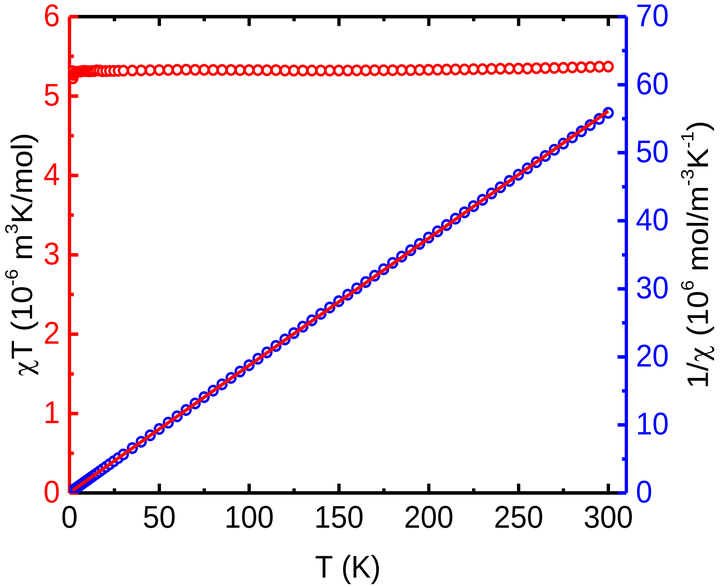

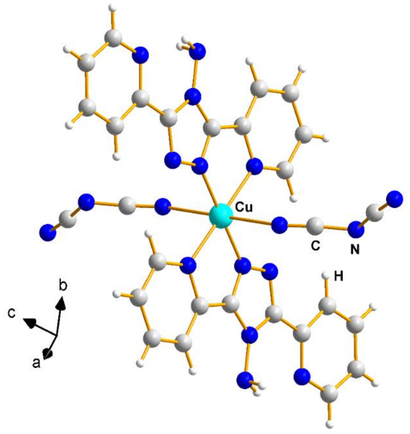

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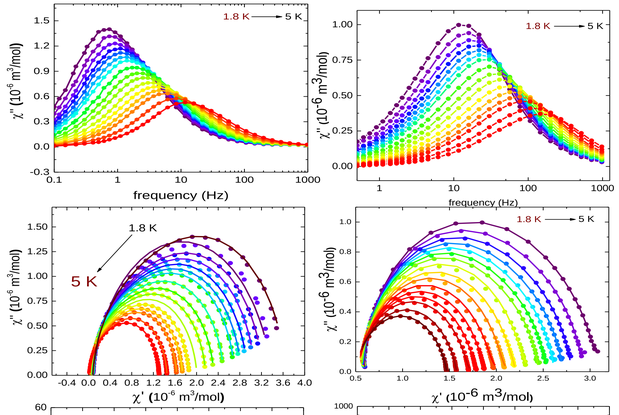

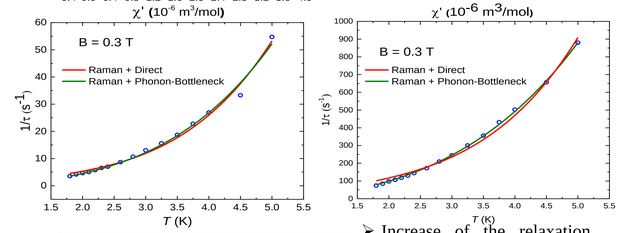

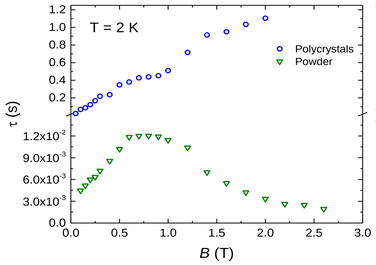

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- The behavior at fields B > 0.8 T is related to the more efficient direct mechanism of relaxation
- At the low fields, the Raman process will dominate the slow magnetic relaxation.

- For polycrystal sample, a maximum is observed at 0.6 Hz (at 1.8 K), whose position shifts to higher frequencies with a further increase of temperature.
- Reducing the size of the crystallites leads to an acceleration of the magnetic relaxation, which is obvious from the observation of the position of the maximum in the imaginary component of the AC susceptibility.
- The Cole-Cole diagrams indicate the presence of a single relaxation channel

Used two models for analysis of the spin dynamics of the system to evaluate the relaxation mechanism:

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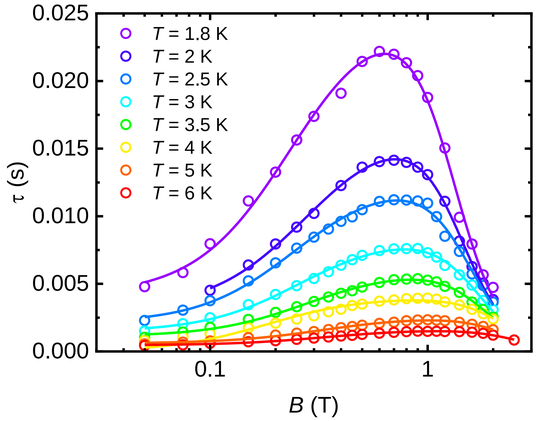

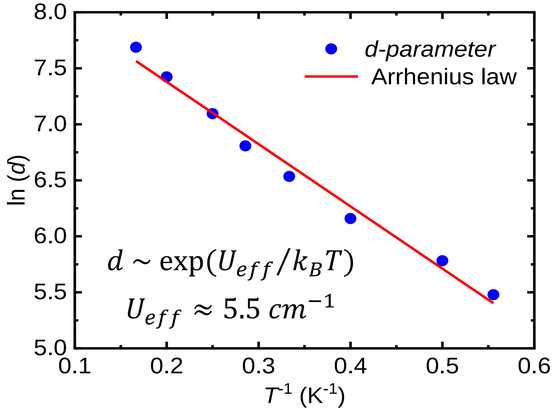

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We studied the compound $Cu(abpt)_2(dca)_2$, which consists of isolated molecules with spin S = ½. The type of magnetic relaxation, the number of processes, and the influence of other factors on the magnetic processes in the sample were determined. The magnetic relaxation characteristics obtained in the compound under study indicate an interesting example of slow magnetic relaxation, where the temperature dependence of the one-phonon direct relaxation process is renormalized under the influence of the PB effect.. A possibility to suppress the observed PB effect allows extracting also the typical energy of the phonons participating in the Raman process from the field-dependence of the relaxation time estimated in the wide temperature and magnetic-field range using the Brons-van Vleck model. The extracted energy of low-lying phonon modes of 11 cm^{-1} is suggested to be related to the local molecular rotation and torsion modes of the molecular units in the crystal structure of $Cu(abpt)_2(dca)_2$.

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