SPIN-TRIPLET EXCITONS IN HAN PURPLE PIGMENT $BaCuSi_2O_6$: ELECTRON PARAMAGNETIC RESONANCE STUDIES

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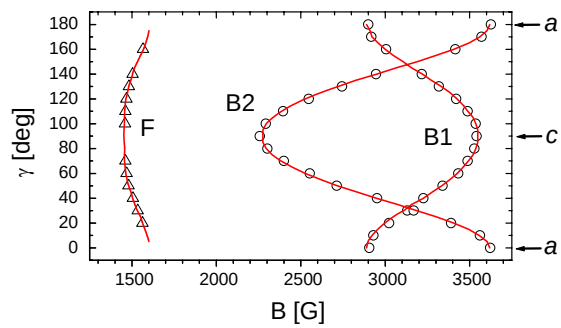
Introduction

$BaCuSi_2O_6$, also known as Han Purple Pigment, can be regarded as an almost ideal realization of the $S = 1/2$ system of weakly-interacting spin dimers with the spin-singlet ground state and gapped excitation spectrum [1]. By application of external magnetic field the gap can be closed, creating a gas of interacting bosonic spin-triplet excitations (triplons). In $BaCuSi_2O_6$ this phenomenon can be effectively described in terms of the field-induced Bose-Einstein condensation of triplons [2]. Here, we focus on another interesting phenomenon associated with interacting excited triplets – but in the low-field quantum-disordered state, employing temperature as a tuning parameter.

Experimental

Experiments were performed using a Bruker Elexsys E680X spectrometer at the X-band frequency (~ 9.4 GHz).

Results and Discussion



A very rich EPR spectrum has been observed in $BaCuSi_2O_6$ at low temperatures. It was revealed that the low-temperature multiple absorptions are intimately related to a thermally-activated spin-triplet exciton superstructure. Analyzing the angular dependence of exciton modes in $BaCuSi_2O_6$ allowed the precise calculation of the anisotropy parameter, $D = 0.07$ cm^{-1}. The figure shows the angular dependence of the electron paramagnetic resonance (EPR) spectrum in $BaCuSi_2O_6$ in the *ac* plane, taken at a temperature of 6 K. γ is the angle between the *a* and *c* axis. Triangles and circles correspond to experimental data, while lines correspond to simulation results. It is argued that spin-triplet excitons are a generic feature of low-dimensional $S = 1/2$ spin systems with the dimerized spin-singlet ground state; the described procedure can be applied for accurate estimating anisotropy constants in a large number of quantum-gapped systems. In addition, the temperature dependence of the EPR intensity and linewidth was measured and discussed. The magnitude of the energy spin gap Δ, determined by analyzing the temperature dependence of the integrated signal intensity, is 53 K.

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References

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