Bose-Einstein Condensation of Magnons in Han Purple $BaCuSi_2O_6$

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Han purple ($BaCuSi_2O_6$) is a valuable model material for studying Bose-Einstein condensation (BEC) of magnons in high magnetic fields [1]. In order to understand the nature of the two-dimensional (2D) BEC phase in $BaCuSi_2O_6$, we performed detailed ^{63}Cu and ^{29}Si nuclear magnetic resonance (NMR) studies [2, 3] above the critical magnetic field, H_{c1} = 23.4 T.

We also investigated the low-temperature crystal structure of $BaCuSi_2O_6$ with high-resolution synchrotron x-ray and neutron powder diffraction techniques [4] and found it to be on average (ignoring the incommensurate modulation) orthorhombic, with the most probable space group *Ibam*. The Cu-Cu dimers in Han purple are forming two types of 2D layers with distinctly different interatomic distances. Subtle changes also modify the interlayer Cu-Cu exchange paths. The two different alternating layers present in the system have very different local magnetizations close to H_{c1}; one is very weak, and its size and field dependence are highly sensitive to the nature of inter-layer coupling. Its precise value could only be determined by "on-site" ^{63}Cu NMR [2], and the data are fully reproduced by a model of interacting hard-core bosons in which the originally suggested strong frustration associated to tetragonal symmetry is lifted, leading to the conclusion that the population of the less populated layers is not fully incoherent but must be partially condensed.

Using precise low-temperature structural data [4] and extensive density-functional calculations, we elucidated magnetic couplings in this compound [5]. The resulting magnetic model comprises two types of nonequivalent spin dimers, in excellent agreement with the $^{63,65}Cu$ NMR data. We further argue that leading inter-dimer couplings connect the upper site of one dimer to the bottom site of the contiguous dimer, and not the upper-to-upper and bottom- to-bottom sites, as assumed previously. This finding is verified by inelastic neutron scattering data and implies the lack of frustration between the layers of spin dimers in $BaCuSi_2O_6$, thus challenging existing theories of the 2D-like BEC of magnons in this compound.

Recently, the substituted system $(Ba_{1-x}Sr_x)CuSi_2O_6$ with a stable tetragonal crystal structure down to 2 K has been discovered [6]. From low-temperature neutron and synchrotron powder diffraction, room- and low temperature NMR, magnetic- and specific-heat measurements it is verified, that the structural phase transition into the orthorhombic structure is absent for the $x = 0.1$ sample.

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