Polarized-neutron investigation of magnetic ordering and spin dynamics in BaCo$_2$(AsO$_4$)$_2$ frustrated honeycomb-lattice magnet

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Abstract

The magnetic properties of the cobaltite BaCo$_2$(AsO$_4$)$_2$, a good realization of the quasi two-dimensional frustrated honeycomb-lattice system with strong planar anisotropy, have been reinvestigated by means of spherical neutron polarimetry with CRYOPAD. From accurate measurements of polarization matrices both on elastic and inelastic contributions as a function of the scattering vector $Q$, we have been able to determine the low-temperature magnetic structure of BaCo$_2$(AsO$_4$)$_2$ and reveal its puzzling in-plane spin dynamics. Surprisingly, the ground-state structure (described by an incommensurate propagation vector $k_1 = (k_x,0,k_z)$, with $k_x = 0.270\pm0.005$ and $k_z \approx -1.31$) appears to be a quasi-collinear structure, and not a simple helix, as previously determined. In addition, our results have revealed the existence of a non-negligible out-of-plane moment component $\approx0.25\mu_B$/Co$^{2+}$, representing about 10% of the in-plane component, as demonstrated by the presence of finite off-diagonal elements $P_{yz}$ and $P_{zy}$ of the polarization matrix, both on elastic and inelastic magnetic contributions. Despite a clear evidence of the existence of a slightly inelastic contribution of structural origin superimposed to the magnetic excitations at the scattering vectors $Q = (0.27,0,3.1)$ and $Q = (0.73,0,0.8)$ (energy...
transfer $\Delta E \approx 2.3$ meV), no strong inelastic nuclear-magnetic interference terms could be detected so far, meaning that the nuclear and magnetic degrees of freedom have very weak cross-correlations. The strong inelastic $P_{xz}$ and $P_{zy}$ matrix elements can be understood by assuming that the magnetic excitations in BaCo$_2$(AsO$_4$)$_2$ are spin waves associated with trivial anisotropic precessions of the magnetic moments involved in the canted incommensurate structure.

Keywords: Condensed Matter Physics, Materials Science

1. Introduction

When the space dimension $D$ is lowered from three, to two (2D, planar system) and finally to one (1D, chain system), the magnetism displays more and more interesting and non-trivial features, as a result of the enhancement of both the thermal and quantum fluctuations. In the extreme cases, this leads to the lack of three-dimensional long-range ordering (LRO) down to $T = 0$ K, the occurrence of spin-liquid states, and finally the emergence of unconventional spin dynamics. As established from numerous theoretical studies, both the ground state (GS) and the excited states of low-dimensional quantum magnets appear more and more exotic as the dimension of the spin-space $n$ increases ($n = 1$ for the Ising system, $n = 2$ for the XY system and $n = 3$ for the Heisenberg system), or the spin quantum number $S$ decreases (from $S = \infty$ for the classical case, down to $S = 1$ and $S = 1/2$ for the extreme quantum spin). The nature of the ground state depends also strongly on the connectivity of the lattice (i.e. the number of next-nearest neighbor spins) and the type of spin–spin couplings which are involved: Ferromagnetic (F), antiferromagnetic (AF) or competing between first and second neighbor spins, frustrating or not frustrating the spin lattice, at short range or at long range. As it is now well admitted, the largest effects are seen for the 1D antiferromagnetic Heisenberg (HAF) chain system, which indeed displays drastically different ground states (GS) and spin-excitation spectra, depending whether the spin value is half-integer ($S = 1/2, 3/2, ...$) or integer ($S = 1, 2, ...$) [1]. More precisely, for the latter Haldane predicted the existence of a non-magnetic $S = 0$ singlet GS separated from the first triplet of excited states by a quantum energy gap $E_G \sim JS\exp(-\pi S)$, $J$ being the inter-spin coupling constant ($E_G \approx 0.41J$ for $S = 1$ from numerical calculations [2]). Conversely, for the $S = 1/2$ HAF chain, the magnetic excitation spectrum should be a gapless continuum of magnetic excitations [3], but the introduction of frustrating AF second-neighbor interactions ($J1 - J2$ model) leads to the opening of a gap above some critical ratio, by spontaneous dimerization of the spin system [4]. Similar effects have been predicted for the $S = 1/2 p$-leg AF spin-ladder system, which indeed realize the cross-over between the $S = 1/2$ HAF chain ($p = 1$) and the $S = 1/2$ HAF square plane ($p = \infty$). For $p$ even-integer ($p = 2, 4, ...$), the GS of the $S = 1/2$ HAF p-
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