Incommensurate modulation of the spin frustration in a triangular lattice of the Et$_2$Me$_2$Sb[Pd(dmit)$_2$]$_2$ molecular conductor

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The charge transfer complexes consisting of a dimer of Pd(dmit)$_2$ anion, Z[Pd(dmit)$_2$]$_2$ (Z is a monovalent cation and dmit=1,3-dithiol-2-thione-4,5-dithiolate), show diverse phases such as metal (M), dimer Mott insulator (DM), charge separation (CS), valence bond liquid, and valence bond solid because of the competition of interactions among charges and spins in a two-dimensional triangular lattice [1].

Among those complexes, the Et$_2$Me$_2$Sb[Pd(dmit)$_2$]$_2$ crystal is rather peculiar since the triangular lattice formed by the interdimer interactions is approximately equilateral. The degree of spin frustration in this case is maximum and is at the origin of its magnetic properties. In particular, the temperature dependence of the magnetic susceptibility can be described by a model of a spin-1/2 Heisenberg antiferromagnet on a triangular lattice.

We investigated the structure of the incommensurate phase in this prototype frustrated triangular lattice crystal using high resolution X-ray diffraction. Below 105K, several satellites Bragg peaks appears revealing an incommensurate phase. We used the superspace formalism to solve the structure in 4D and showed that a competition between charge, magnetic order and structural order exists in this phase. The interdimer transfer integrals couple to the distortion in the dimer are incommensurately modulated suggesting that the origin of this phase is the appearance of an incommensurate modulation of the degree of the spin frustration in a triangular lattice. Although such incommensurate correlations have been theoretically predicted [2, 3], only few experimental evidences of its existence has been reported. Magnetic, spin muon relaxation and optical signature of the incommensurate phase are discussed in the frame of the phenomenological model of incommensurate spin frustration.

Fig. 1. Illustration of spin frustration in a triangular lattice [2], signature of the incommensurate phase (phase II) in the magnetic susceptibility, structure of the incommensurate phase showing a strong steric effect between the cation and the anions.

References