$S_{10}$

# ORDER-DISORDER PHASE TRANSITION, 2d → 3d CROSSOVER, CHEMICAL RANDOMNESS, AND ANNNI MODEL IN DIAMAGNETIC MIXED CRYSTALS: $Rb_x(NH_4)_{1-x}AlF_4$

A. Leblé*, A. Jouanneaux, J. C. Fayet
Laboratoire de Spectroscopie du Solide, U.A. no 807, Faculté des Sciences, Université de Maine, 72017 Le Mans Cédex (France)

and J. L. Fourquet
Laboratoire des Fluorures, U.A. no 449, Faculté des Sciences, Université du Maine, 72017 Le Mans Cédex (France)

Ionic fluorides very often constitute physical examples for which basic theoretical models may apply and can be checked by accurate measurements. This work is a part of a general study of the tetrafluoroaluminates $M^IAlF_4$ [$M^I$ = K, Rb, Tl, $NH_4$] and in this paper we consider $Rb_x(NH_4)_{1-x}AlF_4$ as a typical random-axial-Ising-like system [n = 1]. We report the results of local measurements with the help of an E.S.R. probe : $Fe^{3+}$. The single crystals have been grown by hydrothermal synthesis for various $Rb^+$ atomic concentrations [0 < x < 45%]. In the pure $NH_4AlF_4$ crystal, we have evidenced an order disorder phase transition [I4/mcm → $P4_2$/mbc] and the local measurements unambiguously indicate that the low temperature order is characterised by a parallel order of the $NH_4^+$ pseudo spin in [001] layers and an antiparallel order along [001] axis. The intra layer coupling being stronger than the inter layer one, we have observed a 2d → 3d crossover in the course of critical slowing down, as expected for such quasi 2d systems. In the mixed crystals, the E.S.R. measurements at room temperature show very clearly a random distribution of the $Rb^+$ and $NH_4^+$ ions, and permit an accurate determination of x. At low temperature, the ordering of the ammonium ions depends on the $Rb^+$ concentrations: the determination of the local symmetry at the $Al^{3+}$ site indicates that the ordering of adjacent ammonium layers is no more fully antiparallel for 4% < x < 10% and becomes only parallel for x = 25%. These results are inferred from a sampling of the second shell of ions surrounding the $Al^{3+}$ site by the E.S.R. probe. They can be interpreted by an ANNNI-like background Hamiltonian perturbed by a random spin-glass-like one.