3.7 Anomalous Band Magnetism in $LaCrSb_3$: Evidence for Double Exchange

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The magnetic exchange mechanisms and spin-correlations mediated by itinerant electrons have attracted renewed interest in the last few years, in part stimulated by the promising technological possibilities of the emerging field of spin electronics. The system of intermetallic antimonides $RCrSb_3$ (R=La-Nd, Sm, and Gd-Dy) was recently discovered, crystallizing in a novel layered orthorhombic structure (*Pbcm* symmetry), with planes of Sb, R, and a layer of $CrSb_2$, as shown in Fig. 1(a)[1,2]. Previous studies showed interesting magnetic and transport properties. The parent compound, $LaCrSb_3$ has been described as a band ferromagnet. The saturation magnetization at low-T (M_{sat}) strongly depends on the sample growth procedure, which is typical of itinerant magnets, and values in the range 0.8 μ_B/f.u. $< M_{sat} <$ 1.7 μ_B/f.u. have been reported. The $RCrSb_3$ compounds with R = Ce, Pr, Nd, and Sm also show a net ferromagnetic (FM) moment from Cr spins below T_C ~ 105 K - 147 K, with possible orderings of the R moments below ~10 K - 15 K. In contrast, the compounds with R = Gd, Tb, and Dy show no spontaneous FM moment at any temperature, suggesting a quantum phase transition of the magnetic ground state of this system.

We performed neutron diffraction measurements on a crystal of $LaCrSb_3$ showing M_{sat} = 1.62 μ_B/f.u. and T_C = 126.0(6) K. A coexistence of orthogonal ferromagnetic and antiferromagnetic spin sublattices was observed (1.83(4) μ_B /Cr and 0.54(4) μ_B/Cr, respectively), revealing a close competition between exchange interactions of distinct character. A spin-reorientation transition in the *bc* plane was simultaneously observed for both magnetic sublattices, by either varying T across ~ 95 K or by application of small magnetic fields (~ 0.03 T at 100 K), indicating competing spin anisotropies. The magnetic structures at 10 K and 110 K are shown in Fig. 1(b). Supported by previously published band structure calculations[2], chemical bonding considerations[1], and resistivity studies[3], we invoked a coexistence of localized and itinerant Cr *3d* electrons, and, consequently, of Heisenberg superexchange (virtual-charge-transfer) and double exchange (real-charge-transfer) spin coupling mechanisms in $LaCrSb_3$.[4]

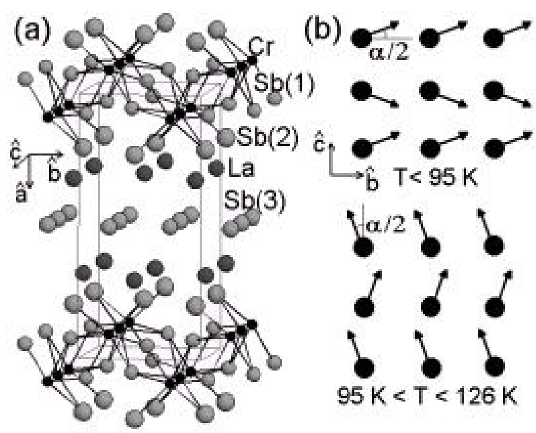

Fig. 1: (a) Crystal structure of $LaCrSb_3$. (b) Projection of the Cr sublattice onto the *bc* plane. The arrows represent the Cr magnetic structures in this compound below 95 K and between 95 K and 126 K. The angle between neighboring spins, 36(4)°, is the same within experimental error for both T-intervals.

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