RKKY-LIKE SPIN CORRELATIONS IN LiV_2O_4*

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We report on full 3-directional neutron polarization analysis studies of the magnetic correlations in LiV_2O_4. The data show the evolution of anti-ferromagnetic spin fluctuations at low temperatures ($T \leq 40\,$K) with a characteristic wave vector of $Q = 0.7\,\text{Å}^{-1}$. The measured magnetic cross section reveals a temperature induced cross-over from predominantly ferromagnetic ($T \geq 40\,$K) to RKKY-like (anti-ferromagnetic + ferromagnetic) spin correlations ($T \leq 40\,$K), as evidenced by reverse Monte Carlo simulations. These results are in agreement with the results of the magnetic relaxation rate on LiV_2O_4.

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1. Introduction

LiV_2O_4 is a narrow band transition-metal oxide that crystallizes in the cubic spinel-type structure [1] with the vanadium ions formally in a non-integral valence state $V^{+3.5}$. Due to the inherent geometrical frustration of the spinel structure magnetic order can easily be suppressed and indeed no sign of magnetic order could be observed for $T \geq 0.02$ K [2, 3]. Based on the results of specific heat, susceptibility and NMR measurements, Kondo *et al.*, proposed LiV_2O_4 to be the first example of a *d*-metal compound

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exhibiting heavy-fermion behavior [2]. Recent transport and specific heat measurements on single crystalline LiV_2O_4 revealed the formation of a heavy mass Fermi liquid below a characteristic temperature $T^* = 20$–$30\,K$ [4].

^{7}Li NMR results, similar to those presented by Kondo *et al.*, [2] were published by Fujiwara *et al.*, [5] but were interpreted in terms of spin fluctuations of a weak ferromagnetic metal. Quasi-elastic neutron scattering experiments showed a dramatic change of the magnetic response of LiV_2O_4 as a function of temperature [6]: at elevated temperatures $T \geq 40\,K$, the quasi-elastic line width and energy integrated cross section reveal a Q-dependence as expected for spin fluctuations of a weak ferromagnetic metal. Below $T \leq 40\,K$ additional anti-ferromagnetic correlations evolve as reflected in a pronounced intensity maximum and a corresponding line width minimum around $Q = 0.7Å^{-1}$. Here we report on a study of the diffuse magnetic scattering of LiV_2O_4 employing polarized neutrons.

2. Experimental results and discussion

The diffuse scattering of well characterized polycrystalline samples of LiV_2O_4 has been studied by full 3-directional polarization analysis on the spectrometer D7 at the Institute Laue Langevin (ILL), Grenoble. The raw data have been corrected for background, detector efficiency and incomplete polarization. The results of these measurements are summarized in Fig. 1. On the left-hand side of Fig. 1, the magnetic scattering cross-section is plotted as a function of momentum transfer Q that falls off monotonously upon increasing Q at elevated temperatures. However, on cooling a pronounced maximum of the scattering intensity evolves around a characteristic wave vector of $Q = 0.7\,Å^{-1}$. To extract the corresponding real-space spin correlations, the magnetic cross-section has been fitted employing reverse Monte Carlo simulation techniques [7]. The final result of the simulations is represented by the full lines in the left column of Fig. 1 and agrees excellently with the measured magnetic cross-sections. On the right-hand side of Fig. 1 the corresponding real-space magnetic correlations are shown. For $T = 80\,K$ and $40\,K$, only positive values of $< S_0 Si >$ are observed, indicative of ferromagnetic correlations in LiV_2O_4 at elevated temperatures. However, for $T = 4\,K$ and $10\,K$, the occurrence of negative values of $< S_0 Si >$ documents the evolution of additional anti-ferromagnetic correlations. The R-dependence of the spin correlations at low temperatures has been fitted according to a RKKY-type magnetic interaction. A characteristic property of the RKKY-interaction is the oscillatory behavior on distance, thus changing from ferromagnetic to anti-ferromagnetic coupling. The result of the fit for $T = 4\,K$ and $10\,K$ is shown as full lines on the right hand side of Fig. 1 and evidently can well describe the spatial variation of the spin correlations.

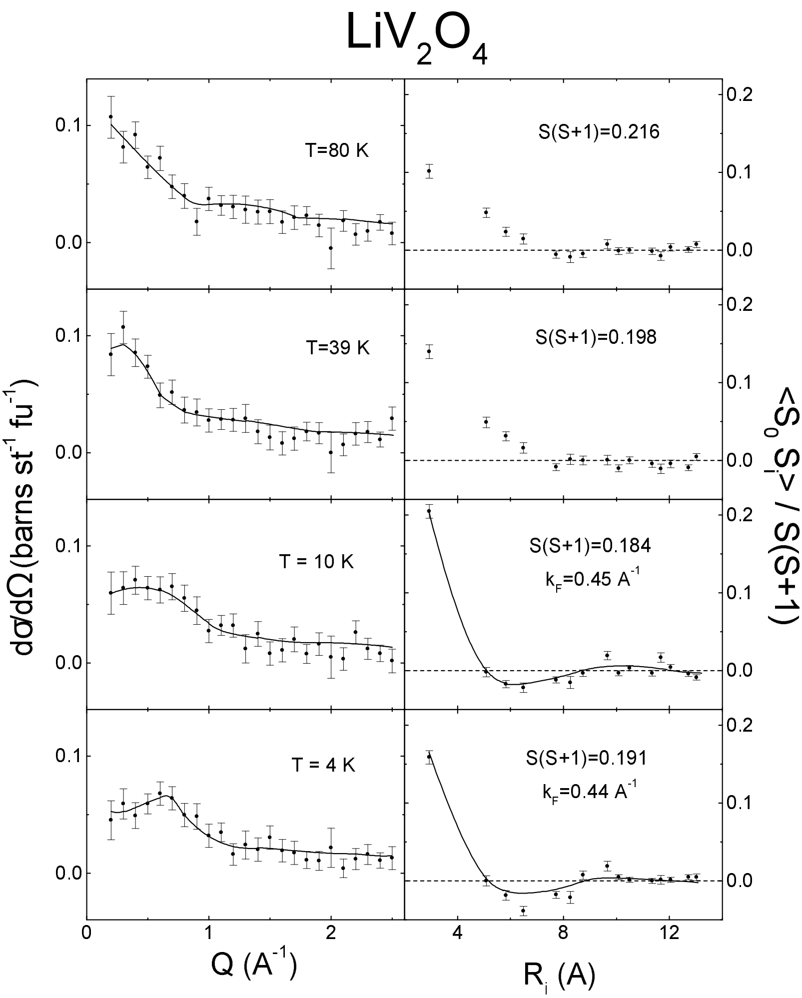

Fig. 1. Left-hand side: Q-dependence of the magnetic scattering cross section of LiV_2O_4 for different temperatures. The magnetic scattering cross section is given in absolute units. The full lines are the result of MC simulations (see text). Right-hand side: the corresponding real-space spin correlations as a function of the interatomic distance R are shown. At low temperatures, the spin correlations follow the characteristic RKKY-type relationship, as indicated by the full lines. Slight changes of $S(S+1)$ upon temperature are not significant.

It should be noted that the only adjustable parameter on fitting the RKKY-function is the Fermi wave vector k_{F} which remains practically constant. This is a further indication for the correctness of the fit. The present results are in agreement with measurements of the magnetic relaxation rate on LiV_2O_4 [6]. We conclude that anti-ferromagnetic spin fluctuations significantly contribute to the spectra at low temperatures. By contrast, above $T \geq 40\,K$, LiV_2O_4 reveals characteristics of a weak ferromagnetic metal.

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