Magnetic properties of the d-metal heavy-fermion system $Li_{1-x}Zn_xV_2O_4$

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LiV_2O_4 is the first d-metal heavy-fermion (HF) compound as determined by specific heat, susceptibility and NMR measurements [1], as well as by an anomalous temperature dependence of the lattice constants and the thermal expansion [2,3] which points towards of a strongly enhanced Grüneisen parameter. Recent μSR studies on LiV_2O_4 [4] indicated a close relationship to spin-glass behavior but without any static freezing-in. In contrast, for ZnV_2O_4, a structural phase transition at $T = 50$ K removes the geometrical frustration of the spinel structure, leading to an insulating, magnetically ordered ground state below $T = 40$ K [5]. The magnetic phase diagram of $Li_{1-x}Zn_xV_2O_4$ has been investigated by Ueda et al. [5]. For a large concentration regime $0.1 < x < 0.9$, the magnetic frustration and the alloying induced disorder leads to a spin-glass state below $T_f \approx 10$ K [5]. Increasing Zn concentration is accompanied by a linear increase of the lattice constants and the unit cell volume, respectively, thereby obeying Vegard's law. One therefore might interpret these results as a transition from a nonmagnetic HF state in LiV_2O_4 via an intermediate spin glass state as a precursor of long-range magnetic order to antiferromagnetic ZnV_2O_4. Traditionally, in strongly correlated f-electron systems, such a transition has been described within the framework of Doniach's phase diagram [7]. It is based on the competion of the RKKY interaction favoring magnetic order and the demagnetizing Kondo effect. Here we report on a systematic neutron scattering study on $Li_{1-x}Zn_xV_2O_4$ in order to establish its magnetic relaxation rate. For pure HF systems, one expects Lorentzian-shaped quasielastic lines with a residual line width for $T \rightarrow 0$ and a monotonous increase for increasing temperature. Samples of $Li_{1-x}Zn_xV_2O_4$ for $x = 0$, 0.05, 0.1, 0.2 and 0.3 were synthesized by a sintering technique of $LiVO_3$, VO, VO_2 and ZnO. X-ray diffraction revealed the nominal FCC spinel structure and no signs of spurious phases could be detected. The lattice constants were in agreement with the values of Ueda et al. [5]. The samples have been investigated by means of quasielastic

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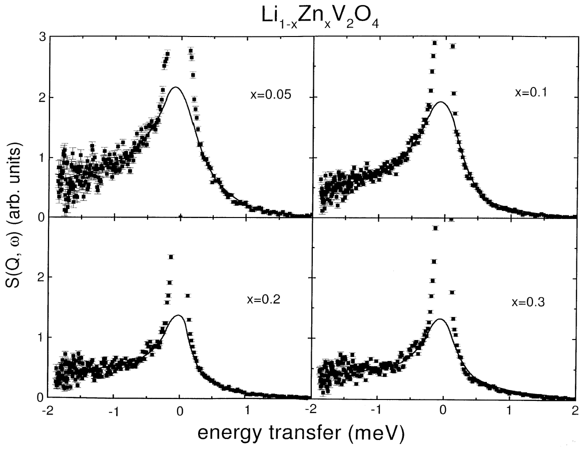

Fig. 1. Quasielastic scattering of $Li_{1-x}Zn_xV_2O_4$ for $x = 0.05$, 0.1, 0.2 and 0.3 at $T = 15$ K. The lines correspond to the Lorentzian fits of the data as described in the text.

neutron scattering for temperatures $1.5 \text{ K} < T < 300$ K. The experiments have been performed on the time-of-flight spectrometer IN6 at the ILL, Grenoble. The data were corrected in a standard way for detector efficiency and background signal. The resulting scattering contributions could be satisfactorily described by a single quasielastic Lorentzian, multiplied with the detailed balance factor and convoluted with the instrumental resolution. The magnetic relaxation rate of LiV_2O_4 revealed a residual quasielastic line width of 0.5 meV and a square root temperature dependence without any significant Q-dependence [6] at low temperatures, characteristic for the formation of a HF ground state. Between 40 and 80 K, the magnetic response changes drastically. At elevated temperatures, LiV_2O_4 displays a linear Q dependence of the line width, as it is expected in Fermi liquid theory and also predicted in spin-fluctuation theories of weak ferromagnetic metals [6,8]. For the Zn-doped samples, the counting statistics did not allow for a detailed analysis of the Q-dependence of the scattering signal. At least, a comparison of the intensities at high and low angles showed that these quasielastic scattering contributions decrease with increasing momentum transfer, thus indicating its magnetic origin. The Zn-doped compounds of $Li_{1-x}Zn_xV_2O_4$ for $x = 0.05, 0.1, 0.2, 0.3$ show a residual quasielastic line width (of approx. 1 meV) and a square root temperature dependence for all samples investigated. The residual line width slightly increases and the intensity at low temperatures strongly decreases with increasing Zn concentration. Fig. 1 shows the strong suppression of the quasielastic scattering upon increasing Zn concentration at low temperature. Such a behavior is also found in ^{7}Li NMR measurements [9]. These mutually consistent results indicate the freezing out of magnetic fluctuations due to the formation of a spin-glass state with increasing freezing temperature T_f for increasing Zn concentration [5]. As evident from Fig. 1, this suppression of magnetic fluctuations is accompanied by a broadening as reflected in a slight increase of the residual quasielastic line width which agrees remarkably well with T_f as determined by Ueda et al. [5]. Summarizing, these results essentially reflect the transition from a nonmagnetic HF system to a spin-glass state upon increasing alloying-induced disorder. This goes along with an increase of the T_f and a concomitant suppression of magnetic fluctuations.

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