Oral Presentation

Competition between heavy fermion behavior and magnetism in the d-metal system $Li_{1-x}Zn_xV_2O_4$

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LiV_2O_4 is the first example of a d-metal compound exhibiting heavy fermion (HF) behavior as first proposed by S. Kondo et al. [1] on the basis of specific heat, susceptibility and NMR measurements. An anomalous temperature dependence of the lattice constant and the thermal expansion [2,3] are significant of a strongly enhanced Grüneisen parameter, again pointing towards HF behavior. Recent μSR studies on LiV_2O_4 [4] indicated a close relationship to spin glass behavior but without any static freezing-in. The magnetic phase diagram of $Li_{1-x}Zn_xV_2O_4$ has been investigated by Ueda et al. [5]. For pure ZnV_2O_4, a structural phase transition at $T = 50$ K removes the geometrical frustration of the spinel structure, leading to a magnetically ordered ground state below $T = 40$ K [5]. For a large intermediate 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. One therefore might interpret these results as a transition from a nonmagnetic HF state in LiV_2O_4 via an intermediate spin glass state to antiferromagnetic order in ZnV_2O_4. Traditionally, in strongly correlated f-electron systems, such a transition has been described within the framework of Doniach's phase diagram [6]. 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 upon expanding the lattice by the substitution of Li by Zn.

Well characterized single-phase material of $Li_{1-x}Zn_xV_2O_4$ for $x = 0$, 0.05, 0.1, 0.2 and 0.3 has been investigated by means of quasielastic neutron scattering for temperatures $1.5\text{ K} < T < 300\text{ K}$. 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, respectively. The data 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 [7], characteristic for

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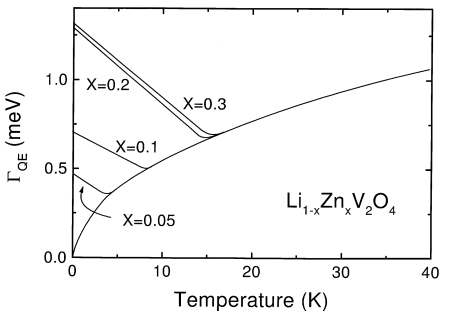

Fig. 1. Temperature dependence of the quasielastic line width of $Li_{1-x}Zn_xV_2O_4$ for $x = 0.05$, 0.1, 0.2 and 0.3.

the formation of a HF ground state. Between 40 and 80 K, the magnetic response changes dramatically. At elevated temperatures, LiV_2O_4 behaves like a weak ferromagnetic d-metal [7]. 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 results of the measurements on $Li_{1-x}Zn_xV_2O_4$ for $x = 0.05, 0.1, 0.2, 0.3$ can be summarized as follows: (i) There is a residual quasielastic line width followed by a square root temperature dependence for all samples investigated. (ii) The residual line width increases and the intensity at low temperatures strongly decreases with increasing Zn concentration. (iii) The temperature dependence of the intensity shows a maximum and $S(Q, \omega)$ (for fixed energy transfer) displays a significant change in slope around $T = 50$ K. Fig. 1 shows the temperature dependence of the quasielastic line for the different concentrations. The principal effect of replacing Li by Zn is twofold. First, the lattice expands which should give rise to a weakening of the hybridization. However, it should be noted that the magnetic vanadium ions are located in the center of oxygen octahedra. Therefore, the change of the vanadium–oxygen bonds may be significantly different from the overall lattice expansion upon Zn doping. Detailed structural investigations are needed to provide reliable atomic positions for band structure calculations. Second, the number of electrons is increased by the substitution of Li by Zn which should result in an increase of the Kondo interaction due to an increasing number of available screening electrons. These two effects compete with each other, giving rise to various available ground state configurations. Usually, the residual quasielastic line is taken as a measure of the Kondo temperature, Thus, the moderate increase of the line width may be interpreted as an increase of the Kondo temperature due to an increasing band filling. However, S. Kondo et al. [1] have determined a Kondo temperature of $T^* \approx 40$ K in pure LiV_2O_4. Around this temperature, a dramatic change of the magnetic response occurs in LiV_2O_4 [7]. Also for the Zn-doped compounds we observe a change of the temperature dependence of either the intensity or of $S(Q, \omega)$ for fixed energy transfer around $T \approx 40$ K. The strong decrease of the intensity when cooling down below $T = 40$ K may be an indication for the onset of strong Kondo compensation effects, thus pointing towards a characteristic temperature of $T^* \approx 40$ K, like in pure LiV_2O_4. Furthermore, the residual quasielastic line width of $Li_{1-x}Zn_xV_2O_4$ agrees remarkably well with the freezing temperature T_F as determined by Ueda et al. [5]. The low-temperature quasielastic response may thus be strongly related to inter-site fluctuations. The other main feature is the strong decrease (with a concomitantly broadening) of the quasielastic intensity upon increasing Zn concentration at low temperatures. Such a behavior is also found in ^{51}V NMR measurements [8]. These mutually consistent results are indicative for a vanishing magnetic moment and therefore is indicative of an increasing itinerancy of the d-electrons. To obtain a more detailed physical insight, improved counting statistics are necessary to allow for a detailed Q-analysis.

Acknowledgements

This work was supported by the BMBF under contracts No. 03-LO5AK1-5 (BEO, Verbund 4) and No. 13N6917 (EKM).

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