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Spin-glass behavior in $\text{Li}_{1-x}\text{Zn}_x\text{V}_2\text{O}_4$

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LiV_2O_4 is a transition-metal based heavy fermion compound close to magnetic order. Mixed crystals of $\text{Li}_{1-x}\text{Zn}_x\text{V}_2\text{O}_4$, $0 \leq x \leq 0.3$, were investigated by zero-field-cooled and field-cooled magnetization and by ac susceptibility measurements at different frequencies and applied external dc fields. The measurements indicate two characteristic temperatures, a freezing temperature T_f which is almost independent of the applied field and a temperature T_{irr} where strong irreversibilities occur and which strongly decreases on increasing fields. Comparison is made to heavy fermion spin glasses.

I. INTRODUCTION

LiV_2O_4 is one of the rare spinel-type compounds revealing metallic conductivity down to the lowest temperatures.¹ In addition, it is the d -electron derived heavy fermion (HF) system with the highest Sommerfeld coefficient of the heat capacity ($\gamma = 420$ mJ/mol K²) and an almost temperature-independent magnetic susceptibility at low temperatures.²⁻⁴ The heavy fermion behavior has been further corroborated by a highly enhanced Grüneisen parameter⁵ and a temperature and wave-vector dependence of the magnetic relaxation rate which is typically found in strongly correlated electron systems.⁶ Like most of the heavy fermion systems LiV_2O_4 is also close to magnetic order and spin fluctuations play an essential role. Consequently the anomalous metallic state in LiV_2O_4 has been explained in terms of Moriyas spin-fluctuation theory by Fujiwara, Yasuoka, and Ueda.^{7,8} However, no signs of magnetic order² were observed down to 20 mK. In HF systems magnetic order is suppressed via Kondo-compensation effects. In LiV_2O_4 in addition, geometrical frustration effects certainly play an important role. And indeed, low concentrations of Zn doping induce spin-glass order⁹ which then extends far into the regime of Zn-rich compounds.¹⁰ The magnetic as well as the electronic properties of LiV_2O_4 , including the origin of the HF behavior, have been explained in a series of band-structure calculations.¹¹⁻¹⁴

To investigate the properties of the spin-glass state in a highly frustrated as well as highly correlated electron system we performed systematic susceptibility and magnetization experiments at low Zn-doping levels, close to the pure compound. A detailed study of the structural and magnetic properties of $\text{Li}_{1-x}\text{Zn}_x\text{V}_2\text{O}_4$ has been performed by Ueda, Fujiwara, and Yasuoka.¹⁰ These authors report on the occurrence of a spin-glass state extending from $0.2 < x < 0.8$. From the cusps in dc susceptibility and from the splitting of the field-cooled and zero-field-cooled magnetization they determined freezing temperatures. However, the spin-glass transition close to $x=0$ seems to be of paramount interest: There exist old but very detailed predictions by Villain¹⁵ about the appearance of exotic frozen-in spin configurations in cubic

B -type spinel compounds like LiV_2O_4 . For the pure but highly frustrated lattice Villain¹⁵ predicts the formation of pairs of spins. On doping with nonmagnetic impurities at the A site a complicated phase diagram of disordered states shows up. In the case of substitution from Li by Zn we replace a nonmagnetic A ion by another one and Villain's phase diagrams are not really appropriate. However, a transition from the paired state ("cooperative paramagnet") to a canonical spin glass can be expected.

A report of heavy fermion spin-glass behavior in URh_2Ge_2 has been presented by Süllow *et al.*¹⁶ Here an explanation in terms of frustration effects produced by stacking faults in the crystallographic lattice has been provided. The physics of spin glasses found considerable attention two decades ago mainly focusing on classical dilute systems like CuMn or AuFe and the state of the art at that time is reported in detail in a review by Binder and Young.¹⁷ And despite the fact that a number of questions concerning the spin-glass physics remained open, e.g., ergodicity or symmetry breaking in external fields, divergence of relaxation times in real systems, etc., not much work has been performed during the last decade and, especially, not much work has been performed on pure but highly frustrated compounds with disordered ground states. Hence the main aim of the present work is twofold: (i) We investigate a spin-glass state which is very close to a coherent HF state and (ii) we present a detailed investigation of the freezing-in in a disordered magnetic system which reveals a series of unusual properties which will be interesting also from the pure standpoint of spin-glass physics.

II. EXPERIMENTAL DETAILS

Polycrystalline samples of $\text{Li}_{1-x}\text{Zn}_x\text{V}_2\text{O}_4$ ($x=0, 0.05, 0.1, 0.2, \text{ and } 0.3$) were prepared by sintering a mixture of powders of LiV_2O_3 , ZnV_2O_3 , and VO with a slight excess of LiV_2O_3 in order to compensate for Li evaporation. Platinum crucibles were used for reaction of the powders at 750 °C for 10 days. In x-ray-diffraction experiments we found the nominally pure fcc spinel structure. These samples

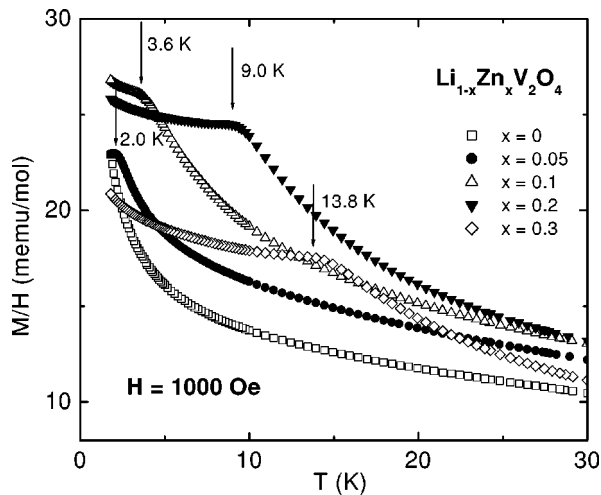


FIG. 1. Temperature-dependent magnetization divided by the applied field of 1000 Oe in $\text{Li}_{1-x}\text{Zn}_x\text{V}_2\text{O}_4$ between 1.9 and 30 K for $x=0, 0.05, 0.1, 0.2,$ and 0.3 and temperatures $1.9 \leq T \leq 30$ K. Arrows indicate the spin-glass temperatures T_f , as derived from χ'_{ac} .

were also used in recent neutron-scattering¹⁸ and NMR measurements.⁹ From electron paramagnetic resonance (EPR) experiments¹⁹ we estimate the number of defect spins to be well below 1% in all samples investigated.

The measurements were performed with a commercial superconducting quantum interference device (SQUID) magnetometer from Quantum Design. The magnetization measurements were made in an external field of 1000 Oe by cooling the sample in a temperature sweep mode. The zero-field-cooled (ZFC) and field-cooled (FC) magnetizations were measured by cooling the sample in zero applied field. Then the external field was set and the sample was heated continuously at a constant rate of 0.1 K/min while measuring to a temperature of 30 K well above the spin-glass temperature. Subsequently the FC data were taken by cooling the sample in nonzero field at the same rate. The ac susceptibility was measured in the same apparatus where an ac field of 4 Oe is applied with an additional copper coil and the magnetization is detected with a second-derivative coil connected inductively to the SQUID sensor.

III. RESULTS

A. Magnetization

Figure 1 shows the magnetization measured in an applied field of 1000 Oe of some $\text{Li}_{1-x}\text{Zn}_x\text{V}_2\text{O}_4$ samples between $x=0$ and $x=0.3$. While pure LiV_2O_4 shows a continuous increase towards low temperatures the magnetization levels off below a given temperature for $x>0$. The temperature of this anomaly increases almost linearly with increasing Zn concentration. While for $x=0.05$ the critical temperature approximately is 2.0 K it increases to 13.8 K for $x=0.3$. The lower value of the magnetization close to the spin-glass transition for $x=0.3$ is due to a different Curie-Weiss law²⁰ and the significantly higher freezing temperature. The leveling off of the magnetization is indicative of static spin freezing occurring in spin-glass systems. The arrows indicate the spin-glass freezing temperatures T_f , which were determined

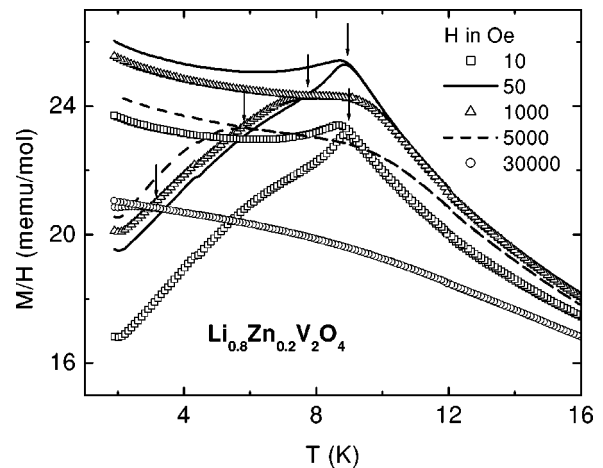


FIG. 2. Zero-field-cooled (lower curves) and field-cooled (upper curves) magnetization divided by the applied field H vs temperature for $\text{Li}_{0.8}\text{Zn}_{0.2}\text{V}_2\text{O}_4$ between 1.9 and 16 K for applied fields of 10, 50, 1000, 5000, and 30 000 Oe. Arrows indicate the temperature T_{irr} where the FC and ZFC curves split.

by measuring for each sample the real part of the ac susceptibility (see Fig. 5) which shows a peak at the freezing temperature. The freezing temperatures of 9 K for $x=0.2$ and 13.8 K for $x=0.3$ are enhanced compared to the results of Ueda, Fujiwara, and Yasuoka,¹⁰ where T_f of 5 K for $x=0.2$ and 9 K for $x=0.3$ have been determined from ZFC measurements. The discrepancies for $x=0.2$ and $x=0.3$ may result from differences in sample preparation, where, e.g., small differences in oxygen stoichiometry are known to produce considerable effects. Our pure sample also does not show the cusplike maximum like the purest samples of Kondo, Johnston, and Miller²¹ which, however, have been measured at higher external fields. In the following we give a representative survey of our experimental results focusing on $\text{Li}_{0.8}\text{Zn}_{0.2}\text{V}_2\text{O}_4$ ($T_f \approx 9.0$ K).

B. Zero-field-cooled and field-cooled magnetization

The ZFC and FC magnetization divided by the applied field H for $\text{Li}_{0.8}\text{Zn}_{0.2}\text{V}_2\text{O}_4$ is displayed in Fig. 2. The ZFC curves in low fields ($H=10$ and 50 Oe) reveal a cusp at the freezing temperature, followed by a broad shoulder close to 5 K on further decreasing temperatures. For this small fields the temperature of the cusp maximum coincides with the onset of the splitting of the ZFC and FC measurements which indicates the onset of irreversibilities (T_{irr}). T_{irr} , as indicated by arrows in Fig. 2, is shifted to lower temperatures with increasing external fields H . But at the same time the cusp maximum becomes smeared out and indicates a change from the Curie-like high-temperature paramagnetic regime to the low-temperature frozen-in state. The temperature of this crossover regime almost remains constant independent of the external field. The small anomaly in the ZFC curves around 4.2 K is due to slightly imperfect temperature ramps while heating through the boiling point of liquid ^4He . This signals that the ZFC magnetization is not in a thermodynamic equilibrium and the values are strongly dependent on the thermal and magnetic history, which is a common feature of canonical spin glasses. On the other hand, no such anomaly is seen in the FC measurements which are believed

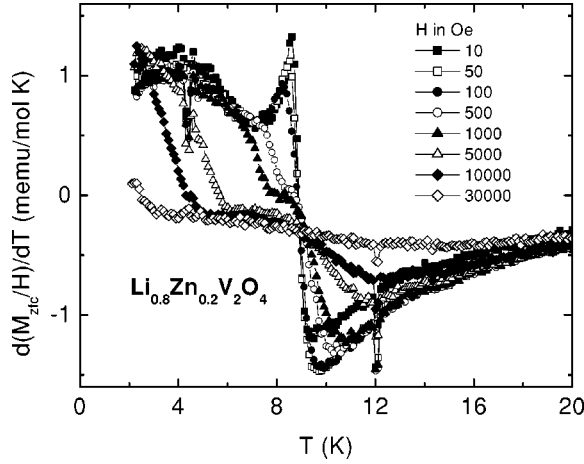


FIG. 3. Temperature derivative of the zero-field-cooled magnetizations divided by the applied field of $\text{Li}_{0.8}\text{Zn}_{0.2}\text{V}_2\text{O}_4$ for applied fields of 10, 50, 100, 500, 1000, 5000, 10 000, and 30 000 Oe vs temperature between 1.9 and 20 K.

to be in thermal equilibrium, where no irreversibilities take place. These measurements indicate that the behavior of $\text{Li}_{0.8}\text{Zn}_{0.2}\text{V}_2\text{O}_4$ is governed by two temperature scales. The temperature of the spin-glass transition T_f indicated by an anomaly in the temperature dependence of the ZFC magnetization and the temperature where irreversibilities occur (T_{irr}), which is determined by the splitting of FC and ZFC curves. While the first seems to be almost independent of applied field, the second decreases from 9 K for $H=10$ Oe which is also the temperature where the maximum in the ac susceptibility occurs, to 3.2 K in 30 kOe.

This behavior is even more clearly demonstrated in Fig. 3. Here the temperature derivative of the ZFC magnetization is plotted vs temperature. We find a resonance-like anomaly at $T \approx 9$ K indicating the onset of freezing, which at low fields is followed by a broad cusp towards lower temperatures. With increasing external fields the well defined resonance becomes considerably smeared out and is followed by a well defined increase of $d(M/H)/dT$ which indicates the onset of irreversibilities. E.g., in fields of 5000 Oe the upturn is seen just below 6 K, which is also the temperature where the FC and ZFC lines split (see Fig. 2). The same is also true for 10 and 30 kOe. It is important to note that all these curves cross at one single point at the spin-glass temperature and reach approximately the high-temperature value of the temperature derivative of the magnetization. The regime in $d(M/H)/dT$ around these inflection points, from the minimum where the first deviations from the paramagnetic behavior occur, to the strong increase which signals the onset of strong irreversibilities broadens considerably on increasing external fields. E.g., for fields of 1000 Oe it extends from 7.5 to 11 K. Figure 4 shows the magnetic-field dependence of these two characteristic temperatures, where for the high-temperature transition which indicates T_f we took the strong broadening into account. We believe that this observation provides experimental evidence for the occurrence of two characteristic temperature scales, the onset of freezing (T_f) and the temperature of the onset of strong irreversibilities (T_{irr}). These two temperature scales coincide in zero field but reveal a significantly different field dependence, following roughly the Almeida-Thouless²² (AT) [$T_{irr}(H) \propto (1 - T/T_f)^{3/2}$], re-

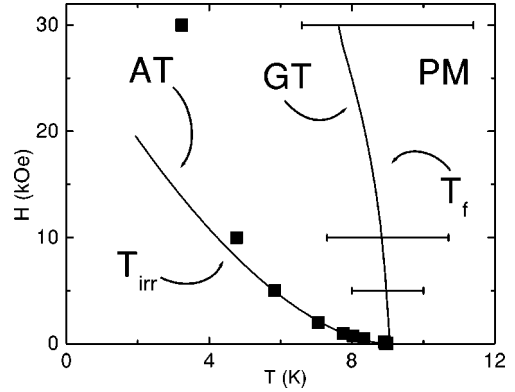


FIG. 4. Magnetic-field dependent phase diagram of $\text{Li}_{0.8}\text{Zn}_{0.2}\text{V}_2\text{O}_4$, where the lines are the AT and GT lines and PM the paramagnetic phase. T_{irr} and T_f are used as defined in the text.

spectively, the Gabay-Toulouse²³ (GT) predictions [$T_f(H) \propto (1 - T/T_f)^{1/2}$]. One line indicates the irreversibility or Almeida-Thouless (AT) line, which describes $T_{irr}(H)$ at fields up to 5000 Oe but fails for higher fields. The field independent line at $T_f(H=0)$ possibly is related to the Gabay-Toulouse (GT) line expected for infinite-range vector spin glasses. We would like to comment that from very similar measurements in AgMn, Chamberlin *et al.*²⁴ determined also different characteristic temperatures of the spin-glass transition. In this detailed work the authors reported on a series of characteristic temperatures in external fields. But we would like to point out that the appearance of two characteristic temperatures, as has been observed in the highly frustrated compounds under investigation, has been predicted by Villain¹⁵ for doped cubic spinels. Here the two temperatures indicate the freezing of a ferromagnetic and of a spin-glass component. However, further detailed experimental and theoretical investigations will be necessary to elucidate this question. Specifically samples with diluted B lattices should be investigated.

C. ac susceptibility

Figure 5 shows the ac susceptibility results for $x=0.05, 0.1, 0.2, \text{ and } 0.3$. From these measurements the freezing temperatures T_f (indicated in Fig. 1) have been determined. For all samples a frequency-dependent maximum in the susceptibility could be observed. The results of the ac susceptibility

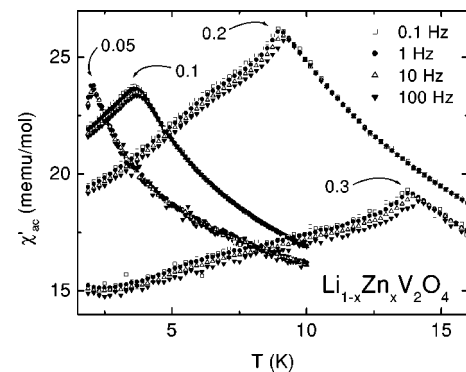


FIG. 5. Temperature dependence of the ac susceptibility of $\text{Li}_{1-x}\text{Zn}_x\text{V}_2\text{O}_4$ for $x=0.05, 0.1, 0.2, 0.3$ at different frequencies.

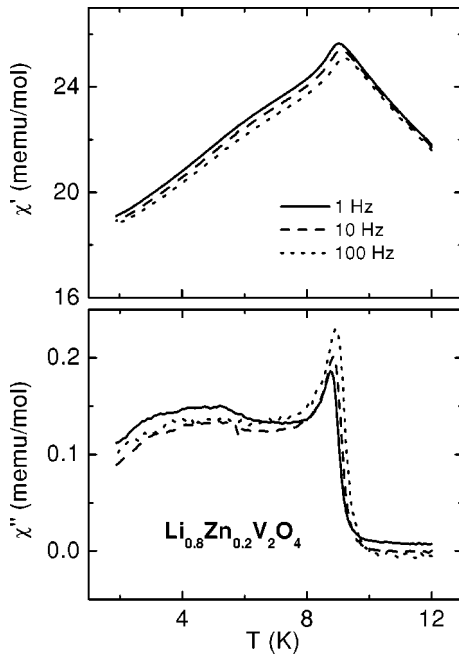


FIG. 6. Real (upper panel) and imaginary (lower panel) part of the ac susceptibility of $\text{Li}_{0.8}\text{Zn}_{0.2}\text{V}_2\text{O}_4$ at frequencies of 1, 10, and 100 Hz vs temperature between 1.9 and 12 K.

measurements of $\text{Li}_{0.8}\text{Zn}_{0.2}\text{V}_2\text{O}_4$ are shown in more detail in the following. Figure 6 shows the real (upper panel) and imaginary part (lower panel) of the ac susceptibility at three different measuring frequencies as function of temperature. χ' reveals a cusp at T_f followed by a broad shoulder at lower temperatures, while in χ'' a narrow peak at 9 K and a broad peak at around 5 K is visible. The peak maxima in the real and imaginary part move towards higher temperatures on increasing frequencies of the applied ac field from 1 to 100 Hz. From the temperature shift of the maxima in the imaginary part we derived a value $\Delta T_f/[T_f(\Delta \log(\omega))] = 0.009$, which is a typical value for prototypical d -metal spin glasses. The corresponding characteristic temperature shifts are 0.005 for CuMn and 0.010 for AuFe (see Mydosh,²⁶ p. 67). The real part of the ac susceptibility resembles almost the ZFC curves at low applied fields (see Fig. 2). With increasing frequency the absolute value of the maximum is reduced and also the values below T_f are lower for higher frequencies. At T_f the imaginary part of the susceptibility χ'' is enhanced for higher frequencies, while for the peak at lower temperatures around 5 K no clear dependence on frequency could be seen. It is interesting to note that the shape of the temperature dependence of the imaginary part is similar to the temperature dependent derivative of the ZFC curves for low fields as shown in Fig. 3. This is surprising because it is the common belief that the ZFC curve is irreversible while the ac susceptibility is fully reversible.

In Fig. 7 we show the temperature dependence of real (upper panel) and imaginary part (lower panel) of the ac susceptibility at 1 Hz in different external dc fields. In the real part the peak broadens but remains almost constant in temperature for applied fields of 100 and 500 Oe. For $H = 10$ kOe this maximum is significantly shifted to lower temperatures. This behavior is qualitatively the same as observed in the ZFC curves at different applied fields (see Fig.

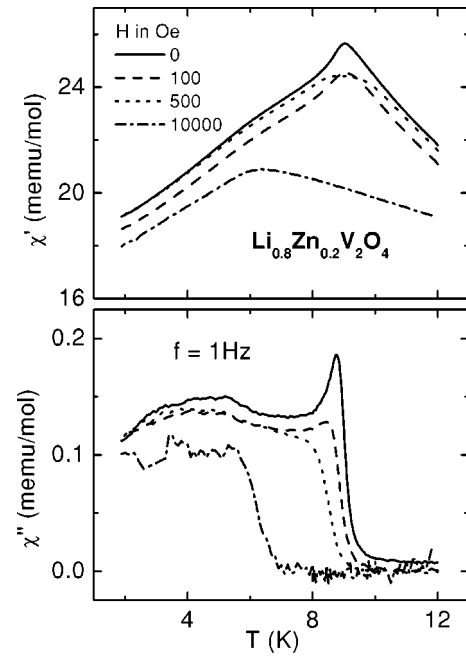


FIG. 7. Real (upper panel) and imaginary (lower panel) part of the ac susceptibility of $\text{Li}_{0.8}\text{Zn}_{0.2}\text{V}_2\text{O}_4$ at 1 Hz for applied dc fields of 0, 100, 500, and 10 000 Oe vs temperature between 1.9 and 12 K.

2). In the imaginary part the peak is suppressed with increasing dc fields and also resembles the shape of the temperature derivative of the ZFC curves at different fields for $T < T_f$. Above the ordering temperature χ'' approaches zero. The broad peak around 5 K is independent of the applied field as long as T_{irr} is high enough and in an applied field of 10 000 Oe the imaginary part is zero above the irreversibility line.

IV. DISCUSSIONS AND CONCLUSIONS

At first we briefly want to summarize the results of the ZFC and FC magnetization measurements which provide experimental evidence for two transition temperatures. For small applied fields a broad hump evolves at about 5 K with is followed by a peak at the freezing temperature of $T_f = 9$ K (see Fig. 2). This is also the temperature where the ZFC and FC curves deviate from each other for small H . The irreversibility temperature T_{irr} is shifted to lower temperatures with increasing fields, while T_f seems to be almost independent of field. This behavior is even more clearly seen in Fig. 3 where the temperature derivative of the ZFC curves are plotted. All curves seem to have one single crossing point, where the ZFC curves have their inflection point.

The ZFC curves resemble the behavior of the ac susceptibilities and behave like the real part of the ac susceptibilities even in applied fields (see Figs. 6 and 7) where the shape changes significantly, e.g., the sharp peak at T_f is smeared out for increasing fields. The comparison holds even for higher fields where T_{irr} is strongly shifted to low temperatures. On the other hand, the temperature derivative of the ZFC curves has the same shape as χ'' which describes the loss and irreversibilities or magnetic hysteresis in the sample. This strong interconnection holds almost independently of

the strength of the external field H . At first sight this relation is surprising, because the ZFC magnetization is measured not in thermal equilibrium and the thermal and magnetic history play an important role, while the ac susceptibility is independent of sample history within experimental resolution. But handwaving one can find a quite natural explanation. The ac susceptibility couples only to the free spins and not to spins that are frozen-in on the timescale of the applied ac field. On the other hand, the ZFC magnetization measures the temperature dependent melting of the spins. Therefore the temperature derivative of the ZFC curve provides a measure of the number of spins, which melt at a given temperature, that also couple to the ac susceptibility. From this argument it is clear that the temperature derivative of χ' should roughly resemble the temperature dependence of χ'' and this behavior indeed is fulfilled independently of the applied dc field.

The appearance of two characteristic temperatures in spin-glass systems has not often been reported so far. Two characteristic temperatures may occur in LiV_2O_4 because it is a highly frustrated magnet and a complex phase diagram has been proposed by Villain¹⁵ for similar compounds. At the phase boundaries the subsequent freezing of the ferromagnetic and the spin-glass component has been proposed. Concerning canonical spin glasses a systematic investigation of dc magnetization and an analysis including derivatives of FC and ZFC curves has been published by Chamberlin *et al.*²⁴ for AgMn and also these authors report on a series of characteristic temperatures. Theoretical predictions exist that in vector spin glasses in external fields where symmetry breaking appears along the GT line and strong irreversibilities occur in a crossover regime at the AT line. This, of course, is highly speculative as on no line were we able to prove a divergence of the corresponding relaxation times.

Concerning the frequency dependence of the ac susceptibility as shown in Fig. 6, we believe that the sharp anomaly around T_f bears the similarities of the logarithmic frequency dependence as predicted in the framework of the droplet model by Fisher and Huse,²⁵ while the susceptibility χ'' at lower temperatures seems to indicate a relaxational behavior, characterized by maxima in the frequency dependence of the imaginary part of the ac susceptibility which shift to lower frequencies as temperature decreases. However, the time window definitely is not wide enough to warrant a conclusive analysis.

From the apparent freezing temperatures in low fields and measured at low frequencies (Figs. 2, 3, and 5) we can construct an (x, T) phase diagram. We find an almost linear increase of the freezing temperature, roughly following $T_f = 44x(\text{K})$. This linear behavior of the freezing temperature

on doping usually is observed in metallic spin glasses²⁶ at low doping levels. In the case of LiV_2O_4 doped with Zn this behavior is rather astonishing. We start from the highly frustrated compound and magnetic short-range order should evolve already at the lowest doping levels. It is interesting to note that the freezing temperatures level off beyond Zn concentrations of $x = 0.3$. This may indicate a transition between different types of spin glasses,¹⁵ but it may also be due to the metal-to-insulator transition which is expected close to this concentration. For future work a detailed comparison of the freezing dynamics of $x = 0.4$ compared to that of $x = 0.2$ would be especially meaningful. In the spirit of Villain's¹⁵ work it also seems interesting to study LiV_2O_4 doped with a nonmagnetic impurity at the V site and doped with a magnetic impurity at the A site.

Finally we would like to make comparison with the heavy fermion spin glass URh_2Ge_2 . We would like to recall that LiV_2O_4 reveals a Sommerfeld coefficient $\gamma = 420 \text{ mJ/mol K}^2$, which becomes continuously suppressed on Zn doping. But γ still amounts to 107 mJ/mol K^2 in $\text{Li}_{0.7}\text{Zn}_{0.3}\text{V}_2\text{O}_4$.²⁰ In URh_2Ge_2 $\gamma = 420 \text{ mJ/mol K}^2$ and also in this compound the ac susceptibility and the FC and ZFC magnetizations reveal the characteristics of canonical spin glasses. The frequency shift of T_f was found to be of the order of 0.025 a value which seems to be enhanced when compared to canonical spin glasses and also slightly seems to be enhanced when compared to the value of 0.009 as observed in doped LiV_2O_4 . In URh_2Ge_2 the frustration effects have been explained assuming crystallographic disorder on an atomic scale while in LiV_2O_4 inherent frustration effects drive the spin-glass phenomena. We would like to point out that similar spin-glass effects have been reported in PrAu_2Ge_2 ,²⁷ another frustrated magnet. In this compound magnetic order is induced on substituting Ge by Si. Here the frequency shift is about 0.016. Also PrAu_2Ge_2 reveals a highly enhanced Sommerfeld coefficient of the specific heat. We conclude that Zn-doped LiV_2O_4 is closer to a canonical d metal than to the heavy fermion spin glasses. Further experiments on doped compounds close to LiV_2O_4 are highly needed to elucidate the unusual ground-state properties.

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