Spin fluctuations probed by NMR in paramagnetic spinel LiV$_2$O$_4$: a self-consistent renormalization theory

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Abstract

Low-frequency spin fluctuation dynamics in paramagnetic spinel LiV$_2$O$_4$, a rare 3d-electron heavy-fermion system, is investigated. A parametrized self-consistent renormalization (SCR) theory of the dominant AFM spin fluctuations is developed and applied to describe temperature and pressure dependences of the low-$T$ nuclear spin–lattice relaxation rate $1/T_1$ in this material. The experimental data for $1/T_1$ available down to $\sim$1 K are well reproduced by the SCR theory, showing the development of AFM spin fluctuations as the paramagnetic metal approaches a magnetic instability under the applied pressure. The low-$T$ upturn of $1/T_1$ detected below 0.6 K under the highest applied pressure of 4.74 GPa is explained as the nuclear spin relaxation effect due to the spin freezing of magnetic defects unavoidably present in the measured sample of LiV$_2$O$_4$.

1. Introduction

The metallic spinel LiV$_2$O$_4$ has attracted much attention since a heavy-fermion behaviour in this material was reported [1–3]. Despite continuous activity in recent years, there is currently no consensus on the mechanism for formation of heavy-fermion quasiparticles in LiV$_2$O$_4$, and the issue is still under debate [4, 5].

At low temperatures, $T < 30$ K, the spin system of LiV$_2$O$_4$ exhibits pronounced short-range antiferromagnetic (AFM) correlations [6–8], but no long-range magnetic ordering was detected at any measured temperatures. The geometrical frustration of the pyrochlore lattice of vanadium ions (in the mixed valence state V$^{3.5+}$) is likely to be a crucial aspect of the problem. The frustration may suppress at any $T$ a long-range ordering of strongly correlated itinerant electrons, but instead, the system is placed near to a magnetic instability. The emergence of largely degenerate low-lying spin excitations in the ground state of LiV$_2$O$_4$ is expected to be responsible for low-$T$ properties of this material, including its heavy-fermion behaviour. This appealing picture has been developed in detail in previous work [9, 10]. It can be examined by considering experimental results obtained by different techniques, like the nuclear magnetic resonance (NMR) and the inelastic neutron scattering (INS), probing low-frequency spin fluctuations.

The $^7$Li-NMR studies of the spin fluctuation dynamics in LiV$_2$O$_4$ were reported in a series of papers [11–15]. For high temperatures, $T > 60$ K, the NMR relaxation data have been successfully explained [12] in terms of a V local moment formalism, while in the low-$T$ region, $T < 30$ K, an approach based on the itinerant and strongly correlated electrons in the paramagnetic metal LiV$_2$O$_4$ is more appropriate. Low-$T$ measurements down to $\sim$1 K on samples of high purity under ambient pressure reveal a nearly constant value of the Knight shift $K$ and linear $T$ dependence of the nuclear spin–lattice relaxation rate $1/T_1$ as for normal metals, but with a very high value of $(1/T_1T)^{-1}$. An estimate [12] of the Korringa relation $K^2T_1T/S = R$, where $S = h\gamma_n^2/4\pi k_B\gamma_e^2$, with $\gamma_e$ and $\gamma_n$ being the electronic and nuclear gyromagnetic ratios respectively, and $R \approx 0.5$ is less than unity, indicates [16, 17] that presumably AFM fluctuations are dominant in the spin–lattice relaxation at low $T$.

The NMR measurements on high purity samples of LiV$_2$O$_4$ under pressure up to $\approx$5 GPa were also reported by Fujiiwara et al [18, 19]. For $T < 10$ K, the value of $(1/T_1T)^{-1}$ which gives information on the $q$ averaged dynamical spin susceptibility becomes larger on applying higher pressure and...
the temperature renormalization of the low-ω fluctuations was proved \[10\] to be a helpful tool in explaining freezing of magnetic defects below \(T\). Because of a lack of the separability of the relaxation effects dynamic spin susceptibility the self-consistent renormalization (SCR) theory \[21\] offers a phenomenological description for spin homogeneous spin fluctuations inherent to magnetically pure dynamics in LiV\(_2\)O\(_4\) derived from INS measurements \[6–8\].

Such a behaviour was proved \[15\] to originate from the spin freezing of magnetic defects below \(T_{\text{peak}}\). With decreasing \(n_{\text{defect}}\), the peak position of \(1/T_{1}\) is apparently shifted to lower \(T\). Remarkably, a proper model analysis of the low-\(T\) NMR data obtained in powder samples of LiV\(_2\)O\(_4\) with varying \(n_{\text{defect}} < 1\) mol\% has shown \[15\] that relaxation effects due to inhomogeneously distributed magnetic defects and homogeneous spin fluctuations inherent to magnetically pure LiV\(_2\)O\(_4\) are separable and thus can be examined independently. It is worth noting that in a single crystal of LiV\(_2\)O\(_4\) containing a small number of magnetic impurities or crystal defects a somewhat different behaviour was observed \[15\], possibly because of a lack of the separability of the relaxation effects mentioned.

A scenario explaining the considerable influence of a weak disorder on the low-ω spin dynamics detected by the low-\(T\) NMR measurements on LiV\(_2\)O\(_4\) was proposed by Johnston et al \[14\], following a more general consideration developed in \[20\]. As already noted, a critical aspect of the problem is the emergence in the ground state of pure LiV\(_2\)O\(_4\) of a large number of low-lying spin excitations, implying a proximity of the system to a magnetic instability. Inhomogeneously distributed magnetic defects may locally lift the degeneracy of low-lying spin excitations and cause their partial condensation, thus giving rise to a strong change of spin dynamics at sufficiently low temperatures. A large degeneracy of strongly enhanced and slow spin fluctuations in pure LiV\(_2\)O\(_4\) was confirmed by a combined analysis of the low-temperature INS data \[6–8\] and the complementary calculations \[9\] of the dynamic spin susceptibility \(\chi(q,\omega)\). In these calculations, performed first at \(T = 0\), the actual electronic band structure of LiV\(_2\)O\(_4\) obtained in the local-density approximation is used and effects of strong electron correlations are treated in the random phase approximation. As an extension for finite \(T\), the self-consistent renormalization (SCR) theory \[21\] of spin fluctuations was proved \[10\] to be a helpful tool in explaining the temperature renormalization of the low-ω spin fluctuation dynamics in LiV\(_2\)O\(_4\) derived from INS measurements \[6–8\].

In the present study, the parametrized SCR theory is applied to elucidate the main features of the temperature and pressure dependences of the spin–lattice relaxation rate observed in the low-\(T\) NMR measurements on LiV\(_2\)O\(_4\). As known \[21\], the SCR theory offers a phenomenological description for spin fluctuations in nearly ferro- or antiferromagnetic itinerant electron systems by taking into account effects of mode–mode coupling between spin fluctuations either at \(q = 0\) or \(q \neq 0\), respectively; the latter case is applicable to the paramagnetic spinel LiV\(_2\)O\(_4\). For details we refer to our recent work \[10\], where the basic equation of the SCR theory is solved numerically and the results are compared with INS data for LiV\(_2\)O\(_4\). There, the values of empirical parameters entering the SCR theory are estimated to provide the best overall coincidence between the theory and INS experiment.

The outline of the paper is as follows. In section 2, an expression for the spin–lattice relaxation rate \(1/T_{1}\) is derived in terms of the SCR theory and, first, an evolution of \((T/T_{1})^{-1}\) down to \(\sim 1\) K with increasing pressure is examined. Next, since the temperature behaviour of \((T/T_{1})^{-1}\) down to much lower temperature \(\sim 60\) mK is available only at the highest applied pressure of 4.74 GPa, these particular data deserve special attention. We argue, contrary to what Fujiwara et al suggested \[19\], that the upturn of \((T/T_{1})^{-1}\) detected below 0.6 K is not entirely due to homogeneous critical AFM spin fluctuations, but more likely is a signature of an additional nuclear spin relaxation mechanism, probably due to the spin freezing of magnetic defects. A summary and concluding remarks can be found in section 3.

2. SCR theory for relaxation rate \(1/T_{1}\) in LiV\(_2\)O\(_4\)

2.1. Background

The nuclear spin–lattice relaxation rate due to electronic spin fluctuations is generally given by

\[
\frac{1}{T_{1}} = \frac{2\gamma_{n}^{2}k_{B}T}{Ng^{2}\mu_{B}^{2}} \sum_{\mathbf{q}} |A_{q}|^{2} \text{Im} \chi(q,\omega_{n}),
\]

where \(A_{q}\) is a \(q\) dependent effective hyperfine coupling; in our calculations, the resonance frequency \(\omega_{n}\) will be taken in the limit \(\omega_{n} \rightarrow 0\). In (1), the \(q\) summation is over the Brillouin zone (BZ) of the fcc lattice inherent to the pyrochlore lattice of V atoms in the spinel structure LiV\(_2\)O\(_4\); \(\chi(q,\omega)\) is the dynamic spin susceptibility calculated per primitive cell (four V atoms) in units of \((g\mu_{B})^{2}\). Calculations suggest \[9\] a rather peculiar \(q\)-dependence of \(\chi(q,\omega)\). The resulting model for a distribution in \(q\) space of dominant spin fluctuations was checked \[10\] to provide a firm ground to describe the low-\(T\) INS measurements for LiV\(_2\)O\(_4\). Main features of the model are discussed and used below to calculate \(1/T_{1}\).

In the low-\(T\) limit, the paramagnetic state of LiV\(_2\)O\(_4\) is characterized by strongly enhanced and slow spin fluctuations occupying a large region in \(q\) space around the surface of a mean radius \(|q| \simeq Q_{c} \simeq 0.6\) Å\(^{-1}\), called the critical surface \[9\] (see figure 1). These strongly degenerate low-ω AFM spin fluctuations dominate over those at smaller \(q\). This rather peculiar distribution in \(q\) space of the dominant AFM spin fluctuations is a consequence of geometrical frustration of the pyrochlore lattice of V atoms, which can be traced back \[9\] to the underlying electronic band structure and the many-sheet Fermi surface of the metallic spinel LiV\(_2\)O\(_4\). With increasing \(T\) (up to 60 K), the AFM fluctuations get suppressed while...
those at the BZ centre remain nearly $T$ independent. As noted in [10], the observed [6–8] warming shift of the low-$\omega$ integrated INS intensity (from $|q|$ $\approx$ $Q_c$ at $T \to 0$ toward low $q$ values at higher temperatures) does not require any significant temperature renormalization of $\chi(q, \omega)$ at small $q$.

2.2. 1/$T_1 T$ obtained from SCR theory

For the pure LiV$_2$O$_4$, two main contributions to the spin–lattice relaxation rate can be written as

$$\frac{1}{T_1 T} = \left( \frac{1}{T_1 T} \right)_{q \to 0} + \left( \frac{1}{T_1 T} \right)_{q \to Q_c}.$$  

As discussed later, at $T \to 0$, the second contribution from AFM spin fluctuations at $|q| \sim Q_c$ is larger than the first one coming from the small $q$ spin fluctuations. For finite but low $T$, because of a comparatively small variation with both temperature and pressure of the Knight shift, and hence, of the intrinsic uniform susceptibility $\chi(q = 0, \omega = 0)$, one expects for the low-$\omega$ and small-$q$ spin fluctuations much weaker temperature and pressure dependences than those observed [6–8] for AFM spin fluctuations. Therefore, in the subsequent analysis the contribution (1/$T_1 T$)$_{q \to 0}$ is assumed to be a constant and treated below as an adjustable parameter of the fit procedure. In this approximation, we relate the experimentally observed temperature and pressure dependence of 1/$T_1 T$ merely to that of dominant AFM fluctuations around the $Q_c$ surface.

Let us choose a wavevector $Q_c$ with the end point lying on the $Q_c$ surface, and consider its vicinity, $q' = Q_c + q$ ($(|q| \ll |Q_c|$), as depicted in figure 1. The inverse dynamic spin susceptibility can now be expanded as

$$\frac{1}{\chi(Q_c + q, \omega; T)} = \frac{1}{\chi(Q_c; T)} + A(q^1)^2 + B(q^\perp)^2 - iC\omega,$$  

where $q^1$ and $q^\perp$ are components of $q$ parallel and perpendicular to the normal $Q_c/|Q_c|$ to the $Q_c$ surface. Since the dynamic spin susceptibility is considered around $q = 0$, only the leading term of $C$ independent of $q$ is taken in the expansion (3). This allows us to avoid an increase of the number of parameters in describing the dynamic spin susceptibility.

In (3), $A, B$ and $C$ are empirical parameters that, together with $\chi(Q_c; T = 0)$, can be estimated [10] from INS and magnetic measurement data. The parameters $A, B$ and $C$ are usually taken to be $T$ independent in the low-$T$ region, where the SCR theory works well. Strong anisotropy in $q$ space of AFM spin fluctuations, i.e., $b = B/A \ll 1$, is assumed and verified [10]. It is also reasonable using a spherical approximation to the $Q_c$ surface, implying that $\chi(Q_c; T)$ does not depend on the direction of $Q_c$, i.e., $\chi(Q_c; T) = \chi(Q_c; T)$.

To be close to the standard notation of the SCR theory [21], we introduce, instead of $A$ and $C$, the following parameters (here, $h = k_B = g\mu_B = 1$):

$$T_A = \frac{A q_B^2}{2}, \quad T_0 = \frac{A q_B^2}{2\pi C},$$  

where $q_B$ is the effective radius of the BZ boundary given in terms of the lattice primitive cell volume $A$ as $q_B = (\pi^2/V_0)^{1/3}$. Next, the reduced inverse susceptibility at $|q| = Q_c$ is defined as

$$y_Q(T) = \frac{1}{T_A \chi(Q_c; T)}.$$  

In this notation, one obtains for $\text{Im} \chi(|q|, \omega_n)/\omega_n$ in the limit $\omega_n \to 0$ and up to a constant factor the following expression:

$$\text{Im} \chi(Q_c + q, \omega_n; T) \sim \frac{1}{T_0 T_A} \left[ \frac{y_Q(T) + (q^1/Q_c)^2 + b (q^\perp/Q_c)^2}{y_Q(T) + (q^1/Q_c)^2 + b (q^\perp/Q_c)^2} \right].$$  

When performing the integration over $q$ in (1), two dimensionless cutoff parameters, $x_c = (q_c^\perp/Q_c)^2$ and $z_c = (q_c^1/Q_c)$, are introduced. For the former, the requirement $x_c < 1$ is sufficient. As the latter cutoff we take $z_c \approx 1/2$, which distinguishes the region of the dominant AFM spin fluctuations from that near the BZ centre. Near the $Q_c$ surface, the hyperfine coupling is assumed to be a constant $A_{Q_c}$. Then the resulting expression for (1/$T_1 T$)$_{q \to Q_c}$ reads as

$$\left( \frac{1}{T_1 T} \right)_{Q_c} = \frac{3\gamma^2 h! A_{Q_c}^2}{\pi k_B T_0 T_A} \left( \frac{Q_c}{q_B} \right)^2 \times \left[ \frac{1}{x_c} \left( \frac{1}{\sqrt{y_Q(T)}} \tan^{-1} \frac{z_c}{\sqrt{y_Q(T)}} \right) - \frac{1}{\sqrt{y_Q(T) + x_c}} \tan^{-1} \frac{z_c}{\sqrt{y_Q(T) + x_c}} \right],$$  

including again the proper-dimensional constants.

2.3. Basic equation of SCR theory and empirical parameters

The reduced inverse susceptibility $y_Q(t)$, where $t = T/T_0$, obeys the following integral equation [10]:

$$y_Q(t) = y_Q(0) + g \int_0^t \frac{d\tau \phi(y_Q(t) + z^2)/t - \phi(y_Q(t) + z^2 + b x_c)/t)}{b x_c/t},$$  

Figure 1. A cross-section in $q$ space of the critical $Q_c$ surface ($|Q_c| \approx 0.6 \ \text{Å}^{-1}$) is shown schematically by the solid line. Strongly enhanced antiferromagnetic spin fluctuations dominating low-$T$ properties of LiV$_2$O$_4$ are located in close vicinity (of width $\Delta q \lesssim 0.2 \ \text{Å}^{-1}$) to the $Q_c$ surface. The meaning of decomposition $q' = Q_c + q_1 + q_2$ for an arbitrary wavevector $q$ near the $Q_c$ surface is explained in the text.
with
\[ \phi(u) = \ln \Gamma(u) - (u - 1) \ln u + u - \frac{1}{2} \ln 2\pi, \]
where \( \Gamma(u) \) is the gamma function.

The present SCR theory includes a set of five parameters which are now denoted as \( y_Q(0) \), \( T_A \), \( T_0 \), \( g_Q \) and \( b_{x_c} \). The parameters \( T_A \) and \( T_0 \) characterize, at \( T \rightarrow 0 \), the momentum and frequency spread of the dominant AFM spin fluctuations, \( g_Q \) is the effective mode–mode coupling constant and \( b_{x_c} \) describes a large anisotropy of the spin fluctuation dispersion (3) in \( q \)-space. From a fit to INS data, we obtained [10] the following estimates:

\[ T_0 \simeq 60 \text{ K}, \quad T_A \simeq 220 \text{ K}, \quad g_Q = 0.16, \quad b_{x_c} = 0.01, \]

and \( y_Q(0) \simeq 0.044 \).

The parameter \( y_Q(0) = [2T_A\chi(Q_c; T = 0)]^{-1} \) is a measure of distance from the magnetic instability. Following earlier studies [22, 23], we consider \( y_Q(0) \) to be the only pressure dependent parameter, assuming that \( y_Q(0) \rightarrow 0 \) as the system approaches a quantum critical point with increasing pressure. In case of ambient pressure, both the empirical parameter \( y_Q(0) \simeq 0.044 \) and the solution of equation (8) are taken the same as in [10], thus providing the agreement with INS data. Here, two remaining fit parameters for \( 1/T_1T \) under ambient pressure are found to be \( (1/T_1T)_{T=0} \simeq 0.55 \) (s\(^{-1}\) K\(^{-1}\)) and \( |A_Q| \simeq 5 \) K G. For NMR data obtained under the applied pressure, the only adjustable parameter \( y_Q(0) \) is estimated by solving first the equation (8) and inserting the solution \( y_Q(T) \) into (7).

2.4. Comparison to experimental data

The experimental data [19] for \( (1/T_1T) \) measured for different applied pressure together with theoretical curves fitting these data are depicted in figure 2. A good agreement between the theoretical results and the available experimental data is found at least down to 1 K. First we note that away from the instability, i.e., \( y_Q(0) > 0 \), the SCR theory predicts the Korringa behaviour \( (1/T_1T)_{T=0} \) in the low-\( T \) limit. On applying higher pressure (or, as \( y_Q(0) \rightarrow 0 \)), \( 1/T_1T \) becomes larger, while a temperature range near \( T = 0 \), where the Korringa relation holds, shrinks and tends to zero at \( y_Q(0) \rightarrow 0 \). These results of the SCR theory are in accordance with the temperature and pressure dependence of \( (1/T_1T) \) observed in LiV\(_2\)O\(_4\).

The SCR theory fails, however, in giving a quantitative description of the low-\( T \) upturn of \( 1/T_1T \) detected below 0.6 K under the highest applied pressure of 4.74 GPa. For instance, as seen from figure 2, the parameter \( y_Q(0) = 0.018 \) fits well in describing the experiment down to \( \sim 1 \) K, while for \( 0 < y_Q(0) < 0.018 \) a strong deviation in the whole temperature range is found (see, for instance, the upper theoretical curve corresponding to \( y_Q(0) = 0.008 \)). It is worth emphasizing that just at the quantum critical point, \( y_Q(0) = 0 \), equation (8) leads to the power law \( (1/T_1T) \sim T^{-3/4} \), in agreement with the earlier SCR theory predictions [22, 23] for the quantum critical behaviour of \( (1/T_1T) \) around the AFM instability in three-dimensional metals. In contrast, the power law \( (1/T_1T) \sim T^{-1/3} \) derived from the experimentally observed behaviour of \( (1/T_1T) \) in a wide temperature range, 0.1 K \( < T < 10 \) K, was reported in [19].

The discrepancy can be clearly explained by taking into account that the SCR theory is invoked to describe homogeneous spin fluctuations in a pure LiV\(_2\)O\(_4\), while a certain number of crystal defects and/or magnetic impurities unavoidably present in the measured powder samples of LiV\(_2\)O\(_4\) may contribute to the nuclear spin relaxation as well. Actually, when approaching a magnetic instability and softening of largely degenerate low-lying spin fluctuations, the system becomes very susceptible to weak perturbations including, for instance, magnetic defects. As known [13–15], the cooperative properties of the paramagnetic LiV\(_2\)O\(_4\) detected by NMR under ambient pressure can be drastically changed at sufficiently low temperatures \( T \sim 1 \) K due to a small number of magnetic defects. We suggest that the low-\( T \) upturn of \( (1/T_1T) \) starting at \( T_{uprun} \approx 0.6 \) K is more likely to be a manifestation of the onset of spin freezing of magnetic defects. If so, this calls for a complete re-examination of the experimental data below \( T_{uprun} \), as done, for instance, in [15] by using a stretched exponential description for the nuclear spin relaxation. We expect that measurement of \( (1/T_1T) \) on a sample with the same quality, but under the ambient pressure, would reveal the onset of spin freezing at a lower temperature, \( T_{uprun} < 0.6 \) K, because of a larger distance from the instability.

Finally, it is helpful to discuss the \( T \) dependence of \( 1/T_1 \) predicted by the SCR theory for higher temperatures, \( T > T_0 \). For instance, a typical behaviour \( 1/T_1 \sim \sqrt{T} \) is expected in the paramagnetic state of nearly and weakly AFM three-dimensional metals [21, 22]. By extending formally the present SCR theory to the region \( T > T_0 \), we obtain \( 1/T_1 \sim T^\alpha \) with \( \alpha > 1/2 \), because of the SCR equation (8) reflecting an
effective quasi-one-dimensionality due to the strong anisotropy in q space of AFM spin fluctuations in our model. We recall, however, that the present SCR theory works well up to 40 K, which is below $T^*$ characteristic to LiV$_2$O$_4$, as evidenced from the comparison of theoretical results with INS data [10]. Actually, for $T > T^*$, the AFM fluctuations at $|q| \simeq Q_c$ are suppressed and no longer distinguished from those at other wavevectors in the BZ; the system enters a spin localized regime compatible with the Curie–Weiss behaviour of $\chi(q = 0)$ observed in LiV$_2$O$_4$ for $T > 60$ K.

3. Summary and conclusions

AFM spin fluctuations located in a large region of q space around the critical surface of a mean radius $|q| \simeq Q_c \simeq 0.6 \, \text{Å}^{-1}$ dominate the low-T properties of the paramagnetic spinel LiV$_2$O$_4$. A parametrized self-consistent renormalization theory of the AFM spin fluctuations was developed and applied to describe temperature and pressure dependences of the low-T spin–lattice relaxation rate $1/T_1$ in this material. Most of the empirical parameters entering the present SCR theory have been estimated earlier and kept fixed in the present study. To simulate pressure effects in our calculations, the inverse static spin susceptibility $\chi^{-1}(Q_c)$ is considered to be the only fit parameter depending on the applied pressure.

Comparison between NMR data and the calculated results has shown that the SCR theory is able to describe correctly the development of AFM spin fluctuations as the paramagnetic metallic state of LiV$_2$O$_4$ approaches a magnetic instability under the applied pressure. In particular, we concluded that up to the highest applied pressure of 4.74 GPa, the spin system is still away from the instability. In this case, the SCR theory predicts the Korringa behaviour ($1/T_1 = \text{constant}$) in a narrow temperature range near $T = 0$; on applying higher pressure, the constant value $1/T_1$ becomes larger and the temperature range near $T = 0$, where the Korringa relation holds, shrinks. The theoretical results were shown to be in a good agreement with experimental data for $1/T_1$ in wide ranges of temperature and pressure. A deviation from the theoretical prediction was interpreted as a signature of change below the characteristic temperature $T_{\text{spin}}$ (5.1 K) of the spin fluctuation dynamics. If for $T > T_{\text{spin}}$ the nuclear spin relaxation is determined by homogeneous AFM spin fluctuations inherent to a magnetically pure LiV$_2$O$_4$, then below $T_{\text{spin}}$ cooperative properties of the detected paramagnetic state are strongly affected by coupling of spin fluctuations to magnetic defects in the measured sample of LiV$_2$O$_4$. A detailed mechanism describing this coupling and the magnetic defect interactions in LiV$_2$O$_4$ remains a challenge [14, 15] for a further work.

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