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LETTER TO THE EDITOR

Spin dynamics in the spin-gap system CaV_4O_9 studied using muon-spin relaxation

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Abstract. We report a muon-spin relaxation study of the two-dimensional spin-gap system CaV_4O_9. We find that the form of the muon-spin relaxation is strongly temperature dependent and attribute this behaviour to the presence of a spin gap. At temperatures below 30 K a root-exponential behaviour is seen for the muon-spin relaxation function, which can be attributed to defect spins. At temperatures above 160 K we see the onset of muon hopping.

Although a number of one-dimensional spin-gap materials are known and have been investigated experimentally (e.g. [1]), the existence of the first (quasi-) two-dimensional spin-gap material, CaV_4O_9, has only been reported recently [2]. This material consists of V_4O_9 planes separated by layers of calcium atoms [3]. Each vanadium atom is at the centre of an O_5 square pyramid and is in oxidation state 4 with $S = \frac{1}{2}$. Magnetic susceptibility [2], NMR [4, 5] and neutron scattering [5, 6] measurements indicate the existence of a spin gap in this material with a spin-gap temperature $T_{sg} \sim 110$ K. These experimental reports have stimulated a number of theoretical studies [7–10]. The magnetic system is described by a Heisenberg model for the vanadium spins for a $\frac{1}{5}$-depleted square lattice. At each site of this lattice two of the bonds belong to the spin plaquettes ('plaquette bonds'), and the third one connects a plaquette with its neighbour (a 'dimer bond'). The inclusion of a frustrating next-nearest-neighbour (NNN) interaction seems to be essential for the formation of the spin gap [8]. Starykh *et al* [9] argue that CaV_4O_9 might represent an example of a two-dimensional 'spin–Peierls'-like system where spin–phonon coupling cooperates with tendencies to form a spin gap due to the frustrating NNN interactions. Later work [10] suggests that the picture of two coupled metaplaquette systems is a realistic way of interpreting the spin-gap behaviour.

In this letter we investigate the effect of the spin gap on the magnetic excitation spectrum using muon-spin relaxation (μSR). Subtle magnetic effects have been studied in a wide range of materials with μSR [11], including spin-gap formation [12, 13].

Powder samples of CaV_4O_9 were prepared by direct reaction of molar ratios of CaO (Johnson–Matthey, puratronic grade) and VO_2 (99.9%, Aldrich Chemical Company). The starting powders were ground together in a glove box and then sealed under vacuum in silica ampoules. Heat treatments of 18 h at 750 °C were employed, following previous reports [2, 3]. The product was reground in the glove box and the heat treatment repeated to ensure homogeneous reaction. After this stage the product appeared uniformly green in colour. We shall see below that our material contains a substantial concentration of magnetic defects, presumably arising from disorder in the vanadium sublattice.

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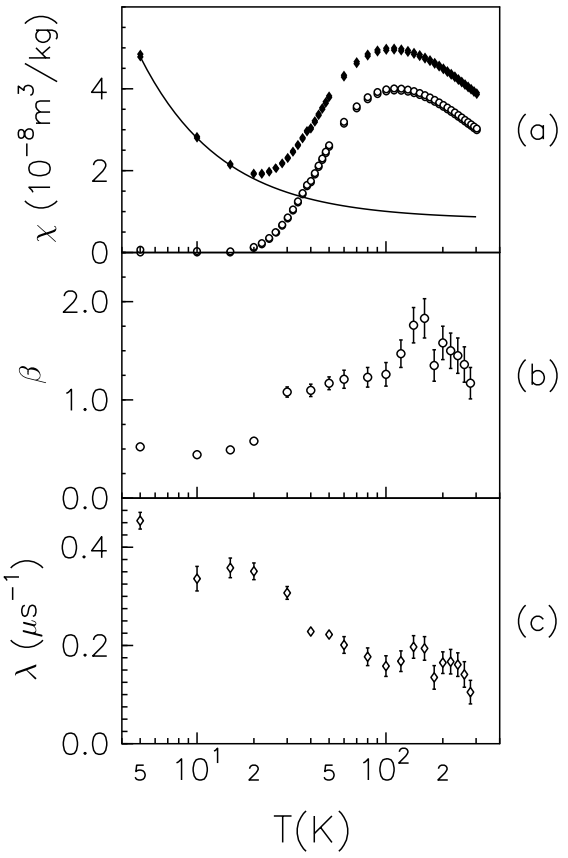

Figure 1. The temperature dependence of (a) the measured mass susceptibility of CaV_4O_9 (diamonds), calculated susceptibility due to defects, diamagnetism and van Vleck paramagnetism (full curve) and spin susceptibility (open circles) (diamonds minus full curve), (b) the parameter β (see the text); $\beta = 2$ corresponds to a Gaussian relaxation, $\beta = 1$ to an exponential relaxation and $\beta = \frac{1}{2}$ to a root-exponential relaxation and (c) the relaxation rate λ (see the text).

Figure 1(a) shows the mass susceptibility $\chi = \mu_0 M/B$ of our sample of CaV_4O_9, where the magnetization M was measured with a SQUID magnetometer under a magnetic field of $B = 0.1$ T; variation of the magnetic field in this region showed a clear linear relation between magnetization M and applied field B. Below ~20 K there is an upturn of χ with decreasing temperature due to defect spins. Below the spin-gap temperature of $T_{sg} \sim 110$ K the value of χ drops. Following reference [2] we split the contributions to the susceptibility into three terms: (i) the spin susceptibility χ_{spin}, (ii) the diamagnetic part and the Van Vleck paramagnetic part χ_0, which are both temperature independent, and (iii) the defect-spin contribution $\chi_{def} = C/T$ with $C = (N/m)(g\mu_B)^2 J(J+1)/(3k_B)$, where N is the number of defect spins in the sample, m the mass of the sample, μ_B the Bohr magneton and k_B the Boltzmann constant. We take $g = 2$ and $J = 1/2$. The expression in (iii) is a good approximation to a Brillouin function in the given temperature and magnetic field range. The values of C and χ_0 have been fitted assuming $\chi_{\text{spin}} = 0$ for temperatures below 15 K [4] (see figure 1(a)). This enables us to plot the contribution to the susceptibility made by the spin system (open circles in figure 1(a)), in good agreement with previous measurements [2].

Using the fitted values we can estimate the defect-spin density to be $N/m \approx 5.1\times10^{20}$ kg^{-1}, giving $\approx 3.3\times10^{-4}$ defect spins per formula unit, and as demonstrated below these lead to observable muon-spin relaxation only at low temperatures.

Zero-field μSR experiments were carried out at the Paul Scherrer Institute (Switzerland). In a μSR experiment spin-polarized positive muons are implanted into a sample and the polarization of an ensemble of muon spins is recorded by monitoring the time dependence of the angle-dependent decay positron emission [11]. This is possible since the decay positrons are emitted preferentially in the direction of the instantaneous muon spin. We present the μSR data in the form of the asymmetry (or relaxation) function G_z. This function compares the positron counts in the forward and the backward detectors. The Earth's magnetic field was compensated to less than 10 μT. The sample was mounted on a silver backing plate, which provides a non-relaxing muon-spin signal.

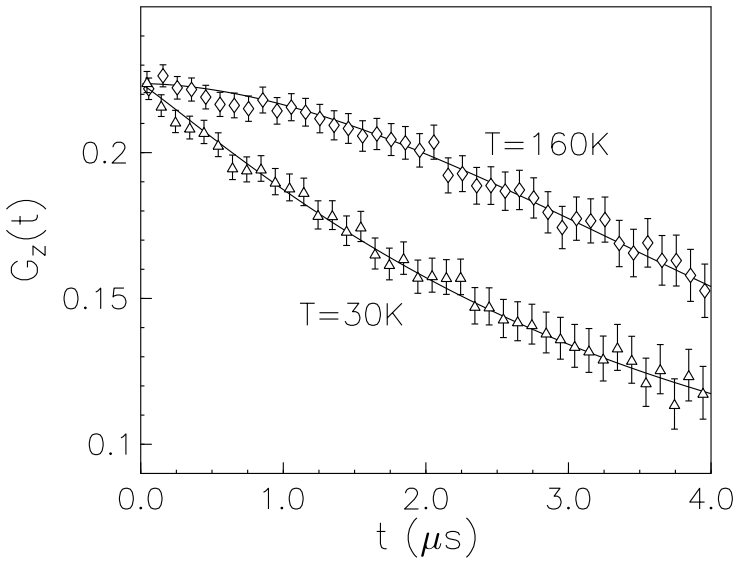

Figure 2. Decay positron asymmetry functions G_z for CaV_4O_9 at 160 K (diamonds) and 30 K (triangles) The data are fitted by a Gaussian (160 K) and an exponential (30 K) form.

Typical $G_z(t)$ values are shown in figure 2 for $T < T_{sg}$ and $T > T_{sg}$. Well below T_{sg} we find that G_z is well fitted by an exponential function and well above by a Gaussian. We can follow the exact behaviour of the relaxation curve as a function of temperature by fitting the data to the stretched-exponential form

$$G_z(t) = A_S e^{-(\lambda t)^\beta} + A_{Ag} \tag{1}$$

where λ is a temperature-dependent relaxation rate; A_S and A_{Ag} are temperature-independent amplitudes, reflecting the relative fraction of muons stopping in the sample ($\sim A_S$) and in the silver backing plate ($\sim A_{Ag}$). A value of $\beta = 2$ corresponds to a Gaussian relaxation, $\beta = 1$ corresponds to an exponential relaxation and $\beta = \frac{1}{2}$ corresponds to a root-exponential relaxation. Equation (1) fits the data well over the entire temperature range studied. In the fitting process A_S and A_{Ag} were kept fixed and λ and β were adjustable parameters.

The complex temperature dependence of β (figure 1(b)) may be understood in terms of the following model.

(i) Above T_{sg} the parameter β rises from $\sim$1 to a maximum value of $\beta \sim 1.9$ at 160 K. This behaviour can be ascribed to the fluctuations of the electronic spins [14]. At low temperatures the electronic fluctuations produce exponential relaxation but, as the temperature rises, these fluctuations become too fast for the muon to follow. Hence the dipole fields of the fast-fluctuating electronic spins average to zero over a time much shorter than the lifetime of the muon. The resulting Gaussian relaxation can be attributed to randomly orientated quasi-static nuclear moments [14] of ^{51}V, the nuclear relaxation times being longer than the typical muon lifetime [5].

(ii) Above 160 K the functional form of the relaxation becomes an exponential again. This drop in β is explained by the onset of muon hopping. Thus, the muon averages the fields of the randomly orientated nuclear spins [14].

(iii) Below T_{sg} the electronic spins begin to freeze out. Between 30 K and 100 K we find $\beta \sim 1$ and $0.17\ \mu s^{-1} < \lambda < 0.30\ \mu s^{-1}$ (figures 1(b) and 1(c)). The increasing relaxation rate below T_{sg} is consistent with slowing spin fluctuations [13]. In the same temperature region the susceptibility decreases with decreasing temperature (figure 1(a)). λ varies exponentially in this temperature regime; the resulting activation temperature is comparable with the spin-gap temperature of 110 K, indicating that the fluctuations are associated with thermal excitations across the spin gap.

(iv) At temperatures below 30 K the shape of G_z is root exponential ($\beta \sim \frac{1}{2}$; figure 1(b)), which is characteristic of a dilute spin system [15]. As the temperature is lowered, more and more of the electrons go into singlet states which are non-magnetic and hence do not affect the muons. In this low-temperature regime, in which the electronic spins are frozen out, the dominant mechanism for relaxing the muon polarization comes from the dilute defect spins. This freezing out of spins below 20 K coincides with the vanishing of the spin susceptibility (figure 1(a) and reference [2]) and the saturation of the transverse relaxation rate in NMR [5].

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