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Au$_4$V – Moment Stability and Spin Fluctuations in the Ordered Phase

K.J. Ellis, R. Cywinski, S.H. Kilcoyne, A.D. Hillier

1. The Gold-Vanadium System

The first investigations (1959) of magnetism in the gold-vanadium system were performed on disordered alloys ranging from 1-15 at.% vanadium [1], in which the temperature dependent susceptibility closely obeyed a Curie-Weiss law incorporating a temperature independent term, χ_0, such that

$$\chi = \chi_0 + \frac{C}{(T - \theta)} \qquad (1)$$

where C is the Curie constant and θ is the critical temperature. Interestingly the critical temperature of these relatively dilute alloys was found to be negative, despite there being no evidence of a magnetic transition. To explain this behaviour a model was proposed in which a fraction of "isolated" vanadium atoms i.e. those without other vanadium nearest neighbours, possess a local moment with the remaining fraction contributing to the Pauli-like temperature independent term.

Creveling et al [2] extended the study of Au-V to higher concentrations ranging from 17-24 at.% V. When annealed at ~500°C, these alloys were found to undergo a structural transition forming an intermetallic compound with the nominal concentration Au$_4$V with a body-centred tetragonal structure (I4/m). This phase effectively isolates all the vanadium atoms and correspondingly a ferromagnetic transition was found at ~45K. In this ordered phase the susceptibility above T$_c$ obeys the Curie-Weiss law given in Eq. 1. However a large disparity in the magnetic measurements between different samples was noted. It has been suggested that this is due in part to magneto-crystalline anisotropy which is not only the result of the non-cubic structure but also crystalline imperfections caused by the arbitrary alignment of the tetragonal c-axis. As generally observed for weak itinerant electron ferromagnets, the ordered moment of Au$_4$V is substantially lower (approximately half that) of the paramagnetic moment; however, annealing under compression promotes alignment of the c-axis, and increases the ordered moment from the usual value of 0.4-0.6μ_B to 0.83μ_B per V atom [3]. Furthermore, in extremely high pulsed magnetic fields (29T) the ordered moment saturates to $M_s = 1\mu_B$ per V atom, indicating a spin-½ moment [4].

The importance of the V-V distance in establishing a localised moment, and consequently ferromagnetic order, is further emphasised by the observation that when Au$_4$V is prepared as a disordered solid solution the V moment, and also ferromagnetism, collapses. Au$_4$V thus appears to belong to the same class of weak itinerant ferromagnets as ZrZn$_2$ and Sc$_3$In. However, in marked contrast to systems such as ZrZn$_2$, for which the application of external pressure rapidly decreases T$_c$ to zero by 20kbar, the Curie temperature of Au$_4$V increases with pressure to 90K at ~180kbar, at which point the magnetic order collapses entirely [5].

Positive muon spin relaxation has proved to be an invaluable tool in investigating itinerant magnetic systems such as MnSi, as demonstrated in the seminal paper by Hayano et al [6]. We have therefore performed zero and longitudinal-field measurements on Au$_4$V using the MuSR spectrometer, (ISIS Facility, UK) to follow the evolution of spin fluctuations with temperature between 5K and 90K.

The polycrystalline Au$_4$V samples were prepared by argon arc melting stoichiometric proportions of gold and vanadium. The resulting 2g ingots were pressed into disks, approximately 5mm in diameter and

1.5mm thick before undergoing two days of homogenisation at 1000°C under reduced argon atmosphere, followed by annealing at 500°C for eight days.

2. Muon Spin Relaxation in Zero-Field

The observed zero-field muon spin relaxation spectra for Au_4V were best modelled by a dynamical Kubo-Toyabe function, $G^{(DKT)}$, representing a nuclear dipole contribution, multiplied by a simple exponential term, representing the contribution from atomic spin fluctuations .

$$a_0 G_z(t) = [a_1 G_z^{DKT}(t) \times \exp(-\lambda t)] + a_b \qquad (2)$$

where a_0 is the initial asymmetry, a_1 is the relaxing asymmetry and a_b is a background.

In the paramagnetic regime, above 50K, the muon relaxation rate σ from the randomly oriented nuclear dipoles is constant at $0.34\mu s^{-1}$. Furthermore, these spectra show a marked divergence of λ from a negligibly small value at high temperatures to a maximum at the reported Curie point of Au_4V.
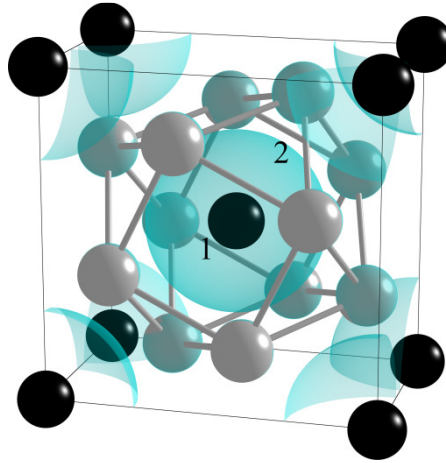

Figure 1: The structure of ordered Au_4V in which vanadium atoms are black and gold atoms silver. The isosurface indicates the positions within the unit cell where the nuclear depolarisation rate, $\sigma = 0.34\mu s^{-1}$. Labels 1 & 2 indicate the likely muon position.

Estimates of the nuclear dipole contributions within the unit cell suggest possible muon sites at the centres of the 3Au-1V tetrahedron or the 5Au-1V octahedron as shown in Figure 1. The slow fluctuation rate rate ($<1\mu s^{-1}$) observed as a damping of the "tail" of the Kubo-Toyabe function is likely to arise from the muon hopping between these interstitial sites.

3. Muon Spin Relaxation in Applied Longitudinal Fields

Spectra from Au_4V were collected in sufficiently high longitudinal magnetic fields to decouple the muon from the nuclear dipole fields, but sufficiently small to offer negligible perturbation of the atomic spins. In Figure 2 we present spectra from a range of applied longitudinal-fields above the transition temperature, clearly showing a full decoupling at 100G. Above this field the μSR spectra are well described by a simple exponential function. The associated relaxation rate, λ, increases rapidly as the transition temperature is approached until below 50K a there is a critical divergence. As found for the weak itinerant helimagnet, MnSi [6] this critical divergence is best described by the simple formula:

$$\lambda = \tau_\infty \frac{T}{T - T_c} \tag{3}$$

A least square fit of Eq. 3 to the data provides $T_c = 42.0 \pm 0.2$K and $\tau_\infty = (3.17 \pm 0.14) \times 10^{-3} \mu s^{-1}$. For comparison τ_∞ for MnSi (T_c=29.5K) was found to be $(6.55 \pm 0.13) \times 10^{-3} \mu s^{-1}$. Figure 3 shows the critical scaling of λ, described by Eq. 3, for both Au_4V and MnSi with the data for the latter taken from [6].

4. Conclusions

The critical scaling of the muon spin relaxation rate observed for Au_4V shows close similarity with that obtained for the archetypal itinerant electron ferromagnet, MnSi, and correspondingly follows the predications of Moriya's self consistent renormalisation (SCR) theory for itinerant systems [7]. In this respect Au_4V may well prove a simpler system with which the mechanisms responsible for moment localisation in itinerant electron magnets can be explored.
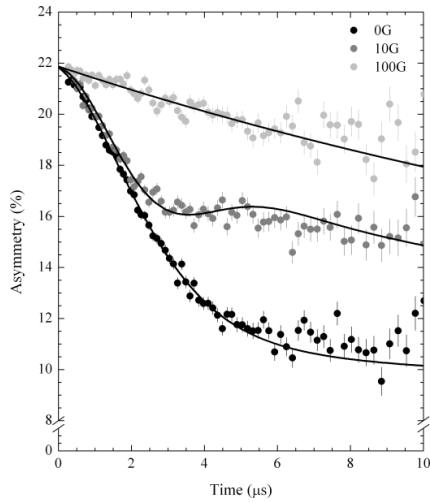

Figure 2: The longitudinal field muon spin relaxation spectra from Au_4V observed at 46K in several applied fields. Eq. 2 was used to fit both 0G and 10G data. The application of 100G fully decouples the nuclear dipole fields such that it can be fitted with a single exponential term.
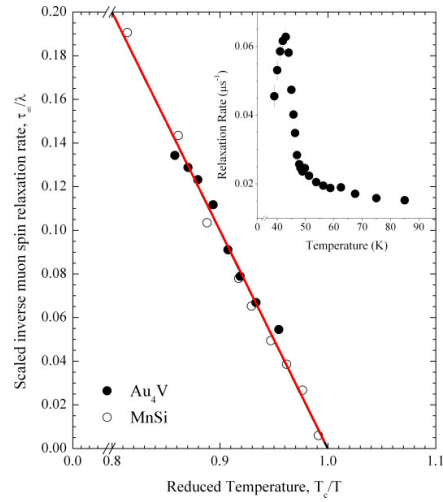
Figure 3: The scaled inverse muon spin relaxation rate (τ_∞ / λ) versus reduced temperature for Au_4V (closed circles) and MnSi (open circles). The solid line represents the fit of Eq. 3, as predicted by SCR theory [7] to the data. The experimental points for MnSi are taken from [6]. Insert: The temperature dependence of λ in Au_4V.

5. Acknowledgements

6. References

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