Evidence for an antiferromagnetic component in the magnetic structure of ZrZn$_2$

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Abstract

Zero-field muon spin rotation experiments provide evidence for an antiferromagnetic component in the magnetic structure of the intermetallics ZrZn$_2$.

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The discovery of superconductivity below 1 K within a limited pressure range in UGe$_2$ provided an unanticipated example of the coexistence of superconductivity and ferromagnetism [1]. Later on, resistivity measurements indicated the onset of superconductivity in ZrZn$_2$ at ambient pressure below $\sim$ 0.3 K, i.e. deep in the ferromagnetic ordered state of this $d$-electron compound [2]. The electronic pairing mechanism needed for superconductivity is believed to be of magnetic origin for both materials.

It is important to ascertain the magnetic structure of these two compounds because a Cooper’s pairing mechanism in compounds with a spin density wave has been described in the literature [3]. The ferromagnetic state of UGe$_2$ is in fact not simple since bulk measurements provide evidence for a magnetic transition within the ferromagnet state. Its origin is unknown, despite a number of neutron diffraction investigations. The general belief that the ground state of ZrZn$_2$ is ferromagnetic below $T_C$ $\approx$ 28 K rests on measurements performed more than 40 years ago [4,5]. A magnetic structure which would be characterised in neutron diffraction by weak magnetic satellites in the vicinity of a
main ferromagnetic Bragg reflection is not excluded experimentally. The satellites would mean that the magnetic structure of \( \text{ZrZn}_2 \) contains an antiferromagnetic (AFM) component. Since the saturation moment is relatively small and the magnetic structure the compound may adopt is unknown, the muon spin rotation (\( \mu \)SR) technique is the most convenient method to determine whether an AFM component is at all possible. Here we report the results of measurements with this technique, which demonstrate the existence of an AFM component in the magnetic structure of \( \text{ZrZn}_2 \).

The \( \text{ZrZn}_2 \) powder sample was made by slow cooling of an optimised alloy of Zr and Zn with atomic ratio \( \text{Zr:Zn = 1:2.006} \) in order to avoid the formation of surrounding secondary phases (\( \text{ZrZn}_3 \) and \( \text{ZrZn} \)). The starting materials were zone refined zirconium and zinc of 6N purity. To prevent the contamination of \( \text{ZrZn}_2 \) by aluminium if an alumina crucible was used, the mixture of zirconium and zinc was introduced in a homemade yttria crucible which was encapsulated in a tantalum crucible to hold the zinc vapor pressure. No further annealing was performed on this sample.

The analysis of x-ray diffraction patterns reveals no parasitic phases. They are consistent with the C15 cubic Laves structure. Figure 1 shows the magnetisation measured at low temperature in a field of 0.1 T. The magnetic ordering temperature was determined from Arrott’s plot (not shown) to be close to 27 K.

![Graph showing magnetisation versus temperature](image)

Fig. 1. Magnetisation versus temperature measured in an external field of \( B_{\text{ext}} = 0.1 \) T on the sample of \( \text{ZrZn}_2 \) used for the \( \mu \)SR experiment.

The zero-field \( \mu \)SR measurements were performed at the \( \pi \)M3 beamline of the Swiss muon source (Paul Scherrer Institute, Villigen, Switzerland) from 1.7 up to 66 K.

The \( \mu \)SR technique consists of implanting polarised (along direction \( Z \)) muons into a specimen and monitoring \( P_{\text{exp}}(t) \) which provides a measure of the evolution of the muon ensemble polarisation projected onto direction \( Z \); for an introduction to this technique, see e.g. Refs. [6,7,8]. The quantity actually measured is the so-called asymmetry corresponding to \( a_0P_{Z_{\text{exp}}}(t) \), where \( a_0 \approx 0.24 \).

For a paramagnet, \( P_{Z_{\text{exp}}}(t) \) tracks the dynamics of the magnetic field at the muon site, reflecting the dynamics of the electronic moments. In the fast fluctuation or motional narrowing limit, \( P_{Z_{\text{exp}}}(t) \) takes the exponential form \( \exp(\lambda_Z t) \) characterised by a relaxation rate \( \lambda_Z \). The measurements above \( T_C \) are fully consistent with this expectation. \( \lambda_Z \) is found to be smaller than 0.01 \( \mu s^{-1} \). The thermal dependence of \( \lambda_Z \) above \( T_C \) has not been studied in details because
Asymmetry:
\[ P_{\text{exp}} = p_1 \exp (-\lambda_X t) J_0 (\gamma \mu B_{\text{max}} t) + p_2 \exp (-\lambda_Z t), \]

with \( p_1 + p_2 = 1 \). The damped oscillation is modeled by the product of two functions: a zeroth-order Bessel function \( J_0 \) and an exponential function. \( J_0 \) is the cosine-Fourier transform of the field distribution at the muon site resulting from a simple incommensurate magnetic structure [9]. However, it is possible to generate a similar distribution out of a complex magnetic structure; see for example Fig. 10 in Ref. [10]. Anyway, \( J_0 \)-like oscillation indicates clearly that ZrZn\(_2\) cannot be a simple ferromagnet. The exponential envelop of the oscillations is thought to account for the finite coherence length, i.e. the disorder of the magnetic structure. \( \gamma \mu \) is the muon gyromagnetic ratio (\( \gamma \mu = 851.6 \text{ Mrad s}^{-1} \text{ T}^{-1} \)), \( B_{\text{max}} \) the maximum value of the magnetic field at the muon site and \( \lambda_Z \) the spin-lattice relaxation rate. Numerically, the fit of the spectrum in Fig. 2 gives \( B_{\text{max}} = 16.5 (1) \text{ mT} \), \( \lambda_X = 1.42 (9) \mu \text{s}^{-1} \) and \( \lambda_Z = 1.63 (11) \mu \text{s}^{-1} \). We find \( p_1/p_2 \sim 2.7 \) instead of the expected value of 2. This discrepancy is a signature of a relatively large texture for our sample, which is not surprising since it is made of relatively large crystallites. Spectra were recorded for increasing temperatures up to \( T_C \). They are all well described by Eq. 1.

To reach a reliable conclusion concerning the possible presence of an AFM component in the magnetic structure of ZrZn\(_2\), we need to determine whether the description of the wiggles displayed by \( P_{\text{exp}}^Z(t) \) with the zeroth-order Bessel function is
safe. We have done a fit of the measured spectrum substituting in Eq. 1 $J_0(\gamma \mu B_{\text{max}} t)$ with the cosine function $\cos(\gamma \mu B_{\text{loc}} t + \varphi)$. We derive $B_{\text{loc}} = 16.0 \pm 2$ mT. Remarkably, $\varphi = -34.5 \pm 2.2$ degrees. The phase shift $\varphi$ is therefore relatively large whereas to get a physically grounded cosine fit, $\varphi$ should be negligible, reflecting only the inherent error of the experimental determination of the time origin. We have checked that point by recording zero-field spectra on the intermetallics PrRu$_2$Si$_2$ which is a collinear ferromagnet below 14 K [11] and which is characterised by $B_{\text{loc}}$ of the same order of magnitude as ZrZn$_2$. As expected, the observed oscillation is well accounted for by a cosine function with a negligible phase shift.

The fact that the fit with a shifted cosine function provides a description of the spectrum with the exception of the short times, is easily understood if we recall that $J_0(x)$ is well approximated by the asymptote $(\frac{2}{\pi x})^{\frac{1}{2}} \cos(x - \frac{\pi}{4})$ if $x$ is not too small.

In conclusion, the $J_0(x)$ Bessel function provides a reliable description of the measured $\mu$SR oscillation in the ordered state of ZrZn$_2$. This means that this compound is not a simple ferromagnet and that an AFM component is therefore present. However, our measurements do not resolve the magnetic structure, as no information on propagation wavevectors could be extracted from the $\mu$SR data. Since the superconductivity of ZrZn$_2$ is thought to be of magnetic origin, a proper characterisation of its magnetic structure is important. This could be achieved using neutron scattering experiments. Among the possibilities for the magnetic structure, a well known example for a compound with a spiral structure is provided by CeAl$_2$ [12]. Interestingly, CeAl$_2$ shares the cubic C15 crystal structure with ZrZn$_2$. However, the length of the wavevector is substantial for CeAl$_2$ whereas, if it exists, it is probably small for ZrZn$_2$.

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