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Muon spin rotation measurements on LaNiSn

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Abstract

The first microscopic investigation of superconductivity in LaNiSn is reported using muon spin rotation. LaNiSn is found to be mainly a type I superconductor in an intermediate state with some evidence for type II behaviour at low temperatures, possibly due to a temperature dependent Ginzburg Landau parameter κ.

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1. Introduction

LaNiSn has recently generated interest due to its structural analogue, CeNiSn. Both CeNiSn and LaNiSn have an orthorhombic crystal structure [1], but LaNiSn is the non-magnetic analogue of the 4f Kondo insulator CeNiSn and therefore a useful comparison. Both NMR [2] and magneto-resistance [3,4] indicate a small pseudo-gap at the Fermi Energy in CeNiSn, which is due to the anisotropic hybridisation between the 4f electrons and conduction electrons [5] in the orthorhombic structure. LaNiSn is known to be a good metal [3,6] with an electronic specific heat coefficient $\gamma = 11.4\,\mathrm{mJ\,mol^{-1}\,K^{-2}}$. Following the availability of high quality single crystals, LaNiSn was found to show evidence of superconductivity [7]. This paper represents the first microscopic investigation of superconductivity in LaNiSn.

2. Experimental results

Until recently [7] previous experiments on LaNiSn were performed on polycrystalline samples [3,4,6]. The sample used here was a high-quality single crystal. High-purity constituent materials La, Ni and Sn were melted into a polycrystalline ingot in a cold copper crucible under a purified argon atmosphere. The single crystal was then grown using the Czochralski pulling method [7] using a radio frequency induction furnace with a hot tungsten crucible. The crystal was cylindrical with a diameter of $\sim 5\,\mathrm{mm}$ and a length of $\sim 25\,\mathrm{mm}$. In order to decrease defects, strains and impurity ions, the as-grown crystal rod was treated using solid-state electrotransport. The rod was heated to 1000 °C by a direct current of $250\,\mathrm{A\,cm^{-2}}$ for two weeks under a vacuum better than $3 \times 10^{-8}\,\mathrm{Pa}$. Crystallographic orientation was determined by the back-scattering X-ray Laue method and resistivity measurements yield a large residual resistivity ratio, indicating the sample is of high quality. The superconducting critical temperature was found to be 550 mK.

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This paper describes TF MuSR measurements of the superconducting state in LaNiSn performed on the LTF instrument at PSI and on the MUSR instrument at ISIS. The temperature was varied over a range of 50 mK–1.1 K, with a typical magnetic field ranging 0–600 Oe applied perpendicular to the sample's cylindrical axis and transverse to the muon spin direction. The sample was mounted on a ~5 mm thick annealed haematite plate (Fe_2O_3) to ensure a rapid, random depolarisation of any muons not hitting the sample. The data was analysed using a Maximum Entropy technique [8].

Fig. 1 shows the field probability distribution, $P(B)$, taken at PSI for a range of temperatures. The sample was cooled in an applied field of 17 Oe from above T_c and measurements were made on heating. An initial, qualitative description of the data shown in Fig. 1 shows an *increase* in internal field as T_c is passed followed by a splitting of the peaks at lower temperatures. We first examine the behaviour of our sample just below T_c. The observed increase in internal field and the corresponding decrease in asymmetry in Fig. 1 can be explained by an intermediate state present in our sample. If a magnetic field is applied to a type I superconductor which has a non-zero demagnetisation factor, the magnetic field over part of the surface may exceed the critical field even though the applied field is considerably less than it. The value of the demagnetisation factor is dependent on the shape of the sample and its orientation with respect to the applied field. For an infinitely long cylinder with the field applied perpendicular to the axis, it is exactly one half. Thus, applying a magnetic field greater than half the critical field at a given temperature will yield an intermediate state, containing large volumes of both normal and Meissner states.

A field of 17 Oe is sufficient to allow an intermediate state, as LaNiSn has a critical field of approximately 30 Oe at 500 mK [7]. Since the Meissner state is one of total flux expulsion, one would expect the asymmetry to reduce as the Meissner volume fraction of the intermediate state increases. One would also expect a greater flux density in the normal part, due to the flux expelled from the Meissner regions. Thus, at temperatures between 550 and 400 mK, it is clear that our measurements are consistent with an intermediate state.

At lower temperatures, however, the flux distribution inside our sample is considerably more complicated. The significant shift of the peak corresponding to the normal fraction to higher fields and the sudden drop in total asymmetry indicates a decreased volume fraction of the normal state and an increased volume fraction of the Meissner state. The appearance of the second peak at a field below that of the normal region's field indicates part of the superconducting region is no longer in the Meissner or normal state. This peak cannot be due to stray muons, as there are no oscillations observed in Fig. 4 in the true Meissner state. Furthermore, there is no peak present in the $P(B)$'s in Fig. 1 between 400 and 550 mK. Finally, at 100 mK and below, the peak has a non-Gaussian structure. If it were a background peak, it should always be present

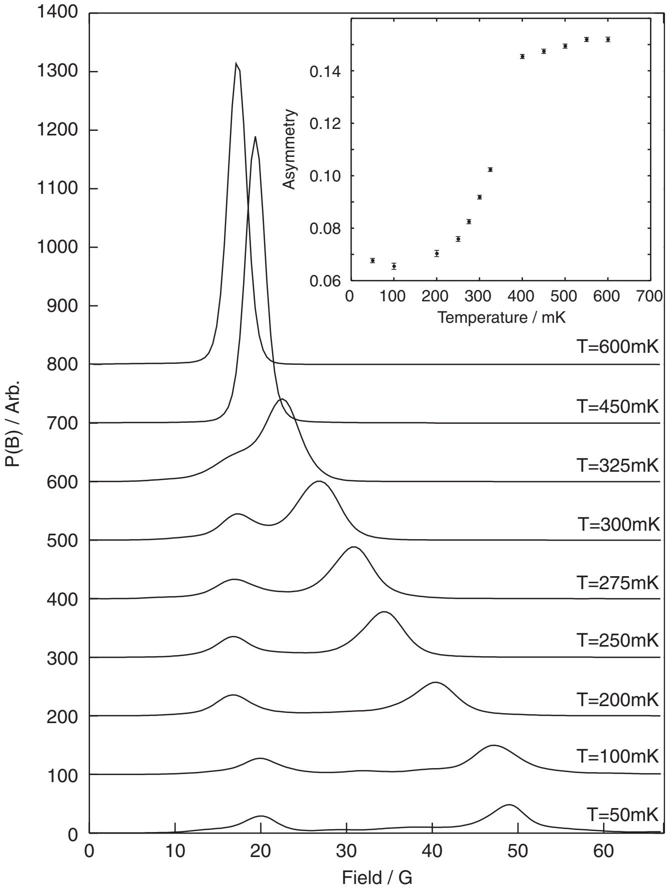

Fig. 1. Temperature dependence of $P(B)$ cooled in a 17 Oe field. Error bars have been removed for clarity. Plots are on the same linear scale with arbitrary units and have been offset from the previous temperature by 100. The $P(B)$'s for $T = 500$ and 550 mK have not been included, but are consistent with the general trends shown here. Inset: The experimental asymmetry for the $P(B)$'s shown in the main panel.

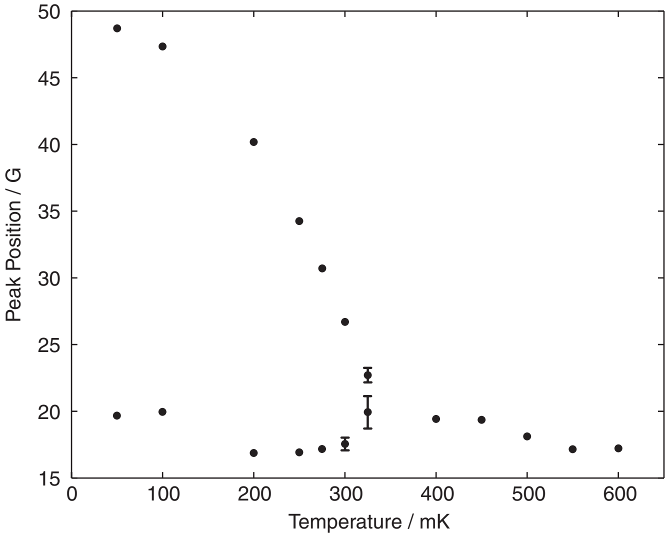

Fig. 2. Position of the peaks in Fig. 1 obtained from fits to a double Gaussian. Error bars are only shown when larger than the points.

and identical at all temperatures for a given field. This peak most likely corresponds to a mixed state within the sample. Fig. 2 shows the position of the peaks as a function of temperature, obtained from fits to a double Gaussian. Although in the lower branch at 200 and 250 mK the points are at a marginally smaller field than the applied field, the rest of the points are not. However, in an intermediate state, the superconductor sees the flux density present in the normal regions or at the sample surface which is higher than the applied field. Thus the lower peaks present in Fig. 1 correspond to a region of partial flux expulsion, but not complete expulsion as in the Meissner state. The drop in total asymmetry is too great to be accounted for by only a mixed state, so the low temperature spectra are consistent with a sample mainly comprised of the Meissner state, but with some normal regions and some regions in the mixed state.

This apparent mixture of type I and II superconductivity could be explained by considering a superconductor on the type I/II boundary, with a Ginzburg Landau (GL) parameter $\kappa(T_c)$ very close to $1/\sqrt{2}$. From the residual resistivity of LaNiSn [7], the mean free path is approximately $l \sim 700\,\text{Å}$, which is considerably less than the GL coherence length ($\xi_0 \sim 1200\,\text{Å}$) [7]. This indicates LaNiSn is in the non-local limit, so $\kappa(T)$ is expected to have an appreciable temperature dependence [9]. Indeed, by finding a gauge-invariant solution to the linearised Gor'kov equations, it was shown that $\kappa(T)$ can vary between $\kappa(T_0) \sim 1.2\kappa(T_c)$ [10,11] and $\kappa(T_0) \sim 1.8\kappa(T_c)$ [12] for a ξ_0 and l similar to our sample.

The inset of Fig. 3 shows the $P(B)$'s at 50 mK, cooled in a variety of fields from above T_c. Plotting the full width of the $P(B)$'s as a function of applied field (Fig. 3) is a useful measure of the subtle changes in the $P(B)$'s. The initial increase in the width (indicated with an arrow) is taken to be the onset of superconductivity. By performing a linear extrapolation of the data, the critical field can be estimated to be $B_c \sim 135$ Oe. After a plateau there is a dramatic increase in width below ~70 Oe, that corresponds to the field at which a combined intermediate/mixed state occurs.

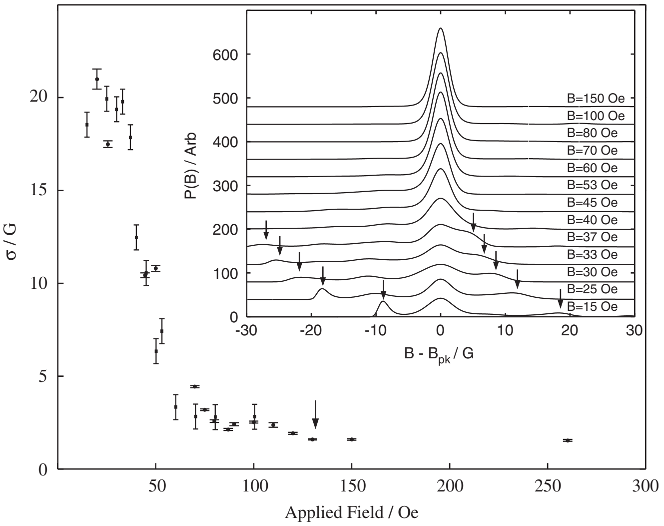

Fig. 3. Square root of the second moment for field cooled measurements at a variety of applied fields. Squares represent data taken at ISIS and circles at PSI. The ISIS points have been scaled down to take account of the background peak which is not present at PSI. Inset: $P(B)$'s obtained from ISIS at a temperature of 50 mK, cooled in a variety of fields from above T_c. In order to make a direct comparison between different fields, it was necessary to plot $P(B)$ as a function of $(B - B_{\text{peak}})$. The central peak evident at low temperatures is a background peak due to stray muons.

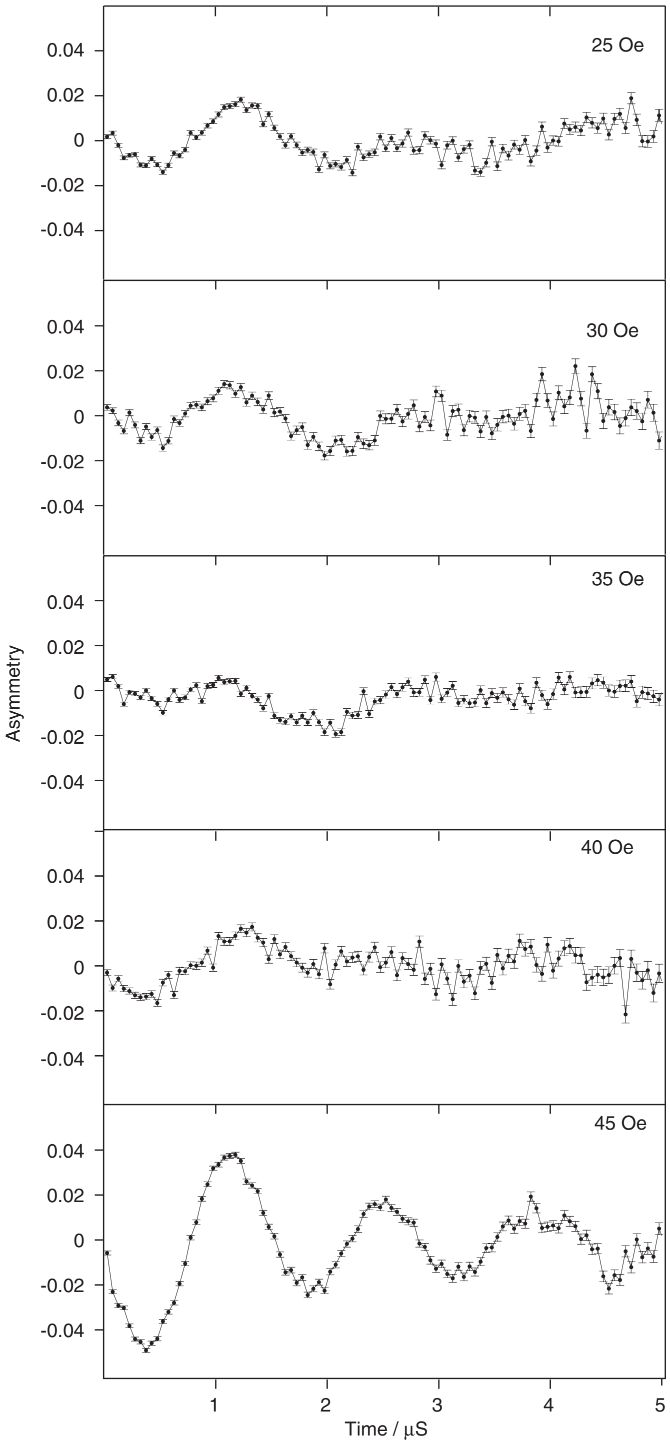

Fig. 4. The asymmetry of LaNiSn initially cooled to 50 mK in zero field, then the field was increased in 5 Oe steps.

Fig. 4 shows time-domain data plotted for three magnetic fields, after cooling in zero applied field to 50 mK and applying the field in steps of 5 Oe. The superconducting magnet was quenched prior to measurements to ensure the measurement was performed in as close to zero magnetic field as possible. At 25 Oe, it is clear there is a weak oscillation in the data, although the asymmetry is rather small, indicating a large fraction of the sample is in the Meissner state. As the field is increased, however, the asymmetry *reduces* to almost nothing at 35 Oe, before increasing slightly at 40 Oe followed by another more significant increase at 45 Oe. This behaviour has been measured multiple times at both ISIS and PSI, although only the PSI data is shown here. This "entrant" Meissner state as the field is increased cannot currently be explained. Curiously only at 35 Oe, and not at lower fields, is a pure Meissner state observed.

In conclusion, we have shown LaNiSn is mainly a type I superconductor with possible evidence for a combined mixed/intermediate state. On cooling in zero field and applying a magnetic field, an "entrant" Meissner state is observed only at an applied field of 35 Oe. The μSR experiments were performed at the Swiss Muon Source, PSI, Villigen and at ISIS, Rutherford Appleton Laboratory, UK. We thank the EPSRC for financial support and the facility staff for technical assistance.

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