# Superconducting properties of $Ca_3Ir_4Sn_{13}$: a $\mu$SR study

**P K Biswas[1], A Amato[1], Kefeng Wang[2], C Petrovic[2], R Khasanov[1], H Luetkens[1] and E Morenzoni[1]**

[1] Laboratory for Muon Spin Spectroscopy, Paul Scherrer Institute, CH-5232 Villigen PSI, Switzerland
[2] Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, New York 11973, USA

E-mail: pabitra.biswas@psi.ch, elvezio.morenzoni@psi.ch

**Abstract.** Muon spin relaxation and rotation ($\mu$SR) measurements have been performed to study the superconducting properties of $Ca_3Ir_4Sn_{13}$. Zero-field $\mu$SR data shows no sign of any magnetic anomaly in $Ca_3Ir_4Sn_{13}$ at the superlattice transition temperature, $T^*$ or in the superconducting ground state. Transverse-field $\mu$SR measurements in the vortex state provided the temperature dependence of the magnetic penetration depth $\lambda$. The dependence of $\lambda^{-2}$ with temperature is consistent with the existence of a single $s$-wave energy gap in the superconducting state of $Ca_3Ir_4Sn_{13}$ with a gap value of 1.51(5) meV at absolute zero temperature. The magnetic penetration depth at zero temperature $\lambda(0)$ is 351(4) nm. The ratio $\Delta(0)/k_BT_c = 2.41(8)$ indicates that $Ca_3Ir_4Sn_{13}$ is a strong-coupling superconductor.

## 1. Introduction

The ternary intermetallic stannide compounds, $M_3Ir_4Sn_{13}$, where $M$ = Ca, Sr, etc. are of particular interest because they exhibit many exotic physical properties such as superconductivity, magnetic or charge order, and structural instabilities [3, 4, 5]. $Ca_3Ir_4Sn_{13}$ one of the most important material among these compounds shows a superconducting ground state below 7 K [1, 2]. Although this material was synthesized some 30 years ago, only very recently it has regained particular attention due to the possible coexistence or competition between superconducting and ferromagnetic spin fluctuation or charge density wave (CDW) states.

Resistivity and susceptibility measurements on $Ca_3Ir_4Sn_{13}$ show a broad peak like anomaly at $T^* \approx 33$ K. A similar anomaly has also been observed in the isoelectric sister compound $Sr_3Ir_4Sn_{13}$ at $T^* \approx 147$ K. Initially these $T^*$ anomalies were attributed to ferromagnetic (FM) fluctuation. The claim was made with the observation that the resistivity follows a non-Fermi liquid temperature dependence, but that the Fermi liquid behaviour can be restored by applying a high magnetic field [3]. Under an applied hydrostatic pressure, the $T_c$ of $Ca_3Ir_4Sn_{13}$ increases up to 4 GPa then falls for higher pressures [4].

Recently, single crystal x-ray diffraction studies [4] showed that the $T^*$ anomaly in $M_3Ir_4Sn_{13}$ ($M$ = Ca, Sr) is related to a second order superlattice transition from simple cubic parent phase, the $I$-phase, to a superlattice variant, the $I'$-phase, with a lattice parameter twice that of the $I$-phase. It has been further argued that this superlattice transition is associated with a charge density wave transition of the conduction electron system. At low temperatures, thermal conductivity and specific heat measurements indicate weakly correlated nodeless superconductivity in $Ca_3Ir_4Sn_{13}$ [5, 6]. On the other hand, recent $\mu$SR study on $Ca_3Ir_4Sn_{13}$ [7] determined a very high gap-to-$T_c$ ratio value $\Delta(0)/(k_B T_c) = 5$, which is unusually large even for a very strongly coupled BCS superconductor. In this contest, it is therefore important to reveal the true nature of the superconducting gap structure in $M_3Ir_4Sn_{13}$ ($M$ = Ca, Sr).

Here, we use the $\mu$SR technique as a microscopic local magnetic probe to investigate the magnetic properties of the superconducting ground state and determine the superconducting gap structures in $Ca_3Ir_4Sn_{13}$.

## 2. Experimental details

Single crystal samples of $Ca_3Ir_4Sn_{13}$ were prepared using a high temperature self-flux method [1, 2]. Sample characterization measurements were performed at the Brookhaven National Laboratory. The transverse-field (TF) and zero-field (ZF) $\mu$SR experiments were carried out at the $\pi$E1 and $\pi$E3 beam lines at the Paul Scherrer Institute (Villigen, Switzerland). The sample was cooled to the base temperature (1.6 K) at $H = 0$ during ZF-$\mu$SR experiments and in series of fields ranging from 2 mT to 0.5 T in TF-$\mu$SR experiments. The typical counting statistics were $\sim$ 8 million events per data point. The ZF and TF $\mu$SR data were analyzed by using the free software package MUSRFIT [8].

## 3. Results and discussion

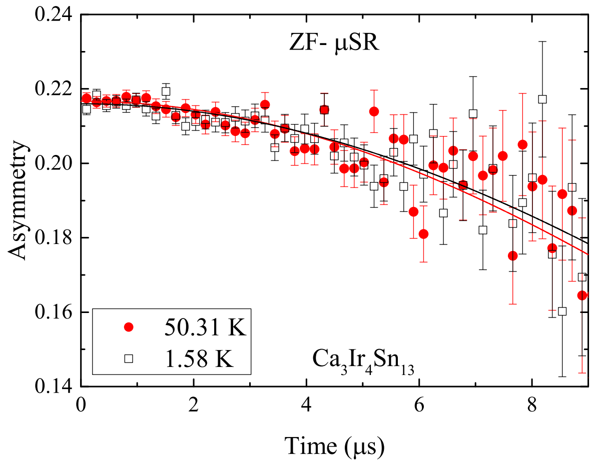


**Figure 1.** (Color online) ZF-$\mu$SR spectra of $Ca_3Ir_4Sn_{13}$ taken at 1.58 and 50.3 K. The solid lines are fits to the data using Eq. 1.

Figure 1 compares the ZF-$\mu$SR signals collected above $T^*$ and below $T_c$. Both signals are practically identical, indicating that no additional magnetic moments (either static or dynamic) appear below $T^*$ and also below $T_c$. ZF-$\mu$SR data can be well described using a Gaussian Kubo-Toyabe relaxation function, [9]

$$A(t) = A(0)\left\{\frac{1}{3} + \frac{2}{3}\left(1 - \sigma^2 t^2\right)\exp\left(-\frac{\sigma^2 t^2}{2}\right)\right\}, \quad (1)$$

where $A(0)$ is the initial asymmetry and $\sigma$ describes the muon spin relaxation rate due to the presence of static nuclear moments in $Ca_3Ir_4Sn_{13}$. The values of $\sigma$ obtained from the fits are 0.048(2) and 0.051(2) $\mu s^{-1}$ for 1.58 and 50.3 K, respectively.

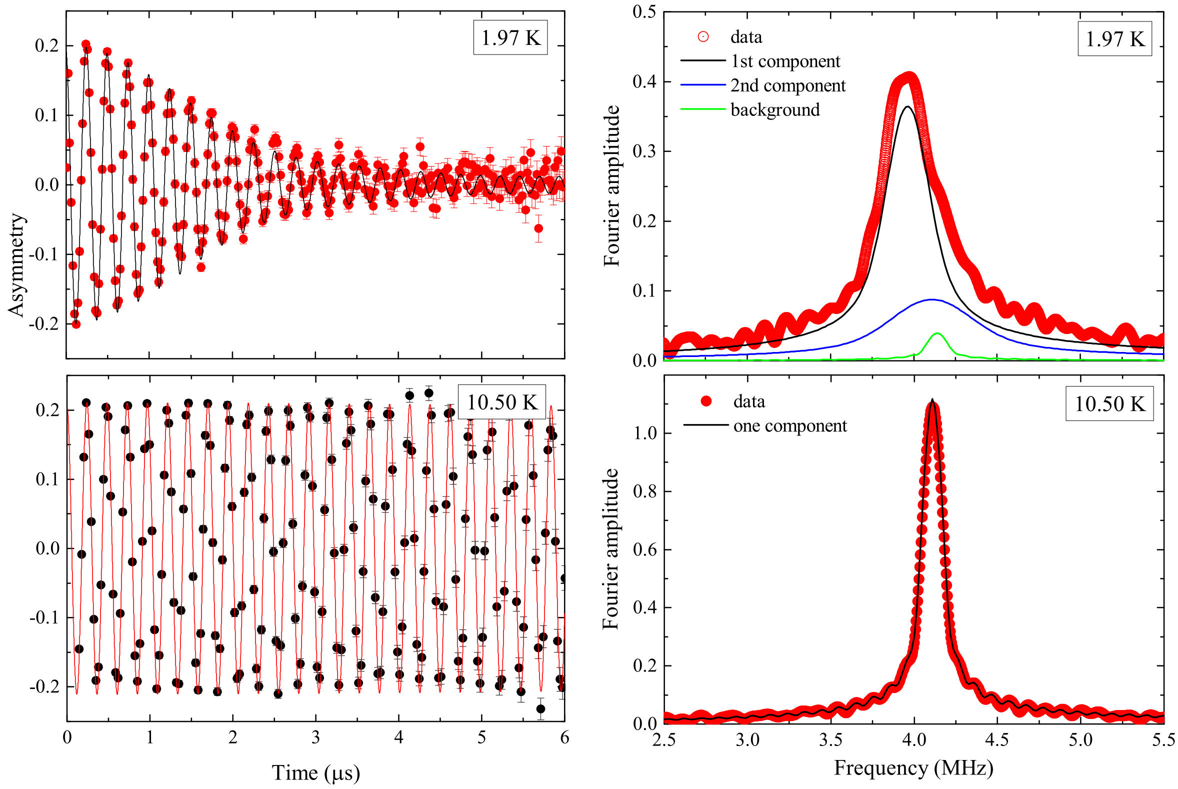


**Figure 2.** (Color online) (Left panel) TF-$\mu$SR signals for $Ca_3Ir_4Sn_{13}$ measured at 1.97 and 10.5 K in an applied field of 300 Oe. The solid lines are the fits to the data using Eq. 2. (Right panel) Corresponding TF spectra plotted in the frequency domain.

The left panel of Fig. 2 shows the TF-$\mu$SR precession signals of $Ca_3Ir_4Sn_{13}$ taken at 1.97 and 10.5 K in an applied field of 300 Oe. The right panel of Fig. 2 shows corresponding TF spectra plotted in the frequency domain. The signal in the normal state exhibit almost no damping due to homogeneous magnetic field distribution inside the sample. However, the signal taken at 1.97 K (below $T_c$) decays very quickly due to the inhomogeneous field distribution generated by the superconducting vortex lattice [10].

We can determine the second moment of the magnetic field distribution associated with the vortex state from the TF-$\mu$SR spectra. To do that the TF-$\mu$SR spectra were fitted using a multi-component Gaussian curve [11, 12]:

$$A(t) = \sum_{i=1}^{N} A_i \mathrm{exp}(-\sigma_i^2 t^2/2)\mathrm{cos}(\gamma_\mu B_i t + \phi) + A_{bg}\mathrm{cos}(\gamma_\mu B_{bg} t + \phi), \tag{2}$$

where $\phi$, $A_i$, $\sigma_i$, and $B_i$ are the initial phase, asymmetry, relaxation rate, and mean field (first moment) of the $i$th Gaussian component, respectively. $A_{bg}$ and $B_{bg}$ are the asymmetry and field, respectively due to the background contribution. We found that two Gaussian components ($N = 2$) are sufficient to fit the muon time spectra data. For $N = 2$, the first and second moments of $P(B)$ are given by

$$\langle B \rangle = \sum_{i=1}^{2} \frac{A_i B_i}{A_1 + A_2}, \tag{3}$$

and

$$\left\langle \Delta B^2 \right\rangle = \frac{\sigma^2}{\gamma_\mu} = \sum_{i=1}^{2} \frac{A_i}{A_1 + A_2} \left\{ (\sigma_i/\gamma_\mu)^2 + [B_i - \langle B \rangle]^2 \right\}, \tag{4}$$

where $\gamma_\mu = 2\pi \times 135.5388$ MHz/T is the muon gyromagnetic ratio and $\sigma$ the muon depolarization rate. Fig. 3 shows the temperature dependence of $\sigma$ of $Ca_3Ir_4Sn_{13}$ for an applied field of 300 Oe.

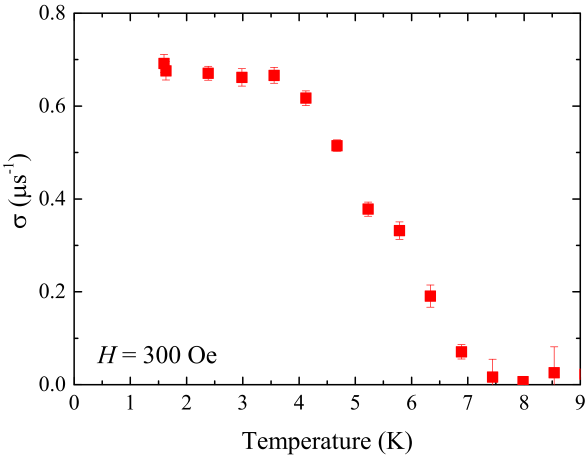


**Figure 3.** (Color online) Temperature dependence of the muon depolarization rate $\sigma$ in $Ca_3Ir_4Sn_{13}$ collected in an applied magnetic field of 300 Oe.

The superconducting contribution to $\sigma$ is obtained by subtracting the nuclear moment contribution (measured above $T_c$) as ${\sigma_{sc}}^2 = \sigma^2 - {\sigma_{nm}}^2$. For an isotropic type-II superconductor

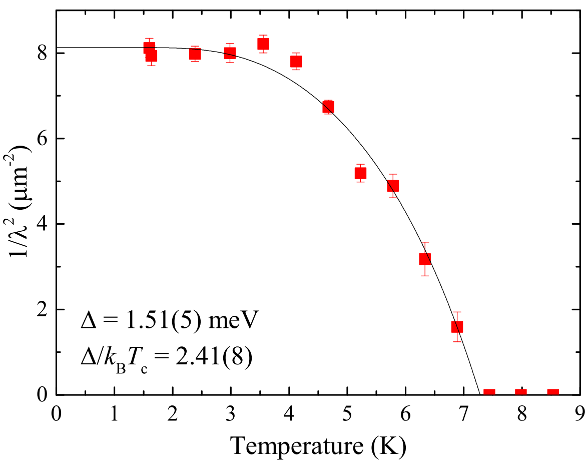


**Figure 4.** (Color online) The temperature dependence of $\lambda^{-2}(T)$. The solid line is a fit to the data using a single $s$-wave BCS model.

with a hexagonal Abrikosov vortex lattice described by Ginzburg-Landau theory, the magnetic penetration depth $\lambda$ is related to $\sigma_{sc}$ by the equation [10]:

$$\sigma_{sc}(b)[\mu\mathrm{s}^{-1}] = 4.854 \times 10^4 (1-b)[1 + 1.21(1-\sqrt{b})^3]\lambda^{-2}[\mathrm{nm}^{-2}], \tag{5}$$

Here $b = \langle B \rangle / B_{c2}$ is a reduced magnetic field. Fig. 4 shows the temperature dependence of $\lambda^{-2}$, i.e. the effective superfluid density, which is nearly flat below 4 K. This suggests that $Ca_3Ir_4Sn_{13}$ is a nodeless fully gapped superconductor. The solid line is a fit to the data using a single gap BCS $s$-wave model [13, 14]:

$$\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = 1 + 2\int_{\Delta(T)}^{\infty} \left(\frac{\partial f}{\partial E}\right) \frac{EdE}{\sqrt{E^2 - \Delta(T)^2}}. \tag{6}$$

Here $\lambda(0)$ is the zero-temperature value of the magnetic penetration depth, and $f = [1 + \exp(E/k_B T)]^{-1}$ is the Fermi function. We approximated the temperature dependence of the gap with $\Delta(T) = \Delta(0)\tanh\{1.82[1.018(T_c/T - 1)]^{0.51}\}$ [15]. The fit yields $\lambda(0) = 351(4)$ nm, and $\Delta(0) = 1.51(5)$ meV. The gap to $T_c$ ratio $\Delta(0)/k_B T_c = 2.41(8)$ is higher than the BCS value of 1.76, suggesting that $Ca_3Ir_4Sn_{13}$ is a strong-coupling superconductor. Our values of $\Delta(0)$ and gap-to-$T_c$ ratio obtained by a microscopic measurements are in good agreement with those obtained from specific heat measurements [16], whereas they are nearly 50 % lower than the values obtained in the previous $\mu$SR measurements by S. Gerber et al. [7].

## 4. Conclusions

In conclusion, ZF-$\mu$SR results do not find evidence of any magnetism in $Ca_3Ir_4Sn_{13}$ below $T^*$ as well as in the superconducting state. TF-$\mu$SR results indicate for a fully gapped superconducting state in $Ca_3Ir_4Sn_{13}$. $\lambda(T)$ can be well fitted using a single BCS $s$-wave model with gap value, $\Delta(0) = 1.51(5)$ meV and penetration depth, $\lambda(0) = 351(4)$ nm. The value of the gap to $T_c$ ratio, 2.41(8) is higher than the BCS value of 1.76 and suggests that $Ca_3Ir_4Sn_{13}$ is a strong-coupling superconductor.

## Acknowledgments

The $\mu$SR experiments were performed at the Swiss Muon Source (S$\mu$S), Paul Scherrer Institute (PSI, Switzerland). Work at Brookhaven is supported by the US DOE under Contract No. DE-AC02-98CH10886.

## References

[1] Remeika J, Esponosa G, Cooper A, Barz H, Rowell J, McWhan D, Vandenberg J, Moncton D, Fisk Z, Woolf L, Hamaker H, Maple M, Shirane G and Thomlinson M 1980 *Solid State Commun.* **34** 923
[2] Espinosa G P 1980 *Mater. Res. Bull.* **15** 791
[3] Yang J, Chen B, Michioka C and Yoshimura K 2010 *J. Phys. Soc. Jpn.* **19** 113705
[4] Klintberg L E, Goh S K, Alireza P L, Saines P J, Tompsett D A, Logg P W, Yang J, Chen B, Yoshimura K and Grosche F M 2012 *Phys. Rev. Lett.* **109** 237008
[5] Zhou S Y, Zhang H, Hong X C, Pan B Y, Qiu X, Dong W N, Li X L and Li S Y 2012 *Phys. Rev.* B **86** 064504
[6] Wang K and Petrovic C 2012 *Phys. Rev.* B **86** 024522
[7] Gerber S, Gavilano J L, Medarde M, Pomjakushin V, Baines C, Pomjakushina E, Conder K and Kenzelmann M 2013 *Phys. Rev.* B **88** 104505
[8] Suter A and Wojek B M 2012 *Physics Procedia* **30** 69
[9] Kubo R 1981 *Hyperfine Interact.* **8** 731
[10] Brandt E H 2003 *Phys. Rev.* B **68** 054506
[11] Weber M, Amato A, Gygax N, Schenck A, Maletta H, Duginov V N, Grebinnik V G, Lazarev A B, Olshevsky V G, Pomjakushin V Yu, Shilov S N, Zhukov V A, Kirillov B F, Pirogov A V, Ponomarev A N, Storchak V G, Kapusta S and Bock J 1993 *Phys. Rev.* B **48** 13022
[12] Maisuradze A, Khasanov R, Shengelaya A and Keller H 2009 *J. Phys.: Condens. Matter* **21** 075701
[13] Tinkham M 1975 *Introduction to Superconductivity* (New York: McGraw-Hill)
[14] Prozorov R and Giannetta R W 2006 *Supercond. Sci. Technol.* **19** R41
[15] Carrington A and Manzano F 2003 *Physica* C **385** 205
[16] Kase N, Hayamizu H and Akimitsu J 2011 *Phys. Rev.* B **83** 184509