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Abstract. The superconducting properties of the graphite intercalation compound CaC$_6$, available as high-quality $c$-axis-oriented polycrystals, were investigated through muon-spin spectroscopy measurements. An unconventional TF-µSR procedure, applicable to superconductors characterized by close-lying critical fields, was successfully adopted for measuring the muon-spin relaxation rates. Field-dependent measurements provide a value $\lambda_{ab}(0) = 62(4) \text{ nm}$ for the in-plane magnetic field penetration depth, $\kappa = 1.4(2)$ for the GL parameter, and hint at the presence of a slight gap anisotropy in the $ab$ plane. The dependence of relaxation on temperature confirms the s-wave character of CaC$_6$ superconductivity, with an average zero-temperature superconducting gap of 2.1(1) meV.

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1. Introduction

Despite many years of study, graphite and graphite intercalation compounds (GICs) continue to hold in store many surprises. Superconductivity in the range 6–12 K was discovered in YbC$_6$ [2], CaC$_6$ [2, 3], and Li$_3$Ca$_2$C$_6$ [4]. Even though the pristine, semi-metallic graphite is not a superconductor, the intercalation process induces a negative charge transfer towards graphene sheets, resulting in partially filled graphite $\pi$-bands. Depending on the intercalant species, different degrees of band hybridization between the donor s-band and host graphite $\pi$-bands are possible, a fundamental fact for the occurrence of superconductivity in these compounds. In the case of first-stage binary GICs, the dramatic increase in the superconducting critical temperature (from 1.9 K for LiC$_2$ to 11.5 K for CaC$_6$) has been ascribed to a higher degree of hybridization of the s-band in alkaline-earth metals with the host’s $\pi$-band and, therefore, to an increased coupling between the out-of-plane C phonons and the metal’s s-band electrons [5, 6]. This picture could become more complex in the first-stage ternary GICs, such as e.g. Li$_3$Ca$_2$C$_6$, characterized by poly-layered (five lithium and two calcium) intercalated sheets [4]. Both CaC$_6$ and Li$_3$Ca$_2$C$_6$ have been characterized as type-II superconductors with slightly anisotropic critical fields [3, 4]. In particular, CaC$_6$ has been extensively studied by means of low- and high-frequency electrodynamic response [7, 8], scanning tunnelling microscopy and spectroscopy (STM/STS) [9], tunnel junction [10] and point-contact Andreev-reflection (PCAR) spectroscopy [11], dc resistivity [12, 13] and magnetization [14, 15] versus temperature and pressure, specific heat [16] and isotope effect [17], Raman [18]–[20] and angle-resolved photoemission spectroscopy (ARPES) [21], far-infrared reflectivity (FIR) [22], small-angle neutron scattering [23] and conduction electron spin resonance (CESR) [24]. From the experimental evidence emerges a coherent picture favoring a Bardeen–Cooper–Schrieffer (BCS) s-wave conventional superconductivity, even though the exact value of the superconducting gap seems to depend critically on the sample quality and varies significantly with the employed technique. Currently, the large spread in the measured gap values, ranging from 1.35 to 2.3 meV, questions strongly the initial consensus on an isotropic, weakly coupled system. In contrast, the above-mentioned distribution of superconducting gaps was indeed predicted by ab initio calculations [25] and justified as due to a moderately anisotropic gap.
In this work, we describe the use of the muon-spin rotation ($\mu$SR) technique [26] in a detailed investigation of the binary CaC$_6$ compound. Implanted muons can randomly sample the vortex-induced field modulation, with a spin dephasing that correlates directly with the magnetic penetration depth. By measuring the muon-spin relaxation rate $\sigma$ as a function of temperature and applied field, one can obtain information on the symmetry of the superconducting gap and, consequently, on the pairing mechanism [27, 28]. In fact, due to the proportionality relation $\sigma \propto n_s \propto \lambda^{-2}$, which connects an experimentally accessible quantity with the superfluid density and the magnetic penetration depth, one can investigate the low-lying quasiparticle density of states under different experimental conditions. The extreme sample sensitivity to air exposure, associated with a narrow range of upper and lower critical fields, makes the investigation of this class of compounds quite challenging, and this has limited $\mu$SR studies to very few attempts [29]. In the following, we discuss the results of temperature- and field-dependent $\mu$SR measurements, carried out on $c$-axis-oriented polycrystals of CaC$_6$. By investigating the in-plane superconducting properties, their $T$ and $H$ evolution, we could independently determine important SC parameters, such as the gap value and the nature of the pairing symmetry.

2. Experimental details

High-quality, high-purity, $c$-axis-oriented CaC$_6$ samples were prepared by immersion (for 10 days) of platelets of highly oriented pyrolytic graphite (HOPG) in a molten lithium–calcium alloy under very pure argon atmosphere (see [3, 30, 31] for details). Like the pristine graphite, also the resulting samples were polycrystalline [32], with the $c$-axes of the crystallites parallel to each other, whereas the orientation of the $ab$-planes was random. X-ray diffraction patterns of the binary GICs show clear and intense (00$l$) Bragg peaks. No residual reflections from the pristine graphite could be observed, and the calculated intensities fully agreed with those reported in the above-cited references. Due to the simple structure of the binary compound, its structural characterization readily confirmed its high degree of homogeneity. The unavoidable stacking faults could at most cause a very small degree of disorder.

Due to the high reactivity towards oxygen and moisture, great care was taken always to keep the samples under a controlled atmosphere (extremely pure argon), even during the $\mu$SR measurements. To this end, a specially designed kapton sample holder, as shown in figure 1, was
Figure 2. Magnetization versus temperature plot for CaC$_6$. After cooling in the absence of fields (ZFC), the measurement was carried out upon warming the sample in a constant magnetic field of $\mu_0 H = 0.5 \text{ mT}$.

realized. It consists of an inner square-shaped frame with two 76 $\mu$m thick cover sheets (front and rear), where a $\sim 6.5 \times 6.5 \text{ mm}^2$ sample mosaic can be hosted (see figure 1). To further protect the sample from degrading, the whole assembly was placed inside an aluminized mylar capsule (not shown) and sealed under argon atmosphere. An aluminium frame with a central hole 19 mm in diameter was used to centre the sample with respect to the incoming muon beam (whose nominal diameter is approximately 6 mm full-width at half-maximum (FWHM)). This configuration also provides a good signal-to-noise ratio, since the fraction of muons which miss the sample and stop in the inner frame is relatively small ($\approx 30\%$, vide infra). After the experiment, the sample was checked again by means of magnetometry and x-ray diffraction: no sample degradation due to possible redox reactions with oxygen or moisture during the sample transport and $\mu$SR measurements could be detected.

3. Dc magnetization measurements

The superconducting critical temperature and transition width for the binary CaC$_6$ samples were measured using a dc superconducting quantum interference device (SQUID) magnetometer (Quantum Design). Figure 2 shows the zero-field-cooling (ZFC) magnetization as a function of temperature. The ZFC measurement exhibits a sharp drop in the magnetization just below 11.5 K. The strong diamagnetic signal, which reaches complete saturation right after the superconducting transition temperature, is indicative of a true Meissner effect. The midpoint of the ZFC curve occurs at 11.1 K, resulting in a total transition width $\Delta T = 0.2 \text{ K}$. By taking into account the geometrical demagnetization factor and the estimated sample volume, a superconducting shielding fraction of $\sim 0.86 \pm 10\%$ could be inferred.

4. Transverse-field muon-spin rotation

TF-$\mu$SR experiments were carried out at the GPS spectrometer ($\pi M3$ beam line) of the Paul Scherrer Institut, Villigen, Switzerland. The relatively large sample thickness ($\geq 0.3 \text{ mm}$) and
Figure 3. (a) For $H_{c1} \ll H_{c2}$, a standard fixed-field scan is used to measure $\sigma(T)$. (b) For $H_{c1} \lesssim H_{c2}$, a ‘zigzag’ path is needed, made of piece-wise sequences of field cooling (horizontal dashed line), followed by a measure scan on heating, at the same field (full line, displaced for clarity), and by a field change (vertical dashed line). The plot is for a ratio $H_{c2}/H_{c1} = 1.5$; the actual value for CaC$_6$ is in the range of 2.5–3.

the use of veto counting (which accounts for only those muons effectively stopped in the sample) both contribute to a significantly improved signal-to-noise ratio. In this configuration, a total asymmetry of about 0.15–0.18 was obtained, still low if compared with the normal asymmetry value expected at the GPS instrument (0.27). The difference can be attributed to muons stopping in kapton, whose asymmetry (apart from a small residual fraction) is strongly suppressed by a delayed radical formation mechanism.

TF-$\mu$SR measurements were carried out by applying a constant magnetic field parallel to the muon momentum, while the muon spin was rotated to form an angle of $\sim 50^\circ$ with momentum. Samples could be oriented in such a way that the magnetic field direction was orthogonal to the graphene planes. This configuration provides straightforward access to the in-plane superconducting properties (hereafter denoted with the subscript $ab$).

Usually, in TF-$\mu$SR measurements, samples are first cooled from above $T_c$ down to the lowest temperature $T_{\text{min}}$ at a certain applied field (field cooling). Then, by keeping the field fixed, a temperature scan from $T_{\text{min}}$ up to $T_c$ is performed (see figure 3(a)). However, this standard procedure proved problematic for the particular samples under test. Indeed, as shown schematically in figure 3(a), in compounds with an extended mixed Meissner state (i.e. with very different low and upper critical fields), a constant-field temperature scan is sufficient to measure $\sigma = \sigma(T)$ over the whole SC temperature range. Unlike the majority of superconductors, CaC$_6$ has an upper critical field $H_{c2}(0)$ that is merely three times its lower critical value [3], hence implying a very narrow mixed state region. By following a constant-field path (top solid line in figure 3(b)), one ends up in the normal state already at temperatures of the order of $T_c/2$.

To solve this problem, we have considered a zigzag temperature scan sequence, where the field is kept piecewise constant and changed in discrete steps around the average value $(H_{c1}^2 + H_{c2}^2)/2$ (figure 3(b)). The temperature variation in the upper and lower critical fields was taken from measurements on samples from the same batch [3]. Each constant step was measured as in the standard case, except when the field is changed, where the sample is heated up to the normal state to avoid remanent fields and then cooled again in the new applied field.
Figures 4(a)–(c) show the muon spin precession signal in our sample. The difference in the relaxation rates above and below $T_c$ is due to the appearance in type-II superconductors of the flux line vortex lattice (FLL), which leads to a spatially inhomogeneous magnetic field induction. In particular, we note that the muon-spin precession signal for $T < T_c$ consists of a superposition of two distinct terms: a fast relaxing component, originating from the superconducting sample, and a small temperature-independent and non-relaxing component, due to a residual fraction of muons stopping outside the sample.

By performing a fast Fourier transform (FFT) of the experimentally measured asymmetry, $A(t)$, one obtains a straightforward picture of the internal magnetic field distribution $P(B)$, both below and above $T_c$, as shown in figures 4(d)–(f). The time evolution of the spin polarization of the muon ensemble, $P(t) = A(t)/A_0$, and its field counterpart, $P(B)$, are related by

$$P(t) = \int P(B) \cos(\gamma \mu B t + \phi) \, dB,$$

with $A_0$ being the maximum value of asymmetry, $\gamma \mu = 2\pi \times 135.53 \text{ MHz T}^{-1}$ the muon gyromagnetic ratio and $\phi$ the initial muon-spin precession angle. In the normal state, CaC$_6$ shows a single line at the position of the externally applied magnetic field $B_{\text{ext}}$, with resolution-limited vanishing relaxation (figure 4(f)).

In the superconducting state, the presence of the FLL vortices induces a typical asymmetric field distribution at low temperatures (figure 4(d)), with a large diamagnetic shift of the average.
internal field $B_i$ ($B_i - B_{\text{ext}} < 0$). The absence of the sharp features expected in the ideal FLL lineshape indicates a moderate effect of FLL disorder.

However, when varying the field and temperature of the experiment, as e.g. along the path sketched in figure 3, the lineshape rapidly evolves to a more symmetric shape (see e.g. figure 4(e)). A consistent analysis of the temperature dependence of the $\mu$SR time-dependent asymmetry (figure 4(a)), after subtraction of the temperature independent small residual contribution described above, is therefore only possible with the following simple Gaussian model,

$$A(t) = a \cdot \exp(-\sigma^2 t^2/2) \cos(\gamma \mu B_i t + \phi). \quad (2)$$

Here, $a$ is the muon-spin precession amplitude and $\sigma$ the relaxation rate. We note that even at $T = 1.5 \text{ K}$, $B_{\text{ext}} = 100 \text{ mT}$, the $\chi^2$ improvement introduced by a second Gaussian component, to account for the non-symmetrical peak shape of figure 4(d), is negligible, as can be seen from the rather good single Gaussian fit in figure 4(a).

5. Results and discussion

5.1. Field dependence of the muon asymmetry relaxation rate

In a superconductor, the relaxation rate of the decaying signal is due to the internal field modulation arising from the FLL. Based on a general Ginzburg–Landau approach, which takes into account also vortex overlap effects at high applied fields, one can express $\mu$ as a function of the reduced internal magnetic field $b = B/B_c$ and of the Ginzburg–Landau parameter $\kappa = \lambda/\xi$. In particular, a good approximation [33] for all $\kappa$ and for $0.25 < b < 1$ is given by

$$\sigma = \frac{4.83 \times 10^4 \cdot \kappa^2 \cdot (1 - b)}{\kappa^2 - 0.069} \cdot \lambda^{-2} \text{ (nm)} \quad (3)$$

(note the different use of symbols here\(^8\) and in [33]).

From systematic measurements of the $\mu$SR depolarization rate $\sigma$ as a function of the applied magnetic field at a fixed temperature ($T = 1.5 \text{ K}$), we could extract the zero-temperature values for both the critical field $B_c(0)$ and for the in-plane magnetic penetration depth $\lambda_{ab}(0)$. Figure 5 shows the field dependence of $\sigma$, together with the fit to equation (3), that yields as best fit values $B_c(0) = 180(20) \text{ mT}$ and $\lambda_0 = 62(1) \text{ nm}$. The latter is $\sim 20\%$ lower than that reported by inductance measurements [7] on samples of the same origin. The value of the upper critical field $B_c(0)$ instead is in very good agreement with previous dc magnetization measurements [3]. A direct check on a small crystal from our sample, in the same field configuration of the $\mu$SR experiment, confirms these findings, as can be seen by inspection in figure 6.

The upper critical field yields directly the zero-temperature limit of the GL coherence length, $\xi_{ab}(0) = (\Phi_0/2\pi B_c^2)^{1/2} = 43 \pm 5 \text{ nm}$, where $\Phi_0$ is the quantum of magnetic flux. By combining this value with our best estimate for $\lambda_{ab}$, we obtain a GL parameter value $\kappa = \lambda_{ab}/\xi = 1.43 \pm 0.16$, also in good agreement with other techniques, which suggest CaC$_6$ to be an intermediate-$\kappa$ superconductor.

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\(^8\) The conversion factor $a = 4.83 \times 10^4$ is obtained from the coefficient $a' = 7.52 \times 10^{-4}$, relating $\langle (\Delta B)^2 \rangle$ with $\lambda^{-4}$ in the original formula (10) in [33], by recalling that $\sigma^2 = \gamma_\mu^2 \langle (\Delta B)^2 \rangle$. This gives $a = \sqrt{a'} \Phi_0 \gamma_\mu$, with $\Phi_0 = h/2e = 2.07 \times 10^{-15} \text{ Tm}^2$ being the quantum of magnetic flux and $\gamma_\mu$ the muon gyromagnetic ratio (see also [28]).

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Figure 5. Field dependence of the $\mu$SR depolarization rate $\sigma$ at $T = 1.5$ K. The solid line represents a best fit to the data using using equation (3), with $\lambda$ taken as a constant. The inset shows the penetration depth $\lambda$ versus $b$, again using equation (3), but with $\lambda$ allowed to vary to give a better fit to the field dependence. The straight line represents a linear fit to the resulting field dependence of $\lambda$.

Figure 6. Temperature dependence of $B_{c2}$ for CaC$_6$. The solid line is a fourth-order polynomial fit.

Once vortex interaction effects have been taken into account by using Brandt’s theory [33] (see equation (3)), any further field dependence in the extracted penetration depth can reveal possible deviations from the simple BCS picture towards unconventional or anisotropic superconductivity [28], [34]–[36]. If nodes or a highly anisotropic SC gap were present, the density of quasiparticles would be different from zero even at low temperatures. In such a case, the Doppler shift of the quasiparticles’ momentum can induce a linear variation in $\lambda$ with the
applied magnetic field [28], [34, 35]. In particular, the slope of the magnetic penetration depth versus the normalized field \( b = B / B_{c2} \),

\[
\eta = \frac{1}{\lambda(0)} \frac{d\lambda(b)}{db},
\]

is close to zero for isotropic superconductors, but it is much larger, up to a factor 6, in the anisotropic case [35]. From the relaxation rate \( \sigma \) measured at different field values, one can calculate \( \lambda_{ab}(b) / \lambda_{ab}(0) \) by means of equation (3), as shown in the inset of figure 5. We obtain \( \eta \sim 0.38(8) \), which may suggest a slight gap anisotropy in the \( ab \)-plane [35], as predicted in [25]. Note that the zero-field extrapolation value \( \lambda_{ab}(0) \) is somewhat lower than the value obtained from the fit of equation (3) and their half-difference (4 nm) may be taken as a conservative error bar on this quantity.

5.2. Temperature dependence of the in-plane superfluid density

The temperature dependence of the depolarization rate, \( \sigma = \sigma(T) \), is used to extract the normalized superfluid density \( n_s(T) \). For magnetic induction values such that \( b > 0.1 \), the vortex overlap becomes non-negligible; consequently, both individual vortex properties and the vortex density become \( b \) dependent [33, 37]. This leads to a field dependence of the scaling coefficient relating \( \sigma \) with \( \lambda^{-2} \), as is evident from the pre-factor in equation (3). This equation, applicable in a large range of magnetic fields \( (0.25 < b < 1) \) and for any value of \( \kappa \), shows that to obtain the behaviour of \( \lambda^{-2}(T) \), the temperature dependence of the upper critical field, \( B_{c2} = B_{c2}(T) \), is required. We extract it from a polynomial fit to the reversible magnetization data of figure 6, taken in the same geometry as the \( \mu \)SR measurements (i.e. with the applied magnetic field parallel to the crystallographic \( c \)-axis).

The temperature dependence of the normalized superfluid density \( n_s(T) = [\lambda_{ab}(0, B) / \lambda_{ab}(T, B)]^2 \) is shown in figure 7. The special zigzag procedure we follow (see figure 3(b)) introduces a systematic error, since \( n_s \) is evaluated using slightly different \( b \) values at each step. This error, however, is small, since the field dependence of the magnetic penetration for \( b \sim 0.5 \pm 0.2 \) is weak \( (\lambda_{ab}(b) / \lambda_{ab}(0) \simeq 1.04 \pm 0.04 \) from the inset of figure 5), resulting in an additional average uncertainty of \( \sim 8\% \) on the superfluid density.

The \( n_s \) data may be compared with the local BCS theory in the weak-coupling limit (locality follows from \( \lambda > \xi \), since \( \kappa = 1.43 \)). The electronic mean free path \( l \) distinguishes three different regimes: clean \((\xi < l)\), intermediate \((\xi \approx l)\) and dirty \((\xi > l)\), as discussed in the appendix. The experimental data shown in figure 7 are best fitted by the dirty limit curve equation (A.3), with the \( T = 0 \) value of the in-plane superconducting gap, \( \Delta_{ab}(0) = 2.1 \pm 0.1 \) meV, as the only free parameter. A clean limit calculation with the same value of the zero-temperature superconducting gap is shown for comparison and it deviates systematically from the data points. A fit of the same data with \( \xi_{ab}(0)/l \) as a free parameter [38] also favours the dirty limit. This picture is further supported by CESR results in the normal state [24], which suggest that the scattering of conduction electrons is dominated by impurities. In contrast, very recent \( \mu \)SR data [29] point to the clean limit. A difference between two CaC\textsubscript{6} specimens grown by similar methods is, however, not entirely surprising, since GICs are characterized by a nanocrystalline in-plane texture. This texture depends strongly on the nature of the pristine graphite and on the thermodynamic growing conditions [32] and, together with Ca vacancies, dislocations and stacking faults [39], it may affect the mean free path. Incidentally, the same
structural texture is probably at the origin of the FLL disorder, apparent from the muon lineshape.

The gap value extracted from our µSR data is in good agreement with tunnelling junction spectroscopy data [10], but slightly higher than that measured by inductance methods [7] on samples of the same origin. We note that, whereas surface degradation issues may affect to different extents the latter technique, implanted muons probe the bulk and are more reliable in this respect. Recent ARPES measurements [21] have evidenced two gaps in CaC₆, with values of 0.2 and 1.8–2.0 meV, and our results seem to agree with the larger one (the much smaller gap would become visible only in very low temperature nₛ data). Furthermore, our estimate of the ratio $2\Delta/k_B T_c$ is equal to 4.3(2), a value substantially larger than the standard BCS expectation (3.5), indicating deviations from the BCS weak coupling towards a strong coupling regime [40]. Our result is in agreement with part of the literature, as for instance the previously mentioned tunnelling spectroscopy [10] and ARPES [21] data. Interestingly, also magnetoresistance measurements [13] find an electron–phonon-coupling constant $\lambda \sim 1.1$. All of these results most likely place this material in the intermediate-to-strong coupling regime.

6. Conclusion

The superconducting properties of the layered graphite intercalation compound CaC₆ were investigated using muon-spin rotation spectroscopy. Standard analysis confirms that this system is a conventional BCS s-wave superconductor in the dirty limit. The origin of the observed dirty limit may be related to the presence of structural defects, characteristic of the pristine HOPG, or introduced during the intercalation process. We could not obtain direct evidence of multiple gaps down to 1.5 K, but from the field dependence of muon-spin relaxation we found indications of a small gap anisotropy. Moreover, we provide additional support for considering this compound in between the intermediate and the strong coupling limit.
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Appendix

In the local, clean limit \((l > \xi)\), the London theory is valid, with the temperature dependence of the superfluid density for a superconducting gap with s-wave symmetry being given as \([41]\)

\[
\left[ \frac{\lambda_L(0)}{\lambda_L(T)} \right]^2 = 1 - 2 \int_0^\infty \frac{\partial f}{\partial E} \left( \frac{E}{E^2 - \Delta^2} \right)^{1/2} dE. \tag{A.1}
\]

Here, \(f = \left[ 1 + \exp(E/k_B T) \right]^{-1}\) is the Fermi distribution and \(\Delta(T)/\Delta(0) = \left[ \cos(\pi/2(T/T_c)^2) \right]^{1/2}\) is an analytical approximation of the temperature dependence of the superconducting gap in the weak coupling limit \([42]\).

When the mean free path is of the same order of magnitude of the superconducting coherence length \((l \simeq \xi)\), the London penetration depth \(\lambda_L\) is described \([41]\) by the relation

\[
\lambda_L(T) = \lambda(T) \left[ 1 + \frac{\xi(0)}{J(0, T) l} \right]^{-1/2}, \tag{A.2}
\]

where \(\lambda(T)\) is the measured magnetic penetration depth and \(J(0, T)\) is the real-space kernel valid for a local electrodynamic response.

Finally, in the dirty limit \((l < \xi)\), following Tinkham \([41]\), equation (A.2) becomes

\[
\left[ \frac{\lambda(0)}{\lambda(T)} \right]^2 = \frac{\Delta_{ab}(T)}{\Delta_{ab}(0)} \tanh \left[ \frac{\Delta_{ab}(T)}{2k_B T} \right]. \tag{A.3}
\]

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