Local Magnetic Measurements of Trapped Flux Through a Permanent Current Path in Graphite

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Temperature and field dependent measurements of the electrical resistance of different natural graphite samples, suggest the existence of superconductivity at room temperature in some regions of the samples. To verify whether dissipationless electrical currents are responsible for the trapped magnetic flux inferred from electrical resistance measurements, we localized them using magnetic force microscopy on a natural graphite sample in remanent state after applying a magnetic field. The obtained evidence indicates that at room temperature a permanent current flows at the border of the trapped flux region. The current path vanishes at the same transition temperature \( T_c \approx 370 \) K as the one obtained from electrical resistance measurements on the same sample. This sudden decrease of the phase is different from what is expected for a ferromagnetic material. Time dependent measurements of the signal show the typical behavior of flux creep of a permanent current flowing in a superconductor. The overall results support the existence of room-temperature superconductivity at certain regions in the graphite structure and indicate that magnetic force microscopy is suitable to localize them. Magnetic coupling is excluded as origin of the observed phase signal.

I. INTRODUCTION

The usual way to probe the existence of superconductivity in a material is by measurement of zero resistance and magnetic flux expulsion below a critical temperature. Experimentally, it is a challenge to prove the existence of superconductivity in very small or in granular regions of a macroscopic sample because typical experimental methods, trying to show nominally zero electrical resistance and/or magnetic flux expulsion, are not well suitable. This is the case where granular superconducting regions are localized within embedded two-dimensional (2D) interfaces, at which no easy access for direct electrical contacts to the regions of interest is possible. Moreover, if the size of the superconducting regions is much smaller than the effective London penetration depth \( \lambda_L \), in addition to the large demagnetization effects expected for 2D interfaces, the flux expulsion, i.e. the Meissner effect, is, strictly speaking, negligible. In any case, a true zero resistance cannot be measured using standard electric current/voltage measurements simply because this would imply using devices with infinite sensitivity.

An alternative proof for the existence of superconductivity can rely on the observation of dissipationless currents that maintain a magnetic flux trapped at certain regions of a sample. Early works using magneto-optical flux imaging (MOI), determined the spatial distribution and the magnitude of the supercurrents in high temperature superconducting YBCO crystals. Recently published results\cite{11} suggest that some regions in natural graphite samples show a superconducting-like transition at unexpected high transition temperatures of \( T_c \approx 370 \) K. The observation of Bragg peaks in X-ray diffraction (XRD) measurements, which correspond to two possible stable stacking orders, i.e. rhombohedral and Bernal graphite, suggests, that the interfaces are the regions where high-temperature superconductivity can be localized\cite{15,20,21} due to the existence of flat bands, which was predicted in theoretical work\cite{51,53}.

The record temperature for superconductivity at 203 K reported recently in a sulfur hydride system at high pressure\cite{5} seems to be consistent with the Bardeen-Cooper-Schrieffer (BCS) theory. A Van Hove singularity was suggested as a possible reason for high-temperature superconductivity\cite{5}. The resulting Khodel-Shaginyan flat bands\cite{41} with dispersionless energy relation\cite{20,21} may lead to superconductivity at high temperatures. Such flat bands also exist at the surface of rhombohedral graphite\cite{15,21,24} or at the interfaces between rhombohedral and Bernal graphite\cite{27}. The low critical temperature for usual superconductors is a result of the exponential suppression in the BCS equation for quadratic dispersion relations. In the case of flat bands, a critical temperature orders of magnitude larger can be expected, assuming similar Cooper pair interaction strengths. In such a case, the critical temperature is proportional to the pairing interaction strength and to the area of the flat band in momentum space. Thus, at certain interfaces between rhombohedral and Bernal graphite\cite{15,21,24}, twisted Bernal layers\cite{11}, or regions under strain\cite{13}, superconductivity with a critical temperature at or above room temperature might be triggered. In the last 43 years, hints for the existence of superconductivity at very high temperatures in graphite-based samples were reported\cite{12,21,24,25}.

In this study we used local magnetic force measurements to find the region of trapped magnetic flux, due to a permanent current, in a natural graphite sample. The temperature dependence and time decay of the phase signal was monitored. The obtained results rule out magnetic order as origin and support the existence of superconductivity at room temperature at certain regions of graphite.
II. SUPERCONDUCTIVITY AND ZERO RESISTANCE

Superconductors exhibit characteristic properties, such as ideal diamagnetism, quantized magnetic flux lines or the vanishing of the electrical resistance. In this study, the main attention will be paid to the latter feature, because it is almost impossible to measure the first two effects on bulk samples where superconductivity is confined to some interfaces embedded in the bulk material. On one hand, the quasi-two dimensional (2D) superconductivity at interfaces, and due to the huge demagnetization factor, impedes to a very large extent the expulsion of a field applied normal to the superconducting area. On the other side, the effective penetration depth $\Lambda = 2\lambda_L^2/d_i$[28], where $\lambda_L$ is the London penetration depth and $d_i$ the thickness of the superconducting interface, gets easily larger than the sample size, making a direct imaging of a single vortex impossible.

a. Zero electric resistance Since superconductivity was observed first in mercury, the magnitude of the decrease of the resistance, when crossing over to the superconducting state, was an important question. At that time, standard methods were used to measure the electrical resistance, i.e. the voltage drop was monitored across a current carrying wire. Hence, it was only possible to state that the resistance dropped below the sensitivity limit of the measurement device, which makes it in principle impossible to prove, even nowadays, that the resistance vanishes and it is exactly equal to zero. In 1911, Kammerlingh-Onnes reported that “while the resistance at 13.9 K is still 0.034 times the resistance of solid mercury extrapolated to 0°C, at 4.3 K it is only 0.00225, while at 3 K it falls to less than 0.0001[29]. New experiments later that year showed that between 4.21 K and 4.19 K the resistance dropped from 0.115 $\Omega$ to less than $10^{-5} \Omega$.

Dealing with the problem of measuring very small resistances, already in 1914 Kammerlingh-Onnes used a technique which is superior to standard resistance measurements. He measured the decay time of an induced current in a closed superconducting loop made of lead. For this purpose, at first the ring is held in the normal state, i.e. above the transition temperature $T_c$. A permanent magnet was used to apply a magnetic field, inside the ring. The ring was then cooled below $T_c$ to 1.8 K, the magnetic field inside the superconducting ring remains unchanged. The magnet was removed, thus inducing a current. Kammerlingh-Onnes used a compass needle placed close to the superconducting ring in order to measure any changes in the magnetic field and thus, in the current flowing through the superconducting ring. He then reported that within an hour, the current (0.6 A) did not decrease, indicating that the resistance has zero value and that the current would continue to flow as permanent current.

In this way, it is possible to estimate a new upper limit for the resistance, which is much more accurate compared to the electrical resistance measurements[21] With $U_i$ being the induced voltage, the self-induction $L$ can be defined as $U_i = -L(dI/dt)$, and the stored energy of a ring with permanent current is $(1/2)LI^2$ $(dP = LI(dI/dt))$. The change of this energy with time is equal to the heating power $RI^2$, thus

$$-LI\frac{dI}{dt} = RI^2.$$  (1)

Hence $-(dI/d_{at}) = (R/L)I$ with solution:

$$I(t) = I_0 \exp \left(-\frac{Rt}{L}\right),$$  (2)

where $I_0$ is the initial current at time $t = 0$. This implies that the decay of the current depends on the shape of the superconducting loop. For a circular ring, the self-induction is given as $L = \mu_0r[\ln(8r/d) - 1.75]$. Assuming that the radius of the ring is $r = 300 \mu$m, the radius of the wire $d_w = 0.5 \mu$m, and that the current decreases less than 1 % per hour, then the resistance must be smaller than

$$R \leq \frac{-\ln(0.99) \cdot 2.53 \mu H}{3600 s} \approx 7.1 \cdot 10^{-15} \Omega.$$  (3)

This shows that, monitoring the magnetic fields created by a permanent current is much more accurate to estimate an upper limit of the resistance, compared to standard measurements of the electrical resistance. In the first experiments a compass needle was used, later more sensitive methods were employed, such as torsion thread experiments or magnetic force microscopy. However, in some cases the resistance does not vanish, e.g. small currents if magnetic flux lines exist or alternating currents.

III. MAGNETIC FORCE MICROSCOPY

a. Basic Principles In FIG.1 the basic principle of magnetic force microscopy (MFM) is shown. The sample, consisting of magnetic domains, emanates magnetic stray fields, which are detected by a magnetized tip. In conventional MFM devices, a laser is used to detect the oscillations of the cantilever, however, self-sensing and self-actuating cantilevers are also used[30]. Assuming the tip as a point dipole, then the force acting on the tip is given by

$$\vec{F} = \mu_0 \nabla \left(\vec{m} \cdot \vec{H}\right),$$  (4)

where $\mu_0$ is the magnetic permeability of free space, $\vec{m}$ is the magnetic moment of the tip and $\vec{H}$ the magnetic stray field from the sample at the position of the tip.

b. Measurement technique In MFM, non-contact mode is used to detect the stray fields, i.e. the distance between tip and sample surface is kept at a constant value. To do so, for each line scan, the topography is
measured using tapping mode prior to the actual non-contact measurement. In tapping mode, the cantilever is oscillating, and the tip is deflected when encountering the sample surface. A line scan is performed and thus the topography of the sample is measured. In a second, non-contact scan, the same line is followed but with the tip kept at constant distance $d$.

Assuming that the tip is a point dipole, the cantilever is parallel to the sample surface and that the tip and sample are independent of each other, then the force and force derivative are

$$F = m_x \frac{\partial B_x}{\partial z} + m_y \frac{\partial B_y}{\partial z} + m_z \frac{\partial B_z}{\partial z}, \quad (5)$$

$$F' = m_x \frac{\partial^2 B_x}{\partial z^2} + m_y \frac{\partial^2 B_y}{\partial z^2} + m_z \frac{\partial^2 B_z}{\partial z^2}, \quad (6)$$

where $m_i, i = x, y, z$, are the effective magnetic moments of tip. Assuming further an infinitely long dipole tip, and if the tip magnetization is perfectly aligned along $z$, then

$$F = m_z \frac{\partial B_z}{\partial z}, \quad (7)$$

$$\varphi = -\frac{Q}{k} \frac{\partial F}{\partial z} = -\frac{Q}{k} \left( m_z \frac{\partial^2 B_z}{\partial z^2} \right), \quad (8)$$

where $Q$ and $k$ are the quality factor of the tip resonance peak and spring constant, respectively.

c. Experimental Details A commercial tip ($k \approx 3$ N/m) with a Cobalt-Chromium coating and a nominal tip radius of 35 nm was used, which was placed in the MFM device (Nanoscope). The graphite sample was fixed on a substrate (Si with 150 nm SiN layer) using varnish. The contacts (four terminal sensing) for the electrical resistance measurement were done using silver paste and gold wires. The resistance measurements were carried out in a magneto-cryostat with a temperature stabilization of a few mK at 300 K, for more details see[19]. After the resistance measurements, the sample was heated up to 390 K, so that the trapped magnetic flux vanishes, followed by a zero-field cool to room temperature. Substrate, including sample, was then placed and fixed with varnish on a copper plate, where a thermometer and heater were at the backside of the plate. The sample and copper plate were connected to ground, to avoid electrostatic influences. MFM measurements were then carried out in the usual way.

d. Characterization of the tip using a current loop According to Eq. (5), we need to calculate the second derivative of the field produced by a current loop. For simplicity, the following substitutions are used[22]: $\rho^2 = x^2 + y^2, r^2 = x^2 + y^2 + z^2, \alpha^2 = a^2 + r^2 - 2a\rho, \beta^2 = a^2 + r^2 + 2a\rho, k^2 = 1 - \alpha^2/\beta^2$, $C = \mu_0 I/\pi$, where $I$ is the current through the loop, $a$ is the radius and origin is placed at the center of the loop, in the $x - y$ plane. The $z$-component of the magnetic field is then given by[23] (assuming the cross section of the conducting path is negligible)

$$B_z = \frac{C}{2a^2 \beta} \left[ \left( a^2 - r^2 \right) E(k^2) + \alpha^2 K(k^2) \right], \quad (9)$$

where $K$ and $E$ are elliptic integrals of first and second kind, respectively. The phase can now be simulated using Eqs. (5) and (8), the calculations were carried out using Mathematica. Further, within the dipole approximation, the phase at the center of the loop is then

$$\Delta \varphi = \frac{3\mu_0 a^2 I m_z Q}{2k} \left[ \frac{a^2 - 4(d + \delta)^2}{(a^2 + (d + \delta)^2)^{7/2}} \right], \quad (10)$$

where $d$ is the lift scan height, $\delta$ is the tip-dipole distance and $\Delta \varphi$ is the difference between the value at the center of the loop and the value at the edge of the measured spectra, i.e. where $\varphi$ is approximately constant, see FIG. 2(e). Note that the shown line scans are taken from the images, the scans used to calibrate the tip were measured across the center of the loop such that there was sufficient space for the phase to saturate. Further, seven lines were saved and used to obtain an averaged scan.

We measured $\Delta \varphi$ as function of distance $d$ and applied current $I$, the data and the fits are shown in Figs. 2(a) and (b). The fits yield $m_z = (1.27 \pm 0.2) \times 10^{-13}$ Am$^2$ and $\delta = (1.31 \pm 0.2) \mu$m, these values agree with what has been observed in the literature[22]. However, it is important to be aware that these are effective results and might differ for different samples. This is due to the different magnetic decay lengths of the magnetic field, which results in a different effective magnetic volume of the tip within the field of the sample. MFM images of another current loop with applied currents of $\pm 3$ mA are shown in Figs. 3(c) and (d), the lines indicate the position of the phase spectra shown in Figs. 2(e) and (f).

e. MFM of ferromagnetic samples MFM is a powerful tool to characterize the magnetic stray fields of a variety of samples, such as thin films or nano/microstructures. This is especially interesting for cases where other measurement techniques are not suitable, e.g. a
SQUID is not practical to measure samples with a large background due to substrates or, as it is the case for interfaces in graphite, due to the bulk contribution. As an example of typical MFM images of ferromagnetic samples, the measurements of a multi-walled carbon nanotubes (MWCNT) filled with iron, see Figs. 2(a)–(h)), and a ZnFe₂O₄ ferromagnetic thin film, FIG. 3(i), are shown. Both samples show clear magnetic domains and domain walls between them. Both samples were not magnetized prior to the MFM scans, thus the domains are pointing in arbitrary directions. FIG. 2(a) shows an image of the topography of the nanotube, FIG. 2(b) the corresponding line scan. The MWCNT was measured at three different scan heights, 50 nm, 100 nm and 150 nm, the images (Figs. 2(c),(e),(g)) and line scans (Figs. 2(d),(f),(h)) are given. The phase line scan profiles show Néel wall features and beside the MWCNT the phase is zero, where no magnetic field is present (or it is a constant).

IV. RESULTS

In this Section we will present the results of measurements on the graphite sample. In Section IV A the result of energy-dispersive X-ray spectroscopy is shown. A comparison of the sample before and after applying a magnetic field is given in Section IV B an investigation on the temperature dependence on the phase signal can be found in Section IV C. Further, the change over an extended period of time is presented in Section IV D.

A. Energy-dispersive X-ray spectroscopy

In order to check for the presence of rhombohedral and Bernal graphite, X-ray diffraction measurements were performed at the initial material, the results can be found in. The existence of both, Bernal and rhombohedral graphite was confirmed. To check for the presence of magnetic impurities, energy-dispersive X-ray (EDX) spectroscopy was carried out at the position of the sample, where the permanent current path was measured. The results of a map scan are shown in FIG. 4. The EDX measurement yields a carbon content of ∼94 At % and ∼6 At % of oxygen. Figs. 4(a) and (b) show the SEM image and the results for Fe and Ni, respectively. We find isolated, homogeneously dispersed impurities, with less than 0.004 At % according to the EDX software analysis.

B. Before/After application of an external magnetic field

Before we started the MFM measurements, the sample has been put into the virgin state. It means, that the sample was placed in the magnetocryostat, for convenience and also to measure the electrical resistance, see Section IV D. Then it was heated to $T = 390$ K followed by a cool down at no field to room temperature. The so-prepared sample was measured for several weeks, in order to cover a large area. A MFM image is shown in FIG. 5(b), and, as it can be seen, there is no sign of magnetic domains. If there was ferro- or ferrimagnetic order with the corresponding magnetic domains, all domains would have a spontaneous orientation and there would be a change in the phase. If the domains were smaller than the lateral resolution, i.e. 50 nm, we would see some unresolved averaged changes in the phase, but changes nevertheless.

In the next step, a permanent magnet (for $t \approx 10$ s, magnetic field $\approx 0.05$ T, measured with a Hall sensor) was placed near the sample with the magnetic field oriented perpendicular to the sample surface. After application of the external magnetic field, the MFM measurements were continued. In FIG. 5(d) the result is shown at the same position as in FIG. 5(b), as the corresponding topography images indicate (Figs. 5(a)–(d)). A clear phase feature appears, FIG. 5(g) shows the phase image at another position, where the feature continued and is clearly visible. From the topography and phase images, it is obvious that there is no relation between surface and phase signal. However, as the naked eye can give a misleading
FIG. 3. The topography ((a)&(b)) and the measured phase ((c)-(h)) of a multi-walled carbon nanotube filled with Fe at different lift heights. (i) is the MFM image of a ZnFe$_2$O$_4$ ferromagnetic thin film.

FIG. 4. (a) Scanning electron microscopy image of the region where the current line is located. (b) is the corresponding energy-dispersive X-ray image for iron and nickel.

FIG. 5. Topography images (top) and phase images (bottom) before ((a)&(b)), and after ((c)&(d)) application of an external magnetic field; (f) and (g) show the topography and phase at another position.

A line scan, at the position indicated in FIG. 5(g) as white line, is shown in FIG. 6; the inset is an optical image of the sample, the line shows the position of the persistent current. We could not measure the complete loop, only up to the edges of some rough surface regions. The line scan looks similar to that of a current loop, see FIG. 2(e) and compare with the scans of magnetic domains, e.g. Figs. 3((d)–(h)). This result strongly suggests that the observed signal is not due to magnetic order: After the application of an external field, magnetic domains would have been aligned in $z$-direction. It means that we would need to have two areas, one magnetically ordered (with a single domain) and one without any domain. But this would imply that magnetic stray field existed from the beginning – before applying an external field – because regardless of the magnetization direction and/or size of the domain(s), at the boundary of the ferromagnetic/nonmagnetic region, stray fields would have been present. As shown before, no stray field signals were measured, neither at the shown position, nor at any scan position of the sample in the virgin state.
A remarkable change occurs at \( T \) to the shown profiles up to convenience not all are shown, however, they look very similar to the shown magnetic field in the interior. Several scans have been performed with a rather constant magnetic field in the interior. An optical image of the scanned area is shown in FIG. \ref{fig:6}f. The scans show a well-defined offset between either side of the loop. This is due to the shape of the loop at this position, which resembles a U-turn, with a rather constant magnetic field in the interior. Several scans have been performed (in temperature steps of 5 K), for convenience not all are shown, however, they look very similar to the shown profiles up to \( T = 370 \) K. The magnitude of the phase remains constant, only a broadening can be seen. A remarkable change occurs at \( T = 377 \) K, where a sudden decrease of the phase sets in, see FIG. \ref{fig:7}i). MFM measurements were continued to \( T = 395 \) K, and then the sample was zero-field cooled back to room temperature. The phase signal did not reappear at any point, the result at room temperature can be seen in FIG. \ref{fig:7}a) as red line.

The sharp transition around \( T = 377 \) K contradicts the expected behavior of ferromagnetism, where a continuous decrease of the magnetic coupling would cause a continuous decrease of the magnitude of the phase. Indeed, the decrease of the remanence with temperature in graphite samples that show magnetic order, follows a nearly linear decrease with temperature from low to about 0.9 of the Curie temperature \( T_C \) at higher temperatures near \( T_C \), the decrease is even stronger. This dependence is compatible with 2D spin waves measured in non-irradiated, as well as irradiated graphite, see also the review in for details and additional literature.

Further, when cooling without applied magnetic field, magnetic domains would tend to order spontaneously in random directions, and a domain structure similar as shown in FIG. \ref{fig:3}(i) would be expected. Yet, no signal in the phase was obtained after heating above this critical temperature and cooling down again. Using \( \Delta \varphi \), see FIG. \ref{fig:7}(a), we plot the temperature dependence of the phase in FIG. \ref{fig:7}(g). The transition is clearly visible around \( T = 370 \) K and the general behavior agrees very well with the resistance measurements of the sample, i.e. \( R(T) \) after a linear background subtraction, and the remanence \( \Delta R(0) = R_B(0) - R_0(0) \). Here, \( R_B(0) \) is the resistance measured at zero field after applying a field of 0.03 T normal to the main sample surface, and \( R_0(0) \) is the resistance of the sample in the virgin state, obtained after zero-field cooling from \( T = 390 \) K, which was done for each temperature. For more details regarding the temperature dependence of this remanence in different graphite samples see.

The two resistance measurements and the MFM scans indicate a critical temperature of \( T_c \approx 370 \) K with a transition width of \( \lesssim 20 \) K. In FIG. \ref{fig:7}(e), the MFM line scan of a current loop is shown, measured at \( T = 380 \) K under the same conditions as the measurements done for the graphite sample. This confirms that the MFM experiments work at temperatures higher than \( T_c \), and that the tip remains magnetized at such elevated temperatures. Note that, if the magnetization of the tip was influenced by field of the current loop, we would have seen a hysteresis-like shape in the phase scans (backward/forward scan direction), which was not the case.

The temperature dependence of the resistance was already measured before the MFM measurements and a transition was found at \( T \approx 370 \) K, in agreement with measurements done in similar samples. For this reason, we measured the temperature dependent MFM and the results are shown in FIG. \ref{fig:7}. In Figs. \ref{fig:7}(a)–(d)) the lines scans at four different temperatures can be seen. An optical images of the scanned area is given in FIG. \ref{fig:7}f. The scans show a well defined offset between either side of the loop. This is due to the shape of the loop at this position, which resembles a U-turn, with a rather constant magnetic field in the interior. Several scans have been performed (in temperature steps of 5 K), for convenience not all are shown, however, they look very similar to the shown profiles up to \( T = 370 \) K. The magnitude of the phase remains constant, only a broadening can be seen. A remarkable change occurs at \( T = 377 \) K, where a sudden decrease of the phase sets in, see FIG. \ref{fig:7}i). MFM measurements were continued to \( T = 395 \) K, and then the sample was zero-field cooled back to room temperature. The phase signal did not reappear at any point, the result at room temperature can be seen in FIG. \ref{fig:7}a) as red line.

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After applying an external magnetic field, it took approximately 22 days to find the position of the permanent current path and to prepare to follow up with the experiments. This period followed an investigation of the time dependence of the phase shift \( \Delta \varphi \). For this purpose, a suitable spot was chosen and measured for 2 days such that approximately every hour, a \((10 \times 10) \mu m^2\) image was obtained. The phase shift was then obtained at the same position of the sample, considering the piezo creep (of the positioners), especially within the first hours of the experiment, see FIG. \ref{fig:6}.

\[ a. \quad \text{Estimate of the resistance} \quad \text{Using} \ \Delta \varphi(T), \ \text{an effective electrical resistance can be estimated with Eq.}\] \[ \text{that would produce the measured decrease in phase shift, i.e., in the current amplitude. Applying the parameters mentioned above, we find a resistance of} \ \approx 8 \times 10^{-17} \ \Omega. \] \[ \text{The intrinsic ohmic resistance of the loop should be much smaller, eventually zero. The current line remains for several weeks which indicates the existence of a permanent current which originates the magnetic field and is also the reason for the observed remanence in the resistance. However, the reduction of } \Delta \varphi \]
can be explained in terms of creep. Since
\[ \frac{\partial m}{\partial t} \propto \frac{\partial \phi}{\partial t} \times \frac{\partial I}{\partial t} \propto \frac{\partial \phi}{\partial t}, \]
the well known logarithmic time dependence can be written as
\[ \frac{\Delta \phi(t)}{\Delta \phi(0)} = 1 - \Delta \phi_1 \ln \left( 1 + \frac{t}{\tau} \right), \]
where \( \tau \) is a time constant and \( \phi_1 \propto k_B T / U_a \) with \( U_a \) being the flux creep activation energy. The results and the fit to Eq. (12) can be seen in FIG. 8, where \( \tau = 10 \text{ s} \) has been taken from [32]. The inset shows \( \Delta \phi(t) \) starting from \( t \approx 0 \), which was calculated using the results of the fit \( \Delta \phi_1 = 0.87 \).

The meandering-like structure of the measured current path is similar to the one observed in high-temperature superconducting oxides, where a modified Bean model, that includes the lower critical field \( H_{c1} \), was used to understand the origin of the Meissner hole formation [31]. However, if superconductivity is localized at the interfaces of the graphite sample, from the Ginzburg-Landau relation for \( H_{c1} \propto 1/\Lambda^2 \) and using \( \Lambda = 2 \lambda_2 / d_i \), one expects \( H_{c1} \) to be negligible due to the huge penetration depth. Further, the pinning of the pancake vortices within the interfaces would also be negligible. In contrast, the here presented sample as well as similar ones, show a maxima in the remanence of the resistance \( \Delta R(0) \) not far from the critical temperature. This indicates that the magnetic field at remanence is produced by macroscopic (or mesoscopic) current loops, i.e. fluxons, not pinned pancake vortices. These fluxons are the origin for the remanent magnetic field and the irreversible behavior observed in the electrical resistance and magnetization.

Examples for the creep of Meissner holes can be seen in Figs. 9(a)–(f). There, remagnetization of superconducting plates in normal fields occurs by closed induction loops centered at the remagnetization front. This formation of the Meissner holes is a consequence of changing the field distribution at the edge of a plate in the normal field. The measurements of the fluxons in the graphite sample are shown in Figs. 9(g)–(h), where time-dependent, thermally activated creep gives rise to a weak dissipation.
FIG. 9. Figures ((a)–(f)) are adapted from \cite{37,38}. (a) shows a sketch of magnetic field of a superconducting plate in remanent state. The Meissner holes with $B = 0$ are shown, the shape (b) in a square-shaped plate, critical currents in the bulk and Meissner current along the boundary of the flux-free cylinder. Magnetic flux patterns in a 40 µm thick YBCO crystal in: (c) remanent state after field cooling in $\mu_0 H = 150$ mT and switching off the field at $T = 20$ K; (d) application of $\mu_0 H = -100$ mT after (c), the remagnetization front moved further inside the sample; (e) remanent state after field cooling in $\mu_0 H = 150$ mT and switching off at $T = 55$ K; (f) application of $\mu_0 H = -23$ mT after (e). (g) and (h) are MFM images of the graphite samples with a time difference of 2 days, the drift is position was corrected. The time-dependent creep of fluxons can be seen. The shape of the fluxons resembles the shape of the Meissner holes in (e) and (f), where also the boundary between two flux directions is narrow and strongly bent.

V. SUMMARY AND CONCLUSION

We have prepared and measured a natural graphite flake (Sri Lanka), which shows a transition at $T \approx 370$ K in the electrical resistance and its remanence. We were able to localize a persistent current using MFM, which is stable for several weeks, yet shows signs of creep. Because of the apparent similarity of the line phase scans to the scans one would obtain around magnetic domain walls, we summarize below the reasons that speak against this possible origin.

a. Reasons against a magnetic origin. The first experimental observation at odds with a magnetic origin, is that the phase is constant over all sample when it is measured in the virgin state, that means after cooling down from above $T_c$ without applied magnetic field. Would there be a magnetic order transition, it is highly unlikely that there were no measurable stray fields from magnetic domains, as all domains would have to be aligned in the same direction. Even if these domains were smaller than the spatial resolution, some unresolved signal should have been measured.

We have measured the phase feature at the apparent current path over a large area with a length of $\approx 600$ µm. It is very unlikely to obtain such a large domain wall in remanence and after application of a field as low as 30 mT at room temperature. If that was the case, the exchange interaction would be strong enough to form large magnetic domains when cooling below the Curie temperature at no applied field, which is clearly not the case.

The sudden vanishing of the phase, i.e. the stray fields, at $T \approx 370$ K is not what one would expect when approaching the Curie temperature, where a continuous decreasing amount of magnetically ordered moments results in a decreasing phase shift and/or more pronounced domain wall structure.

As discussed in \cite{32}, there is no evidence for magnetic order in all measured samples. Neither the amount of impurities is significant, nor the intrinsic disorder, in very different samples, ranging from bulk to multilayered graphene flakes.

The measured scan profile of the phase at the path border is not compatible with the edge of a magnetically ordered domain and/or a domain wall. Part of the scan profile, i.e. the zero crossing and the sign change between the left and right side of the border, appears to be compatible with a Néel wall between two magnetic domains. But, the measured line scan indicates that a homogeneous field remains only in the interior of the loop, which contradicts the expected profile from the two antiparallel domains (after application of an external field perpendicular to the sample surface). Further, it is unlikely that it is a single domain, because parts of the same material would couple ferromagnetically while other areas do not.
In addition, this asymmetry of the phase border implies that even if the domains were pointing all in the same direction (the virgin state) there would have to be stray fields at this position. This was not the case in the virgin measurements and after zero field cool. It is also highly unlikely that those magnetic domains would be produced only after applying a field normal to the graphene planes and that all are antiparallel aligned in such a long path.

The observed change in the position of the current path with time rules out a correlation with any magnetic topographic feature, such as zigzag edges or conglomeration of magnetic impurities. This change was not only observed in the measurements presented above, but was recognizable during all measurement time. This was especially imminent when we returned to the starting position, after we reached the rough edge region, where we were not able to continue the MFM measurements, but had to continue the experiments on the other side of the found current path.

The virgin state is not reached anymore, even changing the amplitude direction of the maximum applied field, see[32]. Further, the magnetoresistance is positive and large at fields of a few mT, in contrast to the negative and weak magnetoresistance at $T \geq 300$ K for magnetic graphite[35].

Our results indicate the presence of a trapped flux through a persistent current, which we interpret as superconductivity. Further, MFM, as well as other scanning magnetic imaging techniques, can be used to identify the regions of interest of the graphite sample. This will undoubtedly assist to further characterize the superconducting-like interfaces in graphite, paving the way for their future device implementations.

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