Advanced Practical Course M

M2.4 Magnetization of a superconductor

January 15, 2019

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Date:	November 20, 2018

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1 Introduction

In this experiment, the magnetization of a type-I superconductor was measured at different temperatures and applied fields. By changing the applied magnetic field, magnetisation curves were made for different temperatures. From this, the critical field and critical temperature could be found, as well as the demagnetization factor. From the Field Cooling and Zero Field Cooling measurements, the shielding effect could be observed and the Meissner fraction could be determined for different applied fields.

2 Theoretical background

2.1 Phenomenon of superconductivity

After successful liquefaction of helium by Kamerlingh Onnes in 1908 it has became possible to investigate properties of metals in the new range of low temperatures. In 1911, he observed a surprising drop in resistance of a mercury sample at about 4 K, which within the accuracy of his experiment was indistinguishable from zero. This phenomenon was named superconductivity and temperature at which it happens - critical temperature. Since that time superconductivity was observed in many others materials.

However, superconductors are not just perfect conductors (defined as a conductor with infinite free path, which is a model for normal conductors in a low temperatures). In 1933 W.Meissner and R.Ochsenfeld discovered, that applied magnetic field is expelled from superconductors after cooling below critical temperature. This result cannot be obtained from Maxwell's equations and Ohm's law assuming only zero resistivity:

$$\boldsymbol{E} = \rho \boldsymbol{j} \Rightarrow \mathbf{E} = \mathbf{0} \tag{1}$$

$$\nabla \times \boldsymbol{E} = -\frac{\partial \boldsymbol{B}}{\partial t} \Rightarrow \frac{\partial \boldsymbol{B}}{\partial t} = 0, \tag{2}$$

so flux through metal cannot change if we cool it below critical temperature, which contradicts Meissner effect. In the case of perfect conductor, if it was subjected to external magnetic field before becoming perfect conductor, the internal fluxes will remain inside even after removal of external field. However, in the case of superconductor, magnetic field will be totally expelled out of metal after transition. This result holds for both possible situations: if superconductor is cooled below critical temperature in zero field and then it was applied and superconductor was cooled in a finite field (Fig.1). This effect is achieved by producing currents on the surface of superconductor, which create magnetic field that cancels the applied one inside the superconductor.

2.2 London equations and coherence length

Because observed Meissner effect cannot be explained directly from basic equations of electrodynamics, some modifications are needed. However, Maxwell's equations are believed to be universal,

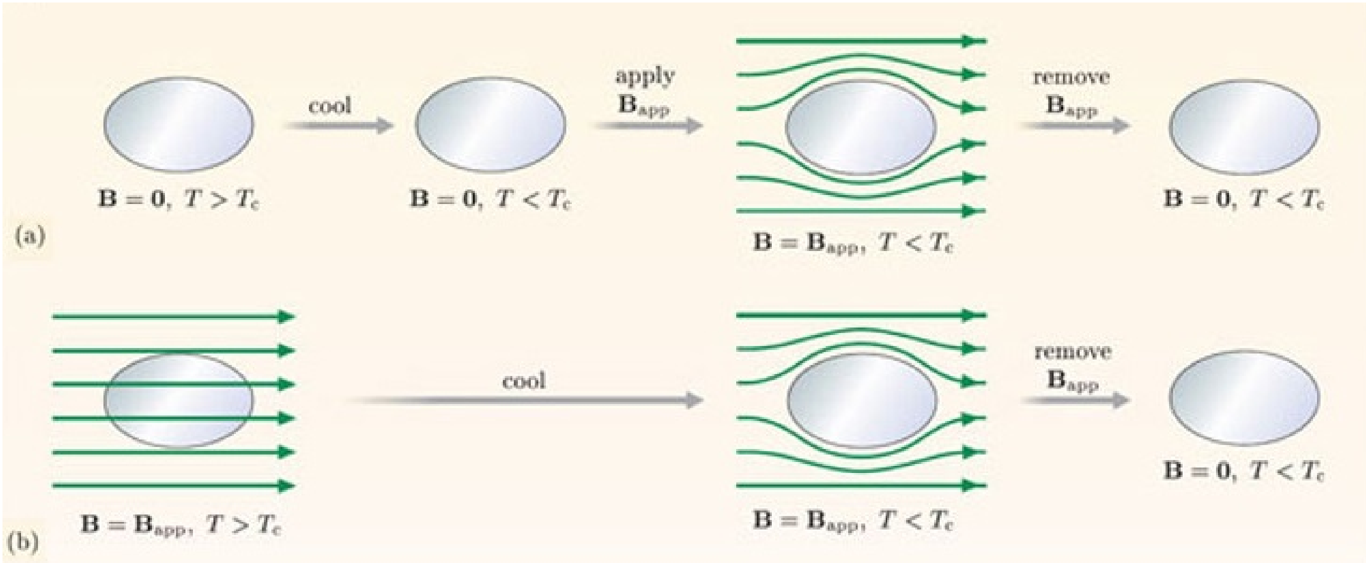

Figure 1: Response of superconductor to applied magnetic field (a) after and (b) before cooling [1]

so some changes to Ohm‘s law should be done. In 1935 F. and H. London postulated equation, which is consistent with Meissner effect:

$$\boldsymbol{j} = -\frac{1}{\mu_0 \lambda^2}\boldsymbol{A} \Rightarrow \nabla \times \boldsymbol{j} = -\frac{1}{\mu_0 \lambda^2}\boldsymbol{B}, \tag{3}$$

where $\boldsymbol{A}$ is a vector potential, λ - constant with dimension of length, μ_0 - vacuum permeability. Together with specifying gauge in which $\nabla \cdot \boldsymbol{A} = 0$, $\boldsymbol{A_n} = 0$ (thus $\nabla \cdot \boldsymbol{j} = 0$ and $\boldsymbol{j_n} = 0$) and Maxwell‘s equations it can be obtained, that:

$$\nabla^2 \boldsymbol{B} = \boldsymbol{B}/\lambda^2 \tag{4}$$

It is obvious from this equation, that uniform magnetic field is not its solution. However, it cannot be obtained, that magnetic field is exactly zero inside. For a semi-infinite superconductor on the positive side of x axis, solution of form

$$\boldsymbol{B}(x) = \boldsymbol{B}(0)\exp(-x/\lambda) \tag{5}$$

can be obtained. So magnetic field decays exponentially with λ being its penetration depth. This constant depends on superconducting electron density, but its typical range is about $10^{-8} - 10^{-9}$m. Because of that, from London equation magnetic field is predicted to be effectively excluded from macroscopic specimens with good agreement with observation of Meissner effect.
However, London equation is a local equation, while superconductivity is a non-local effect with coherent state of many electrons, condensed into macroscopic ground state. Characteristic distance, over which their number density can change in spatially inhomogeneous magnetic field, is known as coherence length ξ_0. It also can be understood as measure of scale, over which we should average $\mathbf{A}$ to obtain $\mathbf{j}$.

In order to obtain approximate expression for coherence length, energy scales of energy gap in superconductors E_g (found at Fermi level using spectroscopy measurements) and Fermi energy

E_F (which represents the highest occupied energy level at zero temperature) should be compared. Coherence length can be defined as

$$\xi_0 = \hbar v_F / 2E_g \tag{6}$$

or, as found using BCS theory, with different numerical constant:

$$\xi_0 = 2\hbar v_F / \pi E_g. \tag{7}$$

Defined in this way coherence length is called intrinsic, because for impure materials real coherence length ξ is shorter then ξ_0. Both ξ and λ depend on the mean free path of the electrons in normal state. Relation between them $\kappa = \lambda/\xi$ plays important role in determining properties of superconductor (it is approximately independent of temperature near phase transition, and, therefore, is a convenient parameter). For $\kappa \gg 1$ London model is a good approximation, but for most pure elements well below their critical temperature $\kappa \ll 1$ and local model is not appropriate.

2.3 BCS theory

London equation (3) does not explain origins of superconductivity, but is a useful relation, using which experimentally observed Meissner effect can be deduced. More fundamental microscopic theory was developed in 1957 by Barden, Cooper and Schrieffer. It relied on previously found result by Cooper, that arbitrarily small attraction between electrons in metal will lead to formation of pairs, which have a lower energy than the Fermi energy. This attraction can be explained as electron-lattice-electron interaction: second electron adjusts itself to a deformation of lattice caused by the first electron. Such Cooper pair consists of two electrons, with momentum $\mathbf{k}$ and spin up and momentum $-\mathbf{k}$ and spin down, so that their total momentum is zero and they have many attributes of bosons. Most importantly, they can condense into Bose–Einstein condensate and form a BCS superconducting ground state.

Fundamental property, which can be obtained from the BCS theory, is a formation of a finite energy gap E_g between ground state and lowest excited state. This makes possible to evaluate

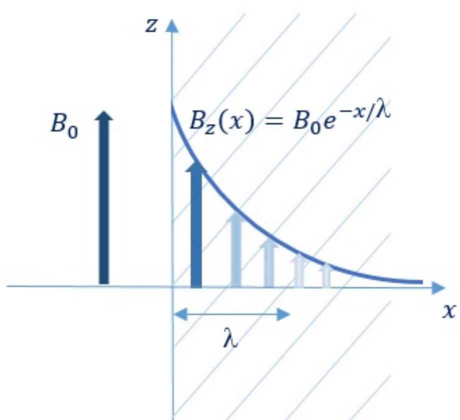

Figure 2: Penetration of magnetic field inside superconductor

the critical field of superconductor, thermal and most of electromagnetic properties. Amount of energy, required to scatter electron from condensate, is larger then the thermal energy, available to it below critical temperature. Because of this, electrons can flow without being scattered and zero resistence is observed. London equation (and thus a Meissner effect), penetration depth and coherence length can be also obtained within this theory.

However, the upper bound of critical temperature, predicted by BCS theory is about 30 K, which turned out to be false, as such materials were discovered later. In this cases some other effects play important role.

2.4 Superconductor in magnetic field

In SI magnetic susceptibility χ and magnetic permeability μ are defined by relations:

$$\boldsymbol{M} = \chi \boldsymbol{H}$$
$$\boldsymbol{B} = \mu_0(\boldsymbol{H} + \boldsymbol{M}) = \mu_0(\boldsymbol{H} + \chi\boldsymbol{H}) = \mu\boldsymbol{H} \tag{8}$$

Due to Meissner effect, discussed in the first section, magnetic field inside superconductor is zero, so (Fig. 3a):

$$\boldsymbol{B} = \mu_0(\boldsymbol{H_a} + \boldsymbol{M}) = 0 \Rightarrow \boldsymbol{M} = -\boldsymbol{H_a}. \tag{9}$$

There are two types of superconductors: for the first type, magnetic field can be applied up to critical value H_c, after which magnetic field penetrates into superconductor and superconductivity is ruined (they have $\kappa < 1/\sqrt{2}$). The temperature dependence of critical field H_c can be quite good approximated by parabolic law (Fig.4):

$$H_c(T) \approx H_c(0)(1 - (T/T_c)^2). \tag{10}$$

For the second type of superconductors (characteristic parameter $\kappa > 1/\sqrt{2}$) two critical fields exist (Fig. 3b): after exceeding H_{c1} the Meissner effect is incomplete and magnetic field can penetrate inside in the form of fluxes and after exceeding H_{c2} superconductivity is ruined. All of pure elements, which can host superconductivity, are type-I superconductors, while superconducting alloys and high critical temperature ceramics are type-II.

Transition between superconducting and normal state is reversible, so it is possible to obtain free energy difference between this two states as a function of applied magnetic field, needed to destroy superconductivity. The work per unit volume, done on a superconductor to take it at constant temperature from infinity to a certain position $\boldsymbol{r}$ in a field of a permanent magnet equals:

$$W = -\int_0^{B_a} \boldsymbol{M} d\boldsymbol{B}_a$$

For a type-I superconductors we can insert eq.(9), so for free energy in superconducting state:

$$dF_s = \frac{1}{\mu_0} B_a dB_a,$$

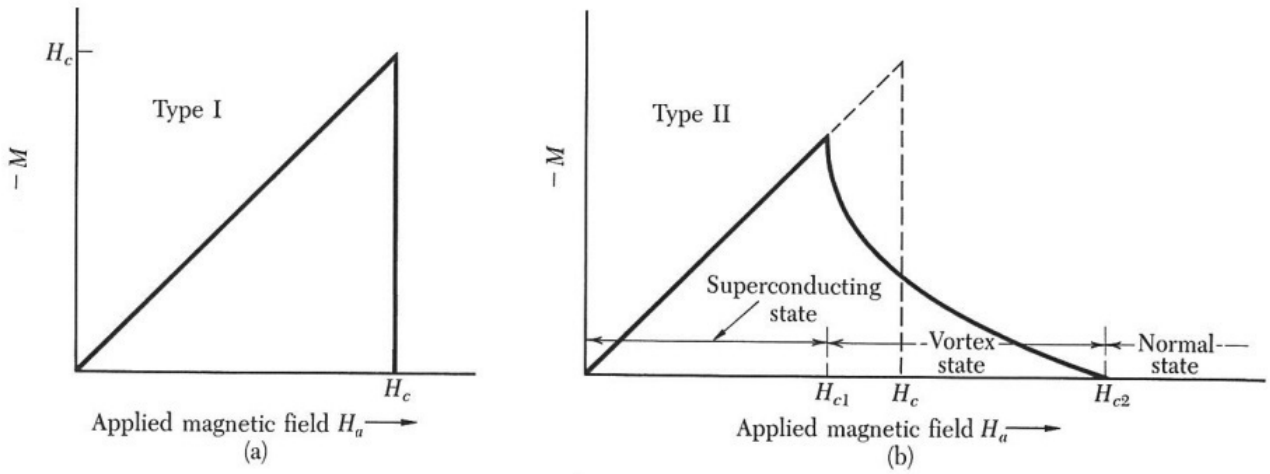

Figure 3: Types of superconductors [2]

and after integration:

$$F_s(B_a) - F_s(0) = \frac{B_a^2}{2\mu_0}. \tag{11}$$

For a normal nonmagnetic metal $M = 0$, so at critical field:

$$F_N(B_{ac}) = F_N(0) \tag{12}$$

Free energies (12) and (11) are equal at critical field, so we can combine them to obtain

$$F_N(B_ac) = F_S(B_{ac}) = F_S(0) + \frac{B_{ac}^2}{2\mu_0} \tag{13}$$

This is depicted in Fig.5. If applied field is stronger, then critical, nonmagnetic normal metal has lower free energy density, so it is a favorable state. However, the situation changes after going through phase transition at B_{ac}, and superconducting state has lower energy with quadratic dependence on applied field.

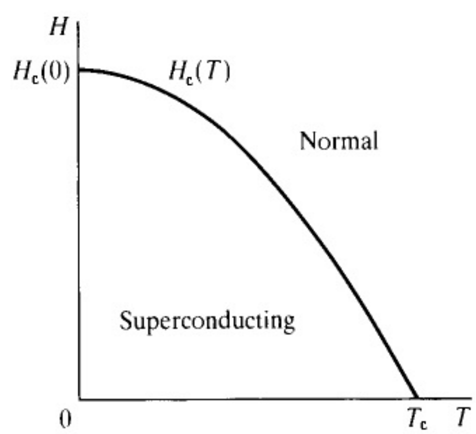

Figure 4: Temperature dependence of critical field [3]

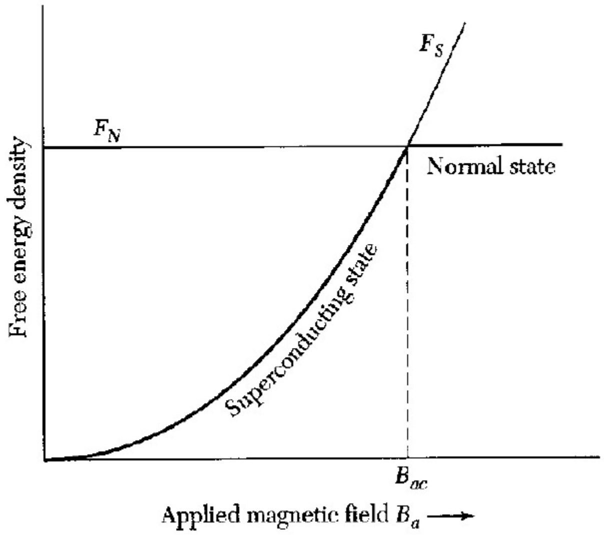

Figure 5: Free energy density in nonmagnetic and superconductor states [2]

Let‘s consider now effect of the shape of a sample. This becomes important, when applied fields are strong enough to destroy superconductivity. The field lines will have a higher density over part of surface and there will be a local increase in magnetic field. Inside the sample:

$$\boldsymbol{H}_i = \boldsymbol{H}_{ext} + \boldsymbol{H}_d, \tag{14}$$

where $\boldsymbol{H}_{ext}$ and $\boldsymbol{H}_d$ are external and demagnetization magnetic fields. The latter is proportional to $\boldsymbol{M}$ with demagnetizing factor D, so:

$$\boldsymbol{H}_i = \boldsymbol{H}_{ext} + \boldsymbol{H}_d = \boldsymbol{H}_{ext} - D\boldsymbol{M}. \tag{15}$$

The apparent magnetic susceptibility is then

$$\chi_{app} = \frac{|\boldsymbol{M}|}{|\boldsymbol{H}_{ext}|} = \frac{|\boldsymbol{M}|}{|\boldsymbol{M}|/\chi + D|\boldsymbol{M}|} = \frac{\chi}{1 + D\chi}. \tag{16}$$

For superconductor $\chi = -1$, so finally we obtain

$$\chi_{app} = -\frac{1}{1 - D}. \tag{17}$$

Possible values for demagnetizing factor lie between 0 and 1 (Fig.6):

cylinder in parallel field	$D = 0$
cylinder in transverse field	$D = 1/2$
sphere	$D = 1/3$
thin plate in perpendicular field	$D = 1$

Table 1: Demagnetizing factors

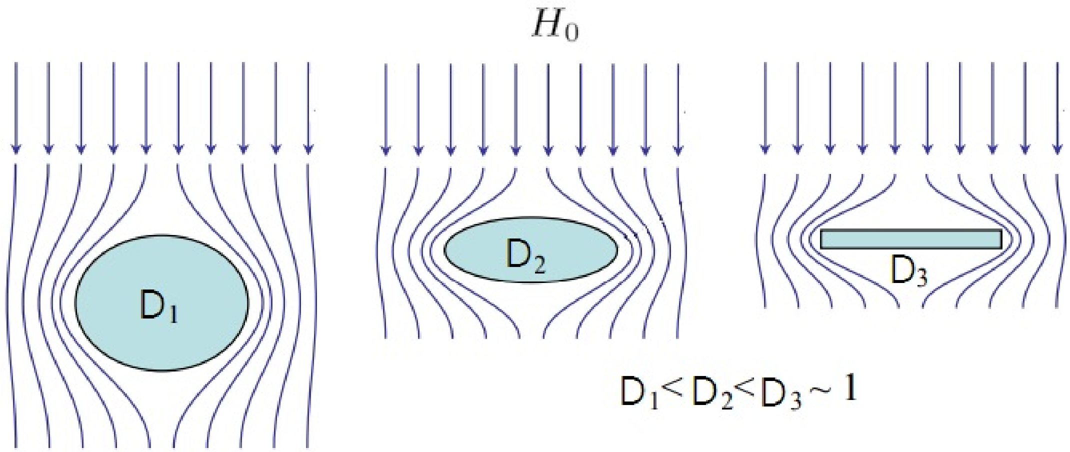

Figure 6: Demagnetizing factors for different shapes [4]

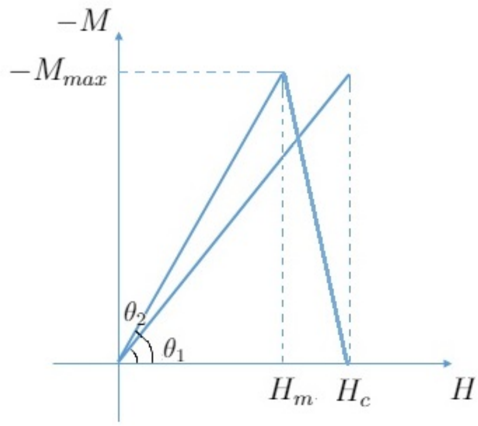

Figure 7: magnetization

Because of nonzero demagnetizing factor for real specimens, dependence of magnetization on magnetic field will have a maximum at $H_m < H_c$ (Fig.7) with

$$\tan(\theta_1) < \tan(\theta_2) = \frac{M_{max}}{H_m} = \frac{H_c}{H_m} = -\frac{1}{1-D}, \tag{18}$$

so demagnetizing factor can be found as

$$D = 1 - \frac{H_m}{H_c}. \tag{19}$$

As expected, the area below magnetization curve remains the same, because it is related to the thermodynamics of process. For a certain range of applied magnetic field, normal regions should coexist with superconducting regions. This state is called intermediate. For example, for a sphere, effective field in equator is $3H_a/2$, while on poles it is approximately H_a.

Now we can consider the effect of non-homogeneity of a sample. Let‘s consider the simplest case, where inner and outer parts of superconductor have different critical temperatures, T_1 and T_2 respectively (for certainty, $T_1 < T_2$). Then, for the temperatures T between T_1 and T_2 in the applied external field B_{ex} outer part will become superconducting and will expel magnetic field (so that $B = 0$ inside it), while the inner will be a normal conductor and magnetic field will have finite value B_{ex}. The total magnetization is then:

$$M_{tot} = -\frac{V_{sc}}{V}\frac{B_{ex}}{\mu_0} \tag{20}$$

Magnetic susceptibility for the case when sample is cooled inside magnetic field is then

$$-\chi_{FC} = -\frac{M_{tot}}{H} = \frac{V_{sc}}{V} < 1. \tag{21}$$

The superconducting volume fraction is called Meissner fraction. If a sample is cooled without external magnetic field and only then it is switched on, then internal flux is zero and total magnetization depends on the distribution of superconducting and normal conducting volume fractions. The following expression is valid:

$$B_{ex} \geq -\mu_0 M_{tot}^{zfc} \geq \frac{V_{sc}}{V} B_{ex} \Rightarrow 1 \geq -\chi_{zfc} \geq -\chi_{fc} \tag{22}$$

2.5 Superconducting ring

Superconductor can be described by using $\psi(\boldsymbol{r})$ - a particle probability amplitude, which equals

$$\psi(\boldsymbol{r}) = n^{1/2} e^{i\theta(\boldsymbol{r})},$$

where n is a pair concentration and $\theta(\boldsymbol{r})$ is a phase. From the Meissner effect, $\boldsymbol{B}$ and $\boldsymbol{j}$ are zero inside the interior of ring, so

$$\boldsymbol{j} = q\psi^* \boldsymbol{v}\psi = \frac{nq}{m}(\hbar\nabla\theta - \frac{q}{c}\boldsymbol{A}) = 0 \Rightarrow \hbar c\nabla\theta = q\boldsymbol{A}, \tag{23}$$

where q - elementary charge in superconductor (found by experiment to be $2e$), $\boldsymbol{A}$ - vector potential. Change of phase after going once around the ring (contour C, Fig.8):

$$\oint_c \nabla\theta \cdot dl = \theta_2 - \theta_1 = 2\pi s,$$

where s is an integer. This is because the probability amplitude $\psi(\boldsymbol{r})$ should be measurable in classical approximation. Integrating left hand side of eq.23 leads to:

$$\oint_c \boldsymbol{A} \cdot dl = \iint_C \boldsymbol{B}\, d\boldsymbol{\sigma} = \Phi$$

Finally we obtain:

$$\Phi = \frac{2\pi\hbar c}{q} s,$$

so the flux through ring is quantized. The smallest possible value of it is called fluxoid and is equal to $\Phi_0 \approx 2 \times 10^{-15}$ tesla m^2. Total flux is a sum of the flux of external sources and flux, created by currents which flow in the surface of superconductor. The first one is normally not quantized, so the second one should adjust itself in a way for total flux be quantized.

[5]

3 Experimental procedure

3.1 Setup

The goal is to apply different magnetic fields to the sample and then measure its magnetization. This is achieved by a set of coils. The setup is shown in figure 9.

A primary coil (1) is used to apply a field to the sample and the astatic pair of coils. The astatic pair (3a) and (3b) together with the magnetometer coil (7) form the flux transformer, which is used to measure a change in the sample's magnetization. Its operation is based on the fact that the magnetic flux though an area surrounded by a superconductor remains constant. Because the whole setup is submerged in a bath of liquid Helium, the flux transformer is lowered to a temperature of about 4.2K and becomes a superconductor. The flux transformer and sample are shown in figure 10. We know that $\phi_{tot} = A_1\phi_1 + A_2\phi_2 + A_3\phi_3$ will remain constant. Furthermore, we know $\phi_1 = \mu_0 H, \phi_2 = \mu_0(H + M)$ and ϕ_3 is measured. From measuring a change in H and ϕ_3, the change in M can be deduced.

The astatic pair (3a) and (3b) are wound in opposite direction and coupled in series to cancel the signal produced by the primary field. This cancellation is not perfect and some background remains, which needs to be subtracted from the data.

Flux heater (6) and sample heater (4 in right side of the picture) are used, as their names suggest, to heat up the flux transformer and sample, respectively. Part of the experiment is to look at the behaviour of the sample at different temperatures. Its temperature is measured by a resistor (5) which has a very well known calibration curve. The heating is also used in between measurements to remove super-currents in the flux transformer and magnetization of the sample by raising their temperature above their critical value. The flux heater only heats up a small part of the circuit to remove the super-current. It is located away from all other coils so its magnetic flux does not affect the measurement.

Finally, a fluxgate magnetometer probe (8) is used to measure a change in magnetic field in the magnetometer coil from which the change of magnetization of the sample can be deduced, as explained above. The magnetometer was invented in the 1930s by Victor Vacquier [6] and is able to measure very small changes in the magnetic field.

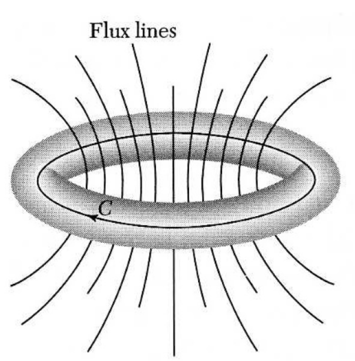

Figure 8: Contour of integration [2]

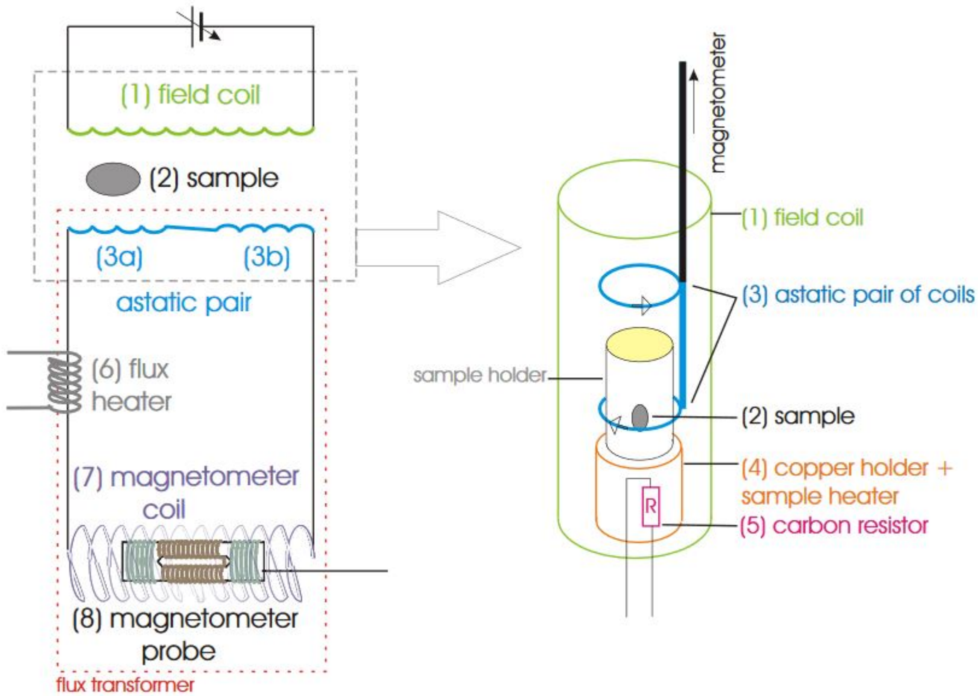

Figure 9: The experimental setup and the coil system around the sample.

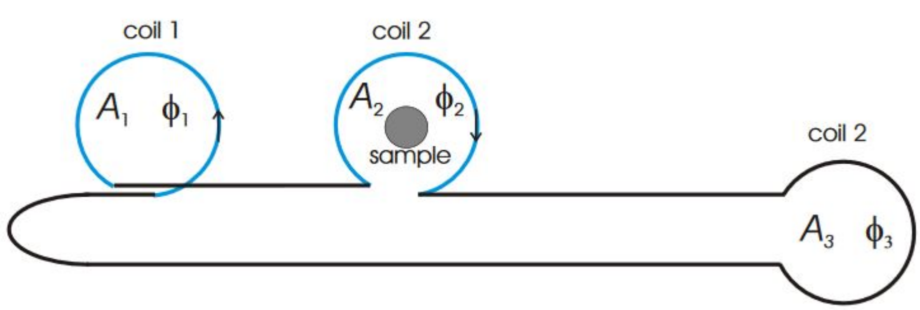

Figure 10: The flux transformer.

3.2 Measurements

Software was used to control the external field (field coil) and sample temperature. It could be used to automatically sweep either the external field or sample temperature and then measure the magnetization of the sample.

Magnetization curves (as a function of external field) were made for 10 different sample temperatures: 4.34, 4.56, 4.63, 4.92, 5.25, 5.55, 5.93, 6.26, 6.68 and 8.08 Kelvin. In between every measurement, the sample and flux transformer were heated to remove superconductor effects. In a low-field measurement, we determined that the critical temperature of the sample above which no superconducting effects show is roughly 6.8K. We made 9 curves below T_c and one above it.

Secondly, the magnetization was measured as a function of temperature for 10 different fields between $H = 0$ and $H = 517$G. Again, the sample and flux transformer were heated in between every measurement. In a low temperature measurement, we measured that the maximum field at which the Meissner-Ochsenfeld effect is still measured is roughly $H_{max} \approx 517$G.

4 Data analysis

4.1 Meissner-Ochsenfeld effect

From the obtained curves of volt (magnetization) as a function of resistance (sample temperature), the background signal was removed. The background signal was assumed to be a constant effect (since the external field was kept constant during these measurements) and equal to the high-temperature values of the magnetization, since at high temperatures the magnetization of the sample should disappear. Furthermore, the resistance was converted to temperature using the given calibration curve:[6]

$$T = 1.017 + 6.07 * \exp(-R/1700) + 40.6 * \exp(-R/170).$$

Results at $H = 146.32$G are shown in figure 11. The sample was first cooled down below T_c after which the field was applied. The blue dots show the magnetization of the sample in reaction to this applied field. After the sample is heated above roughly 6.4K, the magnetization disappears. Now without changing the field, the sample is cooled down again, shown by the orange dots. The sample shows the Meissner-Ochsenfeld effect by spontaneously magnetizing. The small difference (decrease) from the starting value tells us that the effect is not maximal. The curves for the other fields are shown in the appendix.

The size of the MO-effect was determined by dividing the starting (maximum) value by the spontanious magnetization after cooling down. It is shown as a function of the applied field in figure 12. The MOE appears to decrease as a function of the applied field.

Two measurements stand out and are probably false. The effect at 0.548A is higher than would be expected. This is because the initial magnetization was low: lower than the initial magnetization of the measurement before it. It should have been higher due to the higher applied field. The reason for this error could be that something went wrong with the sample heating in between the measurements. It could be that the sample had not cooled down below T_c before the field was

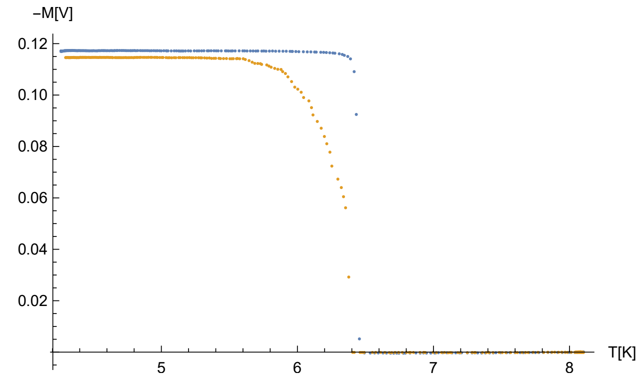

Figure 11: $M(T)$ curve for the sample at $H =$ 146.32G.

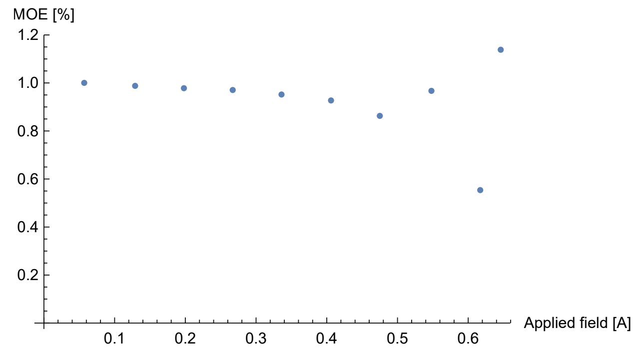

Figure 12: The fraction of the Meissner-Ochsenfeld Effect as a function of applied field.

applied. The last measurement at $H = 0.646$A shows a MOE larger than 1, which should never happen. Measurements at this field strength have been repeated but with no succes. The reason seems to be again that the initial magnetization is lower than expected: lower than the value from the measurement before it.

4.2 Magnetization curves

From the obtained curves of volt (magnetization) as a function of current (applied field) the background signal was subtracted. This time, the background was assumed to be linear with the linearly increasing applied field. The background was fitted to the high-field region where the magnetization was assumed to vanish. Results at temperature $T = 4.56$K are shown in figure 13.

For the different temperatures, the critical field H_c defined as the field at which the magnetization disappears could be determined. Applied primary coil current could be converted to the field H in Gauss using the given relation[6]: $H = 739 * I$ G.

The results are shown in figure 14. They were fitted to the relation

$$H_c \simeq H_{c0}(1 - (T/T_c)^2),$$

which returned the values of $H_{c0} = 805.58$G and $T_c = 7.14$K, in our case.

We recall equation 19 for the demagnetization factor:

$$D = 1 - \frac{H_m}{H_c},$$

where H_c is the field where the magnetization vanishes and H_m is the field at which the magnetization is maximal. The demagnetization factor was determined for all 9 temperatures below $T_c = 7.14$. They are displayed in figure 15. From a theoretical point of view, it should be a geometrical factor and therefore constant with temperature. The found values show quite large deviations but the plotted fit shows that there is some overall increase with temperature. The superconducting properties seem to decrease with temperature.

Finally, the area $A = \int_0^{H_c} M dH$ was determined for every magnetization curve. This area is proportional to the work necessary to convert the sample from a superconductor to a normal

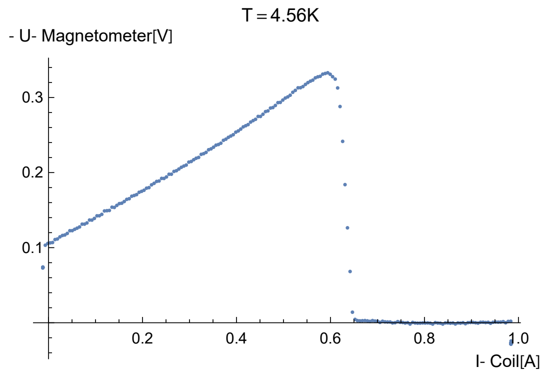

Figure 13: The magnetization, in terms of measured voltage, as a function of the applied field, in terms of ampere applied to the coil. The sample was at an average temperature of 4.56K

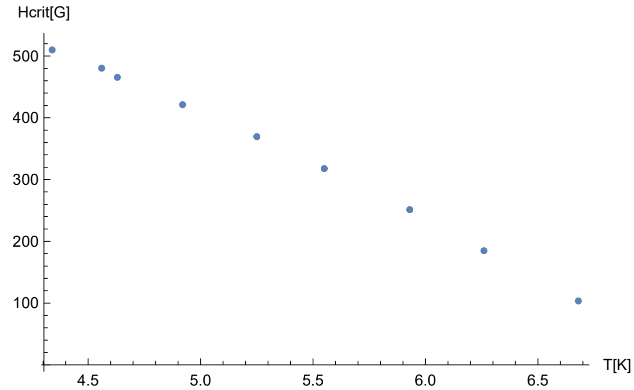

Figure 14: The critical field in units Gauss measured as a function of temperature.

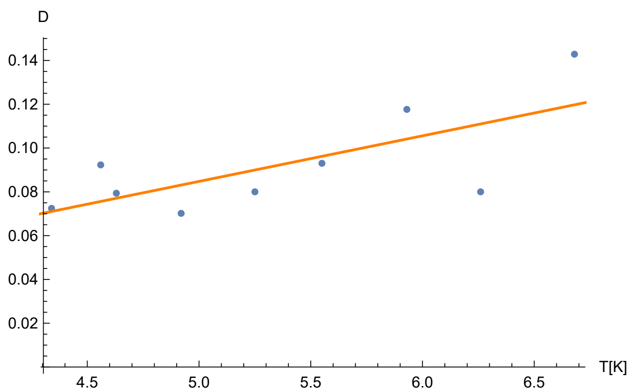

Figure 15: The demagnetization factor as a function of temperature, together with a linear fit.

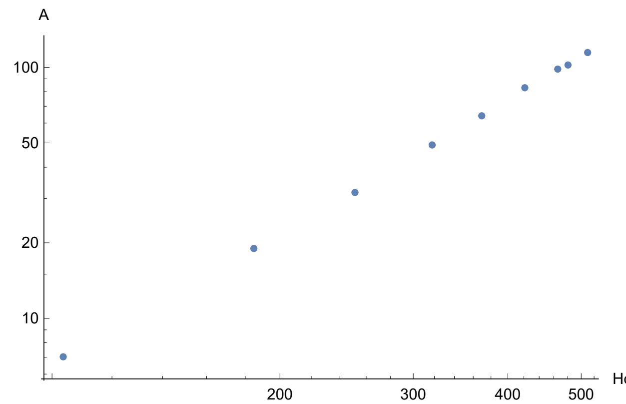

Figure 16: The area underneath the magnetisation curves plotted as a function of the critical field H_c.

state. It should be proportional to H_c^2.[2][6] On a double logarithmic plot, we expect to find a slope of

$$\frac{\log(A_2/A_1)}{\log(H_{c2}/H_{c1})} = \frac{\log(H_{c2}^2/H_{c1}^2)}{\log(H_{c2}/H_{c1})} = 2.$$

The measured values are displayed as a function of H_c in figure 16 on a double logarithmic scale. The slope in this plot was determined to be 1.75.

5 Conclusion

This experiment investigated the behaviour of the magnetization of a superconductor.

First, the magnetization was measured as a function of temperature for 10 different applied magnetic fields. From the heating up and cooling down curves, the Meissner-Ochsenfeld effect could be determined. The effect was shown to decrease for higher applied fields.

Secondly, magnetization was measured as a function of applied magnetic field for 10 different temperatures. From the magnetisation curves, the critical field at which the superconducting properties disappear could be read off. The critical field was shown to decrease with temperature. The critical field H_{c0} was determined to be 805.6G and the critical temperature T_c was determined to be 7.14K. Unexpectedly, the demagnetization factor seemed to increase with temperature. Finally, the area underneath the curves was determined. It was found to be proportional to H_c to the power 1.75, lower than the expected power of 2.

6 References

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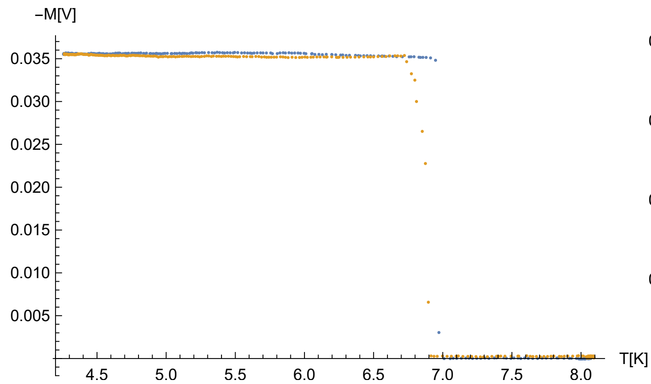
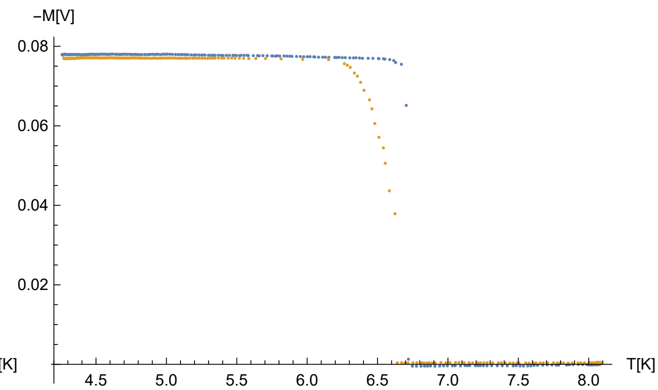
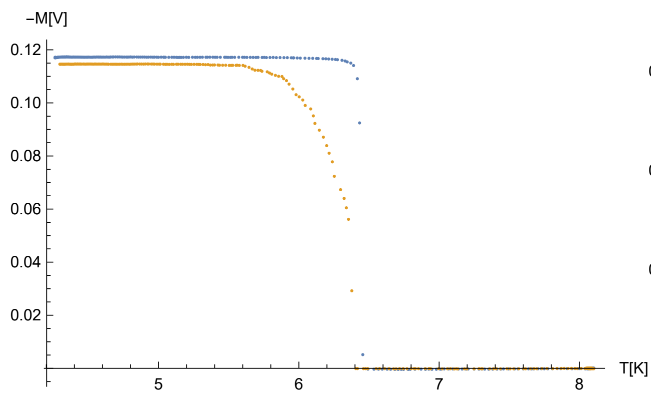
A Supplementary figures



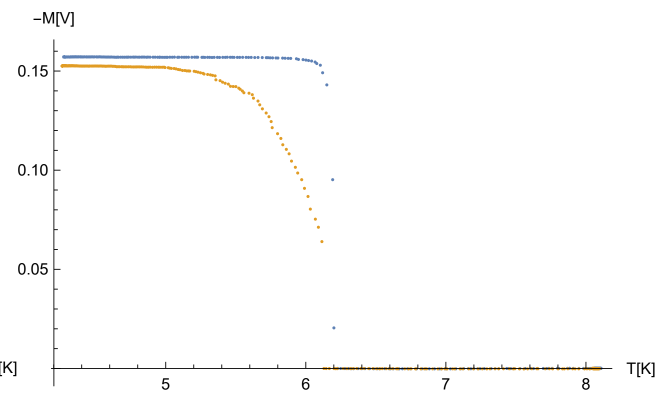
(a) $H = 42.12\text{G}$



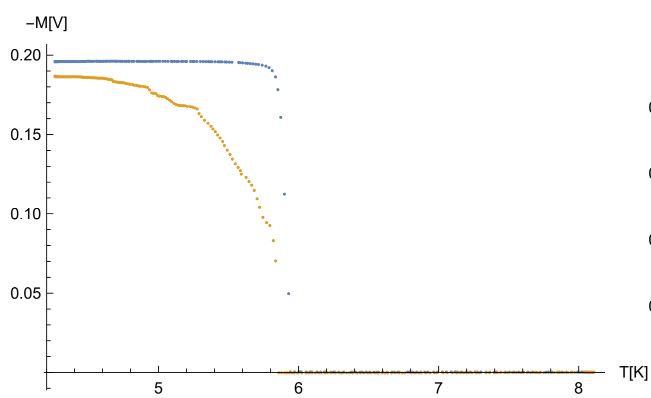
(b) $H = 95.33\text{G}$



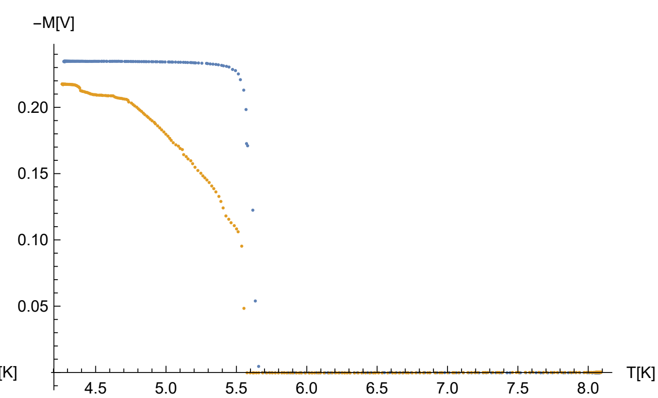
(c) $H = 146.32\text{G}$



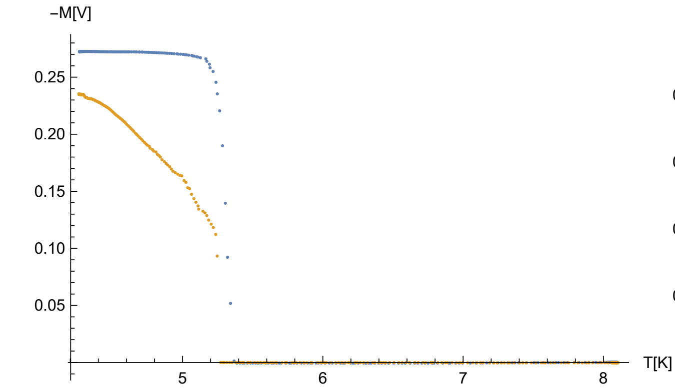
(d) $H = 197.31\text{G}$



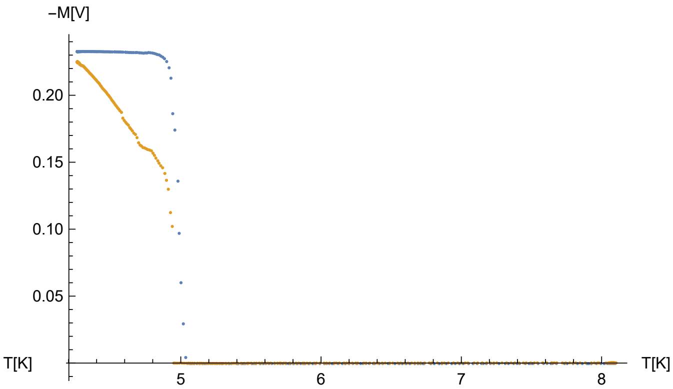
(e) $H = 248.30\text{G}$



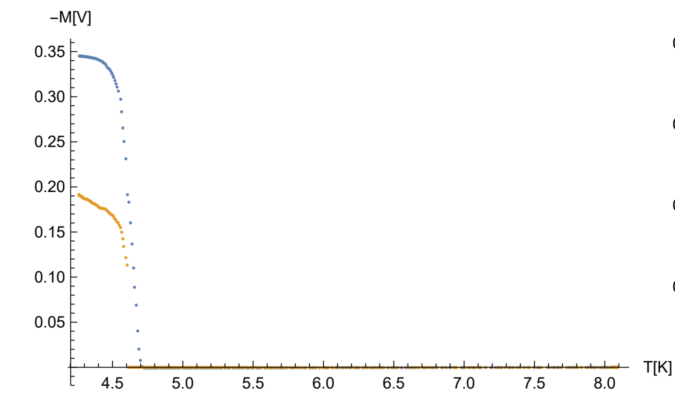
(f) $H = 300.03\text{G}$



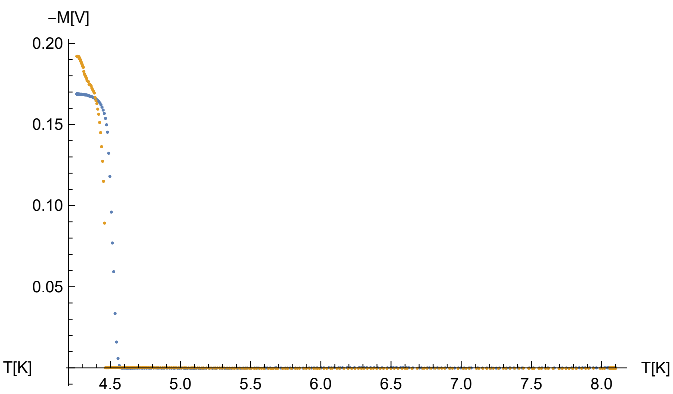
(g) $H = 351.03\text{G}$



(h) $H = 404.97\text{G}$

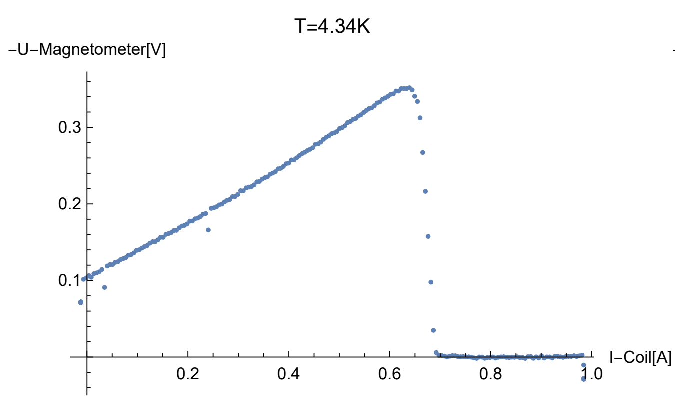


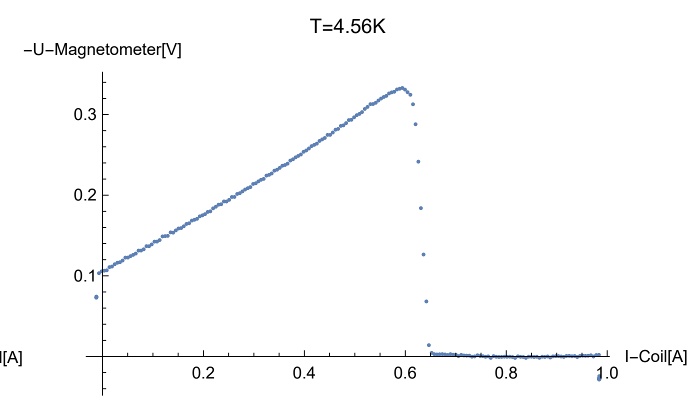
(i) $H = 455.96\text{G}$



(j) $H = 477.39\text{G}$

Figure 16: Magnetisation as a function of temperature for different applied fields.





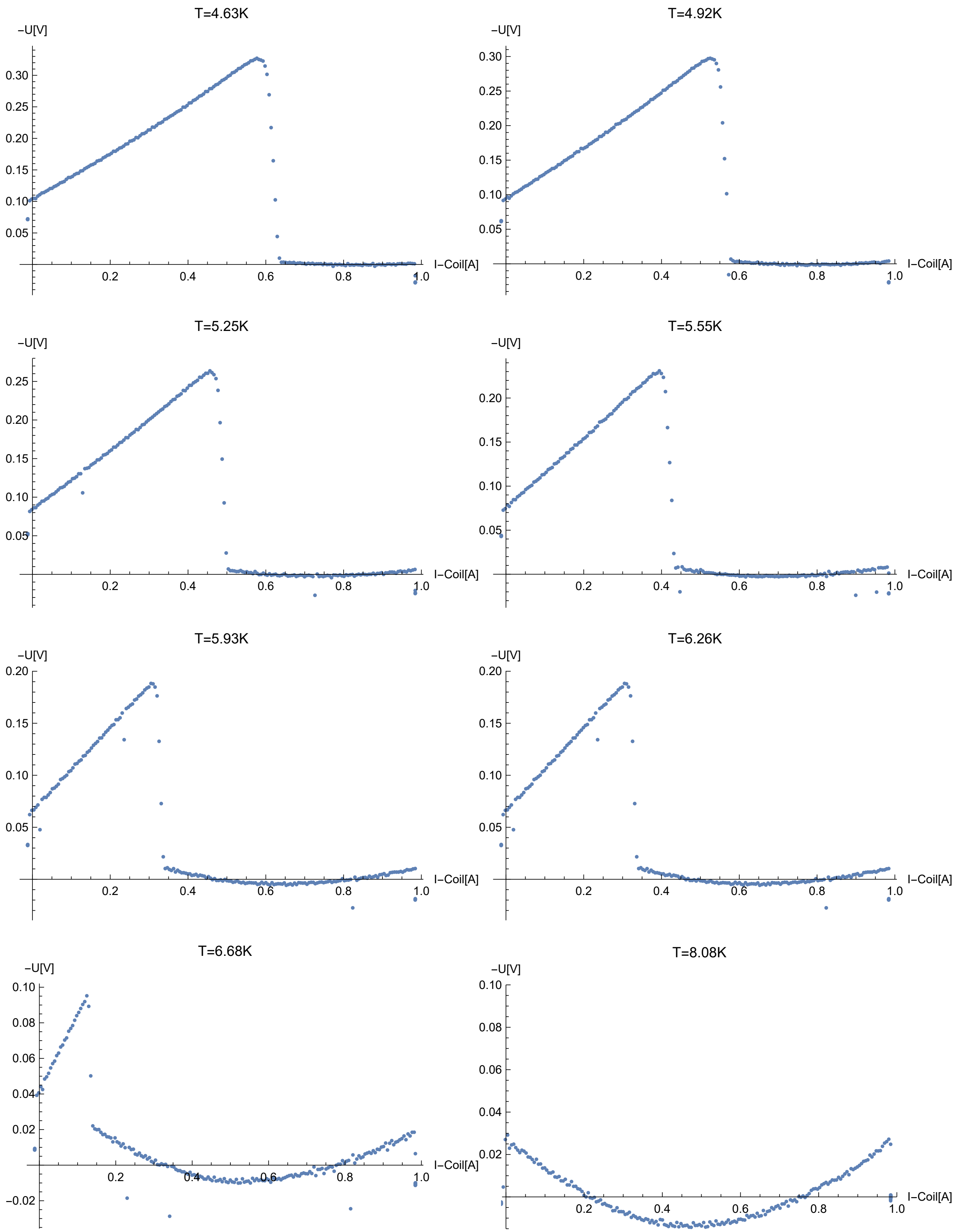


Figure 16: Magnetisation as a function of applied field for diffirent temperatures.