Precise Magnetization Measurements by Parallel Self-Compensated Induction Coils in a Vertical Single-Turn Coil up to 103 T

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We have developed a magnetization measurement technique for use in a vertically aligned single-turn coil. Compensation ratios of an order as high as $10^{-3}$ were achieved by self-compensation in parallel twin-pickup coils without using the electronic compensation circuit mixing of the signal from an auxiliary coil. High performance cryogenics was employed with a liquid Helium container that was also manufactured to fit the system. The high-magnetic-field magnetization measurements were applied to a manganite $\text{Bi}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ at room temperature, and also to a geometrically frustrated spinel oxide, $\text{CdCr}_2\text{O}_4$ at very low temperatures. Not only the transition field, but also the absolute value of the magnetization was shown to be evaluated within a 3% degree of accuracy in magnetic fields of up to 103 T by means of a 14-mm-inner-diameter single-turn coil.

KEYWORDS: magnetization, single-turn coil, ultra-high magnetic field, geometrically frustrated magnet, manganite, low temperature

1. Introduction

A great deal of effort has been made in the technological development of pulse magnets, and recently, a record of a magnetic field above 90 T in a bore of 16 mm was obtained in nondestructive manner.1) Utilizing time variation of the magnetic flux associated with the generation of a pulse magnetic field, the electromagnetic induction method is suitable for the sensitive measurement of magnetization in a variety of magnetic materials. It is widely recognized that a magnetic field above 100 T is only generated in a destructive manner, i.e., by destroying the magnet. The single-turn coil (STC) technique is very useful for solid-state physics measurements in a range of ultra-high magnetic fields of up to 200 T, since the coil only explodes outwards, while the sample and the equipment that holds it are usually kept intact inside the coil, and both the upsweep and downsweep of the magnetic field is applicable in a one-shot measurement. In this regard, the application of various measurements in solid state physics experiments have been reported.2) A peak field of up to 200 T can be generated in a 10-mm-inner-diameter coil with a mega-ampere current injected from 200 kJ fast operating capacitor banks.3) In these circumstances, there is a great need for the development of a precision magnetization measurement system for application to the STC technique in magnetic fields above 100 T.

Application of the electromagnetic induction method to the STC technique was first developed by Takeyama and Amaya, who used the STC with its magnetic field axis aligned in a horizontal direction (called the horizontal STC, abbreviated as H-STC) and obtained the magnetization curve of $\text{CsCoCl}_3$ in magnetic fields up to 90 T at a temperature 4.2 K with a relatively good signal-to-noise ratio.4,5) This direction of research has been continued, with further technical improvement achieved by Goto et al. to obtain a nice magnetization curve of a typical itinerant ferromagnet $\text{YCO}_3$ up to 110 T,6) using the same technique as was used in refs. 4, 5.

Using a single-turn coil installed at Humboldt University in Berlin, where the coil is aligned in a vertical direction (called vertical STC, abbreviated as V-STC),7) Kirste et al. have taken a great care in the winding of the pickup coil and achieved compensation ratios of better than $10^{-3}$ in two alternatively wound parallel pickup coils. In addition, magnetization measurement in magnetic fields of up to 130 T was applied to bismuth-based manganesees ($\text{Bi}_{0.5}\text{M}_{0.5}\text{MnO}_3 \text{M = Ca, Sr}$) in order to investigate the nature of the ferromagnetic phase in terms of its relevance to the charge order phase.8)

All of these data, however, are only sufficient to determine the transition points of a magnetic field from the direct signal of the time derivative of magnetization, $dM/dt$, in the magnetic phase diagram of interest, while the signal-to-noise ratio and the level of the background signal are insufficient to provide details of the absolute values of magnetization, $M$, in magnetic fields. There are three major obstacles associated with measurements in a singular environment of the STC. The first is the trigger starting electro-magnetic noise arising from high voltage (35–50 kV) and from the huge current (2–3 mega ampere) injected into the coil. The second is the poor spatial homogeneity of a magnetic field inside the small STC. The third problem is the deformation of the coil during the pulse duration, due to the explosive nature of the coil. It is particularly distressing to obtain high compensation ratios, which are required for accurate magnetization measurement, in the magnetic pickup coil. In this paper, we present a technical approach that enables us to establish a precision magnetization measurement system in a V-STC, with almost comparable quality obtained by the nondestructive long pulse magnet.

2. Experimental Setup

A schematic diagram of the entire experimental setup is shown in Fig. 1. The energy stored in a fast condenser bank (0.125 mF) is discharged to a V-STC in order to generate a magnetic field that exceeds 100 T. The details are presented in ref. 9. The $dM/dt$ from a sample is detected as a voltage drop that is generated in a pair of magnetic pickup coils and then transferred to an oscilloscope through shielded coaxial
cables. A double-walled shielding box is used in order to shield the digital oscilloscope against the enormous discharge noise that accompanies high field generation. It is placed close to the anti-explosion chamber of the STC in order to minimize the length of the co-axial cables for signal transmission. The outer wall of the shielding box is made of iron with a thickness of 2.5 mm, and the inner wall is made of aluminum with a thickness of 3.0 mm. The magnetic field is measured by the pickup coil, which is wound 4 turns around a polyimide tube (a diameter of 1.62 mm) by a copper wire (a diameter of 60 $\mu$m), which is coated with polyamide imide to provide high electrical insulation.

### 2.1 Magnetic pickup coils

In experiments that employ a pulsed magnetic field, a huge voltage is momentarily induced across the pickup coil. This induced voltage reaches as much as 2 kV in a pickup coil that is wound 20 turns around a rod with a 1 mm diameter in a field of 100 T. Two kinds of parallel-type magnetic pickup coils have been proposed as optimal for STC experiments: one-way counter (parallel) winding and alternative winding, which are shown in Figs. 4(b) and 4(c), respectively, in ref. 4. We have ensured that parallel one-way counter winding with 28 turns can survive magnetic fields of up to 100 T, due to improvement of the electrical insulation of the polyamide-imide-coated copper wire, which ensures insulation up to 6 kV.

We adopted the parallel-type magnetic pickup coils and wound the 60-$\mu$m wire 20 turns around a Kapton tube with a diameter of 1.12 mm diameter and a thickness 0.06 mm. A photograph of the magnetic pickup coil wound in this way is shown in the photo inset of Fig. 1. When the magnetic pickup coil was properly positioned inside a coil, the induced voltage of 2 kV was reduced to less than 1 V, which led to compensation ratios of less than $5 \times 10^{-4}$, without an assist of any auxiliary compensation coils. In the past, the residual uncompensated signal that results from the imbalance between the two opposite windings has thus far been reduced further down to a factor of $10^{-2}$, by means of the electronic compensation circuit mixing of the signal from an auxiliary coil. Unlike the case of a long pulse magnet, the pulse field produced by the STC operates at several MHz. Therefore, complete compensation obtained by the mixing of the signal from the auxiliary coil is made difficult only by the resistance network. It is crucial for self-compensation to take place only through the pair of counter windings in order to achieve actual compensation of the magnetization data in the STC.

### 2.2 Magnetic field distribution inside a V-STC

We measured the magnetic fields of different positions inside a 14-mm-inner-diameter STC simultaneously by means of five pickup coils (see Fig. 2), each of which was calibrated. The coils were wound 4 turns around 1.12-mm-diameter Kapton tubes that were fixed vertically at the same height with horizontal distance of 2 mm between each tube [inset photo of Fig. 2(a)]. A peak magnetic field of over 100 T was generated, which was discharged from a 100 kJ fast operating condenser bank with 40 kV. The results are shown in Fig. 2 as a result of the simultaneous measurement of magnetic fields at 5 positions in a horizontal $x$ direction inside a V-STC with a 14-mm-inner diameter. The $x$ and $y$ (radial) direction is horizontal, and the $z$ direction is that of the magnetic field [as indicated in the inset of Fig. 2(b)]. The overall pulse waveforms from the 5 pickup coils are almost identical [Fig. 2(b)]. However, at around a peak field, when the scale is enlarged [Fig. 2(a)], differences can be noted not only in the maximum value itself, but also in the time at the maximum of a pulse magnetic field. The peak field is the lowest at the center of the coil, and it becomes larger as it is closer to the inner wall of the coil (off-center). We noted an
increasing time delay at the peak for positions away from the center of the coil. These phenomena arise from inhomogeneous deformation of the coil during pulse generation prior to complete destruction.

The distribution of the magnetic field was measured in the $x$, $y$, and $z$ directions, respectively, inside the coil, using the same probe as that shown by the photo in the inset of Fig. 2(a). The results are shown in Fig. 3, in which the peak field of each position of the pickup coil is plotted at the moment when pickup coil No. 3, positioned at the center of a coil, reached its maximum. The obtained results are quite similar to those in refs. 4,5, in which the field gradient near the center of the coil in the $z$ direction is almost twice as it is in the radial ($x, y$) direction. The field distribution of the $y$ direction, which is presented for the first time in this work, shows the decrease in intensity of the field along with the direction toward the injection current feed gap, with a gap size of 2 mm [see the inset of Fig. 2(b)].

Due to the deformation of the coil during the explosive outward movement of the magnetic coil, it is important to study the time dependences of the magnetic field distribution inside the coil. The time evolution of the field deviation relative to that at the center of the coil is plotted for the horizontal $x$ and $y$ directions, in Figs. 4(a) and 4(b), respectively. There is a minimal deviation of the field in time between Nos. 2 and 4 in the $x$ direction [Fig. 4(a)], as compared to the other combinations. Therefore, in order to minimize the uncompensated background signals, the best position for the pair of magnetic pickup coils should be a position between Nos. 2 and 4, along the $x$ direction shown in the inset of Fig. 2.

Schematics of the magnetic probe and its component are presented in Fig. 5. The magnetic pickup coils are positioned precisely at the center of the single-turn coil, with the pair aligned along the $x$ direction. The outer diameter of the tube in which the pickup coils are held was designed to fit the innermost wall of a cryostat with an inner diameter of 6 mm. The use of conductive metals should be avoided near the STC in order to prevent problems caused by eddy-currents. The resin polyoxymethylene (Derlin) was used as the material for the tip of the probe, where the pickup coils were held tight by an adhesive called "Stycast". Reinsertion of a sample from one magnetic pickup coil to the other was carried out by a top loading mechanism, which is shown in Fig. 5(c). The sample is pulled up into the chamber (B) following its measurement by one magnetic pickup coil, and is then reinserted into another tube (C), which guides it to the other magnetic pickup coil via the rotation of an insertion rod (A). This sample exchange process is easily confirmed by the naked eye through a transparent tube (B). A thin Kapton tube with a 0.9 mm diameter that is fit to the inner tube of magnetic pickup coil was used to hold the samples, whether in powder or bulk form, and was fixed in place with Stycast resin.

![Fig. 2.](image)

![Fig. 3.](image)
A cryostat was specifically designed for the V-STC system as shown in Fig. 6. The upper part of the cryostat is made of stainless steel, and is comprised of a 0.36 L liquid He container with a tail section made of glass epoxy (fiber-reinforced plastic; FRP) thin tubes. Because there needs to be an insulating Kapton sheet inside the STC, which has a diameter of 14 mm, the diameter of the outermost tube of the cryostat tail section is limited to less than 13.4 mm. Hence, in order to keep a sample space that is 6 mm in diameter, four thin separating walls with two vacuum insulation layers and one liquid nitrogen layer should be incorporated within a gap of $\frac{(13.4 - 6.0)}{2} = 3.7$ mm. The thickness of the tube walls is 0.7 mm for both the innermost and the outermost walls, and 0.5 mm for the other two walls. Thus, the spaces of the inner vacuum insulating layer, the liquid N\textsubscript{2} layer, and the outer vacuum insulating layer are designed to be 0.3, 0.5, and 0.5 mm, respectively. The FRP thin tubes are connected smoothly to the upper stainless steel tubes by a cryogenic epoxy adhesive (Nitofix SK-229). Liquid He is maintained for 2–3 h after the liquid He container is completely filled.

3. A Manganite Bi\textsubscript{0.5}Ca\textsubscript{0.5}MnO\textsubscript{3} at Room Temperature

Rare-earth manganites with perovskite-type structure have been the subject of large interest during these decades as they show diverse physical phenomena as a result of interplay among charge, spin and orbital orders. With introducing a certain fraction of carriers by chemical substitution of divalent cations for the rare-earth ions, the doped manganites show periodic order of charge and orbital. In general, this charge/orbital order becomes more stable as the average ionic radius of the rare-earth site decreases. However, bismuth-based manganites exhibits unusually stable charge/orbital order beyond this general empirical rule in the rare-earth manganites. To realize the field-induced melting of the charge/orbital order in this class of
materials, extremely high magnetic field is required. We have chosen $\text{Bi}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ as a test sample for measurements at room temperature as it shows strong magnetic like transition at very high magnetic fields.

Magnetization measurements of $\text{Bi}_{0.5}\text{Ca}_{0.5}\text{MnO}_3$ in the mega gauss region using a STC system were first performed by Kirste et al. in a high magnetic field laboratory in Germany. Clear peaks associated with the first order magnetic phase transition with a large hysteresis have been observed in a $dM/dt$ signal in measurements up to 130 T. In the present measurements, a powdered sample was used that had a volume of about 1.3 mm$^3$. The total weight was 6.0 mg, which was expected to produce a value of 0.64 emu as the saturation moment (it is known that the saturation magnetization is 3.5 $\mu_B$/Mn). The measurements were carried out at room temperature (290 K). A magnetic field was generated from a 100 kJ, 40 kV condenser bank with a STC with an inner diameter of 14 mm. During the experiment, a high frequency discharge noise was always recorded at the start of the pulse field, coming mainly from the air gap switches for huge electric current (an order of mega ampere) and high voltage (40 kV) operation. This noise was then reduced to a few percent of the original noise by the subtraction process, followed by signal smoothing. In two experiments, a pair of $dM/dt$ signals was obtained by a pair of magnetic pickup coils, which were the right and left pickup coils under identical conditions. The background signals were almost completely canceled out by subtracting one from the other, and in Fig. 7, the results are plotted versus the magnetic field. A very sharp peak appeared with a negligible amount of a background signal in $dM/dt$. The position of the peak was different in the upsweep and downsweep of the magnetic field, showing a large hysteresis. This is indicative of a first order magnetic phase transition accompanied by a sudden change in magnetization.

A striking decrease in the background signals in Fig. 7 was noted as compared to those in ref. 9. At this point, it was worth integrating the $dM/dH$ data in order to obtain a full magnetization curve; the results are shown with the data taken by a long pulse magnet up to 60 T at a temperature 290 K in Fig. 8. There, we can observe a little deviation between the data in the upsweep and downsweep of the magnetic field at the region of lower magnetic fields. This is caused mainly from deformation of the STC at very end of the pulse operation, which caused changes in spatial distribution of magnetic fields inside of the STC. However, there is almost a coincidence between the upsweep and downsweep signals within an error of 3%, except for the enhanced hysteresis. The quality of the data is comparable to those obtained by a long pulse magnet. The absolute value of the magnetization curve can be evaluated reliably within an error of 3%. The remarkable difference to be noted is the width of the hysteresis. The widening of the hysteresis arises from the fact that the sweep rate of the magnetic field in the STC is much faster (three orders of magnitude) than it is in the long pulse magnet. This increased width of the hysteresis is consistent with the first order transition associated with lattice deformation, which creates slow transition dynamics. This is the first data that reliably shows the magnetization curve in the upsweep and downsweep of a magnetic field of up to 103 T obtained in the STC system.

4. A Chromium Spinel $\text{CdCr}_2\text{O}_4$ at 4.2 K

Geometrically frustrated magnet systems have been a fascinating subject in recent years because they have macroscopically degenerated ground states vulnerable to small perturbations (spin–lattice interaction, quantum and thermal fluctuation, etc.), which results in the emergence of exotic magnetic states. Magnetic fields cause the appearance of peculiar magnetic phenomena such as the magnetization plateaus, magnetic supersolids and crystalline magnon states as a result of interplay between a magnetic field and frustrated spin exchange interaction. Among the numerous frustrated magnets, $\text{CdCr}_2\text{O}_4$ is recognized as one of the typical three-dimensional frustrated magnets exhibiting a very high Curie–Weiss temperature and a low Neel temperature ($T_N/\theta_{CW} = 8 \text{ K}/70 \text{ K} \sim 0.1 \ll 1$). This large discrepancy is caused by suppression of the occurrence of a magnetic order by the geometrical frustration, which requires multiple extreme physical conditions, such as ultra-high magnetic fields and very low temperatures, for thorough understanding of the magnetic ordering.
Ueda et al. reported magnetization of CdCr$_2$O$_4$ in magnetic fields up to 50 T using nondestructive long pulse magnets. They found a first-order phase transition at 28 T followed by a wide magnetization plateau with a half-of-full moment (1/2 plateau). After this, Mitamura et al. have attempted measurements in magnetic fields up to 80 T by the induction method in the H-STC at 6 K. They could have shown a second order phase transition at 61 T and a first order phase transition at 77 T inferred only from the downswEEP data of $dM/dt$. Signals from upsweep data were difficult to analyze due to large ignition noises. Lately, Kojima et al. have applied a Faraday rotation method to this material for evaluation of magnetization. They have carried out systematic measurements in wide temperatures in magnetic fields up to 140 T by the H-STC, and the magnetization processes until its full saturation moment are thoroughly revealed in wide range of temperature.

In the present experiment, powdered samples were prepared for measurements with the net-mass of 5.4 mg. The expected saturation magnetization is 0.71 emu with the assumption that the saturation magnetization is 3.0 $\mu_B$/Cr. The results of the magnetization measured up to 100 T with upsweep and down sweep are presented in Fig. 9 together with the data taken from the Faraday rotation measured at 7 K. The apparent difference noted here is the width of hysteresis associated with the first order transition to the 1/2 plateau at $B = 28$ T, which is caused by the difference in the rate of the magnetic flux change between the two experiments. Another deviations visible in magnetic fields in 30–50 and 65–70 T are supposed to arise from the subtle difference in measurement temperature between the two experiments. Both data coincide well with each other within the error of 3% with respect to the full saturation moment. In the inset, the data is also compared with those measured by the long pulse magnet in magnetic fields of up to 55 T. Owing to slow rate of the flux change subjected to the long pulse magnet, a width of the hysteresis is almost invisible in this figure, but the transition occurred at a midpoint in magnetic fields between the hysteresis observed in the data taken by the present measurement. Striking coincidence is obtained in the two data.

5. Summary

We have developed magnetization measurement techniques for use in the V-STC system. Based upon careful winding and selective positioning in a magnet coil with an inner diameter of 14 mm, compensation ratios of $5 \times 10^{-4}$ were achieved in a pair of magnetic pickup coils without an assist of any external compensation. A cryogenic temperature cryostat containing liquid Helium was designed to fit the V-STC and was then manufactured. The developed system was applied to the magnetization measurement of Bi$_2$O$_2$Ca$_{0.5}$MnO$_3$ at room temperature and the frustrated magnet CdCr$_2$O$_4$ at 4.2 K. Background signals were substantially suppressed in comparison to those obtained in a previous report in which the STC was used. The magnetization curves obtained for Bi$_2$O$_2$Ca$_{0.5}$MnO$_3$ and CdCr$_2$O$_4$ were of almost as high a quality as that obtained by a conventional non-destructive long pulse magnet. This is the first work in which the magnetization curve has been reliably obtained in both upsweep and downswEEP of a magnetic field of up to 100 T. An accurate magnetization curve (not only for $dM/dt$ but also for $M$) can be obtained in magnetic fields of up to 103 T and at temperatures from room temperature down to as low as 4.2 K. Thus, not only the magnetic phase transition point, but also the absolute values were evaluated from the measurements. The system developed here may serve as a substantial contribution to the research in magnetism at the region of mega-gauss magnetic fields.

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