Measuring nuclear magnetization in strong magnetic fields

Akira Harita [a], Takashi Tayama [a], Takahiro Onimaru [a], Toshiro Sakakibara [a,1]

[a] *Institute for Solid State Physics, University of Tokyo, Kashiwa, 277-8581, Japan*

Abstract

Al nuclear magnetization has been measured in magnetic fields up to 100 kOe, by use of a capacitive Faraday magnetometer installed in a dilution refrigerator. The sample employed was an $Al_{0.98}Si_{0.02}$ alloy prepared from high purity (6N) elements. The Si doping was essential in suppressing the de Haas-van Alphen oscillations of conduction electrons. Fine Curie's law was observed at temperatures below 3 K down to ∼60 mK, in several magnetic fields in the range 30 kOe∼100 kOe. The measurement opens a new route towards primary thermometry at low temperature and high magnetic field region.

Key words: thermometry; nuclear magnetization; high magnetic field;

1. Introduction

Cryogenic thermometry in high magnetic fields has been a difficult challenge, in particular at low temperatures accessible by dilution refrigerators. In most cases, resistance thermometers such as ruthenium oxide thick film resistors or zirconium oxynitride thin film resistors are used in these conditions since they offer the possibility of relatively small temperature errors in magnetic field. For precise temperature measurements, these resistors need to be calibrated in magnetic fields because of their non-simple magnetoresistance at low temperatures [1,2].

Regarding primary thermometry, various types of nuclear magnetic thermometers have been used at vary low temperatures and low magnetic fields [3–5]. However not much work has been done in high magnetic fields. Candela *et al.* reported Pt NMR thermometry in high magnetic field of 80 kOe [6]. Very recently, Coulomb blockade based thermometer has been claimed to provide absolute thermometry that is virtually insensitive to magnetic fields [7].

In this paper, we report on an attempt to extend the nuclear magnetic thermometry to high magnetic fields. By use of the Faraday method, dc magnetization of Al nuclear spins could be measured with high resolution at temperatures below 100 mK in magnetic fields up to 100 kOe.

2. Experimental

2.1. *Magnetometer design*

The principle of the magnetization measurement is as follows. A sample, situated in a region where a magnetic field is spatially varying, feels a force proportional to its magnetization M and the field gradient. This force can be detected by a force-sensing parallel-plate capacitor; one of its electrodes can move in proportion to the applied force. The capacitor employed in our experiment is shown in Fig. 1. The upper plate of the capacitor (10 mm diameter) is made of epoxy (Stycast #1266) and its electrode is formed by silver paint. The sample stage of the upper plate is made of 5N copper. These are suspended by four phosphor-bronze wires of 0.1 mm diameter from x and y directions. The loaded capacitance value was about 7 pF, and once cooled below 10 K it was stable to 10^{-5} pF.

[1] E-mail:sakaki@issp.u-tokyo.ac.jp

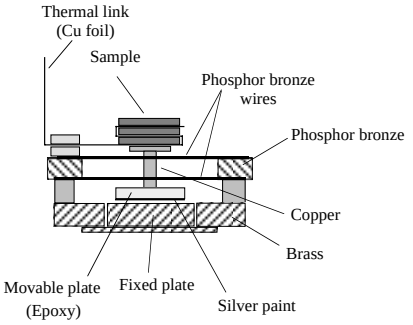

Fig. 1. The force-sensing capacitor for the Faraday magnetometer.

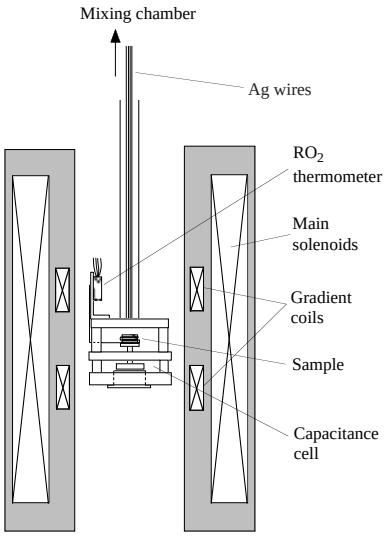

Fig. 2. The experimental setup for the magnetization measurement.

Figure 2 shows the experimental setup of the Faraday magnetometer. Magnetic fields H up to 100 kOe were produced by a superconducting solenoids whose homogeneity at the sample position is 10^{-4} over a 10 mm-diameter spherical volume. A vertical field gradient dH/dz of 800 Oe/cm was generated by gradient coils wound inside the magnet. The capacitor was cooled by a dilution refrigerator via a thermal link of Ag wires connected to the mixing chamber.

We used a calibrated ruthenium oxide thermometer (RO-600, Scientific Instruments Inc.) mounted on the capacitor. The resistance measurements were made with a low power AC resistance bridge (AVS-47, RV-Electroniikka). In this experiment, we tentatively corrected a magnetic field effect on the thermometer using a simple equation provided by the company: $T^*/T^*(H) = 1 + 1.31 \times 10^{-3}H - 2.93 \times 10^{-6}H^2$, where T^* is a corrected temperature and $T^*(H)$ is an apparent temperature reading of the thermometer in magnetic field H (kOe).

In a normal operation, we made two sweeps of temperature or field for each measurement, first with the gradient coils switched on and second with no current in the gradient coils. Taking a difference between these two capacitance data, we could eliminate the effects of a magnetic torque or a field inhomogeneity of the main solenoids. More details of the measurement method are given in ref. [8]. The resolution of the magnetometer was better than 10^{-5} emu in a field of 100 kOe. Absolute sensitivity was calibrated by measuring a Ni standard sample with 1 % accuracy.

2.2. *Sample Preparation*

Various metals have been used as magnetic thermometers at very low temperatures [3–6]. Among these, aluminum (^{27}Al, $I = 5/2$) would be most favorable for the present purpose. One of the merits of using Al is that it has a large value of the unit-mass Curie constant (2.55×10^{-8}emu·K/g), a factor of 5 larger compared to Cu. This is a significant advantage for the Faraday magnetometer. We first attempted the measurements with high-purity (6N) Al metal, and the magnetization curve obtained at 0.4 K is shown in Fig. 3 by open circles. Unfortunately, we observed a strong de Haas-van Alphen (dHvA) oscillations in spite of a polycrystalline sample, and could not detect the nuclear contribution.

In order to suppress the dHvA oscillations, we prepared a 2-at.% Si doped Al-Si alloy by arc melting the high-purity (6N) materials. The dots in Fig. 3 are the magnetization of the Al-Si alloy, indicating that the dHvA oscillations are completely wiped out by the non-magnetic 'impurity' scatterings. Magnetically, Si does not contribute to the Curie constant because of the small nuclear spin of ^{29}Si ($I = 1/2$) and its poor natural abundance (4.7%). The Si doping might cause a quadrupolar splitting of the ^{27}Al nuclear spins due to locally destructing a cubic sym-

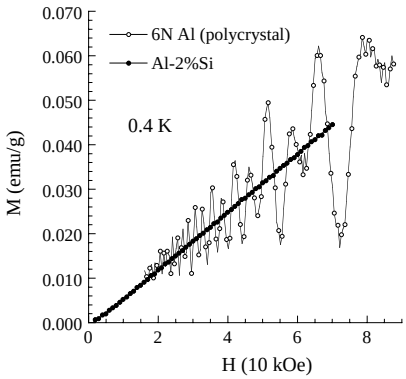

Fig. 3. Magnetization curves of the 6N polycrystalline Al (open circles) and the Al-2%Si alloy (dots), obtained at 0.4 K.

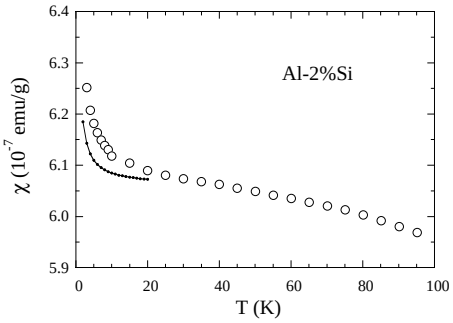

Fig. 4. Magnetic susceptibility of the Al-2%Si alloy above 2 K (open circles) measured by a SQUID magnetometer in a magnetic field of 5 kOe. The dots are the sum of the calculated contribution of Al nuclear spins $2.495 \times 10^{-8} T^{-1}$ emu/g and the Pauli paramagnetic susceptibility of $\chi_0 = 6.06 \times 10^{-7}$ emu/g

metry, but this effect on the nuclear spin susceptibility would be small in the temperature range of interest ($>$ 10 mK). The linear magnetization of the Al-Si alloy observed in Fig. 3 mainly comes from Pauli paramagnetism of conduction electrons. We precisely determined the linear paramagnetic susceptibility at 2 K to be 6.18×10^{-7} emu/g, through the magnetization measurement up to 70 kOe using a commercial SQUID magnetometer (MPMS, Quantum Design). This susceptibility can be decomposed into the Pauli term of $\chi_0 = 6.06 \times 10^{-7}$ emu/g and a calculated nuclear contribution of 1.25×10^{-8} emu/g.

Figure 4 shows the high-temperature behavior of the susceptibility $\chi(T)$ of the alloy, measured by the SQUID magnetometer at temperatures above 2 K in a field of 5 kOe. $\chi(T)$ is a gradually decreasing function of T above 20 K. Below 10 K, a steep increase can be seen in $\chi(T)$. A part of this upturn can be attributed to the Al nuclear magnetization, as indicated by the dots in the figure. It turns out that the sample contains magnetic impurities of order 1 ppm in concentration. While these impurities have detectable effect in this 'high-temperature' low field range, they are fully saturated and will not contribute to the susceptibility at low temperature ($<$ 1 K) and high field (>30 kOe) regions which are of our primary interest.

The alloy ingot was cut into slabs of the size $1.1 \times 5 \times 7$ mm^3, three of which were then stacked together with varnish, with thin Cu foils (10 μm thickness) sandwiched in between them for a thermal contact as shown in Fig. 1. Total weight of the alloy used in the present measurement was 0.319 g.

3. Results and Discussion

We first measured the background magnetization of the empty capacitance. The background is found to be

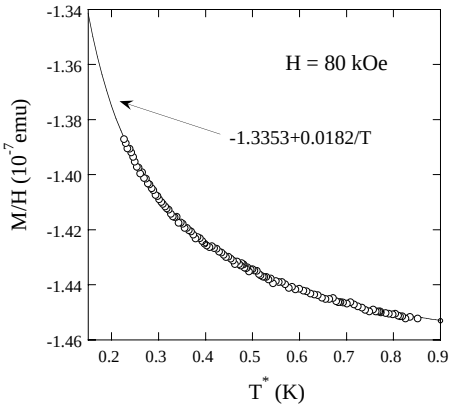

Fig. 5. Background susceptibility of the empty capacitance cell measured at 80 kOe, plotted as a function of the corrected temperature reading T^* of the ruthenium oxide thermometer. The solid line is a fit to the Curie's law with a constant term: $M/H = (-1.3353 + 0.0182/T) \times 10^{-7}$ emu.

linear in H, and Fig. 5 shows the result for M/H obtained under the field of 80 kOe, plotted as a function of the corrected temperature reading T^* of the ruthenium oxide thermometer. The data (open circles) can be well fitted by a small Curie term with a constant negative background: $M/H = (-1.3353 + 0.0182/T) \times 10^{-7}$ emu (solid line). The negative T-independent background is due to diamagnetism of the movable plate, whereas the source of the Curie term would be the nuclear magnetization of the copper stage ($\sim$ 0.35 g weight). The background Curie term is a factor of 5 smaller compared to the expected contribution of Al nuclear spins in the sample. Hereafter the background magnetization is subtracted from all of the data.

Figure 6 shows the observed magnetization change of the sample (open circles) in magnetic field of 50 kOe, plotted as a function of T^*. The data were taken by

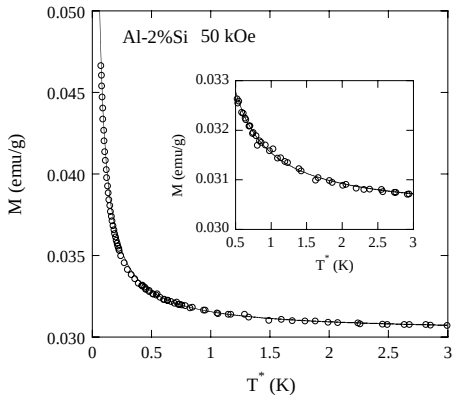

Fig. 6. Magnetization of the Al-2%Si alloy measured in magnetic field of 50 kOe plotted as a function of T^*. The solid line is a sum of the calculated Al nuclear spin moment and the Pauli paramagnetic susceptibility. Inset is an enlarged plot for $T > 0.5$ K.

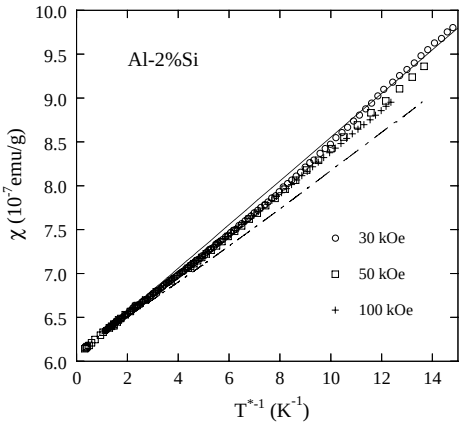

Fig. 7. Magnetic susceptibility of the Al-2%Si alloy measured at 30 kOe, 50 kOe and 100 kOe plotted as function of reciprocal temperature read of a RO_2 thermometer, corrected for the field effect. The solid line is the sum of the calculated Al nuclear spin susceptibility and the Pauli term. The dot-dashed line shows the data for 100 kOe plotted against the uncorrected temperature reading $T^{*-1}(H)$ of the thermometer.

slowly cooling the sample from 4.2 K. Since there was some uncertainty in the initial capacitance value of the magnetometer, we subtracted a small constant background from the data so that the magnetization value at 4.2 K becomes equal to the calculated one. The solid line is a calculated magnetization M(emu/g)= $H(\chi_0 + 2.495 \times 10^{-8}/T)$, which is in agreement with the data points at low temperatures. Temperature resolution was better than ±10 mK and ±1 mK at 0.5 K and 100 mK, respectively. Contribution of the nuclear magnetization is well resolved even at ∼3 K as shown in the inset.

We continued the similar measurements at various magnetic fields and the results for 30 kOe (open circles), 50 kOe (open squares) and 100 kOe (crosses) are shown in Fig. 7 as function of T^{*-1}. The solid line in the figure indicates the calculated variation of the total susceptibility of the sample. We found that the data points show a weak deviation from the expected Curie behavior below ∼300 mK. At 200 mK, the difference between the observed and the calculated magnetization values reached $\Delta\chi \simeq 0.09 \times 10^{-7}$ emu/g, which corresponds to a temperature-reading error of $\Delta T^* \simeq$ 16 mK. ΔT^* became smaller at lower temperatures. A part of ΔT^* (∼2 mK) probably originates from the sensitivity uncertainty of our magnetometer. By monitoring a temperature reading of the mixing chamber where the field is compensated, however, we have a feeling that the field correction for T^* is still insufficient.

The dot-dashed line in Fig. 7 is the data for 100 kOe plotted against the uncorrected temperature reading of the thermometer, $T^*(H)$. Evidently, correction for the magnetic field effect on the thermometer is crucial. Below 100 mK, the data plots become field dependent. This fact implies that the field effect on the thermometer cannot be compensated by the simple correction function, in agreement with the work by Goodrich *et al.* [1] reporting that the magnetoresistance of the ruthenium oxide thick film resistor shows a non-monotonic field dependence below 120 mK. The scattering of the corrected temperature reading for these three fields is however within ±3 mK at 80 mK.

At the field of 10 T, cooling below 1 K down to the base temperature required ∼6 h for equilibrium. While we do not know the origin of this slow rate quite well, a part of this is probably due to a large nuclear specific heat of Al in high magnetic field, $C_{\rm n}$(erg/g·K)= $2.54 \times 10^{-8}(H/T)^2$, and a relatively weak thermal link to the sample which could be made only through a thin Cu foil. The spin-lattice relaxation time of Al expected from the Korringa law ($t_1 = 1.8T^{-1}$ sec) [4] is much shorter in this temperature range.

To summarize, we have successfully measured the nuclear magnetic moment of an Al-Si alloy at temperatures down to below 100 mK in magnetic fields up to 100 kOe by a Faraday method. This technique would lead to absolute thermometry at low temperatures and in high magnetic fields.

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