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# APPLICATION OF SQUID MAGNETOMETER TO NUCLEAR MAGNETIC THERMOMETRY*

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## ABSTRACT

This paper presents an application of a SQUID magnetometer for low temperature thermometry using the magnetic properties of a nuclear paramagnet. The static magnetization of a material which obeys Curie's law provides a very sensitive means of thermometry. Also included in the device is the capability to observe, also with a SQUID magnetometer, the nuclear spin-lattice relaxation time $\tau_1$ using NMR methods. This allows a temperature self-calibration of the thermometer system. As an example of a suitable nuclear paramagnet, magnetization and relaxation time data are presented for aluminum.

## I. INTRODUCTION

SQUID magnetometers have been commonly used to measure temperature dependent changes in the static magnetization of many substances. With various paramagnetic salts, such as CMN, or nuclear spin systems which have known Curie law behavior such a magnetometer arrangement gives one a simple, yet highly sensitive and continuously measuring thermometer. For ultra-low temperatures the paramagnetic behavior of nuclear spins in metals provides a very useful thermometer since Curie's law is expected to hold to well below the MilliKelvin region. Moreover the sensitivity of a SQUID magnetometer makes it ideal for measuring the relatively small nuclear magnetizations. One possible disadvantage lies in the fact that these thermometers are normally calibrated over only a limited range of high temperatures, and any subsequent shifts in the SQUID output would invalidate the calibration. Another problem is one of the proper choice of thermometer material. Even small concentrations of magnetic impurities in some metals cause departures from Curie's law at relatively high temperatures. A selection of material is discussed below.

Many other previous thermometers have relied on conventional NMR, both continuous wave and pulsed, in order to discriminate against signals from magnetic impurities.[1,2] These thermometers have been successfully used in nuclear cooling and other experiments. It is not clear, however, that resonance methods really avoid the magnetic impurity problem. Such impurities affect the linewidth of the resonance; consequently any temperature dependent behavior of the magnetic impurities will change the nuclear spin signal. This difficulty has been pointed out.[3] Resonance thermometers alone do not give a continuous readout of temperature or magnetization; and also they require by their nature some amount of heating in the thermometer material while the temperature is being measured.

The nuclear magnetic thermometer presented here employs a SQUID magnetometer in the conventional sense to measure static magnetization changes. But an additional provision allows the nuclear spins to be pulsed with a resonant r.f. field perpendicular to the static magnetization. The NMR pulse lowers the magnetization seen by the SQUID magnetometer which thus is able to detect the NMR and also to show the spin-lattice relaxation after the r.f. field is turned off. The temperature may then be obtained independently from Korringa's relation for $\tau_1$, the relaxation time: $\tau_1 T$ = constant. The thermometer system is still kept simple and is not only capable of giving continuous temperature readings from magnetization changes but may also be self-calibrated at any time and temperature by a resonance method.

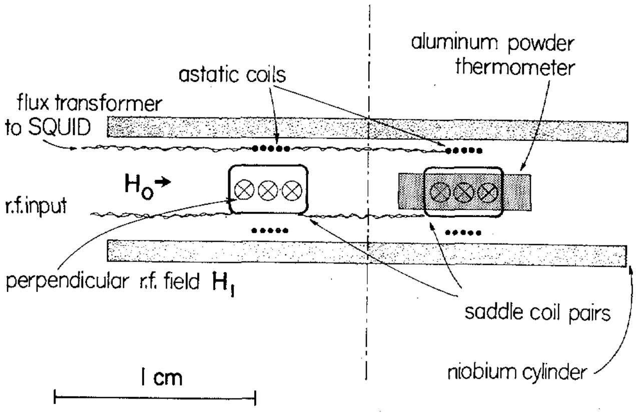

Fig. 1. Basic nuclear magnetic thermometer combining SQUID magnetometer and NMR detection.

## II. APPARATUS

Of initial importance is the choice of metal for the thermometer. It turns out that in certain metallic hosts, magnetic impurities form no local moments; i.e. there is no temperature dependent contribution by the impurities to the nuclear magnetization. This fact has been verified recently in experiments on dilute Al-Fe and Al-Mn alloys down to a temperature of 10mK.[4] The nuclear magnetic susceptibility of aluminum is suitably largest among various candidate metals which include Cu, Pt, and Sn. Aluminum nuclear spin-lattice relaxation times are also the longest of these metals, extending from 1.8 sec at 1K. For showing the feasibility of doing nuclear magnetic thermometry with SQUIDS aluminum was chosen. The advantageous use of other metals for certain applications will be mentioned later.

The thermometer consists of 0.02 cm$^3$ of 5N aluminum powder, 75-150 μ dia. This is placed in the cylindrical sample chamber of the magnetometer which is formed of Stycast 1266 epoxy.[5] In turn the magnetometer is located inside the mixing chamber of a $^3$He-$^4$He dilution refrigerator. The powder provides good thermal contact with the $^3$He-$^4$He bath and is also necessary to reduce eddy current heating from the r.f. NMR pulses.

For the magnetization measurements a constant longitudinal magnetic field $H_o$ is trapped in a superconducting Nb cylinder 0.508 cm i.d. and 2.5 cm long which surrounds the magnetometer. In these experiments the trapped field was 546 Oe. (The aluminum is thus kept normal.) Changes in flux due to the magnetization are sensed and coupled to the SQUID by a superconducting flux transformer. Figure 1 shows the basic arrangement. The flux transformer consists of two pick-up coils astatically wound which reduce most of the stray contributions to the signal. The SQUID magnetometer operates in a flux-locked mode giving a

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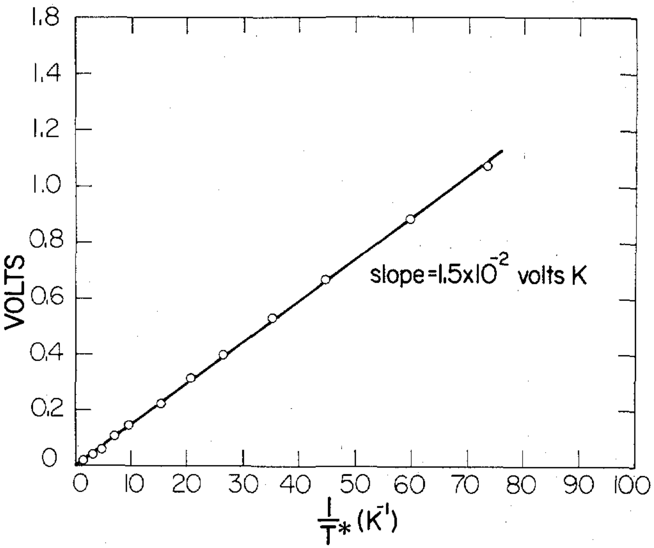

Fig. 2. Nuclear magnetization (expressed in voltage of the SQUID output) of aluminum powder.

continuous output voltage proportional to the nuclear magnetization.[6,7]

The same magnetometer detects changes in the longitudinal magnetization for the NMR part of the experiment when the nuclear spins are perturbed at resonance by an r.f. field $H_1$, perpendicular to the trapped field. To achieve this, small coil pairs of 4 turns each of 0.076 cm dia. Nb wire are wound around the sample holder in a rectangular (0.38 cm x 0.2 cm) saddle "Helmholtz" configuration. Symmetry is maintained by locating one pair of r.f. coils in each of the astatic detection coils, so helping to reduce coupling of the r.f. field into the SQUID. This coupling which has the tendency to unlock the SQUID is further attenuated with a resistive shunt of 0.2 ohms of maganin wire placed across the flux transformer leads. R.F. current for the NMR is fed from a function generator.[8] Pulse lengths of 606kHz r.f. used were 1.3 seconds with typical peak currents through the coil of 1 to 5 ma. These correspond to r.f. fields in the sample somewhat less than a milligauss. Eddy current heating, which is minimized by using short pulse times and low r.f. fields, was not observed with the power levels stated in the experiments on aluminum. Such a method of detecting NMR has interesting possibilities.[9]

## III. RESULTS

In order to demonstrate the nuclear magnetic thermometer, experiments were done to measure the aluminum magnetization and nuclear spin-lattice relaxation times, $\tau_1$, as a function of temperature from 2K to 11mK. Thermometry is performed with a second SQUID magnetometer detecting changes in the magnetization of a powder CMN cylinder (length equal to height). This too, is placed in the $^3$He-$^4$He bath of the mixing chamber.

The magnetization data, shown in Fig. 2 for a magnetic field of 546 Oe, have the expected linear 1/T behavior. Scatter in the data reflects the non-ideal compensation of background signals peculiar to the magnetometer used here. In order to subtract out the background, measurements were repeated with the aluminum sample moved to the other side of the magnetometer. By careful construction and selection of materials, background levels in the magnetometer can be greatly reduced.[10] The Curie law 1/T behavior of the magnetization confirms that for aluminum (5N purity) there are indeed none or few contributions from impurities. This behavior should hold over a very much lower range of temperatures, so long as $kT > \mu H$ holds.

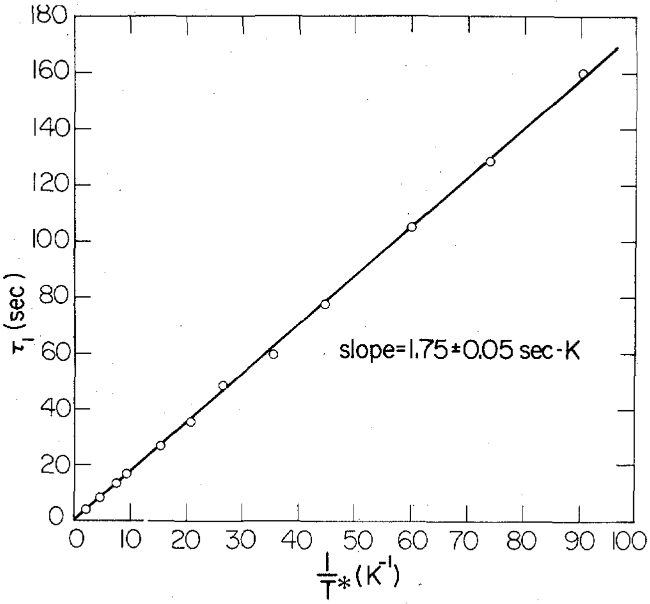

Fig. 3. Nuclear spin-lattice relaxation times $\tau_1$ of aluminum as a function of 1/T.

The nuclear spin-lattice relaxation is measured by observing the exponential return to equilibrium of the magnetization following the time when the r.f. field is turned off. Because aluminum relaxation times are relatively long, the magnetization change is displayed on a chart recorder. From the obtained exponential curves, $\tau_1$ is easily determined. Fig. 3 is a plot of the measured $\tau_1$ as a function of 1/T. The results show the linear Korringa's relation, with $\tau_1 T = 1.75 \pm 0.05$ sec-K in the high field limit. This value is in good agreement with published data[11] at higher temperatures which give a Korringa constant of 1.80 sec-K. The main errors are caused by changes in the baseline of the signal while the relaxation times are measured. This is especially a problem for aluminum since long relaxation times occur for the lowest temperatures and even small temperature and electronics drifts will affect the baseline stability.

From the verification of the 1/T dependence of $\tau_1$ and the nuclear magnetization, it is clear that the nuclear magnetic thermometer can be calibrated by measuring $\tau_1$, once Korringa's constant is known for a given material. Although in the very low temperature limit there will be departures from Korringa's relation, such limits are outside the scope of the present experiment.[12]

## IV. DISCUSSION

The combination of the high sensivity of the SQUID magnetometer with its ability to detect NMR of nuclear paramagnets provides an instrument that will find many applications in low temperature experiments. This thermometer can be further improved by using nuclear spin systems with a shorter spin-lattice relaxation time. These will give quicker response, especially at very low temperatures, and $\tau_1$ measurements will not be affected as much by stray drifts in the baseline of the magnetization. An excellent candidate is metallic tin whose Korringa's constant is 0.03 K sec. This system has nuclear magnetic properties similar to Pt (which is used in conventional NMR thermometers) but with the advantage that magnetic impurities also form no local moments. Tin would particularly be useful in the milli-Kelvin and lower temperature ranges. For such temperatures it would be advantageous to use finer powder to further reduce eddy current heating of the sample. Of course, since the major heat input to the sample would be during the NMR calibration, small as it may be, calibrations could be postponed until one is at higher temperatures.

It is interesting to note that the detection of NMR with a SQUID by looking at the longitudinal component is quite simple for broad lines.

The following advantages are realized in the nuclear magnetic thermometer which has been discussed here:

(1) a simplicity in measuring the static nuclear magnetization,
(2) a high sensitivity and temperature resolution,
(3) continuous read-out of the magnetization, i.e. of the temperature, with essentially no self-heating,
(4) self-calibration from nuclear spin-lattice relaxation time measurements, and
(5) no contribution from magnetic impurities since metals having no local moments are used.

The thermometer and results presented have been utilized here for only a $^3$He-$^4$He dilution refrigerator temperature range where perhaps these features are not always necessary; but extended into the milliKelvin and sub-milliKelvin region each capability may very well be needed for successful thermometry.

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