Simple cryostat for measuring rf susceptibility from 4.2 to 300 K

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We describe a cryostat for measuring the rf (100 kHz) susceptibility of insulators in the temperature range from 300 to 4.2 K. The sample is connected by a tube to room temperature and is moved in and out of the coil of a tunnel diode oscillator located at a constant low temperature. The change in frequency of the oscillator is measured with a digital frequency counter. The oscillator is stable to \( \pm 2 \) Hz over periods of several hours. The counter resolution and the attainable filling factor limit the resolution \( \chi/\Delta \chi \) to about \( 2 \times 10^1 \). The susceptibility of anhydrous NiCl\(_2\) powder was measured to evaluate the apparatus and determine the filling factor for a given shape sample.

INTRODUCTION

In recent years, the availability of inexpensive frequency meters has led to increasing use of oscillators to measure the magnetic susceptibility of insulating materials. The sample is placed in the tank coil of the oscillator, the frequency of which then depends on the susceptibility of the sample. If the frequency is high and the oscillator stable, a resolution of 1 part in \( 10^4 \) is possible.\(^1\) When the oscillator is located at room temperature but the susceptibility is to be measured at lower temperatures, one must take considerable care to insure the stability of the oscillator. Tunnel diode oscillators, however, have the great advantage that the whole oscillator circuit including all critical components can be built into a very compact space, thereby minimizing thermal gradients. A room temperature oscillator can be built with a short-time stability of 1 part in \( 10^7 \) even without regulating the ambient temperature. Tunnel diodes can be operated successfully down to 1 K, and if the oscillator is located at low temperature (e.g., surrounded by liquid nitrogen or liquid helium) then the temperature of all the components will be particularly stable and oscillator stability is almost automatic. Another active device that will operate at low temperatures is the field effect transistor (FET).\(^2\) Stable oscillators using FET's at low temperature have also been used.\(^3\) Several cryostats using tunnel diode oscillators to measure magnetic susceptibility have recently been described in the literature.\(^4\)

In the present apparatus, the susceptibility is measured by observing the frequency change when a sample is moved in and out of the tank coil of a tunnel diode oscillator. We believe the apparatus offers several advantages over those previously described.

DESCRIPTION OF THE APPARATUS

A diagram of the low temperature part of the cryostat (not to scale) is shown in Fig. 1. The tank coil of the oscillator (TC) is tightly wound around a 12 mm od glass tube (GT). Our coil consists of 90 turns of 0.2 mm copper wire close wound. The coil is covered with a layer of Stycast type 2850 GT\(^6\) epoxy (E). Below the coil, a PTFE clamp (CL) is attached to the glass tube and holds a small piece of circuit board on which are mounted the crucial frequency determining components of the oscillator. A small coaxial cable (CC) runs from the circuit to room temperature. This serves to carry the rf signal to room temperature and to bring down the bias voltage for the oscillator from room temperature. During a run, all of the above are immersed in a liquid helium bath at 4.2 K, or for the higher sample temperatures are cooled through exchange gas by an outer liquid air bath. Since the helium was not pumped below the \( \lambda \) point, it was constantly boiling and the epoxy layer on the coil is to prevent bubbles (with variable dielectric constant) from circulating around the coil and causing frequency jitter of the oscillator. If the circuit is rigidly

\[ \text{Fig. 1. Low temperature part of the rf susceptibility cryostat.} \]

- O—O-ring seal; SS—stainless steel support tube; GT—glass tube; SH—sample holder (Mircarta); CC—small coaxial cable to room temperature; S—sample; TC—oscillator tank coil; E—epoxy layer over tank coil; VL—vacuum line to diffusion pump; H—heater coil; T—carbon thermometers; CL—PTFE clamp for oscillator circuit board.
built, a stability of ±2.1% over several hours is usual, even though the sample has been moved in and out of the coil many times and its temperature has been varied from 200 K down to 4.2 K.

The sample (S) is attached to a thermally insulating sample holder (SIH) with cigarette paper and GE 7031 varnish (diluted with a 50–50 toluene–methanol mixture). Two carbon resistors for temperature measurement are located in holes in the sample holder near its upper and lower edges (T). A heater (H) is wound around the upper end (but was not used in our measurements). The sample holder in turn is attached to a long thin-walled stainless steel tube (SS) which runs to room temperature through a rubber O-ring seal. The inside of the tube is used for the thermocouple and heater leads. If the tube is well greased near the O-ring seal, it can be moved up and down the required amount without affecting a vacuum of 10^{-4} mm Hg inside the glass tube.

OPERATION OF THE APPARATUS

Inductance of the tank coil

If the inductance of the tank coil is \( L_0 \) when the coil is empty, then we want to know its inductance when containing the sample, \( L_s \). In general the self inductance of a coil is related to the total stored energy in it by

\[
U = \frac{1}{2} L_i I^2
\]

(1)

when a current \( I \) flows in it. This energy is also equal to the total field energy (in Gaussian units)

\[
U = -\frac{1}{8\pi} \int B \cdot H d^3r.
\]

(2)

Since we are only interested in the change in inductance, we

\[
\Delta U = -\frac{1}{8\pi} \int (B - H_0) d^3r,
\]

(3)

where \( H_0 \) and \( B_0 \) are the initial fields without any sample. Equation (3) can be written in terms of an integral only over the sample,

\[
\Delta U = -\frac{1}{2} \int M \cdot B_0 d^3r,
\]

(4)

where \( M \) is the magnetization in the sample. The change in inductance is thus given by

\[
(L_s - L_0) I^2 = \int M \cdot B_0 d^3r.
\]

(5)

If the sample is an ellipsoid of revolution and is small enough to be in the uniform part of the coil field, then the magnetization inside the sample is uniform and equal to

\[
M = \frac{4\pi x}{1 + \epsilon x} B_0,
\]

(6)

where \( \epsilon \) is the demagnetization factor, and \( x \) is the volume susceptibility. Hence we get

\[
\frac{L_s - L_0}{L_0} = \frac{4\pi x V_s}{1 + \epsilon x V_s},
\]

(7)

where \( V_s \) is the sample volume, \( V \), the volume of the coil and we have taken \( \frac{1}{2} L_0 I^2 = B_0^2 V_0 / 8\pi \) (i.e., the coil is assumed long compared to its diameter). Thus we see that by measuring the inductance change, one does not measure \( x \) directly but rather the factor \( x / (1 + \epsilon x) \). This factor is sometimes referred to as the "measured susceptibility." In our case, however, the sample was cylindrical and was not located in a uniform external field. For cylindrical samples, the demagnetizing field is not uniform even in the case where \( B_0 \) is uniform. This situation can be formally described by letting \( \epsilon \) become a function of position in the sample \( \epsilon(t) \). For a rod of length to diameter ratio of 3 (which corresponds to our samples), \( \epsilon \) at the center of the sample is 0.97 if \( B_0 \) is uniform. In our case where the ends of the sample lie in a region of lower field, there will be fewer induced poles and hence one would expect \( \epsilon \) to be smaller. Thus for nonferromagnetic samples where \( \chi \ll 1 \), we can safely ignore the term \( \chi x \) in the denominator of Eq. (7) compared to 1. Hence we do not need to know \( \chi \).

The integration in Eq. (5), however, still cannot be done easily, but it can be written as a combination of \( L_0 \) and a term which depends only on the geometrical shape of the sample and coil. The final result is

\[
\frac{L_s - L_0}{L_0} = 4\pi x F,
\]

(8)

where

\[
F = \frac{\int r^3 B_0^2(r) dr}{\int r^1 B_0^2(r) dr}
\]

is the geometrical filling factor for the sample, and must be determined in our case by measuring the susceptibility of a standard sample.

Oscillator

The tunnel diode circuit is a conventional parallel oscillator. The oscillation frequency is given by

\[
\omega_0 = [L_0 (C_0 + C_1)]^{-1} (1 - L_0 / r_2 C_1),
\]

(9)

where the components are identified in Fig. 2. \( r_d \) is the negative resistance of the tunnel diode. Since this negative resistance is temperature dependent, it is best to keep the term \( L_0 / r_2 C_1 \) small for good frequency stability. This means one must have \( r_d \) large and \( L_0 \) small. In our case where \( (L_0 / C_1)^{11} \) is about 50 \( \Omega \), we must have \( r_d \gg 50 \Omega \). Since the minimum negative resistance for our 1N3713 was found to be 67 \( \Omega \), the circuit had to be operated with low series resistance so that the diode would operate with a large average \( r_d \). With the large by-pass capacitor \( C_1 \) the main resistance in the circuit should be the diode resistance, which for a 1N3713 is about 4 \( \Omega \). Using this value and the condition for oscillation \( r_d = L_0 / r_2 C_1 \), one obtains \( L_0 / r_2 C_1 = 0.007 \) which satisfies the required condition. If the oscillations
are too nonlinear because of the large value of \(r_d\), then \(C_1\)
must be increased to allow the diode to oscillate over a more
linear portion of its characteristic (where \(r_d\) is smaller).
For the values of the components shown, the oscillations
were reasonably linear. If frequencies lower than 100 kHz
are desired, the above conditions would be more easily
satisfied using a back diode (which in general has a larger
minimum value of negative resistance).

If the frequency of the oscillator when empty [Eq. (9)]
is written as
\[
\omega_0 = \frac{1}{L_0C-a}\]
(10)
then when the sample is inserted, the frequency will decrease
to
\[
\omega_s = \frac{1}{L_sC-a}\]
(11)
where \(\Delta L = L_s - L_0 = 4\pi XF\) as shown above, and \(\Delta C = C_s - C\)
is the change in stray capacitance when inserting the sample.
We have ignored the change in the small term \(a\). If one
assumes \(\Delta L/\Delta C \ll 1\) and \(\Delta C/\Delta C \ll 1\), then subtracting Eqs. (11)
and (10) and simplifying, one obtains
\[
\frac{\Delta L}{\Delta C} = \frac{\omega_0 - \omega_s}{\omega_0 - \omega_s} = \frac{L_0}{L_s} \frac{1}{\omega_0}
\]
(12)
Using Eq. (8) this can be written in the more convenient
form
\[
\frac{\Delta \omega - \Delta \omega_0}{\omega_0} = 2\pi XF.
\]
(13)
This is the basic equation used in analyzing data. \(\Delta \omega/2\pi\) is
the measured change in frequency when the sample is
inserted into the coil. \(\Delta \omega/2\pi\) is a correction term due to the
dielectric constant of the sample or to the presence of the
stainless tube. Using a nonmagnetic sample of \(\text{Mg}_2\text{TeO}_4\),
the change in frequency was only about 2 Hz, thus \(\Delta \omega\) is a
small correction. Even though one expects the factor
\(1 - aL_sC\) to be close to unity, it can be included in the
filling factor \(F\), which has been done in Eq. (13).

Temperature Measurement and Control

Temperature is measured by two calibrated carbon
resistors. The resistors are calibrated against a Ge
thermometer below 90 K and a calibrated Cu–constantan
thermocouple above 90 K. This resulted in a temperature scale
believed to have an absolute accuracy of about 3%. The
insulating sample holder serves two purposes. First, it
keeps the stainless tube well away from the coil so as to
minimize \(\Delta \omega_0\), and second, it provides thermal insulation
against the heat leak coming down the stainless tube from
room temperature. Temperature control of the sample is
achieved by balancing the heat flow through the stainless
tube with the heat loss to the walls of the glass tube (which
is at 4.2 K) through the residual helium exchange gas. The
two thermometers enable one to determine the thermal
gradient across the sample holder, and therefore, to estimate
the magnitudes of possible thermal gradients in the sample.
This gradient was found to be about 2 K at 90 K and to
decrease to 0.1 K at 5 K. Thus any thermal gradients in
the sample are believed to be less than 1% of the sample
temperature.

With this method of temperature control, the range of
temperatures between 200 and 4.2 K is most easily
accessible. When helium is transferred and there is a good vacuum
\((P < 10^{-4} \text{ mm Hg})\) in the glass tube, the sample cools to
about 200 K. Further cooling is obtained by slowly admitting
helium exchange gas. Finer control of the temperature
could be obtained by connecting the heater and one of the
thermometers to a temperature controller.

When liquid air is transferred, the oscillator drifts as it
cools down to 80 K. If the vacuum around the sample is
good, then the oscillator drift will have slowed sufficiently
before the sample cools significantly. Measurements may be
taken in the range from room temperature to 200 K and
then a correction applied for the drift. This is most easily
done if the digital counter has an analog output.

\textbf{NiCl}_2 Susceptibility

As an example of measurement made with the apparatus,
Fig. 3 shows \(f/\Delta f\) for a powdered sample of anhydrous
\textbf{NiCl}_2. As seen from Eq. (13), \(f/\Delta f\) should be proportional
to \(1/\chi\) and for a sample showing Curie-Weiss behavior,
should be proportional to \((T-\theta)/C\) where \(\theta\) is the Curie-Weiss constant and \(C\) is the Curie constant (in kelvins). NiCl\(_2\) clearly shows Curie-Weiss behavior.

In measuring the susceptibility of a powder, one must consider its smaller density. This can be done by noting that for a homogeneous isotropic sample (ignoring small powder demagnetization effects), the ratio of powder susceptibility to crystalline susceptibility

\[
\frac{\chi_p}{\chi} = \frac{\rho_p}{\rho}
\]

where \(\rho_p\) is the powder density and \(\rho\) is the normal crystalline density. This introduces a second filling factor \(f\) into Eq. (13). For the NiCl\(_2\) sample, \(f\) was 0.84 so that using the known molar susceptibility of NiCl\(_2\), one obtains a geometrical filling factor \(F = 0.58\) (the sample powder was compressed into a cylinder 8.4 mm in diameter by 25 mm long).

For other samples with the same geometry, \(F\) can then be used to calculate approximate absolute values of the susceptibility. It should be noted, however, that \(\theta\) can be read off the graph without calibration. The minimum value, which for antiferromagnets corresponds approximately to the Néel temperature, can also be obtained directly without calibration. The error bars shown are not statistical errors, but rather represent twice the magnitude of the 5 Hz correction of the data for the empty frequency shift \(\Delta f_0\) in Eq. (13).

Because of \(\pm 1\) Hz ambiguity in using a 1 sec frequency meter, the resolution in susceptibility \(\chi/\Delta\chi\) is limited to \(5 \times 10^4\). This is reduced by another factor of about two by the attainable filling factor. The absolute calibration accuracy to be expected will depend on what substance is used as a standard and the reproducibility of the geometry of the sample. In our case we expect an absolute relative accuracy of about 30% for \(\chi\). This excludes any consideration of systematic errors such as relaxation time effects in the sample. The apparatus has been used to measure the susceptibility of \(\alpha\)-NaFeO\(_4\) and Mn\(_3\)TeO\(_4\) and the results are within 30% of the free ion \(\rho_{\text{ion}}\).

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