

Rice University
Physics 332

MEASUREMENT OF SUPERCONDUCTOR T_c

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Introduction

After liquefying helium in late 1910, Kamerlingh Onnes set out to extend measurements of the electrical resistivity of pure metals to temperatures of 1-2 K. On the basis of previous measurements to about 15 K with liquid hydrogen, he expected to see a roughly linear decrease from room temperature to a temperature-independent minimum resistivity determined by sample purity. Using a sample of very pure mercury, he found instead that the resistivity abruptly vanished at about 4.0 K. Below this critical temperature, T_c , the resistivity, ρ , was apparently zero, so the phenomenon was called "superconductivity". Later experiments with magnetically induced currents in superconducting rings demonstrated that $\rho < 10^{-21} \text{ } \Omega\text{-cm}$ below T_c , far less than the $10^{-6} \text{ } \Omega\text{-cm}$ typical of good conductors at room temperature.

The magnetic properties of superconductors are also unusual. Onnes soon found that a magnetic field, external or from current through the sample, destroys the perfect conductivity above a temperature-dependent critical value $H_c(T)$. Even stranger, Meissner and Ochsenfeld discovered, in 1933, that fields less than $H_c(T)$ are completely excluded from the interior of the superconductor. Exclusion occurs if the field is applied below T_c , or even if the sample is cooled through T_c in the presence of an external field. This is not expected of a perfect conductor, which would set up circulating currents to oppose any change in the external field.

These odd properties, and the potential for applications, has made superconductivity an active field of research for many years. Many materials, mostly metallic mixtures, were investigated to find higher T_c or H_c materials, but progress was slow even after a microscopic theory was developed in 1957. A metallic compound, Nb_3Ge with $T_c = 23 \text{ K}$, held the record until Bednorz and Mueller found a mixed lanthanum-barium-copper oxide with $T_c = 35 \text{ K}$ in 1986, quickly followed by yttrium-barium-copper oxide with $T_c = 92 \text{ K}$. The current record is held by even more complex oxides with T_c approaching 130 K.

All of these interesting oxide materials are mechanically brittle ceramics, so it is very difficult to fabricate a long solid sample and form reliable electrical contacts to measure resistivity. Because of the complicated compositions and extreme sensitivity to oxygen content, they also tend to be inhomogeneous, with superconducting and normal phases sometimes intermixed. If one is trying to measure bulk properties, resistivity can be misleading because all the current could be carried by a tiny filament of superconductor threading the sample.

In this exercise you will measure the T_c of a ceramic superconductor powder, using the Meissner effect (field exclusion) to detect the onset of bulk superconductivity. The specific technique, AC susceptibility, is explained below. Cryogenic temperature control and measurement is also discussed.

Read the Topical Notes on safety before starting the liquid nitrogen portion of this exercise.

Experimental Methods

To measure the properties of a superconductor, one must cool a sample through T_c and apply a magnetic field, electric current or other stimulus. The response of the material is then measured with appropriate sensors. To study the AC susceptibility of oxide materials, we need to set up cryogenic apparatus to control sample temperature in the range from about 75 K to 150 K, apply a small AC magnetic field, and detect the change in magnetic susceptibility around T_c .

Figure 1 is a block diagram of the apparatus used here. The AC constant-current supply provides an alternating field which is detected by the secondary coils surrounding the sample. Sample temperature is controlled by a flow of liquid nitrogen, warmed as necessary by a resistance heater. Thermometers are provided to measure the sample and heat exchanger temperatures. Operation of these subsystems is described below, along with procedures for automatic data logging.

A. AC susceptibility measurement

In the presence of an external magnetic field all materials develop magnetic polarization. That is, they acquire a net magnetic dipole moment per unit volume, denoted by the vector magnetization \mathbf{M} . For many materials, the magnetization is linearly proportional to the applied \mathbf{H} ,

$$\mathbf{M} = \chi \mathbf{H}$$

(1)

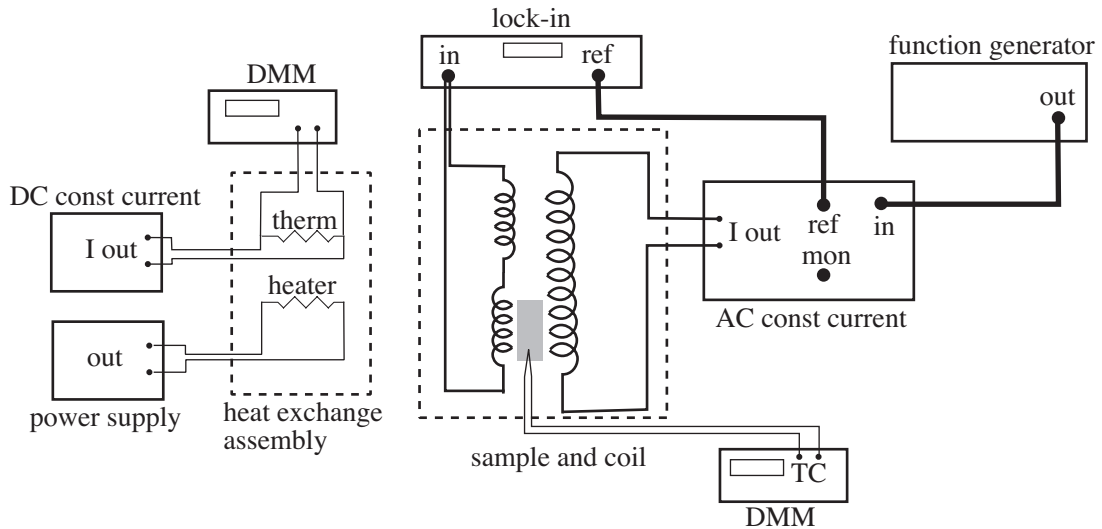


Fig. 1. Block diagram of measuring apparatus. The coils, lock-in and AC current source detect changes in sample susceptibility. A cryostat, heater and thermometer provide temperature control.

where the constant of proportionality is called the magnetic susceptibility. The magnetic field \mathbf{B} inside the sample is therefore

$$\mathbf{B} = \mu_0(\mathbf{H} + \mathbf{M}) = \mu_0(1 + \chi)\mathbf{H} \quad (2)$$

For a superconductor below T_c the magnetic field is totally excluded from the sample interior, so $\chi = -1$, which is several orders of magnitude larger than for a normal metal.

The susceptibility change is rather easy to detect with a pair of coils arranged as in Fig. 2. A changing current in the primary produces an axial \mathbf{H} field in both secondaries, inducing an AC voltage 90° out of phase with the driving current. The identical secondaries are wound in opposite directions and connected in series, so that the net induced voltage is very close to zero. When the sample becomes superconducting it reduces the magnetic flux in one of the coils, breaking the symmetry and producing a measurable signal.

A detailed analysis taking into account many other factors can be found in Ref. 1. For our purposes, the result can be summarized by

$$v = KVfH\chi \quad (3)$$

where v is the voltage measured out of phase with the driving current, V is the sample volume, and K is a geometric calibration factor. \mathbf{H} is proportional to the magnitude of the driving current at frequency f . Since χ is so small in the normal state, the signal we detect is effectively

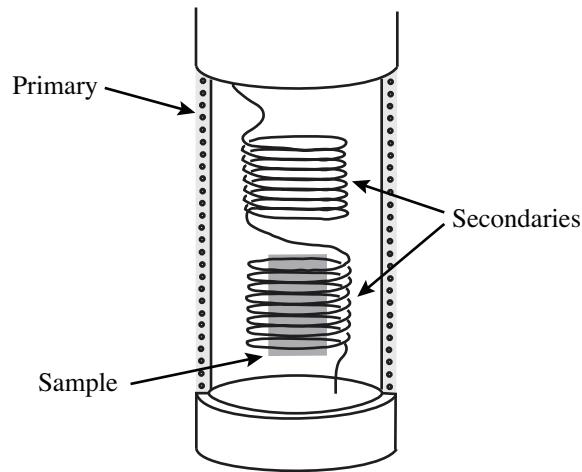


Fig. 2 Coil arrangement for AC susceptibility. (Adapted from Ref. 1) The coils used here are fully described in Appendix A.

proportional to the volume fraction of the sample which has become a superconductor. If the apparatus has been calibrated, this is a good way to test the homogeneity of a given sample.

To set up for a measurement, connect the function generator, current source, and lock-in reference as shown in Fig. 1. Input to the lock-in should be from the monitor terminal of the constant current source. The monitor output is the voltage across a $1\ \Omega$ resistor in series with the primary (see App. C for full circuit). Adjust the lock-in phase to maximize the signal from the monitor and set the generator amplitude to get the desired coil current according to the RMS voltage read by the lockin. Shift the phase by 90° and make small phase adjustments to zero the lockin, thereby detecting the voltage exactly 90° out of phase with the current.

If the secondary coils are now connected to the lock-in you will see a small signal due to residual imbalance between the secondary coils. This is effectively the baseline for changes due to superconductivity.

B. Temperature measurement

Sample temperature is measured with a Type T (Copper-Constantan) thermocouple embedded in the sample powder. Plug the thermocouple connector into the TC input on the DMM. Follow the procedure in the manufacturer's instructions to configure the meter for a Type T input and reading in Kelvin.

The temperature of the cryostat heat exchanger is read with a platinum resistance thermometer. Use the DC constant current source to apply 1 mA and read the voltage with a DMM. The sensor temperature can be found from the supplied resistance-temperature table.

C. Temperature control

The sample and coil assembly are housed in a liquid nitrogen cooled cryostat, diagrammed in Fig. 3. The sample, coil system and thermocouple are supported by a rod that brings the various electrical connections to the top of the cryostat. This fits into a closed tube that can be evacuated or pressurized with dry nitrogen through the sample space pump out valve. The bottom portion of the sample space is surrounded by a copper heat exchanger containing tubing for cold nitrogen flow, a heater, and thermometer. To cool the system, liquid nitrogen is allowed to flow through the heat exchanger coils and out through the flow control valve at the desired rate. Temperature is controlled by adjusting the flow valve and passing current through the heater resistor as needed. The entire system is insulated with a vacuum jacket and thermal radiation shields (not shown).

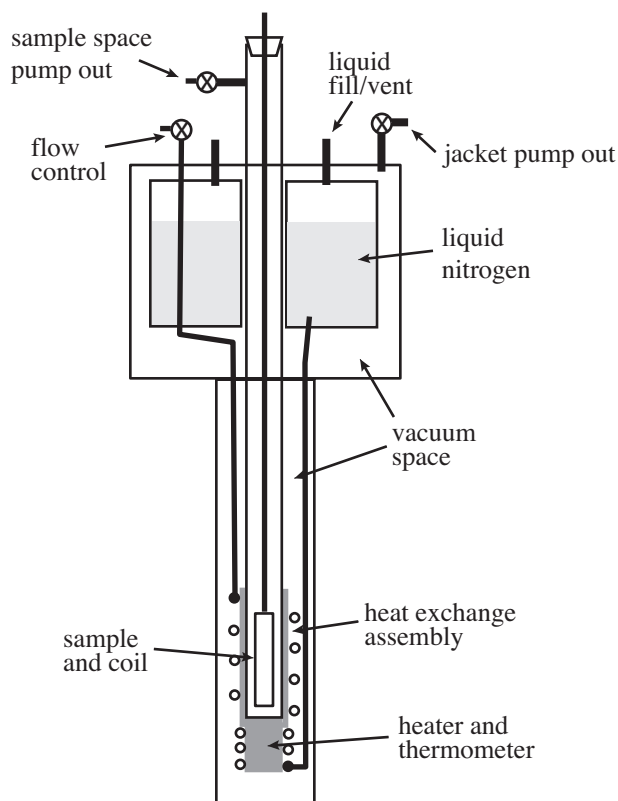


Fig. 3. Overall diagram of the nitrogen flow cryostat.

The cryogenic system must be cooled and operated properly to avoid substantial delays or even damage, so consult the instructor if any part of the procedure is not clear.

Preliminary setup

1. The vacuum space has been evacuated on a separate pumping station and closed off. Do not disturb the jacket pump out valve for any reason. Doing so will release the vacuum, necessitating several days of bake and pump out.
2. At room temperature, check that the sample holder assembly is in place and the vacuum feed-through fitting at the top is finger-tight.
3. Check that both thermometers read room temperature and that the coil signal is as expected. Connect the heat exchanger heater to the DC power supply, but do not apply power.

Nitrogen fill

1. Close the sample space vent valve and open the valve to the pump to evacuate the sample space. Both valves are located on the wooden support structure, below the mechanical pressure gauge.
2. Gently close the flow control valve. Fill the reservoir with liquid nitrogen by pouring it into the funnel on top, very slowly. The liquid will initially boil violently and cold gas will flow out the vent tube. Continue to pour slowly until liquid starts to come out of the vent tube. Once the reservoir is full you can set a stopper into the funnel to direct all the gas out the vent tube, away from the cryostat top. One fill will allow you to take data for 12-18 hours.
3. Fill the sample space with dry nitrogen gas to improve thermal contact between sample and heat exchanger. To do this, seat the vent tube into the top of the nitrogen storage container, close the sample space vacuum valve and open the sample space vent valve. Sample space pressure should remain near 0 psig because excess gas will be vented through the slit rubber tubing near the storage container.

Cool down and control

1. Open the flow control valve $1/3$ - $1/2$ turn to start coolant flow through the heat exchanger. The temperature of the heat exchanger should decrease within a few minutes, with the sample temperature lagging behind.
2. Decrease the flow setting as the heat exchanger approaches the desired temperature. Make further flow adjustments so that the sample temperature continues to fall slowly through the measuring range. Minimum sample temperature is typically 78-79 K, which can be reached in about 2 hours from room temperature.
3. To increase or stabilize the temperature, decrease the flow setting to a small fraction of a turn open. Turn on the heater power supply and increase to a few volts output. *Do not exceed 15 V* to avoid damage to the heater assembly. Monitor the heat exchanger and sample temperatures to obtain the desired rate of change of temperature. Remember that the sample will respond much more slowly than the heat exchanger, so base your control decisions on the heat exchanger temperature.

D. Data recording

The data consist of lock-in readings as a function of sample temperature. These quantities can be read from the instrument displays while setting up, but automatic recording is more convenient for extended runs. The Keithley 2110 DMM and Excel with KI-Link add-in make this fairly easy.

Set up the system by connecting the lock-in analog output to the voltage input of the DMM, and connect a USB cable from the back of the DMM to the PC. Start Excel from the desktop icon, and go to the Add-Ins tab on the toolbar. You should see several KI icons, some of which will be used as follows.

Connect to Device: Choose the desired device by clicking on the name and then click Connect. Use Search first if the list is empty.

Set up Instrument: Select TCOUPLE and choose the thermocouple type and temperature units as above. Check Enable 2ND function and choose DCV. This causes the DMM to read the thermocouple on the primary channel and the lock-in output on a secondary channel. Both will be visible on the DMM display. Click Submit to set the DMM for computer control.

Logging/Charts: Logging tab - Set for immediate start, a 10s sampling interval, and a couple of hours running. Chart tab - Choose strip chart (rolling display of specified number of samples) or graph (fixed display of all samples, probably more useful); Putting the chart on a separate Excel sheet is usually clearer.

Data taking will start immediately after you close the Logging/Charts window. You can pause, resume or stop using the control buttons in the tool bar. Switch to the Chart sheet to see the data plot.

The DMM will remain under computer control when you pause or stop acquisition. To return to manual operation, push SHIFT (LOCAL) on the front panel.

Measurement Program

The objective is to characterize the superconducting transition for a powder sample of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ made at Rice. An ideal, homogeneous, material becomes completely superconducting at a very well defined temperature T_c , but any variations in chemical composition or crystal structure will spread the transition over a finite temperature range. The apparent width of the transition is, therefore, a rough measure of sample homogeneity. Of course, internal temperature gradients and the applied magnetic field must be minimized to properly determine the sample properties, since they would also broaden the transition region.

Temperature gradients can arise for a number of reasons. Most important, the powder grains touch at only a few points, so heat transfer within the sample volume will be mostly through the surrounding gas, which has poor thermal conductivity. For the same reasons, thermal contact between the thermocouple and sample material will be poor. The net result is that the sample and thermocouple may take a long time to come to a uniform internal temperature after a change in external temperature. To minimize the effect, you must heat or cool the sample very slowly, particularly in the region of the transition where $\chi(T)$ is changing rapidly. Experimentally, the rate can be checked by measuring $\chi(T)$ for both increasing and decreasing temperatures. If the rate is slow enough the two curves will coincide.

Magnetic susceptibility measurements require an applied field, but the critical field H_c goes to zero as the temperature approaches T_c , so the applied field should be as small as possible. From Eq. 3, this makes the signal small, so there must be a compromise between signal strength and the resulting perturbation. One way to find an appropriate compromise is to determine signal strength as a function of AC field amplitude at a fixed temperature near but below T_c . For sufficiently large applied field the signal amplitude will fall below the linear dependence expected from Eq. 3 because the superconductivity has been destroyed in part of the sample volume. Operating as far below this level as feasible will give the best measurements in the transition region.

A. Initial exploration

The first step should be to locate T_c , at least approximately. To do this, set up the apparatus and check that the susceptibility and temperature systems seem to be working properly. The AC field should be set for a Gauss or two peak amplitude, using the coil factor of 81 G/A. Any frequency in the range 400-500 Hz should be satisfactory.

When all is in order, start the cool-down according to the procedure above. You can allow the system to cool fairly quickly until you approach T_c , expected to be 90-95 K. Start recording sample temperature and lock-in readings as you near T_c so that you can locate the transition region to a degree or two. Plotting these data will give you an estimate of both T_c and the signal strength compared to the normal state.

Next, you should get a better idea of the allowable magnetic field strength. Allow the temperature to settle to the minimum, since that is quite constant. Plot signal strength on the lock-in vs field amplitude over the accessible range, and look for deviations from linearity. Because of the properties of the AC current source, you will probably have to adjust the lock-in phase slightly for each amplitude. Choose the operating amplitude to give the largest signal possible while keeping the deviation from linearity small relative to the signal.

B. Transition measurements

Using the operating conditions you have established, record data as you raise the sample temperature slowly from the minimum available up through the transition region. A rate of 1 K per minute or less is typically acceptable, although you are free to experiment. Once the temperature is well above the transition, perhaps 110 K or so, you can cool slowly back through the transition.

Plot the heating and cooling curves together, and look for temperature offsets in the steepest part of the transition. If the offsets are significant, you are changing temperature too fast to produce an accurate measurement.

Once the temperature sweep rate is satisfactory you can check for any remaining sensitivity to the applied field. Reset the field amplitude for the lowest level that seems feasible, and again sweep the temperature through the transition in both directions.

Ideally, the lower-field curve would be a scaled-down version of the higher-field data, indicating that the magnetic field has no detectable effect. More commonly, the lower part of the transition will show some suppression of superconductivity by the larger magnetic field. The onset temperature may appear shifted as well, but that is usually less obvious. Unless the apparatus can be made more sensitive, these effects set a lower limit to the transition widths that can be measured.

Reference

1. M. Nikolo, "Superconductivity: A guide to alternating current susceptibility measurements and alternating current susceptometer design" Am. J. Phys. **63**, 57 (1995)

Appendices: Apparatus Details

The following are provided for your information. None of this material would be included in a formal research publication. (Ref. 1 does include many details because it is an instructional document, not a research report.)

A. Susceptibility measuring coils

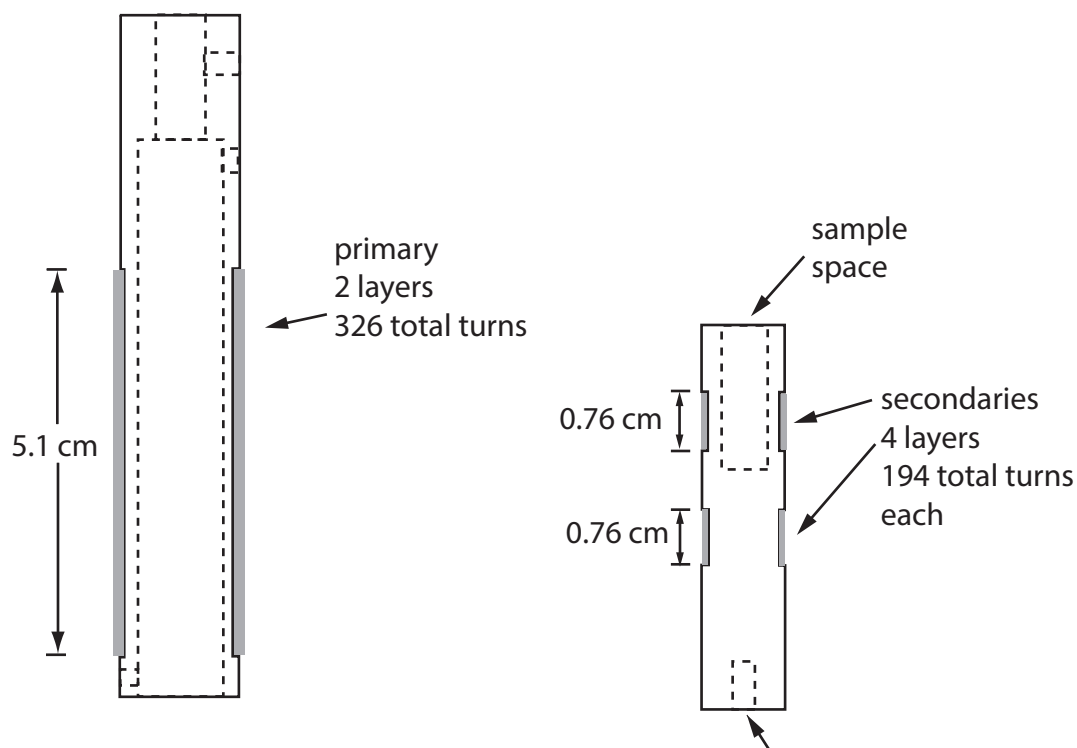


Fig. A1. The measuring coils are wound on non-conductive forms, shown to scale. The sample is loaded into the indicated volume, with the thermocouple wire embedded in the powder and secured with a cotton plug. The secondary coil holder is inserted coaxially into the primary coil so that the two secondaries are located symmetrically about the center of the primary. The whole assembly is supported by a rod extending to the top of the cryostat. The primary produces a field of 81 Gauss/Amp.

B. DC constant current circuit

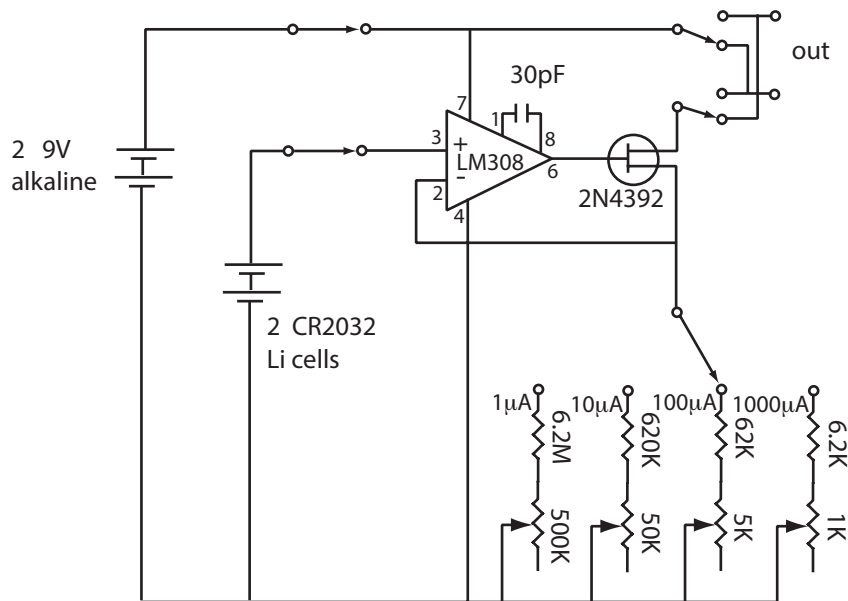


Fig. A2. The op-amp forces the voltage drop across the selected current sensing resistor to equal the reference voltage from the two Lithium cells. A 4-position switch selects a feedback resistor for the desired current, while a toggle reverses the current through the load. The 2N4392 JFET increases the current capability of the op-amp.

C. AC constant current circuit

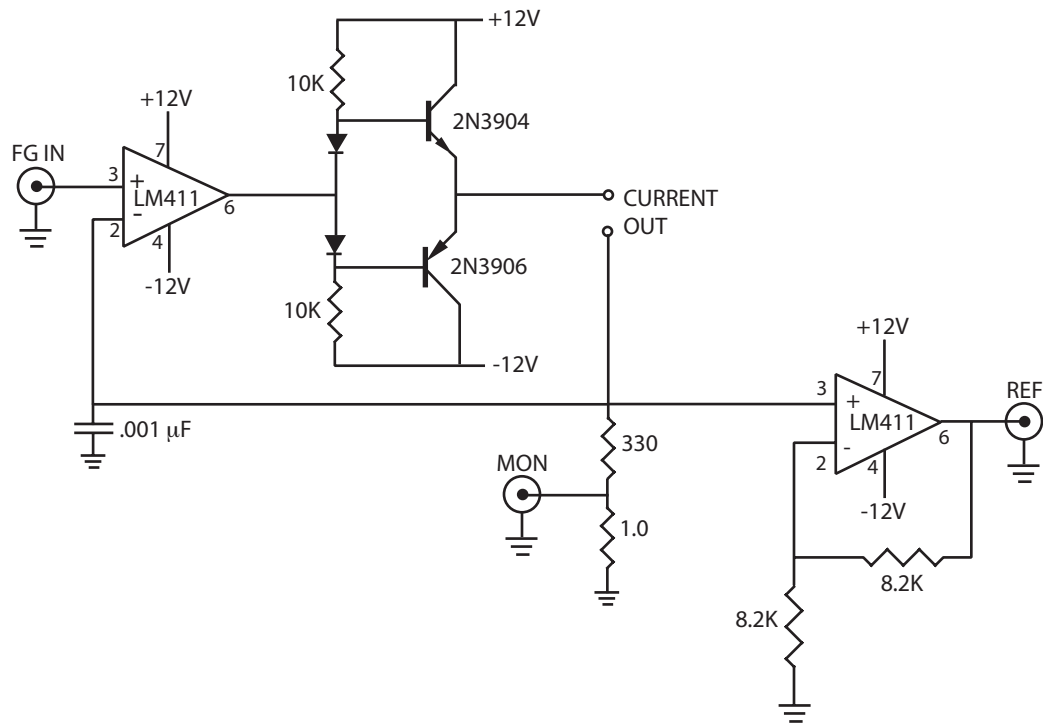


Fig. A3. This circuit is similar to the DC version. An external sine generator provides a reference voltage to compare to the voltage drop across the 331Ω sense resistors. A two-transistor push-pull increases the output capability. The second op-amp provides a slightly amplified signal, REF, nearly in phase with the current to drive a lock-in reference channel. The MON output is used to accurately fix the amplitude and phase of the primary coil current.