

## An oscillating coil system for contactless electrical conductivity measurements of aerodynamically levitated melts

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A new method for the contactless measurement of the electrical conductivity  $\sigma$  of high temperature aerodynamically levitated materials is presented. The apparatus, based on an oscillating coil system, improves considerably on previously reported static coil techniques by eliminating the large errors introduced by temperature drifts in the measurement system. With this system we demonstrate that accurate and fast measurements can be made for liquid metals and semiconductors with conductivities in the range  $10^4$ – $10^7 \Omega^{-1} \text{ m}^{-1}$ . Measurements of  $\sigma$  of liquid Ge as a function of temperature have been made to demonstrate the reliability of the method and give  $\sigma = 1.52 \pm 0.03 \times 10^6 \Omega^{-1} \text{ m}^{-1}$  at 1250 K in good agreement with previous measurements. The speed and precision of the apparatus will allow new studies of the electrical properties of high temperature liquids and glasses including precise observations of melting and glass transitions under quenching conditions. © 2006 American Institute of Physics. [DOI: [10.1063/1.2403939](https://doi.org/10.1063/1.2403939)]

### I. INTRODUCTION

The processing and study of liquids at high temperatures poses significant practical problems. The requirement to use crucibles means the liquid will react partly with even the most inert materials. For example, glasses and metals produced from the melt will often show some contamination from common crucible materials such as platinum or alumina.<sup>1</sup> In addition, measurements of, for example, the electrical conductivity of the material risk contamination from the electrodes used. There is considerable interest in the study of glasses produced from high temperature melts but their production is again affected often by the effects of impurities and nucleation centers in the crucible that can promote early crystal nucleation in the supercooled liquid state preceding the glass transition. In recent years various levitation techniques have been developed to study high temperature liquids, supercooled liquids, and glasses with the aim of avoiding such chemical contamination and nucleation. These methods include, electromagnetic,<sup>2</sup> electrostatic,<sup>3</sup> aero-acoustic,<sup>4</sup> and aerodynamic levitation.<sup>5,6</sup> With the development of these methods there has been an interest to develop contactless ways of measuring the electrical and magnetic properties of these levitated melts. In order to make these measurements some form of electromagnetic coupling (inductive or capacitive) needs to be made to the sample and this leads to significant difficulties when studying electromagnetic or electrostatically levitated liquids due to the need to decouple the levitation fields from the measurements. Despite these problems contactless measurements of metals have been made using induced torque measurements in an electrostatic levitator<sup>7</sup> and an electromagnetic levitation system.<sup>8</sup> However, these systems suffer some significant disadvantages, for example, the vacuum require-

ment of the electrostatic levitator means only very low vapor pressure metals were studied, whereas the electromagnetic levitator is only suitable for metals with a conductivity high enough to be supported in the electromagnetic field. Aero-acoustic levitation avoids these problems but the measurements are difficult due to the small sample size and distortions in the sample due to the acoustic field. In aerodynamic levitation these problems are largely absent and there has been some effort to establish contactless measurement techniques for high temperature liquids supported in this way. Although measurements that measure the changes in resistance ( $R$ ) and self inductance ( $L$ ) of a coil inductively coupled to the sample have been reported previously,<sup>9,10</sup> the methods used are highly susceptible to systematic errors due to the heating of the sensing coil. For example, in order to achieve good signal to noise in the measurements the coil needs to be as small as possible. However, a small coil is inevitably very close to the sample and is rapidly heated giving rise to a large change in its resistance (due to the temperature coefficient for resistance of the coil windings) and its inductance (due to the thermal expansion of the coil) that will swamp the required signal. The alternative of using a larger carefully cooled sensing coil decreases these heating problems at the expense of a severe decrease in signal to noise. In addition, the method relies on a careful pre-calibration of the empty coil (without sample) and the ability to place the sample onto the levitation nozzle and to levitate it without disturbing the coil and hence ruining the calibration. For the successful contactless measurements<sup>9,10</sup> reported for liquid B and Si, advantage was made of the fact that the cold sample was known to be insulating. In this case, by rapidly solidifying the melt, the empty coil calibration could be made immediately after the sample measurement by assuming the solid sample had negligible conductivity. However, this method cannot be extended to materials that remain conducting on cooling (metals) or for continuous conductiv-

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ity measurements. In this paper we describe a new method, extending these ideas, that enables a continuous empty coil calibration to be made by oscillating the sensing coil up and down on the levitated sample. This system substantially reduces the problems due to thermal drift in the apparatus and allows fast, reliable and accurate measurements to be made for a wide range of conducting liquids at temperatures approaching up to 3000 K.

## II. THEORY

The real  $\chi'$ , and imaginary  $\chi''$ , parts of the magnetic susceptibility of a non-ferromagnetic sphere in a homogeneous alternating magnetic field are given (in SI units) by<sup>11</sup>

$$\chi' = \frac{9}{4} \left[ \frac{\sinh 2\alpha - \sin 2\alpha}{\alpha(\cosh 2\alpha - \cos 2\alpha)} - \frac{2}{3} \right], \quad (1)$$

$$-\chi'' = \frac{9}{4} \left[ \frac{\sinh 2\alpha + \sin 2\alpha}{\alpha(\cosh 2\alpha - \cos 2\alpha)} - \frac{1}{\alpha^2} \right], \quad (2)$$

where the parameter  $\alpha$  is the ratio of sample radius  $a$ , and classical skin depth  $\delta$ , given by

$$\alpha = \frac{a}{\delta} = a \left( \frac{\sigma \omega \mu_0}{2} \right)^{1/2}, \quad (3)$$

where  $\sigma$  is the electrical conductivity,  $\omega$  is the angular frequency, and  $\mu_0$  is permeability of free space. These equations are in general valid provided the system satisfies the quasi-static approximation such that the wavelength of the radiation is much greater than the sample size and its period is large compared to the relaxation time of the current carriers in the material. For the sample sizes and frequencies used in this experiment these are satisfied.

The real and imaginary parts of the susceptibility of such a sphere can be found by measuring the change in the self-inductance  $\Delta L$  and resistance  $\Delta R$  when the sphere is moved in and out of a suitable coil (i.e., one that has a uniform field at the sample position). In this case  $\Delta L$  and  $\Delta R$  between the filled and empty coil may be written as<sup>12</sup>

$$\frac{\Delta L}{L_0} = \frac{L - L_0}{L_0} = -\eta \chi', \quad (4)$$

$$\frac{\Delta R}{R_0} = \frac{R - R_0}{R_0} = \frac{\omega L}{R} \eta \chi'', \quad (5)$$

where  $L$  and  $L_0$  are the self inductance of the filled and empty coil,  $R$  and  $R_0$  are the resistance of the filled and empty coil and  $\eta$  is the filling factor (ratio of sample volume to field volume). If the ratio  $\chi'/\chi'' = -\omega \Delta L / \Delta R$  is taken then it is possible to eliminate the contribution of the filling factor by fitting a curve over the measured frequency range to obtain a value for  $a\sqrt{\sigma}$ . Hence, by knowing the sample radius, the conductivity of the sphere can be determined.

The crucial requirement of any conductivity measurement using this technique is that the empty coil measurements  $L_0$  and  $R_0$  need to be accurately determined and to remain constant during the measurement of  $L$  and  $R$ . For typical coil sizes a stability of better than 0.001% is required. Until now the measurements of  $L_0$  and  $R_0$  have been made

before the sample is heated and every effort is made to ensure that they do not change for the duration of the whole experiment. In practice this is not possible to achieve for several reasons. First, extreme care must be taken to avoid any movement of the sensing coil and the leads running to it during the experiment. Even the slightest movements can lead to changes in  $L_0$  and  $R_0$ . Second, the sensing coil must be held at constant temperature otherwise unknown drifts in  $L_0$  and  $R_0$  as the sample is heated will occur. For example, for a simple solenoidal coil, a temperature change of typically 10 K or less is usually sufficient to swamp the required signal due to the temperature coefficient of resistance of the wire in the coil (changes  $R_0$ ) and the thermal expansion of the coil (changes  $L_0$ ). For these reasons it is in practice very difficult to make measurements in which  $R_0$  and  $L_0$  are measured first, followed by a complete set of measurements of  $R$  and  $L$  afterwards.

However, it can be seen from Eqs. (4) and (5) above that the crucial parameters to measure are the changes  $\Delta R$  and  $\Delta L$ . Hence, it does not matter if  $R_0$  or  $L_0$  drift with time provided that the  $(R, R_0)$  and  $(L, L_0)$  pairs are determined for each individual measurement point. There are two ways in which this may be achieved: continuously move the sample in and out of the coil or continuously move the coil up and down over the sample. In our aerodynamic levitation system the former is not possible due to the requirement to keep the sample stable in the levitation gas flow and to maintain its position for laser heating. However, the latter option is experimentally possible. In this case the requirement is that, at any frequency point, the empty coil measurements ( $R_0$  and  $L_0$ ) should be made every time immediately before the filled coil measurements. Hence, provided the time it takes to measure  $(R, R_0)$  and  $(L, L_0)$  is much faster than the thermal time constants of the measuring system, it is possible to eliminate the effects of any thermal drifts from the measurement.

## III. EXPERIMENT

A practical system for establishing suitable continuous  $\Delta R$  and  $\Delta L$  measurements is shown in Fig. 1. In this a carefully balanced, simple pivot system, is used such that the solenoidal sensing coil can be oscillated up and down over the levitated sample. The actuating solenoid for the pivot is sufficiently far away from the sample to avoid any errors due to any stray fields it may produce. The sensing coil itself is approximately 1 cm long and 1 cm in diameter and consists of approximately 20 turns of eight strands 0.25 mm diameter polyimide coated manganin wire. The continuous operating temperature of the polyimide coating is 240 °C and tests show that our coil operates within this range under levitation conditions. The coil was wound onto a 10 mm diam  $\text{Al}_2\text{O}_3$  tube and fixed using Autocrete (Flexbar Corp., New York) high temperature adhesive. The Autocrete also provides a further layer of thermal protection for the coil. The system was aligned so that the levitated sample was situated at the center of coil in the “down” position and  $\sim 3$  mm clear of the coil in the “up” position. The coil assembly was then fixed to a machined Alumino-silicate mount that is attached to the pivot. Due to the proximity of the levitated sample all the

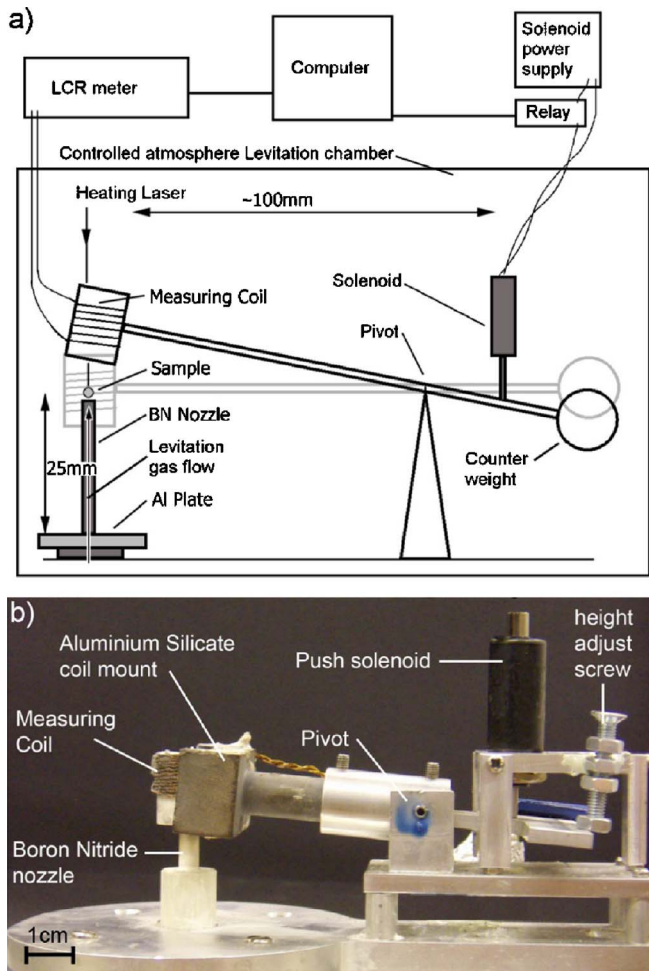


FIG. 1. (a) Schematic of the experimental setup, (b) photograph of the coil and pivot system.

components used in the immediate vicinity of the coil were chosen so that they were capable of sustaining temperatures in excess of 500 °C for long periods of time. This construction assured a high thermal mass for the coil assembly as well as a high filling factor due to the relatively small size of the coil. The coil can be oscillated up and down by computer control up to a maximum frequency of approximately 0.5 Hz. The  $L$  and  $R$  measurements are made using an HP 4285A precision LCR meter which can provide up to five significant figure precision in  $R$  and  $L$  over a frequency range of  $0.1 < f < 5$  MHz. In practice individual ( $L, L_0$ ) and ( $R, R_0$ ) measurements can be made in about 800 ms depending on the time constants set on the HP4285A (long integration times give more accurate results). Cable corrections for the HP4285A were made with the whole apparatus situated in the levitator in order to give accurate  $L$  and  $R$  measurements. In practice, due to large metal components in the levitator nozzle proximity, there is a small difference in the empty coil inductance and resistance in the up and down positions. These errors were minimized by using a tall levitator nozzle and determining any residual offset error before the measurements took place.

Figure 2 shows examples of  $R$  and  $L$  obtained over a period of approximately 50 s from a levitated molten Germanium sample while the coil was being moved up and

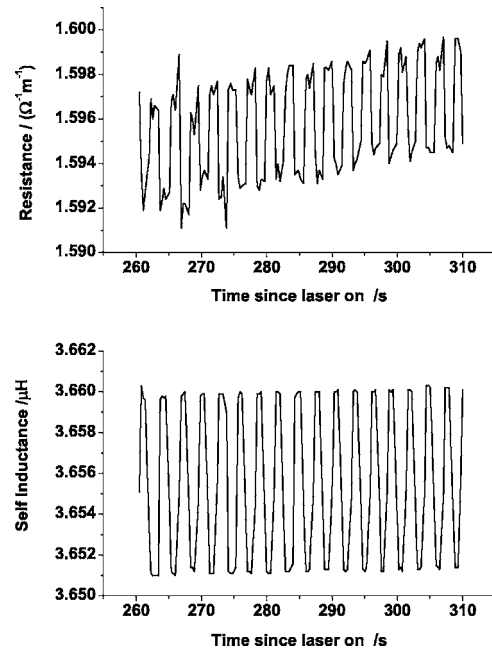


FIG. 2. The resistance ( $R$ ) and self-inductance ( $L$ ) of the coil as it is oscillated up and down on a molten germanium sample.

down. All the measurements are under computer control to allow complete synchronization of the coil position with the measured  $R$  and  $L$  values. In the figure the coil system had been allowed to reach approximate thermal equilibrium with the sample over a period of approximately 2 min. Small drifts in the baseline values of  $R$  and  $L$  can still be observed. However, the crucial parameters, notably  $\Delta R$  and  $\Delta L$  can clearly be observed as the amplitude of the oscillations. In order to complete a conductivity measurement a full set of measurements of  $\Delta R$  and  $\Delta L$  was made over the range  $0.1 < f < 5$  MHz in 100 kHz steps. In practice,  $\Delta R$  and  $\Delta L$  were measured at each frequency point by finding the differences in  $R$  and  $L$  between a measurement made in the “coil” up followed immediately by a measurement in the coil down position. The frequency was then changed and the procedure repeated. In this way, given the thermal mass of the coil system, errors due to temperature drifts were avoided. Hence a single frequency point could be taken in about 1 s and an accurate conductivity measurement in about one to two minutes.

#### IV. RESULTS AND DISCUSSION

Figure 3(a) shows the measurements of  $\chi'$  and  $\chi''$  as determined from  $\Delta R/R$  and  $\Delta L/L$  a function of frequency for liquid and solid Germanium (99.95% purity) along with the fits to Eqs. (1) and (2). The goodness of fit to the theoretical curves and the correct low frequency behavior (for nonmagnetic samples  $\chi'$  and  $\chi''$  should both tend to zero as  $f$  tends to zero) suggest that we have successfully eliminated errors in the measurements due to background thermal drifts.

Figure 3(b) shows the conductivity results obtained for liquid germanium over the temperature range  $1100 \text{ K} < T < 1500 \text{ K}$  [as determined from the corrected pyrometer reading ( $\epsilon=0.55$ )]. In the liquid state (above 1150 K) we observe the conductivity and its temperature dependence in agree-

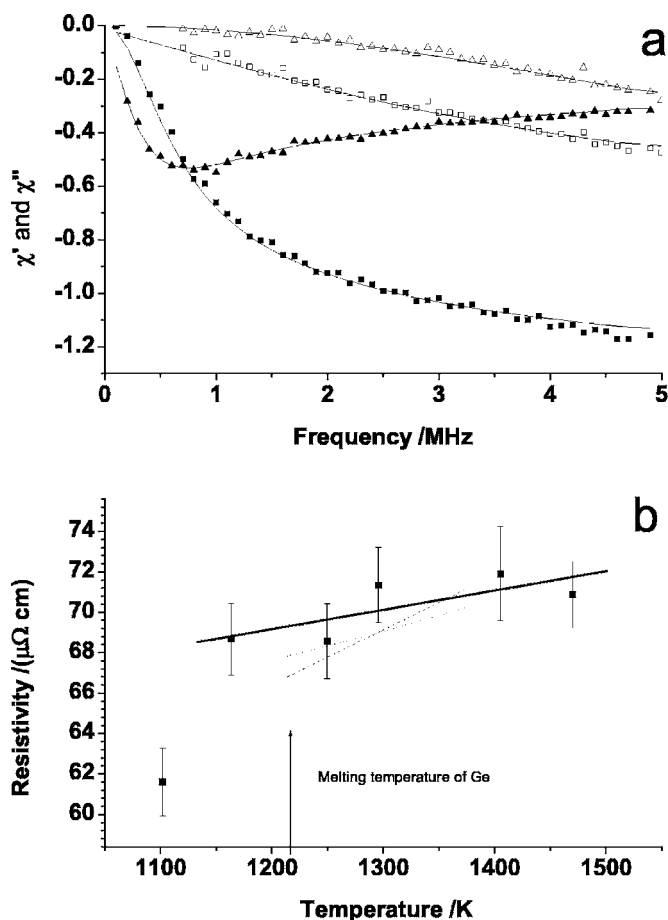


FIG. 3. (a) The susceptibility as a function of frequency of molten germanium at 1250 K (filled symbols) and solid germanium just after cooling 1100 K (open symbols) along with the fit to Eqs. (1) and (2). The triangles represent  $\chi'$  and the squares represent  $\chi''$ . (b) The resistivity of molten and solid germanium as a function of temperature as measured with this equipment. The line shows the least-squares fit to the data for the molten samples. The melting temperature of Ge is indicated. The dashed and dotted lines represent the fits to the data for liquid Germanium as reported by Schnyders and Van Zytveld Ref. 13 and Koubaa and Gasser (Ref. 14) over their measurement range.

ment with those reported previously.<sup>13,14</sup> The relatively high conductivity of the solid Ge sample is consistent with the purity of our sample.

From these measurements we conclude that this new apparatus is capable of measuring reliably the conductivity of small levitated spheres with an accuracy of approximately 2% for samples with  $10^4 < \sigma < 10^7 \Omega^{-1} \text{ m}^{-1}$ . Also, by monitoring  $\chi'$  or  $\chi''$  at a constant frequency (i.e., one chosen for optimum sensitivity) this apparatus is also suitable for measuring changes in the sample state in both equilibrium and nonequilibrium states (i.e., supercooled liquids and glasses) and as such is a useful system for measuring transient phenomena, for example, solidification or glass transitions in high temperature liquids.

The ultimate accuracy of the measurements themselves is dependent on any changes that take place during the levitation process.

For example, a sample with a moderate vapor pressure will lose mass and hence change in size as material is carried away in the levitation gas stream. This can lead to some long term drift in the measurements although the sample size can be continuously monitored in order to correct for this. Similarly it is possible that the levitation gas may react with the sample under study, for example, even for the highest purity argon available there is still likely to be some small surface oxidation of the sample that may lead to systematic errors in the conductivity measurements. This problem can be offset by, for example, introducing small quantities of reducing gas (e.g.,  $\text{H}_2$ ) into the levitation gas stream. Indeed, the system as described is very suitable, by modifying the gas stream, to investigate the *in situ* changes in the sample properties as the environment is changed.

## V. DISCUSSION

We have developed a new system for making accurate electrical conductivity measurements of laser heated high temperature liquids supported in an aerodynamic levitator. The results demonstrate that measurements, accurate to better than 2%, can be made rapidly and reliably. The system will open up new possibilities for the contactless study of liquid metals and semiconductors in the liquid, supercooled and glassy states.

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