The Measurement of RRR and Resistance for Aluminum Alloy Based on a Cryocooler

C.H. Yin, Z.H. Gan, S.H. Wang, D.L. Liu, L.M.Qiu

Institute of Refrigeration and Cryogenics, Zhejiang University
Hangzhou 310027, P.R.China
Key Laboratory of Refrigeration and Cryogenic Technology of Zhejiang Province
Hangzhou 310027, P.R.China

ABSTRACT

RRR (Residual Resistivity Ratio) is a common index for measuring the quality of a metal. This paper reviewed the three main methods of measuring alloy RRR, and compared the respective advantages and disadvantages. A new cryocooler based measurement method is proposed. Theoretical analysis shows this method can measure the resistance at various temperatures. There is no need to reach the liquid helium temperature (4.20K), as achieving temperatures below 38 K is sufficient to measure RRR. The analysis is later verified by experimental results, and the results differ little from PPMS (Physical Property Measurement System) measured results.

INTRODUCTION

Residual Resistivity Ratio or Residual Resistance Ratio (RRR) is defined as the ratio of a metal's electrical resistance at room temperature to that at 0 K, which is usually replaced by liquid helium temperature (4.20K). It is the one of the common indexes for characterizing the quality of a metal, including purity and structural characteristics. RRR can be used to evaluate operational stability of commercial devices made of superconductive materials.[1] Moreover, the RRR of a metal strongly controls its cryogenic conductivity and emissivity, including the conductivity of a superconductive material cavity and the performance of a superconducting resonator cavity.[2] Thus, efficient and accurate measurements of RRR are particularly useful in the cryogenics field.

There are three mainstream conventional methods for RRR measurement: the eddy current decay method, the curve method, and the fixed-point method. Developed in the 1960s, the eddy current decay method is a mature technology with established international standards.[3] This method is based on the relationship between the magnitude and decay of eddy currents and the resistance of a metal sample with a certain size and shape. The eddy current is introduced by a changing magnetic field, and the resistance at liquid helium temperature is then determined according to the time constant of the eddy current decay. The sample is placed in a liquid helium dewar. For instance, when the metal sample is of a cylindrical form, its resistance can be calculated via Eq. (1):

$$\rho = 2.17 r^2 \times 10^{-11} / \tau \tag{1}$$

where, r is the radius of sample in m; τ is the time constant of eddy current decay time in s; and ρ is the resistivity. It is applicable for pure metal samples with a diameter between 5 mm and 20 mm, a length-to-diameter ratio larger than eight, and a resistance ranging from $10^{-12}\,\Omega\cdot$m to $10^{-8}\,\Omega\cdot$m; otherwise a large measurement error may occur.

Cryocoolers 19, edited by S.D. Miller and R.G. Ross, Jr.

Table 1. Comparisons of conventional RRR measurement methods

Method	Measuring principles	Advantages	Disadvantages
Eddy current method	Induced eddy currents are generated when the metal sample is in a changing magnetic field; eddy current decay time constant is measured to determine the resistivity.[4]	Non-destructive to the samples; No welding of electric potential or current wire lead to samples; Impurity distribution can be determined.	Strict requirements of shape and size of samples; Only discontinuous points can be measured; No measurement of resistance varying with time.
Fixed-point method	Constant temperature points achieved by helium dewar and ice-water bath are used to measure a sample's resistance at certain temperature are used to calculate RRR.[1]	Relatively short measuring time; low cost; Suitable for batch measurement.[4]	Only discontinuous points can be obtained, not resistance versus temperature curve.
Curve method	Helium dewar is used for cooling and achieve low temperature condition while heating units are used to control temperature.[1]	Can obtain sample resistance curve changing with temperature.	Complex system setup; Long measuring time and high cost.

The Fixed-point method calculates the RRR based on the measurement of material resistance at fixed temperature points. Room temperature (293.0 K) or ice point (273.15 K) and helium temperature (4.20 K) are typical pairs of fixed points. Chinese and some international manufacturers mainly employ the fixed-point method to measure the residual resistance at low temperature. One characteristic of the fixed-point method is that the residual resistance measurement is conducted at a thermal equilibrium state.[1]

The Curve method employs measurements of a series of temperature points before numerically fitting the curve. The RRR is calculated using the resistance read from the curve.[1] Typically, a liquid helium dewar is used to provide the required temperature (4.20 K).

The advantages and disadvantages of the methods are summarized in Table 1. As can be seen, all three methods require a helium dewar, and thus there exist inherent limitations: the resistance can only be measured at limited temperature points, and the cost is high. Therefore, this paper proposes a new cryocooler-based method for RRR measurement. The temperature of a cryocooler can be continuously adjusted simply while reaching 4.20 K as well. The proposed method is capable of accurate RRR measurement. The method is also applicable to studying the variations of resistance with temperature. Moreover, the accuracy of the measurement depends less on the sizes and shapes of samples, it is more versatile for industrial applications.

BASIC PRINCIPLES OF MEASUREMENT

Principle Analysis

The resistance of metals can be described by the Mathieson rule:

$$\rho = \rho_l + \rho_r \tag{2}$$

where: ρ_l originates from the thermal motion of lattice points and is also called phonon or ideal resistivity; it is related to temperature. The residual resistance ρ_r, which originates from impurities and defects in the metal lattice, is independent of temperature.[5-6] There is no phonon excitation at absolute zero, which means ρ_l=0. Increasing temperature causes faster movement of the lattice points, thus interaction between electrons and phonon will increase ρ_l with increasing temperature.

The relationship between ρ_l and temperature can be expressed by the Bloch-Greisen equation:

$$\rho_l = \frac{AT^5}{M\Theta^6}\int_0^{\Theta/T}\frac{z^5}{(e^z-1)(1-e^{-z})}dz = \frac{AT^5}{M\Theta^6}J_5\left(\frac{\Theta}{T}\right) \tag{3}$$

where

A is constant,

M is the molar mass of the substance,

Θ the characteristic temperature of the metal element and close to the Debye temperature Θ_D.[7]

If the temperature is relatively high or low, the equation can be simplied as follows when $T > \Theta / 2$:

$$\rho_l \approx \frac{AT}{4M\Theta^2} \propto T \tag{4}$$

ρ_l is linear function of temperature.

When $T>\Theta/10$, the integral upper limit of Equation (3) can be set as infinity, thus:

$$\rho_l \approx \frac{124.4AT^5}{M\Theta^6} \propto T^5 \tag{5}$$

The residual resistance ρ_r depends only on impurities and the types and quantities of defects.

The total resistance can be expressed as $\rho = \rho_l + \rho_r$. At high temperature, $\rho_l >> \rho_r$, so $\rho \approx \rho_l$; at very low temperature, for instance T=4.2 K, $\rho_l << \rho_r$,, so $\rho \approx \rho_r$ which means the total resistance is mostly independent of phonon resistance. An alloy can be regarded as a highly impure metal.[8]

RRR is expressed as the ratio of the resistance of the material at ice water bath temperature (273.15K) to that at a low temperature (4.20K):

$$RRR = \frac{\rho_{T=273.15K}}{\rho_{T=4.20K}} \tag{6}$$

The relation between resistance and resistivity can be expressed as:

$$R = \frac{\rho L}{A} \tag{7}$$

where

R is the resistance in Ω;

L is the length in m;

A is the cross-sectional area in m^2.

Since L and A can be considered as constant through the temperature range, the problem of measuring resistivity can be simplified as resistance measurement, which can be expressed as:

$$RRR = \frac{R_{T=273.15K}}{R_{T=4.20K}} \tag{8}$$

The basic principle of resistance measurement follows Ohm's law:

$$R = \frac{U}{I} \tag{9}$$

where

U is the voltage in volts;

I is the current in amps.

The terminal voltage of the sample can be measured when constant current is applied at both ends; therefore the RRR can also be expressed as the ratio of the voltages measured at the two different temperatures:

$$RRR = \frac{U_{T=273.15K}}{U_{T=4.20K}} \tag{10}$$

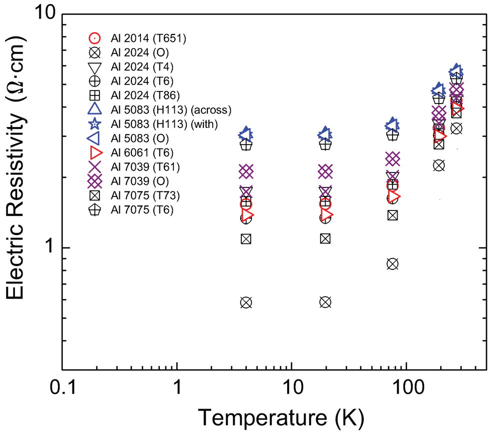


Figure 1. Resistance changes at varied temperature for different types of aluminum alloy.

Aluminum Alloy Resistance Analysis

The Debye temperature of aluminum is 380 K, which means that its resistance is nearly constant under 38 K. The resistance of aluminum alloys at low temperature is over 1000 times greater than that of pure aluminum.[9-10] For different types of aluminum alloys. the resistivity changes as a function of temperature is shown in Figure 1, where resistivity varies between 0.5 and 6 μΩ·cm. Figure 1 shows that aluminum alloy resistivity changes little under 77 K. Especially when the temperature is under 20 K, the resistance remains nearly constant when the temperature decreases. If the sample size (width × thickness × length) is 5.0 ×0.5 ×90 mm, its resistance at low temperature will be (0.18-2.16) $\times 10^{-3}\Omega$. With 100 mA excitation current applied, the terminal voltage will be 18-216 μV.

Measuring Strategy

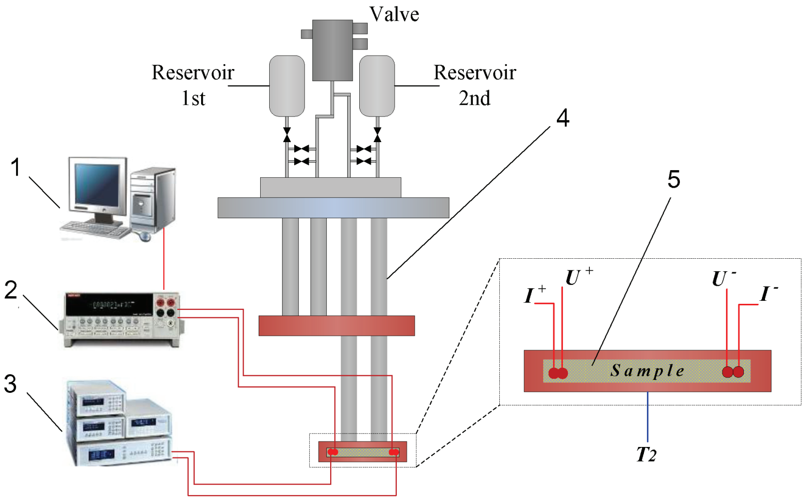
According to the range of signal induced, a six-and-a-half-digit high precision digital multimeter (2700 type) was used in this experiment with a measuring range of 100 mV and measuring precision of 3 μV. A Lake Shore 120 Current Source was used for 100 mA current excitation, which provides constant current within 11 V with precision of ±0.1%.

A Cernox thermometer from Lake Shore Inc. was used, which is applicable from 1.4 to 325 K. The measuring precision is 0.014 K at 4.2 K, 0.009 K at 20 K and 0.046 K at 77 K, which satisfies the requirements for low temperature measurements.

CRYOCOOLER MEASUREMENT PLATFORM

As shown in Figure 2, the cryocooler measurement platform consists of the cryocooler, the data acquisition system, and the sample setup. A self-developed two-stage pulse tube cryocooler was used to achieve the low temperature environment.[11] Its lowest cooling temperature is 2.2 K, thus meeting the measurement requirement. In the experiment, the constant current source for exciting the aluminum alloy sample was attached to the cold head at the second stage of the cryocooler and to measure the voltage at both ends of the sample. By adjusting the temperature of the second stage cold head through the rotary valve and minor orifice valve, the respective voltage of both ends can be measured when the temperature varies.

Four wires were used for the measurements. Two copper wires were welded to both ends of the sample respectively. As shown in Figure 3, to the outer end was welded the current leads (I+,I-), and the inner end was attached to the voltage leads (U+,U-). The samples were wrapped in electrical tape in order to insulate them electrically. Outside the lead end tape was wrapped another highly adhesive aluminum tape to avoid the influence of cryocooler vibration. Samples were placed on the side wall of the copper block at the second stage cold head. To improve the sample attachment to the



1:data acquisition 2:Keithley2700 3:Lakeshore120 4:cryocooler 5:sample

Figure 2. Diagram of cryocooler measurement platform

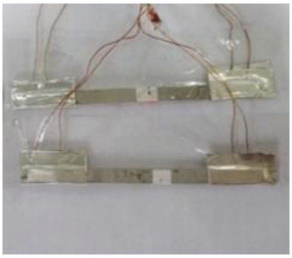


Figure 3. 90 mm samples and four-wire connection method

Figure 4 Spring fastening method on outside of test samples

second stage cold head, a spring fastening method was adopted in addition to the traditional aluminum tape, as shown in Figure 4. This method ensured that the sample was closely attached to the cryocooler cold head at all times during the measuring process, preventing the sample from falling off from unstable adhesion or cryocooler vibration, i.e. it ensured that the temperature of the sample was equal to that of the cryocooler cold head.

In order to eliminate contact potential and effects caused by zero drift, separate measurements were made with currents flowing in opposite directions to obtain two voltages, one positive and one negative. The average voltage value was obtained by subtracting these two values and dividing the result by 2.

$$\bar{U} = \frac{1}{2}(|U^+| + |U^-|) \tag{11}$$

RESULTS AND DISCUSSION

The experimental results are shown in Figure 5, which shows resistance versus temperature curves for samples A and B. Below 38 K, the curves remain almost horizontal and the resistance changes little with temperature. Above 90 K, the aluminum alloy resistance is mostly linearly proportional to temperature. Furthermore, the ratio between resistance at 273.15 K and other temperatures can be obtained as shown in Figure 6, where the resistance ratio is almost constant below 38 K. To further analyze the accuracy of the test results from the cryocooler measurement platform, the

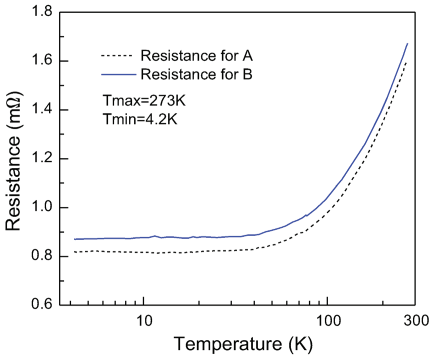


Figure 5. Resistance vs. temperature of the cryocooler measurement platform

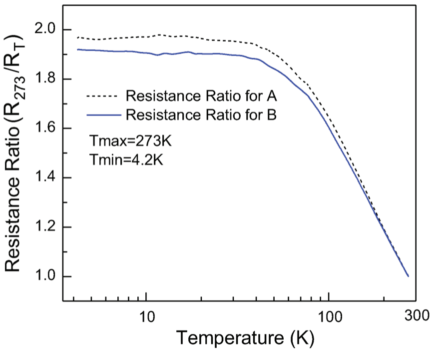


Figure 6. Resistance ratio vs. temperature of the cryocooler measurement platform

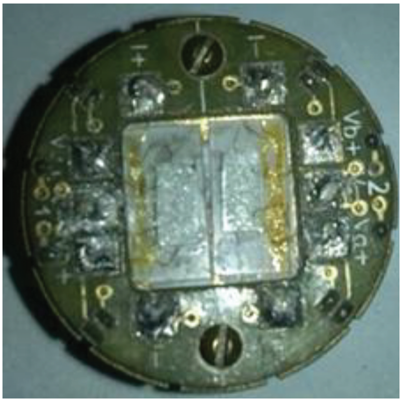


Figure 7. Schematic diagram for sample A and B on the sample holder of PPMS.

electric transport measurement option of PPMS (Physical Property Measurement System) was used to measure the sample resistance. Both samples were 5.0 ×0.5 ×2.5 mm in size and were placed as shown in Figure 7. Gold wires with high ductility were used for four-wire connection method and they were adhered by elargol.

Measurement results using PPMS for the two aluminum alloy samples are shown in Figure 8. The ratio between the resistance at 273.15 K and at other temperatures is shown in Figure 9, where the resistance ratio is also shown to change little below 38 K.

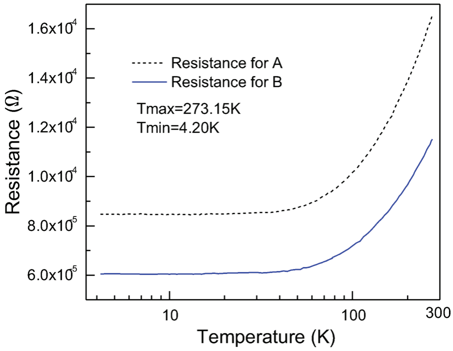


Figure 8. Resistance changes at varied temperature on PPMS

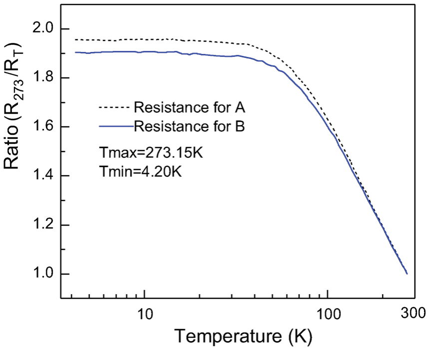


Figure 9. Resistance ratio changes at varied temperature on PPMS

Table 2. Resistance ratio of samples for 273.15K/4.20K (/20K/38K).

Sample	Sample A (4.20K)	Sample A (20K)	Sample A (38K)	Sample B (4.20K)	Sample B (20K)	Sample B (38K)
PPMS	1.957	1.953	1.944	1.905	1.896	1.891
Cryocooler	1.966	1.960	1.952	1.918	1.903	1.892

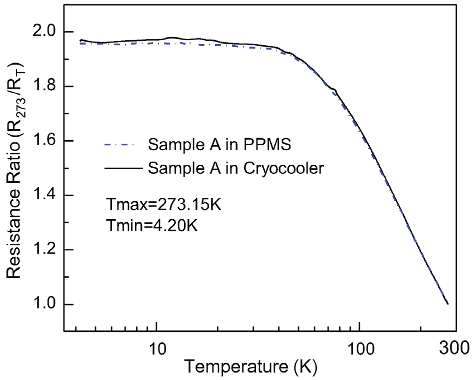


Figure 10. Comparison of Resistance Ratio vs Temperature curve, sample A.

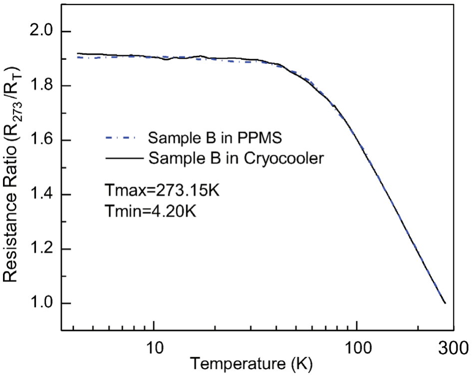


Figure 11. Comparison of Resistance Ratio vs. Temperature curve, sample B

Comparing the results from the cryocooler measurement platform and the PPMS system, the resistance value at 273.15K is divided by the resistance at 4.20K, 20K and 38 K to obtain the resistance ratio, as shown in Table 2. The deviation for samples A and B between measurement results from the cryocooler and PPMS systems is 0.46% and 0.68% at 20 K, respectively. This proves the measurement results of the cryocooler platform to be accurate and reliable.

Note also that the difference in the resistance ratio obtained from cryocooler measurements made at 4.20 K and at 38 K is only 0.66% and 0.73%, respectively, for samples A and B.

For both samples, Figures 10 and 11 compare the resistance ratio versus temperature curves from the two methods. The curves of the PPMS platform agree well with those of the cryocooler platform.

CONCLUSION

Several methods for measuring the RRR of alloy materials have been compared. The respective advantages and limitations are analyzed. The cryocooler measurement method is proposed and verified experimentally. The deviation of this new method is within 1%. On the other hand, the theoretical analysis and experiment results have shown that the resistance remains nearly the same between 38 K and 4.20 K for the alloy examined. If 4.2 K cannot be reached due to limits of the test setup, the resistance value of materials with relatively low RRR values (say RRR< 10) should not be significantly affected by using at any temperature between 38 K and 4.20 K.

ACKNOWLEDGMENTS

This work is supported by the National Magnetic Confinement Fusion Science Program (No. 2015GB121001).

REFERENCES

1. Hua, C.Y, Deng, T.Z, “Discussion about several issues were in the international standards for testing residual resistivity ratio of the compositing superconducting wire NbTi,” Journal of low temperature physics. 1997(06), pp. 21-27. (in Chinese)

2. Wen, M.H, “the RRR testing of niobium material in Superconducting resonator cavity through eddy current nondestructive method,” Journal of low temperature physics. 1998(03), pp. 54-58. (in Chinese)
3. Hua, C.Y, “Determining the content and distribution of impurities in high purity metal through testing residual resistivity ratio,” Rare metal. 1987(02), pp. 126-129. (in Chinese)
4. Ni, Y.M, “The resistivity measurement for metal samples based on eddy current method,” Low temperature physics, 1981(02), pp. 139-143. (in Chinese)
5. Rosenberg, H.M, “Low Temperature Solid State Physics,” Oxford: Clarendon Press, 1963.
6. Fickett, F.R, “Aluminum—A review of resistive mechanisms in aluminum,” Cryogenics. 1971, 11(5): pp. 349-367.
7. Yan, S.S, “Principle and method of physical experiment in low temperature,” Beijing: Science Press, 1985. (in Chinese)
8. Reed, R.P, Clark, A.F, “Materials at Low Temperatures,” Metals Park, Ohio 44073: American Society for Metals, 1983.
9. Clark, A.F, Childs, G.E, Wallace, G.H, “Electrical resistivity of some engineering alloys at low temperatures,” Cryogenics (1970), pp. 295-305.
10. Qiu, L.M, Zhang, K.H, Dong, W.Q, et al, “Study of mass flow distribution between stages in a two-stage pulse tube cryocooler capable of 1.1 W at 4.2 K,” International Journal of Refrigeration. 2012, 35(8), pp. 2332-2338.