Int J Thermophys (2009) 30:316–324
DOI 10.1007/s10765-008-0478-x

Low-Magnetoresistance RuO_2–Al_2O_3 Thin-Film Thermometer and its Application

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Published online: 1 July 2008

Abstract Thermometers consisting of RuO_2–Al_2O_3 composite thin films were prepared by RF sputtering. It was found that different electrode-patterning techniques have dissimilar effects on the magnetoresistance (MR) and the temperature coefficient of resistance (TCR). In general, the thermometers with electrodes fabricated by photo-resist lithography exhibit superior performance compared to those with electrodes prepared using a metal mask. By adjusting the relative compositions of RuO_2 and Al_2O_3, the thermometers can be applied to a wide temperature range from 60 mK to 500 K. In a pulsed magnetic field up to 55 T, the MR at 4.2 K of a typical thermometer for the temperature range from 1.4 K to 300 K increases linearly with magnetic field to a maximum of $\sim$15 %, corresponding to a temperature deviation of $\sim$−4 %. As frequency increases from dc to 1.9 MHz, the MR decreases from −13 % to $\sim$−0.5 % at $T = 1.3$ K and $H = 55$ T. By integrating the thermometer with a heater on a sapphire chip, a micro-calorimeter can be developed and successfully used to measure the heat capacity of small mg-sized sample. The RuO_2–Al_2O_3 composite film can also be employed as an infrared bolometer operated at room temperature.

Keywords Composite thin film · Magnetoresistance · Microcalorimeter · Thermometer

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1 Introduction

The search for good sensors is a common interest of researchers in the field of cryogenics and thermometry. Different types of temperature sensors have been developed for various applications. Sensors are required to meet certain criteria for their practical application to be successful. For example, it should be convenient to use, be robust with respect to thermal cycling with high stability, be applicable to a wide temperature range with appropriate sensitivity, and be minimally influenced by a magnetic field. A good resistive temperature sensor should be applicable to a wide temperature range with acceptable logarithmic sensitivity, $S = \mathrm{d}(\log R)/\mathrm{d}(\log T)$, and should exhibit small field-induced magneto-resistance (MR) $\mathrm{d}R(H)/R(0)$.

Many kinds of resistive temperature sensors have been invented and further improved. Resistive temperature thermometers such as RuO_2 resistors and RuO_2-glass thick-film resistors [1,2] have been widely used at low temperatures, due to their low MR, typically within (2 to 3) % at 2 K for fields up to 10 T [3], and high logarithmic sensitivity, with $S \sim 0.8$ at 1 K. Carbon glass and zirconium oxynitride film (Cernox) thermometers [4] are alternative choices, especially for use in magnetic fields, due to their low MR.

Our resistive RuO_2–Al_2O_3 composite thin-film thermometer [5] has shown its merits in field-induced MR, temperature sensitivity, thermal stability, and ease of preparation. In this work, we extend the MR study to magnetic fields up to 55 T. We also report the influence of electrode preparation on the performance of temperature sensors, especially in minimizing MR effects and maximizing the temperature coefficient of resistance (TCR). For example, the magnetic field-dependence temperature error $\mathrm{d}T/T$ is significantly improved for a thermometer with electrodes fabricated by photo-resist lithography; for a typical thermometer used from 0.3 K to 20 K, its temperature error is $\sim 0.1\%$ for fields up to 8 T at $T = 2.5$ K. A micro-calorimeter made using the thermometer for heat-capacity measurements will also be briefly discussed.

2 Experiments and Results

The thermometer is made of a RuO_2–Al_2O_3 composite thin film, consisting of conducting RuO_2 and non-conducting Al_2O_3, that is fabricated by RF sputtering in Ar gas on a sapphire chip (Al_2O_3 single crystal). The structural similarity between the film and the substrate gives the film and substrate superior bonding and thermal conductance. By varying the relative composition of the components and the control parameters of the sputtering, the thermometers can be fabricated and tailored to a desired temperature region with high logarithmic sensitivity, $S = \mathrm{d}(\log R)/\mathrm{d}(\log T)$. To improve the electric contact and reduce the resistance, patterned electrodes are deposited on the film. Two methods of patterning, metal-mask shielding and photo-resist lithography, are applied to fabricate the electrodes. To represent the two kinds of thermometers in the text later, those with electrodes made using the metal mask will be represented by the index ‘metal’; whereas thermometers with electrodes made by photo-resist lithography will be represented by the index ‘photo’.

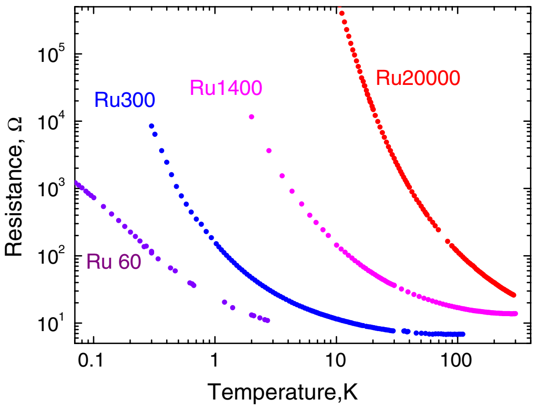

Fig. 1 Temperature-dependent resistances of four RuO_2–Al_2O_3 thin-film thermometers with electrodes made by photo-resist lithography

The temperature-dependent resistances of four thermometers having different compositions with electrodes made by photo-resist lithography is shown in Fig. 1. This result is comparable to other similar types of film resistors composed of conducting and insulating components. Regarding the nomenclature of the thermometers, e.g., Ru1400, the index 1400 represents the lowest temperature (1400 mK) to which the thermometer can be usefully employed. Results for thermometers with electrodes made using a metal mask are shown in Fig. 2 [5]. Obviously the R–T curvature and its smoothness for thermometers with electrodes made by photo-resist lithography is much improved compared to those with electrodes made using a metal mask. For direct comparison, the temperature dependence of resistance for thermometers Ru1400-metal and Ru1400-photo is shown in Fig. 3. With the advantage of delicate patterning techniques by photo-resist lithography, electrode gaps with sharp edges can be made very narrow. Due to the small gap between the electrodes, the resistance of the Ru1400-photo can be dramatically reduced to about one or two orders of magnitude less than that of the Ru1400-metal. For both types, the absolute value of the logarithmic sensitivity, $|S|$, increases rapidly from about 1 to 4 as the temperature decreases from 30 K to 2 K (see inset of Fig. 3). The data of Ru1400-photo also show a smoother profile than that of Ru1400-metal. The field dependence of the MR and the corresponding temperature deviation for thermometers Ru1400-metal and Ru1400-photo at $T = 2$ K to 5 K are shown in Figs. 4 and 5, respectively. The field-induced MR and temperature deviation increase monotonically with increasing magnetic field for all measuring temperatures. For Ru1400-metal at $T = 2$ K and 3 K, the signs of the MR are negative; the maximum $\mathrm{d}R/R$ values are -5.5 % and -0.5 % for 2 K and 3 K, respectively, at $H = 9$ T. The MR becomes positive with values of 1.5 % and 2.5 % for 4 K and 5 K, respectively, at $H = 9$ T. For Ru1400-photo, the signs of the MR are positive for all temperatures; the MR increases from 0.01 % at 2 K to a maximum of 5.2 % at 4 K, and then decreases to 0.04 % as the temperature increases to 5 K.

The field dependence of the temperature deviation can be derived from the equation $\mathrm{d}T/T = (\mathrm{d}R/R)/S$. Since $S > 2$ for $T < 10$ K, the absolute value of $\mathrm{d}T/T$ is much less than $\mathrm{d}R/R$, as shown in Fig. 5. For Ru1400-photo, the maximum value is about 2 % at $T = 4$ K for $H = 9$ T. The sign of $\mathrm{d}T/T$ is opposite to that of $\mathrm{d}R/R$ because

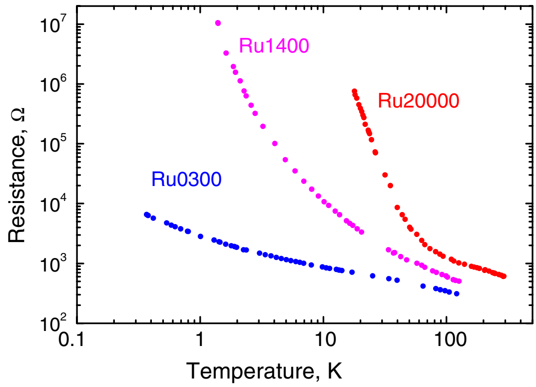

Fig. 2 Temperature-dependent resistances of three RuO_2–Al_2O_3 thin-film thermometers with electrodes made using a metal mask

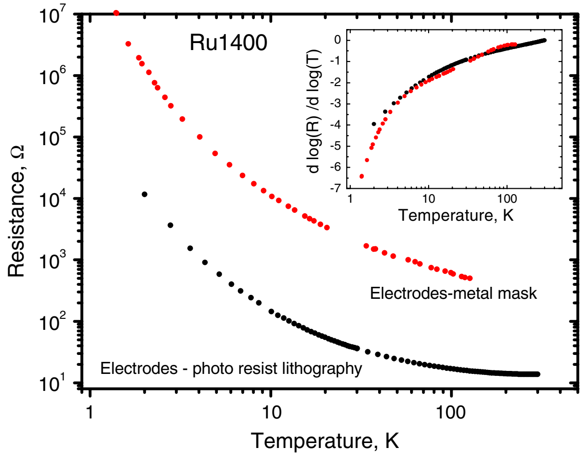

Fig. 3 Temperature-dependent resistances of thermometers Ru1400-metal and Ru1400-photo. Inset: The temperature dependence of logarithmic sensitivity, $S = \mathrm{d}(\log R)/\mathrm{d}(\log T)$

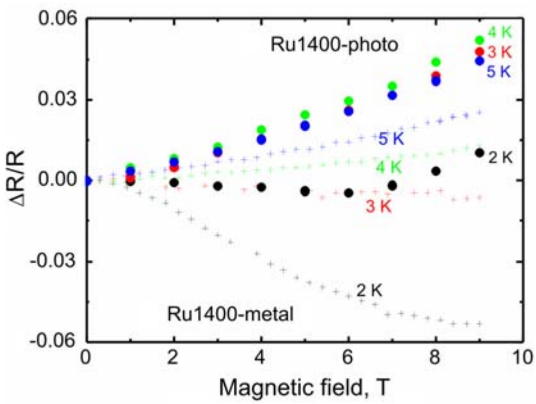

Fig. 4 Field-dependent MRs of thermometers Ru1400-metal (*cross symbols*) and Ru1400-photo (*solid circles*) at $T = (2, 3, 4, \text{and } 5)\,\mathrm{K}$

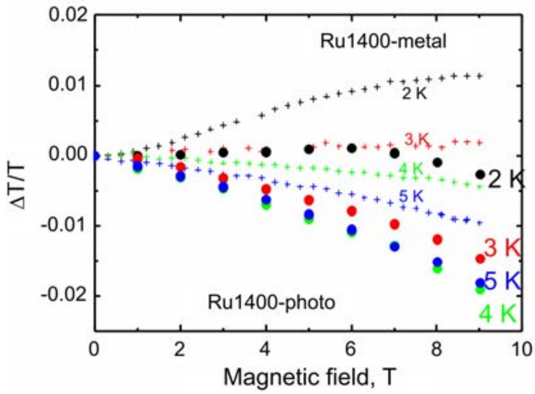

Fig. 5 Field-dependent temperature deviations of thermometers Ru1400-metal (*cross symbols*) and Ru1400-photo (*solid circles*) at $T = (2, 3, 4, \text{and } 5)$ K

Table 1 Temperature deviation in a magnetic field for thermometers Ru1400-metal and Ru1400-photo

T(K)	Type	Senstivity S	$\Delta T/T$ (%)			
			$B = 2$T	$B = 4$T	$B = 6$T	$B = 8$T
2	Ru1400-metal	−4.70	0.15	0.47	0.72	0.87
	Ru1400-photo	−3.95	0.02	0.06	0.11	0.09
	Cernox CX-1050	−1.944	1.1	−2.25	2.77	3.06
4.2	Ru1400-metal	−2.80	−0.06	−0.10	−0.11	−0.14
	Ru1400-photo	−2.68	−0.36	−0.70	−1.11	−1.51
	Cernox CX-1050	−1.33	0.11	0.2	0.13	0.11

Data of a Cernox thermometer CX-1050 from LakeShore Cryotronics, Inc. are also included for comparison

S is negative. The numerical data are listed in Table 1. For comparison, the data of a Cernox thermometer CX-1050 from LakeShore Cryotronics, Inc are also included [4].

To examine the temperature measurement uncertainty, the temperature deviation was statistically calculated from the data taken during thermal cyclings. The temperature uncertainty for the temperature range of 0.5 K to 10 K was estimated to be around ±0.3 %, corresponding to ±0.6 mK at 2 K and ±30 mK at 10 K for a typical Ru1400-photo thermometer. In general, the result is comparable to most other currently available low-temperature thermometers [4,5].

The field dependence of the MR and the corresponding temperature deviation for a Ru300-photo thermometer at $T = (0.3, 1, 2, \text{and } 3)$ K are shown in Figs. 6 and 7, respectively. A similar result to that for the Ru1400-photo thermometer is obtained. The field-induced MR and temperature deviation increase monotonically with increasing magnetic field. $\mathrm{d}R/R$ at $T = 2$ K for $H = 9$ T is about 0.3 %. Due to the large value of the logarithmic sensitivity, $S = -3$, the derived $\mathrm{d}T/T$ is only −0.1 %. Based on the results discussed above, we concluded that the magnetic field-dependent temperature error $\mathrm{d}T/T$ can be substantially improved if the electrodes are fabricated by photo-resist lithography.

It has been reported that the MR of composite materials is independent of the relative orientation of the measuring current to the magnetic field [4]. Figure 8 shows the

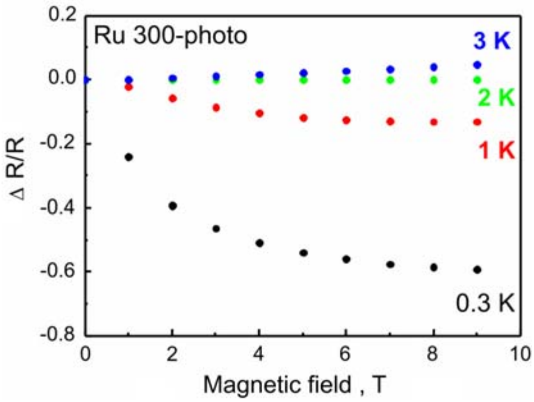

Fig. 6 Field-dependent MR of a Ru300-photo thermometer at $T = (0.3, 1, 2, \text{and } 3)\,\mathrm{K}$

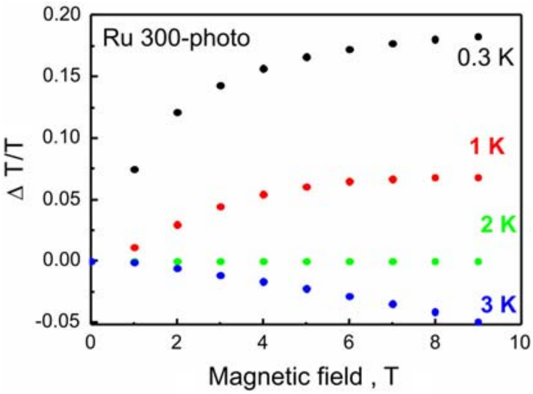

Fig. 7 Field-dependent temperature deviation of a Ru300-photo thermometer at $T = (0.3, 1, 2, \text{and } 3)\,\mathrm{K}$

MR taken for three different orientations of the magnetic field relative to the measuring current and the plane of the film at two different temperatures, 2 K and 5 K. No noticeable difference can be observed for different orientations among magnetic field, measuring current, and film plane.

For a Ru1400-metal thermometer, the field dependence of MR under static fields up to 9 T is shown in Fig. 9. The MR $\mathrm{d}R/R$ increases approximately linearly with the magnitude of the magnetic field with a rate of $-0.005\,\mathrm{T}^{-1}$ and $0.0027\,\mathrm{T}^{-1}$ at 2 K and 5 K, respectively. To see if the evolution of MR with magnetic field either keeps on increasing or saturates at a constant value, measurements at higher magnetic fields are required. The field dependence of MR for magnetic fields up to 55 T was measured at the International Mega Gauss Science Laboratory, ISSP, at the University of Tokyo using a non-destructive pulsed magnet with a duration time of 42 ms. The high-field measurements were performed at temperatures of 1.3 K to 4.2 K using a dc potentiometric method and AC lock-in techniques (Fig. 9). Maxima of $\mathrm{d}R/R \sim 15\,\%$ at $H = 25\,\mathrm{T}$ and 55 T for 1.3 K and 4.2 K are observed, respectively. Comparing the $\mathrm{d}R/R$ taken in a dc magnetic field to that in a pulsed field at $T =\sim 4.2\,\mathrm{K}$, we observed that for the same temperature and magnetic field, the MR in a static field will be slightly larger than

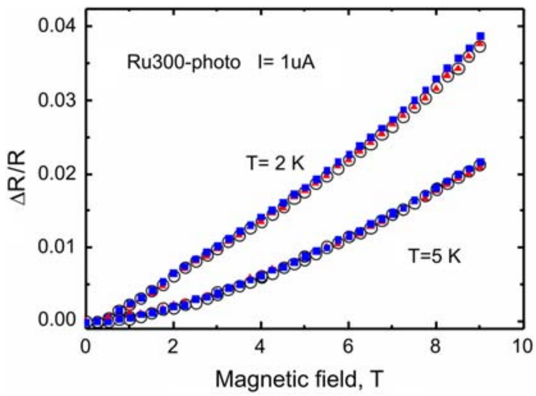

Fig. 8 The MR for different relative orientations of the measuring current to the magnetic field. The circles represent the field perpendicular to the film plane, the triangles represent the field parallel to the current direction, and the squares represent the field in the plane and perpendicular to the current direction

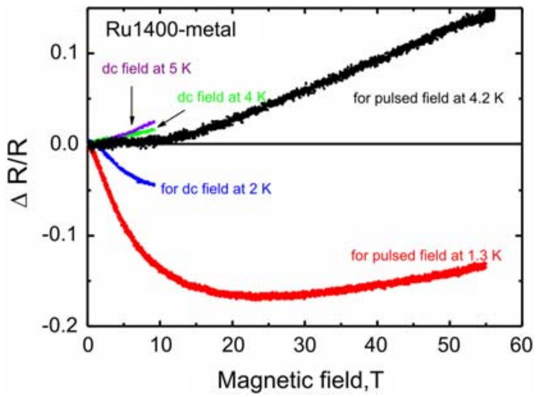

Fig. 9 Field-dependent MR of a Ru1400-photo thermometer

that in a pulsed field. The sign of dR/R is negative for $T \leq 2$ K and becomes positive for $T \geq 4$ K. At the transition temperature (between 2 K and 4 K) where the sign of the MR changes from negative to positive, the MR will approach zero. Since $S > 3.5$ for $T < 4.2$ K, the absolute value of dT/T is much less, as shown in Fig. 10: the absolute value of the maximum dT/T at $H = 55$ T is approximately 4 % for $T = 4.2$ K and 1.3 K. The result is comparable with other currently available low-temperature thermometers. For comparison, the data of field-induced temperature deviation taken at 2 K and 4.2 K for thermometers Ru1400-metal, Ru1400-photo, and a Cernox CX-1050 thermometer are tabulated in Table 1. In general, the field-induced temperature deviation of Ru1400 is comparable to that of the Cernox CX-1050.

The frequency dependence of the MR for a Ru1400-photo thermometer in a pulsed magnetic field up to 55 T is shown in Fig. 11. The maximum MR dR/R at $H \sim 25$ T falls from 16.5 % to 0.5 % as the frequency of the measuring current increases from dc to 1.9 MHz.

Taking advantage of the small mass of the RuO_2–Al_2O_3 composite thin film and its good thermal contact with the sapphire substrate, a thin sapphire chip on which a RuO_2–Al_2O_3 film and a Ni–Cr heater were deposited was employed as a micro-calorimeter for heat-capacity measurements (Fig. 12). The method of heat-capacity

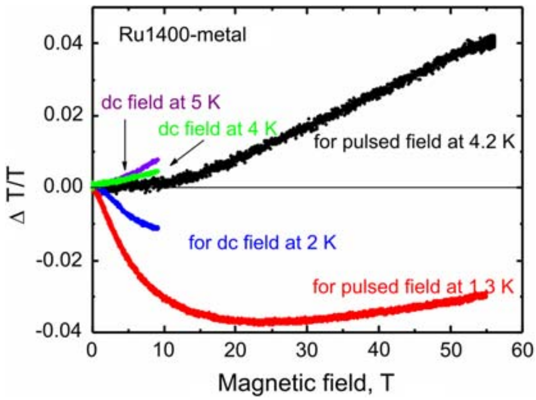

Fig. 10 Field-dependent temperature deviation of a Ru1400-photo thermometer

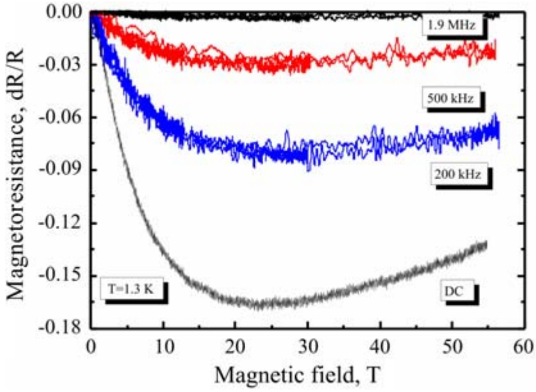

Fig. 11 Frequency dependence of the MR of a Ru1400-photo thermometer

measurement was described previously [5]. Another use for the RuO_2–Al_2O_3 composite thin film is its application to bolometry. With special compositions of RuO_2 and Al_2O_3, the TCR of a selected RuO_2–Al_2O_3 composite film can be as high as 1.6 at room temperature. Integrated with semiconductor industry techniques, an infrared bolometer consisting of a suspended RuO_2–Al_2O_3 composite film with a working temperature near room temperature can also be employed.

3 Conclusion

We have reported a RuO_2–Al_2O_3 composite thin film that exhibits low MR and temperature deviation in a magnetic field. With photo-resist lithography to pattern the electrode, the performance of the RuO_2-Al_2O_3 film thermometer was improved as compared to those having electrodes prepared by means of a metal mask. The field dependence of MR is dependent on the measuring temperature; for the case of a Ru1400 thermometer, its maximum is located at 25 T for $T = 1.3$ K, whereas for $T = 4.2$ K it increases linearly with magnetic field up to 55 T. The MR measured in a pulsed field is slightly smaller than that measured in a dc field. Due to the small mass and superior

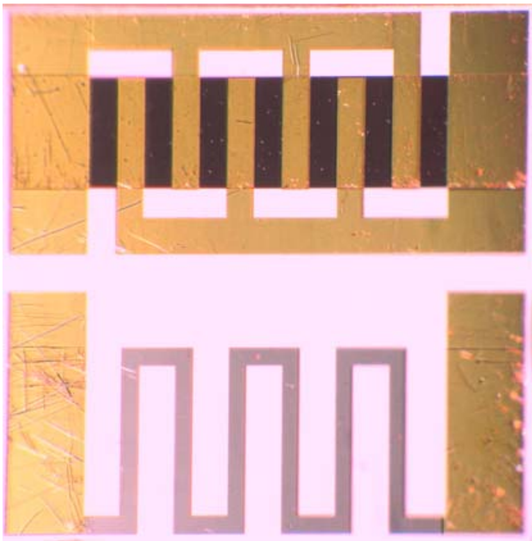

Fig. 12 A micro-calorimeter made by integrating a thermometer (top half) and a Ni–Cr heater (bottom half) on a sapphire single crystal. Dimensions of the micro-calorimeter are 3 mm × 3 mm × 0.2 mm

thermal contact with the sapphire substrate, a highly sensitive microcalorimeter can be made using the RuO_2–Al_2O_3 thin film. The RuO_2–Al_2O_3 composite film with a high TCR value can also be employed as a room-temperature infrared bolometer.

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