Sensitive germanium thermistors for cryogenic thermal detector of Tokyo dark matter search programme

Wataru Ootani, Yutaka Ito, Keiji Nishigaki, Yasuhiro Kishimoto, and Makoto Minowa

*Department of Physics, Graduate School of Science, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113, Japan*

Youiti Ootuka

*Cryogenic Center, University of Tokyo, 2-11-16 Yayoi, Bunkyo-ku, Tokyo 113, Japan*

Abstract

Sensitive n-type and p-type germanium thermistors were fabricated by the melt doping technique and by the neutron transmutation doping (NTD) technique, respectively, aiming at a use for the cryogenic thermal detector, or bolometer of Tokyo dark matter search programme. We report on the measurements of the sensitivities of these thermistors. In particular, the p-type thermistors are sensitive enough to scale up our existing prototype LiF bolometer and realize a multiple array of the bolometers with the total absorber mass of about 1 kg.
I. INTRODUCTION

One of the most important issues of the modern physics is the nature of the dark matter. Weakly interacting massive particles (WIMPs) is considered as the leading candidates for the particle dark matter. A direct search for WIMPs using a cryogenic thermal detector, or bolometer is planned. This programme is now under way and a prototype of the detector is already completed [1]. A bolometer consists of an absorber in which a WIMP scatters off a nucleus elastically and a thermistor which senses the resulting temperature rise of the absorber. Operation at very low temperature leads to small specific heat of the absorber. A bolometer offers significant advantages for the dark matter search because of its high energy resolution, low energy threshold, and flexibility on a choice of the absorber materials.

We developed a prototype bolometer with a 2.8-g LiF absorber [1] on the grounds that an elastic scattering cross section of the axially-coupled WIMPs on $^{19}$F (natural abundance 100%) is fairly larger than on other nuclei like $^{73}$Ge, $^{23}$Na, and $^{27}$Al [1,2]. Its energy resolution was 3.8 keV (rms) for 60-keV $\gamma$ rays. Absorber mass of about 1 kg is, however, required for searching for the WIMPs. We are planning to scale up the prototype bolometer and construct a multiple array of the bolometers with the total mass of about 1 kg. In realizing this scaling-up of the bolometer, we need more sensitive thermistors, as well as a reduction of the noise level.

To our knowledge, there is no ready-made thermistor that can be used below 50mK with a sufficient sensitivity and has a small size which leads to small specific heat. Many attempts to develop sensitive thermistors have, therefore, been made so far by different groups, using superconductors [3], heavily doped semiconductors [4], or thin alloy films [5]. A heavily doped germanium thermistor is used in our prototype LiF bolometer [1].

Electrical conductance in heavily doped and compensated semiconductors at low temper-
ature is dominated by variable hopping conductance \[6\]. In this conduction regime, charge carriers tunnel from an occupied dopant site to an unoccupied dopant site. The uniformity of the dopant distribution and the ratio of the minority dopants to the majority dopants (compensation ratio) are critical parameters as well as the dopant concentration. The temperature dependence of the resistance \( R \) of doped germanium predicted in this conduction regime is

\[
R(T) = R_0 \exp \left( \frac{\Delta}{T} \right)^{\frac{1}{2}},
\]

where \( R_0 \) and \( \Delta \) are constant parameters, and \( T \) is the temperature in Kelvin. The dimensionless sensitivity of a thermistor can be defined by the logarithmic slope \( n \):

\[
n = -\frac{d \ln R}{d \ln T} = \frac{1}{2} \left( \frac{\Delta}{T} \right)^{\frac{1}{2}}.
\]

A fabrication of thermistors by doping semiconductors is quite delicate because an impurity concentration very close to the metal-insulator transition is required. It also needs to be uniform everywhere in a sample. Furthermore, an amount of thermistors with similar characteristics are needed for the construction of a multiple array of the bolometers.

The present paper reports on the measurements of the sensitivities of the germanium thermistors fabricated by the melt doping technique and the neutron transmutation doping (NTD) technique. Particularly, the latter were very sensitive and reproducible as we shall see later in Sec. \[IV\]. The thermistors by the NTD technique have been developed so far by LBL group \[4\], but ours have slightly higher sensitivities.
II. MANUFACTURE OF N-TYPE GERMANIUM THERMISTORS BY MELT DOPING TECHNIQUE

The melt doping is one of the conventional techniques to dope semiconductors. Our n-type germanium thermistors which is used in our prototype LiF bolometer are doped with antimony (n-type dopant) and compensated with copper (p-type dopant).

The starting materials available to us were germanium wafers doped with antimony, whose resistivities ranged from $2.55 \times 10^{-2}$ to $3.22 \times 10^{-2} \Omega \text{cm}$ at room temperature. This resistivity range corresponds to the charge carrier concentration from $1.26 \times 10^{17}$ to $1.00 \times 10^{17} \text{cm}^{-3}$, which is far from the metal-insulator transition. In order to reduce the effective charge carrier concentration the samples were compensated with copper which is opposite type dopant.

The thermistors were manufactured by the following procedure. A chip with dimensions of $1.5 \times 1.5 \times 1^t \text{mm}^3$ was cut from the wafer. A 1% aqueous solution of copper nitrate was smeared on one square face of the chip. The chip was annealed at $800^\circ \text{C}$ for twenty minutes to diffuse copper. A solid solubility limit of copper in germanium at $800^\circ \text{C}$ fixes the copper concentration of the chip at $1.29 \times 10^{16} \text{cm}^{-3}$. The resulting charge carrier concentration and the compensation ratio of each sample are listed in Table I.

In order to form ohmic contacts antimony pads with a thickness of $1000 \text{Å}$ were evaporated on two areas on one square face and gold pads with a thickness of $1000 \text{Å}$ followed, as shown in Fig. I. The chip was annealed at $600^\circ \text{C}$ for thirty minutes to activate the contacts. Gold wires with a diameter of $50 \mu\text{m}$ were attached to the contacts by the spot welding.
The neutron transmutation doping (NTD) which is a technique to dope semiconductors by partial transmutation of its stable isotopes to dopants via thermal neutron capture, was first put into practical use in high power device with silicon [10]. Since the NTD technique provides uniform dopants concentration in the crystal, large area silicon device became available.

The NTD process can provide much more uniform distribution of the dopants in germanium. Impurity conduction in the NTD germanium was experimentally studied by Fritzsche et al [11]. Thermal neutrons from a nuclear reactor are captured by the five isotopes contained in natural germanium; $^{70}\text{Ge}$, $^{72}\text{Ge}$, $^{73}\text{Ge}$, $^{74}\text{Ge}$, and $^{76}\text{Ge}$. The available information of these reactions are listed in Table II [12], which indicates $^{70}\text{Ge}$, $^{74}\text{Ge}$, and $^{76}\text{Ge}$ are transmuted to the p-type majority dopants or n-type minority dopants and the others are not. Net hole concentration can be controlled by the amount of the irradiated thermal neutrons. Furthermore, the moderate cross sections of thermal-neutron capture allow one to dope a large volume crystal of germanium with extremely uniform dopant concentration. The compensation ratio is fixed to 0.32 due to the fixed abundance of germanium isotopes and the cross sections of thermal-neutron capture.

Pure germanium single crystals with resistivities of over 50\,Ω\,cm were exposed to the thermal neutrons in the JRR-3M reactor at Japan Atomic Energy Research Institute. The amount of the irradiated thermal neutrons and the resulting net hole concentration are listed in Table III.

After the exposure a chip with a dimension of $1.5 \times 1 \times 1^t$ mm$^3$ was cut from the crystal. In order to remove the undesired radiation damage due to fast neutrons, the chip was
annealed at 400°C for ninety minutes. Boron (p-type dopant) ions with a dose of $3 \times 10^{14}$ cm$^{-2}$ were implanted on two areas on one face of the chip to a depth of 1200 Å in order to form heavily doped regions which yield ohmic contacts. Gold pads as thick as 1500 Å were evaporated on the implanted sites. The contacts were activated by annealing the chip at 250°C for ninety minutes. Gold wires with a diameter of 50 µm were attached to the contacts in a similar way as the n-type thermistors (Fig. 1).

IV. MEASUREMENTS AND RESULTS

The measurements of the temperature dependence of the resistance of the thermistors were carried out in an Oxford Instruments refrigerator equipped with a $^3$He/$^4$He dilution unit with a cooling power of 600 µW at 100 mK. The whole system except the pumping system is housed in an electromagnetic shielding room.

The fabricated n-type or p-type thermistors were mounted onto a copper plate which was thermally anchored to the mixing chamber of the dilution unit, and cooled down to between 30 and 110 mK. The temperature of the mixing chamber was monitored by a carbon resistor, a $^{60}$Co nuclear orientation thermometer, or both. The thermistor was glued with GE varnish to a Kapton foil with a thickness of 7.5 µm which was glued to the copper plate. A Kapton foil is an insulating foil with a good thermal conductivity.

The thermistor was biased at a current between 50 pA and 100 nA with load resistors by an external battery. Since a nonlinearity of the current-voltage relations of the thermistor was conspicuous at low temperature, the resistance was determined by the zero bias resistance $(dV/dI)_{I=0}$. The measured temperature dependence of the resistance ($R$-$T$ curve) of the n-type germanium thermistors by the melt doping technique is given in Fig. 2 and of the p-type germanium thermistors by the NTD technique in Fig. 3. The solid lines in the figures are the best fits to the data. The parameters $R_0$ and $\Delta$ in Eq. (1) and the dimensionless
sensitivities $n$ at 20 mK extracted from these $R$-$T$ curves are presented in Table I for the n-type thermistors and Table III for the p-type thermistors.

Fig. 2 shows that for the n-type thermistors both the slope of the $R$-$T$ curve and the resistance at the same temperature decrease with the charge carrier concentration. Therefore, one can see that the estimated charge carrier concentration is a parameter to control the sensitivity of the n-type germanium thermistor by the melt doping technique. While the dimensionless sensitivity $n$ is proportional to the slope of the $R$-$T$ curve as Eq. (2) indicates, the resistance from a few MΩ to a few hundred MΩ is desirable at the operating temperature in order to avoid electronic noise such as Johnson noise. From this point of view the sample MDGe3 is most favorable among the measured n-type thermistors and is used in our prototype LiF bolometer. The drawback in the manufacture of the n-type thermistors by the melt doping technique is that the dopants distribution it provides is not extremely uniform, and it leads to poor reproducibility in the $R$-$T$ characteristics. This poor reproducibility was observed in our experiments.

Fig. 3 also shows that for the p-type thermistors both the slope of the $R$-$T$ curve and the value of $R$ at the same temperature decrease with the charge carrier concentration in the same way as the n-type thermistors. From the noise consideration mentioned above, the sample NTDGe3 is most favorable. The sensitivity of the p-type thermistor was much higher than that of the n-type thermistor. Furthermore, the $R$-$T$ characteristics were very reproducible for the thermistors exposed to the same amount of the thermal neutrons. The $R$-$T$ curves of five NTDGe3 thermistors with the same irradiation dose of $3.42 \times 10^{18}$ neutrons cm$^{-2}$ are shown in Fig. IV. It is apparent that these five thermistors have similar $R$-$T$ characteristics. This good reproducibility of the p-type thermistors would provide a significant advantage in constructing a multiple array of the bolometers.
V. SUMMARY AND DISCUSSIONS

In summary, we fabricated n-type and p-type germanium thermistors by the melt doping technique and by the NTD technique, respectively, and measured the $R$-$T$ characteristics of the thermistors. We controlled the sensitivity of the n-type thermistor by the measured resistivity at room temperature before compensating and of the p-type thermistor by the amount of the irradiated thermal neutrons.

The p-type thermistor had much higher sensitivity and better reproducibility than the n-type thermistor. These differences are considered to be primarily due to the facts that the impurity distribution in the NTD is much more uniform than that in the melt doping and the compensation ratio of the p-type thermistor is different from that of the n-type thermistor. The sensitivity of the NTDGe3 thermistor, which has most suitable $R$-$T$ characteristics for our purpose, is by a factor of 1.2 higher than that of the NTD thermistor with similar resistance fabricated by LBL group [4]. The differences in the geometry of the thermistor and the configuration of the contacts may cause this small difference in the sensitivity.

The improvement on the resolution of the prototype 2.8-g LiF bolometer in using the NTDGe3 thermistor can be estimated. Assuming the same input power into the thermistor and the thermistor temperature of 20 mK, the resolution could be improved by an order of magnitude. This improvement could allow us to realize the absorber mass of 30 g and construct multiple array of the bolometers with similar sensitivities to perform the dark matter search with a total absorber mass of about 1 kg.

ACKNOWLEDGEMENTS

We would like to thank Prof. Y. Ito and H. Sawahata for irradiating germanium in the JRR-3M reactor at Japan Atomic Energy Research Institute, Y. Koizumi for very useful
discussions on radioactive contaminations of the irradiated germanium, and Dr. H. Bando for providing us with the dilution refrigerator. This work has been supported by the Inter-University Program for the Joint Use of JAERI Facilities. This work is financially supported by the CASIO Science Promotion Foundation, the Iwatani Naoji Foundation’s Research Grant, the Yamada Science Foundation, and the Grant-in-Aid for Scientific Research (A) and the Grant-in-Aid for Developmental Scientific Research by the Japanese Ministry of Culture, Education and Science.
REFERENCES

[1] M. Minowa et al., Nucl. Inst. Meth. A 327 (1993) 612;
   M. Minowa et al., J. Low Temp. Phys. 93 (1993) 803.
[2] J. Ellis and R. A. Flores, Phys. Lett. B 263 (1991) 259.
[3] P. Colling et al., Nucl. Inst. Meth. A 354 (1995) 408.
[4] É. Aubourg et al., J. Low Temp. Phys. 93 (1993) 289;
   N. Wang et al., Phys. Rev. B 41 (1990) 3761;
   K. M. Itoh et al., J. Low Temp. Phys. 93 (1993) 307.
[5] L. Dumoulin et al., J. Low Temp. Phys. 93 (1993) 301.
[6] B. I. Shklovskii and A. L. Efros, Electronic Properties of Doped Semiconductors, Solid-
   State Science vol.45, Springer-Verlag, Berlin Heidelberg New York Tokyo, 1984.
[7] D. B. Cuttriss, Bell System Technical Journal 40 (1961) 509;
   M. B. Prince, Phys. Rev. 92 (1953) 681.
[8] H. Fritzsche, J. Phys. Chem. Solids 6 (1958) 69.
[9] F. A. Trumbore, Bell System Technical Journal 39 (1960) 205.
[10] Neutron Transmutaion Doping in Semiconductors, edited by J. M. Meese (Plenum Press,
    New York, 1979);
    E. E. Haller et al. in Neutron Transmutaion Doping of Semiconductor Materials, edited
    by R. D. Larrabee (Plenum Press, New York, 1984), p21.
[11] H. Fritzsche and K. Lark-Horovitz, Phys. Rev. 113 (1959) 999;
    H. Fritzsche and M. Cuevas, Phys. Rev. 119 (1960) 1238.
[12] Table of Isotopes, seventh edition, edited by C. M. Lederer and V. S. Shirley (JOHN
    WILEY & SONS, INC., New York· Chichester· Brisbane· Toronto,1978).
TABLES

TABLE I. Parameters of the n-type thermistors by the melt doping technique

| Sample  | Charge carrier concentration (cm\(^{-3}\)) | Compensation ratio | \(R_0\) (\(\Omega\)) | \(\Delta\) (K) | Sensitivity \(n\) |
|---------|------------------------------------------|--------------------|----------------------|----------------|------------------|
| MDGe1   | \(8.1 \times 10^{16}\)                  | 0.19               | 148                  | 7.51           | 9.69             |
| MDGe2   | \(8.9 \times 10^{16}\)                  | 0.18               | 148                  | 5.74           | 8.47             |
| MDGe3   | \(9.1 \times 10^{16}\)                  | 0.17               | 32.1                 | 4.37           | 7.39             |
| MDGe4   | \(1.1 \times 10^{17}\)                  | 0.15               | 6.97                 | 3.64           | 6.75             |
| MDGe5   | \(1.2 \times 10^{17}\)                  | 0.14               | 112                  | 0.192          | 1.55             |
| Isotope  | Abundance (%) | Cross section (barn) | Reaction | Dopant type |
|----------|---------------|----------------------|----------|-------------|
| $^{70}_{32}$Ge | 20.5 | 3.25 | $^{70}_{32}$Ge$(n, \gamma)^{71}_{32}$Ge$^{11.2d} \rightarrow ^{71}_{31}$Ga | p |
| $^{72}_{32}$Ge | 27.4 | 1.0 | $^{72}_{32}$Ge$(n, \gamma)^{73}_{32}$Ge | – |
| $^{73}_{32}$Ge | 7.8 | 15 | $^{73}_{32}$Ge$(n, \gamma)^{74}_{32}$Ge | – |
| $^{74}_{32}$Ge | 36.5 | 0.52 | $^{74}_{32}$Ge$(n, \gamma)^{75}_{32}$Ge$^{82.8m} \rightarrow ^{75}_{33}$As | n |
| $^{76}_{32}$Ge | 7.8 | 0.16 | $^{76}_{32}$Ge$(n, \gamma)^{77}_{32}$Ge$^{11.3h} \rightarrow ^{77}_{35}$As$^{38.8h} \rightarrow ^{77}_{34}$Se | n |
| Sample  | Irradiation  | Charge carrier concentration | Compensation ratio | $R_0$  | $\Delta$ | Sensitivity |
|---------|--------------|-----------------------------|--------------------|-------|---------|-------------|
| NTDGe2  | $2.85 \times 10^{18}$ | $5.70 \times 10^{16}$ | 0.32               | $5.35 \times 10^{-2}$ | 18.2   | 15.1        |
| NTDGe3  | $3.42 \times 10^{18}$ | $6.84 \times 10^{16}$ | 0.32               | $1.74 \times 10^{-2}$ | 10.6   | 11.5        |
| NTDGe4  | $4.11 \times 10^{18}$ | $8.22 \times 10^{16}$ | 0.32               | $1.68 \times 10^{-1}$ | 2.58   | 5.68        |
FIGURES

FIG. 1. Schematic drawing of the fabricated n-type or p-type germanium thermistor.

FIG. 2. Measured temperature dependence of the resistance of the n-type germanium thermistors by the melt doping technique. Solid lines are the best fits to the data.

FIG. 3. Measured temperature dependence of the resistance of the p-type germanium thermistors by the NTD technique. Solid lines are the best fits to the data.

FIG. 4. $R$-$T$ curves of five NTDGe3 thermistors with the same irradiation dose of $3.42 \times 10^{18}$ neutrons cm$^{-2}$. Solid lines are the best fits to the data.
1.0mm doped germanium contact pads

1.5mm
doped germanium

Au wires

contact pads