MAY 1, 1934 PHYSICAL REVIEW VOLUME 45

Thermocouples for the Measurement of Small Intensities of Radiations*

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(Received January 5, 1934)

The use of a.c. amplification of voltages produced in very thin thermocouples is proposed as a means of measuring small intensities of radiation. The theory of the proper design of such a couple is developed and a comparison of theory and experiment given. The sensitivity available is about 100 times greater than with the usual thermocouple galvanometer arrangement.

THE purpose of this paper is to present the theory underlying the design of thermocouples to be used with a.c. amplification. Measurements made with such couples indicate the feasibility of using a.c. amplification for measurement of small amounts of radiation.

In the usual design of sensitive thermocouples employed for the measurement of radiation it is necessary to limit the resistance of the couples; otherwise full advantage of the galvanometer sensitivity cannot be utilized. This limitation results in an appreciable loss due to heat conduction and prevents the use of multijunctions and of a number of materials which have a very high thermal electromotive force.

The use of d.c. amplification is difficult because it does not permit the measurement of small voltages. However, the use of a.c. amplification seems quite promising. In the first place, parasitic voltages can be minimized, and secondly, up to a certain point, the sensitivity is independent of the resistance of the couple. Losses due to Joule heating and Peltier effect are practically negligible with a.c. amplification. Although the voltage available for a.c. amplification is under the best conditions only 0.45 that available for d.c. measurement, the sensitivity attainable with a.c. amplification is greater than with the galvanometers. The most sensitive galvanometers permit readings of about 5×10^{-8} volt; as low as 10^{-9} volt can be detected with a.c. amplifiers.

It should be mentioned that the use of a thermal relay[1] in conjunction with the thermocouple-galvanometer system, or the use of the resonance principle[2, 3, 4] permits the sensitivity of the usual thermocouple-galvanometer system to be increased about 100-fold. Both these systems involve, however, the use of two galvanometers, vibrationless supports and considerable effort in installation.

The usual technique for the construction of thermocouples is not applicable for thermocouples that must respond to alternating radiation intensities. As will be seen from the development presented below, it is necessary that the thickness of metal be of the order of magnitude of 10^{-5} cm and there must be no accumulation of metals at the junction. The necessity for a low heat capacity precludes the use of a separate receiver. Instead a large number of junctions must be used if it is not possible or desirable to concentrate the radiation beam. The thin couples have the advantage that the heat conduction loss is a small or negligible fraction of the total loss; so that when such couples are maintained *in vacuo* (1 micron or better) the reradiation is essentially the only loss. It was decided to make the couples for this work by cathodic sputtering on thin cellulose. The technique of sputtering and of constructing single and multijunction couples of Bi-Sb and Bi-Te has been described elsewhere.[5]

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* Contribution No. 325 from the Research Laboratory of Physical Chemistry.

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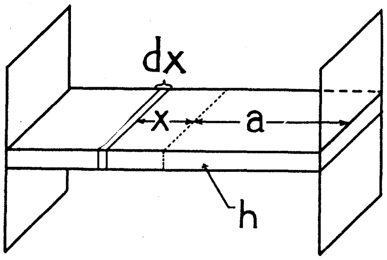

FIG. 1.

Let us now determine what conditions must be satisfied to obtain a couple that will respond to intermittent radiation. A heat balance leads to a differential equation whose solution is found. Consider a strip of metal (on cellulose), 1 cm wide, as illustrated in Fig. 1.

Let Θ = Difference in temperature in degrees Centigrade between metal and surroundings.
h = Thickness of couple (cm).
a = Length from hot to cold junction (cm).
x = Distance from hot junction to the element dx.
c = Heat capacity of metal and cellulose—calories/degrees/gram.
ρ = Density of metal and cellulose—grams/cc.
k = Heat conductivity—calories/second/cm thickness/cm^2/second.
j = Phase factor.
$w = 2\pi f$. f = frequency of intermittent radiation, seconds^{-1}.
t = Time seconds.
A = Intensity of radiation incident upon the couple—calories/cm^2/second.
b = Fraction of incident energy absorbed.
$E\Theta$ = Loss by radiation $= 4\sigma T^3\Theta$ — calories/cm^2/second/degree^4.
T = Temperature degrees Kelvin.
σ = Radiation constant.

Assume that the ends $(x=a)$ are at room temperature. Consider an element of volume along the length of the couple, dx long. The rate of loss by heat conduction is $-k\times 1\times h \times (d^2\Theta/dx^2)dx$. The rate of loss by heat radiation is $E\times\Theta\times 1\times dx$. (The loss from the rear side of the couple is included in E.) The rate of heat absorption $= bAe^{jwt}\times dx\times 1$. The rate of gain of heat energy $c\rho\times 1\times h\times dx\times d\Theta/dt$. Equating the rate of gain to the net gain of heat

$$1\times dx\times Ae^{jwt} - E\Theta\times 1\times dx + k\times h\times dx\times 1 \times d^2\Theta/dx^2 = c\rho\times 1\times h\times dx\times d\Theta/dt,$$

$$c\rho h d\Theta/dt = bAe^{jwt} - E\Theta + khd^2\Theta/dx^2. \quad (1)$$

To solve, let $\Theta = \mathrm{X}e^{jwt}$; X being a function of x alone.

$$d\Theta/dt = \mathrm{X}jwe^{jwt};\quad d^2\Theta/dx^2 = d^2\mathrm{X}/dx^2 e^{jwt}.$$

Substituting into (1)

$$jwc\rho h = bA - E\mathrm{X} + khd^2\mathrm{X}/dx^2,$$

$$d^2\mathrm{X}/dx^2 = -bA/kh + (E+jwc\rho h)/kh. \quad (2)$$

A solution is

$$\mathrm{X} = D - (c_1e^{\alpha x} + c_2e^{-\alpha x}).$$

Boundary conditions: $\Theta = 0$, $x = \pm a$, $\mathrm{X} = 0$.

$$d^2\mathrm{X}/dx^2 = -(c_1\alpha^2e^{\alpha x} + c_2\alpha^2e^{-\alpha x}).$$

Substituting into (2)

$$-(c_1\alpha^2e^{\alpha x} + c_2\alpha^2e^{-\alpha x}) = -bA/kh + [(E+jwc\rho h)/kh](D - (c_1e^{\alpha x} + c_2e^{-\alpha x})). \quad (3)$$

Eq. (3) can be true only if

$$bA/kh - [(E+jwc\rho h)/kh]D = 0;$$

$$D = bA/(E+jwc\rho h).$$

From boundary conditions $(\mathrm{X}=0)$

$$bA/(E+jwc\rho h) - c_1e^{\alpha a} - c_2e^{-\alpha a} = 0,$$

$$bA/(E+jwc\rho h) - c_1e^{-\alpha a} - c_2e^{+\alpha a} = 0,\quad c_1 = c_2$$

and

$$c = [bA/(E+jwc\rho h)][1/(e^{\alpha a} + e^{-\alpha a})]. \quad (4)$$

Substituting into $\Theta = \mathrm{X}e^{jwt}$

$$\Theta = bA/(E+jwc\rho h) \times [1 - (e^{\alpha x} + e^{-\alpha x})/(e^{\alpha a} + e^{-\alpha a})]e^{jwt}. \quad (5)$$

Let H represent the fraction in the bracket. If H can be neglected, the final solution becomes:

$$\Theta = (bA/E)[1/(1+(wc\rho h/E)^2)]^{\frac{1}{2}}e^{jwt} \quad (6)$$

and the solution for the case of uninterrupted light is

$$\Theta = bA/E. \quad (7)$$

The quantity $[1/(1+(wc\rho h/E)^2)]^{\frac{1}{2}}$ called the attenuation factor, is equivalent to an inertia factor and is designated by V.

$$V = \frac{\text{Voltage obtained with intermittent radiation}}{2.2\times\text{voltage obtained with steady radiation}}.$$

In order to have V as large as possible $wc\rho h/E$ must be kept small. Inasmuch as E is constant (at constant temperature) and $c\rho$ does not permit of much choice, the thickness of both metal and cellulose and the frequency of interruption of incident radiation must be kept as low as possible.

The length of each element necessary to keep the value of H small, say below 0.2 of the whole bracketed term in Eq. (5), was determined for the different conditions of operation. If $(e^{\alpha x}+e^{-\alpha x})/(e^{\alpha a}+e^{-\alpha a})<0.2$, then $e^{\alpha a}+e^{-\alpha a}<10$. From Eqs. (3) and (4)

$$\alpha=[(E+jwc\rho h)/kh]^{\frac{1}{2}}$$

and

$$|\alpha|=[(E^2+w^2c^2\rho^2h^2)^{\frac{1}{2}}/kh]^{\frac{1}{2}}.$$

Three cases may be considered

1. $wc\rho h \ll E$ low frequencies and thin couples

$$\alpha=(E/kh)^{\frac{1}{2}}.$$

2. $wc\rho h=E$ $\qquad \alpha=2^{\frac{1}{4}}(E/kh)^{\frac{1}{2}}.$

3. $wc\rho h \gg E$ high frequencies $\qquad \alpha=(wc\rho/k)^{\frac{1}{2}}.$

For thicknesses of metal 5×10^{-5} cm or less the value of H will always be less than 0.2 if a is as much as 0.3 cm. The length of each element of the couple was therefore made 3 to 3.5 mm. The heat conduction loss along the cellulose is negligible for a length of even one mm of cellulose.

It will be observed that the width of the couples has dropped out of the final expression. This is to be anticipated since an increased width results in more energy absorption but simultaneously in an equal amount of radiation loss. The single junction couples were made with the metal strips one millimeter wide, the multijunction couples with the metal strips 0.5 mm wide. The metals overlapped several tenths of a millimeter at the hot junction. The cold junctions were situated on the mica so that they would not respond to the periodic radiations. Figs. 2 and 3 show the completed couples drawn to scale; the cellulose section at the center is sufficiently large to reduce heat conduction loss along it to a negligible quantity.

To prevent breakage in handling it was found necessary to provide mounts for the mica frames supporting the couples. After being sputtered the couples were cemented to the mounts at the edges of the mica frame. Adjustable slits were screwed to the mount so that the radiation beam

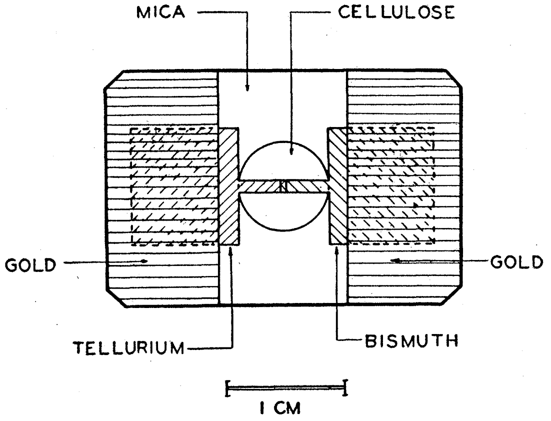

FIG. 2. Single junction thermocouple, to scale.

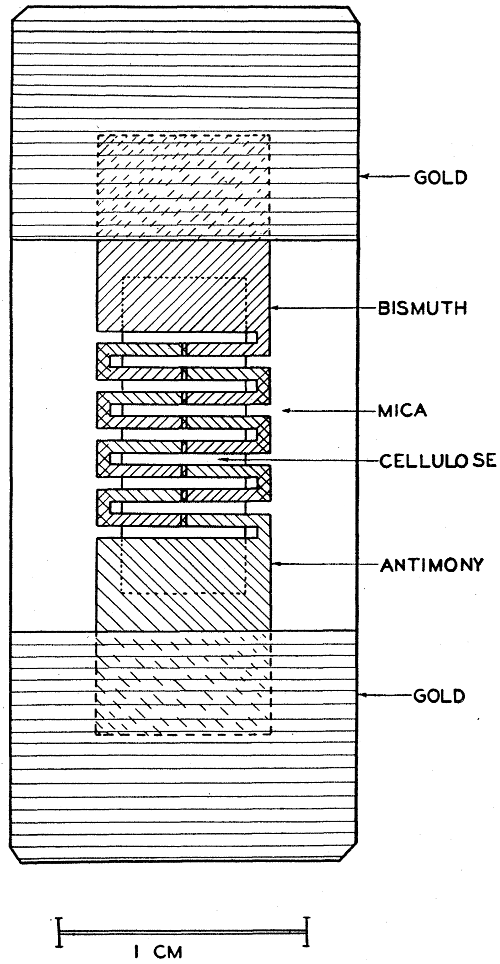

FIG. 3. Eight-junction thermocouple, to scale.

TABLE I.

Metals	Junction	Resistance (ohms)	Volts d.c.	Volts 30∽	Volts 90∽	Volts 270∽	Volts 810∽	Attenuation factor 30∽	Attenuation factor 90∽	Attenuation factor 270∽	Attenuation factor 810∽
Single Junction Couples											
Bi-Sb	Black	25000	3.59×10^{-4}	9.6×10^{-6}	4.62×10^{-6}	1.43×10^{-6}	0.5×10^{-6}	5.9×10^{-2}	2.8×10^{-2}	8.8×10^{-3}	3.1×10^{-3}
Bi-Sb	Bright	1000	1.95×10^{-4}	6.9×10^{-6}	2.76×10^{-6}	9.2×10^{-7}	3.18×10^{-7}	7.8×10^{-2}	3.1×10^{-2}	1.0×10^{-2}	3.6×10^{-3}
Bi-Te	Black	150000	2.5×10^{-3}	3.36×10^{-5}	6.4×10^{-6}	8.05×10^{-7}	7.5×10^{-8}	2.5×10^{-2}	4.8×10^{-3}	6.0×10^{-4}	5.6×10^{-5}
Multijunction Couples											
Bi-Sb	Black	243000	1.3×10^{-3}	2.95×10^{-5}	1.13×10^{-5}	3.8×10^{-6}	8.31×10^{-7}	5.0×10^{-2}	1.9×10^{-2}	6.4×10^{-3}	1.4×10^{-3}
Bi-Sb	Black	6000	1.21×10^{-3}	2.17×10^{-5}	1.55×10^{-5}	5.47×10^{-6}	1.72×10^{-6}	4.0×10^{-2}	2.8×10^{-2}	1×10^{-2}	3.1×10^{-3}
Bi-Sb	Bright	55800	4.7×10^{-4}	1×10^{-5}	1.5×10^{-6}	1.62×10^{-7}	1.56×10^{-8}	4.7×10^{-2}	7.0×10^{-3}	7.6×10^{-4}	7.2×10^{-5}

was defined close to the hot junctions. The mounted couples were screwed into a plate in an apparatus for blackening the junctions, blackened, and then screwed into a similar plate in a housing (see below) for testing. For transportation, the couples were screwed into a piece of flat brass contained in a small box. No couples were broken during transportation (automobile, 140 miles).

The thermocouple housing was evacuated by means of a butyl phthalate pump in series with an oil pump to about 0.1 micron as recorded by a calibrated hot wire gauge. A trap surrounded by "dry ice" was between the "butyl pump" and the housing and gauge.

LIGHT SOURCE

The light from a 108 watt ribbon filament, operated at about 75 watts was focussed on a slit about 0.8 mm wide. A "light chopper" operated by a synchronous motor was directly behind the slit. The "light choppers" were aluminum disks 30 cm in diameter having 1, 3, 9 and 27 equally spaced open sectors respectively near their periphery. The frequencies obtained: 30, 90, 270, 810 cycles, respectively, were chosen so as to minimize any 60 cycle in the fundamental or harmonics which would disturb the amplification due to 60 cycle "pick-up." The light passing through the choppers was focussed on the couples, the spot of light being considerably larger than the area of the couples. The separation of the slit jaws directly in front of the hot junctions was varied; for most measurements the slit width was 1.5 mm.

The a.c. voltage produced was measured with a Rawson milliammeter on the output side of a sensitive tuned amplifier having a range from 30 cycles to 5000 cycles. The housing with its couple was contained in a copper box together with the first stage of amplification. The d.c. voltages were measured with a sensitive galvanometer together with a potentiometer arrangement.

The results are presented in Table I.

DISCUSSION OF RESULTS

Fig. 4 represents the results to be expected from Eq. (5). The upper curve is one obtained for metals without backing material. The (B) curve is one obtained using Eq. (5) and for couples as used here.

cellulose thickness	3×10^{-5} cm
metal thickness	1×10^{-5} cm (single junction); 5×10^{-6} cm (multijunction)
$c\rho$ (cellulose)	0.56
$c\rho$ (metal)	0.3
b	1.0
E	1.5×10^{-4} (Assumes very little reradiation from back of couple.)

The crosses indicate the experimental points.

The experimental values seem to agree with the theoretical equation in the case of four of the six couples: for higher frequencies the attenuation

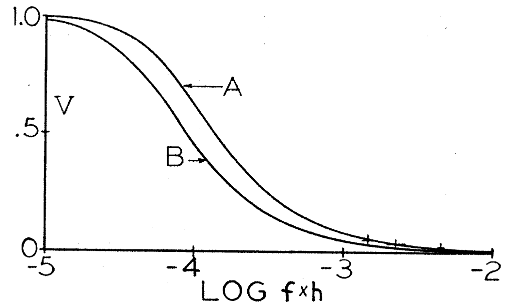

FIG. 4. Attenuation factor as a function of the frequency-thickness product. *A*, for metals without backing; *B*, for metals with cellulose backing (3×10^{-5} cm thick).

factor decreases linearly with frequency while at lower frequencies the decrease of V with increasing frequency is less than linear. In the case of two of the couples the attenuation factor varies as the square of the frequency. This discrepancy may be due to the particular orientation of slit and contacts in these two cases—a capacity effect being introduced which was more pronounced at the high frequencies.

The results show that the sensitivity is independent of the resistance up to several hundred thousand ohms, at least. The extremely high resistance encountered in several cases was due to breaks in the metal films and this was a common fault, were care not exercised to have a smooth edge of mica at the center hole supporting the cellulose.

The results also show that for couples of the thickness used it is necessary to operate at frequencies as low as 1 to 3 cycles to obtain the full sensitivity available. The cellulose is the chief contributing cause to the relatively high inertia and it can be minimized. The cellulose was made thicker than would ordinarily be used due to the transportation requirements. Unfortunately, an amplifier capable of responding to the low frequencies was not available; one is now under construction.

The voltage output of the multijunction couples is only three times as much as that of the single junction couples, whereas eight times the voltage should be obtained. Recently, Johnson and Harris[6] have found that sputtered couples of Bi-Sb showed a thermal e.m.f. of 75 microvolts per degree for thicknesses greater than 10^{-4} cm. Below this thickness the thermal e.m.f. decreased nearly linearly with decreasing thickness. Since the multijunction couples were about one-half the thickness of the single junction couples, the apparent discrepancy is explained. If we assume a value of 50 microvolts per degree for the single-junction Bi-Sb couples we must assign a value of 400 microvolts per degree for Bi-Te couples. Massive couples of Te-Bi have a thermal e.m.f. of 600 microvolts per degree for the β-form of tellurium and 200 microvolts per degree for the α-form.[7] It seems that most of the tellurium is in the β-form, the modification obtained by cooling the nearly molten metal quickly.

To test Eq. (7) the single junction couples were illuminated with the light from a 25-watt lamp without the use of lenses. The lamp had been calibrated against a standard with a commercial thermopile and galvanometer.

	Volts	Calories/ cm^2/ second
Bi-Te	7.6×10^{-5}	44×10^{-6}
Bi-Sb	8.8×10^{-6}	44×10^{-6}

Substituting the value of A in Eq. (7), $\Theta = bA/E$, and assuming $b=1$, $\Theta=0.29°$ temperature elevation of hot junction. The Bi-Sb couple ($50\mu V/°$) gives a calculated temperature elevation of 0.176°C; the Bi-Te couple of 0.19°C. It does not seem reasonable to ascribe the discrepancy to a heat loss. The fact that no increase in voltage was obtained when the slit jaws in front of the couples were opened from 1.5 to 3 mm confirms this belief. If one assumes, however, that the area for reradiation is two times as much as the effective receiving area the agreement is satisfactory.

SENSITIVITY

A single junction Te-Bi couple operated at 1 cycle per second will respond to an energy density of about 2×10^{-10} calorie/cm^2/sec. (1.5×10^{-9} volt), or if all the energy is concentrated on the couple, of about 2×10^{-12} calorie/sec. When it is considered that the most sensitive wire couples used in conjunction with a high sensitivity galvanometer under ideal conditions respond to about 1×10^{-8} calorie/cm^2/sec. (1×10^{-8} volt) (scale 3 to 5 meters distant and no allowance for drift)[8] the use of thermocouples as described here seems highly promising for measurements of small intensities. The resonance radiometer[2, 3] and periodic radiometer[4] appear to have an even higher sensitivity; however, the requisites for their operation are considerably more exacting.

The multijunction couples described here offer a means of attaining still greater sensitivity. It would not be difficult to multiply the number

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[8] E. D. McAlister, Smithsonian Miscellaneous Collections **87**, No. 17 (1933).

of junctions even above the number used here and to make them more concentrated. Due to the poor heat conductivity along the surface of blackening only the region in the immediate vicinity of the junctions is active.

The sensitivity of such couples may be still further increased by minimizing the reradiation loss. This may be done by a change in design and especially by operation at low temperature.[9, 10, 11]

ACKNOWLEDGMENT

It is a pleasure to acknowledge the assistance given during this investigation by Mr. E. A. Johnson of the Department of Electrical Engineering and to Mr. Carl Neitzert of that department for his assistance with the measurements.

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