

References and Notes

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CURRENT PROBLEMS IN RESEARCH

Thermoelectricity at Very Low Temperatures

Kelvin's discovery may be the key today
to electron transport problems.

D. K. C. MacDonald

The experimental discovery of thermoelectricity dates from early in the last century. The Seebeck potential, or thermoelectric force, is the voltage produced in a circuit of two dissimilar elements when one junction is heated relative to the other (Fig. 1). The Peltier heat is the component of heat evolved

or absorbed at the junctions per unit time when unit current flows in a circuit. It was William Thomson (Lord Kelvin), however, who in 1854 essayed a thermodynamic analysis of these effects (1), recognizing that they must be interrelated, and who was then led to make the remarkable prediction that a third

effect (known today as the Thomson heat) must exist in a single conductor when a current flows through it and the conductor is in a temperature gradient. The Thomson heat is reversible in the sense that a component of heat is evolved or absorbed, depending on the relative direction of the electric current and temperature gradient. After his theoretical prediction, Thomson then went on to show the existence of this effect by a painstaking series of experiments. The definition of the Thomson coefficient μ is given by the following equation:

$$\dot{Q} = -\mu J_x \frac{dT}{dx} + \frac{J_x^2}{\sigma} \quad (1)$$

where \dot{Q} is the heat evolved per unit volume per unit time in a conductor; J_x is the current density; dT/dx is the temperature gradient; and σ is the electrical conductivity.

The second term in Eq. 1 corresponds to the irreversible Joule heating, and it

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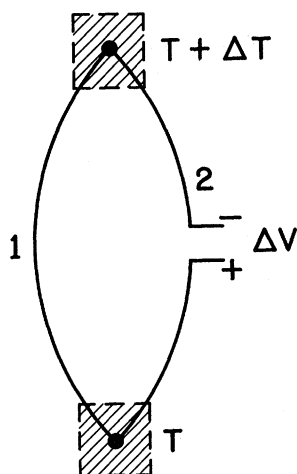


Fig. 1. Schematic thermoelectric circuit. The net thermoelectric power of this circuit is given by $dV/dT = S_1 - S_2$, where S_1 and S_2 are the absolute thermoelectric powers of conductors No. 1 and 2. If $S_1 > S_2$, then the polarity of ΔV is as shown in the figure.

is assumed that the Thomson heat and the Joule heat are independent of one another. It is clear that the Thomson heat (μ), like the electrical conductivity (σ) [or thermal conductivity (κ)], is a bulk property of a conductor, and so indeed are the Peltier heat and absolute thermoelectric power; unfortunately, confusion still sometimes occurs today when it is implied that the Peltier heat and thermoelectric power are "contact" phenomena because they have to be *measured* in a circuit composed of two different conductors.

The thermoelectric power is of great practical significance, since of course the use of thermocouples as thermometers depends on the very existence of this effect. On the other hand, we might perhaps say that thermoelectricity, until rather recently, has been to some extent the "Cinderella" of conduction phenomena from the point of view of yielding fundamental information about electron-transport behavior. In particular, in the last 30 years, a great deal of work has been done on the investigation of electrical conductivity at low temperatures (by which, roughly, we mean liquid-hydrogen temperatures—say, 20°K and below), and since World War II this work has grown to be very intensive at temperatures down to those of liquid helium (say, 1.5° to 4°K). Also, during this period *thermal* conductivity has become a rapidly growing field of interest. However, both of these parameters essentially measure thermodynamically irreversible effects. As a consequence of this, at sufficiently low temperatures (typically, below 4°K in a pure metal)

the irreversible scattering of electrons by chemical impurities and physical defects in the lattice tends to dominate the situation, and it quickly becomes more and more difficult to extract information about the scattering of electrons by thermally activated processes. Of recent years, this has been seen to be particularly unfortunate because the details of thermal scattering of electrons are of great interest to the fundamental theory at these low temperatures. In addition, however, when more than one type of scattering process is involved, each, acting independently, would have to contribute a positive resistivity, since we are dealing with irreversible processes; put in other words, scattering due to physical defects, chemical impurities, excited lattice waves ("phonons") of high energy, low-energy phonons, or

other causes must in each case be essentially positive, and it is not always easy to distinguish experimentally between these types with certainty. Lastly, the resistivities being essentially irreversible effects, little can be said about their expected behavior from a thermodynamic point of view (except, as we have said, that electrical and thermal resistivity must indeed be positive.)

Not so, however, for the thermoelectric effects. It is becoming clear that the puzzling changes of sign observed in some elements, as we vary the temperature, may well be a direct consequence of the changing predominance of different scattering components. It is indeed still true that the thermoelectric power (or Thomson heat) is sensitively dependent, in an almost alarming way in some cases, on temperature, on the type of im-

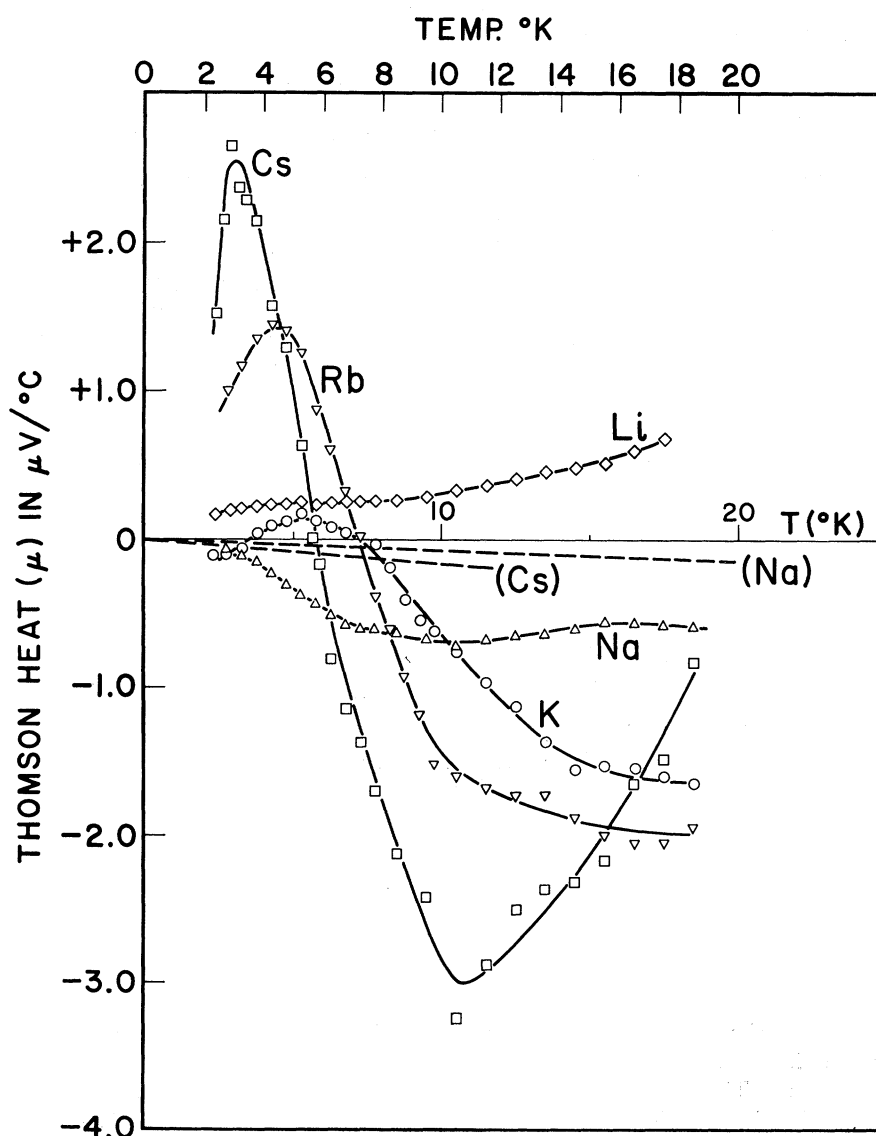


Fig. 2. Thomson heat of alkali metals at low temperatures. The two dashed lines show the detailed predictions of theory at low temperatures, ignoring phonon-drag; A. H. Wilson (13) gives $\mu = \pi^2 kT/3eT_0$ (compare Eq. 4 in text). [After MacDonald, Pearson, and Templeton (14)]

purity present, and so on, but this very sensitivity appears now to offer thermoelectricity as a rather valuable parameter for testing theories of conduction and for obtaining fresh information in this field. At the same time, it is a consequence of the *reversibility* of the thermoelectric phenomena that the behavior is not necessarily limited at low temperatures by the irreversible scattering due to impurities and the like, in the way that the electric and thermal resistivities are. Finally, we can, in some cases, usefully appeal to the laws of thermodynamics for some guidance as to what behavior we must expect from the thermoelectric coefficients.

Let us turn, then, to consider the significance of recent experimental work on thermoelectricity at low temperatures—say, 20°K and below.

Thomson Heat and Thermoelectric Power

William Thomson himself first suggested that the Thomson heat might be regarded essentially as the specific heat of electricity. This is not, however, to be identified immediately with the familiar specific heat at constant pressure or constant volume; it is a rather special specific heat, measured in a temperature gradient under conditions of no net electric current. Now in order to evaluate this properly we must appeal to some detailed theory of electron transport, but it would certainly be reasonable to assume that the Thomson heat should be fairly close in value to the more conventional equilibrium electronic specific heat. Thus, we should expect

$$\mu \sim C \quad (2a)$$

or

$$\mu \sim c_{el}/e \quad (2b)$$

where C is the specific heat per unit (free) charge transported [assumed positive from the defining Eq. 1]; c_{el} is the specific heat per electron; and e is the charge of an electron in sign and magnitude. The relation shown in Eq. 2b follows from Eq. 2a of course if we assume conduction by free electrons. Now, the modern theory of metals gives

$$c_{el} = \frac{\pi^2 k}{2} \left(\frac{T}{T_0} \right) \quad (3)$$

where T_0 is the electron-degeneracy temperature (around 50,000°K for a typical metal), and thus we might expect

$$\mu \sim \frac{k}{e} \left(\frac{T}{T_0} \right) \quad (4)$$

Now k/e is about 10^{-4} volt/°C and we should therefore expect a Thomson heat falling linearly with temperature and diminishing from about 10^{-7} volt/°C in the region below about 20°K. Actually, one usually measures the absolute thermoelectric power (S) of a metal, but since we have the Thomson relation

$$\mu = \frac{TdS}{dT} \quad (5)$$

μ and S should be identical if Eq. 4 is valid. Actually, few metals seem to be “well-behaved”—at first glance, anyway—in this temperature region. Perhaps most striking are the Thomson heats of the alkali metals between 2°K and 20°K, shown in Fig. 2. The variation of thermoelectric behavior with temperature in most of this group of metals appears rather disconcerting, and the magnitudes of the Thomson heat and thermoelectric power, particularly in the heavier alkali metals, are often greatly in excess of the predicted values as indicated by the relation of Eq. 4 above.

“Phonon-drag Effect”

The tentative theoretical explanation today of these results rests on the so-called “phonon-drag” effect [or Gurevich effect

(2)]. When we think of the Thomson heat as a specific heat of electricity, we are confining our attention essentially to the thermal entropy of the conduction electrons themselves. That is to say, under a temperature gradient, we may expect that the “hot” electrons (since they have higher velocities, for one thing) will tend to drift more rapidly towards the “cold” end than the “cold” electrons will drift in the opposite direction, and that this will give rise to a thermoelectric current. However, if we also recall that the electrons collide with the excited lattice waves (or “phonons”), it is not difficult to see that we may be able, in this way, to “tap” the entropy gradient of the lattice waves as well. Perhaps the simplest way to think of this process is as follows: When a temperature gradient is present, the phonons will themselves carry a thermal current (the lattice heat flow) and if phonon-electron collisions are important—as indeed they are in a pure metal at low temperatures—then the phonons may tend to “drag” the conduction electrons with them from hot to cold. Thus arises the “phonon drag” effect, producing an additional component of thermoelectric power.

A simple momentum argument (see, for example, 3) leads us to expect:

$$S \sim \alpha C_{latt}/N_{el} \quad (6)$$

where C_{latt} is the lattice specific heat per unit volume; N_{el} is the density of

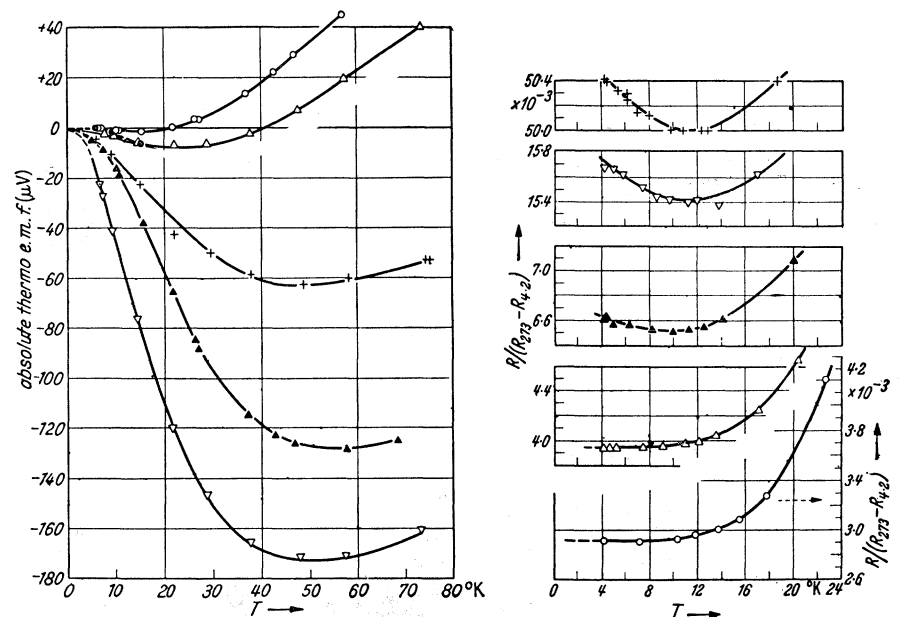


Fig. 3. (Left) Absolute thermoelectric force of copper and alloys. (Open circles) Pure Cu; (open triangles) pure Cu + 0.0009 atomic percent Sn; (solid triangles) pure Cu + 0.0026 atomic percent Sn; (open triangles, inverted) pure Cu + 0.0054 atomic percent Sn; (plus signs) pure Cu + 0.026 atomic percent Sn. (Right) Relative electrical resistance of these copper and copper-tin alloys. [After MacDonald and Pearson (6)]

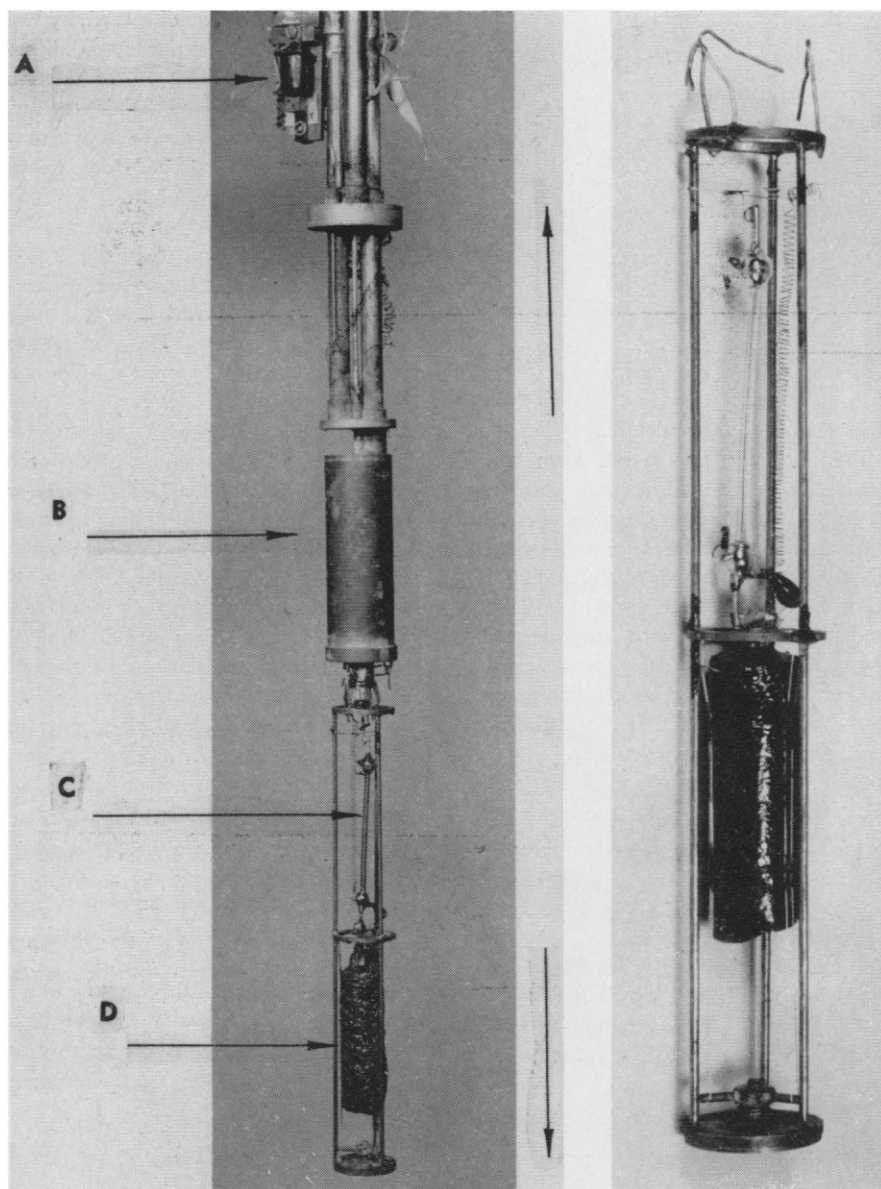


Fig. 4. (Left) Cryostat (with vacuum cans removed) for measurements of thermoelectric force below 1°K . *A*, Superconducting reversing switch; *B*, inner liquid helium bath at about 1°K ; *C*, Alkali metal specimen; *D*, "pill" of paramagnetic salt. Over-all dimension, about 14 in. between arrows. (Right) Close-up of experimental "cage," showing "pill" of paramagnetic salt and alkali specimen mounted for experiment.

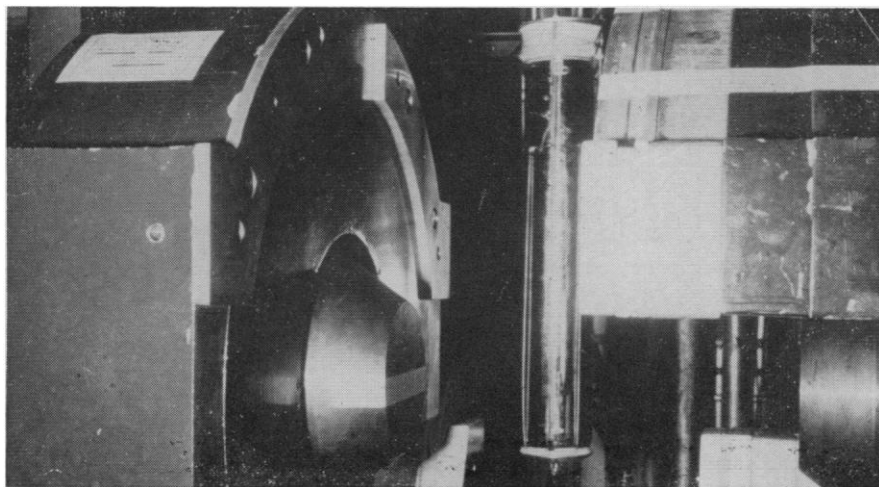


Fig. 5. Cryostat (inside Dewar vessel) in large electromagnet prior to establishment of magnetization cycle for achieving temperatures below 1°K .

free electrons; and α is a parameter lying between 0 and 1; if phonon-electron collisions can be neglected, $\alpha = 0$, while if phonon-electron collisions are dominant, $\alpha \rightarrow 1$.

As the temperature increases, the lattice specific heat grows rapidly [with $(T/\Theta)^3$], and so long as α remains significant, Eq. 6 can give rise to quite a large amount of thermoelectric power. More detailed theories, such as J. M. Ziman and M. Bailyn are now developing, suggest that the behavior of the "phonon-drag" effect could be quite complicated, depending on the precise mechanisms of phonon-electron scattering. Now scattering may take place by direct interchange of momentum between a phonon and an electron (the so-called "normal" process), or the exchange may also involve momentum corresponding to one of the reciprocal lattice vectors. It seems that this latter so-called "Umklapp process" (4) may yield a component of thermoelectric power opposite in sign to a component due to "normal" processes; furthermore, the relative proportion of these two scattering mechanisms may depend quite sensitively on the temperature. It then appears quite possible that a satisfactory explanation for the behavior of thermoelectric power down to, say, liquid helium temperatures may be provided in this way; indeed Ziman has suggested that this may well be one of the most sensitive ways of estimating the shape of the electron Fermi surface in relation to the Brillouin zones, since this is a vital factor in determining the relative frequency of the Umklapp processes.

Anomalous Minimum of Electrical Resistance

At the same time, we should not forget many other peculiar features found in thermoelectric power in the liquid helium and hydrogen temperature region. It has been known for some time (see, for example, 5, 6) that in metals such as copper, very small amounts of impurity (of the order of 0.1 atomic percent) can have a quite violent effect on the thermoelectric power in this region, and it appears rather certain that this behavior is related quite closely to the appearance of an anomalous minimum in the electric resistance of such metals at low temperatures. This is illustrated in Fig. 3. The anomalous minimum of electrical resistance has excited growing interest since it was first discov-

ered in experiments on gold at Leiden (7), but so far no adequate theoretical explanation has been found for this behavior. One is sometimes tempted to suspect that the fundamental explanation, when found, might perhaps be comparable in significance for electron transport theory to the explanation of superconductivity—although the phenomenon is of course less dramatic.

Measurements Below 1°K

Since experiments have proved so fruitful down to liquid-helium temperatures, one is naturally led to extend the work to even lower temperatures. Some results below 1°K have now been published (8, 9); apparatus for use in experiments at these temperatures is shown in Figs. 4 and 5. Since the “phonon-drag” effect should diminish as T^3 (compare Eq. 6) while the purely electronic component should diminish only with T (see Eq. 4), we should certainly expect that at sufficiently low temperatures Eq. 4 would become essentially valid—and, in fact, temperatures below 1°K, as a general rule, ought to be sufficient to achieve this. We have now carried out initial experiments on the alkali metals; on copper, silver, and gold; and on one or two other metals, such as platinum, nickel, and iron. In the alkalis we find that the thermoelectric power has indeed now fallen to the order of magnitude predicted by Eq. 4—that is, to around 10^{-8} to 10^{-9} volt/°C in the region below 1°K. On the other hand, the temperature-dependence is not always linear, as we might expect it to be; indeed, in the case of potassium (compare the two parts of Fig. 6) the temperature-dependence appears almost quadratic, and, so far, we can offer no theoretical explanation for this behavior. Much remains to be done here, even among the alkali metals; in particular the dependence of thermoelectric power on impurity requires careful investigation.

Perhaps the most striking results below 1°K are those for copper, silver, gold, and platinum—particularly those obtained on gold. Figure 7 shows experimental data on two samples of gold—one specimen very much purer than the other—and Fig. 8 shows some experimental results on silver. The most remarkable feature is the high order of magnitude of the thermoelectric power in gold, even at 0.5°K. At this temperature the “phonon-drag” effect should be

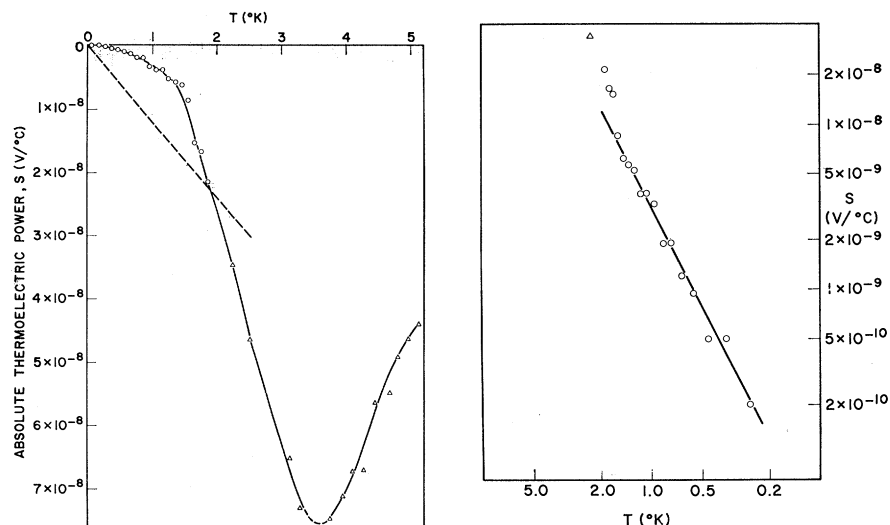


Fig. 6. (Left) The dashed line shows the predicted behavior at very low temperatures (irrespective of phonon-drag); Wilson (13) gives $S = \pi^2 kT / 3eT_0$. (Right) Data replotted on logarithmic scales; line shows T^2 dependence. [After MacDonald, Pearson, and Templeton (8)]

quite negligible, and for the ordinary electronic component we should expect a thermoelectric power of about -4×10^{-9} volt/°C; actually, with the purer gold specimen, a figure about 1000 times greater (about $-4 \mu\text{v}/^\circ\text{C}$) is observed! Again, no theoretical explanation for this remarkable behavior is yet forthcoming; naturally we are most interested to see whether any other metals or group of metals will show thermoelectric powers of this (or perhaps an even greater) order of magnitude.

The very high thermoelectric power

found in gold at these low temperatures makes one consider another question. A thermoelectric power of about 10^{-8} volt/°C, which we might generally expect at 1°K, corresponds to an entropy of about 2×10^{-4} cal/°C per mole of electrons transported (that is, 1 faraday—about 10^5 coul of charge transported). On the other hand, a thermoelectric power as large as $5 \mu\text{v}/^\circ\text{C}$ corresponds to about 0.1 cal/°C Faraday. This suggests at least the possibility of thermoelectric cooling even at these very low temperatures.

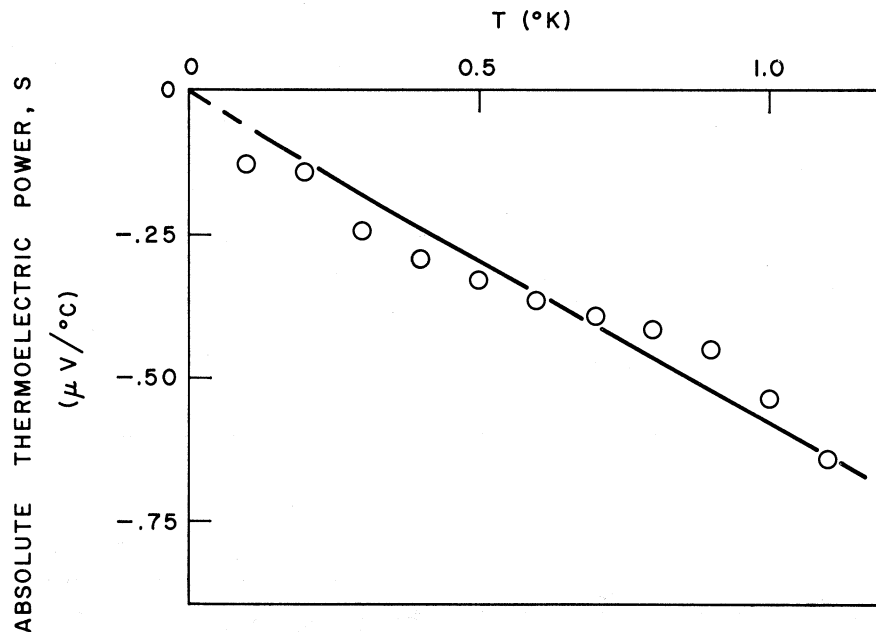


Fig. 7. Absolute thermoelectric power of silver at very low temperatures. [From data of MacDonald, Pearson, and Templeton]

Ideal Limit of Refrigeration

A detailed analysis (see 10) leads to the conclusion that the ideal limit of refrigeration is given by

$$\Delta T/T_0 = S_0^2/2L_0 \quad (7)$$

where ΔT is the reduction of temperature obtained; T_0 is the low temperature (the cold end of the couple); S_0 is the thermoelectric power at the cold end; and L_0 is the Lorenz number ($L = \kappa/\sigma T$) at the low temperature.

For a metal whose conductivity is limited by impurity or defect scattering (as is generally the case at low temperatures)

$$L_0 = \frac{\pi^2}{3} \left(\frac{k}{e} \right)^2 \approx 2.5 \times 10^{-8} \text{ (volt/deg)}^2 \quad (8)$$

If, now, $S_0 \approx 10^{-8}$ volt/°C, then evidently, from Eq. 7, the outlook is quite hopeless for thermoelectric cooling! In the case of gold, setting $S_0 \approx -5 \times 10^{-6}$ volt/°C, we would expect $\Delta T/T_0 \approx 5 \times 10^{-4}$ at temperatures around 1°K—an amount which of course is still far too small to be practically useful. However, at our present state of knowledge we cannot be sure that a metal or alloy is not to be found with even higher thermoelectric power at low temperatures, and if one could be found for which S_0 was perhaps 10^{-4} volt/°C, the situation would become very promising. This naturally provides a further incentive

for experimental work on thermoelectricity at low temperatures.

There is another hopeful aspect; the data of Fig. 8 show that the largest thermoelectric power was obtained in the purer sample of gold. Now, if a metal could be obtained which was sufficiently pure so that the conductivity was not limited, at the low temperatures required, by impurity or defect scattering but was limited rather by thermal scattering, then the Lorenz number (L) could in principle be very much lower than the limiting value quoted above. This is because, if electrons are scattered by thermal vibrations at low temperatures, the electrical conductivity (σ) increases much more rapidly (as T^{-5}) than does the thermal conductivity (κ) (which increases as T^{-2}). Thus it is at least possible that a metal might be obtained which would be sufficiently pure to have both a high enough value of S and a low enough value of L to be of use for low-temperature refrigeration.

If thermoelectric refrigeration could be made a practical possibility at low temperatures (say 1°K), it would be of considerable benefit to experimental research. At present the only feasible method is to use the adiabatic demagnetization of a suitable paramagnetic salt, as first envisaged by Debye and Giauque, in order to achieve temperatures below 1°K. A number of such salts are available—such as iron ammonium alum (which is rather popular), with

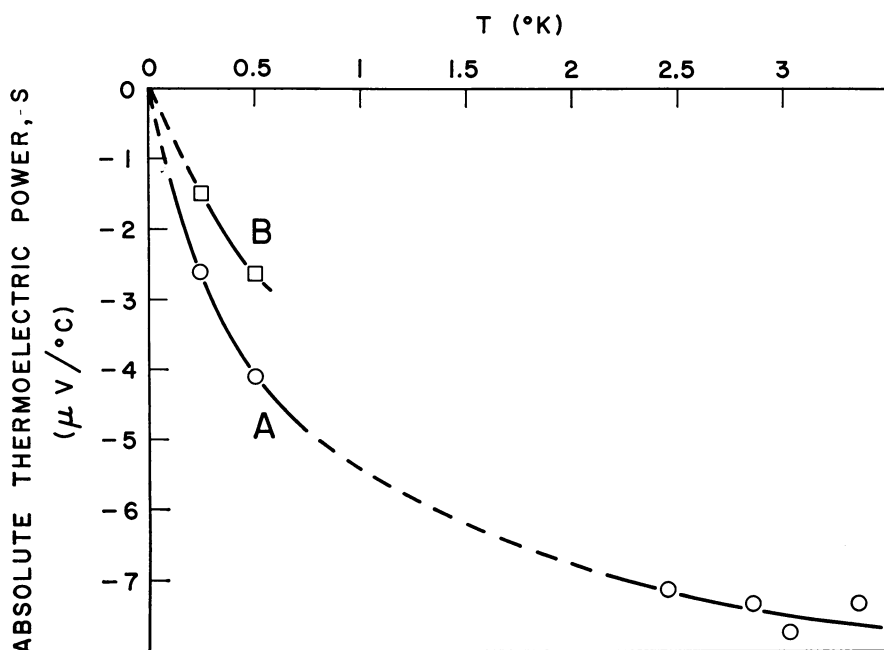


Fig. 8. Absolute thermoelectric power of two specimens of gold at very low temperatures. Sample A ("specpure" quality): $R_{4.2^\circ\text{K}}/R_{294^\circ\text{K}} \approx 3.19 \times 10^{-3}$. Sample B (lower purity, unannealed): $R_{4.2^\circ\text{K}}/R_{294^\circ\text{K}} \approx 6.39 \times 10^{-2}$. [From data of MacDonald, Pearson, and Templeton]

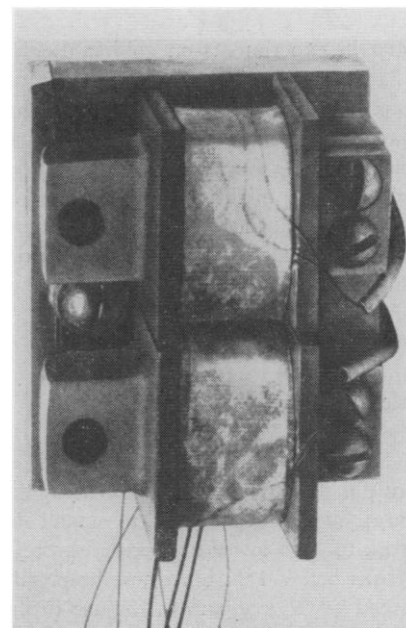


Fig. 9. Superconducting reversing switch designed by Templeton for operation in liquid helium. (About $1\frac{1}{2}$ by $1\frac{1}{4}$ in.; height, about $\frac{3}{4}$ in.)

which one can conveniently work down to about 0.05°K, and, say, cerium magnesium nitrate, with which temperatures of a few millidegrees Kelvin can be reached. Of course such salts are used as refrigerating agents in this work, but there are certain serious difficulties that are met with. As the temperature is reduced, the thermal conductivity of the salts themselves becomes rather poor, and the temperature distribution may thus become rather inhomogeneous through the salt "pill." It is also not easy to achieve satisfactory thermal contact with the salt itself for the purpose of cooling some other material; my co-workers and I usually use the method of recrystallizing the salt directly from a saturated solution onto a silver mesh, but we are by no means certain of the adequacy of thermal contact below, say, 0.1°K.

Another problem of long standing is the question of determining the absolute temperature below 1°K. An approximate temperature is obtained by assuming that the magnetic susceptibility of the salt (χ) obeys Curie's law ($\chi \propto 1/T$). This is in fact an excellent approximation in the liquid-helium region; hence a calibration of magnetic susceptibility against absolute temperature can readily be made in this temperature range. This calibration is then extrapolated to provide a so-called "Curie temperature" down to the low temperatures achieved when the salt is adiabatically demagnetized. However, deviations from Curie's

law must (and indeed do!) occur at sufficiently low temperatures, and thus for accurate work it is necessary to make an absolute calibration in the very low temperature region. This, in principle, can be done by various methods involving the second law of thermodynamics, by taking the salt through a reversible cycle. However, a difficulty arises in the fact that the calibration found does not always agree from laboratory to laboratory, or indeed necessarily from one sample of a salt to another, since the behavior may depend on the previous history of the specimen. One solution is to use as a thermometer an auxiliary sample of a paramagnetic salt (such as cerium magnesium nitrate) which is known to obey Curie's law down to rather low temperatures, but of course this all adds to the experimental difficulties involved. In principle, a very attractive solution would be one in which use was made of the electrical Brownian movement ("thermal noise") in a re-

sistor for thermometry, but there are many difficult technical problems as yet unsolved, not the least of which is that of measuring the very small noise voltage with some accuracy.

Another challenging aspect of the measurement of thermoelectric power at very low temperatures is the problem of measuring the small thermoelectric voltages involved in many cases. The superconducting reversing switch (see Fig. 9) and superconducting modulator developed in these laboratories by Templeton (11) have proved invaluable, and we feel that the limit of sensitivity has by no means yet been reached (12). All in all, it appears that the study of thermoelectricity at very low temperatures is full of promise, and "Cinderella" may yet turn out to be the "belle of the ball."

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News of Science

White House Reports on Scientific Aspects of Radiation Belts Created by Argus Experiments

Three nuclear test explosions, in a series called Project Argus, were set off 300 miles above the earth on 27 and 30 August and 6 September 1958 by a Navy task force in the South Atlantic Ocean. These tests, the first such explosions in outer space, are reported to have had a force measured in kilotons.

Argus was first announced by the New York Times on 19 March. The announcement was promptly confirmed by the Department of Defense, but Deputy Secretary of Defense Donald Quarles, speaking at a news conference, was reluctant to go into any details and expressed regret that the project had been made public. On 25 March the White House released a report on the Argus experiments prepared under the direction of the President's Science Advisory Committee and the International Geophysical Year Committee of the National Academy of Sciences. The text of the report follows.

Committee's Report

This report discusses the scientific as distinct from the military results and implication of the Argus experiments. Because of the fact that many of the experiments performed in connection with these atomic bursts involved both the election trapping phenomenon and classified military effects phenomena, it was considered advisable to withhold all results classified until a proper sorting of the information could be accomplished. Since reports on relevant military aspects have only become available within the last two weeks, it has not heretofore been possible to release any of this information.

The scientific aspects of these experiments, involving three high-altitude small atomic bursts over the South Atlantic in August–September 1958 are regarded by many participants as one of the major achievements of the International Geo-

physical Year. The execution of these experiments engaged the coordinated resources of large segments of the scientific talent of the nation, and it was apparent that the effects of the experiment, if successful, would be recorded by instruments of the far-flung international network of the IGY. The compilation of the observational and interpretative contributions by the many participants will doubtless stand as a durable milestone in the development of man's knowledge of the great natural phenomena of the earth's atmosphere which have engaged his study for many centuries.

Christofilos Proposal

The underlying idea for the Argus experiments was due to Nicholas C. Christofilos, physicist of the Lawrence Radiation Laboratories of the University of California. In October 1957 he called attention to the fascinating physical effects which might be expected to follow an atomic burst in the near-vacuum of outer space, high above the earth and its dense atmosphere. Of the various effects contemplated, the most interesting one promised to be the temporary trapping of high-energy electrons at high altitudes in the magnetic field of the earth. Following the burst there would be thrown off in all directions nuclei of intermediate atomic weight. Most of these nuclei, as is well known, are radioactive and subsequently decay with the release of energetic electrons and gamma rays. Most of the decays occur within a few minutes. The fission fragments themselves are electrically charged and move