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S. R. Harutyunyan
National Academy of Sciences of Armenia

V. H. Vardanyan
National Academy of Sciences of Armenia

A. S. Kuzanyan
National Academy of Sciences of Armenia

V. R. Nikoghosyan
National Academy of Sciences of Armenia

S. Kunii
Tohoku University

See next page for additional authors

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Thermoelectric cooling at cryogenic temperatures

S. R. Harutyunyan, V. H. Vardanyan, A. S. Kuzanyan, and V. R. Nikoghosyan
Physics Research Institute, National Academy of Sciences, Ashtarak, 378410, Armenia

S. Kunii
Department of Physics, Tohoku University, Sendai, 980-8578, Japan

K. S. Wood
Naval Research Laboratory, Washington DC, 20375

A. M. Gulian[a]
Physics Art Frontiers, Naval Research Laboratory, Washington DC, 20375

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Experimental results demonstrating Peltier cooling below 10 K are reported, using crystals of the thermoelectric cerium hexaboride (CeB_6). Direct measurements of the Peltier cooling showed δT up to ~0.2 K in magnitude at $T\sim 4-5$ K. All three kinetic parameters: resistivity (ρ), heat conductivity (k), and Seebeck coefficient (S), characterizing the thermoelectric figure of merit $ZT = S^2T/\rho k$, were measured, giving high-confidence results.
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Recently, it was proposed to use lanthanum (cerium) hexaborides as sensors for low-temperature photon detectors.[1] Another possible application is conversion of electric energy into negative heat fluxes, i.e., thermoelectric refrigeration using the Peltier effect. This short report announces the achievement of Peltier cooling below 10 K using crystals of CeB_6, a Kondo metal well described in the literature.[2–5]

Crystals of CeB_6 were grown by a floating zone method under pressurized high-purity argon gas. Details were similar to those in Ref. 2. Two bars of CeB_6 were cut from two ingots by spark erosion. They have dimensions $9.9\times 0.8\times 0.8\ \text{mm}^3$ (sample No. 1) and $8.1\times 1.46\times 0.93\ \text{mm}^2$ (sample No. 2), with long dimension parallel to the $\langle 110\rangle$ orientation.

We measured independently all parameters required to determine the thermoelectric figure of merit (ZT). Resistivity was measured with a four-probe ac technique. Heat conductivity was measured in a standard steady-state heat flow method. Thermoelectric power was measured by a conventional direct current differential technique using copper leads. As Fig. 1 demonstrates, saturation of the resistivity, the peak Seebeck coefficient, and the behavior of thermal conductivity follow theoretical expectations for Kondo materials.[6]

The setup used for the Peltier cooling measurement is shown in Fig. 2. When the switch is in position 1, direct current through the sample produces heating at the free end. When the switch is in position 2 the free end is cooling. The sizes of current leads and thermocouples were chosen to minimize spurious in- and out-flow of heat, e.g., thermocouple wires were 40 μm diam and lengths 10 cm (for constantan) and 15 cm (for copper).

The other end of the sample, owing to a good thermal contact with the heatbath, remained at temperature T_0. Absolute accuracy is estimated to be about 20%. Consider the end junction area of the cooler: the Peltier effect depends on the sign of the current: $Q_P = STI$. The Joule heat ($Q_J = RI^2$) is released at the junction ends and in the sample volume. We presume that the major contribution to resistance (R) is the junction pad at the free end. In steady state, these processes are compensated by heat transport along the sample, along the thermocouples, along the current leads, and by radiative (blackbody) heat transfer: $Q_P + Q_J = Q$, where $Q = G\,\delta T$ stands for the rate of heat transport due to the corresponding thermal conductance G, representing all channels of heat exchange of the free end with the external world, excluding Peltier and Joule mechanisms. One can show that radiative heat transfer is negligibly small since the Al shield at temperature T_0 protects the whole device. We also omit the Thompson effect, because of the relatively small values of ZT. Experimental values of $\delta T(I<0) \equiv \delta T_{\text{ht}}$ and $\delta T(I>0) \equiv \delta T_{\text{cl}}$ as functions of $|I|$ are given in Fig. 3 for different ambient temperatures. The slope of the "heating" curves decreases with increasing temperatures since $d(\delta T_{\text{ht}})/dI \propto G^{-1}$, and G grows with the temperature. The "cooling" curves $\delta T_{\text{cl}}(I)$ have minima at certain values of the current, which follow from a simple model of the quadratic dependence of Joule heat on current versus the linear dependence of the Peltier effect. Assuming G is temperature independent in a small region δT, one can find $I_{\text{min}} = ST/2R$. At larger values of currents ($I \gg ST/R$) the cooling is not observable: one has a net heating.

Subtracting the experimental curves in Fig. 3, one can isolate the Peltier heat source (Fig. 4), the linear behavior of Q_P/G confirms its nature.

Table I addresses the maximal cooling amplitudes δT^{max} at different ambient temperatures (T_0) obtained at corresponding (optimal) values of the current (I^{opt}). The parasitic heat load grows drastically at higher temperatures when the rate of the heat transfer is large. Since the silver contacts

[a]Author to whom correspondence should be addressed; electronic mail: armen.gulian@nrl.navy.mil

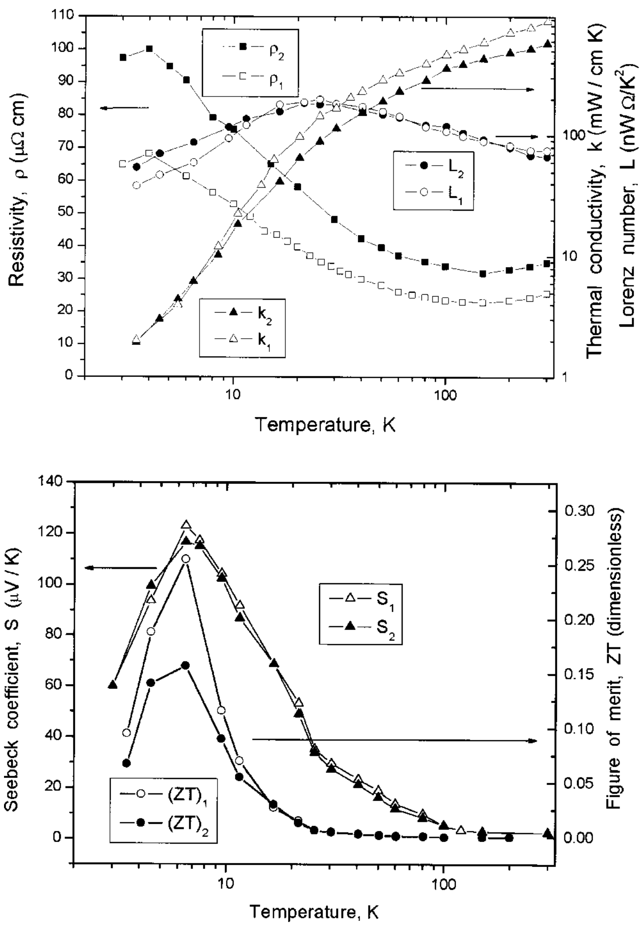

FIG. 1. (Top) Resistivity (ρ), thermal conductivity (k), and Lorenz number ($L \equiv k\rho/T$). (Bottom) Seebeck coefficient (S) and dimensionless figure of merit ($ZT = S^2/L$) of two single crystals of CeB_6.

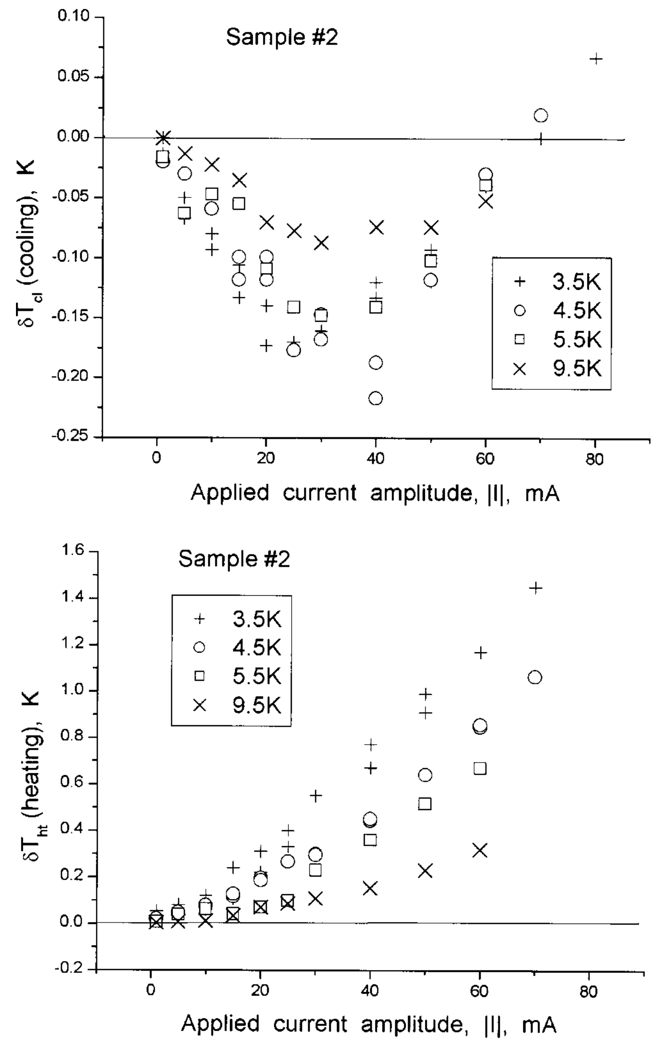

FIG. 3. Cooling and heating of the free end of the crystal. The data were received in the same arrangement where the only difference was the polarity of the dc current.

were silver epoxy these pads create large parasitic heat loads, so the comparison favors the low-resistance indium pads. The greatest value of cooling δT_{cl} is obtained at the temperature $T = 4.5$ K and is equal to $\delta T_{\mathrm{cl}}^{\max} = 0.2$ K at a current $I = 40$ mA. It is important to mention that the optimal performance temperature is not determined by the maximum in the Seebeck coefficient, but rather by the maximum in the ZT value. This experimental result is expected according to theory.

Lorenz numbers measured in this work coincide with the behavior reported in the literature.[3] There is a major discrepancy between different measurements of S on CeB_6 in different publications. Reference 4 reports $S_{\max} \sim 55\ \mu$V/K at $T \sim 9$ K. The same value can be found in Ref. 3. Reference 5 reports $S \sim 265\ \mu$V/K at the same temperatures, hence, the difference is about a factor of 5, unlikely to be an experimental mistake; there is a need to understand this discrepancy. In

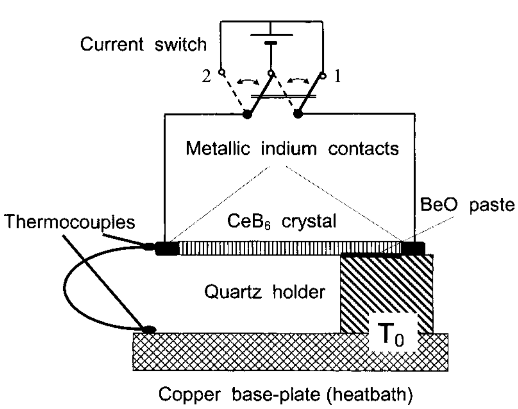

FIG. 2. Schematic of the Peltier experiment.

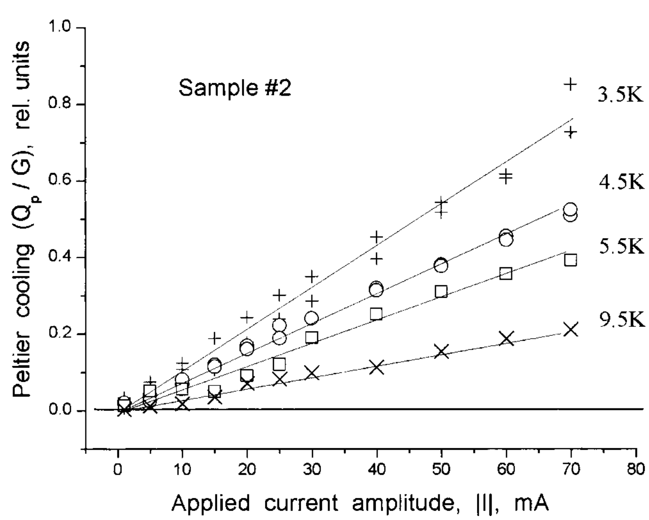

FIG. 4. Justification of the Peltier cooling.

TABLE I. Maximal cooling values.

T_0 (K)	I^{opt} (mA)	δT^{max} (K)	$ZT^2/2$ (K)
	Sample 1, silver contacts		
3.5	12.5	−0.08	0.17
4.5	11	−0.02	0.42
5.5	12	−0.04	0.80
	Sample1,indiumcontacts		
3.5	25	−0.17	0.17
4.5	25	−0.13	0.42
5.5	25	−0.10	0.80
	Sample2,indiumcontacts		
3.5	25	−0.17	0.12
4.5	40	−0.20	0.32
5.5	30	−0.15	0.52
9.5	30	−0.09	0.43

our case, T_{max} is again about 9 K, and $S_{max} \sim 120\ \mu V/K$, intermediate between the referenced data. The Peltier measurements are direct evidence that our crystals are unlikely to have the Seebeck behavior reported in Refs. 3 and 4. Indeed, were $S < 20\ \mu V/K$ at $T \sim 5$ K, we would have $ZT \sim 0.016$ at most, and maximum cooling would not exceed the value $\delta T_{cl}^{max} \equiv ZT^2/2 - 0.03$ K, which is definitely contrary to our observation of $\delta T_{cl}^{exp} \sim 0.2$ K. Therefore, it is likely that there is a pronounced influence of crystalline perfection and impurities onto the Seebeck coefficient. Had the Peltier measurements been performed on the crystal of Ref. 5, a much higher δT_{cl}^{exp} should have been found.

In conclusion, direct measurements of the Peltier effect on CeB_6 crystals have demonstrated efficient thermoelectric cooling at cryogenic temperatures. In view of the neglected parasitic heat fluxes, overall agreement between the experimental δT_{cl} and the theoretical estimate is good. Reducing parasitic heating effects should allow one to obtain a temperature difference $>0.5°$ below liquid helium temperatures in a single refrigeration stage. This can have practical applications in creating efficient solid state cryocoolers starting, say, from 4.2 K liquid helium reservoirs or even higher temperatures.

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