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New Instruments and Measurement Methods

MEASUREMENTS OF INFRALOW TEMPERATURES

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IT is quite easy to measure low temperature with high accuracy by using the vapor tension of liquid He^4 and the 1958 scale[1] and of liquid He^3 with the 1962 scale[2]. These scales, however, cover only temperatures from 0.3 to 5.2°K. There is an international temperature convention relative to the resistance of platinum[3]. It gives the temperature values accurate to 0.01° down to 13.81°K. Recently, germanium resistance thermometers have found wide application[4-7]. The temperature dependence of the germanium thermometer of the "Solitron" company[5] is shown in Fig. 1a. The reproducibility of the graduations of good germanium thermometers is not worse than 0.001° even after two years[7], the sensitivity can be brought to 10^{-5}°K[6], and the lower limit of temperature measurement at $W = 10^{-12}$ W is 10^{-2}°K[7]. The germanium thermometer, however, must be first calibrated in the entire temperature range in which its use is planned. Very stable and convenient in operation are resistance thermometers of superconducting alloys such as phosphor bronze[8] and lead brass[9] (see Fig. 1a). Their temperature reproducibility accuracy is about 2×10^{-5}°K, but they operate in narrow temperature ranges and are very sensitive to the magnetic field and to the measuring current, their resistance increasing by 20% when the current is increased from 1 to 5 mA.

Thermocouples of chromel and gold with small additions of iron can be used to measure temperature differences.

Figure 1a shows the sensitivity of one such thermocouple[10] at zero magnetic field and in a field of 15 kOe. The thermocouples have a fair sensitivity, but since they are made of almost pure gold, much parasitic heat flows through them and thus limits the region of their applicability. At the very lowest temperatures, carbon resistors are used extensively, especially for indicating purposes. Carbon resistors made by the Allen-Bradley Company are used for temperatures above 1°K. Below 1°K, their resistance rapidly exceeds several megohms, and below 0.6°K they cannot be used in practice[11]. Resistors made by the Speer company are widely used. They were calibrated in detail[12] in the range from 4 to 0.02–0.03°K, but different batches of the resistors had different calibrations.

The Institute of Semiconductor Physics of the USSR Academy of Sciences has developed[13] carbon thermometers that are sensitive down to 0.006°K, but their resistance changes after they are heated to room temperature. The temperature dependences of the Speer and of the Semiconductor Physics Institute (SPI) are shown in Fig. 1a. The resistance of carbon thermometers depends little on the magnetic field[14], increasing 20% at H = 150 kG and T = 1.75°K.

The resistance of germanium and carbon thermometers is practically independent of the external pressure[15], $\Delta R/R\Delta p = -2 \times 10^{-4}$ atm^{-1}. It should be noted that owing to poor thermal conductivity and to the temperature jump on the boundary, carbon resistors used at T < 0.1°K should have a very low power dissipation $W < 10^{-12}$ W, and this power should decrease with temperature. At temperatures below 1°K, one of the main methods of determining the temperature, using a scale that differs little from the thermodynamic scale, is to use the Curie law for paramagnetic salts or metal nuclei. If the paramagnetic sample has a low Curie or Neel temperature T_C, then the magnetic moment in the region $T \gg T_C$ is $M = AH/T$, where A is a constant and H is the magnetic field. For cerium-magnesium nitrate (CMN) we can assume, accurate to 3%, that the Curie law is obeyed at T > 0.006°K. The question of satisfaction of the Curie law does not arise for nuclear thermometers (at least down to 10^{-4}°K).

At temperatures such that the field produced by the magnetic moments of the sample becomes comparable with the external field, it is necessary to take into account the difference between the external magnetic field and the effective one. This circumstance, as well as other possible deviations from the Curie law, has led to the introduction of the so-called magnetic tem-

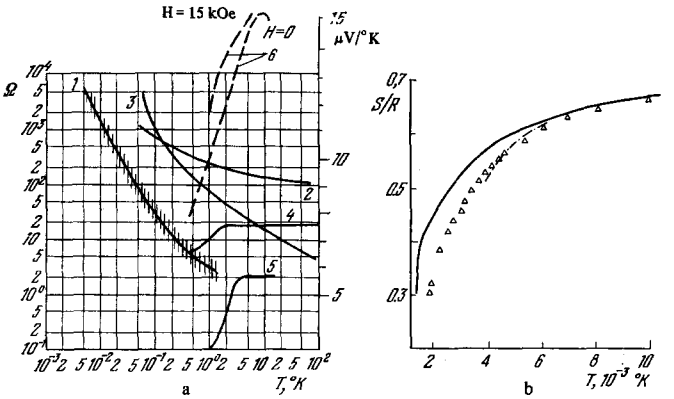

FIG. 1. a) Dependence of the resistance Ω on the temperature of the carbon thermometers made by the ISP (1) and the Speer company (2), of the Solitron germanium thermometer (3), of phosphor bronze thermometers of 30μ diameter (4), and of lead brass thermometers of 50μ diameter (5), and temperature dependence of the sensitivity of a gold (Au + 0.03% Fe)-chromel thermocouple (6). b) Entropy of CMN single crystal vs temperature, obtained from experiments with heating by γ rays (curve) and from the anisotropy of the γ radiation of Cu^{137} (symbols), and calculated by means of the formula $\Delta S/R = \ln 2 - (S/R) = 2.88 \times 10^{-6} T^{-2}$ (dashed curve).

perature $T^* = AH/M$. If one knows T^* and the experimental data on the specific heat of the specimen $d'Q^*/dT^* = C^* = C^*(T^*)$, where $d'Q^*$ is the amount of heat necessary to heat the sample by $d'T^*$, as well as data on the entropy $S^*(T^*)$ obtained by demagnetization from a high temperature T_1^*, where the entropy $S^*(T_1^*)$ is known to low values of T^* under the condition that $S^*(T_1^*) = S^*(T^*)$, then one can determine also the thermodynamic temperature, since

$$T = C\,(\partial S/\partial T)^{-1} = C^*\,(\partial S^*/\partial T^*)^{-1}, \qquad (1)$$

inasmuch as $\partial Q/\partial T = C = (\partial Q^*/\partial T^*)(\partial T^*/\partial T) = C^*\,(\partial T^*/\partial T)$ and $\partial S/\partial T = (\partial S^*/\partial T^*) = dT^*/dT$. Figure 1b shows the dependence of the entropy of single-crystal CMN on the temperature determined from formula (1) on the basis of experiments with heating by γ rays (the solid curve is from[16] and when the temperature is determined from the unisotropy of the γ radiation of Ce^{137} nuclei placed in CMN (the symbols are those used in[17]). The dashed curve was calculated on the basis of the Curie law and corresponds to $\Delta S/R = \ln 2 - (S/R) = 2.88 \times 10^{-6} T^{-2}$. We see that all the curves coincide up to 0.006°K, but there is a discrepancy below this point.

Figure 2 shows a plot, taken from the paper of Mess et al.[18], of the magnetic temperature of CMN single crystals vs the thermodynamic absolute temperature in accordance with data by different workers[16-19]. As seen from the figure, there is no agreement between the different data below 0.006°K.

It should be noted that, owing to the temperature jump on the boundary, the time necessary to establish thermal equilibrium between a single-crystal sphere of CMN of radius r = 1 cm and the liquid helium, in which it is placed amounts to several hours at T = 0.01°K, and this time increases like T^{-5} with decreasing temperature. It is clear that such a sphere cannot serve as a thermometer in helium at temperatures below 0.01°K.

To decrease the time required to establish thermal equilibrium, a number of workers have used pressed thermometers consisting of minute CMN crystallites, but time the results of measurements with CMN in the region T < 0.006°K can hardly be regarded as convincing at present. Thus, a CMN thermometer can measure temperatures from 0.006 up to 1°K with accuracy 1–2%.

Another type of paramagnetic thermometer is a nucleus of copper, platinum, or an other metal possessing magnetic a moment. But the nuclear magnetic moment is smaller than the electron magnetic moment by a factor of almost 2000, and therefore methods of measuring paramagnetism of nuclei are much more complicated. Whereas the sensitivity of a ballistic galvanometer or of a low-frequency bridge suffices in the measurement of the magnetic moment of CMN, to measure the magnetic moment produced by nuclei it is necessary to use nuclear-magnetic-resonance methods or a superconducting quantum interference device (SQUID). Since nuclear thermometers operating in the millidegree region are at present the only ones in which the magnetic scale should coincide with the thermodynamic scale, we shall stop to discuss the procedure for measuring temperatures with them in greater detail. The first to determine a temperature on the order of several microdegrees from the value of the magnetic moment of the a nucleus were Kurti et al.[20], and the method was described in detail by Walstedt et al.[21] The gist of the method is that nuclear spins placed in a homogeneous magnetic field H_0 precess about it with a definite frequency $\nu = \gamma H_0$, where γ is the gyromagnetic ratio.

If a sample in the form of a cylinder assembled from platinum foils, as shown in Fig. 3, is placed in a constant magnetic field $H_0 = 470$, then the platinum, being a nuclear paramagnet, acquires an average magnetic moment $M = AH_0/T$ directed along the field H_0. No signal will appear then in a receiving coil (240 turns) whose axis is perpendicular to H_0, since the

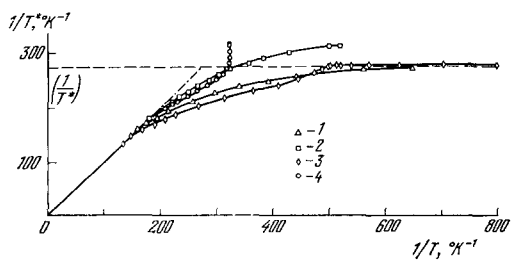

FIG. 2. Connection between the magnetic (T*) and absolute (T) temperatures for spherical CMN samples in accordance with the data of [16] (1), [17] (2), [18] (3), and [19] (4) (dash-dot curve–Curie law; dashed curve–for $(1/T^*)_{sphere}$).

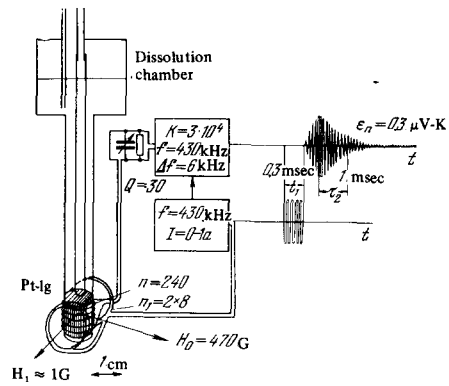

FIG. 3. Diagram of platinum nuclear thermometer.

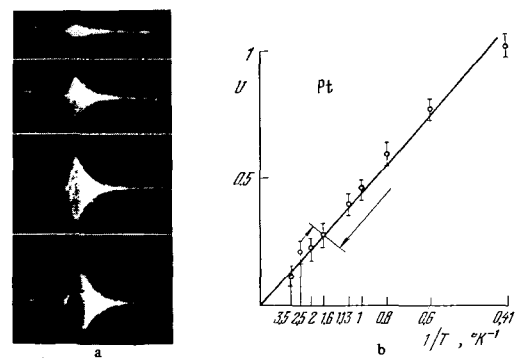

FIG. 4. a) Photographs of signals from the screen of a long-persistence oscilloscope; b– dependence of nuclear signal on the reciprocal temperature.

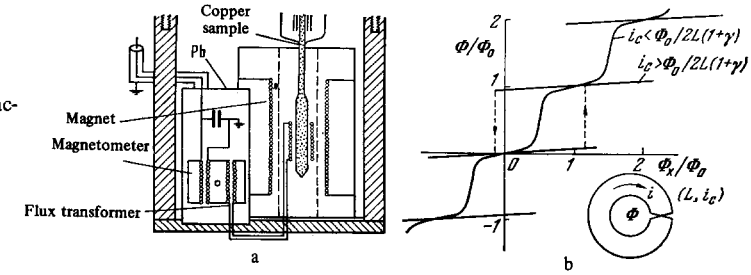

FIG. 5. a) Diagram of quantum magnetometer [25]; b) characteristic of superconducting circuit with Josephson junction [26].

signals of the precessing nuclear spins cancel each other. If now two coils, each of 8 turns, are placed as shown in Fig. 3 with axis perpendicular to H_0 and to the axis of the receiving coil, and a train of waves of radio frequency ν, of alternating-field amplitude H, and duration t is applied to them, then the average magnetic moment will rotate away from the field H_0 to an angle $\theta = \pi\gamma H_1 t$, and will then precess about it at a frequency ν. Now the receiving coils will record a signal proportional to $\nu M\theta$, i.e., $\epsilon \sim \nu H_0 H_1 t/T \sim \gamma H_0^2 H_1 t/T$. This signal, the amplitude of which is proportional to the reciprocal temperature, will attenuate with a nuclear-spin relaxation time τ_2 ($\tau_2 = 0.001$ sec for Pt).

Photographs[22] of signals from Pt^{195} nuclei, amplified and fed to a long-persistence oscilloscope, are shown in Fig. 4a ($T = 2°K$ at the top and $T = 0.01°K$ at the bottom). This determines the temperature in the nuclear-spin system at the instant of the start of the measurements. This is the great advantage of this method. Although the nuclei receive only 10^{-4} erg during the measurement process, their specific heat is very low and they become noticeably superheated (20° at 0.03°K). The next measurement can therefore be performed only after thermal equilibrium is established in the system, i.e., after a time greatly exceeding the time τ_1 of the spin-lattice relaxation. It should be noted that the heating of the spins is proportional to $H_1^2 t$, while the signal is proportional to $H_1 t$, so that it is more convenient to choose t to be of the order of the relaxation time τ_2, of the nuclear spins. A nuclear thermometer is very sensitive to paramagnetic impurities. A thermometer made of technically pure platinum is insensitive at infralow frequencies, so that platinum and copper of high purity are used for nuclear thermometers. Figure 4b shows the calibration of a platinum thermometer made of foil 20 μ thick with $R_{300°K}/R_{1.2°K} = 1000$.

According to the Korringa rule[23], the spin-lattice relaxation time τ_1 for nuclei is inversely proportional to the temperature down to $T \approx \mu H/k \sim 10^{-5}°K$. For platinum $K = \tau_1 T = 0.03$ sec-deg.

Using the Korringa rule, it is possible, by measuring the time τ_1, to determine the temperature $T = K/\tau_1$. Although the accuracy is not very high (about 5—10%), this procedure does provide a new and independent method of checking the temperature scale of a nuclear thermometer.

The lowest temperature obtained and measured in a copper wire sample by Berglund et al.[24] was 0.0006°K. They have demagnetized a sample from 0.016°K and H = 46 kOe to H = 700 Oe in 7.5 hours and obtained a temperature 0.006°K. The temperature subsequently remained below 0.001°K for about 4 hours. The temperature was measured by pulsed nuclear resonance at the ends of copper wires situated outside the demagnetization volume. It should be noted that at $T = 6 \times 10^{-4}°K$ the time τ_1 given by Korringa rule for copper is 0.5 hour, i.e., at a measurement accuracy 25%, only one measurement per hour can be performed.

There is one more very sensitive and accurate but methodologically very complicated method of measuring the temperature, by using nuclear magnetization[25] and a quantum magnetometer.

Figure 5a shows the volume of a quantum magnetometer. The copper sample is placed in a superconducting magnet with a strictly constant field. When the temperature is changed, the change of the magnetic moment of the copper changes the magnetic flux through the coil surrounding the sample, and this changes the flux in the magnetometer. The magnetometer itself is a superconducting circuit short-circuited by a junction through which the electrons pass by tunneling (Josephson junction).

The characteristic of such a circuit is shown in Fig. 5b. It is seen from the figure that the flux through the circuit changes in quanta of value $\Phi_0 = 2 \times 10^{-7}$ G-cm^2. A second coil and a very sensitive circuit with low-noise germanium field-effect transistor operating at helium temperatures are used to measure the number of flux quanta, which determines the magnetic moment M of the copper sample. In principle, the circuit has a sensitivity $\Delta\Phi = 10^{-3}\Phi_0$. Figure 6a shows the results of measurements at temperatures 0.5—0.055°K as a function of T^{-1} determined from the melting curve of He^3. The temperature-measurement accuracy obtained by the authors of[25] in a field of 10 G was 0.5%. The copper used in the experiment had a purity 0.999999. If platinum is used under similar conditions, it should contain not more than 1 paramagnetic impurity atom per 10^7 platinum atoms. It should be noted that the circuit is very sensitive to induced static and to shaking, the production of a Josephson junction having the required characteristic is a very complicated problem, and furthermore the characteristics vary from experiment to experiment.

An instrument of this type is the SQUID. Its operating principle is described in[26,27]. The instrument has unique characteristics. It has a noise level 10^{-22} W and can measure a voltage 10^{-15}—10^{-16}, an inductance change of 10^{-13} H, and a resistance change of 10^{-7}—10^{-8} Ω at a current 10 μA.

For infralow temperatures, a very important question is the establishment of thermal equilibrium be-

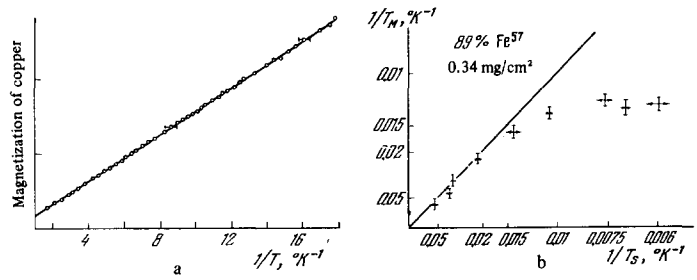

FIG. 6. a) Temperature dependence of the magnetization of copper [25]; b) dependence of $1/T_M$, calculated on the basis of the anisotropy of absorption by two levels of Mössbauer γ quanta, on the reciprocal temperature $1/T_S$ determined from CMN.

tween the medium and the thermometer. If the medium is liquid He^3 or a solution of He^3 in He^4, then at $T < 0.1°K$ their specific heats, like the specific heat of metals, is directly proportional to the temperature, but the bulk specific heats of the liquids is larger by hundreds of times than the bulk specific heat of the metals. Therefore the transient time τ will depend only on the specific heat of the metal and will amount to $\tau = RC\delta/2$, where R is the Kapitza jump on the metal-liquid boundary, C is the specific heat of the metal, and δ is the thickness of the metal plates. According to measurements by Zinov'eva[28], at $T < 0.4°K$ $R = 50/T^3$ deg-cm/W or $C \approx T \times 10^{-4}$ J/cm³deg, $\delta = 20\ \mu$, and $T = 0.03°K$, we have $\tau \sim 1$ sec, and at higher temperatures it is even smaller, since $\tau \sim 1/T^2$.

Thus, nuclear thermometers made of pure metals such as platinum, copper, and others are suitable for temperature measurements in the range from millidegrees to degrees, but the procedure is very complicated and the measurements can be performed only in the presence of a constant magnetic field.

There is another possibility of measuring infralow temperatures with the aid of γ rays is in a strong magnetic field produced by neighboring atoms, then the Zeeman splitting of the levels takes place and its value for iron is $\Delta = k \times 2.2 \times 10^{-3}°K$. The population of the lower levels will be larger than that of the upper levels, and the level population ratio is $N_1/N_2 = e^{\Delta/kT}$. If the temperature is close to Δ/k, then N_1/N_2 will differ noticeably from unity, and the temperature is given by $T = \Delta/k \ln (N_1/N_2)$. The ratio N_1/N_2 can be determined either from the anisotropy of the radioactive γ emission or by the Mossbauer method, if the γ-quantum absorption is measured. It is known[17] that the anisotropy of Ce^{137m} isomorphically crystallized in a CMN lattice is variable down to the lowest temperatures obtainable by CMN demagnetization, which were estimated from the value of the anisotropy at $1.9 \times 10^{-3}°K$.

At the same time, determination of N_1/N_2 by measuring the difference of the Mössbauer line absorptions yields different results. Thus, measurements performed at the ISP of the USSR Academy of Sciences[29] on micron-thick foil of iron enriched with Fe^{57}, placed in the dissolution chamber of He^3 in He^4, have shown that the difference of the absorptions hardly increases with decreasing temperature, starting with $T \sim 0.015°K$, while the form of the Mössbauer spectrum remains unchanged. The dissolution machine operated in a one-shot regime, and the temperature was determined from the susceptibility of the CMN. Figure 6b shows a plot of $1/T_M$, calculated on the basis of the anisotropy of absorption in iron by two Mössbauer γ-quantum levels, against the reciprocal temperature $1/T_S$ determined from the magnetic susceptibility of a sphere made of compressed CMN crystallites.

At 0.01° we have for iron $\mu H/kT \approx 0.11$, i.e., a noticeable magnetization of the nuclei is observed, so that it is possible that this phenomenon is due to the onset of exchange between the nuclear spins of the different domains via spin waves. However, this question calls for more detailed investigations.

Summarizing, we can state that a determination of an infralow temperature close to thermodynamic at $T > 0.006°K$ is possible at the present time by using CMN thermometers and nuclear thermometers. Additional experiments are necessary to establish a temperature scale below 0.006°K.

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