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FUTURE OF QUARTZ RESONATOR THERMOMETRY

by

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ABSTRACT

This paper will attempt to predict the future of precision thermometry based on quartz crystal resonators used as thermal sensors. At present, quartz resonator thermal sensors exhibit considerable hysteresis after temperature cycling and, therefore, are not generally used for precision thermometry. However, we have shown that the sensors can be used to detect temperature fluctuations of approximately 20 μK over many seconds. Moreover, major advances in quartz resonators, including new crystallographic cuts, hold promise of producing quartz resonators with greatly reduced hysteresis. These new advances will be discussed in terms of their implication for thermometry from ≈100 to 400 K. A new technique for utilizing quartz resonators for thermal measurements will be discussed in detail. It is expected that a few of these improved resonators will become available for testing within a few months.

INTRODUCTION

Experimental thermodynamics can be said to be a study of thermometry as much as a study of fundamental properties of macroscopic systems. The present accuracy limit of temperature measurements near 300 K is of order 50 to 100 μK and is obtained using platinum resistance thermometers (14).[2] Thermistors used as thermal sensors yield a resolution of approximately 10 μK and a temperature stability of order 1 mK/100 days (4, 19). Quartz crystal thermometers presently have a temperature resolution of order 2 μK; however, they are plagued by hysteresis effects due to thermal shock and cycling. Several new experimental quartz crystal resonators show promise of achieving temperature resolutions of less than 1 μK with daily variations of less than 10 μK and greatly reduced thermal shock and/or cycling-induced hysteresis.

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[2] Underlined numbers in parenthese refer to items in the list of references at the end of the report.

The development of a convenient thermal sensor with 1-μK resolution would provide an important new tool for many areas of science, especially thermodynamics. This paper will review the background, problems, and capabilities of present quartz crystal resonators used as thermal sensors, describe new advances in resonator design and fabrication which are likely to lead to much greater accuracy, and finally, describe some of the methods of using quartz crystal resonators as thermal sensors.

PRESENT STATUS OF QUARTZ THERMAL SENSORS

Quartz crystal resonators have long been known for their stable resonance frequency. Present high-quality commercial quartz-controlled oscillators have achieved frequency stabilities of order 3×10^{-13}.

The potential for using a quartz crystal resonator to obtain a high-resolution digital thermometer has long been recognized (7-9, 11, 15). Indeed, Smith and Spencer in 1962 used a high-quality quartz resonator as the frequency-determining element of an oscillator and obtained a temperature resolution of approximately 5 μK for a 10-s measurement time and a drift of less than 100 μK per hour, illustrated in figure 1 (15). The frequency dependence was approximately 74.6 ppm/K and was relatively linear from -20° C to +100° C. Presently available quartz thermometers are considerably more linear than the results obtained in references 15 and 9 (fig. 2). However, they suffer from thermal stress effects which result in spurious temperature reading of order 10 mK following thermal cycling (fig. 3). This apparent temperature typically drifts approximately 10 mK over several days immediately following a temperature shock of many degrees. Because of these hysteresis effects, quartz crystal resonators have generally not been used in precision thermometry where temperature cycling is required. An additional problem for using the commercial quartz sensors for μK resolution thermometry is the large heat conduction along the sensor cable. This can be overcome either by replacing the sensor cable or by very careful heat sinking of the sensor cable. The advantages of these sensors are that they dissipate only a few μW, they are easily made into a digital thermometer whose output can be manipulated with great precision using microprocessors, etc., and they have a thermal time constant of less than 1 s. Measurements made with these sensors indicate a temperature resolution approaching 2 μK and a drift of less than 100 μK/day (17).

Figure 4 shows the temperature versus time behavior of a single-stage oven that employs one commercial quartz crystal as the thermal sensor for controlling the heater power and a different quartz sensor for detecting temperature changes. These data were obtained after the system had operated on temperature for 2 days in order to allow some of the thermal-shock-induced transients to die out. Figure 5 shows the fractional temperature fluctuations for the single-stage oven, as measured by the quartz thermometer using experimental electronics (17). Note that for the averaging time of 1 s, the approximate thermal time constant for the sensor, the temperature resolution is approximately 3 μK, while at averaging times of 10 s it is approximately 30 μK. It is expected that temperature stabilities of order 2 μK could be obtained for times of many minutes in a two-stage oven if great care is used in

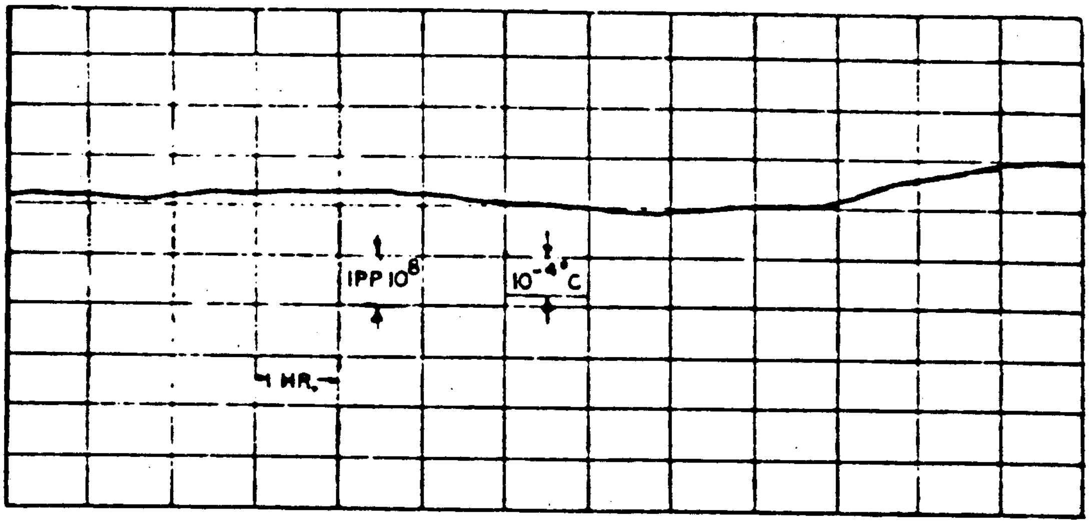

FIGURE 1. - Temperature variation of a double oven versus time measured using 5° Y cut quartz resonator (15).

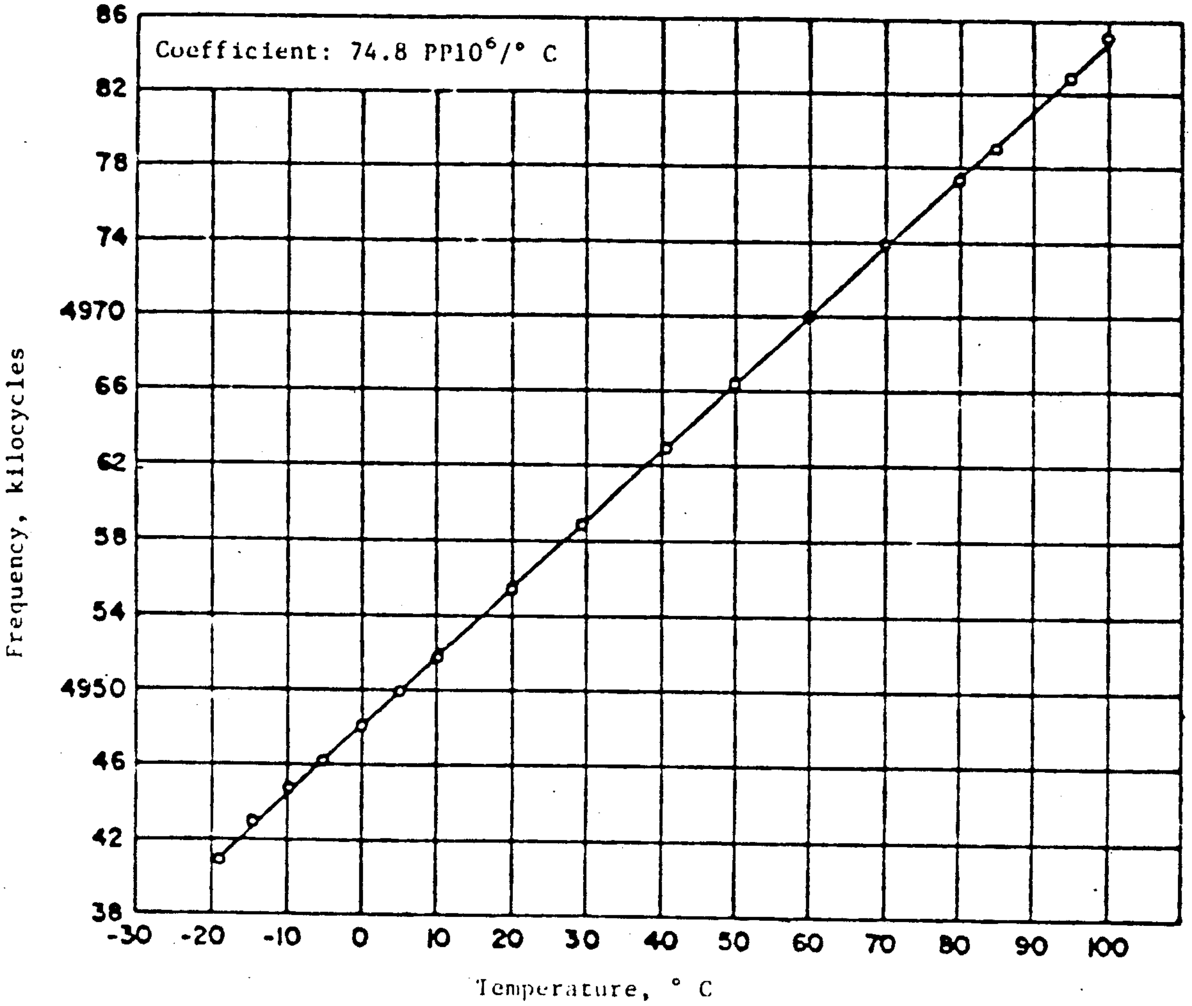

FIGURE 2. - Frequency versus temperature over a 120° C range for 5° Y cut quartz plate (15).

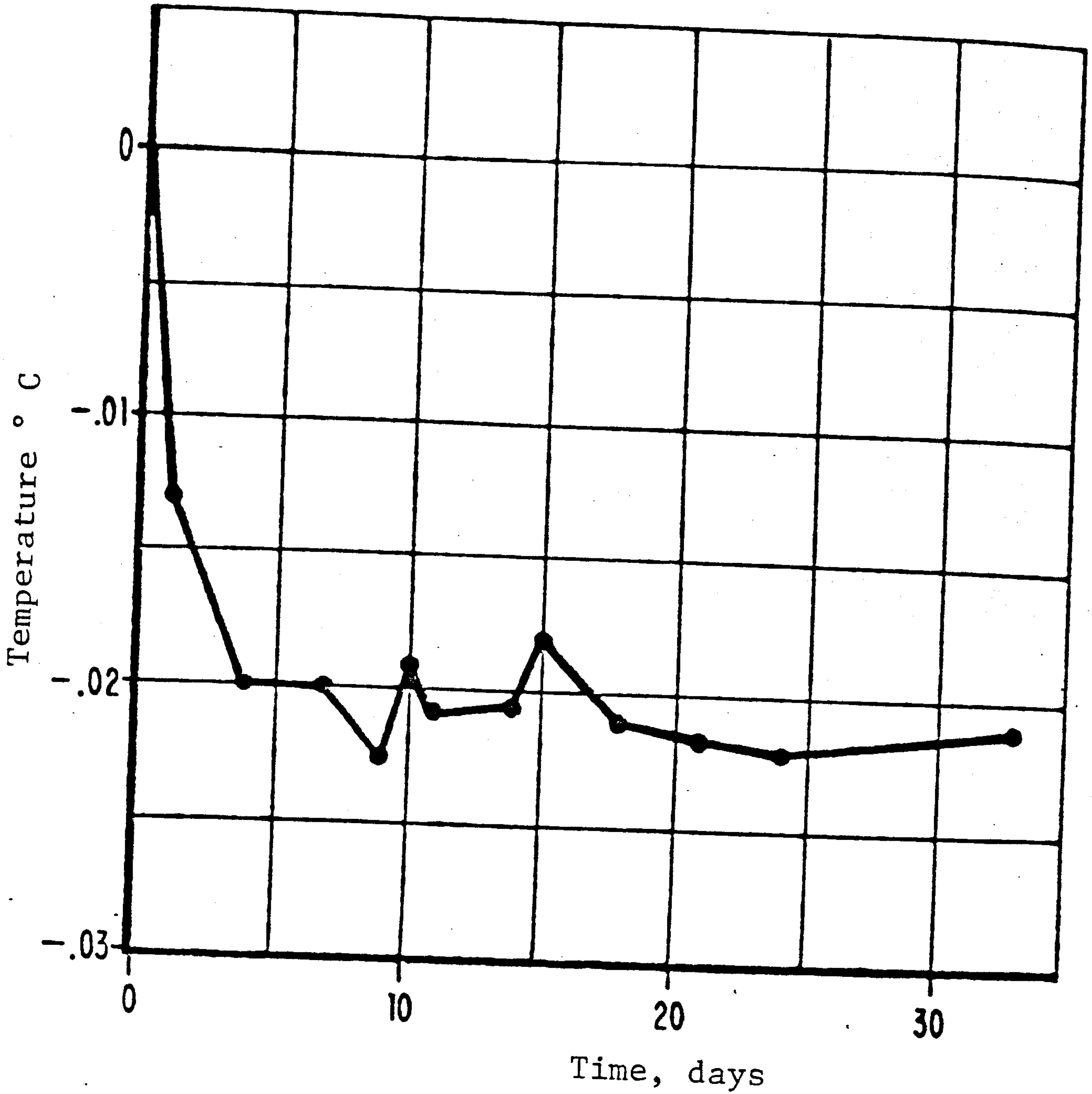

FIGURE 3. - Temperature error of an LC cut quartz thermometer as a function of time (9).

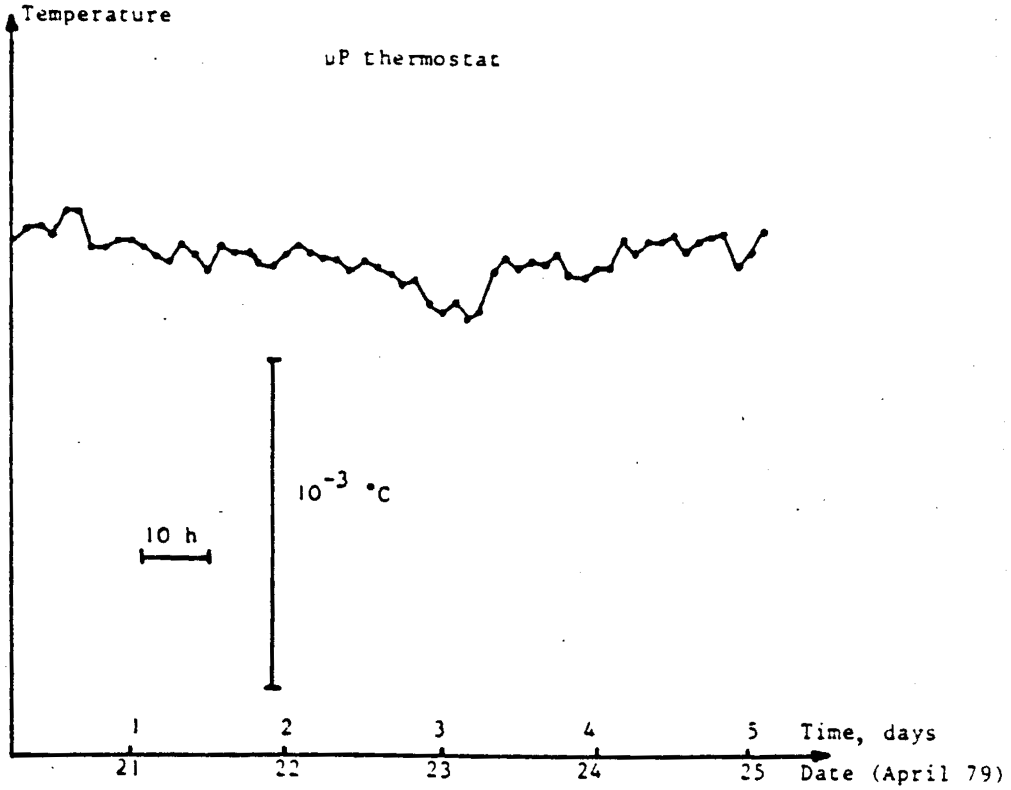

FIGURE 4. - Temperature stability of LC quartz thermometer-controlled oven as measured with a second LC cut resonator (17).

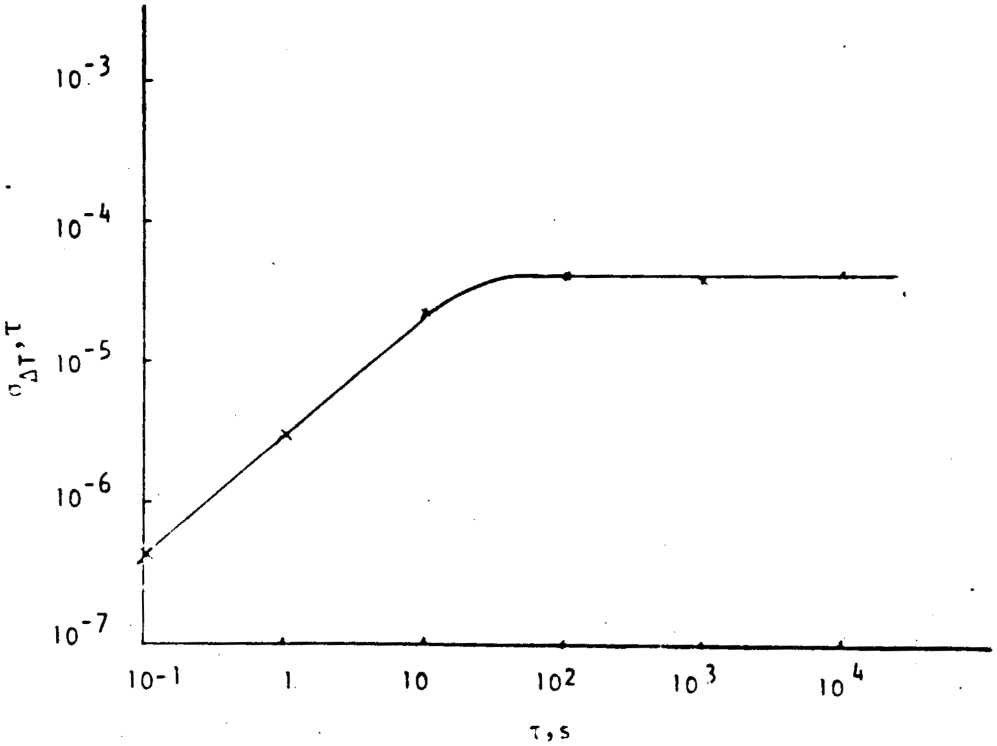

FIGURE 5. - Fractional temperature fluctuation as a function of measurement time (17).

heat-sinking the sensor cables. A resolution of 2 μK is probably close to the limit imposed by stress relaxation in this type of sensor.

NEW DEVELOPMENTS IN QUARTZ RESONATORS

Stress-induced frequency transients have received considerable attention in the past 5 years because they are responsible for much of the frequency instability of high-precision quartz crystal resonators used in frequency control (1, 10, 16-17). It appears likely that they are responsible for the thermal hysteresis as well as the noise in quartz sensors. In the past 2 years, great strides have been made in designing crystallographic cuts that are less sensitive to stress, and to new mounting techniques that help to minimize temperature (i.e., stress) induced transients (2-3, 5-6, 12, 17).

For example, a fifth-overtone, 5-MHz quartz resonator has a temperature sensitivity of approximately $\Delta v/v = 10^{-8}/K$, yielding a linearized sensitivity of ~ $\Delta v/v = 10^{-6}/K$, 10 K away from turnover. Since the frequency stability of such precision units is $\Delta v/v = 10^{-13}$, one would expect to achieve temperature resolution of 0.5 μK; however, the thermal-transient-induced response is $\Delta v/v = 10^{-5}\ dT/dt$, requiring that dT/dt be less that 10^{-8} K/s in order to achieve this 1-μK temperature resolution and stability. Most ovens and test systems do not have the low-temperature transients required to use AT cut resonators for temperature sensors.

The new SC[3] cut resonators (2-3, 12, 17) have a measured transient response which is 50 to 100 times smaller than that for AT cuts. Therefore, dT/dt of only 0.5 to 1 x 10^{-7} K/s is required in order to resolve 1 μK. SC cut resonators are therefore excellent candidates for thermal sensors. These new SC cut resonators have another very interesting feature; namely, they can be made to oscillate on both the B and C mode simultaneously (13). The C mode can be made with $\Delta v/v \approx 10^{-9}/K^2 + 10^{-7}\ dT/dt$, while the B mode has a linear response of $\Delta v/v \sim 2.5 \times 10^{-5}/K$, with a so-far-unmeasured transient response. It may be possible to use the C mode as the frequency reference for the microprocessor-based counter measuring the frequency of the B mode. With such a system, temperature resolutions of approximately 0.1 μK are contemplated. Using only the B mode temperature resolution of 1 μK over hours, and 10 to 20 μK per day resolutions appear likely. Precision SC cut resonator should be commercially available within 9 months.

Additionally, we have been studying the thermal transient process. From measurements on AT and SC cut resonators, it appears that the aging which leads to apparent frequency temperature drift can be greatly reduced by applying an exponentially decaying thermal sign wave to the final temperature. This process anneals out the bonding stress, thereby reducing the hysteresis effect observed in all resonators to date.[4]

Aging rates in the frequency of the C mode of an SC cut resonator are

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[3] For SC and AT cuts, see reference 1.

[4] Private communication from S. R. Stein, C. M. Manney, Jr., and G. M. Kielian, National Bureau of Standards, Boulder, Colo.

dropped from approximately 10^{-10}/day (~10^{-4} K/day) to approximately 10^{-11}/day (~10^{-5} K/day) after a 2-hour annealing cycle. Normally, such an improvement in the aging rate would require more than 40 days. The effect on the B mode has not been measured yet.

A new approach to reducing bonding and plating stress is shown in figure 6 (2-3). The oscillating quartz resonator is the center portion of part C, shown in the cross section. The resonator is fixed at the ends between two auxiliary plates, D1 and D2. D1 and D2 carry the resonator electrodes, which are separated by ~20 µ from resonator C. This design greatly increases the isolation between the resonator C and the bonding points. This technique has been applied to precision SC cut resonators, yielding frequency stablities of order 1 x 10^{-13}, which should make thermal sensors with resolutions of 0.1 µK at 100 s possible.

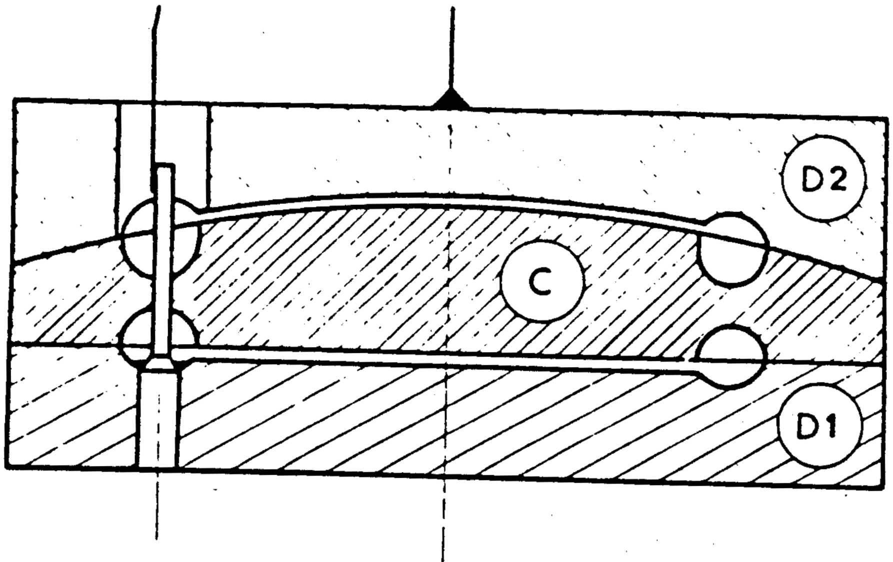
The application of such technology to the LC cut used in the present commercial quartz thermometer (9) or the Y cut used by Smith and Spencer (15) could yield a thermal sensor with resolution of order 0.1 µK, a linearity of order 10 mK/100 K, and a daily stability of order 20 µK.

DISCUSSION OF MEASUREMENT TECHNIQUES

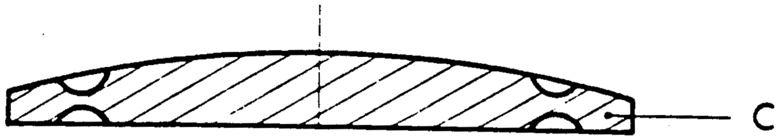
The traditional method of using a quartz crystal as a temperature sensor is to make it the frequency-determining element in a oscillator (fig. 7). The output frequency of the oscillator is then compared with a very stable reference, yielding a difference frequency dependent on temperature. For the resonator of reference 9, the frequency derivation is linear to approximately ± 0.01 pct over the range from 0° to 200° C. AT cut and C mode SC cut resonators have a cubic frequency dependence on temperature which can be locally linearized. To achieve enough resolution to detect 1 µK changes in temperature, it is necessary to detect frequency changes of order 10 to 100 µHz. To resolve 100 µHz with a direct-counting scheme requires a counting time of 10^4 s, or 2.4 hours. The same resolution (100 µHz) can be obtained in 10 s by deterodyning the temperature-controlled oscillator down to 1,000 Hz and measuring its period with a resolution of 1 µs, which is easily done using a frequency synthesizer. Such a scheme gives a direct digital readout of temperature which is easily processed.

The practical problem of such a circuit is that the electrical phase around the oscillating loop must be maintained extremely stable--of order 10^{-5} radians--in order to achieve µK stabilities. This, in turn, requires that the sensor cable be held mechanically very stable and that its temperature be held constant to ~1 K. It is very likely that much of the spurious temperature fluctuations observed with quartz sensors have been due to phase changes along the sensor cable because of mechanical and/or thermal disturbances.

Figure 8 shows the block diagram of a new technique for locking an oscillator to a quartz crystal where the electrical phase length of the sensor cable is relatively unimportant (16). Phase modulation sidebands at Ω_1 are impressed upon a carrier frequency originating from a frequency source. The carrier frequency probes the quartz crystal resonance, while the modulation



BVA_2 Resonator



BVA_2 crystal

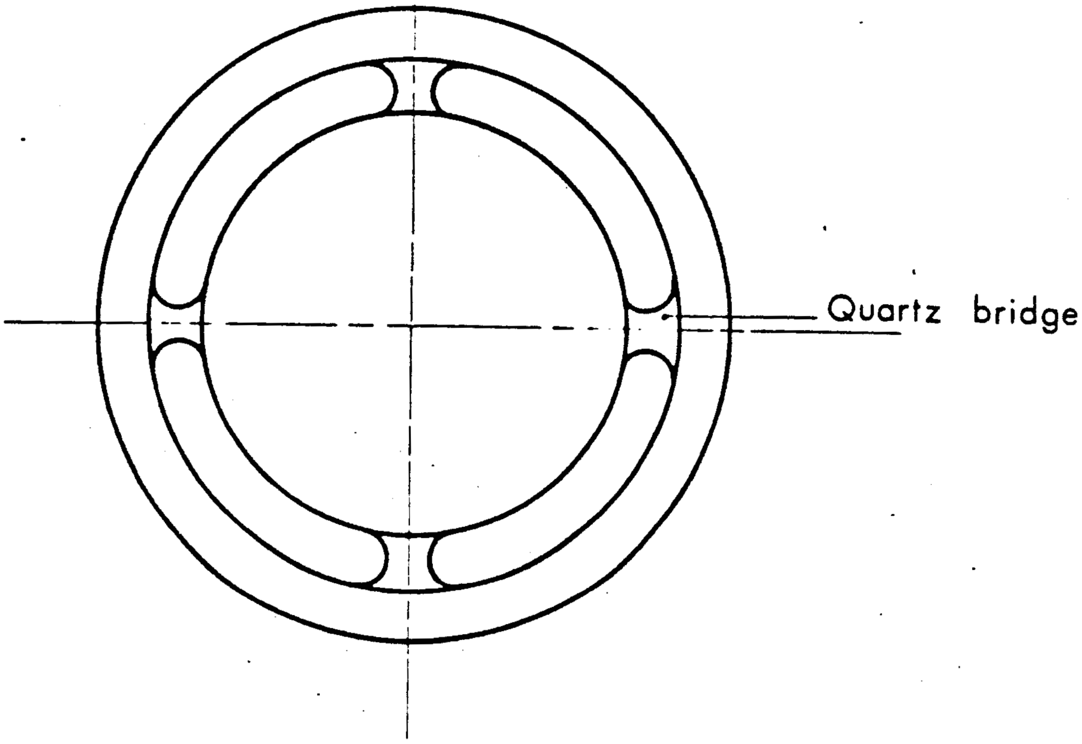


FIGURE 6. - New quartz resonator design (2).

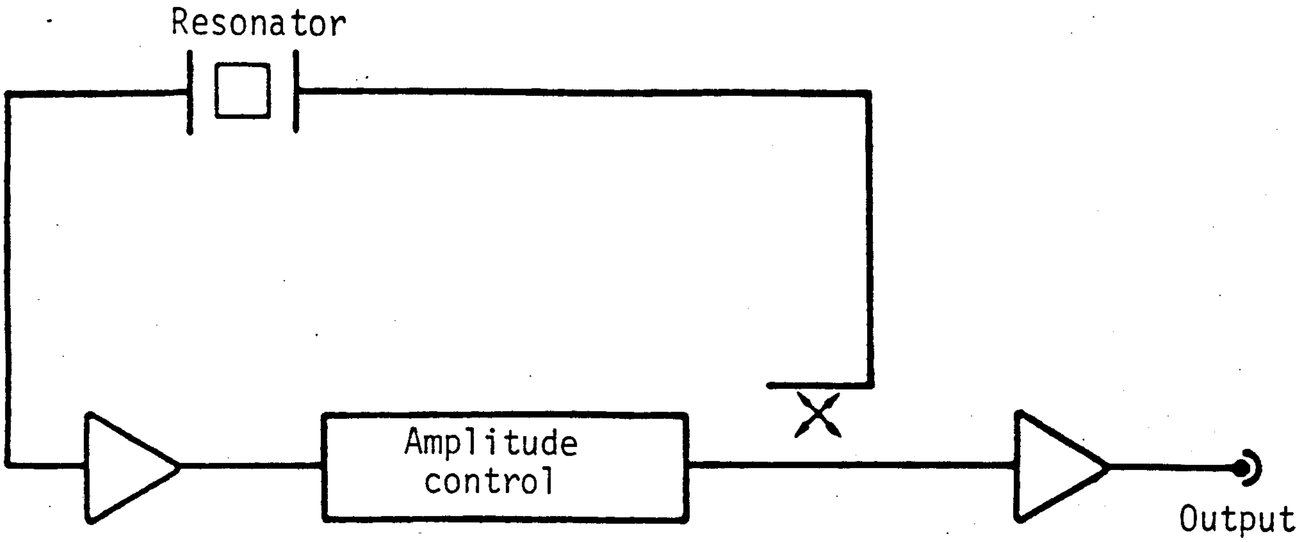


FIGURE 7. - Traditional crystal controlled oscillator.

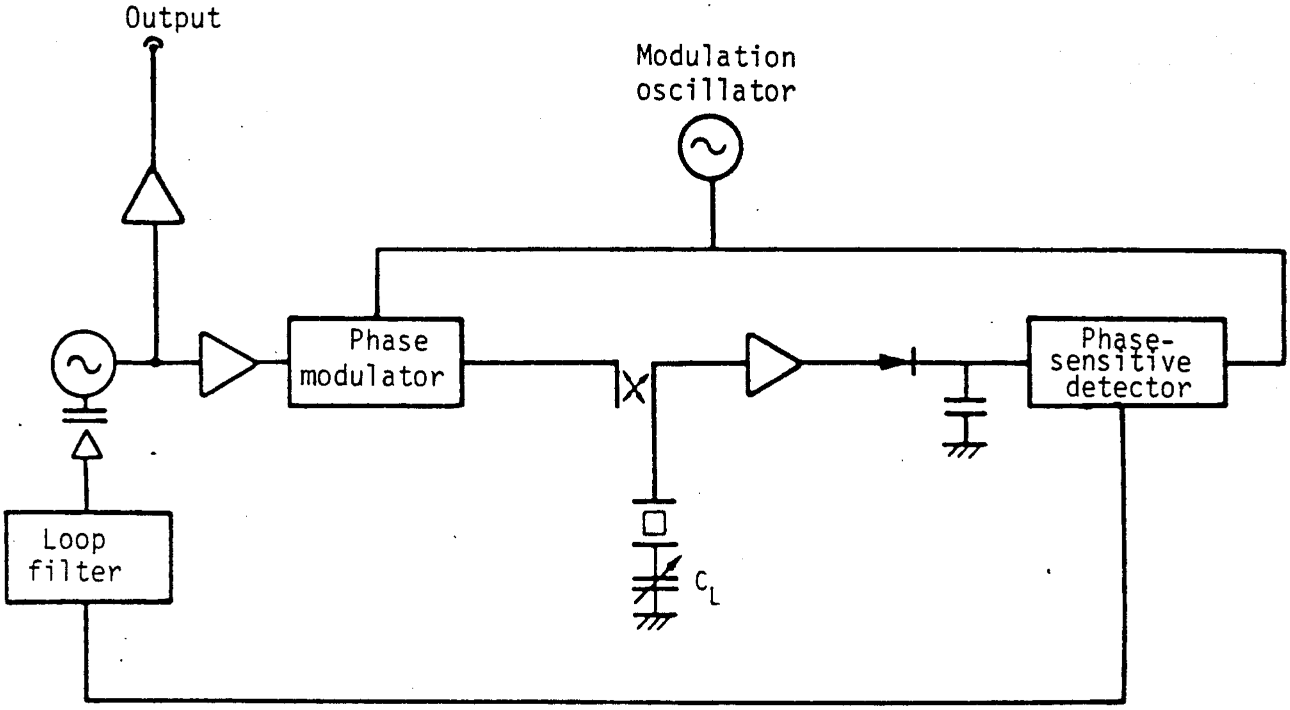


FIGURE 8. - New passive crystal oscillator design (18).

sidebands are reflected off a virtual short since they lie outside the crystal resonance. If the frequency source is detuned from the resonance frequency of the crystal, amplitude modulation at Ω_1 results. The phase of the amplitude modulation relative to the impressed phase modulation depends on whether the frequency source is higher or lower than the quartz crystal, thus enabling one to electronically steer the frequency source to the center of the quartz crystal resonance. The important new aspect here is that the phase of the carrier frequency, v_0, is measured relative to that of the modulation sidebands at $v_0 \pm$ 167 Hz, so that the phase change due to changes in electrical length of the sensor cable is reduced by approximately 10^5 relative to the traditional simple oscillator scheme described above. This technique has been used to obtain a frequency stability of 7×10^{-14} using a 5-MHz SC cut crystal, which is nearly five times better than that obtained previously (16). If the SC crystal had been operated away from its turnover point, this would correspond to a temperature resolution of approximately 0.07 μK.

Using this new technique, the author believes that temperature resolutions of much less that 1 μK, perhaps even as good as 0.1 μK, will be achieved in the near future. It appears that this temperature resolution should be obtainable from approximately 100 to 400 K and perhaps higher.

The above analysis and measurements have primarily addressed temperature resolution without regard to absolute reproducibility. At the present level of technology and understanding, it is likely that hysteresis effects following large temperature excursions will remain at least an order of magnitude above the best resolutions obtainable with the same devices. Therefore, it seems appropriate to suggest that, whenever possible, calorimetry experiments be designed to operate at constant temperature and constant heat loss, and that one measure the heat input necessary to maintain constant temperature. This would appear to have great advantages in terms of maintaining thermal gradients constant in space and time, and minimizing thermal transients, both of which make the measurement of temperatures to μK resolutions extremely difficult.

CONCLUSION

It has been shown that the present commercially available quartz crystal thermometer is capable of achieving a temperature resolution in excess of 20 μK over many seconds, provided care is taken in heat sinking the sensor cable and thermal cycling is avoided. Thermal cycling of the sensor by 100 K typically causes a spurious temperture reading of order 10 mK, which dies away in several days. Temperature stability of presently available quartz thermometers and also AT cut resonators normally used for frequency control application is typically dominated by residual mounting and/or plating stress which changes under thermal shock. New quartz resonator crystallographic cuts and new mounting techniques provide resonators that have thermal-shock-induced transients of order 50 times smaller than traditional resonators. It has been proposed to use these new quartz resonators, which were originally designed for frequency control, as thermal sensors. Calculated resolution based on the best achieved frequency stability at the temperature turnover point indicates an ultimate temperature resolution in excess of 0.1 μK if the same frequency stability can be achieved away from the temperature turnover point.

The traditional oscillator scheme of using quartz crystal resonators as thermal sensors was briefly described, and the most serious cause of spurious temperature readings due to practical electronic problems was discussed. A new technique with several orders of magnitude smaller spurious readings due to electronic problems was described, and preliminary results were presented.

Finally, it is suggested that whenever possible, calorimetry experiments should be designed to operate at nearly constant temperatures, thereby reducing temperature measurement problems due to thermal cycling, changing gradients, etc. Instead of measuring temperatures during a process, one measures the amount of heat necessary to maintain constant temperature in the presence of a known constant heat leak. Under such conditions it was predicted that effective temperature resolutions of much better that 1 μK can be achieved in the near future.

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