

QUANTUM CALORIMETRY

Your opponent's serve was almost perfect, but you vigorously returned it beyond his outstretched racket to win the point. Now the tennis ball sits wedged in the chain-link fence around the court. What happened to the ball's kinetic energy? It has gone to heat the fence, of course, and you realize that if the fence were quite a bit colder, you might be able to measure that heat and determine just how energetic your swing really was.

Calorimetry has been a standard measurement technique since James Joule and Julius von Mayer independently concluded, about 150 years ago, that heat is a form of energy. But only in the past 15 years or so has calorimetry been applied, at millikelvin temperatures, to the measurement of the energy of individual photons and particles with exquisite sensitivity.

Low-temperature microcalorimeters have several unique properties that have led to a variety of applications.¹ Here, to begin, we introduce a few of those applications before focusing in detail on high-resolution astrophysical x-ray spectroscopy. Still relatively young, the technique of quantum calorimetry (the thermal measurement of energy quanta) will no doubt find more applications that will be just as exciting and potentially revolutionary as the advances promised in x-ray astronomy.

Because of the exceedingly small specific heats of many materials at very low temperatures, detectors can be quite large and still be sensitive to small amounts of deposited energy. This energy is sensed after it has been converted to heat, so that even interactions that produce little or no ionization can be detected.

These two properties have made calorimetry an attractive choice in searches for WIMPs, the weakly interacting massive particles that are among the leading candidates for the missing dark matter in the universe. The scattering of a single WIMP is predicted to transfer roughly 1 keV to nuclei of the target material. Such slow nuclei produce much less ionization than electrons of the same energy. By simultaneously measuring both the thermal and ionization signals, it is possible to efficiently glean the real events from the much more numerous background events that interact with electrons and produce relatively more ionization.²

This sensitivity to nonionizing events can also be used to detect DNA fragments in a time-of-flight mass spectrometer. At energies of 10–20 keV, these massive molecules produce little or no ionization on impact. Nevertheless, the energy resolution of a thermal detector is ample for distinguishing singly and doubly charged fragments.³

Novel detectors that operate at 60 millikelvin are now being used to study cosmic gas at millions of kelvin.

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Another useful property of thermal detection is that it does not depend on the charge transport properties of the absorber. Only a very few materials, among them highly pure silicon or germanium, can be used to make ionization detectors, whereas a calorimeter can even incor-

porate the radioactive source being investigated.⁴ This advantage is being exploited in investigations of beta decay spectra to put limits on the neutrino rest mass. Beta spectrometer results are limited by the systematic uncertainties that arise when correcting for energy lost in the source. By contrast, a calorimeter that incorporates the source measures directly the total decay energy minus whatever is carried away by the neutrino—provided no metastable states are created with lifetimes longer than the thermal integration time of the detector.⁵

When a calorimeter measures energy, it is in a low-temperature equilibrium state in which nonthermal excitations have negligible populations. To the extent that a calorimeter can be regarded as a closed system, the event-to-event statistical fluctuations that limit the resolution of an ionization detector do not occur. It is possible to make small calorimeters with resolving powers $E/\Delta E$ of 5000 or more for x rays—almost a factor of 100 better than an ideal silicon solid-state detector and a great leap forward not just for x-ray astronomy, but also for materials microanalysis. (See PHYSICS TODAY, July 1998, page 19.)

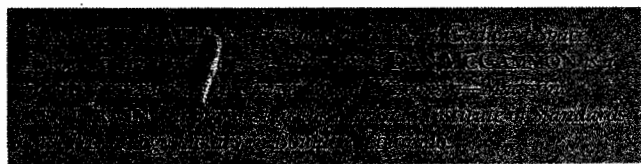
Lower, but still useful, resolving power can be extended all the way down to the near infrared, raising the possibility of fabricating imaging arrays for visible light that simultaneously provide spectra and photometry.⁶

Since much of the early work on quantum calorimeters was inspired by a quest for better astrophysical x-ray spectroscopy, we use this application as a detailed example of the basic physics of calorimeters. But the results are general, and can also be used to determine the suitability of calorimetric methods for other experiments.

X rays reveal the hot universe

Since its beginnings in the 1960s, x-ray astronomy has played a large part in revealing the universe's unexpectedly hot and energetic side. (See David Helfand's article, "X Rays from the Rest of the Universe," PHYSICS TODAY, November 1995, page 58.) Today, we know that most of the universe's nonexotic matter (at least in the forms we can detect) exists as gas at temperatures of tens of millions of degrees. Clusters of galaxies—the largest gravitationally bound structures in the universe—hold huge quantities of this gas, whose combined mass in a typical cluster outweighs that of the constituent galaxies by up to a factor of five. Essentially invisible except at x-ray wavelengths, the hot intracluster medium traces a cluster's gravitational potential well, whose main source of mass is neither hot gas nor galaxies, but the still-mysterious dark matter.

X rays have also provided a way to study physics in the immediate vicinity of black holes (see Roger Blandford

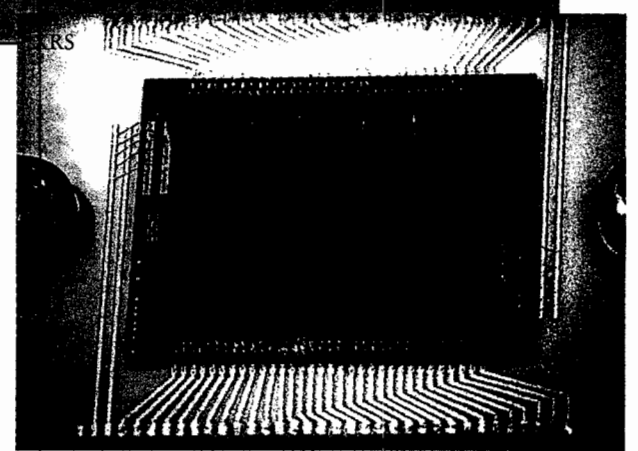
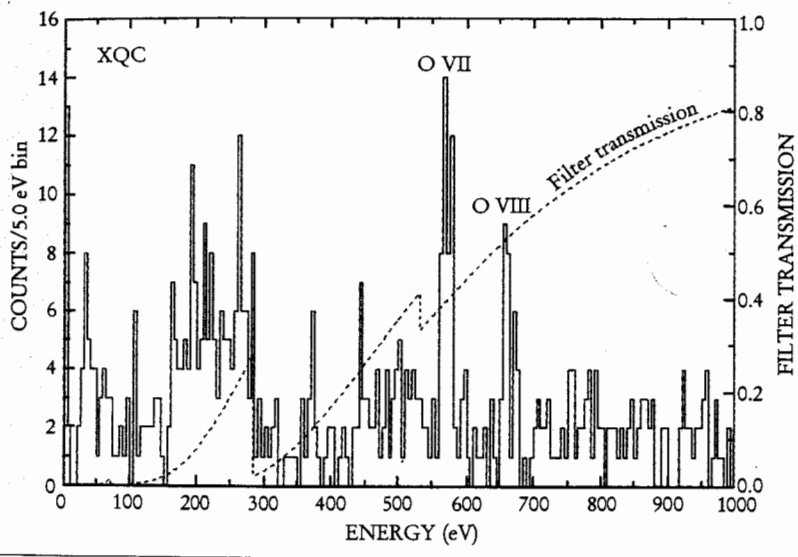


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The First Calorimeters in Space

Two calorimeter arrays were used to measure cosmic X-rays as part of an experiment on the Solar Wind Anomalous Coronal Emission (SWAN) aboard the Solar Wind Anomalous Coronal Emission (SWAN) satellite. The experiment was designed to measure the high-energy x-ray emission from the solar corona and to study the solar wind. The experiment was designed to measure the high-energy x-ray emission from the solar corona and to study the solar wind. The experiment was designed to measure the high-energy x-ray emission from the solar corona and to study the solar wind.

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and Neil Gehrels's article, "Revisiting the Black Hole," PHYSICS TODAY, June 1999, page 40) and neutron stars (see Lars Bildsten and Tod Strohmayer's article, "New Views of Neutron Stars," PHYSICS TODAY, February 1999, page 40).

To see why x rays reveal the extreme environments around these two kinds of compact star, consider the Stefan-Boltzmann equation,

$$P = A\sigma T^4,$$

which gives the radiative power P of a black body in terms of its radiating area A , its temperature T , and the Stefan-Boltzmann constant σ . This equation, combined with the inverse-square law, tells us that small, faraway things will be too faint to see unless they are very hot. For example, a celestial object—a neutron star, active galactic nucleus, or supernova remnant, say—can be 10^{12} times smaller in area or 10^6 times farther away and still be as visible, provided it's also 10^3 times hotter and that we're

looking in the right waveband! Fortunately, the gravitational potential near neutron stars and black holes provides more than enough energy to raise the temperature of infalling matter to x-ray emitting levels.

By measuring this energetic radiation, we can study regions of extreme temperature and gravitational and magnetic fields that cannot be reproduced on Earth. Spectroscopy can be an effective tool in this endeavor, especially for the 0.1–10 keV x-ray band, corresponding to temperatures from 10^6 to 10^8 K. At these temperatures, hydrogen and helium are fully ionized, but bremsstrahlung is still inefficient, and the dominant radiation consists of collisionally excited characteristic lines of partially ionized heavy elements. These lines provide a wealth of diagnostics on the elemental abundances and physical conditions in the gas, and measurements of Doppler shifts and linewidths provide invaluable information about the gas's motion.

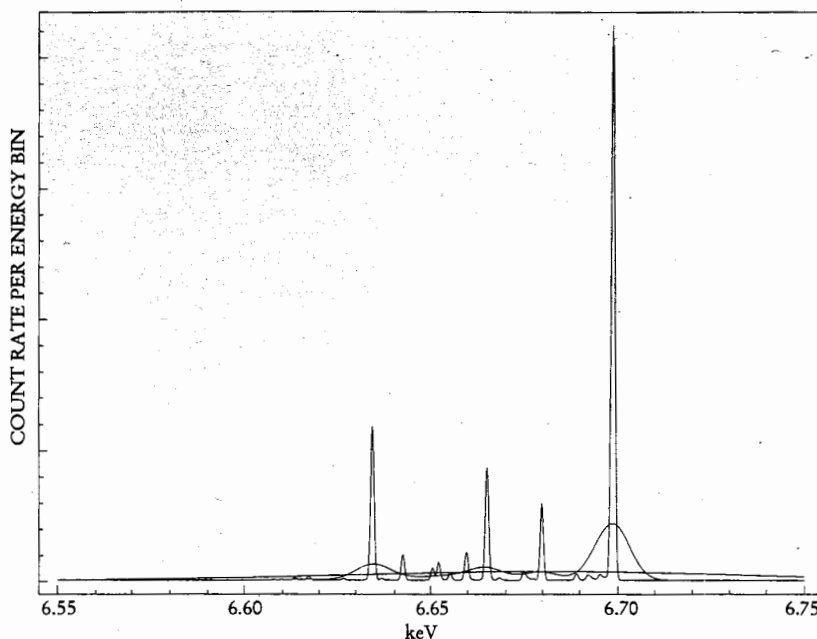
However, the detailed physical information that the spectra contain can only be extracted by a high-resolution

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FIGURE 1. IRON K EMISSION LINES from a model plasma at 7×10^7 K, as viewed with a spectral resolution of 1 (blue), 10 (green), and 100 eV (red). Clearly, high-resolution detectors must be used to exploit the full diagnostic potential of x-ray spectroscopy.



spectrometer like a calorimeter. In the precalorimeter era, the choice between wavelength dispersive devices (diffraction gratings and Bragg crystal spectrometers) and nondispersive spectrometers (proportional counters and solid-state detectors) presented a dilemma. Dispersive spectrometers offer very good energy resolution, but at low throughput. Nondispersive spectrometers, on the other hand, have very high efficiency, but relatively poor resolution.

Gratings have further disadvantages. Their good energy resolution requires optics with very high angular resolution, but, to date, the design of x-ray telescopes has involved a trade-off between angular resolution and collecting area, putting gratings at a sensitivity disadvantage. Furthermore, gratings disperse the spectrum—into several orders—across a position-sensitive detector. But when the x-ray emitting object is itself extended, untangling the dispersed spectra is a daunting prospect.

As a result of these considerations, most astronomical x-ray spectroscopy so far has been carried out with nondispersive detectors that directly determine the photon energy by measuring the amount of ionization produced by each event. Unlike gratings, the energy resolution of these devices is not affected by the acceptance angle, so they can have very high throughput. However, statistical fluctuations in the fraction of a photon's energy that goes into ionization from one event to the next fundamentally limit the resolution.

For silicon, the theoretical limit is about 100 eV at 6 keV, a value that is closely approached by modern x-ray charge-coupled devices. An examination of figure 1, which shows modeled x-ray emission around the iron K lines from a 7×10^7 K plasma as seen with 1, 10, and 100 eV resolution, makes it clear that much higher resolution is needed to take full advantage of the information available. Indeed, the need for higher resolution is what inspired our quest for a better detector.

The basics of x-ray calorimetry

As shown in figure 2, an x-ray calorimeter is conceptually a simple device, whose basic principles are obvious from the outset. Those principles are as follows:

- ▷ The calorimeter should operate at a low temperature so that the energy deposited is large relative to the thermodynamically unavoidable random transfer of heat across the weak link.
- ▷ The absorber should be opaque to x rays, and yet have a low heat capacity so that a small deposition of energy is translated into a measurable temperature change.
- ▷ The absorber must thermalize well—that is, it must reproducibly and efficiently distribute the energy of the initial photon across a thermal distribution of phonons (or electrons, depending on the thermometer).
- ▷ The thermometer must be sensitive.
- ▷ And the thermal link should be weak enough such that

the time for the base temperature to be restored is the slowest time constant in the system (compared with thermalization and diffusion times), yet the link should not be so weak that the device is too slow to handle the incident x-ray flux.

How well can a calorimeter measure the energy it absorbs? Consider a calorimeter with a thermometer that translates changes in temperature into a voltage. It has a heat capacity C and a link with thermal conductance G to a heat sink. An instantaneous deposition of energy into the absorber produces a voltage pulse with an exponential decay time constant τ equal to C/G .

In the frequency domain, the signal is nearly independent of frequency f when $f \ll 1/2\pi\tau$, and falls off as $1/f$ above this corner frequency. The magnitude of the noise due to the random transfer of energy across the link to the heat sink (an elementary calculation in statistical mechanics) has the same frequency distribution as the signal. If this so-called phonon noise were the only source of noise, the signal-to-noise ratio would not depend on frequency, and the measurement error could be made arbitrarily small by employing a sufficiently large bandwidth.

In reality, a frequency-independent noise term and a finite detector response time both reduce the signal-to-noise ratio at high frequencies, effectively limiting the usable bandwidth. This state of affairs is illustrated in figure 3.

To understand the bandwidth limitations and calculate the highest energy resolution attainable, we need to consider specific implementations of the calorimeter concept, which, to date, have been based on resistive, capacitive, inductive, paramagnetic, and electron tunneling thermometers. The best energy resolution so far has been achieved with resistive thermometers—specifically, semiconductor thermistors and superconducting transition-edge sensors.

Johnson noise and Joule heating are important effects in resistive calorimeters. An electrical analog of Brownian motion, Johnson noise is frequency-independent voltage noise that scales with the square root of temperature and resistance. Joule heating occurs because the bias current or voltage used to convert a change in resistance to an electrical signal is continually dissipating power in the thermometer. (See figure 4 for representative circuit dia-

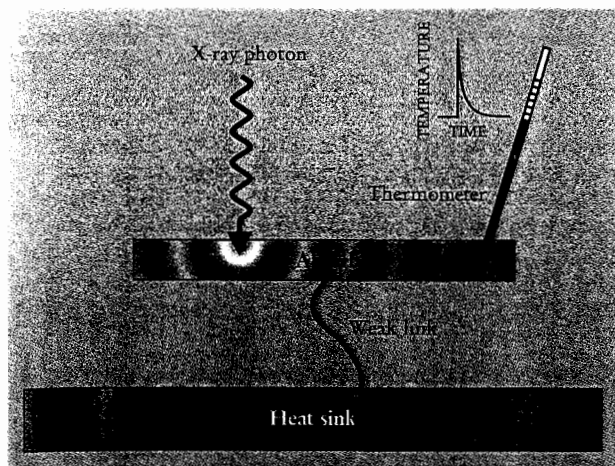


FIGURE 2. AN X-RAY CALORIMETER basically consists of a thermal mass to absorb incident x-ray photons, a thermometer to measure the resulting temperature rise, and a weak link to a low-temperature heat sink that provides the thermal isolation needed to sense a temperature change.

grams illustrating the role of the applied bias.)

These two physical effects lead to the concept of optimal bias. Increasing the bias raises the signal and phonon noise relative to the Johnson noise, extending the useful bandwidth, until the decrease in signal-to-noise resulting from self-heating offsets that gain. Also contributing to the optimization is the increase in heat capacity with temperature.

Fifteen years ago, Harvey Moseley (the US father of quantum calorimetry) and his colleagues⁷ calculated the energy resolution attainable in an ideal calorimeter with a resistive thermometer, following the similar optimization done by John Mather for infrared bolometers.⁸ Moseley obtained an expression for the energy resolution of the form

$$\Delta E = f(t, \alpha, \beta, \gamma) \sqrt{k T_b^2 C_b},$$

where k is Boltzmann's constant, T_b is the temperature of the heat sink, and C_b is the heat capacity evaluated at that temperature.

Writing the reduced temperature T/T_b as t , the heat capacity and thermal conductance functions are $C(t) = C_b t^\gamma$ and $G(t) = G_b t^\beta$, respectively, but the thermal conductance does not appear in the expression for ΔE except through its exponent β . ΔE depends on the resistance through the logarithmic sensitivity α , which is defined by $\alpha \equiv d \log R / d \log T$.

For simplicity, we have not included the definition of $f(t, \alpha, \beta, \gamma)$ above, but, for most practical values of α and t , $\Delta E \propto \sqrt{k T_b^2 C_b} / |\alpha|$. Minimizing the full expression in t determines the optimal bias temperature and the highest resolution for a given detector.

It is assumed that an incident photon changes the temperature so little that it is valid to neglect the temperature dependence of C , G , α , and the noise terms during the pulse. External sources of noise, such as from a signal amplifier, are also assumed to be negligible compared with the intrinsic noise. Though amplifier noise requires careful attention, it can be rendered negligible for resistive thermometers by the right choice of detector resistance and by the use of low-noise junction field effect transistors (JFETs) for high-impedance devices and superconducting quantum interference devices (SQUIDs) for low-impedance devices.

Another property of resistive thermometers, electrothermal feedback (ETF), occurs because the bias power changes as the resistance changes. Consider a thermistor with a negative α that is biased with a constant current I , as in the first example in figure 4. As T increases, R will fall, causing $I^2 R$, the Joule power, to drop. This loss makes the pulse recovery time faster than the value of C/G expected in the absence of feedback. In devices with large $|\alpha|$, this effect can be very important.

Semiconductor thermistors

Doped semiconductor thermistors—in particular, ones made from ion-implanted silicon and neutron transmutation doped (NTD) germanium—were the first thermometers used in x-ray calorimeters. An advantage of using silicon is the existence of well-established micro-machining techniques for fabricating devices in which the active pixels and their links to the heat sink form a monolithic array. NTD germanium's chief advantages are the reproducibility and uniformity of its doping density.

In doped semiconductor thermistors, electricity is conducted by thermally activated charge carriers that hop between localized impurity states through a mechanism called variable range hopping (VRH).⁹ In VRH, the average hopping distance increases as the temperature is lowered. It does so because an electron tends to tunnel farther to a site requiring less change in energy than to a nearby site with a difference in energy that is large compared with that available from the phonon spectrum.

In doped crystalline semiconductors, which have a Coulomb gap in their density of states, VRH produces the resistance law $R = R_0 \exp \sqrt{T_0/T}$, which results in a logarithmic sensitivity given by $\alpha = -1/2 \sqrt{T_0/T}$. T_0 is a constant that depends sensitively on the doping density, increasing from 0 K at the metal-insulator transition to about 120 K at half the critical impurity concentration. R_0 depends on the resistor geometry and only weakly on doping density.

Calorimetrists originally hoped they could use semiconductor thermistors to build a soft x-ray detector with an energy resolution of better than 1 eV (full width-half maximum, FWHM). They based their expectations on being able to operate the calorimeter at temperatures below 0.1 K and on early estimates that the calorimeter's heat capacity could be kept below 0.01 pJ/K. However, those estimates assumed that all noise sources, apart from phonon noise and Johnson noise, were negligible and that resistance was a function of temperature alone.

In fact, actual devices have proved to suffer from nonideal effects that have invalidated these assumptions. For instance, resistance decreases with increasing bias power—even at fixed temperature (nonohmic behavior).¹⁰ And at low frequencies, excess $1/f$ noise is present.¹¹ These two effects limit how small the thermistor can be made, and, hence, its contribution to the heat capacity, since both effects worsen as the thermistor's size is reduced. They also worsen with decreasing doping density, which limits T_0 . Given these effects, the limiting resolution has not yet been determined.

The resolution is not the only property affected; the nonohmic behavior also limits practical device speeds, since higher thermal conductance values require that more power be dissipated in the thermometer to reach the optimal bias temperature.

For a practical semiconductor calorimeter, the total heat capacity should be kept below 0.1 pJ/K, yet the detection area should be about 0.1 to 1 mm². As a result, the selection of an absorber is highly constrained, as follows:

> Insulators and semiconductors have in their bandgaps impurity levels that can trap electrons, thereby forming metastable states that lead to incomplete and variable

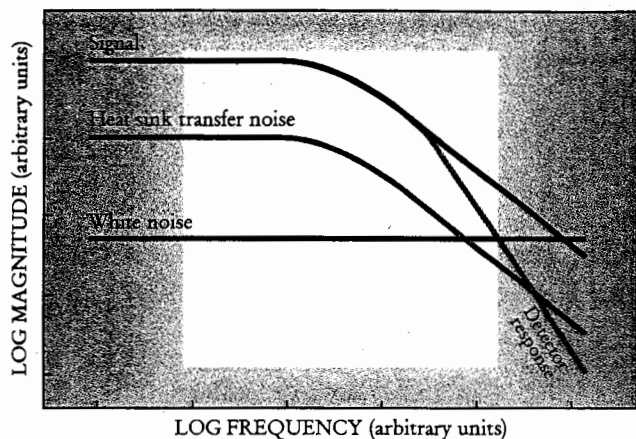


FIGURE 3. AN INSTANTANEOUS DEPOSITION of energy into a calorimeter produces a signal pulse that decays exponentially with time constant τ . In the frequency domain, shown here, the signal (red) is roughly constant until $f \approx 1/2\pi\tau$, after which it falls off as $1/f$. The noise due to the random transfer of energy across the link to the heat sink (blue) has the same frequency distribution as the signal. If this so-called phonon noise were the only source of noise, the signal-to-noise ratio would be frequency independent, and the measurement error could be made arbitrarily small by employing a sufficiently large bandwidth. In reality, a frequency-independent noise term (green) and the effect of a finite detector response time (orange extension from red line) reduce the signal-to-noise ratio at high frequencies, effectively limiting the usable bandwidth.

thermalization.

▷ Normal metals thermalize well, but their large electronic specific heats limit their use to extremely small detectors.

▷ Semimetals, such as mercury telluride, thermalize well, but have relatively high lattice specific heats.

▷ Superconductors with high atomic numbers and high Debye temperatures appear attractive, but thermalization in a superconductor can be delayed by the slowness of quasiparticle recombination at temperatures far below the critical temperature.¹²

Although the search for the optimal absorber for use with semiconductor thermistors is still under way, the best performance so far has been obtained with absorbers made from mercury telluride and tin. Using NTD germanium thermistors and tin absorbers, Ettore Fiorini and his group at the University of Milan currently hold the resolution record for semiconductor calorimeters of 5 eV FWHM at 6 keV. (Fiorini and Tapio Niinikoski pioneered quantum calorimetry in Europe independently of, but at the same time as, Moseley in the US.) Eric Silver and his coworkers at the Harvard-Smithsonian Astrophysical Observatory are achieving comparable resolution with similar kinds of detectors.

Transition-edge sensors

Superconducting transition-edge sensors (TES) operate in the narrow region between the normal and superconducting states of a metal film. (See the second example in figure 4.) For a TES thermometer, the logarithmic sensitivity α can be more than an order of magnitude higher than for a practical semiconductor thermistor.

There are two principal means of developing a voltage (and hence a resistance) across an electrically biased superconducting film undergoing its phase transition. If the superconductor is nonuniform, or has a temperature gradient due to self-heating, the resistance is determined by the size of a normal region that grows with temperature. A voltage can also be generated across a superconductor by magnetic flux flow across the film. Depending on the physical parameters of the film, this voltage generation can be characterized by the nucleation of flux channels, called phase slip lines, over which the superconducting order parameter slips in increments of 2π .¹³

The energy resolution of a calorimeter is determined by the ratio of the signal to the phonon noise and by the useful bandwidth. The use of a thermometer with a higher α increases the useful bandwidth by raising both the signal and the phonon noise above the level of white noise, thereby improving the energy resolution for a fixed heat capacity. (Recall figure 3 above.)

In the small-signal limit, for the same heat capacity,

TES calorimeters therefore significantly outperform semiconductor calorimeters in energy resolution. For x-ray applications, however, the heat capacity of a calorimeter is constrained by the onset of nonlinearity in the detector response to energetic photons.

In both TES and semiconductor thermistors, the sensitivity depends nonlinearly on temperature. For TES calorimeters, however, sensitivity falls abruptly once the normal state has been reached. To keep the temperature excursion due to the absorption of an x-ray photon from exceeding the dynamic range of the detector, the heat capacity C of a TES calorimeter must be increased by a factor proportional to α . Since, for large values of α , the resolution scales as $\sqrt{C/|\alpha|}$, the fundamental limits on the energy resolution of a TES x-ray calorimeter are similar to the original predictions made for semiconductor calorimeters.¹⁴

In fact, the main advantage of a TES calorimeter lies in such practical issues as the design flexibility that a large heat capacity budget allows. TES calorimeters can use materials with high specific heats, such as normal metals, that thermalize deposited energy quickly and efficiently, increasing the probability of actually achieving the predicted resolution.

The concept of optimal bias acquires an interesting twist when applied to a TES calorimeter. Clearly, for a particular superconductor, the bias temperature must lie within the superconducting transition. With the use of the proximity effect, however, it is possible to engineer a TES thermometer with a critical temperature T_c at any convenient temperature. To achieve this feat, a bilayer is made by combining a thin superconducting film that has a T_c much higher than is useful with a thin film of a normal metal to push that T_c down. The value of T_c is determined by the thicknesses of the two layers, the properties of the two films, and the resistance of the interface.

Given a practical refrigerator temperature and the ability to obtain a particular value of α at any T_c , the choice of T_c for the highest energy resolution is determined by the same bias optimization as for any resistive device. In designing a TES calorimeter for an expected maximum incident photon energy, we choose a value for the heat capacity C that avoids large-signal nonlinearity, yet fully uses the linear part of the dynamic range. It works out that, for a given α and a maximum photon energy E_{\max} , we choose C such that $C \propto E_{\max}\alpha/T_c$. This choice counteracts the usual heat capacity penalty for operating at a higher bias, and it results in an optimal bias temperature that is higher than that for the semiconductor devices and in much slower degradation in resolution with temperature above that optimum. The much larger scale of the heat capacity makes it practical to design a TES calorimeter that

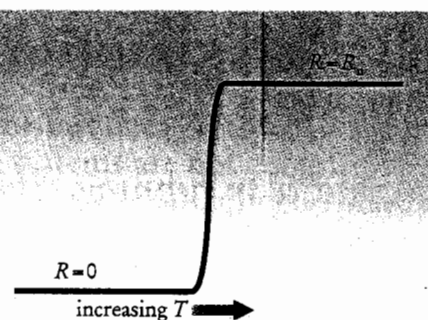
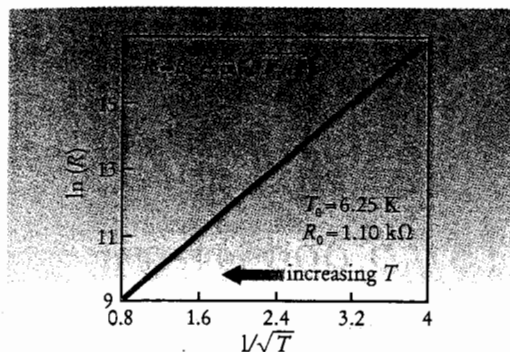
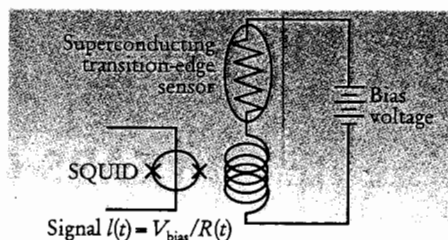
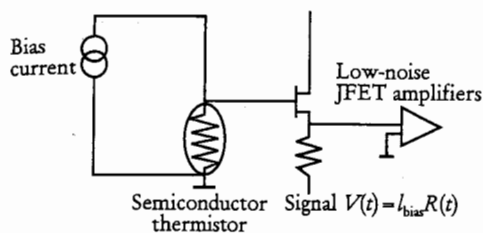


FIGURE 4. SCHEMATIC bias circuits for two kinds of resistive thermometer. A semiconductor thermistor (left) shows a decrease in resistance with increasing temperature, whereas a superconducting transition-edge sensor (right) increases in resistance with increasing temperature within its superconducting transition. The choice of constant current bias for the former and constant voltage bias for the latter results in both devices exhibiting negative electrothermal feedback—that is, the Joule power ($I^2 R = V^2/R$) decreases as the temperature is raised.

has a lower heat capacity, but is biased at a higher temperature.

What is there to gain from designing a TES with a higher bias temperature? Operating in such a way increases the magnitude of the electrothermal feedback (already substantial because of the large α) because more bias power is required, which increases the dynamic range of the feedback reduction in power. With negative ETF, the TES will self-regulate within its transition, and increasing the bias temperature relative to that of the heat sink improves the stability of the bias point.

Increasing the ETF also makes the temperature recover faster, and it is useful to do that until the corner frequency of the thermal response matches the useful bandwidth of the device. The temperature can then be recovered in a time close to the limiting thermalization and diffusion times, thereby raising the maximum x-ray flux that can be spectroscopically analyzed before the onset of pileup. (Pileup is what happens when photons arrive at a detector faster than they can be individually read out.)

The fabrication of TES x-ray calorimeters by one of us (Irwin) and the National Institute of Standards and Technology group with energy resolution (FWHM) of 2.4 eV at 1.5 keV and 4.5 eV at 6 keV and at count rates higher than 400 counts per second is a significant advance for high-throughput x-ray spectroscopy. These results have been obtained with aluminum-silver and molybdenum-copper proximity-effect TES thermometers fabricated on silicon nitride membranes.

The future of x-ray calorimetry

Although x-ray calorimeters have already been deployed on sounding rockets making brief flights (see the box on page 33), the first major x-ray calorimeter instrument, XRS, will be launched early next year on the Japanese-US Astro-E spacecraft. Once in orbit, XRS will inaugurate the field of high-resolution, high-throughput astrophysical x-ray spectroscopy.

Looking further ahead, NASA and the European Space Agency are already investigating future mission concepts—Constellation-X and XEUS, respectively—that plan to include an imaging calorimeter with 2 eV resolution at 6 keV and about 1000 pixels. Since such technology does not exist yet, condensed matter physics and materials engineering will feature prominently in the development of the two missions.

In this article, we have tried to show that continuing research in low-temperature physics leads to a greater understanding of high-temperature astrophysics. Questions about the universe place demands on detectors that, in turn, pose questions that condensed matter physicists can—or strive to—answer. Adaptations of the resulting spectrometers will be useful tools for fields of research beyond astrophysics. Physics thrives on this type of feedback.

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