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High resolution microcalorimeters as detectors for inelastic scattering (invited)

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Small thermal calorimeters operated at temperatures near 100 mK are sufficiently sensitive to detect single photons, measuring the deposited energy to accuracies approaching 1 eV. A test device has demonstrated 17-eV FWHM for 6-keV x rays—almost an order of magnitude better than a conventional Si(Li) solid state detector. Further improvement requires the development of monolithic fabrication techniques to reduce the parasitic heat capacities introduced in assembling a discrete device. Current technology should permit the construction of a detector about 1/2 mm^2 with good efficiency for 35-keV x rays and a resolution near 25 eV. The ultimate resolution achievable in practice and the tradeoff of collecting energy for resolution depend primarily on the low-temperature specific heat of the material used to absorb the x rays and efficiently thermalize their energy. Finding the optimum absorber is particularly important at higher photon energies. We are studying absorbers that may offer considerably improved performance.

I. X-RAY SPECTROSCOPY FOR INELASTIC SCATTERING

One of the only direct ways to determine the structure of the energy and momentum states available to electrons in a sample is by scattering experiments. In one type of these experiments, photons of known energy and momentum are used to scatter electrons into excited states. The distribution of energies and momenta of the photons scattered out of the sample are measured and used to infer how much of the initial energy of the photon beam was taken up by electrons. When the incident photons are from the x-ray region of the spectrum, the electrons may respond collectively (plasmon scattering), may be excited into a discrete higher state (Raman scattering), or may be excited into the continuum of free states (Compton scattering).

The cross sections for these inelastic scattering processes are small, so that practicable experiments require incident beams of relatively high flux. In the past, high flux, monochromatic beams were produced either by using large radioactive sources or by using the characteristic K lines from powerful x-ray tubes. Even with the brightest available of these types of sources, the time required to carry out one of these scattering investigations is often prohibitive. The high luminosities possible with the new generation of synchrotron light sources has renewed interest in x-ray scattering-experiments. In addition to the higher fluxes, the tunability and high collimation of synchrotron sources allows studies not possible with other types of x-ray sources.

Since beam time will continue to be a scarce resource for the foreseeable future, there is still reason to measure the distribution of the scattered x-rays as efficiently as possible. In the past, one could choose either a conventional double-crystal Bragg spectrometer, which offers high energy resolution at low throughput, or a solid state detector, which has very high efficiency but at a fairly low energy resolution. A novel type of x-ray spectrometer, the microcalorimeter, which combines high efficiency with higher energy resolution than can be obtained with the existing forms of nondispersive spectrometers, may offer an attractive alternative for x-ray scattering experiments.

II. CALORIMETER CONCEPTS

Our development work, which has primarily focused on the use of microcalorimeters as x-ray spectrometers for astrophysical applications, has been described in a series of articles.[1-6] We will merely present an overview of that work, with some emphasis on what future developments might make possible for terrestrial applications. The basic principle behind the microcalorimeter is implied by its name; the energy of the x ray is determined by measuring the temperature rise obtained when that energy is converted to heat in a mass of known heat capacity. As the energy of an individual x ray is quite small relative to the more familiar energies of macroscopic objects, x-ray calorimetery requires very small heat capacities and very sensitive thermometers. The fundamental thermodynamic limitation on the resolution of a calorimeter is set by the fluctuations in the heat flowing to the thermal reservoir, which can be thought of as the thermal analog to electrical shot noise. The corresponding uncertainty these fluctuations place upon the determination of the energy of an x ray increases with the square root of the heat capacity of the device and increases linearly with the operating temperature. At low temperatures, the heat capacity of crystalline dielectrics varies as T^3, and so very small heat capacities can be obtained by cooling the detectors to very low temperatures. We have chosen to operate the current generation of calorimeters at temperatures around 0.1 K. Those not familiar with the technology might imagine that such low temperatures would be dauntingly difficult to achieve, but, in actuality, the cryogenic techniques are relatively well developed and straightforward.

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There are several cryogenic thermometry techniques which could be used to transduce the temperature rise of the x-ray absorber. We have chosen to concentrate our efforts on semiconductor thermistors, operating in a conduction mode known as phonon-assisted hopping. The thermistor is created by doping a region of the semiconductor with impurities at a density such that the overlap of the potential wells between sites is very small, and the charge carriers cannot easily move from impurity to impurity. An interaction with phonons in the solid provides the extra impetus needed for a carrier to make the hop to the next impurity site. In actuality, the conduction probably involves near-simultaneous hops by many carriers. When the energy of the x-ray is thermalized, there are more phonons present to assist the electrons across the thermistor, and the resistance drops. One figure of merit for the responsivity of a thermistor is $\alpha \equiv -d \log R / d \log T$, and our thermistors typically have $\alpha \approx 6$.

We have chosen to construct most of our test devices out of silicon, which has several advantages for this work. Silicon has a very high Debye temperature, so the undoped regions of the detector contribute very little heat capacity. We use fairly standard integrated circuit techniques for much of the construction, and by using silicon one can take advantage of the vast experience of the semiconductor industry. High purity silicon is readily available, as is the apparatus for handling it. The doping of our thermistors is accomplished by implanting ions into the silicon, and the techniques and parameters for implanting silicon are well established. We are planning to create arrays of calorimeters using micromachining techniques, where elaborate mechanical structures can be created using selective etchants. While these processes are under development for several materials, the field is most mature for the processing of silicon.

The absorbing body containing the thermistor must be isolated by a weak thermal link from a heat sink. While the theoretical prediction for the energy resolution of a perfect calorimeter does not depend upon the thermal conductance of this link,[1] real-world considerations limit the range of desirable conductances. Most of the calorimeters we have tested, so far, have been small (circa $0.5 \times 0.5 \times 0.01$ mm) silicon chips supported on thin aluminum bonding wires. At low temperatures, the aluminum becomes superconducting and the thermal conductance drops to the required range. Superconducting aluminum has a very low heat capacity so that not much of the x-ray energy is lost to heating the leads. While the best energy resolution we have achieved so far was obtained with a wire suspended device, this suspension technique is not well suited to creating arrays. Recent work has focused upon studies of the thermal conductivities that can be obtained using various microfabrication techniques.

In addition to the above elements, a microcalorimeter must have one more component before it will be a useful x-ray spectrometer. A constant fraction, preferably 100%, of the x-ray energy must be thermalized in a time short compared to the measurement interval. We have found that rapid, complete thermalization does not occur in some materials, and in particular, does not occur in silicon. When an x ray is absorbed into a semiconductor, much of the energy initially goes into breaking hole-electron pairs. While many of these charge carriers quickly recombine and create phonons, a few become trapped on long-lived impurity sites in the band gap, and are lost to the measurement. Fluctuations in the number of trapped carriers gives a large uncertainty to the determination of the energy, and so the x-ray must be absorbed in a material which will consistently thermalize its energy.

Many of the materials which will rapidly thermalize x rays, such as normal metals, cannot be used since their heat capacities would be too large if they were applied in the volumes required to stop most of the x-rays. To avoid the energy losses from carriers getting trapped on impurities near the edges of a relatively wide band gap, we have been investigating systems with small or nonexistent gaps. So far, the most promising results have been obtained with the ternary alloy HgCdTe. In this alloy the band gap can be tuned from 1.5 to 0 eV by changing the relative concentrations of Hg and Cd. So far we have attached absorber samples to the calorimeter bodies with small amounts of epoxy. This is not optimal since soft and disordered materials such as epoxies have very high heat capacities. Techniques are being developed for depositing HgCdTe directly upon silicon, which would avoid the heat capacity penalty incurred by using adhesives.

The fundamental thermodynamic limitation on the resolution of an ideal calorimeter is determined by its heat capacity, which can be estimated by summing the contributions from the various components. Estimates of this limiting resolution for 0.25×1.0 mm calorimeters being designed for an x-ray spectrometer for astrophysical use[6] are under 10-eV FWHM. We have not actually brought together the implanting, etching, and absorber deposition technologies, so do not yet know how closely we can approach this fundamental limiting resolution. We have identified several nonideal effects, which add (in quadrature) to the thermodynamics limited resolution, and our experimental efforts have provided some upper limits on the magnitudes of these contributions.

III. PRESENT STATUS

The best resolution obtained so far, was with one of our aluminum suspended calorimeters with a small (4×10^{-4} mm^3) piece of $Hg_{0.8}Cd_{0.2}Te$ epoxied to it (see Ref. 5 for experimental details). This relatively crude device has an energy resolution of 17 eV (FWHM) for 5.9-keV Mn $K\alpha$ x rays.

As an example of the merits of microcalorimeter x-ray spectrometers, Fig. 1 compares an x-ray spectrum taken with a Si(Li) spectrometer with one taken with another micro-calorimeter. The source was a sample of Tb^{158} supplied by R. Raghavan of Bell Labs, Murray Hill. Some of the lines, such as the ones labeled Ni, are not directly from the source, but are from fluoresence or escape peaks, and were not produced in both experimental setups. The absorber on the calorimeter was not designed to be opaque at these energies, so that the relative strength of the lines in the calorimeter spectrum decreases with photon energy. So, while the experiment does not permit a direct comparison of the spectra, the

impressive improvement calorimeters can offer for nondispersive spectroscopy is still evident.

It takes several hundred microseconds for the temperature to return to near its equilibrium value in these wire suspended devices. While the limit this imposes on the counting rate is reasonably high compared to fluxes from the majority of astrophysical x-ray sources, it is probably disappointingly small for many scattering experiments. The fall time of the pulses can be decreased by decreasing the ratio of the heat capacity of the detector to the conductance of the thermal links. Making the body of the detectors smaller will reduce both their heat capacity and their active area, which will decrease the counting rate for a constant flux beam. The control over the aspect ratio and configuration of the etched support legs should allow tailoring of their thermal conductance. The conductivity cannot be increased without limit though, since high conductance requires higher bias power for optimum responsivity, and measuring our thermistors with large voltages has been found to reduce their resistance, and hence their responsivity. Pulses riding on the tails of previous pulses can be analyzed, but at some decrease in resolution. This analysis becomes much more difficult if the detector is operating in the nonlinear response range.

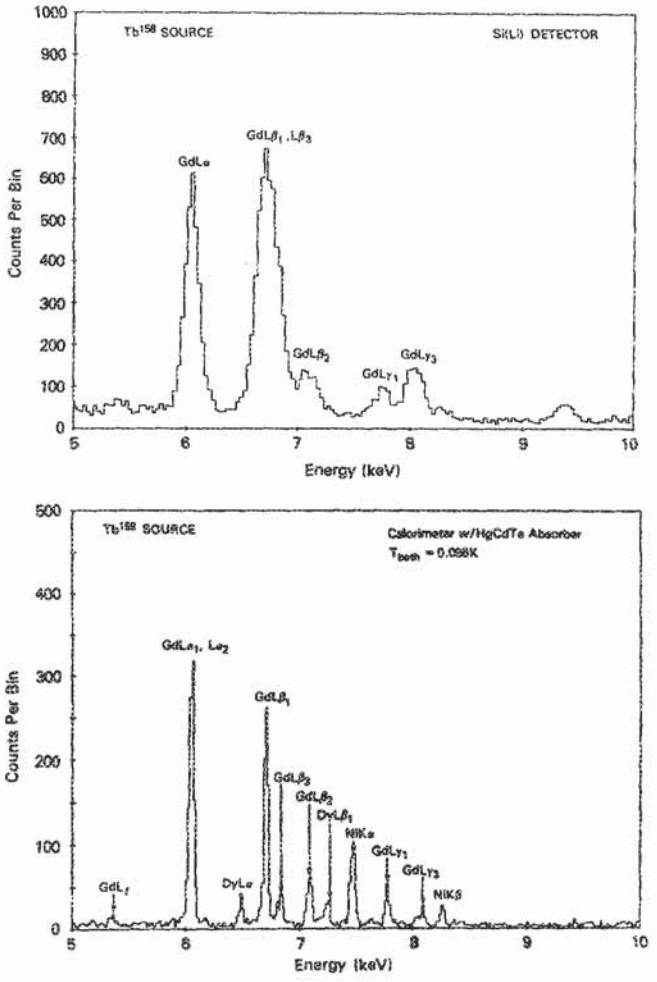

FIG. 1. Pulse height spectra from Tb^{158} illuminating a conventional Si(Li) spectrometer and a calorimeter. There was nickel present only in the setup with the calorimeter, and thus those lines do not appear in the Si(Li) spectrum.

The noise-limited resolutions obtained with these devices are fairly close to the limiting resolutions that one predicts from the heat capacities, which indicates that the magnitudes of the nonideal effects are fairly small. While still better resolution could be achieved by improving details of the wire suspended construction, most of our recent efforts have been directed toward examination of various thermalization materials and toward construction of arrays of lower heat capacity detectors, using monolithic construction techniques.

IV. FUTURE DEVELOPMENTS

Monolithic devices offer several advantages over composite designs. Monolithic calorimeters will have lower heat capacities than can easily be obtained with previous construction techniques. For example, the thermal links in an all silicon device should not have the excess heat capacity that can arise from small normal-state regions around magnetic impurities in superconductor links. The etching techniques allow the construction of smaller calorimeters than could be easily handled during the assembly of a composite unit. Many applications require, or can benefit from, larger sensitive areas than an individual detector, so it is natural to try to construct arrays of calorimeters. It is cumbersome to construct such arrays by hand, so several microfabrication techniques are being examined for feasibility.

Figure 2 shows the construction of a linear array of 12 calorimeters. The 0.25×1 mm calorimeter bodies, and their four narrow ($2.3\ \text{mm} \times 20\ \mu\text{m}$) support legs are patterned out of a section of the wafer which is initially thinned to a thickness of $\sim 12\ \mu$m. The test mask set has various sizes of thermistors to study the tradeoff between the heat capacity of the implant and the magnitude of excess noise and nonohmic conductivity effects. The protrusions from the sides of the support beams were created to study the reduction in thermal conductance that can be obtained by increasing phonon scattering. We will be testing such detectors in the near future.

V. CALORIMETER X-RAY SPECTROMETER SYSTEMS

The remainder of the equipment required for a complete x-ray spectrometer is reasonably simple. The thermistors are put in series with load resistors with about the same resistance ($\sim 50\ \text{M}\Omega$). These load resistors must be mounted in the cryostat to keep their Johnson noise small. A bias voltage is applied to the top of this voltage divider. The high impedance of the signal node cannot tolerate much load impedance, so it is buffered through a JFET source follower. Since the high impedance line is very susceptible to capacitive pickup, the JFET should be located as physically close to the detector as possible. This adds some cryogenic complexity since most of the available types of quiet JFETs cannot run at temperatures below ~ 100 K. In the future, germanium JFETs may become available which could operate at much lower temperatures. The output from the follower is then brought out of the cryostat to a warm amplifier. At present

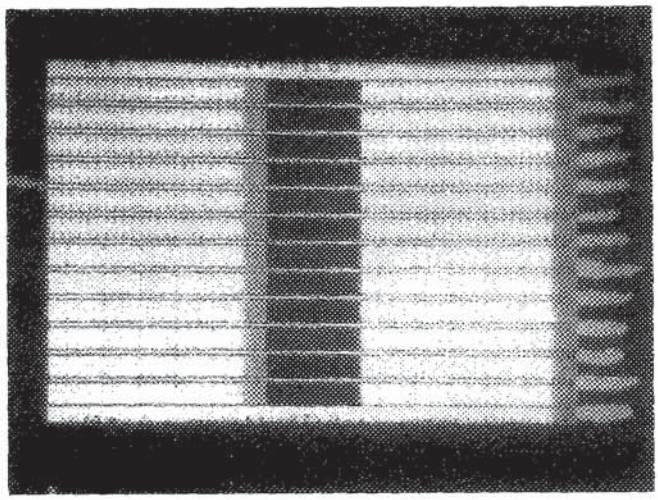

FIG. 2. Micrograph of a 1×12 array of monolithic silicon calorimeters. The rectangular calorimeter bodes are $0.25 \times 1.0\ \text{mm} \times 12\ \mu\text{m}$. The support legs are 3 mm long and have a cross section of $12 \times 20\ \mu\text{m}$.

we digitize the amplifier output and store the pulse records for digital signal processing on a mainframe, although we are developing a stand-alone system to produce pulse heights directly from the ADC output.

The cryostat must have windows which will allow the x rays to penetrate down to the detectors without transmitting lower energy radiation which will heat up the detectors and increase the background noise level. The engineering of these windows for a Compton scattering experiment, where the scattered x rays may have energies near 40 keV, is relatively simply compared to our astrophysical applications, where, while the windows are not required to hold against atmospheric pressure, they must transmit 300-eV x rays while blocking out relatively large UV fluxes. A spectrometer for scattering measurements could use a relatively thick beryllium window for the pressure window, with a thinner, cooled internal window to block the thermal radiation from the room temperature window.

Several different refrigeration techniques are suitable for cooling calorimeters. Temperatures near 0.3 K can be achieved in a cryostat which cools by pumping on He^3. We have been using a commercial dilution refrigerator, which cools by using the entropy of mixing He^3 into He^4, for much of our laboratory work. In addition, we have recently begun using an adiabatic demagnetization refrigerator, which uses the relaxation of the spin system of a paramagnetic salt for cooling.

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