ADVANCES TOWARD HIGH SPECTRAL RESOLUTION QUANTUM X-RAY CALORIMETRY

S.H. Moseley, R.L. Kelley, R.J. Schoelkopf and A.E. Szymkowiak,
Laboratory for High Energy Astrophysics,
NASA/Goddard Space Flight Center, Greenbelt, MD 20771
and
D. McCammon and J. Zhang,
Department of Physics,
University of Wisconsin, Madison, WI 53706

Abstract

Thermal detectors for X-ray spectroscopy combining high spectral resolution and quantum efficiency have been developed. These "micro-calorimeters" measure the energy released in the absorption of a single photon by sensing the rise in temperature of a small absorbing structure. The ultimate energy resolution of such a device is limited by the thermodynamic power fluctuations in the thermal link between the calorimeter and isothermal bath, and can in principle be made as low as 1 eV. The performance of a real device is degraded due to noise contributions such as excess (1/f) noise in the thermistor and incomplete conversion of energy into phonons. We report here some recent advances in thermometry, X-ray absorption and thermalization, fabrication techniques, and detector optimization in the presence of noise. These improvements have allowed us to produce a device with a spectral resolution of 17 eV FWHM, measured at 6 keV.

I. Introduction

The use of micro-calorimetry as a means of measuring the energy of individual events with high precision has been successfully demonstrated[1-6], and has applications to a range of problems. For the past several years, our group has been developing micro-calorimeters which can measure small amounts of energy to a precision of $\approx 10^{-17}$ joules (corresponding to about 10 eV). Our main emphasis has been on developing micro-calorimeters as the sensing elements in non-dispersive X-ray spectrometers for use in astronomical observations. Cryogenic calorimeters with low-energy thresholds will also be useful in nuclear and particle physics, and as detectors for weakly-interacting particles, massive neutrinos, or other dark matter candidates.[1,2]

An ideal calorimeter consists of a small, low-heat-capacity mass connected to a heat sink by a thermal link. In our case, the temperature of the mass is monitored by recording the voltage drop across a thermistor with a large temperature coefficient of resistance. In the small signal limit, the change in voltage is proportional to the change in temperature and thus to the total energy absorbed. The most fundamental noise source in such a system is due to the thermodynamic power fluctuations in the thermal link connecting the absorber and the bath. The total noise of a properly optimized calorimeter is

$$\Delta E = \xi (kT^2 C)^{1/2} \qquad (1)$$

where ξ is a parameter depending on thermal characteristics and is typically ~ 2, k is Boltzmann's constant, T is the temperature of the heat sink, and C is the heat capacity of the mass. The noise consists of approximately equal contributions from transducer Johnson noise and phonon noise. If the device is constructed of dielectric materials, the heat capacity will vary as T^3, and thus the noise can be reduced dramatically by operating at low temperatures. For the devices we are developing, it should be possible to reduce this phonon noise to less than 1 eV full-width half-maximum (FWHM) by operating at temperatures below 0.1 K.

In any real detector, however, there are additional noise terms due to the thermistor which transduces the temperature, the amplifier for the transduced signal, and any fluctuations in the conversion of the X-ray to phonons. Furthermore, there may be effects which limit the sensitivity of the thermometer. We discuss some of these non-ideal effects and steps to reduce their importance in Section II. More sophisticated fabrication methods and near-term plans for further improvements of these devices are described in Section III.

II. Experimental Results

Following the realization that infrared bolometer techniques could be used to measure the energy of single X-ray photons, several proof-of-concept devices were fabricated and tested.[2] These tests demonstrated that the calorimetric approach was viable for X-ray spectroscopy, provided that devices with sufficiently low heat capacity could be constructed.

Early tests with relatively low heat capacity Si calorimeters suspended on Al wires (25 μm x ~3 mm) showed that there was a large additional noise term that limited the energy resolution but did not increase the thermal noise. In particular, one device was tested that had a measured baseline noise with a width of 11 eV, while the energy resolution for 6 keV X-rays was found[3] to be ~ 250 eV.

It was suspected that the additional broadening was due to variations in the amount of energy being converted to heat from event to event. When an X-ray is

0018-9499/88/0200-0059$01.00 © 1988 IEEE

absorbed in silicon, much of the energy initially goes into breaking electron-hole pairs. Even in high-purity silicon, the relatively wide (1.2 eV) band-gap contains many impurities that can serve as trapping sites for some of the electrons or holes liberated by the X-ray. Simple statistical fluctuations in the amount of charge produced cannot explain the large effect seen. The additional broadening probably depends on whether charge traps on shallow impurities in the bulk of the Si, or on deeper traps near the surface of the crystal. Therefore, X-rays which are absorbed near the surface release systematically more energy than those absorbed in the bulk of the Si. We thus began a search for materials that would convert X-rays to phonons with high efficiency. This was done by attaching small samples of materials with low heat capacity and small or nonexistent band-gaps (e.g., metals, semimetals, narrow-gap semiconductors, and superconductors) to working detectors. It was found that some materials can convert essentially 100% of the X-ray photon energy into heat.[4-6] However, in these tests the material was attached to the calorimeter with a high-specific-heat adhesive which contributed greatly to the heat capacity.

Several tests have now been carried out using the narrow gap semiconductor $Hg_{1-x}Cd_xTe$ (x = 0.2, equivalent to a band-gap of ≈ 60 meV) as an X-ray absorber. A pulse-height spectrum was obtained[5] with an Al-suspended Si calorimeter operated at a cold-plate temperature of 0.098 K. [A description of the pulse-height determination procedure is given in the Appendix.] The absorber had a volume of 6 x 10^{-3} mm^3, while the Si substrate had a size of 0.5 x 0.5 x 0.1 mm, and the Al leads had a diameter of 25 μm and a length of ~ 3 mm. The detector system was exposed to an Fe^{55} source and to X-rays produced by fluorescing a Mg target with alpha particles from a ^{244}Cm source. The energy resolution (FWHM) achieved on the 6 keV Mn lines was 35 eV. The width of the baseline was determined by taking samplings of the output signal asynchronously with the X-rays and is ~33 eV, which implies a residual noise broadening at the level of ~11 ± 5 eV. This could be due to some remaining charge trapping in the HgCdTe, variations in the pulse height with position across the detector, or intrinsic pulse shape changes.

In Figure 1 we show a spectrum obtained with this same detector exposed to a ^{158}Tb source. The Tb isotope decays 82% of the time by the capture of an L-shell electron into Dy and 12% of the time by beta emission into Gd.[8] The source was obtained courtesy of R. Raghavan and illustrates the power of cryogenic spectroscopy for laboratory spectra of weak sources. Because of the large atomic number of the HgCdTe, the quantum efficiency of the device was nearly 100% over the energy band shown.

Despite the comparatively large volumes and heat capacities of the HgCdTe, the Si substrate, and the epoxy used to attach the HgCdTe, the energy resolution of this detector was not limited by thermodynamic fluctuations, but by a combination of excess low frequency noise ("1/f noise") and a loss of signal amplitude due to a reduction of thermistor sensitivity with increasing bias power.

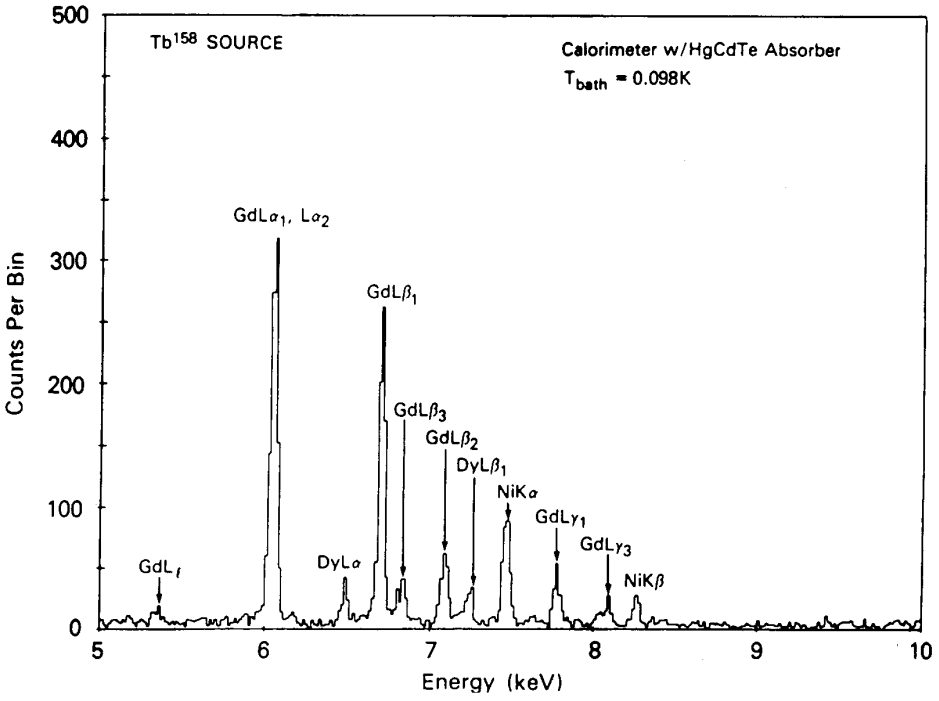

Figure 1: Spectrum of "33 eV" calorimeter exposed to ^{158}Tb source, showing X-ray lines from Gd and Dy L-lines, and K-fluoresence lines from a Ni aperture excited by K and nuclear lines from the Tb.

The loss of responsitivity could plausibly result from either or both of the following effects. First, a thermal impedance might exist between the lattice and the electrons in the thermistor. In this picture, the electrons would be at a higher temperature than the lattice, thus making the thermistor less sensitive to changes in the lattice temperature. This can be modelled by allowing the electrons and the lattice to have separate temperatures, and solving the heat flow equations for the two component system. In another model, the sensitivity of the thermistor would be reduced by an "electric-field effect," in which the applied bias voltage alters the potential near impurity sites in the semiconductor and increases the tunneling rate. This modifies the expression for the electrical conductivity of the thermistor with a term that depends on applied electric field :[10,11]

$$R(T) = R_0 \exp\left(\sqrt{T_0/T}\right)\ G(T,\varepsilon) \qquad (2)$$

where

$$G(T,\varepsilon) = \frac{e\varepsilon\lambda/kT}{\sinh(e\varepsilon\lambda/kT)} \qquad (3)$$

and k is Boltzmann's constant, e is the electron charge, ε is the electric field, and λ is a parameter with dimensions of length, known as the "hopping range." Both of these nonlinearities, however, can be reduced by constructing a device that has a lower thermal conductance so that the optimal bias power is reduced.

We have found that the level of excess noise at low frequencies varies inversely with the net concentration of the thermistor implant.[6] By changing the aspect ratio of our ion-implanted thermistors we can achieve the correct electrical resistance (~ 50 MΩ) at the operating temperature (~ 100 mK) while employing a higher net implant concentration. We fabricated a variety of thermistor geometries on 0.5 x 1.0 mm Si chips 75 μm thick. With the doping densities achieved in a recent implant run we found the 80 by 800 μm thermistors to give a resistance of 40 MΩ at ~80mK. A small (4 x 10^{-4} mm^3) piece of HgCdTe was attached to one of these chips, which was then suspended by two lengths of 0.7 mil (18 μm) diameter Al wire. As desired, the 1/f noise of this detector was substantially smaller than in our previous attempts. To illustrate the signal-to-noise ratio obtained with this detector, the inset to Figure 2 shows the response of the detector to a single 5.9 keV X-ray (no averaging or filtering was used).

Figure 2 shows the pulse height spectrum of an ^{55}Fe source obtained with this device. The spectral resolution of this detector is sufficiently good that it is necessary to include the $K\alpha_1$ and $K\alpha_2$ components of the

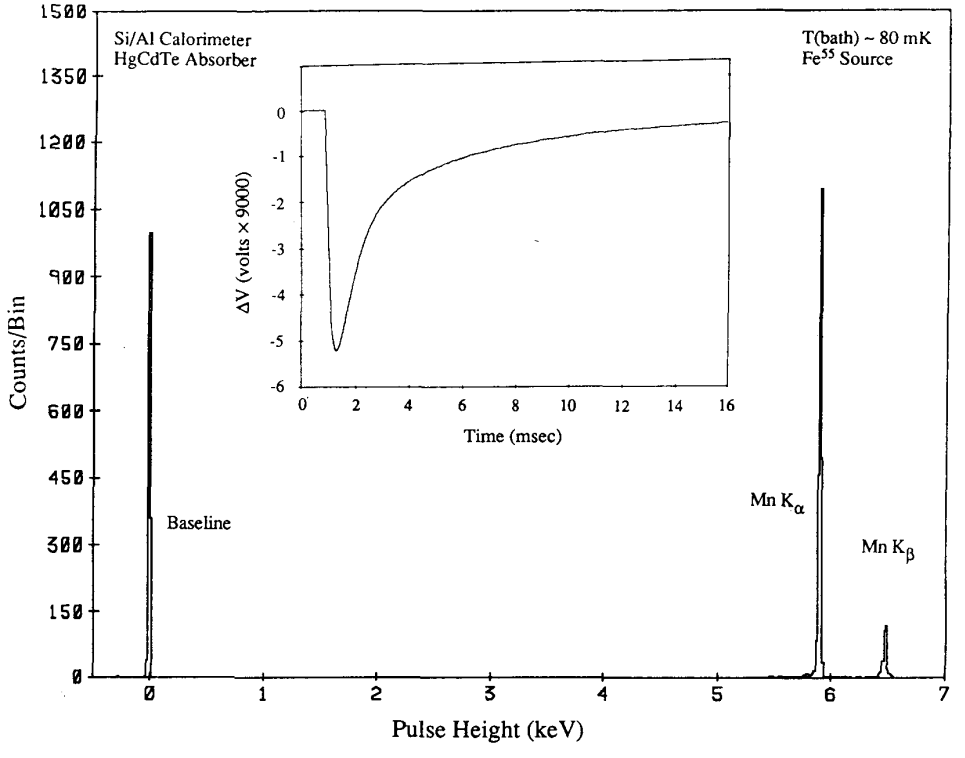

Figure 2: Pulse-height spectrum of Si/HgCdTe/Al calorimeter with high net-concentration implant designed to reduce excess noise. The resolution at the MnKα lines is 17.4 eV FWHM. The baseline width is 13eV, implying about 11eV of remaining excess broadening. Inset shows unfiltered single X-ray pulse.

Mn Kα"line" (which are separated by only 11 eV) in order to obtain the best-fit width (FWHM) of 17.4 eV (see Figure 3). This represents a factor of two improvement over the energy resolution of the device described above, and is sufficiently good to permit many important spectral measurements in high-energy astrophysics.

The baseline width for this device is ~13.5 eV, which implies an excess broadening of 11 ± 1 eV. This is consistent with that obtained for the 35 eV detector, and supports the notion that the excess broadening results from charge trapping in the HgCdTe.

The thermal conductivity of the Al leads was evaluated by fitting the I-V curves of the detector with a model which assumes a temperature dependence of the resistance given by eqs. (2) and (3), and a power law dependence on temperature for the thermal conductance of the link. Using this model, we estimate the conductance to be ~5 x 10^{-11} W/K at 100 mK. This value is consistent with that obtained from published measurements for the thermal conductivity of Al at these temperatures.[7] Using this conductance and an effective pulse time constant of 1 ms, we obtain an effective heat capacity of ~ 5 x 10^{-14} J/K. This is consistent with an estimate calculated by summing the heat capacities of the individual components (see Table 1). More accurate measurements are required, however, and must be carried out using separate heaters and thermistors on the detector in order to avoid difficulties associated with the thermistor nonlinearities.

III. Ongoing Efforts:

The optimization of a micro-calorimeter in the presence of nonideal effects requires accurate empirical data on the scaling of these effects with temperature,

Table 1: Thermal Budget for 17eV FWHM Composite Calorimeter

Component	Volume (mm^3)	Heat Capacity (J/K) at 100 mK
Si	6.2 x 10^{-2}	3.5 x 10^{-14}
HgCdTe	4.0 x 10^{-4}	6.8 x 10^{-15}
Ion implant and contacts	1.0 x 10^{-5}	1.0 x 10^{-15} a
Al leads*	6.6 x 10^{-4}	5.0 x 10^{-16}
Epoxy	1.8 x 10^{-6}	5.5 x 10^{-15} b
Total		5.4 x 10^{-14}

*Heat capacity calculated assuming Debye contribution only.
[a]see Reference 1
[b]Epotek H-74, see Reference 9

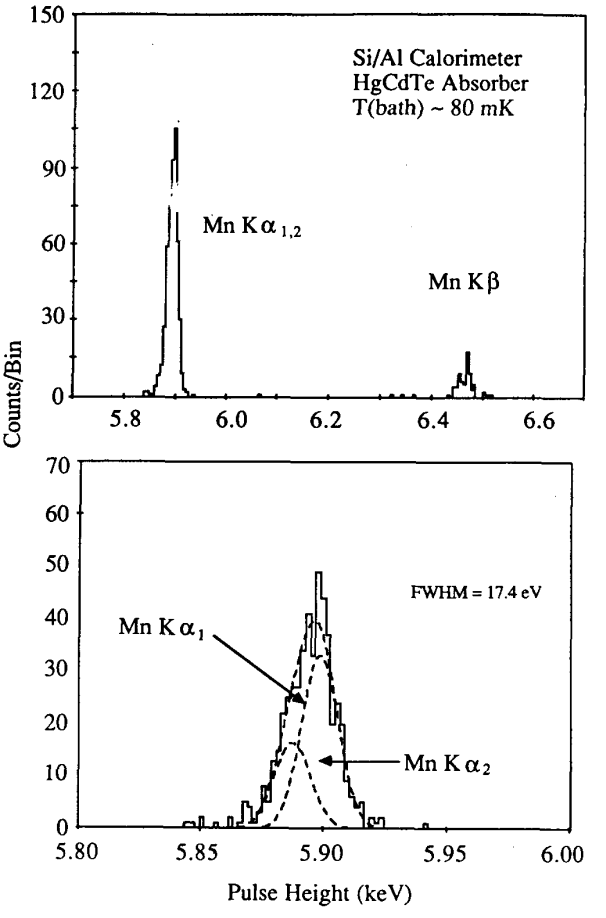

Figure 3: Detail of ^{55}Fe spectrum taken with "17 eV" calorimeter, showing the two components of the fit to the Mn Kα line.

thermistor impurity concentration, and thermistor volume. In the design of a micro-calorimeter, the thermal conductance is closely coupled to the thermistor optimization. Reductions in thermal conductance from the current levels will substantially reduce the nonideal effects in the thermistor, but will increase the recovery time constant unless the overall detector heat capacity is also reduced.

Our immediate goal is thus to complete a numerical model for a micro-calorimeter which includes all the known factors which affect the system performance. We have reasonably complete data on these nonideal effects in a variety of thermistors operated near 0.3 K, but we have only measured a few samples at 0.1 K. We intend to extend this characterization down to 0.1 K for thermistors having a range of impurity concentrations to provide input data for our models.

For NASA's Advanced X-Ray Astrophysics Facility, individual detector areas of ~0.25 mm^2 are required, and the detectors must be thick enough to be at least partially opaque to 8 keV X-rays, so the minimum dimensions are fixed. The calorimeters must be designed so that they can be configured as an array without large "dead" areas,

and, at the same time, be rugged enough to survive launch into orbit. We are therefore interested in achieving the minimum possible heat capacity within these constraints.

While we have achieved good results with the silicon/aluminum composite devices described above, we believe that the best way to meet the performance requirements of the flight instrument is with monolithic detectors. We have been investigating several "micro-machining" techniques for producing linear arrays of detectors, complete with thermal and electrical links, from one silicon wafer. In one approach, anisotropic etchants are used to form the sensing mass and the legs. The thermal conductance of the leads of our first attempts at monolithic devices was disappointingly large. The phonon mean free path derived from this conductance was found to be much larger than the traverse dimensions of the link, implying that phonons were being specularly reflected at the surface of the thermal link.

We are investigating two approaches in order to reduce the thermal conductance. In one approach, we explicitly etch small scattering structures on the thermal link to reduce the phonon mean free path to ~50 μm. We expect this to reduce the conductance by about the necessary factor of 10. In another approach, the sensing mass would be supported by micro-bridges constructed of poly-silicon. One advantage of this technique is that the bridges can be made quite short (~ 100 μm) which permits the sensing elements to be placed close together. Even with the high aspect ratio the bridges must have in order to yield a sufficiently small thermal conductance, they still possess excellent strength and rigidity. Using a similar idea, some test bridges have been constructed out of evaporated aluminum at the University of Wisconsin Micro-electronics Laboratory, which also seem to have good mechanical properties.

Although HgCdTe is effective at providing prompt and complete conversion of an X-ray into phonons, it has the disadvantage of a relatively low Debye temperature ($\theta \approx 150$ K) and therefore a comparatively high heat capacity. An optimal X-ray absorber has the minimum heat capacity per unit volume which is consistent with efficient absorption and phonon conversion. It would also be desirable to have a material which could be grown or evaporated directly onto the Si substrate, to avoid the additional heat capacity due to the epoxy used to attach the converter. To this end, we are having HgTe epitaxially grown directly onto Si wafers. Some materials which may be superior to HgCdTe are α–SnGe, a zero band-gap semimetal which has a higher Debye temperature than HgCdTe, or superconductors such as Re and Os. These superconductors have a negligible electronic contribution to the specific heat at the temperatures where we operate the device (T is much less than the superconducting transition temperature), since essentially all of the thermally excited quasiparticles are frozen out. They have the advantages of high Debye temperatures, which implies a low heat capacity per volume, and high atomic numbers, which implies that only thin layers will be required to stop the incident X-rays. These materials may not promptly thermalize all the energy, since much of the energy will go into the creation of quasiparticles, which can have recombination times of hundreds of microseconds or longer. Adding magnetic impurities or threading the superconductor with a few flux lines may aid conversion to phonons, but will increase the heat capacity. There is thus a trade-off between thermal conversion efficiency and heat capacity in these materials, and the recombination process will have to be studied to determine the optimum ratio of normal to superconducting regions.

References

[1] S.H. Moseley, J.C. Mather, and D. McCammon, "Thermal Detectors as X-ray Spectrometers," J.Appl.Phys., Vol. 56, pp. 1257-1262, September 1984.

[2] D. McCammon, S.H. Moseley, J.C. Mather, R.F. Mushotzky, "Experimental Tests of a Single-photon Calorimeter for X-ray Spectroscopy," J.Appl.Phys, Vol. 56, pp.1263-1266, September 1984.

[3] S.H. Moseley, R.L. Kelley, J.C. Mather, R.F. Mushotzky, A.E. Szymkowiak, and D. McCammon, "Thermal Detectors as Single-photon X-ray Spectrometers," IEEE Transactions on Nuclear Science, Vol. NS-32, pp.134-138, February 1985.

[4] D. McCammon, M. Juda, D. Reeder, R.L. Kelley, S.H. Moseley, and A.E. Szymkowiak, "A New Technique for Neutrino Mass Measurement," in Neutrino Mass and Low Energy Weak Interactions: Telemark 1984, eds. V. Barger and D. Cleine (World Scientific), p. 329.

[5] D. McCammon, M. Juda, J. Zhang, R.L. Kelley, S.H. Moseley, and A.E. Szymkowiak, "Thermal Detectors for High Resolution Spectroscopy," in IEEE Transactions on Nuclear Science, Vol. NS-33, p. 236, 1985.

[6] D. McCammon, M. Juda, J. Zhang, S.S. Holt, R.L. Kelley, S.H. Moseley, and A.E. Szymkowiak, "Thermal Detectors for High Resolution Spectroscopy," in Proc. 18th Int. Conf. on Low Temperature Physics, Kyoto, 1987, Japanese Journal of Applied Physics, Vol. 26, in press.

[7] S. Sahling, J. Engert, A. Gladun, R. Knoner, "The Thermal Boundary Resistance Between Sapphire and Aluminum Monocrystals at Low Temperatures," J.Low Temp. Phys., Vol. 45, pp.457-469, 1981.

[8] R.S. Raghavan, "Ultralow-Energy K-Capture Decay of ^{158}Tb: A New Neutrino Balance," Phys.Rev.Lett., Vol. 51, pp. 975-978, September 1983.

[9] S. Weyhe, B. Junge, F. Petzoldt, S. Bruns, and W. Gey, "Specific Heat of Delta-Bond 152, Epo-Tek H20E, and Wood's Metal at Low Temperatures," Cryogenics, pp. 166-167, March 1983.

[10] P.M. Downey, "The Low Temperature Conductivity of Ion Implanted Silicon and Its Application in a Cryogenic Far-IR Monolithic Bolometer," Ph.D. Thesis, Massachusetts Institute of Technology, 1980.

[11] S. Meyer, private communication.

[12] B. Sadoulet, "Prospects for Detecting Dark Matter Particles by Elastic Scattering," in Proceedings of 13th Texas Symposium on Relativistic Astrophysics, Singapore: World Scientific, 1987, pp. 260-264.

Appendix: Pulse Height Determination

Equation (1) was derived by optimizing the detector performance with respect to the known noise sources.[1] For laboratory measurements of pulse height spectra, we first digitize the output signal voltage into 12 bit samples at typically 100 kHz, and record 2048 samples per X-ray event for later processing. Data are also taken in the absence of X-ray pulses to obtain a measurement of the detector noise. For a linear detector, the energy of each event is proportional to the pulse amplitude. In this case it is straightforward to perform an optimal (i.e., minimum χ^2) fit for the amplitude of each event. The model for the total output signal can be described by the expression

$$P(t) = V_0 + H(E) \times S(t) + N(t), \qquad (A.1)$$

where V_0 is the DC output signal, S(t) is the intrinsic pulse shape (e.g., $e^{-t/\tau}$ for an ideal calorimeter), H(E) is the pulse height for photon energy, E, and N(t) is the total noise (phonon, Johnson, amplifiers, etc.). In the frequency domain, the model pulse shape is simply:

$$P(f) = H(E) \times S(f), \qquad \text{for } f>0.$$

Because of the linearity in H(E), the signal power at each frequency can be used as an independent estimator of H(E). We perform a minimum χ^2 analysis for H(E) in the frequency domain using the measured noise spectrum:

$$\chi^2 = \sum_{f>0} \frac{[\,D(f) - P(f)\,]^2}{N^2(f)} \qquad (A.2)$$

where N(f) is the measured total noise spectrum and D(f) is the Fourier transform of an X-ray pulse. By minimizing this expression with respect to H(E), it can be shown that H is given by (to within a normalizing constant):

$$H(E) = \sum_{f>0} D(f) \times \left[\frac{S^*(f)}{N^2(f)}\right] \qquad (A.3)$$

This can also be written as (again to within a normalizing constant):

$$H(E) = \sum D(t) \times T(t), \text{ where } T(f) = \frac{S(f)}{N^2(f)}$$

Thus, the procedure for determining the pulse height spectrum is as follows. The shape function, S(t), is constructed by averaging a large number of pulse events. Next, the noise spectrum, N(f), is computed by averaging many individual noise spectra. The optimal template function T(f) is computed in the frequency domain, and then transformed back into the time domain, and is convolved with each X-ray pulse record. The maximum of this convolution is the best estimate of the pulse height. This process has the advantage of being applicable for any (non-zero) spectrum, and can be thought of as a "notch filter" that weights the contributions to H(E) in the frequency domain depending on the S/N ratio. It is worth emphasizing that this approach is valid only under the assumption that the intrinsic pulse shape does not vary, which should be true as long as $\Delta T/T \ll 1$. For non-linear detectors, it is necessary to parameterize the pulse shape, and thus the photon energy, in terms of several parameters and fit for these simultaneously.