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Radiation detectors for X-ray and gamma-ray spectroscopy

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The detectors used primarily for the measurement of X-rays and gamma-rays are discussed because of their predominance in analytical measurements, and having the most dramatic improvements.

Introduction

Radiation measurements have a key role in radioanalytical and nuclear chemistry. Continuous progress in the development of instruments designed to detect and measure radiation have led to improvements in their capability in many different applications. In this article, emphasis will be placed on detectors used primarily for X-rays and gamma-rays because of their predominance in analytical measurements. While there has also been progress in devices designed to measure alpha- and beta-radiation, the most dramatic improvements have come about for measurements of X- and gamma-rays.

Portable instruments capable of making spectroscopic measurements of single photons of X- or gamma-rays first appeared in the post-WWII era, with the introduction of thallium-activated sodium iodide scintillators. For the first time, radioisotopes could be readily identified through the pulse-height spectrum observed from such a scintillator. Initially, these spectra were recorded in a laborious fashion by scanning a discriminator or single-channel analyzer across the full range of pulse amplitudes. Beginning in the late 1950s, multichannel analyzers (MCAs) began to provide a method of recording all the pulses from the detector in parallel and sorting them into appropriate amplitude channels. The first commercial MCAs based on ferrite core memories appeared in about 1960. When used with a sodium iodide scintillation detector, these multichannel analyzers ushered in the era of modern gamma-ray spectroscopy. It is significant to note that advances in both the detector and the electronics needed for its readout were both important in this case. A similar interdependence of the detector and electronics remains important in the development of the field today.

The next major step in gamma-ray spectroscopy occurred in the early 1960s with the appearance of lithium-drifted germanium detectors. These devices had a revolutionary impact on gamma-ray spectroscopy since they could provide more than an order of magnitude better energy resolution compared to scintillators. It then became possible to examine complex decay schemes and mixtures of isotopes that would result in hopelessly overlapped spectra using scintillators. The fact that these detectors needed to be operated and stored at liquid nitrogen temperature added a complication, but the vastly superior energy resolution provided from germanium easily justified this inconvenience in many applications.

Beginning in the 1970s, advances in germanium purification techniques allowed the introduction of large-volume germanium detectors in which the lithium drifting step could be omitted. These so-called high-purity germanium detectors had quite similar performance characteristics compared with the lithium-drifted germanium devices they replaced, but the high purity detectors offer one important operational advantage: they can be allowed to warm to room temperature between uses. The instability of lithium in germanium at room temperature prohibited the warming of lithium-drifted detectors at any time following their fabrication, so there was the additional burden of cooling these devices continuously whether they were in use or not.

Since their introduction, typical sizes for germanium detectors have steadily increased. It is not unusual to incorporate a germanium mass of a kilogram or more, and with a detection efficiency that can be as much as twice that of a 3"×3" sodium iodide scintillation crystal. The "relative efficiency" that is quoted by manufacturers is just this ratio measured at 1.33 MeV, and values of 200% or more have been reached. This increase in the typical size of germanium spectrometers has several beneficial effects. The increase size obviously results in an increased solid angle for point sources so that more of the gamma-rays are intercepted by the detector than for the case of the smaller volume. Furthermore, the increased mass enhances the interaction probability of the gamma-rays so that the intrinsic efficiency of the detector also increases. The large volume and mass also promotes multiple interactions of incident gamma-rays that more frequently lead to full absorption of the gamma-ray energy. As a result, the fraction of all gamma-rays that deposit their full energy and appear in the full-energy peak in the recorded spectrum is

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also enhanced. This effect is illustrated in Fig. 1. Not only does this improvement result in higher counting rates in the full-energy peak, but the associated Compton continuum is suppressed relative to the peak. Weak gamma-ray lines at lower energies are thus more likely to be detectable above this continuum.

Germanium detectors that are available commercially generally fall into two categories: *n*-type or *p*-type detectors. Of the two, *p*-type are historically the more common and are usually available at a somewhat lower cost. They have one disadvantage in the nature of the electrical contact that must be formed on the outer surface of the detector. For *p*-type detectors, this contact is a relatively thick lithium-diffused contact that serves as a dead layer through which the incident gamma-rays must pass. As a result, the detection efficiency of *p*-type detectors falls off dramatically at energies below about 100 keV (see Fig. 2). If the crystal is grown from material that is slightly *n*-type rather than *p*-type, a different contact can be formed through ion implantation that is much thinner. As a result, *n*-type detectors have an extended response to low energies that can be useful when applied to low-energy gamma-ray emitters or to the measurement of X-rays.

Nearly all germanium detectors are operated in conjunction with a storage Dewar containing liquid nitrogen. The user must then periodically refill the Dewar (typically at weekly intervals). For field use or at remote locations where liquid nitrogen may not be available, an alternative is the use of mechanical cooling through refrigerator units that are also available commercially.

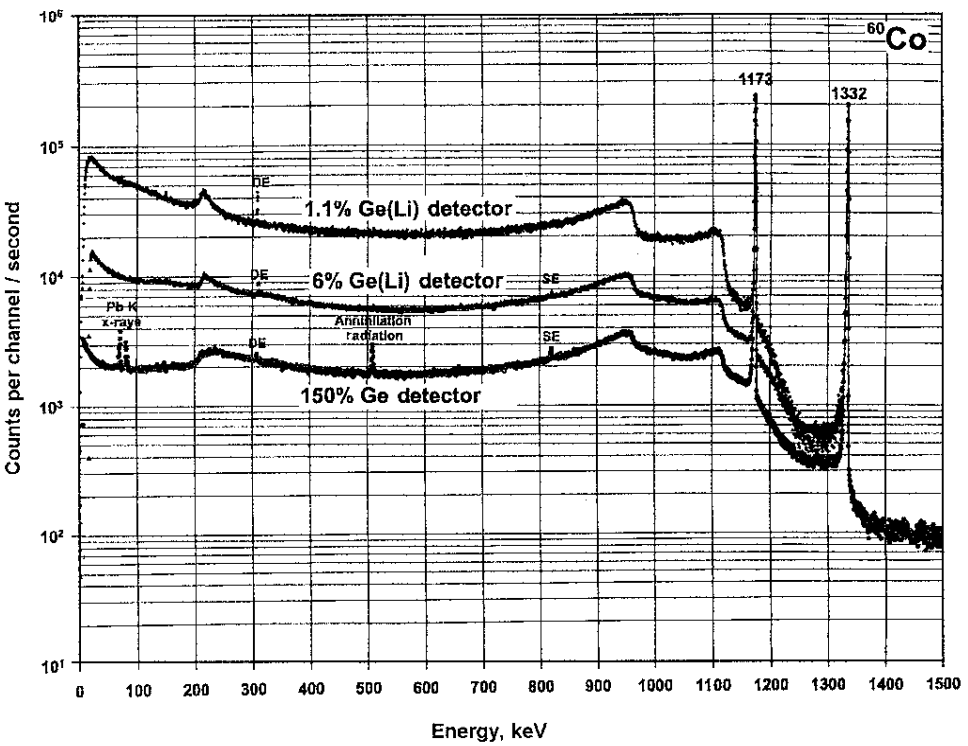

Fig. 1. Illustration of the effect of detector size on gamma-ray spectra from ^{60}Co. Each of the spectra shown is normalized to the same area of the upper full-energy peak. The large detector with 150% relative efficiency results in a greatly suppressed continuum relative to those from the detectors with smaller efficiencies. SE and DE designate single escape and double escape peaks. (Plot from HELMER et al., Gamma-Ray Spectrum Catalog, CD-ROM published by Idaho National Engineering and Environmental Laboratory, 1999)

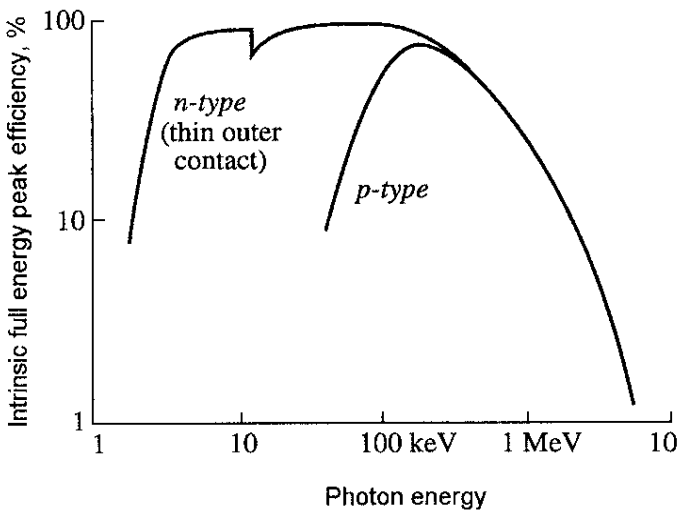

Fig. 2. Typical variation of the full-energy peak efficiency with photon energy for *n*-type and *p*-type coaxial germanium detectors of equivalent volume. The thicker outer contact on the p-type detector attenuates low-energy incident photons

Alternative scintillators

Thallium-activated sodium iodide, NaI(Tl), has remained by far the most widely used scintillation material for almost five decades because of its combination of several attractive properties. It has the highest light output per unit deposited energy for any scintillator when measured using conventional photomultiplier tubes. Since light yield is closely coupled to achievable energy resolution, a high light yield is always an advantage. Sodium iodide can also be grown as a crystal in rather large sizes that retain good optical quality. For gamma-ray applications, the iodine component (with an atomic number of 53) serves as the primary converter of gamma-ray photons to photoelectrons. A disadvantage of NaI(Tl) is its relatively long decay time of 230 ns. For many timing or coincidence applications or for operation at high rates, one would prefer a faster decay time.

Several new scintillators have appeared over the past decade that offer some interesting alternatives. Many of these are inorganic crystals in which the Ce^{3+} ion has been added in small concentration to serve as a luminescence center. Some of these materials with their characteristics are listed in Table 1. They share the useful property that their decay times are generally much shorter than that of NaI(Tl). Several also have high atomic number constituents that aid in promoting photoelectric absorption of gamma-rays. All are somewhat inferior in light yields to NaI(Tl), and so the achievable energy resolution is generally somewhat poorer. There are also problems with the availability of large-size crystals with good optical quality. Nonetheless, these new scintillators are making some inroads in applications where the faster time performance outweighs some of their other disadvantages.

Room temperature semiconductors

Scintillators operate at room temperature, but provide relatively poor energy resolution. Germanium has excellent energy resolution, but must be operated at liquid nitrogen temperature. For the past several decades, there has been a quest for an alternative semiconductor material that would retain (or at least come close to) the excellent energy resolution of germanium while not requiring cooling below room temperature. This goal remains largely unfulfilled today, although there has been interesting progress in this direction.

Table 1. Some alternative fast inorganic scintillators (all activated with Ce)

Material	Acronym	λ_{max}, nm	Decay time, ns	Light yield, % NaI(Tl)
$Gd_2(SiO_4)O$	GSO	440	56	24
$YAlO_3$	YAP	370	27	47
$Y_3Al_5O_{12}$	YAG	550	88, 302	45
$Lu_2(SiO_4)O$	LSO	420	47	66
$LuAlO_3$	LuAP	365	17	45

In order to be operable at room temperature, the semiconductor material must have a relatively wide band gap of at least 1.5 eV or more. As possible candidates, several semiconductor materials that are compounds made up of two or more elements have received the most attention. Development work continues on both cadmium telluride and mercuric iodide, both of which would have sufficiently wide band gap to allow room temperature operation. Difficulties have been encountered in the past with obtaining material of sufficient purity and crystalline perfection so that charges can be efficiently collected from crystals with more than very small volume, and also in the growth of crystals with sizes greater than a centimeter or so in dimension. More recently, considerable attention has been paid to a ternary compound, cadmium-zinc-telluride (CZT), that has the potential of being grown in larger size crystals. Figure 3 shows a gamma-ray spectrum taken at room temperature from a 1 cm^3 CZT crystal. This result was obtained using a novel electrode configuration (known as a coplanar grid anode) to help overcome some of the basic limitations of poor hole transport in this material. The observed energy resolution is much better than achievable in scintillators, but still well short of that from germanium. Although these results offer hope for the future, large-volume CZT crystals that are capable of good spectroscopic performance are not yet routinely available.

X-ray detectors

Probably the most common X-ray detector in use over the past several decades is the lithium-drifted silicon [Si(Li)] detector. Silicon, because of its low atomic number ($Z=14$), is of little interest in gamma-ray spectroscopy where germanium ($Z=32$) is much more attractive. However, for incident photon energies of tens of keV or less, even a low-Z material like silicon will show an appreciable photoelectric absorption probability. In order to have reasonable detection efficiency up to these energies, the silicon detector must have a thickness of at least 3 or 4 mm. Using silicon of the highest available purity, active thicknesses are limited to no more than about 1 mm using simple diode structures. Therefore, the lithium-drifting process, which has been abandoned for germanium, is still an important process to produce thicker silicon detectors. The lithium ions that are slowly drifted into the crystal serve to compensate the residual acceptor impurities in the *p*-type silicon, and a much thicker depletion region (or active thickness) can be produced. Si(Li) detectors of a few millimeters in diameter are commonplace to record the characteristic X-rays emitted from a sample using an electron beam in standard electron microscopy facilities. Because the charge released by a single X-ray photon is quite small, all sources of noise in the measurement must be minimized. For that reason, Si(Li) detectors are normally cooled to liquid nitrogen temperatures to suppress thermally-generated leakage current in much the same manner as for germanium detectors.

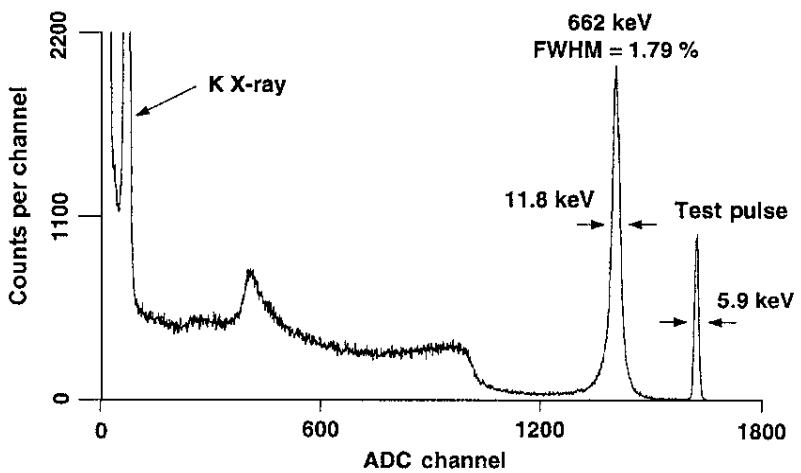

Fig. 3. Gamma-ray spectrum from a room temperature 1 cm^3 CZT detector fitted with a "coplanar grid" anode. (From HE et al., Nucl. Instrum. Meth., A388 (1997) 180)

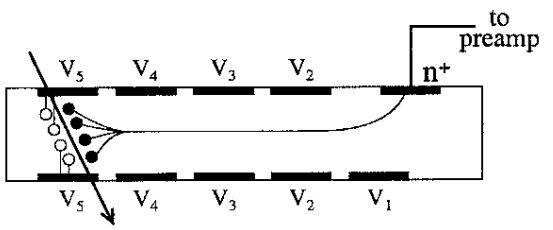

Fig. 4. Cross section through a silicon drift detector. The bulk of the silicon is *n*-type, and *p*-type strip or ring contacts are provided on both surfaces. Holes (open circles) created by an incident photon are collected at either surface. A voltage gradient between strips drives electrons (dark circles) along the potential minimum at the midplane of the wafer to a small-area anode

Furthermore, careful attention has to be paid to electronic noise associated with the preamplifier. This area of application has stimulated some of the most innovative developments in low-noise preamplifier designs.

Another common X-ray detector is a proportional counter filled with xenon gas. These detectors offer much larger volumes and resulting detection efficiency, but have poorer energy resolution compared with Si(Li) detectors. One interesting variant of the proportional counter design is the gas proportional-scintillation detector. In this device, the electric fields through the gas are kept low enough so that no charge multiplication occurs as in a proportional counter. Instead, the signal is derived from the light that is generated through electron collisions with neutral gas molecules, leaving them in an excited state. The subsequent de-excitation photons are detected in a photomultiplier tube in the same manner as in a scintillation counter. This approach has the advantage of resulting in superior energy resolution compared with proportional counters because it avoids the fluctuations introduced in forming avalanches in the charge multiplication process. These X-ray detectors have proven useful in space-based measurement of X-ray spectra, and are still undergoing considerable research in aspects of their design.

Portable X-ray spectrometers with Peltier cooling

Within the past few years, useful X-ray detectors for portable use have become commercially-available. They are based on the use of either silicon or CZT sensors that are cooled to about –40 °C using an electrical Peltier cooler. The cooler is built into the detector probe geometry, and its operation is essentially invisible to the user. While not reaching the energy resolution of standard Si(Li) systems, these portable units have sufficient resolution to separate the characteristic X-rays from light elements.

Silicon drift detectors

In a normal silicon diode, electrodes are fabricated on both surfaces of a disk-shaped wafer, and charges formed by incident radiation are cause to drift in a direction that is perpendicular to the wafer surfaces. As these diodes are made larger in area, their capacitance increases and eventually becomes the limiting factor in determining the electronic noise level of the system. An alternative configuration has been developed that takes a fundamentally different approach to collecting the charge. As shown in Fig. 4, rectifying junctions are provided at both surfaces of the silicon wafer so that a potential minimum for electrons is formed near the wafer midplane. When electrons and holes are created by an incident particle or photon, the holes are drawn to the nearer surface. The electrons, however, drift to the midplane of the wafer and are confined by the potential minimum. If one or both of the surface electrodes are fabricated as a series of strips or rings, this potential minimum can be "tipped" to one point on the wafer by applying different voltages to each of the strips or rings. In this way, the electrons can be made to drift substantial distances that are parallel to the wafer surface, before arriving at a small area anode.

A significant advantage of this configuration is that the capacitance of the readout is determined only by the small area anode, and is not dependent on the area of the detector. Therefore, detectors with substantial surface area can be employed while keeping their capacitance at very low levels. To suppress leakage current, these silicon drift detectors are generally operated at temperature that is below room temperature but well above that of liquid nitrogen. As a result, electrical coolers can be used that lend themselves much more readily to incorporation in portable instruments. The thickness of these devices is typically on the order of 300 μm of silicon. Therefore, their application is limited to very low energy X-rays where the probability of interaction within that thickness of silicon is still reasonable. Because of their low capacitance, silicon drift detectors have shown excellent energy resolution that approaches the limit set by charge carrier statistics (about 110 eV for the typical X-ray calibration energy of 5.9 keV from ^{55}Fe). They also allow high rate operation because the shaping times that can be used are much shorter (due to the small capacitance) than those required for lithium-drifted silicon detectors.

Microcalorimeters and transition edge sensor X-ray detectors

A relatively new development in the spectroscopy of X-rays exploits the very low heat capacity of materials at extremely low temperature. If a small target of material is held at a cryogenic temperature typically well below

1 K, then even the minuscule amount of energy deposited by a single particle or photon can raise its temperature by an amount that becomes measurable using sensitive techniques. These "microcalorimeters" represent a fundamentally different method for the detection of radiation since no charge carriers are collected. Instead, the energy of the radiation is simply converted directly into heat. This approach avoids one of the limits on energy resolution that always applies to conventional devices that are based on collecting charges. Because a limited number of these charges are produced per radiation interaction, their number is subject to statistical fluctuation. Many detectors including germanium gamma-ray spectrometers and cooled silicon detectors approach this limit in some applications, and, therefore, no further improvement in their energy resolution can be expected. In microcalorimeters, this statistical limit is no longer significant since charge carriers do not play a role in forming the output signal. The result has been the creation of a new class of detectors that, while still in the development stage, have demonstrated a real breakthrough in energy resolution. The targets are necessarily limited to very small volume and must be operated in conjunction with an elaborate cryogenic cooling system, but these microcalorimeters represent a truly innovative way to measure radiation.

In many of the early experiments, the temperature rise was measured by incorporating an ion-implanted thermistor as part of the low-temperature target. A newer approach exploits the unique properties of superconductivity at low temperatures. As shown in Fig. 5, a thin strip of superconducting material is placed in thermal contact with the energy-absorbing target. This material is superconducting below a certain transition temperature, and above that temperature reverts to having a finite resistance. If the system is started at a temperature above the transition temperature and a voltage is applied across the strip, the resulting current will produce a small amount of ohmic heating in the strip. If the heat flow to the low-temperature sink exceeds that of ohmic heating, then the temperature of the system will begin to drop. As the temperature of the superconducting strip reaches its transition range, there will be a sudden decrease in its resistance. The current flow thus increases, also increasing the ohmic heating. An equilibrium point is then reached in which the ohmic heating exactly balances the heat flow to the thermal sink, and that point can be made to lie in the steeply changing transition region between superconductivity and normal conductivity of the strip. Under these conditions, the superconducting strip becomes an extremely sensitive thermometer. If a small amount of energy is deposited in the target by an incident particle, the target temperature will tend to rise. The resulting increase in the superconducting strip temperature will increase its resistance and reduce the ohmic heating rate. The net result is only a tiny rise in temperature, but a measurable drop in the current that flows through the superconducting strip. That change in the current can be recorded in the external circuit using sensitive methods and interpreted in terms of the amount of energy deposited in the absorbing target. These transition edge sensor detectors have proven capable of making measurements on 5 keV X-rays with an energy resolution of better than 8 eV. That energy resolution is almost a factor of 20 better than that achievable in typical lithium-drifted silicon detectors.

Trends for the future

While some of the common instruments in use today for X- and gamma-ray detection are many decades old, there continues to be a significant evolution in their capabilities. New scintillation materials offer combinations of decay time and detection efficiency that were not available a decade ago. The size of available germanium gamma-ray spectrometers continues to increase, adding to their performance capabilities.

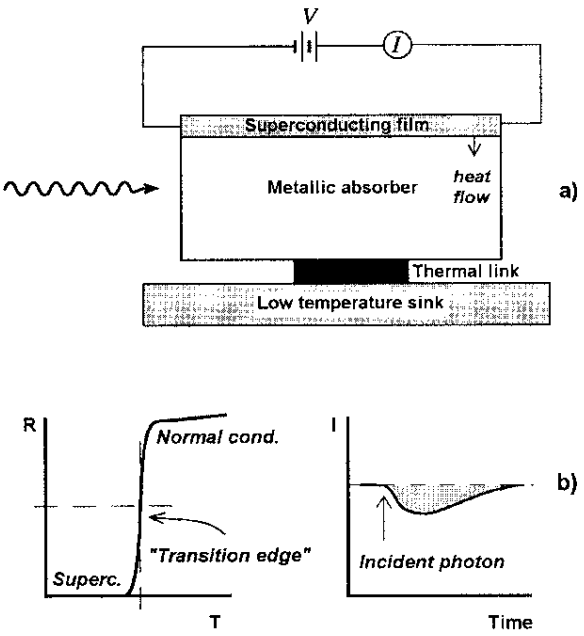

Fig. 5. (a) A schematic representation of a microcalorimeter using a thin film of superconducting material as a transition edge sensor. (b) At the left is a plot of the resistance R of the superconducting strip as a function of temperature T. The operating point is shown by the dashed lines and is on the steeply-changing transition edge. On the right is a plot of the current I measured in the external circuit when a voltage V is applied across the strip. The momentary drop in current caused by an incident photon is proportional to the energy deposited in the absorber by that photon

New silicon-based systems show excellent energy resolution and usable detection efficiency when applied to soft X-rays that are characteristic of the lighter elements. Fundamentally new techniques based on thermal measurements are emerging as a new class of devices that offers breakthroughs in energy resolution not possible in devices based on charge collection. There has also been a dramatic increase in the capability of the supporting electronics for detector systems. It is now possible to support hundreds of detector elements with low-noise electronics integrated onto a single readout chip. This capability opens the door for innovative detector designs in which segmented electrodes permit unique modes of operation and/or sensing of the position of interaction of the radiation. The techniques used to process pulses are also evolving into digital systems that exploit the availability of extremely fast analog-to-digital converters. These digital pulse-processing systems have the advantage of flexibility and stability that can prove to be advantageous in demanding circumstances where analog systems have reached their limit.