sible mechanisms, or suggest new ones, but they cannot prove mechanisms true. Further, in practice, problems associated with low sigin practice, problems associated with low signal-to-noise ratios, or the many possible mechanical and electronic artifacts, frequently limit the complete analyses of data—especially with regard to multistep reactions.
35. R. Lumry, R. Legare, W. G. Miller, Biopolymers 2, 489 (1964).
36. E. Hamori and H. A. Scheraga, J. Phys. Chem. 71, 4147 (1967).
37. T. K. Saksena, B. Michels, R. Zana, J. Chim. Phys. 65, 597 (1968).
38. G. G. Hammes and P. B. Roberts, J. Amer. Chem. Soc. 91, 1812 (1969).
39. G. Schwarz and J. Seelig. Biopolymers 6.

- 39. G. Schwarz and J. Seelig, *Biopolymers* 6, 1263 (1968).
- 1263 (1968).
 40. G. Schwarz, J. Mol. Biol. 11, 64 (1965).
 41. E. M. Bradbury, C. Crane-Robinson, H. Goldman, H. W. E. Rattle, Nature 217, 812 (1968); J. A. Ferretti and L. Paolillo, Biopolymers 7, 155 (1969).
 42. R. Ullman, Biopolymers 9, 471 (1970); J. A. Ferretti, B. W. Ninham, V. A. Parsegian, Macromolecules 3, 34 (1970).
 43. J. Wetmur and N. Davidson, J. Mol. Biol. 31 349 (1968).

- J. Wetmur and N. Davidson, J. Mol. Biol. 31, 349 (1968).
 D. Pörschke, quoted by M. Eigen, in Fast Reactions and Primary Processes in Chem-ical Kinetics, S. Claesson, Ed. (Wiley, New York, 1967), pp. 358-367.
 R. Römer, D. Riesner, G. Maass, W. Win-termeyer, R. Thiebe, H. G. Zachau, Fed. Eur. Biochem. Soc. Lett. 5, 15 (1969).
 H. J. Li and D. M. Crothers, J. Mol. Biol. 39, 461 (1969).
 R. Bittman, ibid. 46, 251 (1969).
 B. Anfinsen, Harvey Lect. Ser. 61, 95

- 48. C B. Anfinsen, Harvey Lect. Ser. 61, 95 48. C. B. Annisch, Harvey Lett. Ser. 61, 93 (1967).
 49. F. M. Pohl, Eur. J. Biochem. 7, 146 (1969).
- J. W. Allis and J. Steinhardt, Biochemistry 8, 5075 (1969).
- 51. A. N. Schechter, R. F. Chen, C. B. Anfinsen, Science 167, 886 (1970).
- 52. P. Hauschka, thesis, Johns Hopkins Univer-
- F. Hadschka, thesis, Johns Hopkins Chivelsity (1969).
 C. Tanford, Advan. Protein Chem. 23, 121 (1969); C. Tanford, *ibid.* 24, 1 (1970).
 H. Theorell, B. Chance, Y. Yonetani, J. Mol. Biol. 17, 513 (1966).

- 55. B. Chance, A. Ravilly, N. Rumen, *ibid.*, p. 525; B. Chance and A. Ravilly, *ibid.* 21, 195 (1966).
- 56. B. Chance, Rev. Sci. Instrum. 22, 634 (1951). K. Kirschner, M. Eigen, R. Bittman, B. Voigt, Proc. Nat. Acad. Sci. U.S. 56, 1661 (1966).
- 58. J. Monod, J. Wyman, J.-P. Changeux, J. Mol.
- Biol. 12, 88 (1965); D. E. Koshland, G. Nemethy, D. Filmer, *Biochemistry* 5, 365 (1966).
 G. G. Hammes and F. G. Walz, *J. Amer. Chem. Soc.* 91, 7179 (1969), and preceding 59.
- papers. 60. G. C. K. Roberts, E. A. Dennis, D. H. Meadows, J. S. Cohen, O. Jardetzky, Proc. Nat. Acad. Sci. U.S. 62, 1151 (1969).
 61. H. W. Wyckoff, D. Tsernoglou, A. W. Han-
- H. W. Wyckoff, D. Isernogiou, A. W. Haffson, J. R. Knox, B. Lee, F. M. Richards J. Biol. Chem. 245, 305 (1970).
 H. Gutfreund and J. M. Sturtevant, Biochem J. 63, 656 (1956); S. A. Bernhard and H. Gutfreund, Proc. Nat. Acad. Sci. U.S. 53, 1238 (1965); B. H. Havsteen, J. Biol. Chem. 242, 769 (1967).
- 63.
- 242, 769 (1967).
 E. D. Barber and H. J. Bright, Proc. Nat. Acad. Sci. U.S. 60, 1363 (1968).
 M. Iwatsubo and D. Pantaloni, Bull. Soc. Chim. Biol. 49, 1563 (1967); J. M. Jallon, A. Di Franco, M. Iwatsubo, Eur. J. Biochem. 13, 428 (1970); H. F. Fisher and J. R. Bard, Biochim. Biophys. Acta 188, 168 (1969); C. Y. Huang and C. Frieden, Proc. Nat. Acad. Sci. U.S. 64, 338 (1969); G. H. Dodd and G. K. Radda, Biochem. J. 114, 407 (1969). (1969).
- (1969).
 (1969).
 (5) R. S. Criddle, C. H. McMurray, H. Gutfreund, Nature 220, 1091 (1968); H. Heck, J. Biol. Chem. 244, 4375 (1969); T. Wuntch, R. F. Chen, E. S. Vesell, Science 167, 63 (1970); H. Suzuki and Y. Ogura, J. Biochem. (Tokyo) 67, 277 (1970).
 (6) H. Theorell, A. Ehrenberg, C. de Zalenski, Reicherg, Biochem. 27, 200
- H. Incorell, A. Enrenberg, C. de Zaleński, Biochem. Biophys. Res. Commun. 27, 309 (1967); J. Shore, Biochemistry 8, 1588 (1969); S. A. Bernhard, M. F. Dunn, P. L. Luisi, P. Schack, *ibid.* 9, 185 (1970).
- Schack, 101a. 9, 163 (1970).
 K. Yagi, M. Nishikimi, N. Ohishi, A. Takai, J. Biochem. (Tokyo) 67, 153 (1970).
 N. Higashi, H. Shoun, K. Hiromi, K. Yano,
- K. Arima, *ibid.*, p. 749.

- P. W. Taylor, R. W. King, A. S. V. Burgen, *Biochemistry* 9, 2638 (1970).
 B. Finlayson and E. W. Taylor, *ibid.* 8, 802 (1969); B. Finlayson, R. W. Lymn, E. W. Taylor, *ibid.* 9, 811; R. W. Lymn and E. W. Taylor, *ibid.* 9, 2975 (1970).
 G. G. Hammes and J. L. Haslam, *ibid.* 8, 1591 (1969).
 D. Storm and D. E. Koshland Proc. Nat.

- (1969).
 D. R. Storm and D. E. Koshland, Proc. Nat. Acad. Sci. U.S. 66, 445 (1970); J. H. Wang, ibid., p. 874.
 Q. H. Gibson, Annu. Rev. Biochem. 35, 435 (1966); B. Chance, D. DeVault, V. Legallais, L. Mela, T. Yonetani, in Fast Reactions and Primary Processes in Chemical Kinetics, S. Chemer Ed. Wilter, New York 10(7), pre-Claesson, Ed. (Wiley, New York, 1967), pp. 437-468.
- 437-468.
 74. R. W. Noble, Q. H. Gibson, M. Brunori, E. Antonini, J. Wyman, J. Biol. Chem. 244, 3905 (1969); M. Brunori and T. M. Schuster, *ibid.*, p. 4046.
 75. Q. H. Gibson, *ibid.* 245, 3285 (1970).
 76. M. Brunori, E. Antonini, C. Phelps, G. Amiconi, J. Mol. Biol. 44, 563 (1969); M. Brunori, G. Amiconi, E. Antonini, J. Wyman, K. H. Winterhalter, *ibid.* 49, 461 (1970).
 77. F. Antonini Science 158, 1417 (1968). F.

- K. H. Winternater, *ibia.* 49, 461 (1970).
 E. Antonini, *Science* 158, 1417 (1968); E. Antonini and M. Brunori, *Annu.* (1968); E. Antonini and M. Brunori, *Annu. Rev. Biochem.* 39, 977 (1970).
 R. E. Cathou and C. T. O'Konski, *J. Mol. Biol.* 48, 125 (1970).
 R. C. Valentine and N. M. Green, *ibid.* 27, 615 (1967). 77.
- R. C. Vale 615 (1967). 79.

- K. C. Patchine and M. M. Bielen, University, 615 (1967).
 J. Yguerabide, H. F. Epstein, L. Stryer, *ibid.* 51, 573 (1970).
 Ph. Wahl, J. Paoletti, J.-B. LePecq, *Proc. Nat. Acad. Sci. U.S.* 65, 417 (1970).
 G. Weber, *Methods Enzymol.* 16, 380 (1969).
 M. Cohn, *Quart. Rev. Biophys.* 3, 61 (1970).
 E. Charney, J. B. Milstien, K. Yamaoka, J. Amer. Chem. Soc. 92, 2657 (1970).
 C. F. Meares, K. G. Bryant, J. D. Baldeschwieler, D. A. Shirley, *Proc. Nat. Acad. Sci. U.S.* 64, 1155 (1969).
 Y. Yeh and R. N. Keeler, *Quart. Rev. Biophys.* 2, 315 (1969); Y. Yeh, J. Chem. Phys. 52, 6218 (1970).
- **52**, 6218 (1970). G. G. Hammes, *Science* **151**, 1507 (1966).
- 88. I thank my colleagues for their criticisms and suggestions,

Semiconductor Radiation Detectors

Basic principles and some uses of a recent tool that has revolutionized nuclear physics are described.

Fred S. Goulding and Yvonne Stone

Just as the theory of atomic structure grew from spectroscopic observations of transitions in the atomic shell, so nuclear theory has developed from studies of the radiations emitted in nuclear transitions. But while the light produced or absorbed in atomic transitions is easy for us to see and measure, we have no simple way of observ-

ing nuclear radiations, which may be charged particles (electrons, positrons, protons, and heavier nuclear constituents), or electromagnetic radiations of very short wavelength (gamma rays). Necessarily then, the progress of nuclear physics has closely followed the development of new means of detecting and measuring nuclear radiations.

In the past decade, we have witnessed a major revolution in nuclear experiments as a new device, the semiconductor radiation detector, has ap-

peared on the scene, providing relatively simple and accurate methods of measuring the energy of many of the radiations produced in nuclear processes. Knowledge of fission and of the structure of nuclei, detection of new transuranic elements and determination of their properties, and exploration of the nuclear surface all have advanced because of the development of these new detectors.

Even outside the field of nuclear physics, the influence of these detectors is being strongly felt. In archeology, because of semiconductor radiation detectors, specimens can now be analyzed in fine detail by observing the gamma rays emitted from a sample after neutron bombardment in a reactor, or by observing the characteristic fluorescent x-rays produced when a sample is exposed to an x-ray or gamma-ray source. Biology, geology, mining, criminology, and many types of industrial processing have taken advantage of this new analytical tool.

The power of semiconductor detectors in analysis is graphically illustrated by comparing the gamma-ray spectra

The authors are with the Lawrence Radiation Laboratory, Berkeley, California 94720. Fred Goulding is leader of the semiconductor detector group, and Yvonne Stone is a science writer with the information division.

shown in Fig. 1. The ordinate is a measure of the intensity of the gamma rays emitted from the sample; the abscissa shows their energy. The wealth of fine detail seen in the spectrum produced by the semiconductor detector is totally absent in that produced by its predecessor, the scintillation detector.

Radiation Energy Determination

by Ionization

The methods of detecting and measuring the energy of nuclear radiation depend finally on the effects of charged particles, even if this radiation is electromagnetic in nature (gamma rays) or consists of uncharged particles (neutrons, neutrinos, neutral mesons). The conversion process by which charged particles are produced from the others might be a "knock-on" effect, such as the production of high-energy protons by neutrons passing through hydrogenous material; or a photoelectric or other interaction in which gamma rays release electrons; or possibly a nuclear reaction in which a charged particle is one of the end products. In all cases, the conversion process must be taken into account in interpreting the results.

Only two fundamental methods of determining the energy of charged particles are available. If the particles are bent in a precisely known uniform magnetic field, as in a magnetic spectrometer, their momentum can be measured; then if their mass is known, their energy is determined. While this technique can provide very precise determinations of energy (accuracies ~ 0.01 percent), magnetic spectrometers are large and expensive and cover only a limited range of energies. The second basic method of finding the energy of charged particles utilizes the ionization produced by their passage through matter. Although individual exchanges of energy between a particle and the electrons in matter vary greatly in size, a well-defined average amount of energy is absorbed from the particle for each elementary charge it releases along the ionization track. Therefore, if the particle's energy is totally absorbed in a piece of material (that is, if the particle stops in it), the ionization produced in the material is a direct measure of the particle's energy.

Ionization occurs in all materials. In making detectors, our problem is to select materials in which the products of ionization—positive and negative 16 OCTOBER 1970

free charges-can be measured. Sometimes, a secondary effect of the free charge can be utilized; the operation of scintillation detectors, for example, depends on light emission from certain crystals, plastics, or liquids after their excitation by the ionization process. In certain cases, direct collection of the charge to produce a measurable current in an external circuit can be achieved: only gaseous ionization detectors and semiconductor detectors have adequate charge collection properties to warrant their being considered for spectrometers. Semiconductor detectors have many advantages over gaseous ionization chambers and scintillation detectors; the fact that virtually all nuclear spectroscopy now uses semiconductor detectors is proof of their value. The basic reasons are the following.

1) Solids are about 1000 times as dense as gases, and therefore much smaller thicknesses are needed to absorb radiation. Because of this, gaseous ionization chambers can be used only for measurements of slow-moving highly charged particles which produce short dense ionization tracks, but semiconductor detectors can be used for a much wider range of types of radiation.

2) The energy required to produce a single pair of elementary charges is much smaller in solids than in gases; in fact, the solids normally used in detectors produce about ten times the amount of ionization, compared with gases, for the same amount of absorbed energy. Consequently, electrical signals are larger, statistical fluctuations less important, and the energy resolution significantly better.

3) In principle, the above advantages also apply to scintillation detectors; indeed heavy materials of high atomic number used as scintillators give even better absorption efficiencies than those achieved with our two common semiconductors, germanium and silicon. Whereas the average energy required to produce a fundamental excitation or ionization in a scintillator is less than in a gas, inefficient processes are unfortunately involved both in the conversion of free electrons into light quanta by the scintillator and in the conversion of light quanta into electrons by the electron multiplier tube. Statistical fluctuations are very large, and the energy resolution suffers accordingly. Semiconductor detectors usually exhibit improvement in energy resolution of one to two orders of magnitude compared with scintillators.

Charge-Production Process

Production of ionization in a detector material is vital to detector operation and determines the basic limitations on performance. As it passes through a detector, a heavy charged particle interacts with atomic electrons via Coulomb forces, producing a large



Fig. 1. A complex gamma-ray spectrum, due to gross fission products, observed by a germanium detector (upper curve) and a scintillation detector (lower curve).



Fig. 2 (above). Passage of a heavy charged particle through a solid. Most of the energy is absorbed in producing electron-hole showers; at the end of the track, the particle becomes neutral and loses its remaining energy in atomic collisions. Fig. 3 (right). Three interaction mechanisms between gamma rays and the electrons in a material. In the photoelectric process nearly all the gamma-ray energy is transferred to the electron, which then produces a hole-electron shower. In the Compton process (billiard-ball collision), only a portion of the gamma-ray energy is given to the electron. Pair-production requires the expenditure of 1.02 Mev to produce an electron-positron pair, which can happen only with high-energy gamma rays. The 1.02-Mev reappears as a pair of 511-kev gamma rays when the positron annihilates. One or both of these 511-kev gamma rays may escape from the detecting volume. The ionization signal will therefore depend on whether single or double escape occurs.



number of small energy exchanges (~ 100 electron volts) all along its track. Each electron set free initiates a shower of holes and electrons (Fig. 2). As the heavy particle slows down, electrons attach to it and it becomes neutral. Beyond this point its remaining energy is dissipated primarily in atomic collisions that set lattice atoms into vibration or permanently displace them from their normal lattice sites. In normal circumstances, however, the fraction of an incident particle's energy lost in this radiation damage mechanism is very small.

When gamma rays enter a detector,

any of three primary interactions may take place between the gamma rays and electrons: a photoelectric interaction, a Compton collision, or pair-production (Fig. 3). Whereas a photoelectric event produces an amount of ionization corresponding directly to the gamma-ray energy, Compton events produce a variable amount of ionization. Only if the degraded (less energetic) secondary gamma ray is fully absorbed can we get useful information from Compton interactions about the distribution of gamma-ray energies. Therefore, unlike the case of heavy charged particles, where the amount of

ionization is almost the same for each member of a monoenergetic group of particles, the ionization produced by monoenergetic gamma rays is not constant. Instead, a statistical distribution of pulses of ionization is produced, containing well-defined peaks that can be used to determine gamma-ray energies. The rest of the ionization pulses constitute an undesirable background which can be reduced only when a larger detecting volume is used to prevent the escape of degraded gamma rays. The difference between the ionization-pulse distribution for gamma rays and that for heavy charged par-





Fig. 4 (left). The gamma-ray spectrum from 60 Co observed by a germanium detector. Note the two full-energy peaks due to the 1.16-Mev and 1.33-Mev gamma rays, and the Compton distribution at energies less than that of the peaks. The sharp drops in the curve at channels No. 680 and No. 790 are the so-called "Compton edges" corresponding to the maximum collision energy given to electrons by 1.16-Mev and 1.33-Mev gamma rays. Fig. 5 (above). The 42-Mev proton spectrum in germanium. There is a virtual absence of back-ground at energies lower than the peak.





Fig. 6 (left). Gamma-ray efficiency as a function of energy for a germanium detector 5 cm² and 0.8 cm thick. Peak efficiency is defined as the ratio of counts appearing in the peak to the total number of photons entering the detector. Fig. 7 (above). The range of protons, deuterons, tritons, ⁸He, and alpha particles in silicon.

ticles is seen by comparing Figs. 4 and 5.

Another striking difference between gamma rays and charged particles is that gamma rays may pass through a detecting medium without interacting at all. In fact, the efficiency for detecting gamma rays and the probability of the energy of a gamma ray being totally utilized to produce ionization may be quite small, as shown in Fig. 6. On the other hand, all heavy charged particles are detected, even by very thin detectors. For their total energy to be absorbed, however, the thickness of the detector must exceed the range of the particle in the detector material. Rangeenergy curves for a range of energies and several particles in silicon are shown in Fig. 7.

The primary effect of radiation is to



of a semiconductor. Experimental values are shown and compared with the theory of Klein (1). Fig. 9 (above). The energy resolution as a function of energy for silicon and germanium detectors. These curves take into account the basic statistical limitations involved in the charge-production process and in atomic collisons. The Fano factor F is assumed to be 0.08 for germanium, and 0.1 for silicon—the best experimental values yet observed. No account is taken of the amplifiernoise contributions to resolution.

0

In Sb

1

2

3

Band gap energy (ev)

4

5

6

release electrons and holes which travel at high velocities initiating showers of free electrons and holes, the final members of which are traveling at thermal velocities in the lattice. The energy of the radiation is expended partly in exciting electrons from the valence band of the solid-this we may consider useful work from the point of view of detectors-and partly in wasteful excitation of lattice vibrations. The performance of a detector material depends on how energy is shared between useful and wasteful work, and on the statistical fluctuations in this process. We can characterize the performance with the following variables:

1) ε , the mean energy required to produce a hole-electron pair; and

2) F, the Fano factor, a smoothing factor applied to Poisson statistics to obtain the value of the fluctuations in the ionization produced in a material. Physically, the factor F represents the statistics of sharing of energy between ionizing processes and excitation of lattice vibrations. If the average number of carriers produced is N, the mean square deviation, $\langle n^2 \rangle$, is given by

$\langle n^2 \rangle = FN$

Theoretical (1) and experimental (2) studies of these parameters permit prediction of the value of ε for different materials (Fig. 8) and accurate estimates of the energy resolution attainable in semiconductor detectors (Fig. 9). Allowance is made, in the case of heavy particles, for the statistics associated with the atomic-collision processes at the end of the track as analyzed by Linhard (3). No allowance has been made for the effects of noise in the electronic amplifier used with the detector.

Charge-Collection Process

A detector can be thought of as a block of material with electrical contacts attached to opposite faces, a structure analogous to the parallel-plate gaseous ionization chamber. Free charges produced in the detector are collected by applying a high voltage across the block. The impulse of current flowing in the external circuit while the free charges are in transit measures the energy of the photon or particle producing the signal. This simple picture of the operation of a semiconductor detector focuses attention on two important requirements.

1) The free charges-electrons and holes-must not recombine or be lost by becoming trapped before being collected; otherwise the total charge flowing in the external circuit will not be a true measure of the original ionization. In other words, the average lifetime of a carrier before recombining, or being trapped, must be long compared with its transit time across the sensitive region, which can range from 10^{-9} second to almost 10^{-6} second, depending on the design of the detector. The detector material must be such that the mobility of charge carriers is high, so that their velocity will be high in reasonable electric fields.

2) Spurious currents in the detector must be very small because signals produced in the external circuit may be minute. As an illustration of the magnitude of the quantities involved, we can now measure 1-kev x-rays, each releasing an average of only 300 electrons, with a resolution [full width at half maximum (FWHM)] of about 100 ev. This implies a root-mean-square error of only about 15 electrons in the whole

signal-measuring process, including contributions from the signal amplifier and those arising from spurious charges flowing in the detector. The need to reduce spurious current flow determines the structures chosen for detectors and influences the types and quality of the materials used.

The ideal semiconductor detector consists of a thick slice of very pure material having almost no free carriers of either type, with a positive electrode containing no free positive holes and a negative electrode containing no free electrons. A close approximation to such a structure is shown in Fig. 10. Heavy doping of the n layer almost totally suppresses its hole population; doping the p layer suppresses its free electrons.

When voltage is applied to this structure in the direction shown in Fig. 10, an electric field is produced throughout the bulk of the material, with the field lines terminating on the heavily doped surface layers. The direction of the electric field is such as to oppose injection of majority carriers from either contact into the bulk, although aiding injection of the minority (almost nonexistent) carriers. Contacts of the above type reduce the currents injected at the contacts almost to zero. The main sources of leakage in a detector then become charge generation and injection at the side surfaces of the material where the lattice suddenly terminates, and generation of holes and electrons in the main bulk of the material by thermal lattice vibrations that excite carriers from the valence into the conduction band (usually through the intermediary of traps). Lowering the temperature of the device, commonly to liquid-nitrogen temperature



Fig. 10 (left). An ideal, fully depleted detector with heavily doped surface layers of opposite types. Fig. 11 (right). The common junction detector in which the material is not fully depleted.

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(77°K), reduces the current due to thermal excitation, although surface effects may still be present. Well-made small silicon detectors, for example, cooled to 77°K and shielded from infrared radiation from the warm walls of the enclosure, exhibit leakage currents as low as 10^{-14} ampere, corresponding to less than ten electron-hole pairs flowing across the detector in the few microseconds needed for measuring the ionization induced by radiation.

A less ideal structure is shown in Fig. 11. Here the bulk p-type material is used for one noninjecting contact and the electric field only partially penetrates the material. In so doing, it drives free electrons and holes away from a region called the "depletion layer," whose thickness is proportional to the square root of the applied voltage. The depletion layer forms the sensitive region of the detector, from which the free carriers produced by ionization can be collected.

Detector Materials

Our choice of materials for semiconductor detectors is very limited, principally by charge-collection considerations. For charge-carrier losses to be small during their transit across the detector, crystals that are nearly perfect are required; lattice imperfections can trap charge in transit and cause excessive leakage currents by acting as intermediaries in the thermal excitation of carriers into the conduction band. So far, only silicon and germanium have performed adequately, although cadmium telluride shows promise (4). Materials with constituents of high atomic number are desirable as they are efficient absorbers of gamma rays by the photoelectric process. However, other considerations enter into the choice of material: for example, germanium cannot be used if room-temperature operation is required, because thermal excitation of carriers into the conduction band produces excessive leakage. If operation at still higher temperatures (>100°C) is necessary, even silicon is unusable; then silicon carbide or cadmium telluride become possibilities.

Another factor has played a big part in the development of semiconductor detectors: namely, that the highest purity silicon and germanium produced by standard zone-leveling and crystalgrowing techniques contain about 10^{12} impurities (either acceptors or donors)



Fig. 12 (left). Fluorescence spectrum of x-rays from a target containing Mg, Al, and Si as seen by a silicon detector. The exciting source was a small x-ray tube (14). Fig. 13 (right). Fluorescence spectrum of x-rays from a Al_2O_3 target bombarded with Al x-rays from the Al anode of a small x-ray tube. The target was contained in the same vacuum chamber as the detector so that no absorbing material was present between the target and the detector (15).

per cubic centimeter. Application of reasonable voltages to a detector of the type shown in Fig. 11, made from this material, produces a depletion layer only 0.5 to 2 mm thick. Such detectors can be used in many applications; but their gamma-ray efficiency is low, and they are not thick enough to stop many of the particles of interest in nuclear physics.

The inherent limitations of this material have been circumvented by "lithium drifting," a process invented by Pell (5) for compensating acceptor impurities with an interstitial donor (lithium). By this method both germanium and silicon can be produced with thick regions that exhibit almost no net impurity concentration. To make lithium-drifted detectors, lithium is evaporated onto and diffused into the face of a slice of p-type material at about 400°C to produce an n-p junction; then with the temperature held high enough for the lithium ions to be reasonably mobile (20° to 50°C for Ge and 120° to 150°C for Si), a positive voltage (500 volts) is applied to the lithium-doped side of the junction. Lithium ions drift in the electric field in such a way as to almost exactly compensate the acceptor impurity level in the material; in a week or two, a compensated region about a centimeter thick is produced.

Such detectors are now used in a variety of applications; lithium-drifted silicon detectors are particularly important in nuclear-reaction studies with particles produced by accelerators, and in low energy x-ray work, while lithium-drifted germanium detectors are used mostly for gamma-ray spectroscopy. Germanium detectors having very large volumes can be made by means

of the coaxial-drift technique (6), whereby lithium is drifted from the outer surface of a cylindrical block of germanium toward the middle; sensitive volumes of 50 cm³ are common, resulting in reasonably high gamma-ray detection efficiencies. In all work at very high resolution, cooling the detector and the first signal-amplifying stage is necessary; cryostats cooled with liquid nitrogen are usually employed for this purpose. In other cases thermoelectric coolers are sometimes adequate.

In spite of the value of lithium for compensating impurities, interest is increasing in programs aimed toward producing materials pure enough to make lithium drifting unnecessary, for several difficulties have arisen that are due specifically to the presence of lithium.

1) Radiation damage is exaggerated by the tendency of lithium ions to precipitate at damage sites and thereby to become electrically inactive.

2) Charge trapping is now believed largely due to lithium-defect pairs (7).

3) Since lithium ions are quite mobile in germanium even at room temperature, handling problems are serious, and storage of lithium-drifted germanium detectors at room temperature is out of the question.

Recent work has shown that germanium crystals can be produced with impurity concentrations substantially below 10^{11} impurity atoms per cubic centimeter (8). If 10^{10} atoms per cubic centimeter can be achieved, detectors with sensitive thicknesses of 1 cm can be made without lithium drifting. This advance will change many aspects of the use of germanium detectors, enabling them to be applied in areas where only silicon has been used so far.

Spectrometer Systems

Fully utilizing the properties of semiconductor detectors has demanded advances in signal amplifiers and the associated electronics (9, 10). In particular, electrical noise in the amplifier, which results in broadening of peaks in pulseheight spectra, has been reduced over the past 4 years by a factor of 10 This improvement has principally come from the use of field-effect transistors at low temperatures (11). Significant advances have also been necessary in the data-acquisition aspects of nuclear electronics. The fine detail produced by semiconductor detectors has resulted in a large increase in the number of channels in pulse-height analyzers; the large data flow has made the use of on-line computers almost a necessity.

Semiconductor detectors are now

used extensively for spectrometer systems in all areas of nuclear physics. In the following brief account of some of these applications, we have avoided the standard ones and have chosen, instead, a few cases where use of the detectors has resulted in totally new capabilities in physics or other sciences and other cases that illustrate the present and future potential of these detectors.

X-ray Fluorescence Analysis

Determining the chemical elements in a sample by means of their characteristic x-rays has been an analytical tool for many years, but the advent of high-resolution x-ray spectrometers in which semiconductor detectors are used, combined with the availability of

low-energy x-ray sources such as ⁵⁵Fe, have converted an expensive, cumbersome tool into a convenient one. The method relies on radiation to remove electrons from the inner atomic shells of atoms in the sample; when the resulting vacancies fill, x-rays characteristic of the elements in the sample are emitted. The characteristic x-ray spectra are much simpler and therefore are easier to interpret than optical spectra since only the few electrons in the inner shells are involved. For this type of analysis, crystal-diffraction spectrometers are being replaced by semiconductor-detector x-ray spectrometers, which give much higher detection efficiency and permit simultaneous observation of a wide range of energies. The increase in detection efficiency makes possible the use of much smaller exciting sources than before-radioactive materials such



Fig. 14 (left). The spectrum of alpha particles scattered from a ¹⁵⁴Sm target (beam energy 50 Mev). Fig. 15 (right). The Dopplershift effect used in short lifetime measurements (16). The gamma rays are emitted by decay from the 871-kev excited level of ¹⁷O formed in the reaction $d(^{10}O, p_{\gamma})^{17}O$. Flight of the ¹⁷O nuclei is stopped by an adjustable plunger. In A, the plunger is very close to the target and nearly all decays occur with the ¹⁷O nuclei at rest. In B, the plunger is 2.54 mm away from the target. About 50 percent of the decays occur while the nuclei are flying away from the detector, producing a Doppler shift to lower energies in the gamma-ray spectrum. In C, the plunger is 12.5 mm away from the target; nearly all gamma rays are then Doppler shifted.

as 55 Fe or 241 Am, small x-ray tubes, electron-beam tubes, and small accelerators of charged particles are all being used to advantage. Since 1965 when semiconductor detectors were first suggested for this application (12), a substantial industry has grown up to exploit the potential of x-ray fluorescence systems for a wide variety of analytical problems.

When a scanning electron microscope or an electron microprobe is used for excitation, elemental distributions can be determined in regions as small as 1 micrometer. Typical applications include determination of the distribution of chemical elements in biological tissue and studies of grain boundaries and microscopic nonuniformities in materials. Using proton beams, or preferably heavy-ion beams, which are more efficient than x-rays or electrons in exciting fluorescence, one can detect subnanogram quantities of many elements. A Swedish group has recently described the use of this technique for measuring the elemental composition of particulate matter precipitated from the atmosphere (13). For medical research or diagnosis, an "x-ray photograph" of the thyroid gland can be obtained by scanning the gland with a detector system while exciting the iodine it contains with a suitable gamma-ray source, such as ²⁴¹Am. Determining the composition of pottery sherds has helped archeologists to trace the trade routes of ancient peoples. X-ray fluorescence analysis has even been used to check the authenticity of "old" paintings, by nondestructive analysis of pigments, and also to detect fake coins.

The latest performance capabilities of detectors for x-ray analysis are illustrated in Figs. 12 and 13. In the first of these spectra (14), separation of the x-rays of magnesium, aluminum, and silicon is shown, together with a peak produced by a constant-amplitude electronically generated pulse. The electronic-pulse peak shows the contributions of electronics to the resolution. while the difference between it and the x-ray peaks indicates the charge statistics and collection properties of the detector. The spectrum in Fig. 13 illustrates the abilities of a silicon x-ray detector at very low energies (15). Here the oxygen x-ray (525 ev) produced by x-ray excitation of an Al₂O₃ target is clearly separated from noise counts at the bottom end of the spectrum, despite the difficulty of produc-

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ing characteristic x-rays in elements of low atomic number. Detection of carbon x-rays appears likely to be achieved in the very near future. For measurements of the characteristic K x-rays of the heavier elements, thin-window germanium detectors must be used, since the efficiency of silicon detectors falls rapidly for x-rays with energies greater than 30 kev. In this energy range, and with good low-noise amplifiers, chargeproduction statistics determine the energy resolution of the system, as shown in Fig. 9.

High-Energy Nuclear Spectroscopy

Many of the experiments at cyclotrons and Van de Graaff accelerators involve bombarding a target material with a beam of particles of fairly high energy (~ 50 Mev) and measuring the energy and angular distribution of the particles that emerge. Studies of this nature serve to elucidate the structure of nuclei and their energy levels. Figure 14 shows the energy spectrum of emerging alpha particles, observed with a silicon detector at a fixed angle when



Fig. 16. The spectrum of isotopes produced in fragmentation of uranium nuclei by 5-Gev protons as observed by a detector telescope and particle identifier. This spectrum represented the first observation of the short-lived isotopes ¹¹Li, ¹⁴B, and ¹⁵B (19).



Fig. 17. K⁻-mesonic x-ray spectrum for lead produced by kaon bombardment of a lead-oxide target (20). The highest energy x-ray seen represents a transition to the innermost kaon orbit outside the nuclear surface. The transitions involved in producing the x-rays are indicated on the figure.

a 154 Sm target was bombarded with 50-Mev alpha particles. The peak of higher energy is due to alpha particles elastically scattered from 154 Sm nuclei; the almost equally intense peak lying 82 kev below the main peak is caused by alpha particles whose energy has been reduced by giving up 82 kev to excite the samarium nuclei.

The energy resolution of the system used in this experiment was just adequate to separate the peak due to the 82-kev excited state from that due to the elastic group. Much better energy resolutions (20 kev FWHM at 50 Mev, that is, 0.04 percent) have been obtained in very carefully controlled experiments where the beam quality, target thickness, and slit scattering were all ideal; where the detector was selected and operated at constant low temperature; and where the associated electronics was used under optimum conditions.

It is difficult to make silicon detectors thick enough to detect particles having a longer range, such as 50-Mev protons. Entry of particles into the side of a detector has been used, but the variable energy-absorbing layer at the surface worsens the energy resolution. Experiments with germanium detectors for long-range particles have been fairly successful-resolutions of 18 kev have been obtained for 42-Mev protons. But radiation damage and the problems associated with handling and cooling germanium detectors have inhibited their use. However, the development-probably quite soon-of very pure germanium requiring no lithium drifting may well change the situation drastically. The use of germanium detectors for even higher energies has been demonstrated (16) and promises exciting applications in the future.

Measurement of Very Short

Decay Lifetimes

Although nuclear reactions produce excited states with lifetimes ranging from 10^{-20} second to many years, standard electronic timing methods permit determination only of those longer than 10^{-9} second. A novel method for determining some lifetimes in the range 10^{-13} to 10^{-9} second utilizes the highresolution capabilities of a germanium gamma-ray detector system to measure the Doppler shift in the energy of gamma rays emitted by nuclei that are in high-velocity flight. The method, proposed by Litherland and described by Alexander and Allen (9, 17), has been used, for example, to study the

lifetime of the 871-kev state of ¹⁷O produced by bombarding a deuterium target with 20-Mev ¹⁶O. In the d(¹⁶O, $p\gamma$)¹⁷O reaction, the emerging ¹⁷O nuclei travel predominantly in a narrow forward cone. A plunger stops them at an adjustable distance (ranging from 20 μ m to about 2 cm) from the target. A germanium gamma-ray detector, situated in a backward direction with reference to the beam, observes the gamma rays emitted when the excited ¹⁷O nuclei decay to their ground state. If decay occurs after the nucleus is stopped, the detected gammaray energy is 871 key; the energy is less if decay occurs while the nucleus is in flight (Fig. 15). By measuring the ratio of the intensities of the two gamma rays, and knowing the fragment velocity and the distance from target to plunger, one can determine the lifetime of the excited level in ¹⁷O.

This technique has been extended by the same experimenters to even shorter lifetimes by accurately observing the shape of the gamma-ray peak that results when a nuclear species stops inside a target. Correlation of the observed and the calculated peak-shapes gives information on the lifetime. The method has also been used in studies of the types of fragments produced in nuclear fission.

Particle Identification

High-energy nuclear reactions generally produce many types of particles (Fig. 16), making it necessary to select the specific reaction products desired for study. Passing each product particle through a very thin silicon transmission detector (often less than 20 μ m thick) generates a signal proportional to the rate at which the particle loses energy in silicon; absorbing the remainder of its energy in a second detector, and suitably processing the two signals, permits unique identification of each particle up to a mass number of about 10 (18).

For mass numbers above 10, this identification is not unique: signals due to the heaviest isotopes of one element overlap those due to the lightest isotopes of the next. To eliminate this uncertainty, the first transmission detector in a telescope is moved some distance in front of the remaining detectors, and the time of flight of the particles between detectors is measured. The additional information permits unique identification of mass numbers through 20 (19).

Mesonic X-rays

When a target material is placed in a flux of negatively charged mesons, hybrid atoms are formed in which a meson replaces an electron and goes into orbit very close to the nucleus, far inside the inner shell of atomic electrons. Because mesons are so much heavier than electrons, some of these orbits are so small that they overlap the nucleus itself, acting as a sensitive probe of the fields close to the nucleus and of the nuclear surface. Germanium and silicon detectors give adequate energy resolution to observe the x-ray spectrum generated as these mesons cascade inward, from orbit to orbit, toward the nucleus.

Recent work has studied π^{-} -mesonic and K--mesonic atoms, and Σ --hyperonic atoms, in isotopes covering a wide range of atomic number. X-rays from K--mesonic atoms have yielded interesting data on the distribution of nuclear matter near the surface of nuclei and have shown that a "halo" of nuclear matter extends well beyond the conventional nuclear radius (20). As the K⁻ mesons fall inward, the x-rays produced are seen as a series of lines of increasing energy in the spectrum recorded by the semiconductor-detector system (Fig. 17). Finally, as the meson reaches the nucleus, it is absorbed by a surface particle, and the x-rays suddenly stop. Hence the x-ray line having the highest energy indicates the energy of the innermost transition; a relatively simple calculation yields the radius of the orbit closest to the nucleus. When this measurement is carried out for a range of nuclei of slowly increasing atomic number, the nuclei for which a given x-ray line is just "snuffed out" can be determined, which gives the precise radius of these nuclei.

Activation Analysis

For many years nuclear reactors have been used to "activate" materials. After capturing slow neutrons from the reactors, materials can be analyzed for trace elements by observing the gamma rays emitted upon decay of excited nuclear states. Other methods of activation are also possible, and have specific advantages for the detection of certain elements; for example, ³He activation is a powerful tool for detecting traces of oxygen in materials.

Until the advent of germanium gamma-ray detectors, decay energies could

be observed with reasonable efficiency only by using scintillation detectors. However, as can be seen by comparing the gamma-ray spectra of Fig. 1, germanium detectors increase by a large factor our ability to analyze complex samples and therefore have made a significant difference to all types of activation analysis. Recently, these methods have been applied to the analysis of pottery and other artifacts of interest to archeologists (21).

Conclusion

The past decade has seen the rapid development and exploitation of one of the most significant tools of nuclear physics, the semiconductor radiation detector. Applications of the device to the analysis of materials promises to be one of the major contributions of nuclear research to technology, and may even assist in some aspects of our environmental problems. In parallel with the development of these applications, further developments in detectors for

nuclear research are taking place: the use of very thin detectors for heavyion identification, position-sensitive detectors for nuclear-reaction studies, and very pure germanium for making more satisfactory detectors for many applications suggest major future contributions to physics.

References and Notes

- 1. C. A. Klein, IEEE Trans. Nucl. Sci. NS-15,
- No. 3, 214 (1968).
 R. H. Pehl, F. S. Goulding, D. A. Landis, M. Lenzlinger, Nucl. Instr. Methods 59, 45 M. Lenzlinger, Nucl. Instr. Methods 59, 45 (1968); J. E. Eberhardt, *ibid.* 80, 291 (1970); H. R. Bilger, Phys. Rev. 163, 238 (1967); R. H. Pehl and F. S. Goulding, Nucl. Instr. Methods 81, 329 (1970).
 J. Linhard and V. Nielsen, Phys. Lett. 2, 209 (1962).
 R. O. Bell, N. Hemmat, F. Wald, IEEE Trans. Nucl. Sci. NS-17, No. 3, 241 (1970); J. W. Mayer, M. Martini, K. R. Lanio, I. L. Fowler, *ibid.*, p. 221.

- J. W. Mayer, M. Martini, K. R. Lanio, I. L. Fowler, *ibid.*, p. 221.
 E. M. Pell, J. Appl. Phys. 31, 291 (1960).
 A. J. Tavendale, *IEEE Trans. Nucl. Sci.* NS-12, No. 1, 225 (1965).
 G. A. Armantrout, *ibid.*, NS-17, No. 1, 16 (1970).
- R. D. Baertsch and R. N. Hall, *ibid.*, No. 3, 235 (1970).
 J. W. T. Dabbs and F. J. Walter, Eds., *Semiconductor Nuclear Particle Detectors* (National Conductor Nuclear Particle Detectors). Academy of Sciences-National Research Council Publ. 871, 1961); proceedings of the Tenth Summer Meeting of Physicists at Herceg-Novii, Nucl. Instr. Methods 43, entire volume (1966).

- 10. G. T. Ewan, in Progress in Nuclear Techniques and Instrumentation, F. J. M. Farley, Ed. (North-Holland, Amsterdam, 1968), vol. 3, p. (North-Holland, Amsterdam, 1968), vol. 3, p. 69; W. L. Brown, W. A. Higinbotham, G. L. Miller, R. L. Chase, Eds., Semiconductor Nuclear-Particle Detectors and Circuits (Na- tional Academy of Sciences-National Research Council Publication 1593, 1969).
 E. Elad, Nucl. Instr. Methods 37, 327 (1965). Re-
- H. R. Bowman, E. K. Hyde, S. G. T. R. C. Jared, Science 151, 562 (1966) . Thompson.
- T. B. Johanssen, R. Akselsson, S. A. E. Johanssen, Lund Institute of Technology, Lund, Sweden, private communication.
- 14. D. Landis, F. S. Goulding, J. M. Jaklevic, "Performance of a pulsed-light feedback preamplifier for semiconductor x-ray spectrom-eters," Lawrence Rad. Lab. Rep. UCRLeters," Lawr 19796 (1970).
- 15. J. M. Jaklevic, private communication.
- C. R. Grun, T. Kuo, B. Gottschalk, S. Kan-nenberg, N. S. Wall, *Phys. Lett.* 24B, 266 (1967).
- (1507).
 17. T. K. Alexander and K. W. Allen, Can. J. Phys. 43, 1563 (1965).
 18. F. S. Goulding, D. A. Landis, J. Cerny, R. H. Pehl, Nucl. Instr. Methods 31, 1 (1964); IEEE Trans. Nucl. Sci. NS-13, No. 3, 514 (1965). (1966).
- 19. A. M. Poskanzer, S. W. Cosper, E. K. Hyde, *Phys. Rev. Lett.* 17, 1271 (1966). 20. C. E. Wiegand, *ibid.* 22, 1235 (1969).
- 21. I. Perlman and F. Asaro, Archaeometry 11, 21 (1969).
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Air Pollution Surveillance **Systems**

George B. Morgan, Guntis Ozolins, and Elbert C. Tabor

The activities of man, the intellectual species, must be in harmony with the natural environment. Such harmony, however, will be achieved only when precepts for managing the renewable resources are as firmly entrenched in our individual lore of survival as the tutored reflex to "look both ways before crossing the street." Until that day arrives, organized society will have to compensate collectively for indifferent, careless, or deliberately negligent individuals. Policing the potability of drinking water is representative of such community action on behalf of the individual. Likewise, management of our air resources is increasingly being recognized as a new community responsibility.

Surveillance of air pollution is an integral and very important part of the total effort to control air pollution. The data derived from atmospheric monitoring and emission measurements are required throughout the various stages of the abatement effort. Atmospheric surveillance efforts serve to identify the pollutants emitted to the air, to establish their concentrations, and to record their trends and patterns. Subsequently, after air quality and emission standards have been legislated, surveillance systems may be used to evaluate the progress being made in meeting standards,

and to facilitate direct enforcement activities including the activation of emergency control procedures during episodes of high air pollution.

In the following discussion of surveillance we shall briefly summarize the types of pollutants and their sources, describe the current instrumentation and anticipated developments of more specific and sensitive sensors, and discuss the contrast between the air quality in our nation's urban environment and that of the rural environment or so-called background air quality.

Nature and Origins of Pollutants

Air pollutants can occur in the form of gases, solid particles, or liquid aerosols. These forms can exist either separately or in combinations; for example, gases may be sorbed on particulates or in liquid droplets (1). Gaseous pollutants constitute about 90 percent of the total mass emitted to the atmosphere, and particulates and liquid aerosols make up the other 10 percent.

Gaseous pollutants are evolved primarily from combustion of fuels and refuse. In the case of sulfur oxides, the burning of high-sulfur fuels in stationary sources is the primary source.

The authors are on the staff of the U.S. De-National Air Pollution Control Administration, Bureau of Criteria and Standards, Division of Air Quality and Emission Data, Cincinnati, Ohio.