References and Notes

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Doppler Shift Measurements of Nuclear Lifetimes

The concept of a Doppler shift is utilized in the measurement of lifetimes of nuclear excited states.

Jack R. MacDonald

The study of the excited states of nuclei represents a major area of interest in nuclear structure physics. By virtue of the motion of the protons and neutrons in the nucleus, either individually or collectively, most nuclei can exist in "excited" states with a prescribed (quantized) amount of energy in excess of that possessed by the nucleus in its "ground" state or state of lowest energy. In addition to its energy of excitation, a nuclear excited state can be characterized by its spin (angular momentum), its parity (spatial symmetry), and its decay properties, which include its lifetime and mode of decay. Each of these properties of nuclear excited states can be determined by appropriate experimental techniques. Of particular interest is the lifetime of the excited state, that is, the mean time a nucleus exists with a particular excess energy before it decays to a state of lower energy by the emission of one or more particles or the emission of gamma rays (relatively high-energy electromagnetic radiation). From a knowledge of nuclear lifetimes and other measurable properties of nuclear states, one attempts to deduce a model or theoretical description of the nucleus which may not only explain in a consistent fashion the observed properties of nuclei but may also lead to a greater understanding of the forces which hold nuclei together.

The measurement of nuclear lifetimes by use of a variety of experimental approaches has long been a study of considerable importance. One such technique, which utilizes the concept of a Doppler shift (*1*), was developed to a high degree of sophistication by Devons and others (*2*) in the early 1950's. This technique can, in principle, be applied to the measurement of lifetimes of a large and important class of nuclear states, namely, those which decay by gamma-ray emission. Until recently, however, these measurements were possible only in a relatively few cases. Recent technological advances have greatly facilitated lifetime measurements which utilize the principle of Doppler shifts to such an extent that the techniques are now routinely applied to the study of nuclear structure. In this article, I shall discuss the principle of the method and the experimental techniques and give some examples of experiments which have indeed furthered our knowledge of the structure of nuclei.

The Doppler Shift Principle

The measurement of time or of time intervals always involves the comparison of the unknown interval with a known time interval. For example, the age of a fossil can very often be determined by comparison with a known half-life of a radioactive isotope found in or near the fossil. Even more familiar is the measurement of time by comparison with a standard time interval produced by a mechanical or electrical device known as a clock. By sophisticated techniques, it is possible to measure intervals of time as short as a few tens of picoseconds (1 picosecond $= 10^{-12}$ second) with a form of electronic clock. However, many nuclear excited states have lifetimes much shorter than this, and it is in the region of 10^{-11} to 10^{-14} second that Doppler shift methods for the measurement of nuclear lifetimes have been of enormous importance.

The most familiar example of a Doppler shift is the change in pitch of a train whistle as the train moves toward and away from a listener. This difference in pitch or frequency of the sound waves depends upon the velocity of the train relative to the listener. It is this velocity-dependent change in frequency which is termed Doppler shift. The same concept applies to electromagnetic radiation emitted from a moving source—the frequency (and hence

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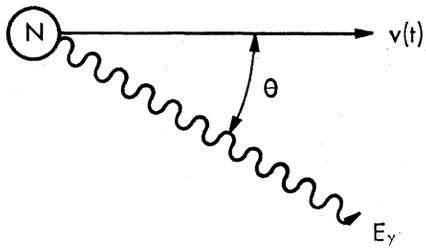


Fig. 1. Schematic diagram of a nucleus moving with velocity $v(t)$ and emitting a gamma ray at angle θ relative to its direction of motion.

the energy) of the radiation is different from what would be measured if the source were at rest. For example, light emitted from a star that is moving away from the earth is observed on earth to have a slightly lower frequency than light emitted from similar atoms on the earth. This phenomenon is known as the "red shift," for the frequency change is equivalent to a shift in wavelength toward the red end of the visible spectrum of electromagnetic radiation. A measurement of this frequency shift indicates the speed at which the star is moving away from the earth. Many radar systems utilize the principle of the Doppler effect to determine the velocity of a moving object. For example, the astronauts aboard the Apollo 11 lunar module measured the velocity of their spacecraft as it approached the lunar surface by use of such a radar system.

The electromagnetic radiation emitted as gamma rays in the decay of many low-lying excited states of nuclei is equally susceptible to Doppler shift, provided that the nucleus decays while in motion with respect to the observer. Consider, for example, a nucleus, moving at velocity $v(t)$, which emits a gamma ray at an angle θ relative to its direction of motion (Fig. 1). If we neglect relativistic effects, the energy of the gamma ray ($E\gamma$) observed at angle θ is related to the energy (E_0) that would be observed if the nucleus decayed at rest by the expression

$$E_\gamma = E_0\left[1 + \frac{v(t)}{c}\cos\theta\right]$$

where c is the velocity of propagation of electromagnetic radiation. The difference between the shifted and unshifted gamma-ray energies (that is, the Doppler shift) is

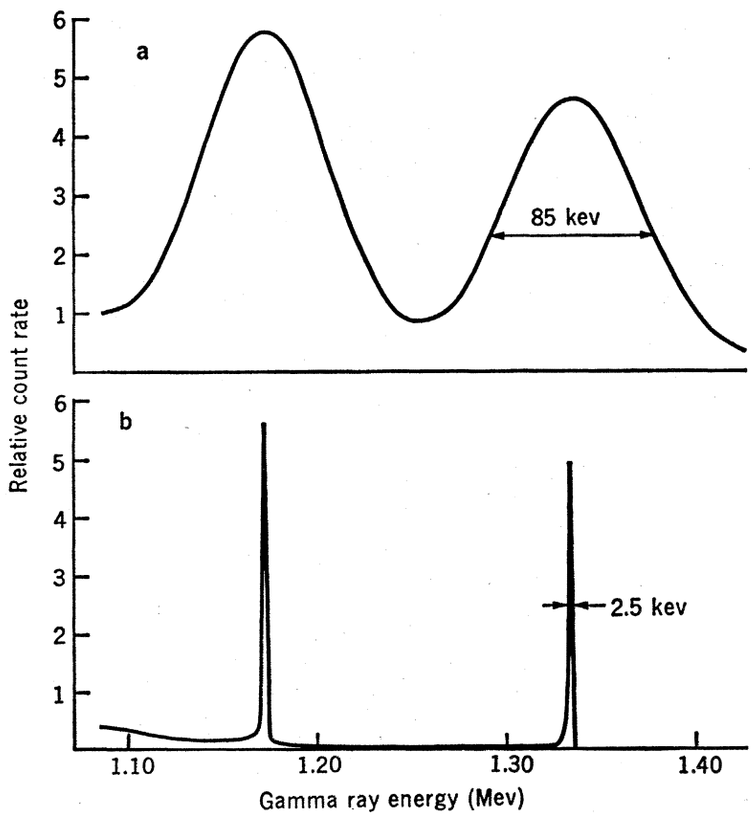
$$\Delta E = \frac{v(t)}{c} E_0 \cos\theta \qquad (1)$$

The extraction of a nuclear lifetime from the measurement of a Doppler shift is dependent upon a knowledge of the velocity of the nucleus as a function of time after the nucleus has been excited to the excited state of interest. A measurement of the Doppler shift establishes the velocity, and hence the time at which the nucleus decays, provided that $v(t)$ changes significantly on a time scale comparable to the lifetime of the nuclear state. This principle of the comparison of lifetimes with velocity is common to all methods of measuring lifetimes by Doppler shift techniques.

Technological Advances

Before I proceed with a more detailed discussion of Doppler shift techniques and experimental measurements, it is useful to comment upon recent technological advances that have made these measurements possible. The accuracy of a Doppler shift measurement, indeed its practicality, depends upon the measurement of small shifts in gamma-ray energies. In a typical case, the Doppler shift as calculated from Eq. 1 may be a few thousand electron volts (kev) for a gamma ray with an energy of 1 million electron volts (Mev).

Prior to 1963, the most practical method for the detection of gamma rays and the measurement of their en-



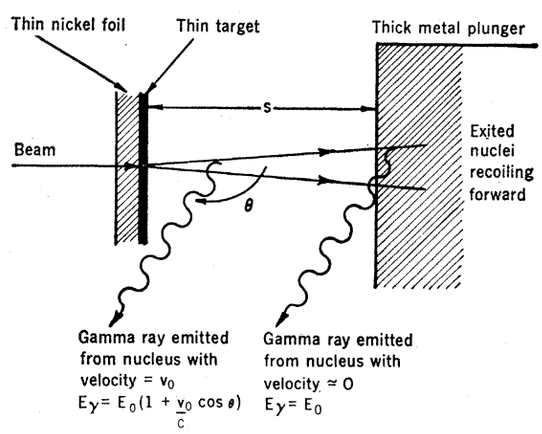


Fig. 2 (left). (a) Response of a NaI(Tl) detector (7.6 centimeters in diameter by 7.6 centimeters long) to the 1.173- and 1.332-Mev gamma rays of ^{60}Co. Only that part of the spectrum which includes the photopeaks is shown. (b) Response of a Ge(Li) detector (effective volume, 30 cubic centimeters) to ^{60}Co gamma rays. Spectra in Fig. 2, a and b, were taken with the detectors at equal distances from the source and for the same period of time. The efficiency of the Ge(Li) detector is approximately 4 percent of the efficiency of the NaI(Tl) spectrometer.

Fig. 3 (above). The recoil-distance method of measuring lifetimes of nuclear excited states.

ergy was to observe their interaction in a thallium-activated sodium iodide crystal, [NaI(Tl)]. The energy deposited in the crystal is given up to electrons which interact with the atoms in the crystal to produce light. The light is observed in a photomultiplier tube, which ultimately produces a voltage pulse whose height is proportional to the energy of the incident gamma ray. Large-volume NaI(Tl) detectors have efficiencies that approach 100 percent for the detection of low-energy gamma rays. However, as a result of the processes by which energy deposited by a gamma ray is converted into a voltage pulse, there is a statistical variation in the height of the voltage pulse for a given incident gamma-ray energy. Figure 2a shows a region in the spectrum of gamma rays (pulse height distribution) of a ^{60}Co radioactive source as measured by a NaI(Tl) cylindrical detector (7.6 centimeters in diameter by 7.6 centimeters long). The energy resolution of the system [full width of the peak at half maximum height (FWHM)] is typical (approximately 85 kev).

Until 1962, the semiconductor detectors which had been used to detect charged particles with an energy resolution of a few tens of a kiloelectron volt were too inefficient to detect gamma rays. However, with the development of the lithium-drift process in silicon (*3*), and more recently in germanium [see, for example, Tavendale and Ewan (*4*)], detectors of larger volume became available and were used for gamma-ray detection. In particular, lithium-drifted germanium detectors [Ge(Li)], which are much more efficient than silicon devices, have demonstrated much better energy resolution than NaI(Tl) crystals. The superior energy resolution is a result of a statistically more favorable energy conversion process in the detector and of a more direct method of obtaining an energy-proportional current or voltage pulse from the device. The parallel development of low-noise amplifiers and high-resolution multichannel pulse height analyzers has enabled experimenters to take full advantage of these remarkable devices. Figure 2b is a pulse height spectrum showing the response of a Ge(Li) detector with a volume of 30 cubic centimeters to the gamma rays of ^{60}Co. This spectrum demonstrates the excellent energy resolution obtainable with such a counter, a property of utmost importance in the accurate measurement of gamma-ray energies. Although the size of such devices at present limits their efficiency to less than 10 percent of that of the NaI(Tl) crystal used to obtain the spectrum of Fig. 2a, their use in the field of Doppler shift measurements has truly revolutionized the study of nuclear lifetimes.

In order to observe the Doppler shift of a gamma ray from the decay of an excited state, the state itself must be excited. One usually brings about this excitation by means of a nuclear reaction which typically can be written as

$$X(x,y)Y^*$$

This notation describes a reaction whereby target nuclei X are bombarded by energetic projectiles x to form the nucleus Y in an excited state (as denoted by the asterisk) and a particle y. In the reaction, total charge, mass-energy, and linear and angular momentum are conserved. Consequently, the number of nuclear states that can be excited by a given reaction at a particular bombarding energy is limited. Early Van de Graaff and cyclotron particle accelerators were severely restricted in the type and energy of projectile that could be accelerated. The development of the Tandem Van de Graaff accelerator in the past 15 years and its subsequent use to accelerate a variety of projectiles to bombarding energies in the range of a few million electron volts to tens of millions of electron volts has enabled experimenters to investigate many nuclear states that could not be formed conveniently with earlier accelerators.

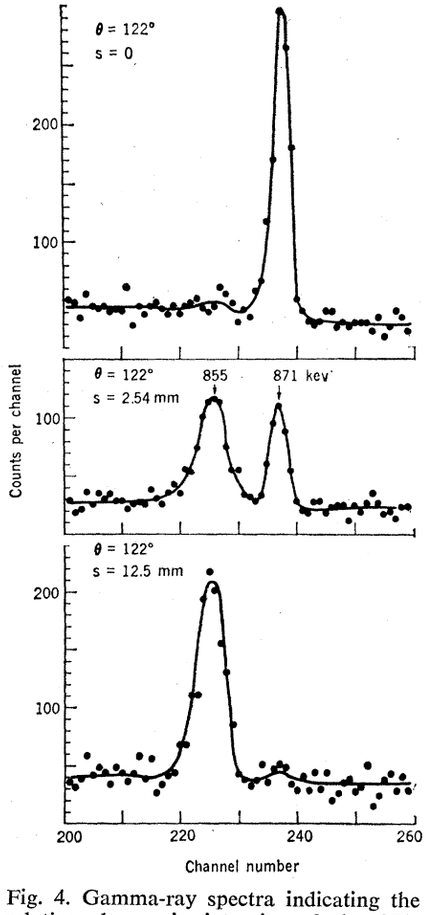


Fig. 4. Gamma-ray spectra indicating the relative change in intensity of the 871- and 855-kev peaks as a function of plunger-to-target distance s.

Recoil-Distance Technique

It has been pointed out above that from a knowledge of the velocity of a recoiling nucleus at the time of decay one can determine the lifetime of a nuclear state from a measurement of the Doppler shift. A particularly simple application of this principle utilizes a stopping plunger to define the velocity distribution and to measure lifetimes as short as a few picoseconds. Alexander and Allen (*5*) have used this technique to measure the lifetime of the first excited state of ^{17}O. Moreover, the experiment indicates the importance of Tandem Van de Graaff accelerators and Ge(Li) detectors in Doppler shift measurements of lifetimes of excited states of nuclei.

Figure 3 illustrates the principle of the method. A beam of 20-Mev ^{16}O ions from a Tandem Van de Graaff accelerator was incident upon a thin deuterium (D) target supported by a nickel foil. The ^{17}O nuclei were produced in their first excited state of excitation energy (871 kev) by the reaction

$$D(^{16}O,p)^{17}O^*$$

and recoiled into a vacuum. As a consequence of the conservation of energy and linear momentum in the reaction, the ^{17}O nuclei were constrained to recoil into a narrow cone with essentially uniform velocity v_0. Gamma rays from those nuclei which decay in flight suffer a Doppler shift given by Eq. 1. Nuclei which strike a metal plunger at a variable distance s from the target stop in a time of the order of 10^{-12} second and decay at rest, and the gamma ray produced in the decay suffers no Doppler shift. The number of nuclei $N(t)$ which have not decayed at time t is given by the radioactive decay law

$$N(t) = N_0 \exp(-t/\tau) \qquad (2)$$

where N_0 is the initial number of excited nuclei and τ is the mean lifetime of the excited state. Consequently, the fraction of nuclei which have not decayed at time $t = s/v_0$ and hence strike the plunger and decay at rest is given by $\exp(-s/v_0\tau)$. The measurement consists of determining the ratio of in-flight (shifted) to stopped (unshifted) events as a function of the target-to-plunger distance s.

At an angle of observation $\theta = 122°$ and for an average recoil velocity of $1.07(10)^9$ centimeters per second, Eq. 1 gives a value of -16.5 kev for the expected full Doppler shift of the 871-kev gamma ray from the decay of the first excited state of ^{17}O. Clearly, the measurement is possible only if one can determine the number of gamma rays observed with shifted energy 854.5 kev relative to those with unshifted energy 871 kev. Figure 4 presents gamma-ray spectra taken with a Ge(Li) detector at various target-to-plunger distances and shows how the fraction of shifted gamma rays changes as a function of this distance. The shifted peak is somewhat broader than the unshifted or stopped peak because the Ge(Li) detector subtends a range of angles near 122° with respect to the cone of recoiling ^{17}O nuclei. The relative intensity of the unshifted peak to the total intensity as a function of the plunger displacement is plotted in Fig. 5. The intensity decreases exponentially with plunger displacement and falls by a factor of e in a distance $s_m = 2.50 \pm 0.22$ millimeters. The error in s_m is the statistical uncertainty in the number of counts in each peak. The mean lifetime of the state is

$$\tau = s_m / v_0$$

or $(2.33 \pm 0.26)(10)^{-10}$ second. In this particular case, the observed lifetime is long enough to permit measurement by direct electronic timing methods; the result is $\tau = (2.51 \pm 0.03)(10)^{-10}$ second. The agreement between the results of the two types of experiments is reasonable and gives considerable confidence that the recoil-distance method can be applied in cases of shorter lifetimes where direct timing measurements are not possible.

The recoil-distance method of defining $v(t)$, and hence of measuring lifetimes, is restricted in its application. Clearly, the technique requires a velocity distribution of recoiling nuclei that is reasonably well defined in both magnitude and direction so that the relative contributions of "stopped" and "shifted" peak components can be determined and related to the lifetime. In many cases nuclear states cannot be excited by a reaction which satisfies this condition. More important is the mechanical limitation on the minimum target-to-plunger distance that can be obtained, and consequently, the limit on the shortest lifetime that can be measured. For recoil velocities of the order of $(10)^9$ centimeters per second, this minimum lifetime is approximately 10^{-12} second. In many cases where the lifetime is shorter than 10^{-12} second or where the recoil-distance technique is not otherwise applicable, it is possible to measure the nuclear lifetime by comparison with the slowing-down time of the nucleus in a solid or gas; this technique is the basis of the Doppler shift attenuation method.

Formulas for the Doppler Shift Attenuation Method

Consider an ensemble of N_0 nuclei produced in an excited state at time $t = 0$. Furthermore, let us assume that these nuclei have an initial velocity v_0, well defined in both magnitude and di-

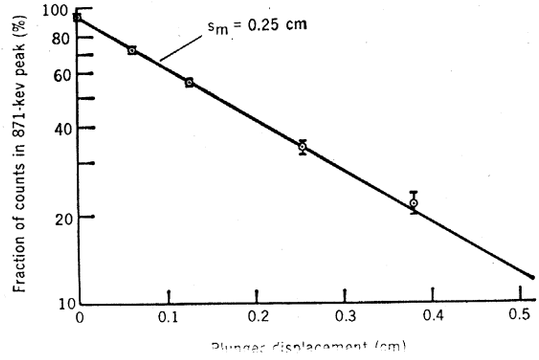
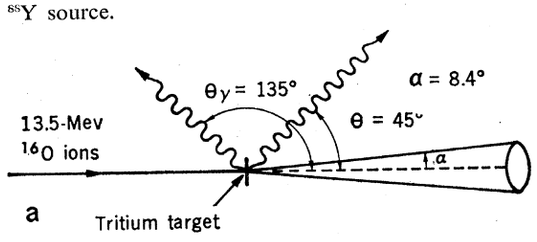
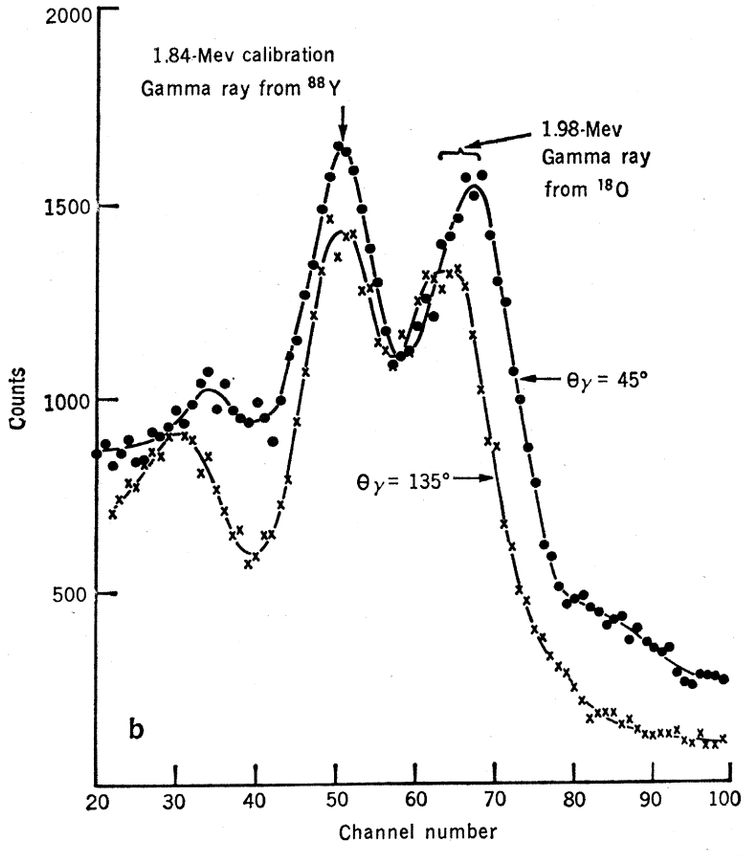


Fig. 5 (above). A plot of the fraction of counts in the 871-kev peak as a function of plunger displacement.

Fig. 6. (Below) (a) The experimental geometry used in the measurement of lifetimes of ^{18}O. The reaction produces ^{18}O nuclei recoiling in a cone of half-angle 8.4°. (Right) (b) Gamma-ray spectra indicating the Doppler shift of the 1.98-Mev gamma ray of ^{18}O relative to the 1.84-Mev gamma ray of a radioactive ^{88}Y source.





rection, and that they decay with a mean lifetime τ by emission of a gamma ray whose unshifted energy is E_0. If these nuclei are moving in some stopping medium, they will slow down and ultimately stop. If the stopping medium is a solid, the time required for the nuclei to come to rest is at most a few picoseconds; if the nuclei are moving in a gas, the stopping time is of the order of tens of picoseconds. As the nuclei slow down, their rate of decay is given by the differentiation of Eq. 2

$$\frac{dN(t)}{dt} = -\frac{N_0}{\tau} \exp\left(-\frac{t}{\tau}\right) \quad (3)$$

where $N(t)$ is the number of nuclei that have not decayed at time t. If the nuclei retain their initial direction of motion while they are slowing down, the average Doppler shift can be obtained in the following manner. The fraction of nuclei which decay between t and $(t+dt)$ is given by Eq. 3; the Doppler shift associated with the decay of these nuclei is given by Eq. 1. The average Doppler shift is obtained by multiplying the fraction of nuclei by the shift and summing or integrating over all time (hence all decays). The result is

$$\Delta E = \frac{E_0 \cos\theta}{c\tau} \int_0^\infty v(t) \exp(-t/\tau)\, dt \quad (4)$$

One obtains a more convenient form of Eq. 4 by relating this average shift to the maximum possible shift

$$\Delta E_{\max} = \frac{v_0}{c} E_0 \cos\theta$$

that would be observed if all nuclei decayed at velocity $v(t) = v_0$. The ratio of observed shift to maximum shift is termed the "attenuation factor" and is given by

$$F(\tau) = \frac{1}{v_0\tau} \int_0^\infty v(t) \exp(-t/\tau)\, dt \quad (5)$$

Equation 5 shows how one may relate the measurement of an attenuated Doppler shift to the mean nuclear lifetime through a knowledge of the velocity of the nuclei as a function of the time during which they are slowing down. The slowing-down time is, in effect, the time standard to which the lifetime is compared. In its present form, Eq. 5 is of limited use because of the restrictions on the initial velocity distribution and subsequent slowing-down characteristics. Experimentally, nuclei are prepared in the excited state of interest by a nuclear reaction which may not produce a monoenergetic, unidirectional beam of recoiling nuclei. Thus it may be necessary to modify the expression for $F(\tau)$ by averaging over the initial velocity distribution. Furthermore, the assumption that nuclei travel in straight lines while slowing down is valid only in special cases. Deviations from this simple behavior require a further modification which follows from a detailed consideration of the mechanism of energy loss of an ion moving in a stopping medium.

There are two mechanisms by which a moving ion (recoiling nucleus with its atomic electrons) loses energy in coming to rest. In the first of these mechanisms energy is lost to the atomic electrons of the stopping material through ionization and excitation—electronic stopping. For ion velocities less than a few percent of the velocity of light, the electronic stopping power or energy loss per unit path length is directly proportional to ion velocity; that is

$$\frac{dE}{dx} = -kv$$

or

$$\frac{d(\frac{1}{2}Mv^2)}{dx} = -kv$$

where dE is the energy lost in the path length dx by an ion of mass M moving at velocity v, and k is a constant of proportionality. Hence

$$\frac{dv}{dx} = -\frac{k}{M}$$

Equivalently

$$\frac{dv}{dt} \cdot \frac{dt}{dx} = -\frac{k}{M}$$

But

$$\frac{dt}{dx} = \frac{1}{v}$$

so that

$$\frac{dv}{dt} = -\frac{k}{M} v$$

This differential equation has the solution

$$v(t) = v_0 \exp\left(-\frac{t}{\alpha}\right) \quad (6)$$

where α (the characteristic slowing-down time) $= M/k$ and $v_0 = v(t=0)$. Insertion of Eq. 6 into Eq. 5 results in the following expression for $F(\tau)$

$$F(\tau) = \frac{\alpha}{\alpha+\tau} \quad (7)$$

As the interaction of heavy ions with electrons results in negligible deflection in the direction of motion, there is little deviation from straight-line motion arising from the electronic loss mechanism. Consequently, Eq. 7 is directly applicable in cases where electronic stopping dominates and the initial conditions on recoil velocity distribution are satisfied. Although these conditions are rarely completely satisfied in practice, the simplicity of Eq. 7 renders it useful as a starting point for more complicated analysis.

As the velocity of an ion decreases, the other stopping mechanism becomes important and often dominant. This mechanism involves a transfer of energy from the moving ion to the translational motion of atoms of the stopping material through elastic scattering processes. As these interactions involve collisions between particles of comparable mass, the moving ion undergoes deflection or scattering through relatively large angles. As a result of this scattering, the initial distribution of direction of velocities is changed and this effect must be taken into account. Blaugrund (*6*) has derived an expression for the time dependence of the average component of ion velocity in the direction of the initial velocity. Equation 5 must be further modified by the replacement of $v(t)$ with the component of $v(t)$ in the direction of the initial motion.

In the general case, recoiling nuclei lose energy by both electronic stopping and large-angle atomic or nuclear scattering processes. Each of these mechanisms for energy loss can be described in terms of a stopping power, dE/dx (energy loss per unit path length traveled), from which $v(t)$ can be extracted. These stopping powers are a function of the atomic numbers and the masses of the moving ion and stopping material and are also dependent upon the velocity of the moving ion and the density of the stopping medium. In some cases, experimental measurements of dE/dx have been made from which $v(t)$ can be inferred. However, in the majority of cases, experimental dE/dx data are not available and it is necessary to use theoretical estimates for both electronic and nuclear stopping powers. The theory of Lindhard *et al.* (*7*) fits most experimental data to an accuracy of approximately ± 20 percent with notable but known exceptions. Velocity distributions obtained by the use of this theory to interpolate between experimentally determined stopping

powers can thus be used with considerable confidence to obtain nuclear lifetimes from a measured Doppler shift.

There are many variations of experimental technique for the determination of lifetimes which depend upon the observation of an attenuated Doppler shift. These differences arise as a result of differences in the methods used to excite a given state by the use of different nuclear reactions, in the method of specifying or limiting the initial recoil velocity distribution, in the stopping material or materials chosen, and in the choice of detector geometry in a given experiment. There are, however, two basic variants of the method in common use. In the first of these one determines the shift in mean energy (or the centroid of the measured gamma-ray energy distribution) of gamma rays observed at two angles and then compares this shift to the maximum possible shift. In the second variant, the gamma rays are observed at one angle and comparison is made for nuclei recoiling into different stopping materials or into a vacuum. An example of the latter approach is given in a later section which deals with the analysis of the shape of an observed gamma-ray peak. In the following discussion I shall describe two of the many variations of the former technique.

Centroid Shift Analysis

The early measurement (1963) of Litherland *et al.* (*8*) of lifetimes in ^{18}O and ^{18}F is a classic example of the use of NaI(Tl) detectors in Doppler shift measurements. These workers populated states of ^{18}O and ^{18}F by means of the reactions

$$T(^{16}O,p)^{18}O^*$$

and

$$T(^{16}O,n)^{18}F^*$$

This type of reaction produces an essentially monoenergetic unidirectional beam of nuclei recoiling with high velocity, ideal for Doppler shift measurements. In particular, the 1.98-Mev first excited state of ^{18}O was populated by the bombardment of a tritium (T) target with 13.5-Mev ^{16}O ions. Excited ^{18}O nuclei, produced with an initial velocity $v_0 = 1.1(10)^9$ centimeters per second, were slowed down and ultimately stopped in a magnesium target backing (Fig. 6). Gamma rays from the decay of the 1.98-Mev level were detected in a NaI (Tl) scintillation counter (12.5 centimeters in diameter by 15 centimeters long) positioned at angles of 45° and 135° with respect to the incident beam. The spectra of Fig. 6 show the shift in energy of the total absorption peak of the 1.98-Mev gamma ray relative to an unshifted gamma ray produced by a radioactive source used for calibration. The observed energy shift of 22 kev is 20 percent of the maximum possible shift

$$\Delta E_{max} = \frac{v_0}{c} E_0 [\cos 45° - \cos 135°] = 110 \text{ kev}$$

that is, $F(\tau) = 0.20$.

In this case the initial recoil velocity is high enough so that electronic stopping dominates. In fact, the initial velocity is so high that the electronic stopping power is no longer proportional to velocity. As a result, Eqs. 6 and 7 are not applicable and the data were analyzed by means of Eq. 5 with a $v(t)$ distribution derived from a combination of experimental dE/dx data and theoretical estimates of the stopping power. The lifetime of the 1.98-Mev state in ^{18}O was found to be 3.7 picoseconds. This and other measured lifetimes in ^{18}O have provided valuable information on the structure of this nucleus.

The ^{40}Ca nucleus has a configuration of neutrons and protons which makes its nuclear structure, and hence the lifetimes of excited states, particularly interesting from a theoretical point of view. In a series of experiments utilizing Ge(Li) detectors, MacDonald *et al.* (*9*), have measured the lifetimes of many of the levels of ^{40}Ca. The states were excited by means of the reaction

$$^{40}Ca(p,p')^{40}Ca^*$$

This reaction, termed inelastic scattering, is one in which protons bombard a calcium target, give up energy to a nucleus, thus leaving it in an excited state, and emerge with correspondingly less energy. The energy possessed by the inelastically scattered protons is indicative of which state has been excited. Figure 7 indicates the experimental arrangement for these measurements. A beam of protons from a Tandem Van de Graaff accelerator passes through an annular solid-state particle detector and is incident upon a self-supporting ^{40}Ca target with an approximate thickness of 2 milligrams per square centimeter (0.03 millimeter). Protons inelastically scattered to 170° are detected in the solid-state counter, thus signifying the excitation of a given nuclear level. Furthermore, as a consequence of the conservation of energy and linear momentum, the associated ^{40}Ca nuclei recoil in the forward direction with a well-defined, uniform velocity. These nuclei ultimately come to

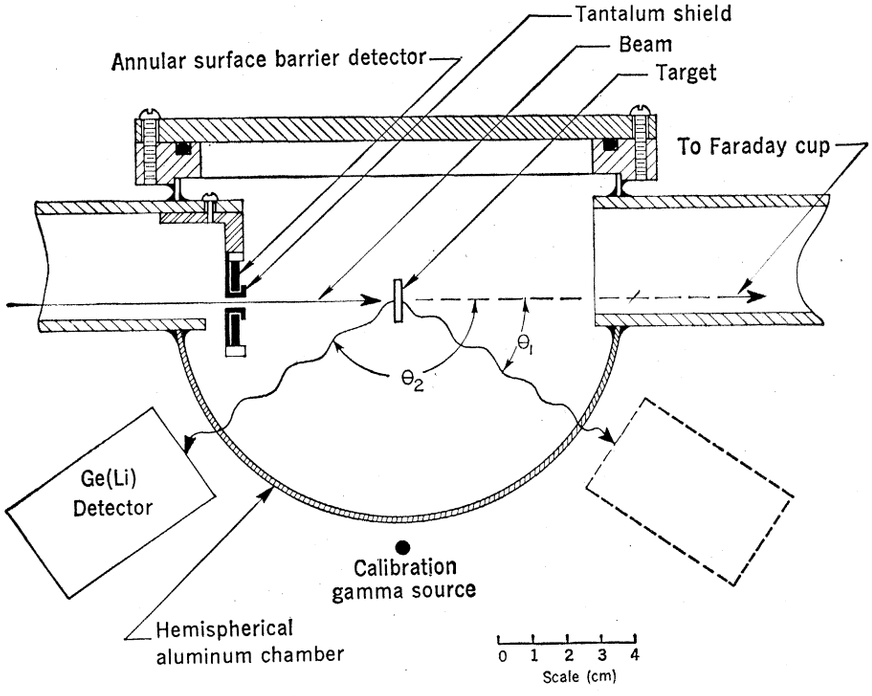


Fig. 7. Experimental arrangement used by groups at Oxford and Rutgers universities to measure lifetimes through excitation of states by inelastic scattering reactions. Gamma rays are observed at forward and backward angles in coincidence with particles backscattered into an annular particle detector.

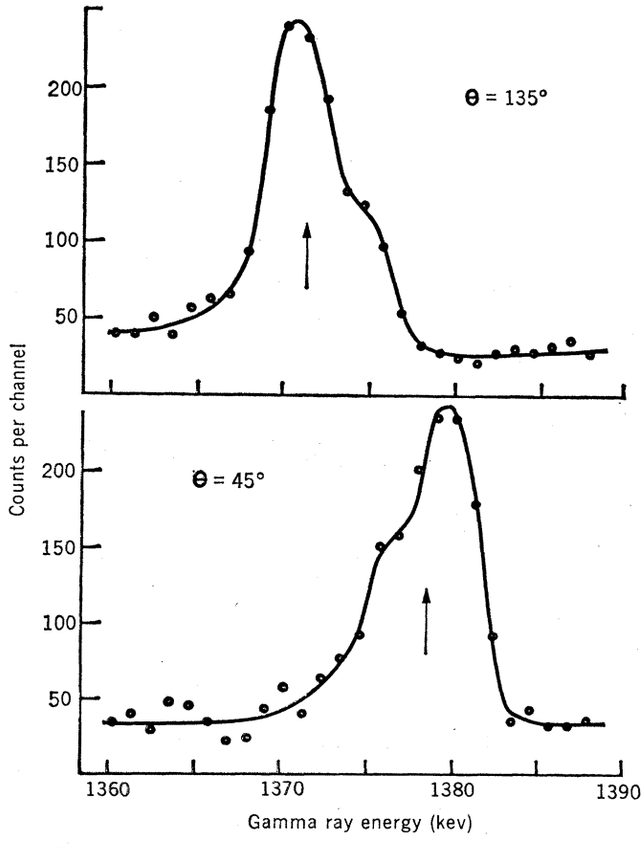
rest in the target material itself. Gamma rays, in time coincidence with a given proton, are detected in a Ge(Li) detector at forward and backward angles of observation. Figure 8 shows gamma-ray spectra in coincidence with protons populating the 5.28-Mev level of ^{40}Ca. The state is observed to decay by emission of a 1.37-Mev gamma ray to a state at 3.91 Mev, which in turn decays to the ground state. The observed shift of 7.15 ± 0.35 kev represents an attenuation factor of 0.65 ± 0.03. As the initial nuclear recoil velocity is only $2(10)^8$ centimeters per second, the mechanism by which energy is transferred to the atoms of the stopping material through elastic scattering is dominant and large-angle deflections of the recoiling nuclei must be taken into account. Figure 9 shows the result of a computer calculation of the attenuation factor F as a function of mean lifetime τ based on theoretical estimates of the velocity-time distribution and the average angle of deflection. The observed attenuation corresponds to a mean lifetime of $(3.3 \pm 0.4)(10)^{-13}$ second.

The sequence of levels in ^{40}Ca at 3.35, 3.91, and 5.28 Mev have excitation energies and angular momentum properties characteristic of a deformed (ellipsoidal) rather than a spherical shape. This, in turn, implies that these states are the result of the collective motion of many nucleons rather than that of a single nucleon. The decay properties of these states should reflect this collective behavior and should indicate the amount of deformation. For example, the 5.28-Mev level should decay to the 3.91-Mev level at a rate considerably faster than that for a non-collective type of transition. The decay rate or lifetime of the 5.28-Mev state has been calculated, based on the assumption that the initial (5.28-Mev) and final (3.91-Mev) states involved in the transition have a collective structure. The calculated lifetime is in good agreement with the experimentally observed value. This result, and similar agreement for decay of the 3.91-Mev level, is extremely strong evidence for the existence of deformed states in ^{40}Ca.

Line Shape Analysis

In addition to the fact that the spectra in Fig. 8 exhibit shifts of the peak centroids, these spectra are also characterized by shapes which do not resemble the fully shifted or unshifted peaks of Fig. 4. This shape is characteristic of the nuclear lifetime and, in addition, is dependent upon details of the stopping mechanism which must be assumed in a centroid analysis. In those cases where the shift in gamma-ray energy is comparable to or greater than the intrinsic energy resolution of the detector, it is thus desirable that in the data analysis the peak be fitted with a calculated line shape in order that information on both lifetime and stopping power may be extracted.

As one can see from Eq. 1, there is a one-to-one correspondence between the observed gamma-ray energy and the instantaneous velocity $v(t)$ (more precisely, the projection of that velocity on the direction of initial recoil). There is, therefore, a continuous distribution of gamma-ray energies corresponding to recoil velocities from v_0 to 0, and this distribution of gamma-ray energies or the peak line shape is thus dependent upon the velocity-time relation and the mean lifetime. In the general case of electronic and nuclear stopping mechanisms and recoil ion deflections, $v(t)$ does not have a convenient analytical form and the line shape must be calculated by computer analysis. It is instructive, however, to see what line shape results from the assumption that the velocity decreases exponentially



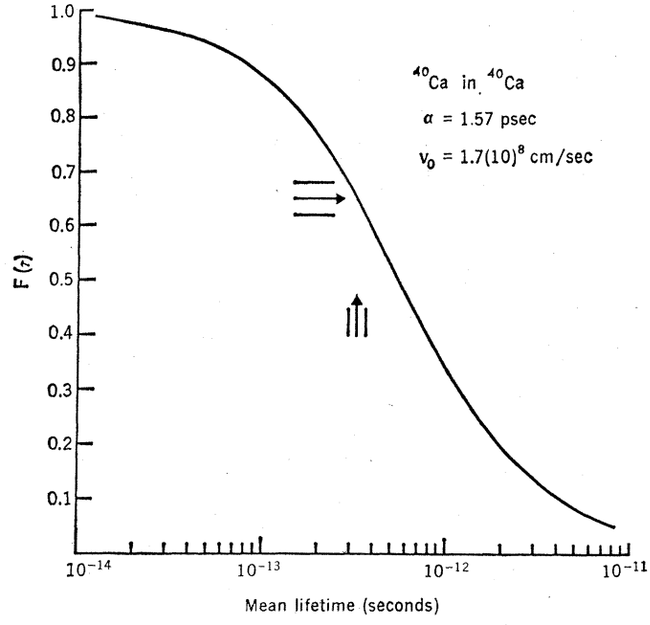


Fig. 8 (left). The Doppler shift of the 1.37-Mev gamma ray (5.28- to 3.91-Mev transition) in ^{40}Ca. The indicated peak centroid shift of 7.15 kev is to be compared with a maximum possible shift of 11.0 kev.

Fig. 9 (above). A graph of the attenuation factor $F(\tau)$ as a function of mean nuclear lifetime τ for ^{40}Ca stopping in ^{40}Ca. The experimentally determined value of $F(\tau) = 0.65 \pm 0.03$ corresponds to a mean lifetime of $(3.3 \pm 0.4)(10)^{-13}$ second for the 5.28-Mev level.

with time and that the nuclei travel in straight lines during the stopping process. From Eqs. 2 and 6 one can calculate the fraction of nuclei which decay with velocity between v and $(v+dv)$. If we define V as the ratio $v(t)/v_0$, we obtain

$$\frac{dN(V)}{N_0} = \frac{\alpha}{\tau}(V)^{\alpha/\tau - 1}\, dV \qquad (8)$$

Equation 8 is an expression for the fraction of nuclei which decay with a mean lifetime τ and with velocities between $v = Vv_0$ and $(v + dv)$; therefore, Eq. 8 represents the energy distribution of gamma rays. Figure 10 shows histograms of the theoretical line shapes for various ratios of α/τ and indicates the attenuation factor in each case. These are not the line shapes that are observed experimentally, however, because of the finite energy resolution of the gamma-ray detectors. Furthermore, the restrictions implied in the derivation of Eq. 8 are rarely satisfied. Indeed, it is from a consideration of experimental line shapes that one is able to extract information on the distribution of velocities as a function of time and hence to lessen the dependence of the Doppler shift method on theoretical estimates of stopping powers.

Warburton and his co-workers (*10*) have developed techniques for the analysis of line shapes and have applied their method to the measurement of many lifetimes of states in ^{22}Na. These states have been excited by means of the reaction

$$^{19}\mathrm{F}(\alpha,\mathrm{n})^{22}\mathrm{Na}^*$$

This reaction is endothermic; that is, the bombarding energy of the alpha particle must be greater than approximately 2 Mev before even the ground state of ^{22}Na can be formed. As a result, the formation of ^{22}Na states could be controlled by variation in the beam energy. Furthermore, near the threshold for the excitation of a given state, there is little spread in the energy of the recoiling excited nuclei and the nuclei recoil in a narrow angular cone with respect to the initial direction of the beam. These favorable initial conditions make possible measurements of the Doppler shifts of gamma rays without recourse to coincidence techniques, and, as a result, the high counting rates provide peaks with good statistical accuracy amenable to line-shape analysis.

Figure 11 shows the observed line shapes of the gamma ray produced by the decay of the nucleus from the 1.528-Mev state to the ground state. The three spectra are from experiments in which the recoiling nuclei were stopped in three different materials—calcium fluoride (CaF_2), strontium fluoride (SrF_2), and what was believed to be isotopically pure metallic lithium-6 ("^{6}Li"). Each spectrum exhibits a "stopped" peak at the unshifted energy $E\gamma_0$ and a broad tail on the high-energy side, corresponding to gamma rays emitted at a finite recoil velocity. The ratio R indicates the relative proportion of "stopped" to shifted contributions to the peaks and changes by a factor of 2 as one goes from CaF_2 to "^{6}Li" backings. The solid curves superimposed on the CaF_2 and SrF_2 data were obtained by treating the mean lifetime and the ratio of electronic to nuclear stopping as parameters which were varied to obtain the best fit to the data. The experimenters used a value of α which was consistent with all available information on experimental and theoretical electronic stopping powers. An attempted fit to the "^{6}Li" data, based on the expected value of $\alpha = 2.1$ picoseconds for ^{6}Li and a lifetime as determined by the first two measurements, gave unsatisfactory results. This can be explained on the assumption that the chemically active ^{6}Li had oxidized or was otherwise chemically altered, and an effective value of $\alpha = 1.37 \pm 0.3$ picoseconds was extracted from the

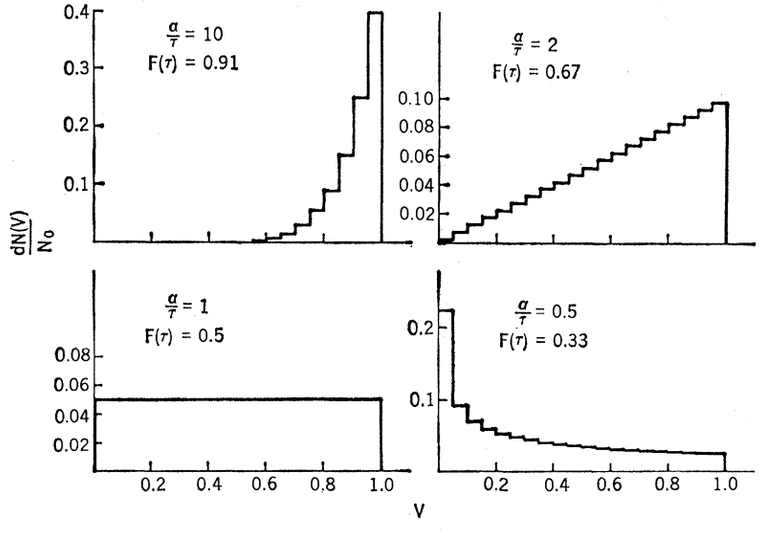


Fig. 10 (above). Theoretical line shapes for various ratios of stopping time to mean lifetime based on the assumption that electronic stopping is the only stopping mechanism.

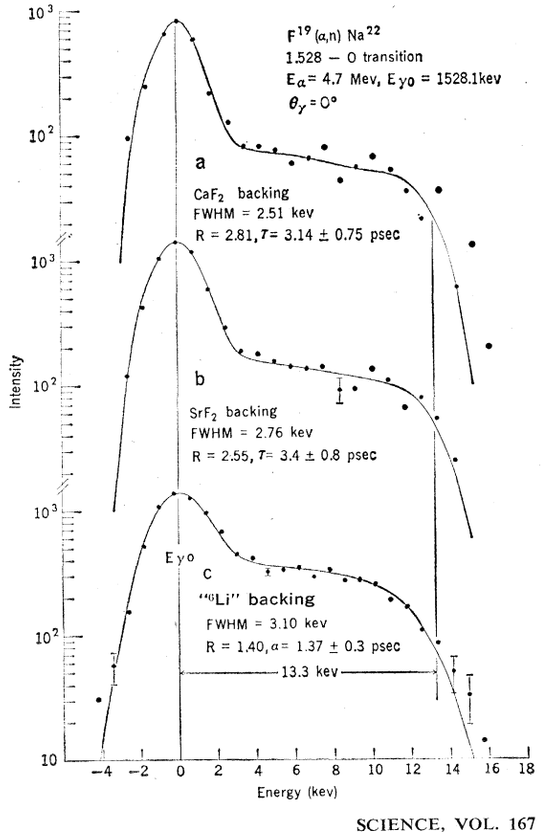


Fig. 11 (right). The line shape of the 1.528-Mev gamma ray for recoil into different target backings. The solid curves are theoretical fits to the data.

measured line shape and known lifetime. This procedure facilitated the use of "^{6}Li" as a backing in other experiments where the long slowing-down time led to increased experimental accuracy. This case demonstrates the caution that must be taken in these measurements, for a single determination of the lifetime with a backing assumed to be pure ^{6}Li metal and with $\alpha = 2.1$ picoseconds would clearly give an incorrect result.

In general, analysis of the same data by line shape and centroid methods yields the same value of the nuclear lifetime, although the former method is usually more accurate. Furthermore, the ratio of electronic to nuclear stopping determined by line-shape analysis usually shows good agreement with estimates based on a combination of available stopping power data and theoretical estimates. This agreement indicates the adequacy of theories of the stopping mechanism used in the centroid shift method of lifetime analysis.

Summary

It has been my intent to show how the concept of a Doppler shift can be utilized in the measurement of nuclear lifetimes. The emphasis has been placed on the mechanics of the experimental techniques with only passing comment on the significance of the results and their importance in nuclear structure physics. Other techniques for the measurement of lifetimes have not been discussed. In many cases, these techniques are more direct and accurate and are used, whenever possible, in preference to Doppler shift methods. In circumstances where a lifetime is accurately known from other experiments, the Doppler shift attenuation technique may be used to extract information on the stopping of ions in solids. This reverse application of the method has been little explored as yet. Nonetheless, measurement of nuclear lifetimes by use of a concept 127 years old is an intriguing and important branch of physics.

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Blood Flow and Diffusion through Mammalian Organs

Stochastic methods used for chemical reactor analysis are readily applicable to complex biologic systems.

James B. Bassingthwaighte

In recent years a quiet revolution has been occurring in the medical sciences. While intuitive and empirical therapeutics will retain its important role in the art that the individual physician applies to the practice of medicine, the capabilities and the incentives to apply scientific approaches increase each day. The appropriate tools are being provided by the physical scientists —primarily engineers—but also by theoreticians.

Because of the complexity of biologic systems, it is difficult to describe them in terms of deterministic models in which one attempts to define the behavior of the physical and chemical components by sets of equations. Complexity and accuracy must be sacrificed to simplicity and approximation in order to produce working hypotheses.

One approach that physiologists and many others have found satisfactory is to describe components of a system in terms of their responses to known inputs. The response to an impulse or a brief pulse input may be an output function which has a magnitude varying with time after the input. Thus, the response is expressible as a probability density function of magnitudes at a sequence of times; the empirically determined impulse response is a "stochastic" description of the behavior of a system. There are other types of stochastic responses—for example, those which occur after a constant time interval but with variable magnitudes. Until we can fathom the nature of the *components* of such systems, we must work with various empiric stochastic descriptions of their behavior.

As knowledge of a system grows, parts of it become describable in a deterministic fashion, usually in terms of simplified mathematical models, while other parts remain stochastic. One reason that biologic systems are so resistant to description by deterministic models is that the responses to a given input change from moment to moment: the systems are nonstationary. The "nonstationarities," and, indeed, apparent nonlinearities, are usually due to uncontrolled and unrecognized inputs or to cyclic fluctuations in subsystems. Such unrecognized inputs may be of a wide variety of physical or chemical forms and may be continuous or quantized variables or even pulse trains of varying frequency.

The system with which we are ultimately concerned is the intact hu-

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