[9]H. Becker, E. Dietz, U. Gerhardt, and H. Angermüller, Phys. Rev. B 12, 2084 (1975).
[10]N. V. Smith and L. F. Mattheiss, Phys. Rev. B 9, 1341 (1974).
[11]U. von Barth and L. Hedin, J. Phys. C 5, 1629 (1972).
[12]J. Hermanson, Solid State Commun. 22, 9 (1977).
[13]G. D. Mahan, Phys. Rev. B 2, 4334 (1970).
[14]W. Kohn and L. J. Sham, Phys. Rev. 140, A1133 (1965); R. Gaspar, Acta Phys. Acad. Sci. Hung. 3, 263 (1954).
[15]V. Korenman, J. L. Murray, and R. E. Prange, Phys. Rev. B 16, 4032 (1977).
[16]D. G. Dempsey and Leonard Kleinman, Phys. Rev. Lett. 39, 1297 (1977).
[17]W. Eib and S. F. Alvarado, Phys. Rev. Lett. 37, 444 (1976).

Quantum Beats of Recoil-Free γ Radiation

Gilbert J. Perlow
Argonne National Laboratory, Argonne, Illinois 60439
(Received 9 February 1978)

Recoil-free γ rays from the decay of ^{57}Co are frequency modulated by vibrating the source with a piezoelectric crystal and one of the lines of the resulting multiplet emission spectrum is absorbed. The remaining radiation displays a time-dependent counting rate whose harmonic composition and relative phases are sensitive to small energy shifts and can be used for their measurement.

When a γ-ray emitter is vibrated sinusoidally with amplitude x_0 along the direction of observation, and with angular frequency Ω, the time dependence of the radiation field can be expressed as

$$E(t,t_0)=\begin{cases}\exp[-\lambda(t-t_0)/2+i(\omega_0 t+a\sin\Omega t)], & t\geq t_0,\\ 0, & t<t_0,\end{cases} \tag{1}$$

where λ^{-1} is the mean lifetime of the excited nuclear state (the lower state is assumed to be stable). The quantity $a\equiv\omega_0 x_0/c$ is called the modulation index. The origin of time has been chosen as a zero of the sine, and the decaying state was formed at $t=t_0$. Irrelevant normalization and phase factors have been omitted. In what follows, t_0 is never measured and must be averaged over. If one forms the average, $\langle|E(t,t_0)|^2\rangle_{t_0}$, the variable t disappears, so that, as expected, there is no time dependence of the intensity. The spectrum corresponding to Eq. (1) is obtained by squaring its Fourier transform and averaging over t_0. It is the familiar sum of Lorentzian-shaped carrier and sidebands, first shown with Mössbauer radiation by Ruby and Bolef[1] and observed and discussed by others since.[2-6]

$$I(\omega)=\sum_{n=-\infty}^{\infty}J_n^2(a)/\{[\omega-(\omega_0+n\Omega)]^2+\lambda^2/4\}, \tag{2}$$

where the J_n are Bessel functions of the first kind.

If we now interpose a resonant absorber between the vibrating source and the γ-ray detector, so that there are alterations in phase or amplitude among the components, a time dependence appears in the intensity. It contains the frequency Ω and its harmonics.

In Fig. 1(a), we see the ordinary Mössbauer velocity spectrum of a source of ^{57}Co diffused into a 12-μm foil of Cu which is cemented to one face of a 0.5 mm×9 mm diameter X-cut quartz crystal. The opposite face is cemented to an aluminum backing. The spectrum is made by scanning with a (slowly) moving absorber of ^{57}Fe-enriched sodium ferrocyanide. There is no voltage across the piezoelectric crystal. In Fig. 1(b), an rf generator has supplied 10 V at 9.95 MHz to the crystal and one sees the carrier and sideband pattern described by Eq. (2). In Fig. 1(c), the central carrier has been nearly eliminated by interposing a thick stationary absorber of ^{57}Fe in Be just after the source. Fe-Be has a broad resonance, actually an unresolved doublet, whose centroid corresponds closely to the energy of the ^{57}Co-Cu emission line. The ferrocyanide analyzing absorber is now removed and the radiation responsible for Fig. 1(c) is counted with a thin NaI scintillation counter in a fast timing circuit.

 © 1978 The American Physical Society

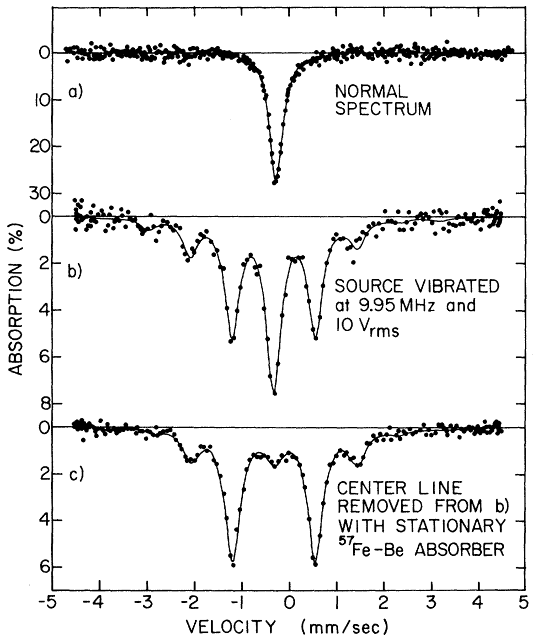

FIG. 1. Velocity spectra (a) unmodulated, (b) frequency modulated at 9.95 MHz, (c) frequency modulated and filtered.

While in principle one need only record the clock time of the detection of selected 14.4-keV γ rays, it is enormously easier to measure the time with respect to synchronizing pulses repeated at a subharmonic of the rf generator frequency. The latter produce the "stop" pulses for a time-to-amplitude converter (TAC). The "start" pulses come from a fast pulse-amplitude discriminator which makes a rough selection of the desired events without destroying the timing accuracy. Finally, the TAC output goes to a pulse-height analyzer gated by pulses from a slow differential discriminator. The net result is a spectrum of intensity versus time for the frequency-modulated and filtered 14.4-keV γ radiation.

A typical case is seen in Fig. 2. The data have been fitted with a sum

$$N = N_0\left[1 + \sum_{m=1}^{6} D_m \cos m\Omega(t-\tau_m)\right]. \qquad (3)$$

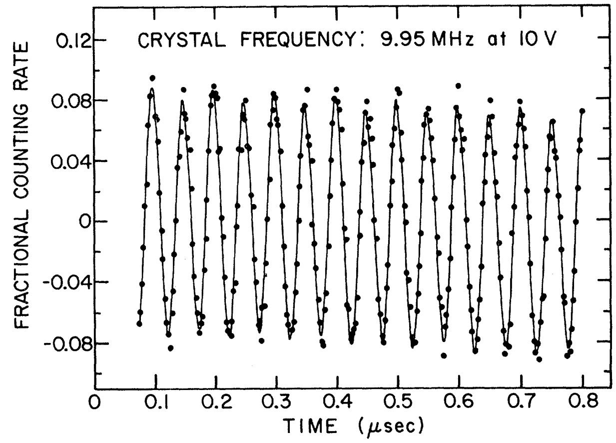

FIG. 2. Time spectrum with the radiation of Fig. 1(c).

The fitting program searches on N_0, the D_m, the time phases τ_m, and Ω, the latter to allow for small errors in the calibration. In addition, because of a deadtime effect in the TAC, it was found necessary to allow N_0 to have a linear variation with time. (A slight downhill slant may be seen in the plot.) If the crystal voltage is increased to 25 V from 10 V, the index a, and therefore the number of sidebands showing significant intensity, increases. Time data with this voltage were also obtained and results for both voltages are summarized in Table I. It is striking that the odd harmonics, including the intensity D_1 of the fundamental, are quite small relative to the even ones. In the 10-V case, the only significant term is D_2 (7.9%), while for 25 V D_4 is also important (1.0%). D_6, which corresponds to a frequency of 59.7 MHz, is not well determined because of insufficient time resolution. The odd harmonics D_1 and D_3 are 0.4% and 0.2% respectively. The suppression of odd harmonics occurs because the absorption or transmission is an even function of $\Delta\omega$, the frequency separation of the centroids of absorber and emitter.

Passage through the resonant medium changes a frequency-modulated photon into one with some

TABLE I. Experimental Fourier amplitudes in %.

Crystal voltage (V)	D_1	D_2	D_3	D_4	D_5	D_6
10	0.38(6)	7.91(6)	0.23(6)	0.27(7)	0.15(10)	0.08(8)
25	0.38(4)	6.60(4)	0.15(4)	1.03(4)	0.06(4)	0.07(4)

amplitude modulation.[7] Independent of the time of formation of the nuclear state, the modulation maintains a fixed phase with respect to laboratory time. It is an individual quantum phenomenon. The quantity $1/\lambda$ is 0.14 μsec, while the mean spacing between recorded events is typically 2500 μsec. The overlap between successive quanta is negligible and in any case, incoherent in phase.

The time dependence could be obtained in a manner analogous to that used by Lynch, Holland, and Hamermesh in analyzing time filtering in a coincidence experiment.[8] The Fourier transform of Eq. (1) would be multiplied by a frequency-dependent transmission function to describe the amplitude after passage through the resonant medium, then transformed back to the time domain, squared, and averaged over t_0. If the absorption of a line is nearly complete, however, it is considerably simpler to subtract from Eq. (1) a term representing the amplitude of the absorbed line, say the kth one, $E \rightarrow E'$ and, for $t \geqslant t_0$:

$$E'(t,t_0) = \exp[-\lambda(t-t_0)/2]\{\exp[i(\omega_0 t + a\sin\Omega t)] - J_k(a)\exp[i(\omega_0 + k\Omega)t]\}, \tag{4}$$

$$I(t) = \langle|E'(t,t_0)|^2\rangle_{t_0} = 1 + J_k^2(a) - 2J_k(a)\,\mathrm{Re}\{\exp[i(a\sin\Omega t - k\Omega t)]\},$$

$$I(t) = 1 + J_k^2(a) - 2J_k(a)[J_0(a)\cos k\Omega t + 2\cos k\Omega t\sum_{n=1}^{\infty} J_{2n}(a)\cos 2n\Omega t + 2\sin k\Omega t\sum_{n=0}^{\infty} J_{2n+1}(a)\sin(2n+1)\Omega t]. \tag{5}$$

If $k=0$, the spectrum takes the simple form

$$I(t) = 1 - J_0^2(a) - 4J_0(a)\sum_{n=1}^{\infty} J_{2n}(a)\cos 2n\Omega t. \tag{6}$$

It contains only the even harmonics, as observed.

One must take into account in Eq. (6) the background from 14.4-keV γ rays that have suffered recoil, and from Compton scattering of higher-energy radiation. Each contributes a factor of about 0.25. With this, the maximum of D_2 would be ~ 0.3 at $a \sim 1.2$. Actually, a unique value of modulation index and of modulation phase is not to be expected.[1-5] There is a variation over different parts of the source. If φ is a random phase added to the argument of the sine in Eq. (1), then $2n\varphi$ is added to the argument of the cosine in Eq. (6). If φ had uniform probability over the interval $0-2\pi$, the beats would vanish.

The derivation gives insight into the origin of the phenomenon. It appears here as interference between the frequency-modulated photon and the spectral hole produced by its partial absorption. All the observed time dependence is, in fact, due to interference between the different frequency components of the photon wave function, and it is for this reason that the term quantum beats applies. The interference exactly cancels when all components are present.

There is an application of the beat phenomenon that is potentially important for sensitive experiments, for example, for the measurement of relativistic energy shifts.[9] If the central emission line is partially absorbed, the ratio D_1/D_2 of the fundamental to second harmonic of the oscillator frequency is a rapidly varying function of $\Delta\omega$, and is zero at exact resonance. Simple arguments lead to the expectation of linear behavior

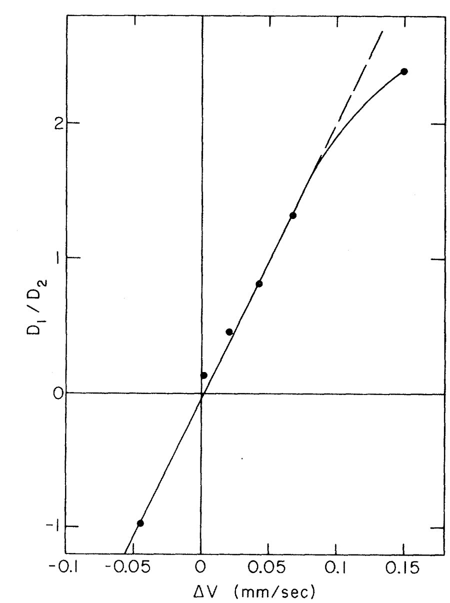

FIG. 3. Ratio D_1/D_2 vs Δv, the velocity displacement of an absorber from resonance with the central line of the ^{57}Co-Cu source.

near $\Delta\omega = 0$. To test this, the Fe-Be absorber was removed and a mock stationary absorber was made of the moving sodium ferrocyanide absorber, by gating the data pulses so that they were counted only over a narrow interval of velocity. By using various velocity intervals and observing and fitting the beat spectra, the curve of Fig. 3 was obtained. Note that D_1/D_2 changes from ~0 to 2 in a shift $\Delta v = 0.1$ mm/sec. Note also the change in phase ($D_1 \rightarrow -D_1$) through resonance. Thus both the magnitude and the sign of the shift can be obtained.

The quantum beats are directly related in phase to the mean phase of the motion of the source. One may set a time gate over some interval of the beats by use of a differential pulse-height discriminator set on the output pulses of the TAC. With data pulses counted only during the gate interval, one may then take the usual Mössbauer velocity spectrum. Interesting and strange looking spectra result from such a mixture of the time and frequency domains, whether one employs the stationary absorber or not. In the latter case, there are no beats, but the time gating still operates. In Fig. 4 one sees such a spectrum (with the stationary Fe-Be absorber in place) gated over a portion of the beat cycle. Without the restrictive gating it would be exactly the spectrum of Fig. 1(c). One now sees dispersionlike transmission curves with counting rates in *excess* of the background at some velocities.

The writer is indebted to H. J. Lipkin, J. E. Monahan, and M. Peshkin for enlightening discussions. He is grateful to W. Potzel for the dialogue that led to the experiment, and to J. E. Breitling for some preliminary calculations. H. E. Stanton and G. F. Kara rendered invaluable service in the data analysis. Some brief but perceptive comments by M. Blume were most welcome.

This work was supported under the auspices of the U. S. Department of Energy, Division of Physical Research.

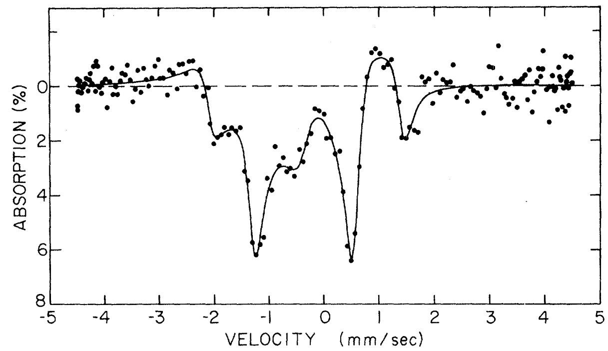

FIG. 4. Velocity spectrum with the conditions of Fig. 1(c) but gated during a short portion of the beat cycle.

[1]S. L. Ruby and D. I. Bolef, Phys. Rev. Lett. 5, 5 (1960).

[2]T. E. Cranshaw and P. Reivari, Proc. Phys. Soc. (London) 90, 1059 (1967).

[3]A. Abragam, *L'Effet Mössbauer* (Gordon and Breach, New York, 1964), pp. 22–24.

[4]L. Mishory and D. Bolef, in *Mössbauer Effect Methodology*, edited by I. Gruverman (Plenum, New York, 1968), Vol. IV, pp. 13–35.

[5]L. Pfeiffer, N. D. Heiman, and J. C. Walker, Phys. Rev. B 6, 74 (1972).

[6]G. J. Perlow, W. Potzel, R. M. Kash, and H. deWaard, J. Phys. (Paris), Colloq. 35, C6-197 (1974).

[7]Amplitude-modulated Mössbauer radiation can also be obtained by mechanically interrupting the radiation from a source. See for example S. L. Ruby, R. S. Preston, C. E. Skov, and B. S. Zabransky, Phys. Rev. A 8, 59 (1973); U. Hauser, W. Neuwirth, and N. Thesen, Phys. Lett. 49A, 57 (1974); V. K. Voitovetskii and S. B. Sazanov, Pis'ma Zh. Eksp. Teor. Fiz. 23, 112 (1976) [JETP Lett. 23, 97 (1976)]. The modulation is observed in the velocity spectrum.

[8]F. J. Lynch, R. E. Holland, and M. Hamermesh, Phys. Rev. 120, 513 (1960).

[9]See for example R. V. Pound, in *Workshop on New Directions in Mössbauer Spectroscopy*, AIP Conference Proceedings No. 38, edited by G. J. Perlow (American Institute of Physics, New York, 1977), p. 41.