fall into two categories: There is one isolated zero, $E(q)$, which lies below the quasicontinuum of p-h states $\omega_k(q)$ and is determined by

$$\Omega_d \int dk/[\omega_k(q)-E]=1, \quad \Omega_d = UV_c/(2\pi)^d. \quad (22)$$

Here, $V_c$ is the volume of the unit cell, $d$ is the number of dimensions, and the integral extends over the whole Brillouin zone. All other zeros of (20) fall inside the limits of the band and can be obtained by contour integration.[6] One finds

$$E_p(q)=\omega_p(q)-\frac{1}{\pi}\Delta_p(q)\tan^{-1}\left\{\frac{\mathfrak{N}(E_p(q))}{D_0(E_p(q))}\right\}, \quad (23)$$

where

$$\mathfrak{N}(E,q)=\pi\Omega_d\int_{BZ} dk\,\delta\{E-\omega_k(q)\}, \quad (23a)$$

$$D_0(E,q)=1+\Omega_d \mathrm{P}\int_{BZ} dk\{E-\omega_k(q)\}^{-1}. \quad (23b)$$

In Eqs. (23), $\Delta_p(q)$ is the spacing of two successive poles $\omega_k(q)$ at wave vector $p$, and P denotes the principal value integral. Equations (21)–(23) are the desired expressions for the pair wave functions and energies. It can be seen from Eqs. (22) and (23) that for $U \gg t$ (atomic limit), the energies $E(q)$ (homopolar states[5]) are separated from the $E_p(q)$ (ionic states[5]) by a gap of $\sim U$.

In one dimension, where $\epsilon_k = -2t\cos(ka)$ ($a$ is the lattice constant), Eqs. (22) and (23) reduce to

$$E(q)=U-\{U^2+(4t\sin\tfrac{1}{2}qa)^2\}^{1/2} \quad (24)$$

and

$$E_p(q)=U-2t\{\cos(\mathfrak{L}_p a)-\cos(\mathfrak{L}_p+q)a\}, \quad (25)$$

where

$$\mathfrak{L}_p = p \pm \frac{\pi}{Na} - \frac{2}{Na}\tan^{-1}\left\{\frac{2t}{U}[\sin(p+q)a-\sin(pa)]\right\}. \quad (25a)$$

In (25a), the upper or lower sign applies according to whether the argument of the arctangent is positive or negative, respectively. Equation (25a) is correct up to, and including, terms of the order $1/N$. It can be shown[9] that the energies given by (24) and (25) are identical to those derived by Lieb and Wu[2] for $M=1$. Although the details are too lengthy to present here, essentially this result follows because, as previously mentioned, the interaction term of (19) yields no contribution in this case. A more detailed account of the present work will be published elsewhere.[9]


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## Coherent Transient Effects in Mössbauer Spectroscopy


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Coherent transient effects in Mössbauer spectroscopy due to phase modulation of recoilless $\gamma$ radiation are considered both theoretically and experimentally. Absolute calibration of the source motion in the angstrom range and separation of the source and absorber contributions to the experimental linewidth were obtained from a single transient Mössbauer spectrum. A new phase-modulation method for generating short enhanced recoilless $\gamma$ pulses is introduced.


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In a recent Letter[1] anomalous line shapes were reported in Mössbauer experiments with sinusoidal phase modulation at frequencies close to the linewidth of the Mössbauer state. Decaying interference oscillations appear in such transient Mössbauer (TM) spectra. Here, a general formula is derived for the time dependence of Mössbauer transmission due to phase modulation of $\gamma$

radiation. The formula is applied to sinusoidal and stepwise phase modulation. Experimentally the modulation is generated by moving the source mechanically. An accurate calibration of the source motion in terms of the wavelength of the $\gamma$ radiation is obtained from the oscillations present in the TM spectra. The source and absorber contributions to the linewidth are separated from the decay constant. Stepwise modulation is utilized in generation of short enhanced $\gamma$ pulses, caused by the phase coherence of the $\gamma$ radiation. These effects have not been reported before, but they bear similarities to transient effects observed in other resonance phenomena, such as NMR[2] and laser spectroscopy.[3,4]

In this work the intensity of the transmitted $\gamma$ radiation is measured as a function of time synchronized to the phase of the source motion. A similar method is used in conventional and in quantum-beat Mössbauer measurements.[5] Because of the low frequency of the motion no transient effects appear in conventional measurements, whereas in quantum-beat measurements at high frequencies only steady-state oscillations appear.

A series expansion for the time dependence of Mössbauer transmission has been derived for the interpretation of quantum-beat spectra.[6] Fast time-dependent effects have also been considered in coincidence measurements,[7] where the time reference comes from the instant $t_0$ at which the excited state is formed, and in theoretical studies on synchrotron radiation.[8] There is a difference of principle between our measurement in the time domain and measurements where only the energy spectrum of the phase-modulated $\gamma$ radiation is determined.[9,10]

The time dependence of the intensity of recoillessly emitted $\gamma$ radiation transmitted through a resonance absorber is derived classically. A quantum mechanical calculation analogous to that of Harris[11] would give similar results. The electric field of $\gamma$ radiation emitted recoillessly from a Mössbauer source is

$$E_s(t)=\exp\{i\omega_s(t-t_0)-\Gamma_s(t-t_0)/2+i[\varphi(t)-\varphi(t_0)]\}\theta(t-t_0), \tag{1}$$

where $\omega_s$ is the center frequency, $\Gamma_s$ is the width of the source line, $\theta(t)$ is the step function, and $\varphi(t)$ is the time-dependent phase shift. If the phase shift is generated by changing the displacement $x(t)$ between the source and the absorber, then $\varphi(t)=x(t)/\lambda$, where $2\pi\lambda$ is the wavelength of the $\gamma$ radiation.

The amplitude $E(t)$ of the radiation transmitted through a resonance absorber can be calculated as a convolution of the absorber function $A(t)$ and $E_s(t)$. The former is[8]

$$A(t)=\delta(t)-b\exp(i\omega_a t-\Gamma_a t/2)\sigma(bt)\theta(t), \tag{2}$$

where $\sigma(x)=J_1(2\sqrt{x})/\sqrt{x}$. Here $\omega_a$ is the center frequency and $\Gamma_a$ the width of the absorber line. The parameter $b=T_M\Gamma_0/4$ depends on the Mössbauer thickness of the absorber $T_M$ and on the natural width of the resonance $\Gamma_0$. If there is no inhomogeneous line broadening $\Gamma_a=\Gamma_s=\Gamma_0$. Equations (1) and (2) are strictly valid only if the inhomogeneous broadenings in the source and absorber are Lorentzian. For a thin absorber ($T_M<4$) $\sigma(x)\simeq\exp(-x/2)\cos(\alpha x)$, where $\alpha=\pi/2x_1\simeq 0.43$ and $x_1$ is the first zero of $\sigma(x)$. The main effect of this approximation is to increase $\Gamma_a$ to $\Gamma_a+b$.

The transmitted intensity is proportional to $\langle|E(t)|^2\rangle_{t_0}$ where an integral over $t_0$ has been taken (in contrast to coincidence experiments). A calculation gives a general result for the relative transmission:

$$[N(t)-N_0]/N_0=2f_s\,\mathrm{Re}\Big(-bF_+(t)\int_{-\infty}^{t}dt'\,\sigma(b(t-t'))/F_+(t')$$
$$+b^2\exp(-\Gamma_a t)\int_{-\infty}^{t}dt'F_-(t')\sigma(b(t-t'))\int_{-\infty}^{t'}dt''\sigma(b(t-t''))/F_+(t'')\Big), \tag{3}$$

where

$$F_\pm(t)=\exp[-(\Gamma_s\pm\Gamma_a)t/2-i\Delta\omega t+i\varphi(t)], \tag{4}$$

$N(t)$ is the transmitted intensity, $N_0$ is its off-resonance value, $f_s$ is the recoilless fraction of the source radiation, and $\Delta\omega=\Delta\omega_a-\omega_s$. The first term in Eq. (3) can be understood as the interference between the source radiation and the radiation scattered coherently in the forward direction. The second term is solely due to coherent forward scattering. It is a retarded term which does not react instantaneously to rapid changes in $\varphi(t)$.

We shall first consider sinusoidal phase modulation: $\varphi(t)=(x_0/\lambda)\cos\Omega t$, where the modulation is generated by sinusoidal vibration with an amplitude $x_0$. An approximate formula for the relative trans-

mission was calculated from Eq. (3), under the assumption that the resonance is passed rapidly (transient effects are seen after the resonance). The result is

$$[N(t)-N_0]/N_0 = -f_s 2b[2\pi/\ddot{\varphi}(t_r)]^{1/2}\exp[-\Gamma_T(t-t_r)]+\cos[\varphi(t)-\varphi(t_r)-\Delta\omega(t-t_r)-\pi/4] + [f_s b^2 2\pi/\ddot{\varphi}(t_r)]\exp[-(\Gamma_a+b)(t-t_r)], \quad (5)$$

where $2\Gamma_T=\Gamma_s+\Gamma_a+b$ is the total linewidth and $t_r$ is the time at which the system is in resonance: $\dot{\varphi}(t_r)=\Delta\omega$. Equation (5) is valid when $t-t_r > [\ddot{\varphi}(t_r)]^{-1/2}$ and $\ddot{\varphi}(t_r) > \Gamma_T^2$. The dots denote derivatives with respect to time. The first term of Eq. (5) shows oscillations which decay with time constant $\Gamma_T$. The frequency of these oscillations is mainly determined from the phase difference $\varphi(t)-\varphi(t_r)$. The second term causes a nonoscillating background which decays with a time constant $\Gamma_a+b$. Because the two terms of Eq. (5) have different time and thickness dependencies they can be separated when the measurement is suitably performed. Thus much more information can be obtained from such transient spectra as compared with conventional Mössbauer measurements.

A transient Mössbauer spectrum is shown in Fig. 1. The 93.3-keV Mössbauer resonance of $^{67}$Zn is utilized. The damped oscillations predicted by the theory are evident. It should be noted that most of the radiation entering the absorber is scattered in the forward direction. The spectrum was fitted with the aid of Eq. (3). The coherency of the motion at frequencies even higher than those used in the measurement of Fig. 1 was confirmed by studying the amplitude dependence of the transmitted intensity.[9,12] From the fit (solid line) the parameters listed in Table I were obtained. The determination of the vibration amplitude $x_0$ is absolute in terms of $\lambda$, and the achieved accuracy, 0.002 Å, cannot easily be obtained by other methods. In this measurement, the amplitude $x_0$ was chosen in such a way that the influence of the other spectral lines is still small. In contrast to conventional measurements the values of the total linewidth $2\Gamma_T$ and the center shift $\Delta\omega$ are quite independent of the $x_0$ determination. The values of $f_s$ and $T_M$ can be determined in spite of inhomogeneous broadenings. In addition, the source and absorber linewidths were also fitted separately with the following results: $\Gamma_s=1.60(44)\Gamma_0$ and $\Gamma_a+b=2.07(42)\Gamma_0$. With better statistics even $\Gamma_a$ and $b$ could be separated. From a conventional measurement such separations are extremely difficult.

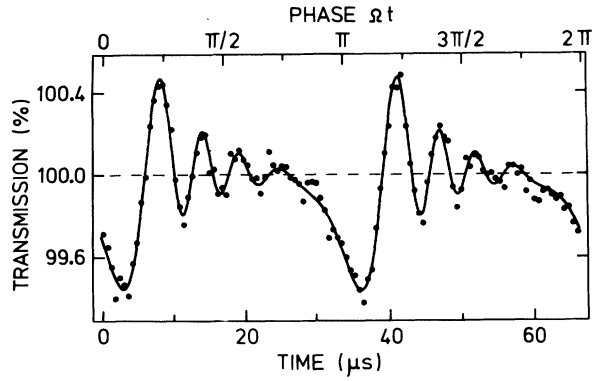


FIG. 1. A transient Mössbauer spectrum with a sinusoidal phase modulation at frequency $\Omega/2\pi=15$ kHz. The source is a $^{67}$Ga:ZnO single crystal and the absorber is made of 90%-enriched $^{67}$ZnO powder with a thickness of 1.0 g $^{67}$Zn/cm$^2$. The motion was generated with a piezoelectric PZT transducer. The $\gamma$ rays were detected using a NaI scintillator. The spectrum was recorded with a time-to-digital converter. The measurement temperature is ≃4 K. The broken line indicates the nonresonant background intensity.

In Fig. 2 a TM spectrum is shown for the 14.4-keV Mössbauer resonance of $^{57}$Fe. Now much higher modulation frequencies are needed since for $^{57}$Fe $\Gamma_0/2\pi \simeq 1.1$ MHz, whereas for $^{67}$Zn it is only 12 kHz. Full coherency of the source motion can no longer be assumed. Because $\ddot{\varphi}(t_r) \simeq \Gamma_T^2$, the transient oscillations predicted by Eq. (5) are efficiently damped. In conventional Mössbauer measurements $\dot{\varphi}(t)$ is approximately constant during the lifetime of the Mössbauer state and no transients appear. Then Eq. (3) reduces to a conventional transmission integral.

Let us next consider the following phase modulation: $\varphi(t)=\varphi_0+\Delta\varphi\,\theta(t)$, where $\varphi_0$ and $\Delta\varphi$ are constants. This can be realized by moving the source at time $t=0$ a distance $\Delta x=\Delta\varphi\lambda$. Before and after the step the source is stationary relative to the absorber. The change in the transmission was

TABLE I. Results from the fit of Fig. 1.

| | |
|---|---|
| $x_0$ | $13.6(1)\lambda = 0.287(2)$ Å |
| $\Delta\omega$ | $-0.20(4)\,\Gamma_0 \cong -0.032(6)$ μm/s |
| $2\Gamma_T$ | $3.6(2)\,\Gamma_0 \cong 0.58(3)$ μm/s |
| $f_s$ | 0.020(3) |
| $T_M$ | 2.8(5) |

calculated from Eq. (3), under the assumption that $\Delta\omega = 0$:

$$\frac{N(t) - N(t<0)}{N_0} \simeq 2f_s b(1 - \cos\Delta\varphi)\exp(-\Gamma_T t)\frac{\Gamma_T \cos(\alpha b t) - \alpha b \sin(\alpha b t)}{\Gamma_T{}^2 + (\alpha b)^2} \times\left(1 - b\frac{\Gamma_d - \exp(-\Gamma_d t)[\Gamma_d \cos(\alpha b t) - \alpha b \sin(\alpha b t)]}{\Gamma_d{}^2 + (\alpha b)^2}\right)\theta(t), \quad (6)$$

where $\Gamma_d = (\Gamma_a + b - \Gamma_s)/2$. The transient has its maximum value when $\Delta\varphi = (2n+1)\pi$ (i.e., the displacement $\Delta x$ is equal to an odd number of half wavelengths) and vanishes if $\Delta\varphi = 2n\pi$. After the jump at $t = 0$ there is an approximately exponential decay in the intensity with time constant $\Gamma_T$. The maximum value of the jump for a thick absorber is $4f_s$, thus being as much as 4 times the maximum of the conventional absorption. Especially if $f_s \simeq 1$, the change in the transmitted intensity can be drastic.

An experimental verification of these effects is shown in Fig. 3. As predicted by the theory, associated with the phase changes there are simultaneous intensity changes which die out exponentially. With $\Delta\varphi = \pi/2$, the slight asymmetry between the transmission steps corresponding to forward and backward motions is due to nonzero $\Delta\omega$.[13] This asymmetry can be used in an accurate determination of the center shift $\Delta\omega$. When $\Delta\varphi = \pi$, the effect is more than 2 times the absorption reached in conventional measurements. The width of the pulses observed at $\Delta\varphi = 2\pi$ is determined by the finite rise time of the modulation steps. The solid curves are least-squares fits by Eq. (3), with $\varphi(t)$ shown in the upper part of Fig. 3.

The microsecond pulses generated by stepwise modulation [Fig. 3(c)] can be applied for timing experiments. These pulses are far more inten-

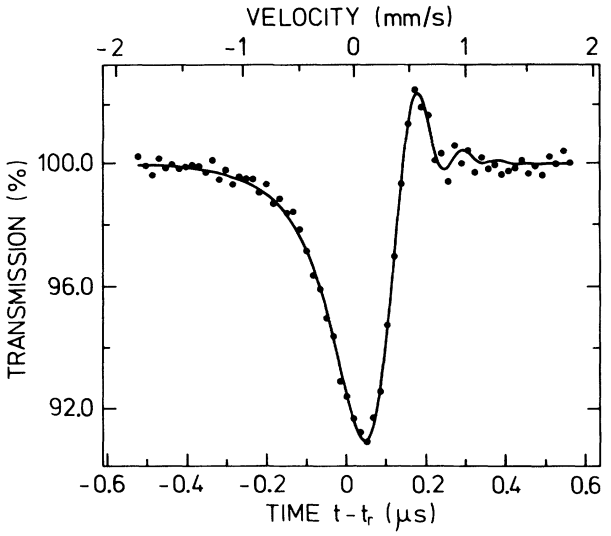


FIG. 2. Part of a transient Mössbauer spectrum measured with sinusoidal phase modulation at a frequency of 190 kHz. The maximum velocity is 3.0 mm/s. The line is shown unfolded in time scale. The source is $^{57}$Co:Rh and the absorber is made of $K_4Fe(CN)_6 \cdot 3H_2O$ powder with a thickness of 0.24 mg $^{57}$Fe/cm$^2$. The spectrum was recorded using a time-to-amplitude converter.

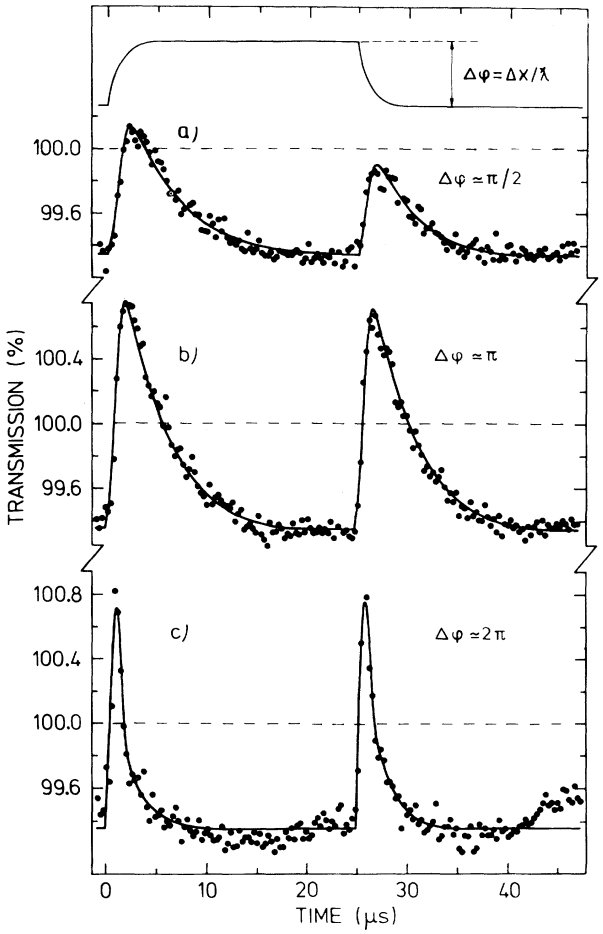


FIG. 3. Mössbauer transmission curves obtained using stepwise phase modulation. The interval between the steps is 25 μs. The phase changes were generated by moving the source by an amount $\Delta x$ with a quartz transducer. The source is a $^{67}$Ga:ZnO single crystal and the absorber is made of enriched $^{67}$ZnO powder with a Mössbauer thickness $T_M \simeq 2.4$.

sive than pulses produced by chopping the beam mechanically, which would also otherwise be very difficult. Classically, the enhanced transmission can be explained as an interference between the source field and the field of the radiated absorber. As compared with the proposed use of synchrotron radiation the present pulse method offers a considerably smaller energy spread which can be controlled with the aid of the pulse length.

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