we get for the path distribution

$$p(l) = (3/8R^3)l^2 \quad \text{for } l \leq 2R ,$$

$$p(l) = 0 \quad \text{for } l > 2R .$$

Since there is no correlation between the directions and the velocities of the atoms, the distribution function for τ, $W(\tau)$ is given by the following expression:

$$W(\tau)\, d\tau = d\tau \int_0^{2R/\tau} w(v)\, p(\tau v)\, v\, dv ,$$

where v is the Jacobian of the coordinate transformation $(l, v) \rightarrow (\tau, v)$ and

$$W(\tau) = \frac{3\pi\tau^2}{2R^3}\left(\frac{m}{2\pi kT}\right)^{3/2} \int_0^{2R/\tau} v^5 \exp\left(-\frac{mv^2}{2kT}\right) dv ,$$

which leads to the final expression

$$W(\tau) = \frac{27}{2\pi^2}\,\frac{\tau}{\tau_f^2}\left[1 - \exp\left(-\frac{4\pi}{9}\,\frac{\tau_f^2}{\tau^2}\right) \times \left(1 + \frac{4\pi}{9}\,\frac{\tau_f^2}{\tau^2} + \frac{8\pi^2}{81}\,\frac{\tau_f^4}{\tau^4}\right)\right] ,$$

where $\tau_f = \frac{3}{2} R(2m/\pi kT)^{1/2} = \langle l\rangle_{av}\langle v^{-1}\rangle_{av}$ is the average time of flight, under the assumption that there is no correlation between the path distribution and the velocity distribution.

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Mechanically Chopped γ Rays; Quantitative Treatment of the Sideband Intensities*

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We report the generation of sidebands in the energy spectrum of 14.4-keV γ rays by a periodic modulation of the properties of the medium between the emitting and absorbing atoms. The observed spectrum agrees in detail with a "chopping" calculation that encompasses both amplitude and phase modulation, although the modulator may equally well be viewed as a moving multislit grating. This is the first experiment in which the observed sidebands in the γ-ray spectrum have been quantitatively understood.

INTRODUCTION

Since the advent of the Mössbauer effect, it has become possible to observe a number of interesting phenomena connected with the modulation of γ-ray wave packets. Lynch *et al.*[1] attenuated certain frequency components in the γ radiation and observed that the distribution of delays between the formation of the nuclear state and the detection of the γ ray was no longer exponential. Ruby and Bolef[2] produced a sinusoidal phase modulation of the wave packets and observed that the usual single-line Lorentzian frequency distribution was replaced by a central line with a succession of uniformly spaced sidebands on either side. Grodzins and Phillips[3] shifted the phase linearly with time and observed the shift in the frequency of the single Mössbauer line. Champeney[4] has observed the irregular frequency distribution of γ rays that have traversed finely divided matter moving at right angles to the direction of propagation. In each of these investigations, the γ ray was treated as a classical damped oscillation modulated by a particular interaction.

Until recently, periodic amplitude modulation of γ rays had not been attempted. Then Kamenov[5] reported that he had tried to amplitude-modulate Mössbauer γ rays by sending them through a rotating chopper, but had been unable to observe any perturbation of the frequency distribution. In a similar experiment, Isaak and Preikschat[6] observed a line broadening which they attributed to a new pair of lines, each separated from the central line by a spacing equal to the modulation frequency.

We have also performed an experiment of this kind. The purpose of this paper is to demonstrate that in such experiments there is phase modulation as well as amplitude modulation, and also that once the dimensions of the chopper are known, the details of the frequency spectrum of the chopped radiation are completely and accurately determined by the chopping frequency and by the complex indices of refraction of the chopper materials. We wish to emphasize that, contrary to a recently published argument,[7] it is not necessary to consider this experiment as a case of diffraction from a moving grating. Rather, it is perfectly valid and sometimes more convenient to calculate what the chopping does to the amplitude and phase of individual rays emerging from the chopper, and then to determine the frequency spectrum by Fourier analysis.

EXPERIMENT

The experimental arrangement is shown in Fig. 1. The apparatus included a usual Mössbauer spectrometer working with ^{57}Fe. A single-line source was moved relative to a single-line absorber which was fixed in front of a proportional counter. Between the source and absorber was the rotating chopper (the modulator). This was a 15-cm-diam wheel made of 12 sectors. Each sector consisted of a single layer of parallel (nearly radial) copper wires of diameter $d = 0.0022$ cm and with a center-to-center spacing $S = 0.0028$ cm. The copper wires were imbedded in plastic resin and sandwiched between two 0.013-cm Mylar sheets. The γ-ray beam collimation was such that only the outermost centimeter of the chopper intercepted the beam. By rotating the wheel at rates up to 500 rev/sec, the modulation period T_m could be made as short as 128 nsec, which is 90% of the mean life τ of the nuclear excited state.

The Mössbauer spectra obtained at different rotational speeds of the chopper are shown in Fig. 2. It is immediately obvious that the periodic modulation of the γ radiation by the chopper has changed the spectrum from a single Lorentzian line to a central line with a series of symmetrically placed sidebands on each side. The clarity of the observed sidebands, as compared with those in Ref.

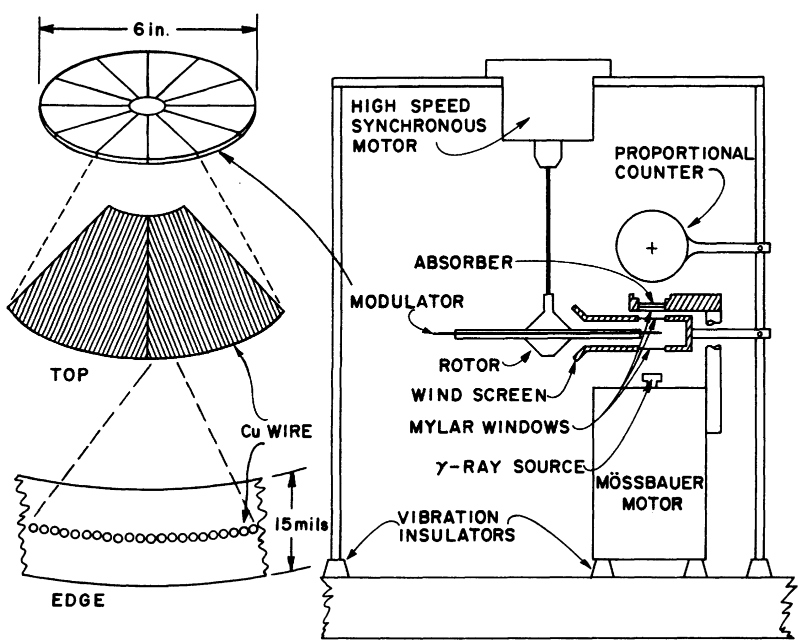

FIG. 1. Schematic diagram showing the physical arrangement of the experiment.

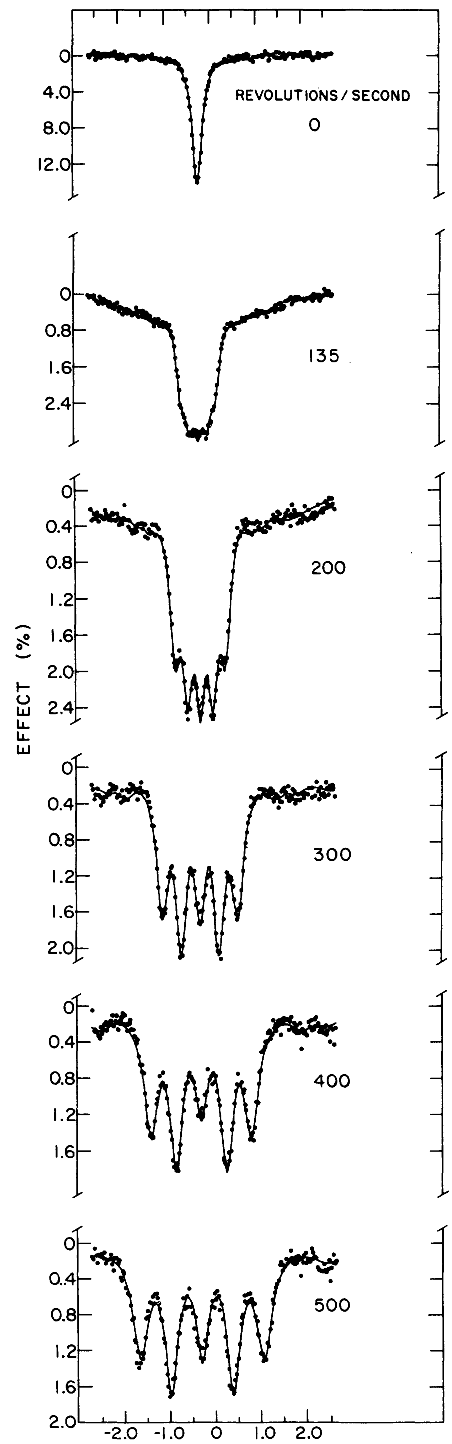

FIG. 2. Experimental data (points) and theoretical curves, illustrating both the high quality of the experimental demonstration of the effect and the detailed theoretical fit. The velocity plotted along the abscissa is that of the Mössbauer source and is proportional to the change in γ-ray energy.

6, stimulated a theoretical calculation of their intensities. This has not previously been reported.

CALCULATION OF THE MODULATED SPECTRUM

This perturbation of the frequency spectrum by the modulator can be understood by considering the effect of the chopper on the amplitude of the radiation emitted by a single excited nucleus. At first this wave propagates out spherically with velocity c and radial frequency $\omega_0 = E/\hbar$, where E is the energy of the excited nuclear state; as the wave passes, its amplitude at any fixed point decays exponentially with decay constant $\Gamma = 1/2\tau$, but the part of the wave that passes through the chopper is quite changed, as can be seen from a calculation of the time variation of the wave amplitude at a point immediately above the chopper.

In this calculation, the segment of the spherical wave that is incident on the chopper is represented by a plane wave normal to the chopper. It will be realized that this condition is not restrictive; in fact, our results can be generalized to include finite distances from source to chopper. If the wheel were solid plastic L cm thick, we might represent the wave amplitude at any point just above the wheel by

$$\begin{aligned} A(t) &= 0, & t<0 \\ &= A_0\, e^{i(\omega_0 + i\Gamma)t}\, e^{ik_p L}, & t>0 \end{aligned} \tag{1}$$

where k_p is the propagation vector for this radiation in plastic and $t=0$ at the instant when the leading edge of the plane wave arrives at the point. But with copper wires (propagation vector k_c) substituted for some of the plastic, the part of the wave arriving at such a point has had its amplitude and phase altered by amounts dependent on k_p, k_c, and the instantaneous path length X in the copper. The wave amplitude at this point is now given by

$$\begin{aligned} A(t, T_0) &= A_0\, e^{i(\omega_0 + i\Gamma)t}\, e^{ik_p[L - X(t,T_0)]}\, e^{ik_c X(t,T_0)} \\ &= A(t)\, e^{i(k_c - k_p)X(t,T_0)} \\ &= A(t)\, \alpha(t, T_0), \end{aligned} \tag{2}$$

where $\alpha(t, T_0)$ is a time-dependent complex attenuation factor representing this amplitude and phase modulation. Refraction effects are negligible since the index of refraction is very nearly unity. The instantaneous value of X depends not only on t, but also on T_0, which corresponds to the phase of the chopper relative to the particular point under consideration. The explicit dependence of $X(t, T_0)$ on $t - T_0$ is readily found from the velocity $v = \omega_{\text{wheel}}\, r$ of the wheel and the diameter d and the spacing S of the wires. $X(t, T_0)$ is periodic in time with period $T_m = S/v$, and the basic modulation frequency is $\omega_m = 2\pi/T_m$.

Since our experiment was performed with the frequency analyzer (the ^{57}Fe atoms in the absorber) about 1 cm beyond the chopper, it is necessary to consider whether our calculation for a "point just above the wheel" is appropriate. Recalling physical optics, we apply the concept of a geometric ray: When a plane wave propagates beyond a scatterer, most of the amplitude at a point at a distance l beyond the scatterer is contributed by a region of diameter $D=(\lambda l)^{1/2}$ on the incident wavefront; if the thickness X of the scatterer is constant over a region with a diameter greater than D, then an expression such as Eq. (2) is sensible. If X changes significantly in distances less than D, however, a more detailed and complicated equation would be needed. With $\lambda = 10^{-8}$ cm and $l \approx 1$ cm, the diameter of the contributing region is only $D \approx 10^{-4}$ cm. Since the diameter d of the chopper wires is 20 times this D, the thickness X is nearly constant over any region of diameter 10^{-4} cm and hence Eq. (2), an equation from ray optics, is a fair approximation in the present experiment.

On the other hand, if the detector distance were increased to $l = 10^3$ cm, then the part of the wave front contributing to the amplitude would have a diameter D wider than each wire so that X, as used in Eq. (2), would be very poorly defined. This raises the question: Does the distance from the modulator to the detector affect the frequency spectrum? We shall return to this question again, but the (mainly) negative answer helps to reduce our anxiety about the actual placement of the absorber.

The time-varying attenuation factor $\alpha(t, T_0)$ can be expanded in the Fourier series

$$\alpha(t, T_0) = \sum_{n=-\infty}^{\infty} C_n e^{in\omega_m(t-T_0)}, \qquad (3)$$

where the C_n are completely determined by the physical properties of the modulator. Thus the amplitude at the point just above the wheel may be rewritten

$$A(t, T_0) = A(t) \sum_{n=-\infty}^{\infty} C_n e^{in\omega_m(t-T_0)}. \qquad (4)$$

The Fourier transform of $A(t, T_0)$ is

$$\hat{A}(\omega, T_0) \propto \sum_{n=-\infty}^{\infty} [C_n e^{-in\omega_m T_0}]/i(\omega - \omega_0 - n\omega_m - i\Gamma). \qquad (5)$$

For all the points in the analyzer, T_0 takes on values uniformly distributed between 0 and T_m. Therefore, by forming $\hat{A}^* \cdot \hat{A}$ and averaging over T_0 (still considering the decay of only a single excited nucleus), the energy spectrum present in all the radiation emerging from the entire chopper is found to be

$$I(\omega) \propto \sum_{n=-\infty}^{\infty} C_n{}^* C_n/[(\omega - \omega_0 - n\omega_m)^2 + \Gamma^2]. \qquad (6)$$

Equation (6) shows that the frequency spectrum $I(\omega)$ is an infinite set of sidebands, that each sideband is of natural width Γ, and that the spacing is uniform and equal to the modulation frequency ω_m.

The C_n are found from Eqs. (2) and (3) to be

$$C_n = (1/S) \int_{-S/2}^{S/2} e^{i(k_c - k_p)X(s)} e^{-in2\pi s/S} ds, \qquad (7)$$

where s is the distance in the direction of the modulator velocity $\vec{v}$ and the path length $X(s)$ in the copper is

$$X(s) = (d^2 - 4s^2)^{1/2}, \quad s^2 \leq (\tfrac{1}{2}d)^2$$
$$= 0, \qquad (\tfrac{1}{2}d)^2 \leq s^2 < (\tfrac{1}{2}S)^2.$$

It should be noticed that the C_n do not depend on the motion of the modulator, but only on its static properties. The coefficients $C_n{}^2$ in Table I were calculated by substituting appropriate values of k_p, k_c, d, and S in Eq. (7). Though the spectrum clearly is dominated by the five center lines ($n=0$, ± 1, ± 2), the absorption due to the higher-order lines is not negligible; some 20% of the total absorption is contributed by $n > 8$.

DISCUSSION OF A "MOVING-GRATING" CALCULATION

A different approach is to consider the process as diffraction from a moving grating. Here one finds the position of the sidebands by appropriately Doppler shifting the beams diffracted into different directions.[7] Using $n\lambda = S\sin\theta$, where $\lambda/S \ll 1$, one finds that the frequency shift of the nth diffraction maximum is

$$\Delta\omega_n = \omega\frac{v}{c}\sin\theta_n = \frac{2\pi c}{\lambda}\left(\frac{v}{c}\right) n \frac{\lambda}{S} = n\omega_m, \qquad (8)$$

in agreement with Eq. (6). As for the intensities in the different orders from a grating made of varying thickness of material with a complex index of refraction, we have not found in a brief search

TABLE I. Sideband intensities $C_n{}^2$ for the indicated sideband numbers n. These values were computed with $k_c - k_p = [2\pi(804) + i\,390]$ cm^{-1}. The value of $\sum_{-\infty}^{\infty} C_n{}^2$ was computed to be 0.43, in agreement with the measured transmission of the chopper while stationary.

n	0	±1	±2	±3	±4	±5	±6	±7	±8
$C_n{}^2$	0.0305	0.0617	0.0562	0.0005	0.0106	0.0092	0.0048	0.0088	0.0101

an explicit formula to refer to, but it will be essentially the same as Eq. (7). Thus the "moving-grating" calculation suggests that the incident beam will be diffracted into different directions and that each beam has a different energy. Thus, if one could work with small, highly directional detectors at great enough distances to pick out only a single order, then the spectrum would not be that corresponding to Eq. (6) but would consist of only one sideband. However, if the spectra obtained at all possible detection angles were added together, the result would again agree with Eq. (6). In our experiment the collimation is such that all radiation leaving the modulator is analyzed, regardless of direction. Thus Eq. (6) is applicable.

It is well known in optical spectroscopy that the diffraction grating constant (S in the present paper) determines the separation into various orders, and the structure of each slit [$X(s)$ in the present paper] determines the envelope and therefore the intensity of the various lines. Recalling from the paragraph after Eq. (2), we see that $\omega_m = 2\pi/T_m = 2\pi v/S$ agrees with the first statement above. The second statement is verified by Eqs. (6) and (7) if "envelope" is appropriately defined.

Instead of the two limiting cases, chopped radiation and diffraction from a moving grating, each of which is fairly simple, it is possible to do a more satisfactory calculation nearer to basic principles. In outline, Eq. (4) can be easily generalized to give not only the wave amplitude at one point just above the chopper, but at all points on the surface just above the chopper which the collimator allows to be illuminated. Since the amplitudes can be taken as zero outside the collimator aperture, Kirchoff's integral theorem[8] allows us to find the amplitude at the detector by summing the waves coming to it from this surface. Fourier-transforming this amplitude gives the amplitudes of the frequency components, and squaring these amplitudes gives the intensities of the sidebands.

The simplifying assumptions by which the two approximate calculations depart from the above "correct" procedure correspond to quite different limiting cases. In the moving-grating calculation, distances are taken large enough that all waves are nearly plane and the scatterer is made large enough that the diffraction pattern collapses into δ functions of the direction. In the chopping calculation, the distance from chopper to detector is taken small enough so that tiny parts of the modulator can be considered quite separately. Properly interpreted, both give the same result so the "correct" calculation is not needed.

A final point is that if one is interested in diffraction from a solid (perhaps to determine its structure), then one usually measures angular distributions. The above discussion clarifies the idea, already utilized by Champeney,[4] that a translational motion of the solid imparts different Doppler shifts to rays emerging at different angles and thereby transforms the experiment into a measurement of an energy distribution.

COMPARISON BETWEEN CALCULATION AND EXPERIMENT

For a particular modulator, the only experimental variable is the rotational frequency. The main unknown quantity in the calculation is the propagation constant $k_c - k_p$ with real part $2\pi\mathfrak{N}/\lambda = 2\pi(\mathfrak{N}_p - \mathfrak{N}_c)/\lambda$ and imaginary part $\mu/2\rho = \frac{1}{2}(\mu_c/\rho_c - \mu_p/\rho_p)$, where $\mathfrak{N}_p$ and $\mathfrak{N}_c$ are the indices of refraction, μ_p and μ_c are the mass absorption coefficients, and ρ_p and ρ_c are the densities of plastic and copper, respectively.

The calculated spectrum—Eq. (6)—is a sum of Lorentzian lines, all of the same width. In the experimental spectrum, however, the lines are broadened by an amount that increases with the order n of the sidebands. This part of the instrumental broadening arises because the collimator selects a quite appreciable range of r on the chopper wheel, so there is a corresponding spread in the value of $v = \omega_{\text{wheel}}\, r$ and hence of $\omega_m = 2\pi v/S$. The calculation of this blurring involves no free variables nor adjustable parameters, and our calculation of the spectrum included it from the beginning.

First the experimental spectrum at each rotation frequency was fitted with the theoretical spectrum, with $\mathfrak{N}$ and μ/ρ as variables. Then when the best-fit values of $\mathfrak{N}$ and μ/ρ were found to be nearly constant from one spectrum to another, a single pair was chosen and used in calculating the curves for all the measured spectra. We emphasize that once these were chosen, the solid lines of Fig. 2 were calculated with *no significant free variable.* (Three unimportant variables were allowed in the fittings. The first measured the baseline, which is merely the duration of the particular measurement. The second was for the experimental linewidth, which increased from 0.26 to 0.31 mm/sec as a result of mechanical vibrations associated with increasing angular velocity of the modulator. The third variable allowed extra intensity in the central line in recognition of a collimator misalignment which allowed some γ rays to skirt the wheel. This extra intensity was never more than 6% of the counting rate because γ rays traversed the chopper, and was completely eliminated in check runs in which additional collimation was used.)

That the calculated spectra fit the data is quite evident from Fig. 2. One must keep in mind that the lower five spectra have in effect no free variables since the linewidth and vertical scale were determined by the zero-velocity case. The values of χ^2 fall between 260 and 390 for 200 points, with some 3×10^6 counts/channel. MISFIT, a normalized χ^2 which takes proper account of the amount of the signal to be fitted,[9] ranges over $0.14\% < M < 0.25\%$ with an error of about 0.05%. First, $M\approx 0.1\%$ represents a good fit in the sense that a perfect one ($M=0$) will not appear better to the eye. Second, the remaining discrepancy in the fit can be traced to the use of Lorentzian line shapes in the calculated spectra. This is not quite true after the lines have been broadened by (i) the finite range in r and (ii) vibrations from the the siren-like sounds of the rotating chopper. Our experience with fitting procedures says that Eq. (6) is sufficient to explain the measured data.

More importantly, the values of the parameters chosen by the fitting routine are quite reasonable. The mass absorption coefficient found from our fitting procedure was $\mu = 87\pm 8$ cm^2/g, in agreement with standard values. The large uncertainty in μ can be traced back to the ratios of the $C_n{}^2$. For example, a 5% increase in μ_c increased $C_0{}^2/C_1{}^2$ by only 1.5%. In contrast, a 5% increase in $\mathfrak{N}_p - \mathfrak{N}_c$ increased the calculated value of $C_0{}^2/C_1{}^2$ by 313%! Without correcting for systematic errors, we found $\mathfrak{N}_p - \mathfrak{N}_c = (6.92\pm 0.02)\times 10^{-6}$, which is to be compared with $\mathfrak{N}_p - \mathfrak{N}_c = 6.85\times 10^{-6}$ calculated on the assumption that the binding energies of the electrons in both copper and plastic are negligible. Correcting the calculation to include the effect of the 8-KeV binding energy of K electrons in copper would increase this estimate of $\mathfrak{N}_p - \mathfrak{N}_c$ by another percent or so. Thus our experimental method allows accurate measurement of $\mathfrak{N}_p - \mathfrak{N}_c$ for any material.

CONCLUSION

Previous experiments involving γ-ray spectra with sidebands have involved either acoustic phase-modulation effects[2,10] or magnetostrictive effects[11] leading indirectly to phase modulation. In all the acoustic cases so far reported, it has not been experimentally possible to get the atoms vibrating with a *known* distribution of amplitudes. Thus, while the frequency shifts are understood, a detailed testing of the relative intensities of the sidebands has not been possible. The same is true *a fortiori* for the magnetostrictive work. In the present case, however, the experimental modulation technique producing the sidebands is fully controlled, and Fig. 2 shows the first quantitative treatment of the spectrum resulting from amplitude and phase modulation of a γ ray at megahertz frequencies.

There has been, among ourselves as well, considerable confusion about how to relate the "chopping calculation" to "diffraction from a moving grating." In particular, the chopping calculation is formally the same as a time-varying index of refraction k, but with no velocity at all. We hope that this discussion has clarified the matter of deciding when it is adequate to use ray optics and chopping, or when to use wave optics and motion of the scatterer.

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