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## APPARENT WEIGHT OF PHOTONS\*

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As we proposed a few months ago,<sup>1</sup> we have now measured the effect, originally hypothesized by Einstein,<sup>2</sup> of gravitational potential on the apparent frequency of electromagnetic radiation by using the sharply defined energy of recoil-free  $\gamma$  rays emitted and absorbed in solids, as discovered by Mössbauer.<sup>3</sup> We have already reported<sup>4</sup> a detailed study of the shape and width of the line obtained at room temperature for the 14.4-keV, 0.1-microsecond level in Fe<sup>57</sup>. Particular attention was paid to finding the conditions required to obtain a narrow line. We found that the line had a Lorentzian shape with a fractional full-width at half-height of  $1.13 \times 10^{-12}$  when the source was carefully prepared according to a prescription developed from experience. We have also investigated the 93-keV, 9.4-microsecond level of Zn<sup>67</sup> at liquid helium and liquid nitrogen temperatures using several combinations of source and absorber environment, but have not observed a usable resonant absorption. That work will be reported later. The fractional width and intensity of the absorption in Fe<sup>57</sup> seemed sufficient to measure the gravitational effect in the laboratory.

As a preliminary, we sought possible sources of systematic error that would interfere with measurements of small changes in frequency using this medium. Early in our development of the instrumentation necessary for this experiment, we concluded that there were asymmetries in, or frequency differences between, the lines of given combinations of source and absorber which vary from one combination to another. Thus it is ab-

solutely necessary to measure a change in the relative frequency that is produced by the perturbation being studied. Observation of a frequency difference between a given source and absorber cannot be uniquely attributed to this perturbation. More recently, we have discovered and explained a variation of frequency with temperature of either the source or absorber.<sup>5</sup> We conclude that the temperature difference between the source and absorber must be accurately known and its effect considered before any meaning can be extracted from even a change observed when the perturbation is altered.

The basic elements of the apparatus finally developed to measure the gravitational shift in frequency were a carefully prepared source containing 0.4 curie of 270-day Co<sup>57</sup>, and a carefully prepared, rigidly supported, iron film absorber. Using the results of our initial experiment, we requested the Nuclear Science and Engineering Corporation to repurify their nickel cyclotron target by ion exchange to reduce cobalt carrier. Following the bombardment, in a special run in the high-energy proton beam of the high-current cyclotron at the Oak Ridge National Laboratory, they electroplated the separated Co<sup>57</sup> onto one side of a 2-in. diameter, 0.005-in. thick disk of Armco iron according to our prescription. After this disk was received, it was heated to 900°-1000°C for one hour in a hydrogen atmosphere<sup>6</sup> to diffuse the cobalt into the iron foil about  $3 \times 10^{-5}$  cm.

The absorber made by Nuclear Metals Inc., was composed of seven separate units. Each

unit consisted of about 80 mg of iron, enriched in  $\text{Fe}^{57}$  to 31.9%, electroplated onto a polished side of a 3-in. diameter, 0.040-in. thick disk of beryllium. The electroplating technique required considerable development to produce films with absorption lines of width and strength that satisfied our tests. The films finally accepted, resonantly absorbed about 1/3 the recoil-free  $\gamma$  rays from our source. Each unit of the absorber was mounted over the 0.001-in. Al window of a 3 in.  $\times$  1/4 in. NaI(Tl) scintillation crystal integrally mounted on a Dumont 6363 multiplier phototube. The multiplier supply voltages were separately adjusted to equalize their conversion gains, and their outputs were mixed.

The required stable vertical baseline was conveniently obtained in the enclosed, isolated tower of the Jefferson Physical Laboratory.<sup>7</sup> A statistical argument suggests that the precision of a measurement of the gravitational frequency shift should be independent of the height. Instrumental instability but more significantly the sources of systematic error mentioned above are less critical compared to the larger fractional shifts obtained with an increased height. Our net operating baseline of 74 feet required only conveniently realizable control over these sources of error.

The absorption of the 14.4-keV  $\gamma$  ray by air in the path was reduced by running a 16-in. diameter, cylindrical, Mylar bag with thin end windows and filled with helium through most of the distance between source and absorber. To sweep out small amounts of air diffusing into the bag, the helium was kept flowing through it at a rate of about 30 liters/hr.

The over-all experiment is described by the block diagram of Fig. 1. The source was moved sinusoidally by either a ferroelectric or a moving-coil magnetic transducer. During the quarter of the modulation cycle centered about the time of maximum velocity the pulses from the scintillation spectrometer, adjusted to select the 14.4-keV  $\gamma$ -ray line, were fed into one scaler while, during the opposite quarter cycle, they were fed into another. The difference in counts recorded was a measure of the asymmetry in, or frequency-shift between, the emission and absorption lines. As a precaution the relative phase of the gating pulses and the sinusoidal modulation were displayed continuously. The data were found to be insensitive to phase changes much larger than the drifts of phase observed.

A completely duplicate system of electronics, controlled by the same gating pulses, recorded

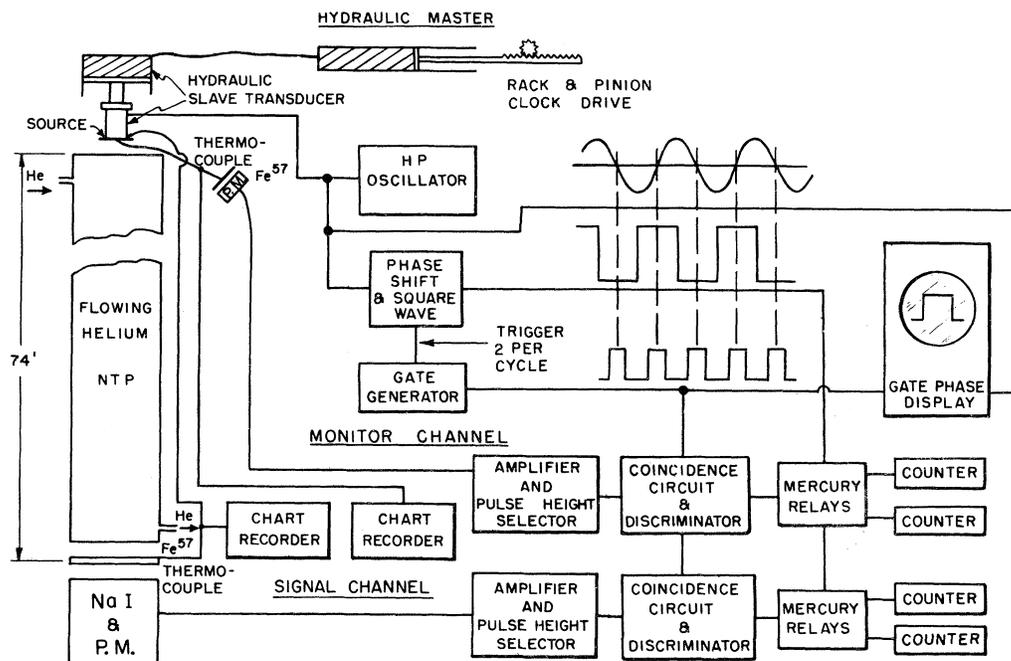


FIG. 1. A block diagram of the over-all experimental arrangement. The source and absorber-detector units were frequently interchanged. Sometimes a ferroelectric and sometimes a moving-coil magnetic transducer was used with frequencies ranging from 10 to 50 cps.

data from a counter having a 1-in. diameter, 0.015-in. thick NaI(Tl) scintillation crystal covered by an absorber similar to the main absorber. This absorber and crystal unit was mounted to see the source from only three feet away. This monitor channel measured the stability of the over-all modulation system, and, because of its higher counting rate, had a smaller statistical uncertainty.

The relation between the counting rate difference and relative frequency shifts between the emission and absorption lines was measured directly by adding a Doppler shift several times the size of the gravitational shift to the emission line. The necessary constant velocity was introduced by coupling a hydraulic cylinder of large bore carrying the transducer and source to a master cylinder of small bore connected to a rack-and-pinion driven by a clock.

Combining data from two periods having Doppler shifts of equal magnitude, but opposite sign, allowed measurement of both sensitivity and relative frequency shift. Because no sacrifice of valuable data resulted, the sensitivity was calibrated about 1/3 of the operating time which was as often as convenient without recording the data automatically. In this way we were able to eliminate errors due to drifts in sensitivity such as would be anticipated from gain or discriminator drift, changes in background, or changes in modulation swing.

The second order Doppler shift resulting from lattice vibrations required that the temperature difference between the source and absorber be controlled or monitored. A difference of 1°C would produce a shift as large as that sought, so the potential difference of a thermocouple with one junction at the source and the other at the main absorber was recorded. An identical system was provided for the monitor channel. The recorded temperature data were integrated over a counting period, and the average determined to 0.03°C. The temperature coefficient of frequency which we have used to correct the data, was calculated from the specific heat of a lattice having a Debye temperature of 420°K. Although at room temperature this coefficient is but weakly dependent on the Debye temperature, residual error in the correction for, or control of, the temperature difference limits the ability to measure frequency shifts and favors the use of a large height difference for the gravitational experiment.

Data typical of those collected are shown in Table I. The right-hand column is the data after

correction for temperature difference. All data are expressed as fractional frequency shift  $\times 10^{15}$ . The difference of the shift seen with  $\gamma$  rays rising and that with  $\gamma$  rays falling should be the result of gravity. The average for the two directions of travel should measure an effective shift of other origin, and this is about four times the difference between the shifts. We confirmed that this shift was an inherent property of the particular combination of source and absorber by measuring the shift for each absorber unit in turn, with temperature correction, when it was six inches from the source. Although this test was not exact because only about half the area of each absorber was involved, the weighted mean shift from this test for the combination of all absorber units agreed well with that observed in the main experiment. The individual fractional frequency shifts found for these, for the monitor absorber, as well as for a 11.7-mg/cm<sup>2</sup> Armco iron foil, are displayed in Table II. The considerable variation among them is as striking as the size of the weighted mean shift. Such shifts could result from differences in a range of about 11% in effective Debye temperature through producing differences in net second order Doppler effect. Other explanations based on hyperfine structure including electric quadrupole interactions are also plausible. Although heat treatment might be expected to change these shifts for the iron-plated beryllium absorbers, experience showed that the line width was materially increased by such treatment, probably owing to interdiffusion. The presence of a significant shift for even the Armco foil relative to the source, both of which had received heat treatments, suggests that it is unlikely one would have, without test, a shift of this sort smaller than the gravitational effect expected in even our "two-way" baseline of 148 feet. The apparently fortuitous smallness of the shift of the monitor absorber relative to our source corresponds to the shift expected for about 30 feet of height difference.

Recently Cranshaw, Schiffer, and Whitehead<sup>8</sup> claimed to have measured the gravitational shift using the  $\gamma$  ray of Fe<sup>57</sup>. They state that they believe their 43% statistical uncertainty represents the major error. Two much larger sources of error apparently have not been considered: (1) the temperature difference between the source and absorber, and (2) the frequency difference inherent in a given combination of source and absorber. From the above discussion, only 0.6°C of temperature difference would produce a shift

Table I. Data from the first four days of counting. The data are expressed as fractional frequency differences between source and absorber multiplied by  $10^{15}$ , as derived from the appropriate sensitivity calibration. The negative signs mean that the  $\gamma$  ray has a frequency lower than the frequency of maximum absorption at the absorber.

Period	Shift observed	Temperature correction	Net shift
Source at bottom			
Feb. 22, 5 p. m.	$-11.5 \pm 3.0$	-9.2	$-20.7 \pm 3.0$
	$-16.4 \pm 2.2^a$	-5.9	$-22.3 \pm 2.2$
	$-13.8 \pm 1.3$	-5.3	$-19.1 \pm 1.3$
	$-11.9 \pm 2.1^a$	-8.0	$-19.9 \pm 2.1$
	$-8.7 \pm 2.0^a$	-10.5	$-19.2 \pm 2.0$
Feb. 23, 10 p. m.	$-10.5 \pm 2.0$	-10.6	$-21.0 \pm 2.0$
	Weighted average = $-19.7 \pm 0.8$		
Source at top			
Feb. 24, 0 a. m.	$-12.0 \pm 4.1$	-8.6	$-20.6 \pm 4.1$
	$-5.7 \pm 1.4$	-9.6	$-15.3 \pm 1.4$
	$-7.4 \pm 2.1^a$	-7.4	$-14.8 \pm 2.1$
	$-6.5 \pm 2.1^a$	-5.8	$-12.3 \pm 2.1$
	$-13.9 \pm 3.1^a$	-7.5	$-21.4 \pm 3.1$
Feb. 25, 6 p. m.	$-6.6 \pm 3.0$	-5.7	$-12.3 \pm 3.0$
	$-6.5 \pm 2.0^a$	-8.9	$-15.4 \pm 2.0$
	$-10.0 \pm 2.6$	-7.9	$-17.9 \pm 2.6$
	Weighted average = $-15.5 \pm 0.8$ Mean shift = $-17.6 \pm 0.6$ Difference of averages = $-4.2 \pm 1.1$		

<sup>a</sup>These data were taken simultaneously with a sensitivity calibration.

Table II. Data on asymmetries of various absorbers in apparent fractional frequency shift multiplied by  $10^{15}$ . In the third column we tabulate the Debye temperature increase of the absorber above that of the source which could account for the shift.

Absorber	$(\Delta\nu/\nu) \times 10^{15}$	$\Delta\theta_D$ in $^\circ\text{K}$
No. 1	$-8.4 \pm 2.5$	$+15 \pm 4$
No. 2	$-24 \pm 3.5$	$+41 \pm 6$
No. 3	$-28 \pm 3.5$	$+48 \pm 6$
No. 4	$-19 \pm 3.5$	$+33 \pm 6$
No. 5	$-24 \pm 3.5$	$+41 \pm 6$
No. 6	$-17 \pm 2.5$	$+29 \pm 4$
No. 7	$-19 \pm 3.5$	$+33 \pm 6$
Weighted mean of No. 1-No. 7	$-19 \pm 3.0$	$+33 \pm 5$
Monitor absorber	$+0.55 \pm 0.15$	$-0.95 \pm 0.26$
Armco foil	$+10 \pm 3.5$	$-17 \pm 6$

as large as the whole effect observed. Their additional experiment at the shortened height difference of three meters does not, without concomitant temperature data, resolve the question

of inherent frequency difference. Their stated disappointment with the over-all line width observed would seem to add to the probability of existence of such a shift. They mention this broadening in connection with its possible influence on the sensitivity, derived rather than measured, owing to a departure from Lorentzian shape. Clearly such a departure is even more important in allowing asymmetry.

Our experience shows that no conclusion can be drawn from the experiment of Cranshaw et al.

If the frequency-shift inherent in our source-absorber combination is not affected by inversion of the relative positions, the difference between shifts observed with rising and falling  $\gamma$  rays measures the effect of gravity. All data collected since recognizing the need for temperature correction, yield a net fractional shift,  $-(5.13 \pm 0.51) \times 10^{-15}$ . The error assigned is the rms statistical deviation including that of independent sensitivity calibrations taken as representative of their respective periods of operation. The shift observed agrees with  $-4.92 \times 10^{-15}$ , the predicted gravitational shift for this "two-way" height difference.

Expressed in this unit, the result is

$$(\Delta\nu)_{\text{exp}}/(\Delta\nu)_{\text{theor}} = +1.05 \pm 0.10,$$

where the plus sign indicates that the frequency increases in falling, as expected.

These data were collected in about 10 days of operation. We expect to continue counting with some improvements in sensitivity, and to reduce the statistical uncertainty about fourfold. With our present experimental arrangement this should result in a comparable reduction in error in the measurement since we believe we can take adequate steps to avoid systematic errors on the resulting scale. A higher baseline or possibly a narrower  $\gamma$  ray would seem to be required to extend the precision by a factor much larger than this.

We wish to express deep appreciation for the generosity, encouragement, and assistance with details of the experiment accorded us by our colleagues and the entire technical staff of these laboratories during the three months we have

been preoccupied with it.

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<sup>1</sup>R. V. Pound and G. A. Rebka, Jr., Phys. Rev. Letters **3**, 439 (1959).

<sup>2</sup>A. Einstein, Ann. Physik **35**, 898 (1911).

<sup>3</sup>R. L. Mössbauer, Z. Physik **151**, 124 (1958); Naturwissenschaften **45**, 538 (1958); Z. Naturforsch. **14a**, 211 (1959).

<sup>4</sup>R. V. Pound and G. A. Rebka, Jr., Phys. Rev. Letters **3**, 554 (1959).

<sup>5</sup>R. V. Pound and G. A. Rebka, Jr., Phys. Rev. Letters **4**, 274 (1960).

<sup>6</sup>We wish to thank Mr. F. Rosebury of the Research Laboratory of Electronics, Massachusetts Institute of Technology, for providing his facilities for this treatment.

<sup>7</sup>See E. H. Hall, Phys. Rev. **17**, 245 (1903), first paragraph.

<sup>8</sup>T. E. Cranshaw, J. P. Schiffer, and A. B. Whitehead, Phys. Rev. Letters **4**, 163 (1960).

## TEMPERATURE-DEPENDENT SHIFT OF $\gamma$ RAYS EMITTED BY A SOLID

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Recent experiments by Mössbauer<sup>1</sup> have shown that when low-energy  $\gamma$  rays are emitted from nuclei in a solid a certain proportion of them are unaffected by the Doppler effect. It is the purpose of this Letter to show that they are nevertheless subject to a temperature-dependent shift to lower energy which can be attributed to the relativistic time dilatation caused by the motion of the nuclei.

Let us regard the solid as a system of interacting atoms with the Hamiltonian

$$H = \sum p_i^2/2m_i + V(r_1, r_2, \dots).$$

The Mössbauer effect is due to those processes in which the phonon occupation numbers do not change. It might appear that in such cases the energy of the solid is unaltered, but this is not so, as the nucleus which emits the  $\gamma$  ray changes its mass, and this affects the lattice vibrations. Suppose the nucleus of the  $i$ th atom emits a  $\gamma$  ray of energy  $E$ , its mass changing by  $\delta m_i = -E/c^2$ .

The change in energy,  $\delta E$ , of the solid is given by

$$\begin{aligned} \delta E = \langle \Delta H \rangle &= \delta \langle p_i^2/2m_i \rangle = -\delta m_i \langle p_i^2/2m_i^2 \rangle \\ &= (\delta m_i/m_i) T_i = (E/m_i c^2) T_i, \end{aligned}$$

where  $T_i$  is the expectation value of the kinetic energy of the  $i$ th atom. The energy of the  $\gamma$  ray must accordingly be reduced by  $\delta E$  so there is a shift of relative magnitude  $\delta E/E = T_i/m_i c^2$ . The same formula can be deduced by regarding the shift as due to a relativistic time dilatation.

To estimate  $T_i$  we make the following assumptions: (i) The atoms all have the same mass, and the kinetic energy is equally distributed among them. (ii) The kinetic energy is half the total lattice energy, i.e., we assume that the forces coupling the atoms are harmonic. Under these assumptions  $T_i/m_i = \frac{1}{2}U$ , where  $U$  is the lattice energy per unit mass. The relative shift is thus given by  $\delta E/E = U/2c^2$ . For Fe at 300°K