TOTAL EXTERNAL REFLECTION OF GAMMA-RAYS AND MÖSSBAUER SPECTROSCOPY

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Résumé.- Il est possible de construire des guides pour les rayons-γ en employant la réflexion externe totale. La radiation incidente à un angle rasant plus petit que l'angle critique $\theta_c \approx \lambda(N_e e^2/\pi m_e c^2)^{1/2}$ peut être presque complètement réfléchie dépendant de la qualité de la surface et de son coefficient d'absorption. Des guides ont été construits à partir de longs tubes de verre de laboratoire ayant un petit diamètre et une rectitude aussi parfaite que possible. Une mesure de la transmission en fonction de la position, alors que la source est déplacée le long d'un diamètre a fourni une technique qui a été d'une aide appréciable pour réaliser l'alignement des sections successives d'un guide. L'observation, dans le spectre de rayons-γ du ^{57}Fe, d'une raie de résonance non-élargie entre une source et un absorbant a permis de conclure que la réflexion est essentiellement élastique. Des calculs sur ordinateur ont analysé les propriétés des guides de section carrée qui pourraient être assemblés à partir de plaques de verre planes montées dans un tuyau où l'on aurait fait le vide. Un long guide construit de cette façon pourrait permettre une mesure améliorée du décalage gravitationnel vers le rouge. L'ouverture effective de capture par un guide étant proportionnel à λ^2, celui-ci a donc les propriétés d'un filtre passe-bas. Plusieurs petits tubes capillaires placés entre une source et un détecteur ont permis d'observer une réduction marquée de l'intensité de la radiation de haute énergie atteignant le détecteur en comparaison avec celle de basse énergie. L'emploi de verres contenant du plomb a réduit la pénétration directe des rayons-γ de hautes énergies dans le verre et a aussi augmenté l'ouverture pour les basses énergies en augmentant les angles critiques.

Abstract.- Guides for γ-rays can be constructed by using total external reflection. Radiation incident at grazing angles less than a critical angle $\theta_c \approx \lambda(N_e e^2/\pi m_e c^2)^{1/2}$ may be nearly completely reflected depending upon the surface quality and the absorption coefficient. Sample guides have been constructed from long small-diameter glass laboratory tubing of good straightness. Determination of the transmission vs. position, as a source was moved across a diameter, provided a technique that greatly helped adjustment of successive sections to colinearity. Observation of an unbroadened ^{57}Fe γ-ray resonance between a source and absorber separated by such a γ-ray guide demonstrated that the reflection process is essentially elastic. Computer calculations have analyzed the properties of guides of square cross-section which would be assembled from plates of float glass supported in a vacuum pipe. A large guide constructed in this way could facilitate an improved measurement of the gravitational red-shift. The proportionality to λ^2 of the effective aperture of capture by a guide gives to it the properties of a low-pass filter. A bundle of small capillary tubing placed between a source and a detector resulted in a striking reduction in the intensity of high energy radiation reaching the detector relative to that of low energy. Use of lead glass reduced the direct penetration of the high energy γ-rays through the glass and also increased the aperture for low energy by increasing the critical angles.

Some applications of Mössbauer's recoil-free resonant absorption suffer from the fact that the γ-rays involved are difficult to guide, to deflect or to focus. A prime example where a guide would yield important benefits is the measurement of the energy shift caused by a gravitational potential difference /1-6/. For a vertical distance h between a source and absorber small compared to the earth's radius at its surface, the contribution to any frequency shift caused by gravity should be proportional to h. So long as the source strength and the detector aperture are limited by other factors, perhaps cost, there appears at first glance to be little advantage in designing an experiment that operates with h greater than sufficient to allow detection of the γ-rays without overload. This is because, so long as the intensity of the radiation reaching the detector varies as h^{-2}, the statistical uncertainty in a measured frequency shift grows as the inverse square root of the intensity and, thus, also linearly with h. It is assumed that absorption over the path can be made unimportant by the use of either a vacuum chamber /3,6/ or a container for a gas like helium /4,5/ having low absorption at STP.

The initial proposal /1/ to employ Mössbauer resonance for the detection of the gravitational red-shift called attention to the possibility of employing long vertical paths through the use of γ-ray guides, or light pipes, employing total external reflection. The practical realization of the resonance in ^{57}Fe /7,8/ made available a medium of such resolution that path lengths depending upon light pipes were not required for the first generation of experiments. More recently the resonance in ^{181}Ta has been developed /9,10/ to realize some of its potentially even higher resolution. Because of the

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low 6.25 keV energy of the ^{181}Ta γ-ray, it is an especially favorable example for the employment of light pipes in applications, as will become apparent below. Bernstein and Campbell reported /11/ extensive studies of the contribution to the total reflection at several angles from an iron mirror enriched in ^{57}Fe, of ^{57}Fe γ-rays scanning the source with a Doppler transducer. We will not discuss such applications of reflection.

We have given some thought to and carried out some experiments on the design of suitable devices. Some of these considerations have been briefly reported previously /12/. Some similar thoughts and observations have been made independently in relation to X-rays of longer wavelength in medical applications /13/.

1. Design considerations.- Most of the electrons of the atoms of materials are bound weakly compared to γ and X-ray energies. As a consequence, as shown by A.H. Compton in 1923 /14/, the index of refraction n is less than one, and rays incident from a vacuum onto a plane surface nearer to grazing incidence than a critical angle θ_c suffer "total external reflection". This reflection, in practice, is less than total because the index is complex, expressing the presence of absorption. Imperfect surfaces also result in losses of intensity in the reflected wave.

The Drude-Lorentz theory predicts the index of refraction to be given by $n = 1 - \delta$, where $\delta \approx Ne^2\lambda^2/2\pi mc^2$, and N is the total number of essentially free electrons per unit volume, e is the charge on an electron, λ is the wavelength, m the electronic mass, and c the velocity of light. Accordingly, the critical angle $\theta_c \approx (2\delta)^{1/2}$ which, for 6 keV photons, corresponds to $\theta_c^2 \approx 1.1 \times 10^{-5}\rho$ radians^2 where ρ is the density in grams cm^{-3}. If μ is the X-ray absorption coefficient, Parratt /15/ showed that $\mu\lambda/4\pi\delta = \beta/\delta$ is a parameter useful for describing the reflection coefficient for a perfect surface. Figure 1 illustrates the dependence of the intensity reflection coefficient $R(\theta)$ on θ/θ_c for several values of β/δ. For purposes of constructing a light pipe one seeks a material of large δ and small β to maximize the capture angle and minimize the loss per reflection, respectively.

In applications that have been made to focussing devices, such as were developed for extracting a beam of synchrotron radiation /16/ and telescopes for X-ray astronomy /17/ only one reflection is utilized. Therefore most of the importance in the choice of material is associated with maximizing δ through the use of a gold film on a glass or plastic backing. For multiple reflections, absorption is compounded and it is therefore less tolerable. Among the possible materials, ordinary borosilicate glass is a convenient initial choice because its surface tends to be smooth and because it has a low value, about 0.025, of β/δ at 6 keV, which, according to figure 1 should result in 95% or more reflection up to within a few percent of the critical angle.

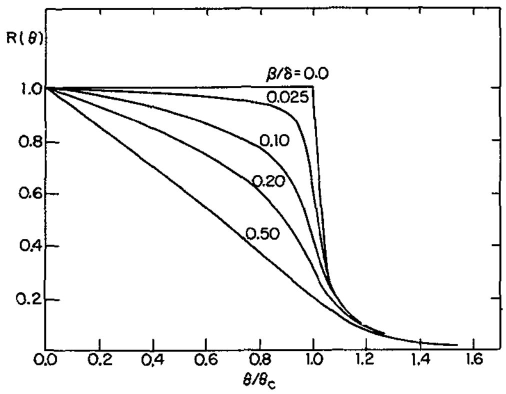

Fig. 1 : The reflection coefficient of intensity $R(\theta/\theta_c)$ for several values of β/δ.

Unfortunately δ itself, and therefore θ_c, are low. For use at 6.25 keV, the energy of the ^{181}Ta γ-ray, the metals Fe, Co, Cu, Ni and Zn have small values of μ. A sufficiently smooth coating of nickel ought nearly to double the critical angle of glass with little degradation of the reflection. Unfortunately it is difficult to obtain adhesion of nickel to a polished surface, particularly inside long, narrow tubes, and we have not yet studied a nickel coated surface.

It is straightforward to calculate the transmission through a long straight tube of circular cross-section from a point source on its axis, given reflection coefficients behaving as in figure 1. Figure 2 describes the intensity transmitted vs. length L, in units of $\theta_c L/d$, where d is the tube diameter, for several values of β/δ. The parameter $\theta_c L/d$ is approximately the number of reflections suffered by a photon travelling at θ_c relative to the axis. The curves are normalized to $I_o\theta_c^2/4\pi$, where I_o is the source intensity in all directions. For $\theta_c L/d < 1$, direct transmission increased steeply with decreased length.

It might be supposed that impossible requirements are placed on the surface quality by the small wavelengths involved. Most mirrors and focussing devices have used surfaces polished at least to optical quality. It would not be economically feasible to construct large scale guides to such specifications. The nearness of the incidence to parallism to the surface reduces the sensitivity to variations of the height of the surface.

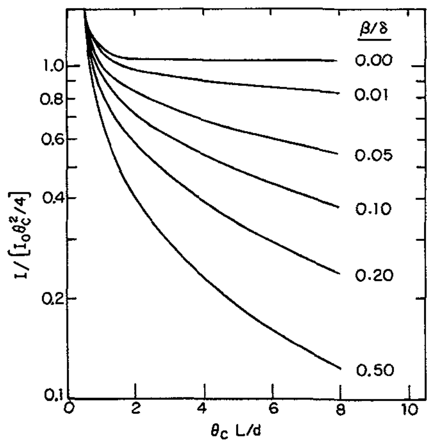

Fig. 2 : Normalized transmission of circular pipes vs. a length parameter $\theta_c L/d$ for several values of β/δ.

Because the absorption coefficient is small, γ-rays pass through obstructions and the effective surface is an average over some distance. In the averaging, the main effect of surface roughness is a fluctuation of the direction of the normal to the net effective surface. The ability to guide radiation through the pipe is degraded by the losses that occur when the angle fluctuates so as to cause some incidence angles to exceed θ_c. Calculations and experimental data indicate /18/ that moderate surface roughness produces a reduction of the reflection coefficient vs. θ very much like that which would result from an increase of β/δ.

2. Experimental Trials.- Our first trials were made with a piece of laboratory Pyrex tubing 120cm long and of 4mm bore. We utilized the 6 keV X-rays from an ^{55}Fe source and soon found that great care was required to avoid bends that destroyed reflections by changing the angles of incidence for successive reflections by more than θ_c, about 0.005 radians for Pyrex glass. We were able to transmit 9 times as intense a beam through the helium filled tubing to a 4mm detector window as arrived there through helium without the tubing. An 11-fold enhancement would have been expected for the critical angle without absorption.

To simplify the problem of straightness a pair of glass tubes was hung vertically, connected in series. Some care was needed to obtain adequate alignment at the joint between the two. Counting rates from the ^{55}Fe source, which covered an area across the entrance end, were recorded for each length as the pipe was broken off, a few centimeters at a time. The resulting data are shown in figure 3.

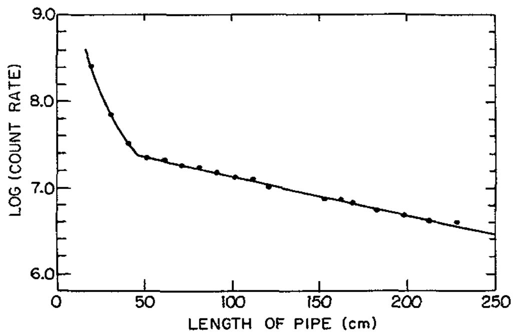

Fig. 3 : Logarithm of the counting rates observed for various lengths of 4 mm-bore Pyrex tubing.

At about 45cm of length, the decrease with length as the inverse square is over ridden by the reflective ducting and the intensity decreases exponentially. Plotted on a logarithmic scale, that region becomes a straight line of slope $d(\ln C)/d\ell = 4.5 \times 10^{-3}\,cm^{-1}$.

Data were collected from measurements of the external dimensions of many examples of Pyrex tubing nominally 11mm outside diameter, 1mm wall and 120cm long, in the interest of identifying those with the most straight inner bores. For this size, external dimensions varied along the lengths in such a way that the angular departures from the axis were on the scale of 0.2 milliradians r.m.s. How this compared for the inner bores is not known. Some pipes of larger diameter were also examined in this way. Those that were 12cm in diameter showed angular deviations typically seven times larger, which would be prejudicial to good performance as a light pipe for 6 keV γ-rays.

A guide was constructed from the six best of the 9mm-bore tubes. They were cleaned successively with detergent, water, hydrogen peroxide, CP acetone, and distilled water and dried with flowing N_2. One centimeter of length was cut from each end to eliminate the distortions in the fire-polished ends. Each length was strapped to a milled recess in an aluminium bar to ensure straightness and these were adjustably supported on a track of Dexion. The adjustment to align the several sections proved difficult and, in the course of trying, it was discovered that the transmission of the pipe for off-centered sources could be utilized as an aid to alignment. Figure 4 illustrates the transmission through the pipe vs. the transverse position of the ^{55}Fe source, of diameter 2mm, before and after successful alignment.

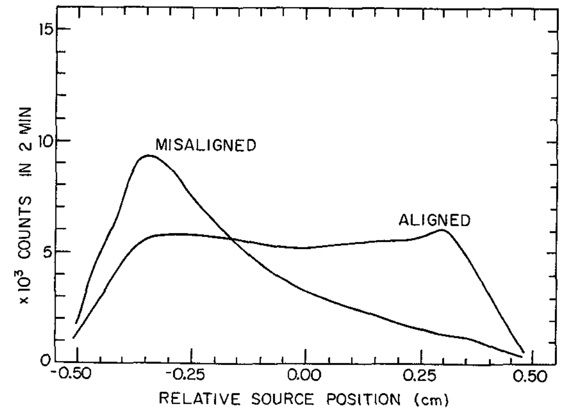

Fig. 4 : Transmission through six sections of 9mm-bore, 115cm long Pyrex vs. transverse position of the source, misaligned and aligned.

After the final alignment of the six sections, the transmission vs. transverse position was observed as shown in figure 5, as it was shortened down to a single section.

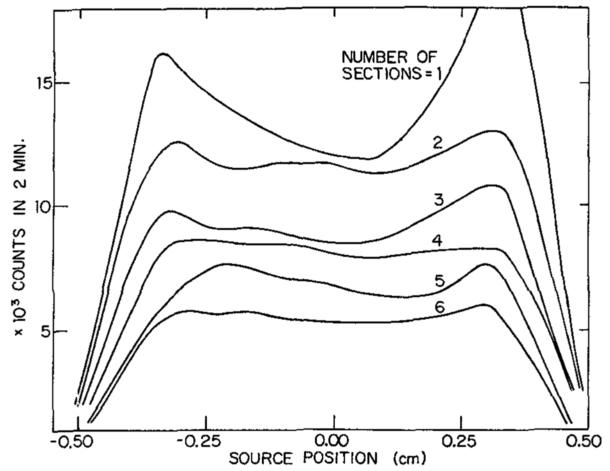

Fig. 5 : Transmission vs. transverse position of the 2mm diam. ^{55}Fe source for 6, 5, 4, 3, 2 and 1 lengths of 9mm bore, 115cm long, Pyrex.

The transmission from the centered source taken from these curves yields the points plotted in figure 6. The solid lines of figure 6 are drawn on the basis of those of figure 2. The experimental data yield a curve of slope close to that for $\beta/\delta = 0.10$, considerably more lossy than the 0.025 based on the calculated value of μ. A very similar number fits the data from the 4mm-bore tubing of figure 3 and it seems probable that surface roughness accounts for the increase in losses.

The reflections by the walls of the pipe must be fully elastic for it to be used in Mössbauer resonance.

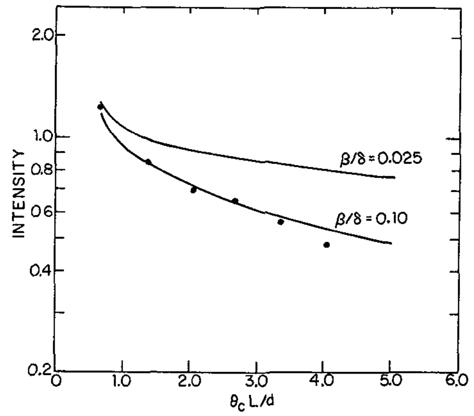

Fig. 6 : Transmission vs. length through the 9mm-bore tubing for a centered source. The solid lines are calculated as in figure 2.

To demonstrate that there was negligible energy loss on reflection and that the quality of a resonance was not degraded, the resonant absorption by an enriched ^{57}Fe absorber, in the form of iron, of ^{57}Fe γ-rays from a ^{57}Co source in iron was observed directly and with the 7 meter long glass tube interposed between source and absorber. Of course the 14.4 keV radiation was used with the correspondingly smaller critical angle and number of reflections. It was deduced that 80% of the 14.4 keV radiation reaching the detector was reflected at least once. The source strength and the solid angle of capture $\pi\theta_c^{\,2}$ were small, so the data collection rate was low. Figure 7 reproduces data accumulated with a velocity spectrometer for a full cycle of sinusoidal motion of the source, showing the resonance for each direction of acceleration of the source through zero velocity. Analysis of the separation of the line centers for 17 runs, similar to that shown, found that any difference, compared to a similar measurement through a three meter path in air, set an upper limit to any energy loss on reflection of 1.4×10^{-15} The resonance lines using the 7-meter pipe were numerically slightly narrower but the difference was within the statistical standard deviations. The fractional depth of the absorption for the piped radiation was 0.36 compared to 0.27 for the air path. This probably results from a reduced background of 122 keV γ-rays using the pipe, which suggests another use for the pipes.

The transmission of the circular pipes for a small source off-center, as shown in figure 5 especially for the short lengths, rises as the source nears the walls.

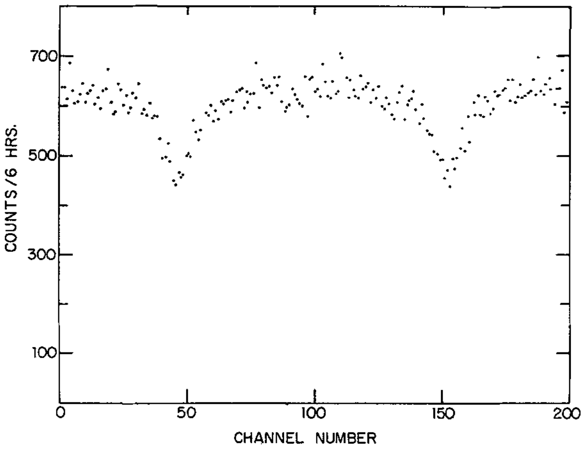

Fig. 7 : Data from a Mössbauer velocity spectrometer for an ^{57}Fe iron source and absorber with a 7-meter light pipe interposed. A full cycle of the source motion is included and the two resonances observed are the central lines with positive and negative acceleration.

Two different things contribute to this effect. First, for a pipe not much longer than giving one reflection for a centered source, all of the rays within the cone making the angle θ_c to the longitudinal axis are transmitted from an off-centered source and some others can directly strike the exit aperture as well. The other contribution applies even in long pipes although it is quickly suppressed by losses because many more reflections are involved than for the ordinary rays. Rays emanating in skewed directions from an off-centered source can make angles greater than θ_c to the longitudinal axis and yet strike the wall nearer to grazing incidence than θ_c. For example, a point source nearly at the wall can illuminate the wall in a direction at a large angle to the longitudinal axis but near grazing incidence on the wall. Such a ray travels in chords in a helical spiral along the tube. As a result, the effective capture solid angle rises steeply as the source nears the wall. The rise is suppressed by losses on reflection and by length. An evaluation by computer of the transmission vs. distance off-center, for several lengths $\theta_c L/d$, assuming a constant reflection efficiency of 0.95 for $\theta \leqslant \theta_c$, is shown in figure 8. It is to be noted that the additional aperture disappears for moderate lengths or for lossy reflections. The source employed in a practical application would likely be of linear dimension not insignificant compared to d and the aperture of acceptance would be an average over the source.

The unavailability of glass tubing of large size and of good surface leads one to investigate the construction of a guide by building it from flat glass plates with a square or rectangular cross-section.

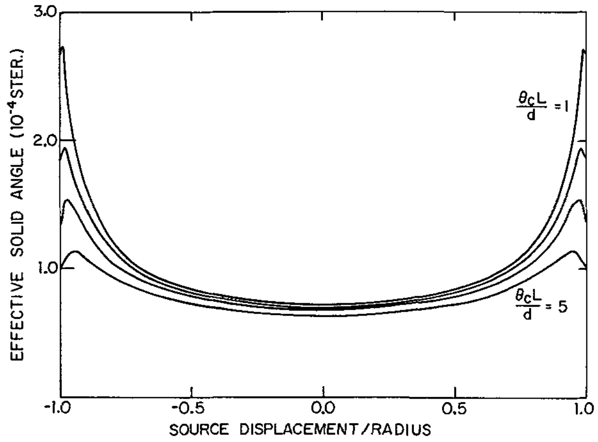

Fig. 8 : The calculated transmission through a circular pipe vs. transverse position of the source for lengths with values of $\theta_c L/d$ of 1, 2, 3 and 5. The reflection is assumed to be 95% for $\theta \leqslant \theta_c$ and to vanish for $\theta > \theta_c$.

Guides for low energy "cold" neutrons, with very similar wavelengths and refractive indices, are built from polished optically flat plates /19/. Float glass has been reported to be of good quality for image formation in X-ray telescopes using glancing incidence /20/ and it should be good for our purposes. Such a structure would be enclosed in a vacuum pipe to avoid bending of the glass plates which would result if they were required to support pressure differences.

A square pipe should capture into the guide mode the radiation entering within a solid angle $4\theta_c^2$ centered on the axial direction. The corresponding aperture is $\pi\theta_c^2$ for the circular pipe, for a small centered source, and the rectangular aperture is greater by $4/\pi$. The increase results because most planes of incidence, even for a centered source, are slanted to the longitudinal axis. The same aperture holds for all off-centered points and all points in the end plane are therefore equivalent.

Some data showing the transmission of 6 keV γ-rays of ^{55}Fe through two aligned lengths of square glass tubing, of side 0.10mm and total length 2.4m for various source positions is given in figure 9. The independence of the transmission on source position is confirmed. It is planned to conduct some tests of guides built up from float glass, which we expect to have improved transmission because of its better surface.

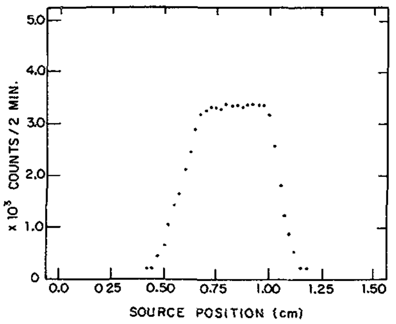

Fig. 9 : The transmission at 6 keV through 2.4m of square glass tubing of 1cm width outside vs. transverse source position.

3. Low-Pass filters.- In reporting, above, the tests of the Mössbauer resonance it was remarked that the 122 keV background was relatively reduced by the use of the γ-ray light pipe. This points up the possibility of designing and using light pipes as low-pass filters to allow greater transmission of low energy radiation than of contaminating high energy. Most absorbers tend to attenuate low energy photons more than they do high. Neutron guides have been used in this way to select long wave-length, or cold, neutrons. In several decay schemes useful for Mössbauer resonance there are very intense γ-rays or X-rays of higher energy than the γ-ray of interest. For example, the 10 times more copious 122 keV γ-ray of ^{57}Fe and the X-rays near 50 keV and 8 keV that are perhaps 100 times more numerous than the 6.25keV γ-ray of ^{181}Ta.

At the end of a light pipe of length sufficient to require at least one reflection of the radiation with the shortest wavelength, the aperture for capture is proportional to λ^2 and therefore higher energy radiation is discriminated against by a factor $(\lambda_0/\lambda_1)^2$. Reflection efficiency at the higher energy might be more degraded by surface roughness but that effect is offset by reduced photoelectric absorption.

Filters of this kind unfortunately accept only a small solid angle of even the lowest energy radiation. If the source is large, a correspondingly large and therefore very long pipe might be required. An alternative is to use a bundle of parallel small diameter tubes in a close packed array or small square passages between sets of intersecting flat plates. Ideally the walls should absorb the radiation not reflected. Each aperture serves a small area of the source and the wall thickness and the interstices result in a further loss in efficiency.

Experiments were carried out with a bundle of Pyrex tubes 30cm long, 1.5mm outside and 0.9mm bore kept straight by insertion in a large evacuated glass tube. The effect on the spectrum of ^{57}Co of interposing this bundle between the source and an intrinsic germanium detector was studied. Records of the low energy part of the spectra with and without the filter are shown in figure 10.

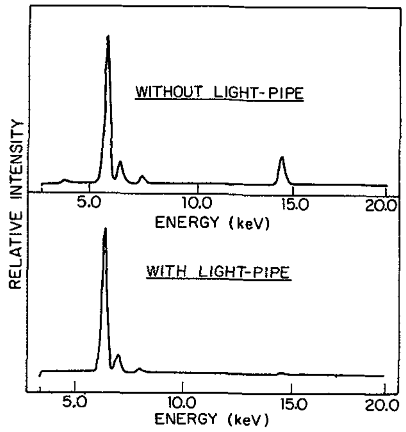

Fig. 10 : Comparison of the X-rays in the 6 keV region and the 14.4 keV γ-ray from ^{57}Co in copper as observed by an intrinsic Ge detector directly and through a bundled Pyrex low-pass filter. The peak below 5 keV is believed to be an escape peak associated with the 14 keV γ-ray and therefore is reduced with it.

The spectra are normalized to equal counts at 6 keV and the 14.4 peak is reduced by the filter by a relative factor of 6, which is very close to $(14.4/6)^2$. The small peak near 4 keV is thought to be an escape peak related to that at 14.4 and is correspondingly reduced.

This filter did not suppress, relatively, the 122 keV or 136 keV γ-rays because these were able to traverse the glass with little absorption. To overcome that we constructed a similar filter using "lead" glass (type 0120 Potash Soda Lead Glass, obtained from Houde Glass Co., Newark, N.J.). These tubes were 121cm long with outside diameters 3.2mm and bores 1.75mm. Figure 11 reproduces the parts of the spectrum below 20 keV and between 120 and 140 keV observed through this filter. In the higher energy seqment the ordinate is expanded 32-fold. A reduction of the relative intensity at 14.4 keV compared to that at 6 keV is similar to the result with Pyrex tubes. The 122 keV and the 136 keV photons are cut by about 25 times the reduction of the 6 keV, a factor determined by the solid angles subtended by the exit apertures. The tubes are too short to cause reduction to the critical angles for those energies. Little radiation traverses the lead glass itself to the detector.

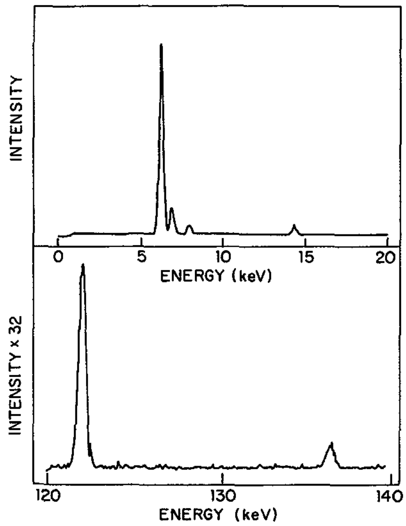

Fig. 11 : The spectra from a ^{57}Co-in-copper-source observed through a bundle of lead glass capillary tubes. The ordinate for the 120-140 keV region is expanded 32-fold. The transmission at 122 keV is determined from the solid angles subtended by the exit apertures.

The effective aperture of these filters for distributed sources can be increased by etching away the walls of the tubes in acid to allow more to be packed into the filter. Unfortunately, little can be done to increase the small solid angles accepted although the higher density of lead glass helps some. A good nickel coating should be better.

A single light pipe used as a low-pass filter from a small source can be further enhanced in performance, or tuned to a particular γ-ray, by use of a stop inside the bore. Such a stop can prevent all radiation from reaching the exit aperture. It is so designed that only rays striking the wall in a small range of angles of incidence, can by reflection, reach the far end. In that case, radiation for which the critical angle is less than that range will be cut off. The transmission efficiency at energies below the cut-off will be independent of energy. The transmission of 6 keV X-rays through a long 18mm inside diameter Pyrex tube equipped with a movable centered cylindrical brass stop 8mm in diameter was studied. Data collected from this experiment is reproduced in figure 12. When near the source end, the stop blocks all radiation and begins to allow radiation to strike the wall and be reflected when it is moved so far that its radius divided by its distance from the source is less than θ_c. The transmission continues to rise as the distance is increased and the rays at smaller angles are let by but, suddenly, the most steeply first reflected rays run into the stop, with a sudden decrease in the transmission. A similar second period begins as the stop continues to move further down the pipe.

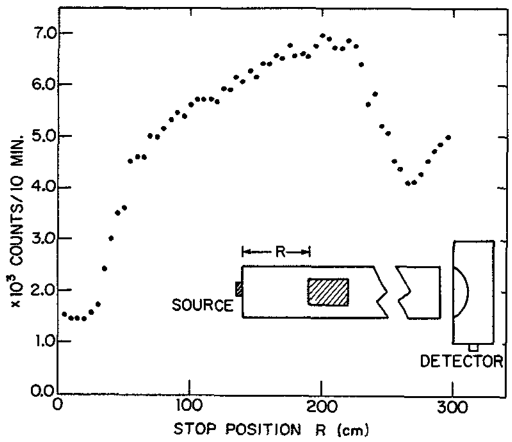

Fig. 12 : Intensity transmitted through a glass pipe from a small source vs. the distance from the source to a cylindrical brass stop. The stop diameter is 8mm and the glass bore is 18mm.

One must take into account the source diameter and symmetry to fit the pattern in detail. By appropriate development and analysis this scheme could be used to determine R(θ) functions for the wall materials.

4. Conclusions.- Light pipes can be constructed to enable the transmission of highly monochromatic Mössbauer γ-rays over reasonable distances. Ordinary laboratory tubing serves surprizing well but better ducts, for diameters more than a centimeter or so, could be assembled from plates of float glass suitably coated with a film of a metal such as nickel or copper for the 6 keV region. Light pipes can also be used as low-pass filters, for the preferential transmission of the low energy components from a multicomponent source. It is estimated that the statistical uncertainty in a measurement of the gravitational red-shift could be reduced to less than one in 10^4 using a suitably designed light pipe in conjunction with a ^{181}Ta source and absorber /21, 22/.

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