Nuclear Resonance Fluorescence in Ni^{60}†

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(Received May 4, 1956)

The gamma radiation emitted by a gaseous source of $Co^{60}Cl_2$ has been used to excite the 1.33-Mev level in Ni^{60}. The angular distribution of the resonance radiation characterizes the spin of the 1.33-Mev excited state as 2 and the spin of the Ni^{60} ground state as 0.

A resonance fluorescence self-absorption experiment has been performed and from it a mean life of $(1.1\pm0.2)\times10^{-12}$ second has been calculated for the 1.33-Mev excited state of Ni^{60}.

INTRODUCTION

EXPERIMENTS[1] with the delayed coincidence method indicated that the mean life of the 1.33-Mev excited state of Ni^{60} is shorter than 10^{-11} second. In view of this short upper limit for the lifetime, it appeared feasible to observe nuclear resonance fluorescence in Ni^{60} by using Co^{60} as the source of the exciting radiation. The combined momenta from the beta decay of Co^{60} and from the 1.17-Mev gamma ray leading to the 1.33-Mev level in Ni^{60} are sufficient to compensate for the recoil energy losses in the emission and absorption of the 1.33-Mev radiation.[2] In solid or liquid Co^{60} sources, however, collisions with the surrounding atoms will slow down the recoiling excited Ni^{60} nuclei before they emit the 1.33-Mev radiation. The 1.33-Mev gamma rays from such sources will consequently be off resonance and the resonance fluorescence effects will be too small to be observed above the background counting rate. In a gaseous source of approximately atmospheric pressure, on the other hand, the collision times are long compared with the nuclear lifetimes and the recoils due to the radiations preceding the 1.33-Mev level will be fully effective and will, at least for some of the 1.33-Mev gamma rays, restore the resonance condition.

Experiments with a gaseous source of $Co^{60}Cl_2$ were successful and permitted the determination of the angular distribution of the resonance radiation. However, as far as the measurement of the lifetime was concerned, the use of a molecular vapor introduced considerable difficulties. In contrast to the angular distribution, the evaluation of the lifetime depends on the detailed knowledge of the shape of the emission line, i.e., it depends on that fraction of the emitted gamma rays which fall into the energy region of the absorption line. In the case of Co^{60}, the dependence on the line shape is especially great because the absorption line is located in the very steep tail of the emission line. The resonance fluorescence effect which one observes with a source of gaseous $CoCl_2$ will, therefore, be very sensitive to the details of the complicated recoil phenomena. Under these circumstances the determination of the lifetime to within one order of magnitude appeared to be the best one could hope for. Fortunately, however, the resonance fluorescence effect turned out to be so large, i.e., the lifetime proved to be so short, that another approach became feasible: a self-absorption experiment. As long as the exciting line shows a smooth behavior in the region of the absorption line, the evaluation of the self-absorption experiment does not demand a detailed knowledge of the line shape. The resonance fluoresence absorption experiment will be useful in all those cases in which the lifetime is short ($\lesssim10^{-12}$ sec) and in which the use of a molecular vapor renders the evaluation of a resonance scattering experiment difficult.

PREPARATION OF THE SOURCES

High-specific-activity Co^{60} ($\sim$20 mC/mg) was obtained from Oak Ridge National Laboratory in the form of $CoCl_2$ in HCl solution. A solution containing approximately 7 millicuries was dried in an all-quartz vacuum system, the dry $CoCl_2$ distilled into a quartz ampoule of $\sim$0.6 ml volume and the ampoule sealed off. The sealed ampoule usually contained about 5 millicuries of Co^{60} corresponding to 0.9 mg/ml of $CoCl_2$. This amount of $CoCl_2$ was expected[3] to be totally volatilized at 1010°C, the pressure being 550 mm Hg. Later tests showed good agreement with this expected behavior. The quartz ampoules were used in vacuum tight steel containers. For the self-absorption experiments, cold-rolled steel containers were used because they do not contain nickel. For the angular distribution experiments, stainless steel containers were preferred because the quartz ampoules seemed to last longer when heated in stainless steel than when heated in cold-rolled steel.

ANGULAR DISTRIBUTION OF THE RESONANCE RADIATION

The experimental arrangement used for the measurement of the angular distribution is shown in Fig. 1 for an average scattering angle of 90°. The nickel ring scatterer was one-half of an inch thick, four inches high,

† Assisted by the joint program of the Office of Naval Research and the U. S. Atomic Energy Commission.

[1] Bay, Henry, and McLernon, Phys. Rev. **97**, 561 (1955).

[2] For a discussion of these problems, see, for example, E. Pollard and D. E. Alburger, Phys. Rev. **74**, 926 (1948).

[3] *Handbook of Chemistry and Physics* (1955–56) (Chemical Rubber Publishing Company, Cleveland, Ohio, 1955), p. 2156.

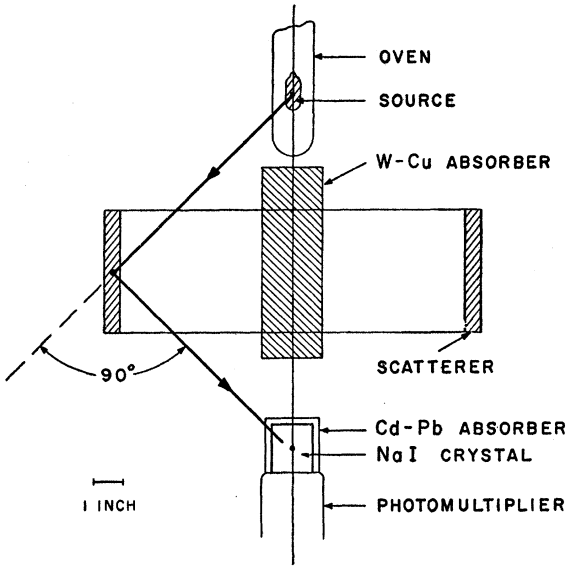

FIG. 1. Experimental arrangement. The relative position of source, scatterer, and detector depicted is the one corresponding to a mean scattering angle of 90°.

and measured one foot in diameter. The copper scatterer used for comparison purposes had the same dimensions.

Because of the high gamma-ray energy and the low atomic number of the scatterers, the contribution of elastic scattering to the counting rates was small, amounting to less than ten percent of the background counting rate. The total background for a 5-mC source was itself only about one-fifth of the resonance fluorescence effect when the full width at half-maximum of the 1.33-Mev peak was accepted in the channel of the single-channel pulse-height analyzer.

A typical pulse-height distribution of the scattered radiation for the geometry of Fig. 1 is compared in Fig. 2 with the pulse-height distribution of the incident radiation from Co^{60}. In the scattered radiation only the 1.33-Mev transition to the Ni^{60} ground state is present; the 1.17-Mev transition is not noticeable because elastic scattering is negligible compared with resonance scattering. This experiment clearly confirms the established order of emission of the two gamma rays in Ni^{60}, and points out another possible application of resonance fluorescence in the study of disintegration schemes of radioactive nuclei.

By moving source, ring scatterer, and detector relative to each other along the axis of symmetry, scattering angles ranging from 90° to 150° were realized. The differential cross sections, measured for five representative angles, are compared in Fig. 3 with the distribution expected for a spin sequence 0-2-0 when the actual angular resolution is taken into account. In order to insure that 0-2-0 is the only spin sequence fitting the experimental data, a study of the angular distribution for all possible spin sequences J_0-J-J_0, including mixtures of dipole and quadrupole radiation,[4] has been carried through. It was found that the coefficient A_4/A_0 in the angular distribution $1+(A_2/A_0)P_4+(A_4/A_0)P_4$ reaches its largest value for $J_0=0$, $J=2$. Actually the value $A_4/A_0=1.143$ for the 0-2-0 sequence exceeds by more than a factor of two the largest value (0.51) for any other spin sequence. Experimentally one finds a ratio $A_4/A_0=1.10\pm0.1$, i.e., clearly >0.51. This means that $J_0=0$; $J=2$ are the only spin values agreeing with the experimental results.

The angular distribution experiment thus determines the spin of the ground state of Ni^{60} as zero and the spin of the 1.33-Mev excited state of Ni^{60} as two.

DETERMINATION OF THE LIFETIME OF THE 1.33-Mev STATE

A. From the Number of Resonance Scattered Quanta

The lifetime τ_γ and the cross section σ_{Av}, averaged over the incident line, are related by the expression[5]

$$\tau_\gamma=\frac{g_2}{g_1}\frac{2.53}{(E_{res})^2\sigma_{Av}}\frac{N(E_{res})}{N}, \quad (1)$$

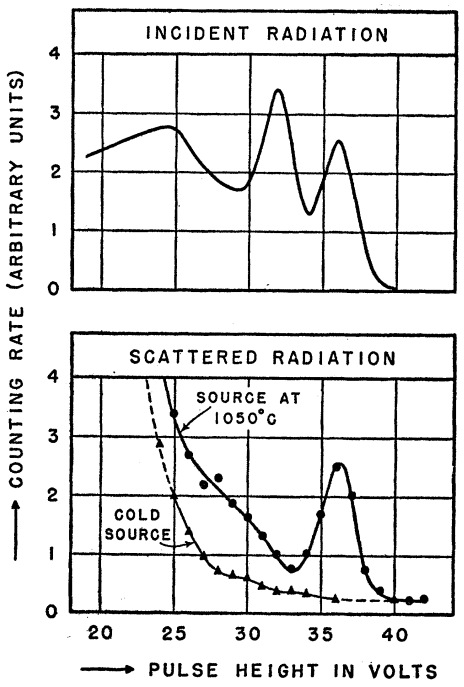

FIG. 2. Resonance fluorescence in Ni^{60}. The pulse-height distribution of the incident radiation (top) clearly shows two peaks corresponding to the 1.17-Mev and the 1.33-Mev gamma rays emitted in the decay of Co^{60}. The scattered radiation with the gaseous source consists mainly of 1.33-Mev resonance radiation, indicating that this gamma ray is the one leading to the ground state of Ni^{60}.

[4] In view of the short lifetime of the level, higher multipoles were excluded.

[5] See, e.g., F. R. Metzger, Phys. Rev. **101**, 286 (1956)

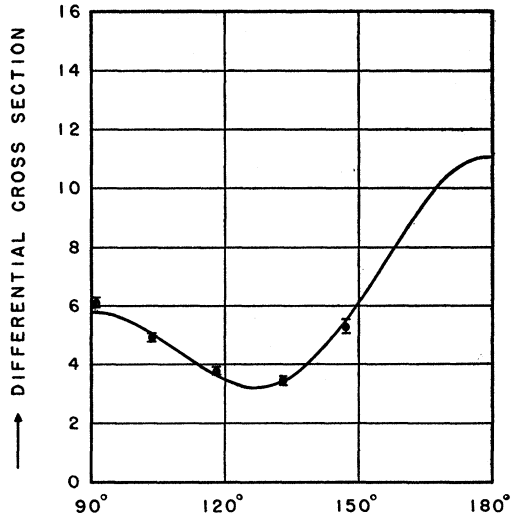

FIG. 3. Angular distribution of the resonance radiation from the 1.33-Mev level in Ni^{60}. The solid line represents the theoretical angular distribution for an excited state with spin 2 and a ground state with spin 0, corrected for the finite angular resolution. The differential cross section is given in arbitrary units.

where τ_γ is the mean life in seconds, g_2 and g_1 are the statistical weights of the excited state and the ground state, respectively, E_{res} is the resonance energy in ev, σ_{Av} the average cross section in barns, and $N(E_{\text{res}})/N$ is the fraction of the gamma rays of the 1.33-Mev line falling into a one-ev interval at the resonance energy.

Once the angular distribution of the resonance radiation is known, the determination of the average cross section is straightforward: σ_{Av} is obtained from a comparison, at a given angle, of the number of resonance scattered quanta with the total number of incident 1.33-Mev gamma rays.

The evaluation of $N(E_{\text{res}})/N$, on the other hand, presents considerable difficulties if the source is not in the form of monatomic vapor. In the case of $CoCl_2$, all one can do is give an upper limit for $N(E_{\text{res}})/N$ and thus an upper limit for τ_γ. One arrives at this upper limit by observing that $N(E_{\text{res}})/N$ assumes its largest value when the radioactive Co^{60} nuclei are not bound to other atoms which might share the recoil momentum and thus narrow the emission line. From the line shape for free Co^{60} nuclei, reproduced in Fig. 4, an upper limit of 0.0051 is obtained for the fraction of the incident line falling into a one-ev interval at E_{res}. If this value is combined with the experimental value for the average cross section, $\sigma_{\text{Av}}=(1.6\pm0.1)\times10^{-26}$ cm^2, the upper limit for the mean life τ_γ is calculated as 2.3×10^{-12} sec.

A lower limit cannot be set, because for suitably chosen properties of the $CoCl_2$ molecule, about which little is known, $N(E_{\text{res}})/N$ could be very small indeed and thus any lifetime shorter than the upper limit could be explained.

The result of the measurement of σ_{Av} is, therefore, that $\tau_\gamma<2.3\times10^{-12}$ second.

B. From a Resonance Self-Absorption Experiment

If the radiation emitted by a gaseous $CoCl_2$ source passes through a nickel absorber before it strikes the nickel scatterer, the line shape of the 1.33-Mev line will be modified because, in addition to the "normal" absorption by Compton effect, photoeffect and pair creation, selective absorption in the region of the 1.33-Mev absorption line has taken place. Because of its restriction to a small portion of the incident line, (see Fig. 4), the effect of the selective absorption on the intensity of the transmitted beam will, in general, be very small compared with the effect of the "normal" absorption. If, however, a detector is used which is only sensitive to the narrow region of the absorption line—and resonance scattering represents such a detector—then the selective absorption becomes measurable. However, even if one uses self-indication, the "normal" absorption is still considerably larger than the selective absorption and it is advantageous to use a comparison method which eliminates the "normal" absorption. The procedure finally adopted was the following: Two absorbers of similar shape, one made of nickel and one of nickel-free steel, are compared, as far as their normal absorption is concerned, using the direct 1.33-Mev gamma ray beam from a cold $CoCl_2$ source. By changing the thickness of one of the absorbers, the attenuation of the beam by the two absorbers is made to agree to within better than 1 percent. Then, with a gaseous $CoCl_2$ source and a nickel scatterer, the resonance fluorescence effect is measured while the nickel absorber and the steel absorber are alternately inserted between source and scatterer. The ratio of the two readings, corrected for the initial mismatch, gives then directly the selective

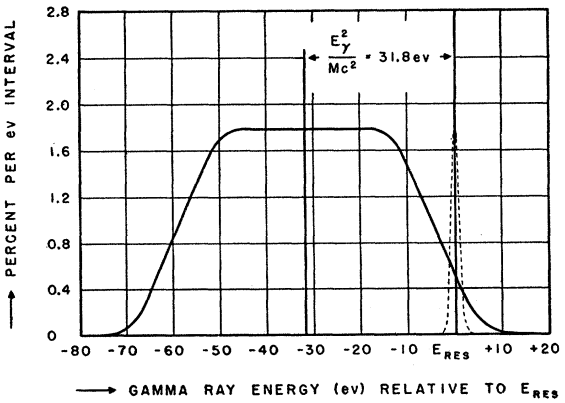

FIG. 4. Line shape of the 1.33-Mev radiation emitted by a gaseous source of Co^{60} atoms, neglecting the angular correlation of the Ni^{60} gamma rays. The absorption line for a nickel absorber at room temperature is indicated by the dashed curve.

TABLE I. Results of the self-absorption experiment. The counting rates with the nickel absorber are quoted relative to those with the steel absorber (=100).

	Direct beam	Resonance effect
Steel absorber	100	100
Nickel absorber	100.9±0.1	95.8+0.7

attenuation of the resonance radiation by the nickel absorber.

The results of the actual measurements are summarized in Table I. One notices that the nickel absorber which, on the basis of the direct beam experiment, is actually slightly too thin, is more effective than the steel absorber in reducing the resonance effect. From the data summarized in Table I, it is concluded that the nickel absorber attenuates the resonance fluorescence effect by a factor 0.949±0.007. The evaluation of this attenuation in terms of the mean life τ_γ is straightforward if one can assume the absorbers and the scatterer to be thin. In the present experiment, this assumption was justified because scatterer and absorber were of approximately the same thickness and from the value of the attenuation in the absorber this thickness is seen to give rise to only 5% absorption.

The weakness of the resonance absorption also indicates that the natural width Γ must be rather small, i.e., $\ll$1 ev. This means that the natural width is small compared to the Doppler width $\Delta=(E/c)(2kT/M)^{\frac{1}{2}}$ caused by the thermal agitation of the absorbing nuclei. The effective absorption cross section[6] has, therefore, the pure "Doppler form"

$$\sigma_D(E)=\sigma_0\frac{\sqrt{\pi}}{2}\frac{\Gamma}{\Delta}\exp\{-[(E-E_r)/\Delta]^2\}, \qquad (2)$$

where $\sigma_0=(g_2/g_1)(\lambda^2/2\pi)$ is the individual cross section at exact resonance.

The resonance effect from the scatterer is proportional to

$$R=\int N(E)\sigma_D(E)dE,$$

if $N(E)dE$ is the number of gamma rays in the energy interval dE. An absorber of thickness d, with n Ni^{60}-nuclei per cm^3, will reduce the incident beam by a factor $[1-n\sigma_D'(E)d]$ and will thus give rise to an attenuation of the resonance effect

$$A=\int[1-n\sigma_D'(E)d]N(E)\sigma_D(E)dE\Big/\times\int N(E)\sigma_D(E)dE. \qquad (3)$$

[6] That is, the resonance cross section $\sigma=\sigma_0/\{1+[(E-E_r)/\frac{1}{2}\Gamma]^2\}$ averaged over all the absorbing nuclei, which are assumed to exhibit a Maxwellian velocity distribution.

A different effective cross section σ_D' was introduced for the absorber because scatterer and absorber might be at different temperatures (different Doppler widths Δ_a and Δ_s).

If we assume that, compared with the rapidly varying σ_D, $N(E)$ is a slowly varying function of the energy, $N(E_r)$ can be taken out of the integral. Using the expression (2) for the cross section, the integrals in (3) can then be evaluated and one obtains for the attenuation, after expressing σ_0 in terms of λ

$$A=1-nd\frac{(g_2/g_1)\lambda^2\Gamma}{4[\pi(\Delta_a^2+\Delta_s^2)]^{\frac{1}{2}}}. \qquad (4)$$

If absorber and scatterer were in the gaseous phase, the actual temperatures would be used to calculate the Doppler widths. If, however, solid absorbers and scatterers are employed, the crystalline binding, characterized by the Debye temperature, has to be taken into account. This was done by Lamb[7] for the case of neutron absorption, and his results can be directly applied to the absorption of gamma rays. For weak binding, Lamb showed that the actual temperature has to be replaced by an effective temperature which is somewhat larger than the actual temperature and which depends on the ratio of actual temperature and Debye temperature.

In the present absorption experiment, the temperature of the absorber was 340°K, the temperature of the nickel scatterer 300°K. Taking for the characteristic (Debye) temperature of nickel the value 375°K, one reads off Lamb's graph the effective temperatures 392° and 350°K, respectively. The Doppler widths corresponding to these temperatures are $\Delta_a=1.46$ ev and $\Delta_s=1.38$ ev. After taking into account the finite angular spread of the beam striking the scatterer, the effective thickness of the nickel absorber was calculated to be $d=1.18$ cm. For an excited state with spin 2 and a ground state with spin 0, $g_2/g_1=5$. Finally $\lambda^2=8.63\times10^{-21}$ cm^2, and $n=2.39\times10^{22}$/cm^3. If all these values, together with the experimental attenuation $A=0.949$, are inserted into (4), a natural width $\Gamma=5.97\times10^{-4}$ ev$=9.56\times10^{-16}$ erg is obtained for the 1.33-Mev level of Ni^{60}. This width corresponds to a mean life $\tau_\gamma=\hbar/\Gamma=1.1\times10^{-12}$ second.[8] In view of the statistical uncertainty in A and the correction for the binding of the absorbing nuclei, the standard deviation in our value of τ_γ is estimated to be 0.2×10^{-12} second. Thus the final result of the self-absorption experiment is $\tau_\gamma=(1.1\pm0.2)\times10^{-12}$ second.

Upon inspecting Fig. 4, one might have some

[7] W. E. Lamb, Phys. Rev. 55, 190 (1939).

[8] The difference between this value and the one (7.5×10^{-13}) quoted at the 1956 New York Meeting of The American Physical Society [Franz R. Metzger, Bull. Am. Phys. Soc. Ser. II, 1, 40 (1956)] is the result of an arithmetical mistake.

misgivings as to the validity of the assumption that $N(E)$ is constant in the region of the absorption line. It should be pointed out that, in view of the symmetry of σ_D with respect to E_r, $N(E)$ does not need to be constant over the region of the absorption line as long as it is linear, i.e., as long as the higher derivatives of $N(E)$ with respect to E are small. Some estimate of the magnitude of the higher derivatives of $N(E)$ may be obtained by a study of the change in the resonance fluorescence effect upon heating of the nickel scatterer. This temperature effect depends on the higher derivatives in closely the same way as the self-absorption experiment. A small temperature dependence of the resonance effect implies a small correction to the simple expression (4) used for the analysis of the self-absorption.

In a separate experiment the scatterer was heated from room temperature to 300°C and the change in the resonance fluorescence effect was found to be smaller than 5 percent. This indicates that the error introduced in the self-absorption experiment by assuming $N(E)$ to be constant over the region of the absorption line is small indeed.

CONCLUSIONS

The resonance fluorescence self-absorption method allows the determination of lifetimes even if the line shape of the exciting radiation is not known because of the complexity of the recoil phenomena.

The mean life of $(1.1\pm0.2)\times10^{-12}$ second measured for the 1.33-Mev excited state of Ni^{60} is approximately five times shorter than the single-particle estimate.[9] Compared with other electric quadrupole transitions in the weak coupling region of the periodic table,[10,5] this transition is slowed down by a factor of from two to five. This slowing down is not unexpected in view of the magic number of protons in Ni^{60}.

ACKNOWLEDGMENTS

The author wishes to thank Mr. W. B. Todd for his help in taking the data and in maintaining the equipment, Dr. Leonard Eisenbud for valuable discussions, and Mr. F. B. Thiess for aid with the computations.

[9] V. F. Weisskopf, Phys. Rev. **83**, 1073 (1951).
[10] G. M. Temmer and N. P. Heydenburg, Phys. Rev. **99**, 1609 (1955).

PHYSICAL REVIEW VOLUME 103, NUMBER 4 AUGUST 15, 1956

Decay Constants of K^{40} as Determined by the Radiogenic Argon Content of Potassium Minerals

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(Received March 28, 1956)

It is shown that the potassium-argon age of young minerals depends almost linearly on the decay constant for electron capture in K^{40} and is very insensitive to the decay constant for beta emission. This fact permits calculation of λ_e by comparing the concordant uranium-lead age of cogenetic uraninite with A^{40}/K^{40} ratios found in young samples of mica. It is found that $\lambda_e=(0.557\pm0.026)\times10^{-10}$ yr^{-1}. Similar comparisons with older mica samples indicate that satisfactory agreement with the uraninite ages are obtained by use of this value of λ_e together with $\lambda_\beta=(0.472\pm0.05)\times10^{-9}$ yr^{-1}. It is concluded that there is no conflict between the decay constants inferred by this geological method and those found by direct counting experiments.

I. INTRODUCTION

A NUMBER of papers have appeared in which the branching ratio of K^{40} has been determined by measurement of the radiogenic argon content of potassium minerals. Aldrich and Nier[1] showed, by semi-quantitative measurements of the ratio of radiogenic A^{40} to K^{40} in minerals of approximately known geological age, that the branching ratio was of the order of 0.1. Earlier counting experiments[2] had indicated that the branching ratio was of the order of 1. Inghram, Brown, Patterson, and Hess[3] determined the branching ratio to be 0.126 by measuring the ratio of radiogenic argon to radiogenic calcium in sylvite (KCl). Subsequently Mousuf[4] and Russell, Shillibeer, Farquhar, and Mousuf[5] published a branching ratio of 0.06 determined by comparing A^{40}/K^{40} ratios from feldspars with various age

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[1] L. T. Aldrich and Alfred O. Nier, Phys. Rev. **74**, 876 (1948)..
[2] E. Bleuler and M. Gabriel, Helv. Phys. Acta **20**, 67 (1947).
[3] Inghram, Brown, Patterson, and Hess, Phys. Rev. **80**, 916 (1950).
[4] A. K. Mousuf, Phys. Rev. **88**, 150 (1952).
[5] Russell, Shillibeer, Farquhar, and Mousuf, Phys. Rev. **91**, 1223 (1953).