Resonant Scattering of X Rays from Magnesium and Silicon

RALPH A. TOBIN
Nucleonics Division, U. S. Naval Research Laboratory, Washington, D. C.
(Received May 26, 1960)

Energy levels at 10.15±0.06 Mev in magnesium and 11.40±0.06 Mev in silicon have been studied using resonant scattering from a continuous spectrum of x rays by samples of these materials. Using tentative spin assignments and assuming no branching, level widths of 4.80 (+1.6, −1.4) electron volts and 2.89 (+1.0, −0.8) electron volts for magnesium and silicon, respectively, have been determined from absorption measurements.

I. INTRODUCTION

IT was suggested by Schiff[1] in 1946 that a continuous spectrum of x rays, such as the bremsstrahlung obtained from targets in electron accelerators, could be used to excite some nuclear levels. In 1956 Hayward and Fuller[2] observed such level excitation when 15.1-Mev x rays were selectively scattered by a carbon sample being irradiated with bremsstrahlung from a betatron. Since that time several investigators have observed this kind of scattering from other elements.[3] This paper describes studies of the 10.15-Mev level in magnesium and the 11.40-Mev level in silicon made by means of the resonant scattering of x rays.

II. EXPERIMENTAL ARRANGEMENT

The system used for this experiment consisted of a bremsstrahlung source, scattering sample, a detector for the scattered x rays and a pulse-height analyzer to sort and record the data. A sketch of the experimental geometry is shown in Fig. 1. Here, a bremsstrahlung spectrum is generated when electrons accelerated in a betatron strike a platinum target at (A). The resultant radiation is monitored by the ionization chamber at (B) and collimated by the lead block (C) before passing through the shielding wall (D) on its way toward the absorber (E) and scattering sample (F). The detector cubicle contains a 4-inch by 4-inch NaI(Tl) crystal, multiplier phototube and pre-amplifier (G) shielded by lead (H) and borax (I). The plug (K) in the hole through the lead shielding is used to help discriminate in favor of radiation in the 10 to 12-Mev region. Brass, from which this filter is made, has the minimum in its attenuation versus energy curve at about 10 Mev.

The radiation from the platinum target in the betatron was collimated along its axis of symmetry so that at the positions of the scattering sample (F) and the absorber (E) the beam had diameters of 4 and 2.25 inches, respectively. The axis of symmetry was determined by irradiating copper samples at an energy above the threshold for the reaction $Cu^{63}(\gamma,n)Cu^{62}$ and then using the positron activity associated with the decay of Cu^{62} to make an auto-radiograph. The copper samples were cut from $\frac{1}{16}$-inch flat stock. Irradiations of 10 minutes duration were made at two positions, one at a point 15 cm in front of (A) and the other at (B). After an irradiation, the sample was placed on top of a film holder containing a piece of "x-ray" type film and left for 10 minutes. After development the film was examined to determine the position of maximum exposure which was assumed to coincide with the axis of symmetry of the primary x-ray beam.

The shielding shown in Fig. 1 was necessary in order to reduce the background to the extent where the radiation of interest could be resolved. The background comes from two principal processes, neutrons from (γ,n) reactions and x rays (direct or scattered) from the platinum target of the betatron. The neutron component gives rise to capture gamma rays which are distinguished from the other component here because of the difference in arrival time at the crystal. The direct or scattered radiation from the target is almost simultaneous in time with the primary x-ray pulse and can be reduced only by bulk shielding. However, the neutron component has a measurable build-up and decay time that is usually characteristic of the moderator material and geometry in the vicinity of interest.

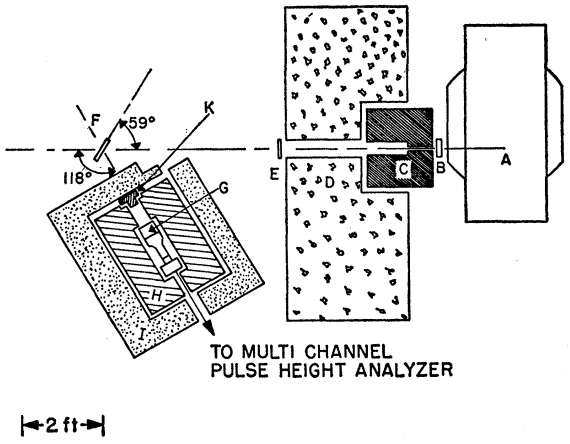


FIG. 1. Experimental arrangement. A—x-ray source; B—ion chamber; C—lead collimator; D—concrete shielding wall; E—absorber; F—scatterer; G—NaI(Tl) crystal, multiplier phototube, and pre-amplifier; H—lead shielding; I—borax shielding; K—brass filter.

[1] L. I. Schiff, Phys. Rev. **70**, 761 (1946).
[2] E. Hayward and E. G. Fuller, Phys. Rev. **106**, 991 (1957).
[3] L. Cohen, R. A. Tobin, and J. McElhinney, Phys. Rev. **114**, 590 (1959); L. Cohen and R. A. Tobin, Nuclear Phys. **14**, 243 (1959); F. D. Seward *et al.*, Bull. Am. Phys. Soc. **5**, 68 (1960).

Much of this latter background can be eliminated by efficient gating of the detection system.

The primary x-ray pulse used in this experiment was triangular in shape (when displayed as intensity versus time the slopes of the rise and decay were similar) with a half microsecond duration and was repeated with a regular period of 1/180 second. Efficient gating can be realized by using a gating pulse whose duration corresponds to that of the primary x-ray pulse and has the correct phase with respect to this primary pulse. If these conditions are satisfied then the background caused by gamma rays resulting from neutron capture will be at a minimum.

The shape of the primary x-ray pulse was measured by observing with a multiplier phototube the Čerenkov radiation from the secondary electrons resulting from the passage of the x-ray pulse through a thin slab of Lucite. The gating pulse was then adjusted to be slightly longer in duration. The gating was accomplished by operating the pulse-height analyzer in the coincidence mode and supplying a gate pulse (or coincidence pulse) only for the duration of the x-ray pulse. The correct phase between the x-ray pulse and the gate was determined by observing the recorded pulses due to annihilation radiation and Compton scattered x rays from a sample as a function of phase between the x ray and gating pulses.

Another source of background was electrical pickup. This was associated principally with the modulator used to perturb the magnetic field in the betatron for the initiation of an x-ray pulse. Because of the impedance (measurable though small) between the ground side of the modulator and earth ground, a portion of the modulator pulse appeared on the ground side of all circuits associated with the modulator. This included the pulse-height analyzer by way of the gate-generator trigger pulse which was obtained from the modulator system. Whenever the amplitude of this pickup was above the baseline setting of the analyzer it was analyzed and counted. This source of background was minimized by reducing the modulator to earth impedance and decoupling, as much as possible, the recording system from the modulator system. This procedure resulted in a pickup signal at the input to the analyzer of 0.009 volt which corresponds to an 18-kev gamma ray giving up its full energy to the crystal. The baseline on the analyzer was then set just above this level.

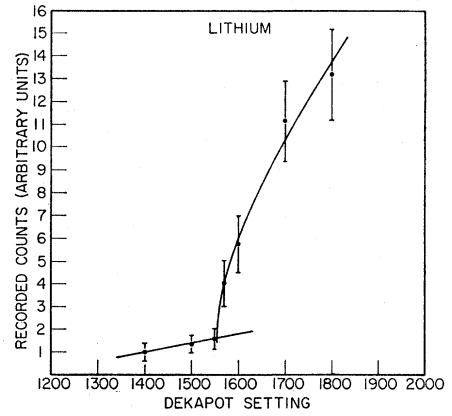


FIG. 2. Thresholds for Li^6 (3.56 Mev) and C^{12} (15.1 Mev) used in calibration of the betatron.

III. EXPERIMENTAL PROCEDURE

The scattering samples and absorbers, made of powdered metal, were contained in Lucite boxes having entrance and exit windows for the primary x-ray beam made of $\frac{1}{32}$-inch Lucite. Magnesium samples were made of 70–80 mesh metal powder and the scattering sample and absorber had areal densities of 1.20 grams/cm^2 and 1.29 grams/cm^2, respectively. The silicon samples were made of 100 mesh metal powder with areal densities of 2.23 grams/cm^2 for the scattering sample and 1.79 grams/cm^2 for the absorber. Samples of high purity aluminum having the same areal densities as the magnesium and silicon scattering samples and contained in similar Lucite boxes were used for background determinations.

The energy scale of the betatron was calibrated using the thresholds for excitation of the 3.56 Mev and 15.1-Mev levels in Li^6 and C^{12}, respectively by means of resonance absorption of x rays. The thresholds for these are shown in Fig. 2.

The levels in magnesium and silicon discussed in this paper were first observed during a survey of several materials[4] for resonant scattering of x rays. After this survey, these levels were studied more intensively in an effort to measure some of their characteristics. Shown in Fig. 3 are the spectra obtained when the Mg, Si, and Al samples were irradiated at a peak bremsstrahlung energy of 13.1 Mev. The shaded spectra represent the background obtained with the aluminum scatterer. This energy was chosen for the irradiation since it is below

[4] R. A. Tobin and L. Cohen, Report of Naval Research Laboratory Progress, July 1959 (unpublished).

the thresholds for the 15.1-Mev level in C^{12} (from the Lucite boxes) and the (γ,n) reaction in Al^{27}. From these spectra it was decided to use those counts above channel 70 as a measure of the yield from the two levels of interest.

The thresholds for these levels are shown in Fig. 4 and were obtained by plotting those counts appearing above channel 70 versus the peak energy of the bremsstrahlung spectrum. For magnesium there is an abrupt change in the slope of the yield curve at 10.10 Mev. The gradual increase of yield with energy below this point is associated with capture gamma rays resulting from the neutrons associated with the $Mg^{25}(\gamma,n)Mg^{24}$ reaction which occurs above the threshold energy of 7.33 Mev. The change observed at 10.10 Mev is attributed to gamma rays scattered from a level in magnesium. The sharp change in the slope of the yield curve for silicon that occurs at 11.35 Mev is considered to be associated

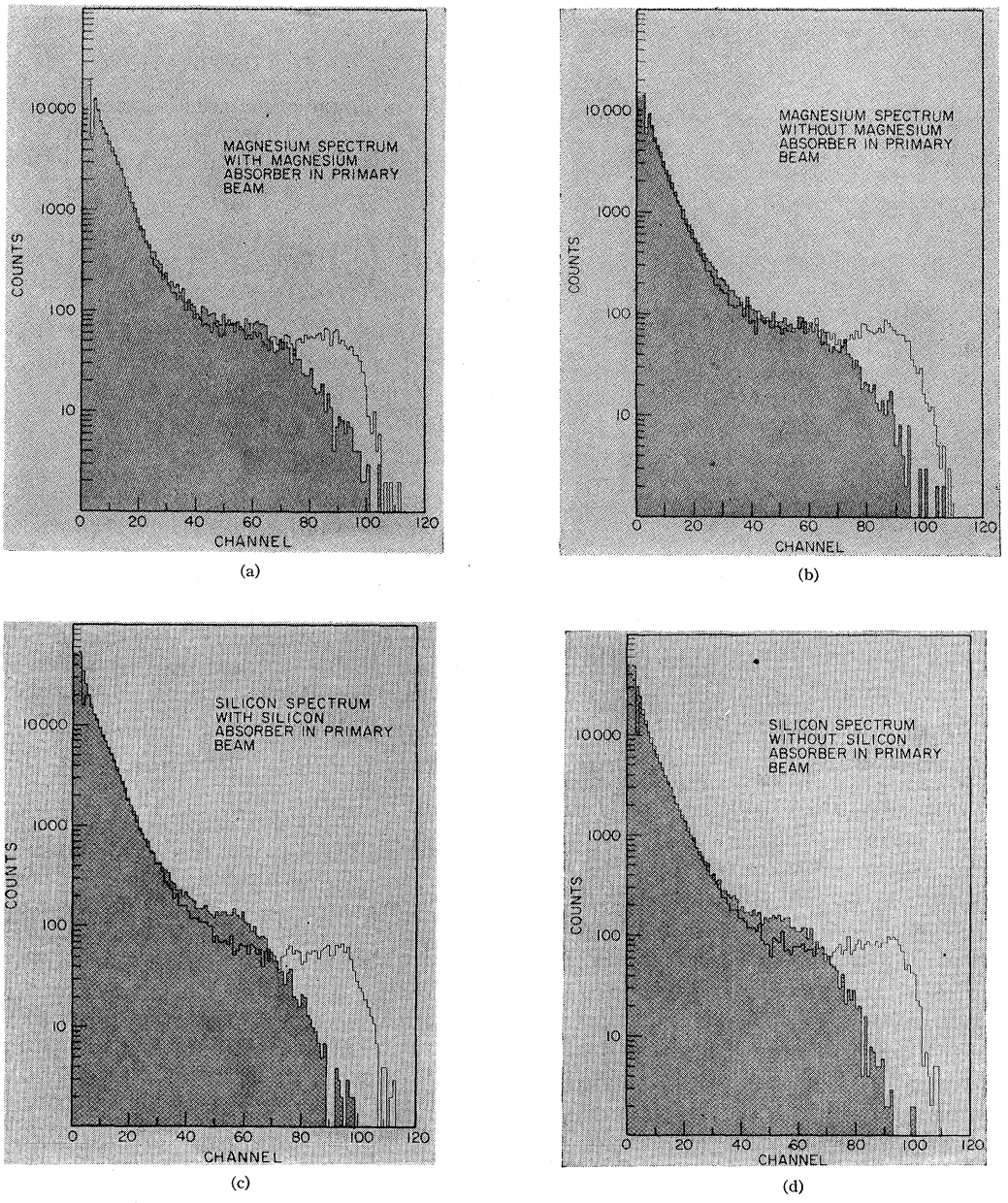
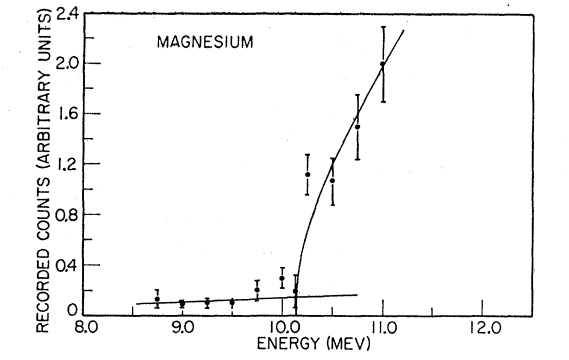
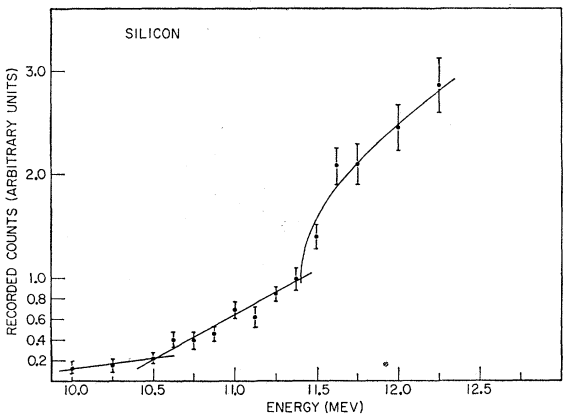


FIG. 3. Shown above are the spectra obtained from magnesium and silicon for irradiations made with and without an absorber of similar material in the primary x-ray beam. The shaded spectra were obtained using the equivalent number of grams/cm² of aluminum as a scatterer for background determination. The energy scale for all these spectra is such that the peaks for the 0.51-Mev annihilation radiation and the 4.43-Mev gamma ray from the decay of C^{12} (observed using a Po-Be neutron source) occur in channels 5 and 50, respectively.



(a)



(b)

FIG. 4. Thresholds for the levels in Mg and Si.

with gamma rays scattered by a level at this energy. The change that occurs at 10.52 Mev is associated with capture gamma rays resulting from neutrons associated with the $Si^{30}(\gamma,n)Si^{29}$ reaction which has a threshold at this energy.

An indirect measure of the level widths was obtained by observing the attenuation produced when a sample of the material under investigation was placed in the primary beam. This was accomplished by using the scattering sample as an indicator and observing the yield from the level of interest when an absorber of similar material was alternately placed in and out of the primary beam. This procedure was repeated using aluminum as a scatterer. The differences between these two yields above channel 70 were used to determine the attenuation. Shown in Fig. 3 are the spectra resulting from these runs from which we get transmissions of

$$\text{Transmission}(T)=\frac{\text{Yield(absorber-in)}}{\text{Yield(absorber out)}}$$

$$=\frac{1574-608}{1868-589}=0.75\pm0.05,$$

for magnesium and

$$T=\frac{1744-436}{2117-486}=0.80\pm0.04,$$

for silicon.

If there were no nuclear absorption and only the electronic effects, the calculated transmission would be (0.98) for magnesium and (0.96) for silicon. These were determined using $I/I_0=e^{-ux}$ with $(ux)=(0.0296)$ and (0.0433) for magnesium and silicon, respectively.

IV. COMPUTATIONS

When a sample of material is irradiated with a spectrum of x rays, the number of interactions per cm^2 between a particular level of nuclear excitation and the x rays is given very nearly by

$$Y'=\int_{x_1}^{x_2}\left(\frac{\Gamma}{2}\right)(I_0)(1-e^{-u})\left(\frac{\sigma_n}{2(N_e/N)\sigma_e+\sigma_n}\right)dx, \quad (1)$$

where $u=(2\sigma_eN_eT+\sigma_nNT)/\cos\alpha$, Γ is the full width of the level measured at half amplitude in units of electron volts, I_0 is the number of photons per cm^2 contained in the incident bremsstrahlung spectrum having an energy corresponding to that of the level and is measured in units of photons per cm^2 per electron volt, σ_e is the cross section for electronic absorption or scattering in units of cm^2 and is considered constant over the level under investigation, σ_n is the cross section of the level for absorption of x rays in units of cm^2 and is a function of energy (or x) as noted below, N_e is the number of atoms per cm^3 which participate in the electronic absorption, N is the number of nuclei per cm^3 which contain the level being investigated, T is the thickness in cm of the scattering sample, and

$$x=\left(\frac{E-E_r}{\Gamma/2}\right),$$

where E_r is the energy of the level at resonance and E is the energy of the incident photon. The normal to the plane of the scattering sample divides the angle between the axes of the x-ray beam and crystal into equal angles α. The limits of integration (x_1,x_2) are chosen such that they bracket the level and are far enough removed from E_r so that the cross section is relatively small. If now we interpose between the x-ray source and the scattering sample an absorber of the same material as the scattering sample then the number of interactions will be reduced and is given by

$$Y''=\int_{x_1}^{x_2}\left(\frac{\Gamma}{2}\right)(I_0)(e^{-v})(1-e^{-u})\left(\frac{\sigma_n}{2\sigma_e+\sigma_n}\right)dx, \quad (2)$$

where $v=(\sigma_nN'T'+\sigma_eN_e'T')$ and the primed symbols refer to the absorber. With these expressions we can

express the transmission as

$$T=Y''/Y'. \tag{3}$$

For the energy range covered between the limits (x_1,x_2), the only dependent variable in these expressions is σ_n and this can be described using the Breit-Wigner single level formula for values of E near E_r, as follows,[5]

$$\sigma_n(E)=\sigma_0\Big/\left[1+\left(\frac{E-E_r}{\Gamma/2}\right)^2\right], \tag{4}$$

where $\sigma_n(E)$ is used to indicate the functional dependence on energy and σ_0 is the value of the cross section at the resonance energy E_r. This expression describes the shape of the cross section as a function of energy and width Γ and could be used for σ_n if it were not for the thermal motion of the nuclei in the sample material that causes a "Doppler" broadening of the level. Expressed in terms of energy there is a change from an initial energy E to an energy relative to the source E' given by

$$E'=E(1\pm v/c). \tag{5}$$

This then is the energy the photon has relative to the sample nuclei which have a velocity component v toward or away from the photon source and must be used instead of E in the Breit-Wigner formula above. If we assume that the distribution of velocities produced by the thermal agitation can be represented by a Maxwellian distribution function and express the motions in terms of kinetic energy,[6] then the probability for finding a nucleus with a velocity component that will produce a relative energy E' with respect to an incoming photon of energy E is given by

$$W(E')dE'=\frac{\pi^{-\frac{1}{2}}\exp[-(E'-E_r)^2/\Delta^2]dE'}{\Delta}, \tag{6}$$

where

$$\Delta=\left(\frac{E_r}{c}\right)\left(\frac{2kT_e}{M}\right)^{\frac{1}{2}},$$

for photons and is called the "Doppler" width. In this expression c is the velocity of light, k is Boltzmann's constant, M is the mass of the nucleus with which the level is associated and T_e is the "effective" temperature which will be identified later.

The observed cross section then will be the product of the "Doppler" broadened Breit-Wigner cross section $\sigma_n(E')$ and the probability for finding nuclei with this relative energy $w(E')$. Reference (5) expresses the result of this combination as

$$\psi\left(\frac{\Gamma}{\Delta},x\right)=\frac{1}{2\pi^{\frac{1}{2}}}\left(\frac{\Gamma}{\Delta}\right)\times\int_{-\infty}^{+\infty}\frac{\exp[-\frac{1}{4}(\Gamma/\Delta)^2(x-z)^2]}{(1+z^2)}dz, \tag{7}$$

[5] H. A. Bethe, Revs. Modern Phys. **9**, 140 (1937).
[6] This treatment is described by Bethe, reference 5.

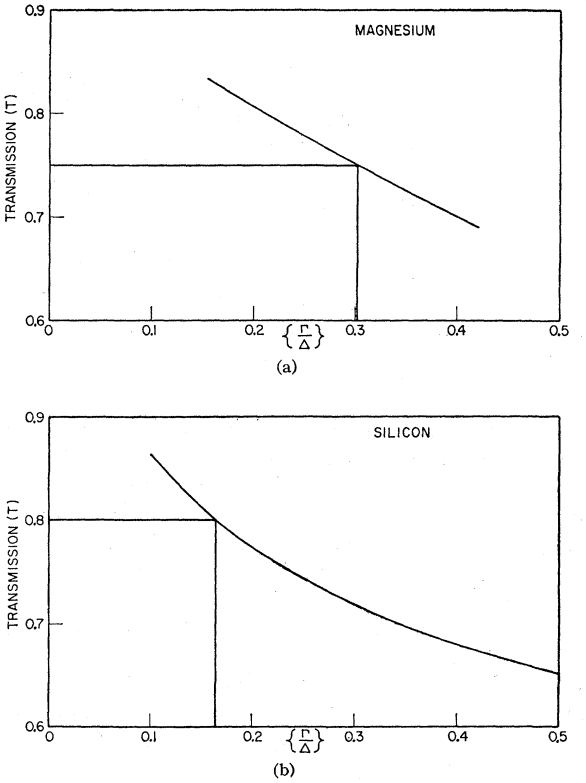


FIG. 5. Plot of transmission T versus Γ/Δ.

where

$$z=\frac{(E'-E_r)}{\Gamma/2}.$$

This integral has been evaluated by Rose *et al.* and is available in tabular form.[7] The expected cross section at a given x is then found from

$$\sigma_n\left(\frac{\Gamma}{\Delta},E\right)=\sigma_0\psi\left(\frac{\Gamma}{\Delta},x\right), \tag{8}$$

and these values are used in (3).

The value of the cross section at $E=E_r$ is given by the Breit-Wigner single level resonance relation and for the case under consideration here $(\Gamma_\gamma/\Gamma=1)$

$$\sigma_0=4\pi\lambda\!\!\!^{-2}\frac{(2I_c+1)}{2(2I_t+1)}, \tag{9}$$

where $\lambda\!\!\!^{-}=\lambda/2\pi$ is the rationalized wavelength of the radiation at E_r, I_c is the spin of the compound nucleus and I_t is the nuclear spin of the ground state of the target nucleus. For isotopes studied here I_t was known and I_c was based upon arguments presented in the discussion.

[7] M. E. Rose *et al.*, Westinghouse Electric Corporation Atomic Powers Division Report WAPD-SR-506, Vols. I and II, available from Office of Technical Services, Department of Commerce, Washington 25, D. C. (unpublished).

TABLE I. Pertinent characteristics of the isotopes in the scattering samples. As mentioned in the text, the assignment of the observed magnesium line to Mg^{24} is somewhat weak. If it is associated with Mg^{26} instead, the natural width would be 4.48 Mev.

Element	Isotope	Percent abundance	(γ,n) Threshold (Mev)	Observed threshold for γ ray (Mev)	Tentative spin assignment	Ground state spin[a]	Cross section at resonance σ_0 (Barns)	Debye temp. θ (°K)[b]	Doppler width Δ (ev)	Natural width Γ (ev)
Magnesium	24	78.8	16.53	10.15±0.06	1	0^+	72.0	406	15.89	$4.80\begin{cases}+1.56\\-1.43\end{cases}$
	25	10.1	7.33			$\frac{5}{2}^+$				
	26	11.1	11.11			0^+				
Silicon	28	92.17	17.17	11.40±0.06	1	0^+	56.9	658	17.63	$2.89\begin{cases}+0.99\\-0.78\end{cases}$
	29	4.71	8.47			$\frac{1}{2}^+$				
	30	3.12	10.60			0^+				

[a] P. M. Endt and C. M. Braams, Revs. Modern Phys. **29**, 683 (1957).
[b] Charles Kittel, *Introduction to Solid State Physics* (John Wiley & Sons, Inc., New York, 1953), 2nd ed.

The treatment used in obtaining (6) is valid only for a gas, but according to Lamb[8] we can consider the solid as a gas if we use an effective temperature T_e that is higher than the actual temperature T provided $\Delta+\Gamma\gg 2k\theta$ which is true in both cases. The relationship between T and T_e is given in reference (8).

For analysis of the experimental data, Eq. (3) was integrated numerically for several values of Γ/Δ. Plotted in Fig. 5 are the results of these integrations from which a value of Γ/Δ has been associated with an observed transmission. This and the calculated Doppler width Δ were used to determine Γ. Shown in Table I are the known, calculated and estimated characteristics of the isotopes in the scattering samples.

V. DISCUSSION

The isotopic assignment of the observed levels is based principally upon the assumptions that, if the energy of the incoming photon is above the threshold for particle emission, then particle emission is preferred over gamma-ray emission as a mode of decay and that the isotope must be relatively abundant or levels associated with it would not have been detected. It should be pointed out that in calculating the value of the cross section at resonance using (9) it has been assumed that there is no branching and decay is 100% to the ground state by way of gamma-ray emission.

As noted previously, levels in several other elements have been excited using the technique described here.[2,3] For those instances where the spins of both the ground state and the excited level were firmly established, all were excited by magnetic dipole ($M1$) interactions. Another piece of evidence in favor of $M1$ interactions is the unsuccessful attempt to excite the 4.43-Mev level in C^{12} (which requires $E2$ radiation) in spite of the the fact that it has a larger peak cross section (as given by 9) than the 4.46-Mev level in B^{11} which was observed. For these reasons tentative values of spin have been assigned to the levels observed in magnesium and silicon assuming they were excited by $M1$ radiation.

The gamma rays observed in this experiment are presumably the same as the "10.5 Mev" and "11.2 Mev" gamma rays from magnesium and silicon, respectively, and recently observed by Seward *et al.*,[3] in an experiment similar to that described here.

The 10.15-Mev gamma ray observed in this experiment may be the same as the 10.08-Mev gamma ray Compion and Bartholomew[9] attribute to Mg^{26}. The decision to associate the line with Mg^{24} in the work reported here was determined largely by the relative abundance of the two isotopes. It must remain until separated isotopes become available in larger quantities or the sensitivities of experimental techniques can be improved upon before we can positively identify the isotope responsible for the gamma ray from magnesium observed in this experiment.

The isotopic assignment of the silicon line was justified by the facts that Si^{28} is preponderantly abundant and is the only silicon isotope having a threshold for particle emission that is higher than the observed line. For these reasons it is felt that the assignment is a rather firm one.

Endt and Braams[10] list in their compilation, levels at 10.3 Mev and 11.8 Mev in Si^{28}, both of which are far enough removed not to be confused with the level observed at 11.4 Mev in the work reported here.

VI. ACKNOWLEDGMENT

The author is deeply indebted to Dr. Leslie Cohen of this Laboratory for many fruitful discussions regarding the method of analysis and conclusions drawn from the data.

[8] W. E. Lamb, Phys. Rev. **55**, 190 (1939).
[9] P. J. Campion and G. A. Bartholomew, Can. J. Phys. **35**, 1361 (1957).
[10] P. M. Endt and C. M. Braams, Revs. Modern Phys. **9**, 4, 683 (1957).

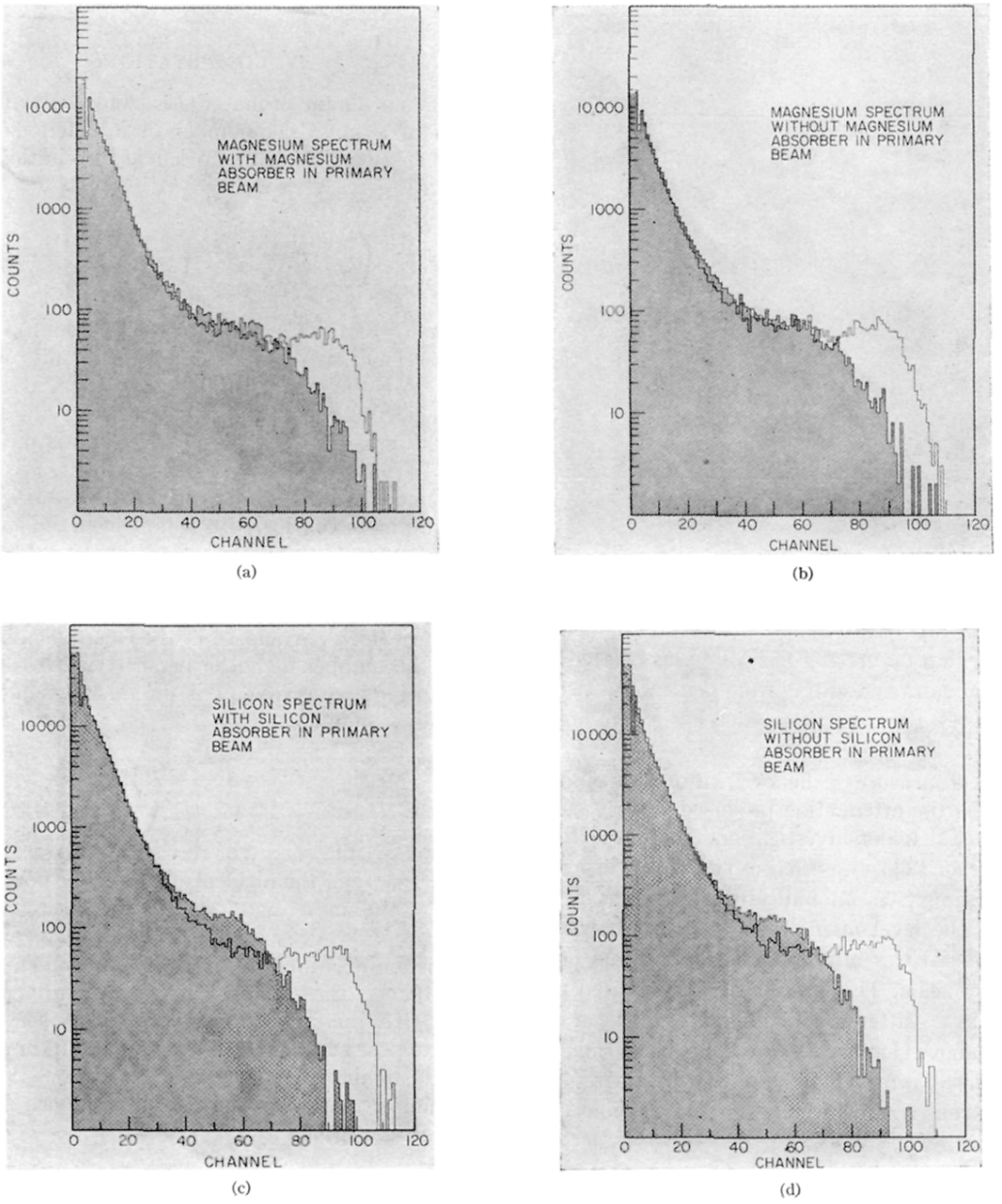


FIG. 3. Shown above are the spectra obtained from magnesium and silicon for irradiations made with and without an absorber of similar material in the primary x-ray beam. The shaded spectra were obtained using the equivalent number of grams/cm^2 of aluminum as a scatterer for background determination. The energy scale for all these spectra is such that the peaks for the 0.51-Mev annihilation radiation and the 4.43-Mev gamma ray from the decay of C^{12} (observed using a Po-Be neutron source) occur in channels 5 and 50, respectively.