

RECOILLESS FRACTION OF GAMMAS
IN TIN DIOXIDE (SnO_2)

Leonidas O. Barbopoulos

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ABSTRACT

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by

Leonidas Odysseus Barbopoulos

The Mossbauer recoilless fraction of 23.8 KeV gammas in SnO_2 is investigated as function of temperature ranging between 78°K and 297°K , with the Sn^{119} source being kept at room temperature. The absorbed fraction, f_a , varies between 0.9 at 78°K and 0.59 at 297°K . f_a is determined by fitting Mossbauer spectrum with the absorption integral and from the line shape of the spectrum. It is found that the area method gives unsatisfactory results because of unresolved quadrupole splitting of the resonantly absorbed line. Also, a study of variation on Debye temperature of SnO_2 with temperature is made, which is found to be in good agreement with other authors.

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ABSTRACT

The Mössbauer recoilless fraction of 23.8 KeV gammas in SnO_2 is investigated as function of temperature ranging between 78°K and 297°K , with the Sn^{119} source being kept at room temperature. The absorbed fraction, f_a , varies between 0.9 at 78°K and 0.59 at 297°K . f_a is determined by fitting Mössbauer spectrum with the absorption integral and from the line shape of the spectrum. It is found that the area method gives unsatisfactory results because of unresolved quadrupole splitting of the resonantly absorbed line. Also, a study of variation in Debye temperature of SnO_2 with temperature is made, which is found to be in good agreement with other authors.

INTRODUCTION

After the discovery of recoilless resonant absorption in atomic nuclei in 1958 by R.L. Mössbauer, it was soon recognized as a powerful tool in the investigation of a wide range of phenomena. One important aspect of such investigations is the measurement of the Mössbauer fraction, f , of the emitted or the absorbed gamma radiation intensity which is simply related to the localization of the atom in its host material. Like many other measurements of radiation intensities, is difficult to measure with precision. The measurement requires that one compare the intensities in a very narrow energy interval with the total intensity, in a much more diffused part of a characteristic gamma ray.

The temperature dependence of recoilless fraction using Sn^{119} has been the subject of many recent attempts. The objective of these studies is to elucidate the nature of the mean square vibrational amplitude of the nucleus in a source and absorber.

Alikhanov and Lyubimov⁽¹²⁾ first observed the resonance absorption effect in Sn^{119} at liquid nitrogen and at room temperature. Delyagin, Shpinel et al⁽¹³⁾ measured the recoilless absorption fraction by Sn^{119} nuclei in different crystals for different absorber thicknesses, at liquid nitrogen temperature. Shirley and Kaplan⁽¹⁴⁾ discussed theoretically three methods of measuring the recoilless fraction and applied them successfully to Au^{197} . The purpose of our work is to apply these methods to Sn^{119} in order to compare their relative merits in the accurate determination of f by measuring at different temperatures and different absorber thicknesses.

It was found that the line width method gives good results for SnO_2 . The method of line width dependence on apparent thickness which is only limited to very thin absorbers, did not give satisfactory results with our enriched thick absorbers. Because of unresolved quadrupole splitting of the absorption line, the area method also gave very unsatisfactory results. The measurements in this work suggest that the most suitable method for determination of f in SnO_2 is the line width dependence on absorber thickness.

This work also includes the determination of the Debye temperature of SnO_2 crystal and its dependence on temperature. This investigation supports the general belief that there is a considerable variation in the Debye temperature of the crystal with temperature. Few experimental data are available for quantitative comparison with our measurements.

Chapters I and II of this work contain the general theory of the Mössbauer effect. Chapter III gives a brief description of the apparatus used. Chapter IV deals with the analysis of the experimental data and the various methods of calculating the recoilless fraction. The later part of this chapter deals with the discussion of results. The theoretical derivation for recoilless fraction is given in the appendix.

CHAPTER 1MOSSBAUER SPECTRUM1.1 Recoilless emission.

The phenomenon of photon emission is a well known quantum effect in which a system emits a photon in a transition from one state to another state of lower energy. A reverse transition process can also take place where a photon is absorbed and transition to a higher energy state occurs. This process is called resonant absorption because the photon must have just the right energy to cause the transition. In optical resonance phenomena the transition takes place between electronic levels, while in case of nuclear resonance, nuclear levels are involved. To meet the condition of resonance, sources and absorbers must be made of identical atoms or nuclei.

The transition energy, E_0 , is the energy difference between two nuclear levels. However, the energy of the emitted photon usually differs slightly from the transition energy E_0 , because of the energy loss due to recoil of the nucleus. To understand this, let us consider a source to be at rest initially; then, as the system undergoes a transition from state $m \rightarrow n$, the transition energy is

$$E_{\gamma} = E_n - E_m \quad (1)$$

and the momentum of emitted photon of energy E_{γ} is given by

$\frac{E_{\gamma}}{c}$, c being the velocity of light.

From the principle of conservation of energy and linear momentum one gets

$$E_o = E_\gamma + \frac{1}{2}MV^2 \quad (2)$$

$$MV = \frac{E_\gamma}{c}$$

where V is the recoil velocity of the atom of mass M . Elimination of V from equations in (2) yields

$$E_\gamma = E_o - \frac{E_\gamma^2}{2Mc^2} \quad (3)$$

Since $E_\gamma^2/2Mc^2$ is small compared to E_o , the photon takes away most of the energy of the transition, and therefore putting $E_\gamma \approx E_o$ in the second term on the right hand side of equation (3) gives

$$E_\gamma = E_o - \frac{E_o^2}{2Mc^2} \quad (4)$$

$E_o^2/2Mc^2$ is the energy of recoil, E_R . Therefore the energy required for the gamma ray to be resonantly absorbed by an identical nucleus is $E_o + E_o^2/2Mc^2$, and the energy difference between the emitted and the absorbed photons in an identical nucleus is twice the recoil energy, that is

$$\Delta E \approx \frac{E_o^2}{Mc^2} = 2E_R \quad (5)$$

The absorption of the emitted gamma ray by an identical nucleus will depend on the recoil energy, E_R and the line width, W , of the gamma-ray at half maximum (Figure 1).

From Figure 1 (a), it is clear that absorption is possible if $W > 2E_R$, and that the probability of recoilless emission is given by the shaded area. Figure 1 (b) shows that resonant absorption is not possible if the line width is less than twice the energy loss due to recoil.

1.2 Doppler Broadening

In the previous section 1.1, the emitting and the absorbing systems are assumed to be at rest. In fact, the source and absorber atoms are in thermal motion; which introduces additional widening of the emission and absorption lines, called Doppler broadening.

To see this effect quantitatively, consider a source atom with initial momentum \vec{P}_i emitting a photon of momentum \vec{p} . The momentum of the source after emission is, $\vec{P}_i - \vec{p}$. The energy E_R , gained by the source and hence lost by the gamma ray is in good approximation given by

$$E_R = \frac{(\vec{P}_i - \vec{p})^2}{2M} - \frac{P_i^2}{2M} = \frac{p^2}{2M} - \frac{\vec{p} \cdot \vec{P}_i}{2M} \quad (6)$$

In the above, it is assumed that the mass of the quantum system does not change during emission. The first term on the right hand side in equation (6) represents the recoil energy E_R of the initially

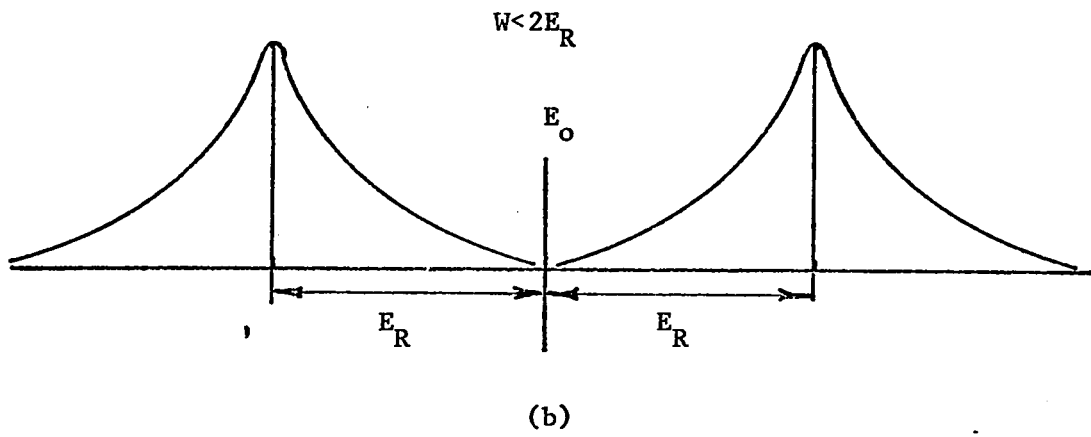
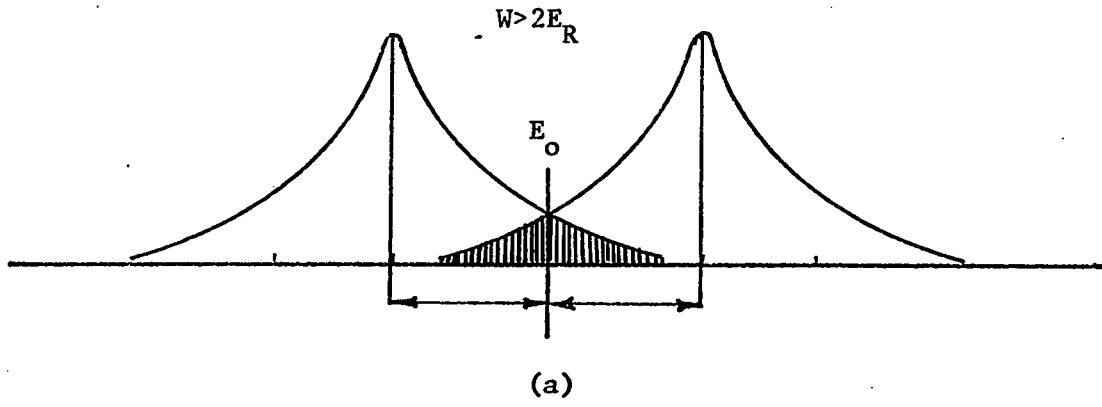


Figure 1

stationary system; and the second term represents the increase in the line width due to thermal motion of the atom.

Maximum absorption of incident photons by an identical nucleus will take place when the energy loss due to the recoil of emitting nucleus is zero. This may be achieved when the decaying nucleus is bound in a matrix in such a way that the recoil momentum is absorbed by the whole lattice, rather than by a single atom. The recoil energy and Doppler broadening for such γ -rays are then characterized by the mass and random velocity of the whole crystal and are therefore completely negligible.

The resonance width D at half maximum as a function of temperature, and recoil energy is given by the following expression (17)

$$D = \frac{1}{c} \sqrt{\frac{2kT}{M}} E_R \quad (7)$$

It is clear from this expression that D is small due to the large mass of the crystal lattice. Thus the Doppler broadening term drops out at the same time as the recoil energy E_R .

When the gamma ray passes through a material containing identical nuclei, resonant absorption will take place provided the recoil momentum is absorbed by the lattice. However, if the environments of the emitting and the absorbing nuclei are different, the nuclear transition energy, E_0 , may differ slightly. This energy difference is known as the isomer shift.

To observe the Mossbauer effect we look for a condition that will destroy resonance absorption. Mossbauer used Doppler's Principle to achieve this condition. Due to the narrow line-width of the resonance it is sufficient to give a small Doppler velocity to the source (or absorber). The source is usually moved with velocity V relative to the absorber. The gamma ray then suffers an energy shift, ΔE given by

$$\Delta E = \frac{VE_R}{c} \quad (8)$$

The velocity V is defined as positive if the source moves towards the absorber. The gamma intensity, I_{exp} , in the detector is determined as a function of V . At large velocities no resonance absorption takes place, whereas maximum absorption occurs at zero velocity.

1.3 Cross Section for Recoilless Absorption

The expression for the cross section of recoilless nuclear resonant absorption is given by J.D. Jackson (1) and W. Heitler (2). It takes the form

$$\sigma_{RA}(E) = \frac{\sigma_o f_a}{1 + \left(\frac{E - E_{oA}}{\gamma_A} \right)^2} \quad (9)$$

$\sigma_{RA}(E)$ is the cross section of recoilless resonant absorption of quanta of energy E by a nucleus in the ground state (the nucleus usually is bound in a crystal lattice). σ_o is the maximum resonance

cross section. E_{OA} is the transition energy between the nuclear ground state and the excited state, γ_A is the half width at half maximum of the excited state ($\gamma_A = \Gamma/2$). The subscript A stands for absorption. The recoilless fraction, f_a , which is a measure of the probability of recoilless absorption, is of the form

$$f_a = \exp \left[- \frac{E_{oa}^2}{2M_a c^2} \cdot \frac{1}{kT_o} \cdot Q(T, T_o) \right] \quad (10)$$

where M_a is the mass of the absorbing nucleus, c is the speed of light, and $Q(T, T_o)$ is an increasing function of the absorber temperature T ; k is the Boltzmann constant. T_o is the characteristic temperature of the crystal in which the absorbing nucleus is bound. $Q(T, T_o)$ increases with temperature, at low temperature ($T \ll T_o$) Q varies slowly with temperature, but rises rapidly with high temperatures ($T > T_o$).

CHAPTER 2

2.1 Recoilless Fraction

The recoilless fraction f is a measure of the probability of recoilless absorption or emission of gammas and is expressed in the form (Appendix A)

$$f = e^{-\langle x^2 \rangle / \lambda^2} \quad (11)$$

where $\langle x^2 \rangle$ is the mean square amplitude of the vibration in the direction of emission of the gamma ray, averaged over an interval equal to the lifetime of the nuclear level involved in the gamma-ray emission process. If $\langle x^2 \rangle$ is not bounded, the recoil-free fraction will vanish, i.e., it will not be possible to observe the Mossbauer effect in liquids where the molecular motion is not restricted. This has been demonstrated by A.J.F. Boyle et al ⁽³⁾ in experiments where the study of recoil-free fraction in Sn¹¹⁹ both below and above the melting point, failed to show any resonant effect.

In the derivation of equation (11) a harmonic motion of the atoms is assumed, which appears to be a good approximation except at temperatures above the Debye temperature. The recoilless fraction can also be expressed in terms of the photon frequency distribution.

A solid containing N atoms is assumed to be a system of $3N$ harmonic oscillators. There are therefore $3N$ harmonic modes of vibration of frequencies ω_j . The range of frequencies is from

zero to a maximum frequency ω_m . The average energy associated with each oscillator is

$$(\bar{n}_j + \frac{1}{2}) \hbar \omega_j \quad (12)$$

where \bar{n}_j is given by the Plank's distribution function

$$\bar{n}_j = \frac{1}{\exp(\hbar \omega_j / kT) - 1} \quad (13)$$

The energy of the crystal attributed to the j th oscillator is

$$NM \omega_j^2 \langle r_j^2 \rangle = (\bar{n}_j + \frac{1}{2}) \hbar \omega_j \quad (14)$$

where r_j is the displacement of the atom due to the j th oscillator.

Dividing by $NM \omega_j^2$ and summing over j , yields for the average displacement.

$$\langle r^2 \rangle = \frac{\hbar}{NM} \sum_j \frac{(\bar{n}_j + \frac{1}{2})}{\omega_j} \quad (15)$$

To evaluate this expression we replace the summation with an integral and introduce the density of vibrational states $g(\omega)$.

This gives

$$\langle x^2 \rangle = \frac{\hbar}{3MN} \int_0^{\omega_m} \frac{g(\omega)}{\omega} \left(\frac{1}{2} + \frac{1}{e^{\hbar \omega / kT} - 1} \right) d\omega \quad (16)$$

For a Debye solid the density of states of the vibrational spectrum is given by

$$g(\omega) = \frac{9N\omega^2}{\omega_m^3} \quad (17)$$

which is normalized so that

$$\int_0^{\omega_m} g(\omega) d\omega = 3N \quad (18)$$

Using $E_\gamma = pc$, $\lambda = \frac{hc}{E_\gamma}$ in equation (11) we get

$$f = \exp\left(-\frac{2E_R}{3N\hbar r}\right) \int_0^{\omega_m} \frac{g(\omega)}{\omega} \left(\frac{1}{2} + \frac{1}{\exp\left(\frac{\hbar\omega}{kT}\right) - 1}\right) d\omega \quad (19)$$

Two properties of the frequency distribution can be determined from the recoilless fraction. They are the mean inverse frequency, $\langle \omega^{-1} \rangle$ and the mean inverse square frequency $\langle \omega^{-2} \rangle$. The first is obtained from the zero degree limit (0°K) in which equation (19) becomes

$$f = \exp\left(-\frac{E_R}{3N\hbar r}\right) \langle \omega^{-1} \rangle \quad (20)$$

The second property resulting from equation (19) in the high temperature limit is

$$f = \exp\left(-\frac{E_R kT}{3N\hbar r^2}\right) \langle \omega^{-2} \rangle, \quad (T > \frac{\hbar\omega_m}{k}) \quad (21)$$

$\langle \omega^{-1} \rangle$ and $\langle \omega^{-2} \rangle$ can be determined experimentally from the intercept and shape of the experimental plot of mean square displacement versus temperature. If one assumes a Debye frequency distribution

$$\begin{aligned} g(\omega) &= \alpha \omega^2 & (\omega < \omega_m) \\ g(\omega) &= 0 & (\omega > \omega_m) \end{aligned} \quad (22)$$

where

$$3N = \alpha \int_0^{\omega_m} \omega^2 d\omega \quad (23)$$

one gets

$$f = \exp\left(-\frac{6E_R}{k\theta}\right) \cdot \left(\frac{1}{4} + \frac{\hbar^2}{k^2\theta} \cdot \int_0^{\omega_m} \frac{\omega d\omega}{e^{\hbar\omega/kT} - 1}\right) \quad (24)$$

The Debye temperature is defined as $\theta = \hbar\omega_m/k$. The following conclusions can be drawn from the above equations:

- (a) The recoilless fraction increases as the temperature decreases; this is what Mossbauer observed with Ir^{119} .
- (b) The higher the Debye temperature θ of the solid, the larger will be the Mossbauer recoilless fraction.
- (c) The Mossbauer effect is limited to relatively low-energy gamma radiation, since in equation (24), f decreases rapidly with large E . The effect has been observed up to 155 keV gamma radiation.

2.2 Mean Square Displacement

The mean square displacement can be calculated by using experimental values of the recoilless fraction in the relationship

$$\langle x^2 \rangle = - \lambda^2 \ln.f \quad (25)$$

Classically, the mean square displacement is linear with temperature. The slope of $\langle x^2 \rangle$ in the classical limit and the zero-degree intercept gives two bits of information related to the frequency distribution. These are $\langle \omega^{-1} \rangle$ and $\langle \omega^{-2} \rangle$ which are related to the experimental intercept, $\langle x^2 \rangle_0$, and slope of $\langle x^2 \rangle$ versus T plot as follows:

$$\langle \omega^{-1} \rangle = \frac{h\nu}{\lambda^2 E_R} \cdot \langle x^2 \rangle_0 \quad (26)$$

$$\langle \omega^{-2} \rangle = \frac{h\nu^2}{k\lambda^2 E_R} \quad (\text{slope})$$

2.3 Mossbauer Spectral Lineshape

The process of gamma emission from a source is either recoilless or with recoil. In the first case the gamma can be absorbed resonantly or electronically. In the second case it may only be absorbed electronically. The count rate per unit energy $I_{RS}(E)$ of recoilless radiation of energy E emitted from the source is given by

$$I_{RS}(E) = \frac{f_s I_{os}}{1 + \left(\frac{E - E_{os}}{\gamma_s}\right)^2} \quad (27)$$

where f_s is the source recoilless fraction and I_{os} is the peak value of counts per unit energy of both the recoilless and nonrecoilless radiation emitted by the source, and does not include extraneous background. E_{os} is the energy of transition from the excited state to the ground state in the source, and $\gamma_s = \Gamma_s/2$ is the half line width at half maximum of the Mossbauer level.

The fraction of gamma rays of energy E which will be resonantly absorbed by the absorber is

$$O_{RA}(E) = 1 - \exp[-n_A \sigma_{RA}(E)] \quad (28)$$

where

$$\sigma_{RA}(E) = \frac{f_a \sigma_o}{1 + \left(\frac{E - E_{oA}}{\gamma_A}\right)^2}$$

and n_A is the number of absorbing nuclei per unit area in the absorber.

In actual experiment the gamma rays emitted by the source are given an additional energy, e , by the Doppler shift mechanism, so that if the source emits photons of energy E the absorber in fact receives photons of energy $E + e$. Therefore the counts per unit energy of gamma emitted by the source and recoillessly absorbed by the absorber are

$$I_{RA}(E, e) = I_{RS}(E) O_{RA}(E + e) \quad (29)$$

and the intensity of absorption observed at this Doppler velocity of the source is

$$\begin{aligned}
 I_{RA}(e) &= \int_0^{\infty} I_{RA}(E, e) dE \\
 &= \int_0^{\infty} I_{RS}(E) Q_{RA}(E + e) dE \\
 \text{or } I_{RA}(e) &= \int_0^{\infty} \left[\frac{f_s l_{os}}{1 + \left(\frac{E - E_{os}}{\gamma_s}\right)^2} \right] \cdot \left[1 - \exp - \frac{f_a n \sigma_a o}{1 + \left(\frac{E + e - E_{oA}}{\gamma_A}\right)^2} \right] \cdot dE \quad (30)
 \end{aligned}$$

This equation is known as the Mossbauer spectral line shape.

2.4 Determination of the Recoilless Fraction in the Absorber by Line area Method.

The area A of a line in Mossbauer spectrum is obtained by integrating the absorption rate $I_{RA}(e)$ in equation (30) over all Doppler velocities,

$$A = \int_{-\infty}^{\infty} I_{RA}(e) de = \int_{-\infty}^{\infty} f_s \pi \gamma_s l_{os} \left(1 - \exp \left(- \frac{\tau_A}{1 + \left(\frac{E}{\gamma}\right)^2} \right) \right) \cdot dE$$

$$A = f_s \pi \gamma_s l_{os} \gamma_A \int_{-\infty}^{\infty} \left(1 - \exp \left(- \frac{t_A}{1+x^2} \right) \right) \cdot dx \quad (31)$$

where $t_A = f_a n_A \sigma_o$

Ignoring the extraneous background, the transmitted intensity is composed of two parts: (a) the fraction $(1-f_s)$ of radiation from the source which is not recoillessly emitted; (b) the recoilless radiation from the source which is not resonantly absorbed in the absorber. Then the transmitted intensity $I_T(e)$ is given by

$$I_T(e) = \int_0^{\infty} \frac{(1-f_s) l_{os}}{1 + \left(\frac{E-E_{os}}{\gamma_s} \right)^2} dE +$$

$$+ \int_0^{\infty} \frac{f_s l_{os}}{1 + \left(\frac{E-E_{os}}{\gamma_s} \right)^2} \cdot \exp \left(- \frac{t_A}{1 + \left(\frac{E+e-E_{os}}{\gamma_A} \right)^2} \right) dE \quad (32)$$

The off-resonance intensity is given by

$$I_T(\infty) = \int_0^{\infty} \frac{l_{os} dE}{1 + \left(\frac{E-E_{os}}{\gamma_s} \right)^2} = \pi \gamma_s l_{os} \quad (33)$$

The area then becomes

$$A = f_s I_T(\infty) \gamma_A \int_{-\infty}^{\infty} \left[1 - e^{-\left(\frac{t_A}{1+x^2} \right)} \right] dx \quad (34)$$

This has been evaluated by G.A. Bykon and Pham Zuy Hien⁽³⁾ in the form

$$\int_{-\infty}^{\infty} \left[1 - \exp \left(-\frac{t}{1+x^2} \right) \right] dx = \pi t e^{-t/2} [I_0(t/2) + I_1(t/2)] \quad (35)$$

where I_0 and I_1 are the Bessel functions of imaginary argument.

Thus

$$A = \pi f_s I_T(\infty) \gamma_A t_A e^{-t/2} [I_0(t_A/2) + I_1(t_A/2)] \quad (36)$$

The actual count rate B, of resonance (baseline), is

$$B = I_T(\infty) + N \quad (37)$$

where N is the count rate from extraneous noise.

CHAPTER 3

3.1 Apparatus

In general the apparatus consists of; (1) transducer which provides relative velocity between the source and absorber, (2) a gamma-ray detector and associated electronic equipment for collecting, sorting, and presenting the data, (3) cryostat and a temperature control device.

Doppler Shifting System

The energy of the gamma rays recoillessly emitted from the source is varied by Doppler shifting them; that is, by superimposing on the source a periodic velocity. If gamma rays are recoillessly emitted from the source with energy E_0 and if at any time the source velocity is V with respect to an absorber, then the energy of the incident gamma rays seen by the absorber is $E_0(1+V/c)$, where positive V denotes velocity of the source towards the absorber. Thus we have in effect a source of gamma rays, whose energy is swept periodically in time, and which is used in conjunction with a time-synchronized sequence of counters (a multichannel analyser) to investigate the resonance absorption spectrum of similar nuclei present in the absorber (Figure 2).

Transducer

The transducer is an electromechanical unit which provides the relative motion between the source and absorber, used to observe Mossbauer effect spectra. The transducer moves the source and at the same time produces a signal proportional to the velocity.

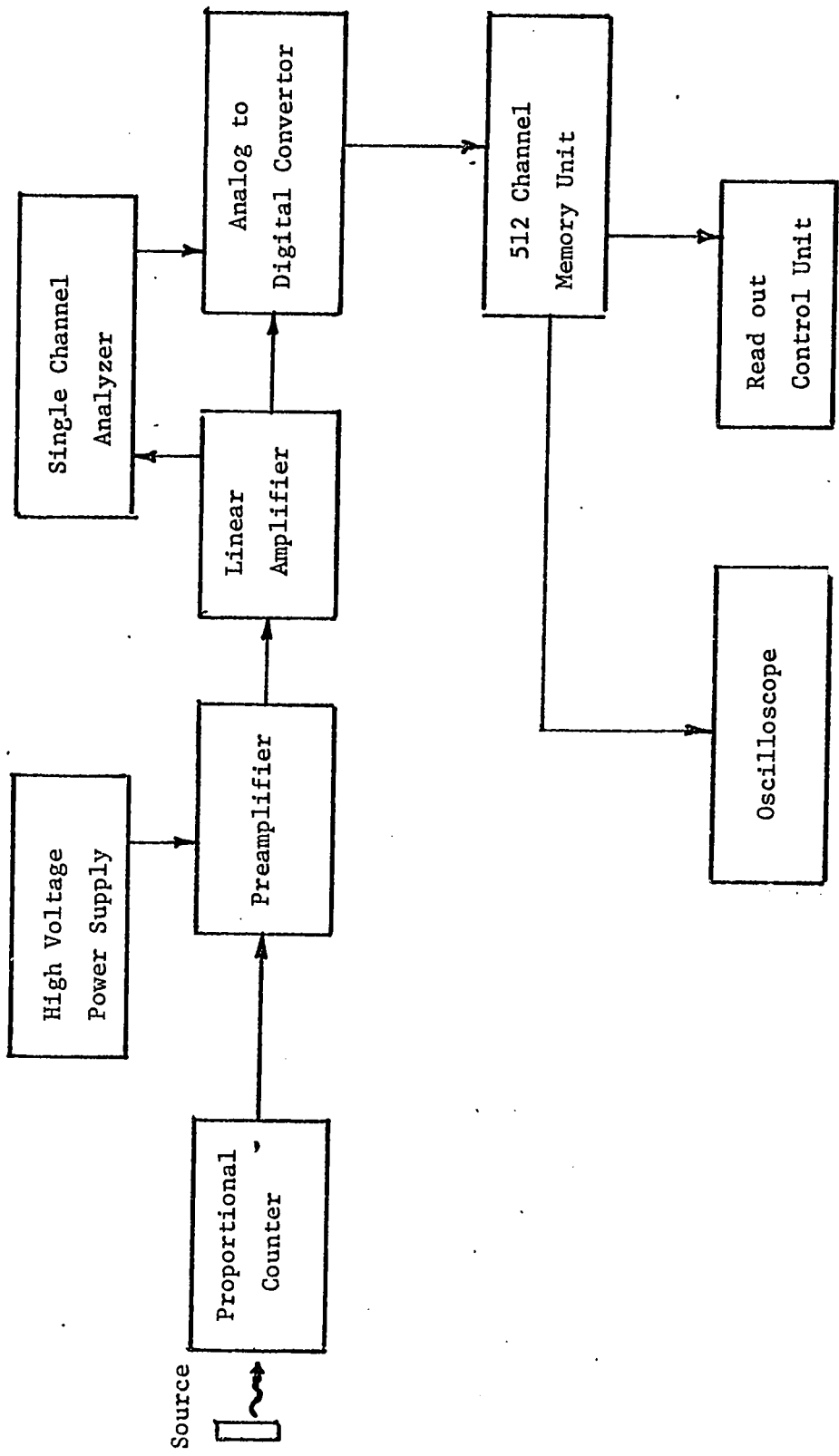


Figure 2. Block diagram for pulse-height measurement and window adjustment (S.C.A.)

The Control Unit

The control unit is a solid state electronic unit which accepts a square wave signal from the address register of the multichannel analyser (512 channels). It consists of, shaper and integrator, difference amplifier and d.c. power amplifier to drive the transducer. The velocity control unit is used to produce a velocity varying linearly with time in a parabolic motion with constant acceleration. The displacement, velocity and acceleration of the parabolic motion as a function of time are illustrated in Figures 3 and 4.

3.2. Proportional Counter

The choice of the detector is one of the main factors in determining the Mossbauer effect spectrum. The proportional counter is chosen because of its great efficiency and high resolution in detection of low-energy gamma rays. In our experiment it is required to identify the 23.8 KeV gamma rays.

The proportional counter operated at 2000 V, was (N.S.E.C. type) filled with a mixture of Xe-CH₄ gases. The equipment used for amplification and pulse-height measurement is shown in Figure 2.

The resolution of the counter for the 23.8 kev energy peak is given by the expression

$$\begin{aligned} \text{Resolution} &= \frac{\Delta E}{E} \times 100 \\ &= \frac{15.1}{122.71} \times 100 \\ &= 12.5\% \end{aligned}$$

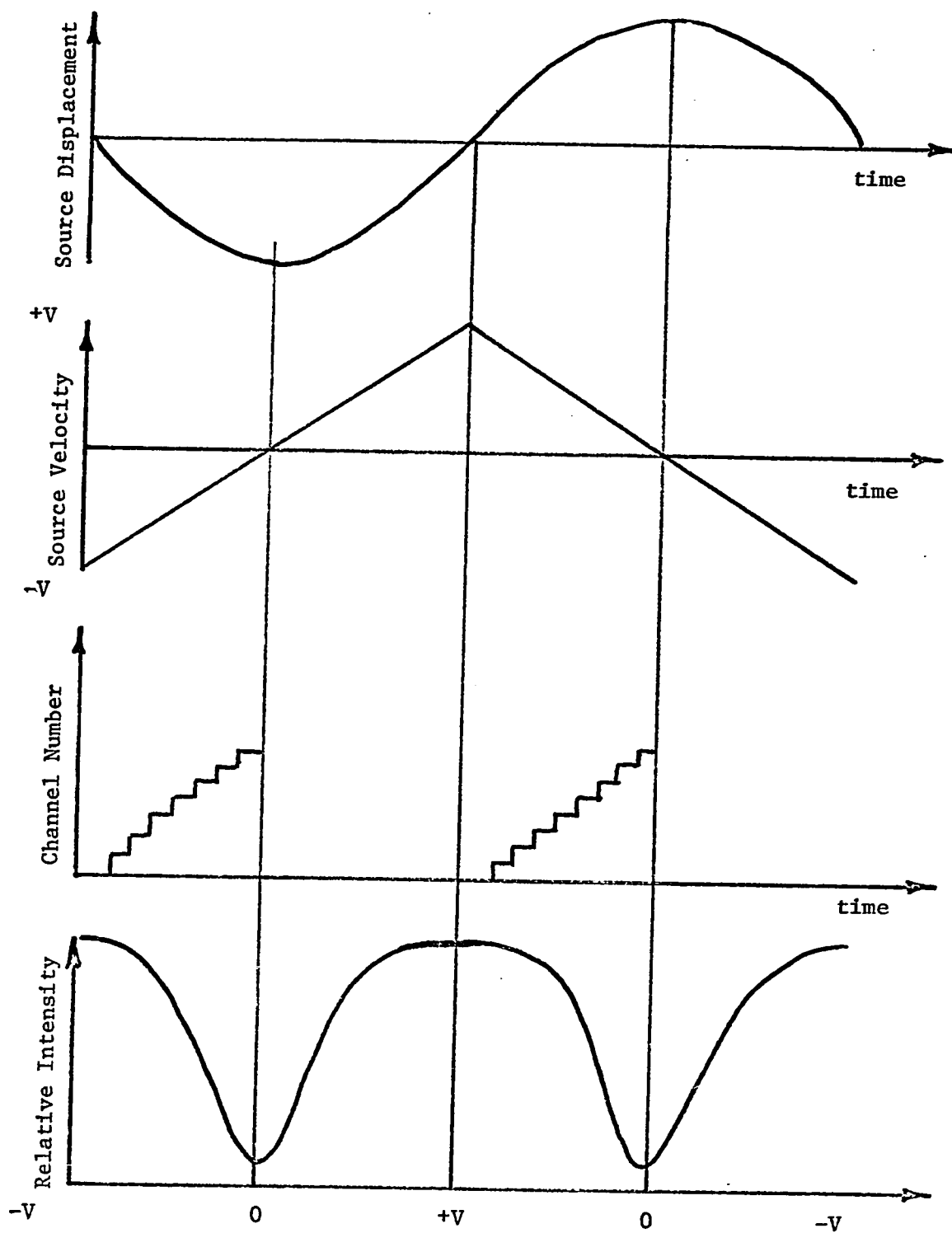


Figure 3. Recording of a Mössbauer Spectrum.

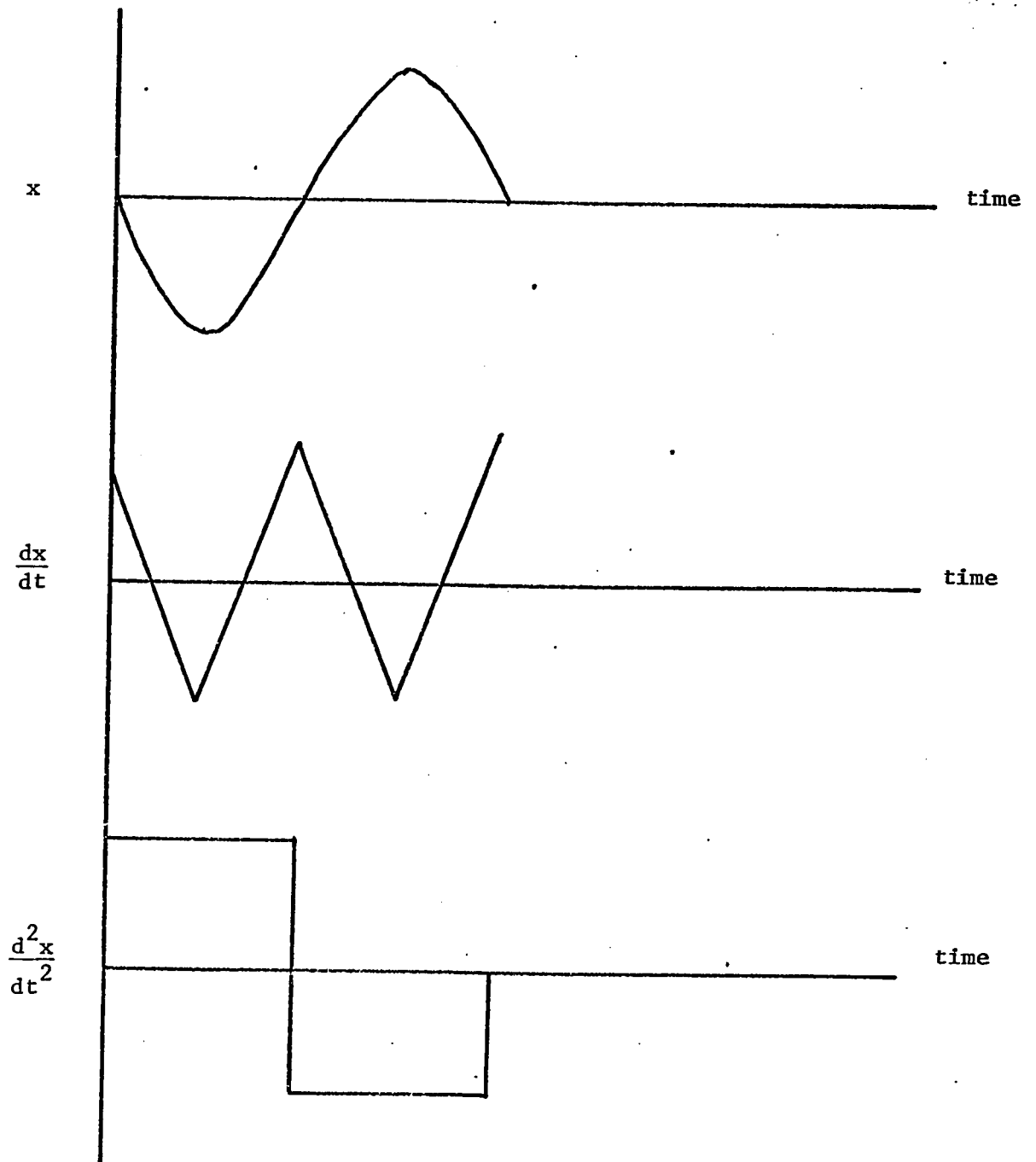


Figure 4. The displacement, velocity, and acceleration as a function of time.

where ΔE is the half width at half maximum of the peak in channels and E is the channel number of the position of the 23.8 keV Mossbauer peak.

3.3 Temperature

The absorber temperature was varied over a range of 77° K to 300° K. The source was held approximately at room temperature (298° K). The width of the absorption peak is independent of the source temperature. However, the depth of the peak is greater for lower source temperature, so it is advantageous to maintain the source at constant temperature.

A schematic diagram of the apparatus used to cool the absorber is shown in Figure 5.

3.4 Source and Absorber

The source used in this experiment was prepared by the New England Nuclear Corporation and consisted of 10 mc of Sn¹¹⁹ in Barium stannate. The active area is one inch in diameter.

The source exhibited reasonably small resonant absorption, as seen in the following. The apparent thickness due to self absorption in the source is

$$T_s = f_s \sigma_o n$$

where f_s is the source recoilless fraction, σ_o is the maximum nuclear resonance absorption cross section, and n is the number of resonantly absorbing nuclei of Sn¹¹⁹ per unit area in the source.

The number of Sn¹¹⁹ atoms in the source at any time is

$$n(\text{Sn}) = \alpha_o (1 - e^{-t/\tau})$$

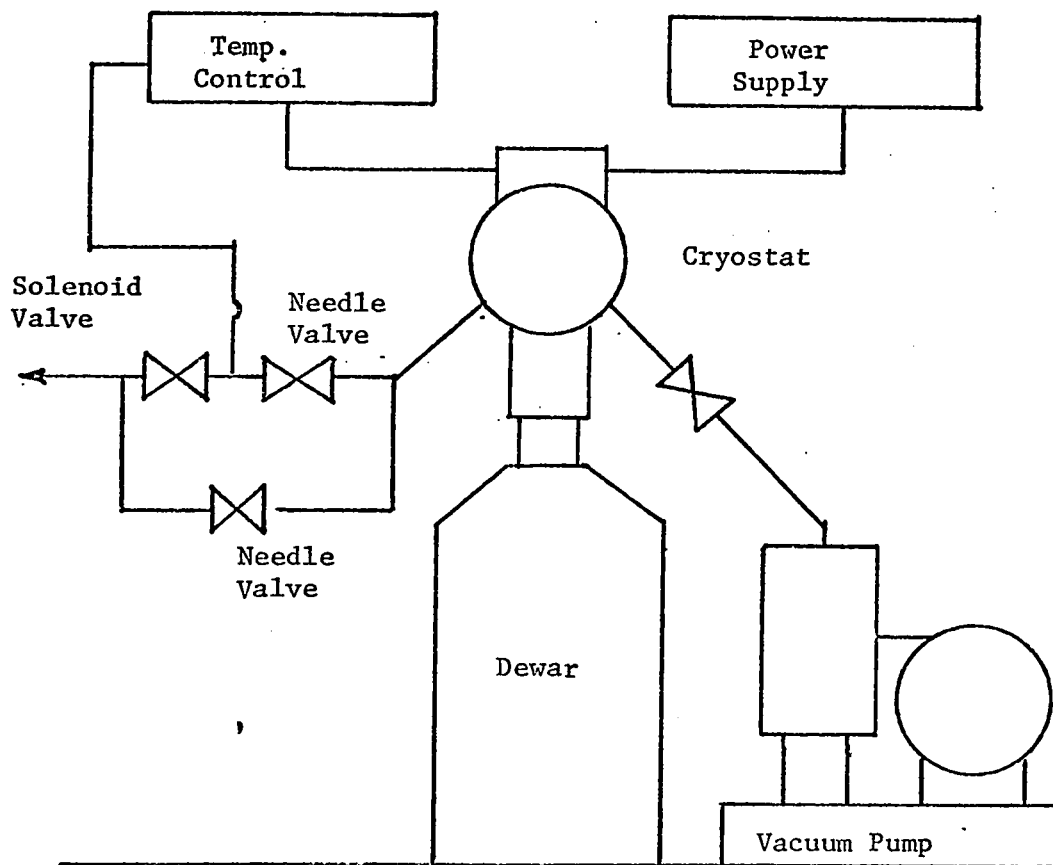


Figure 5. Schematic flow diagram of the heating apparatus.

Where α_0 and τ are the initial activity and the half life of Sn^{119} respectively. Initially 10 mc of fresh Sn^{119} was diffused into the source and it was about one half-life (245 days) old when the Mossbauer experiments were performed. One millicurie activity means 3.7×10^7 disintegrations per second. Therefore, $n(\text{Sn}) = 0.566 \times 10^{16}$. For the 23.875 kev level of Sn^{119} in Barium stannate $f_s = 0.26$ and $\sigma_0 = 1.32 \times 10^{-18} \text{ cm}^2$. The active area is 5.02 cm^2 .

These numbers yield $T_s = 0.038 \times 10^2$ resonant absorption lengths in the source. Margulies ⁽⁴⁾ indicates that for $T_s \ll 1$ the source is considered to be thin, and effects only the off-resonance part of the spectrum. The magnitude of this effect is not larger than the estimated errors in the values of f_a and f_s determined in the present experiment (the calculation requires knowledge of the background corrected off-resonance count rate).

3.5 Absorber

The absorbers used were SnO_2 of 2.54 cm diameter and of 0.15 cm thickness supplied by the New England Nuclear Corporation. The mass per unit area for the six absorbers used was 0.5, 1.0, 1.5, 2.5, 7.5, and 10.0 mg/cm^2 respectively.

CHAPTER 4

4.1 Methods of Analysis of Data

In order to determine accurately the widths of Mossbauer absorption peaks it is necessary to fit numerically the absorption line shape to the experimental data. Assuming a Lorentzian shape, the function used in this experiment is

$$I_T(e) = B - \frac{I_m}{1 + \left(\frac{E_o - e}{W/2}\right)^2} \quad (39)$$

where

$$E_o = E_{oA} - E_{os}$$

E_{os} and E_{oA} are the energies of transition from the excited state in the source and absorber respectively, B is the number of counts at an off-resonance point, I_m and W are the depth and full width half maximum (FWHM) of the spectral absorption dip respectively.

A least square fit program is used to fit the experimental data to equation (39). There are four parameters to be determined, namely (1) the base line (the counting rate at infinite velocity), (2) the height of the peak in counts per channel, (3) the position of the peak and (4) the width of the peak in channels (Figure 6 and 13).

The analysis of the problem is as follows: Experimentally there are Z_i^{exp} counts per channel i ($i = 1, 2, \dots, 256$). Theoretically the counts per channel are given by equation (39) where E_o now becomes the channel position of the peak of the experimental dip

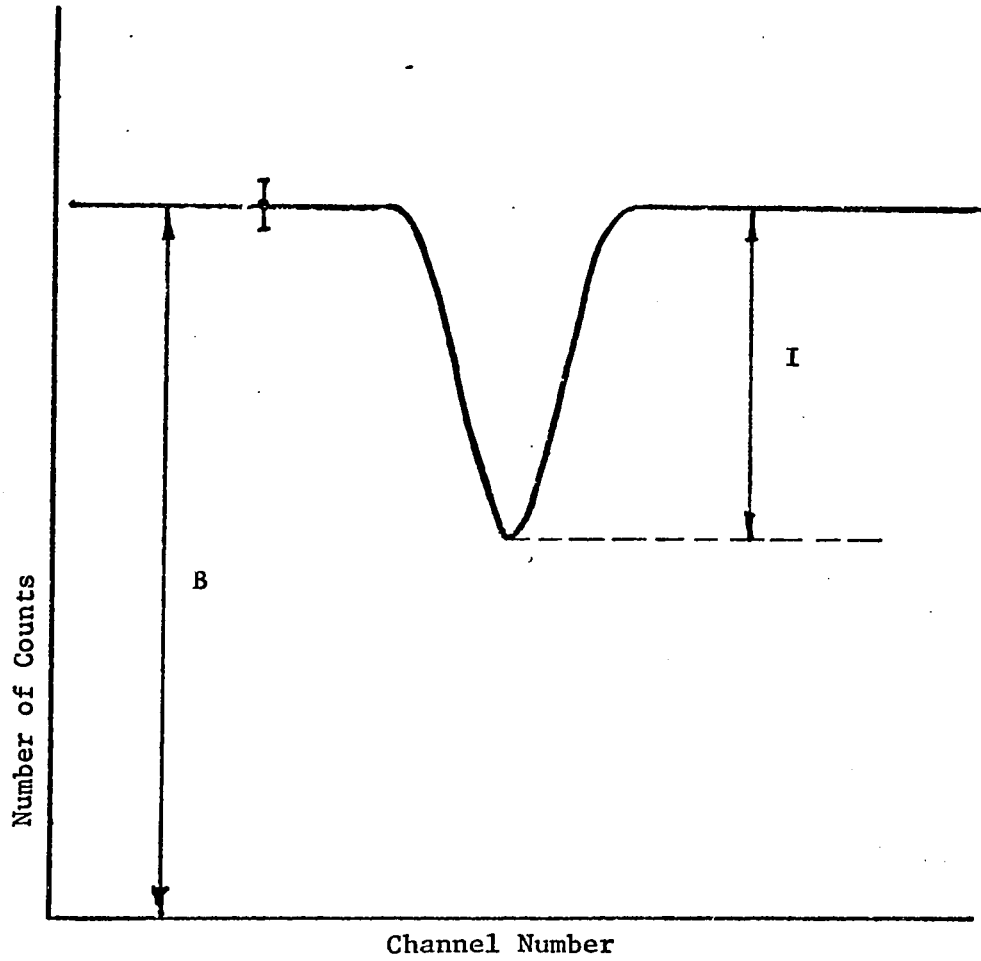


Figure 6. Mössbauer absorption spectrum

and Doppler energy, e , takes the channel number values from 1 to 256.

According to the least square criterion the "best" values of the parameters are given by minimizing the expression

$$Q = \sum_{i=1}^{256} \frac{(Z_i^{\text{exp}} - Z_{(pj)}^{\text{th}})^2}{\sigma_i^2} = \sum_{i=1}^{256} R_i^2 / \sigma_i^2 \quad (40)$$

where σ_i is the standard deviation of Z_i^{exp} and R_i is a residual term. For each individual channel there is a unique Poisson distribution and the number of counts recorded in that channel is a random sample from it. The standard deviation of this distribution is the square root of its mean and if each channel is considered in isolation the best value for this mean is simply the number of counts recorded. Hence the relation

$$\sigma_i = \sqrt{Z_i^{\text{exp}}}$$

The necessary condition for minimizing χ^2 with respect to a parameter P is given by (5)

$$\frac{\partial \chi^2}{\partial p_j} = 0 \quad (41)$$

Where $j = 1, \dots, 3M$, $3M$ being the maximum number of parameters, and the second derivative matrix has all positive eigen values. This assures us that the extremum defined by equation (41) is a minimum

and not a maximum. Actually, condition (41) is only satisfied to a certain degree, say, $\Delta\chi^2 = \epsilon$ where ϵ is a small quantity depending on the precision of the fitting.

The observables $Z_i(p)$ in the expression for χ^2 in Equation (40) are expanded about the point p_0 to the first order of Δp .

$$Z_i(p) = Z_i(p_0) + \left[\sum_j \frac{\partial Z_i}{\partial p_j} \right]_{p_0} \Delta p$$

Equation (40) then becomes

$$\chi^2 = \sum_i \left(\frac{Z_i^{\text{exp}} - Z_i(p_0) - \sum_j \left[\frac{\partial Z_i}{\partial p_j} \right]_{p_0} \Delta p_j}{\Delta Z_i^{\text{exp}}} \right)^2$$

The solution for $\vec{\Delta p}$ which minimizes χ^2 is

$$\vec{\Delta p} = -\alpha^{-1} \Delta \chi^2$$

Where α is the second derivative matrix or error matrix. The parameters p_j are stepped up by successive increments of Δp and the process is repeated until a solution is reached ($\Delta p \rightarrow 0$).

4.2 Methods of Measuring Recoilless Fraction

(a) Line Width Method

The most appropriate parameter for determining the recoilless fraction in the absorber is the full width at half maximum of the Mossbauer absorption peak as it is independent of the source, and the background (the non-Mossbauer gamma-radiation and the X-rays seen by the detector). These factors simply scale the amplitude and do not effect the width

or general shape of the peak. A theoretical expression due to Visscher⁽⁶⁾, relating the absorber line width, W , and the apparent thickness t of the absorber, is

$$W = 2\sqrt{(1.00 + 0.135t)} \text{ for } 0 < t < 5, \quad (42)$$

$$W = 2\sqrt{(1.01 + 0.145t - 0.0025t^2)} \quad (43)$$

for $4 < t < 10$.

A more accurate expression by J. Heberle⁽⁷⁾ is

$$W = 2\sqrt{(1 + 0.1288t + 4.733 \times 10^{-3}t^2 - 9.21 \times 10^{-4}t^3 + .63 \times 10^{-5}t^4)} \quad (44)$$

for $0 < t < 12$

where $t = n f_a \sigma_o$. From measured W , the quartic (44) can be solved (using the Newton-Raphson method) for t , from which f_a can be easily obtained.

(b) Line Width Dependence on Absorber Thickness

A linear dependence of experimental line width on absorber thickness is given by the expression (3)

$$W = \Gamma_a + \Gamma_s + 0.27 \sqrt{n \sigma_o} f_a \quad (45)$$

A graph of $W/\sqrt{n \sigma_o}$ as a function of $n \sigma_o$ will be a straight line of slope $0.27 f_a$. This gives a value of f_a which does not involve knowledge of the three other unknowns. The intercept $W = 0$ gives a value of $\Gamma_a + \Gamma_s$, where Γ_s is the source line width

and ∇_a is the absorber line width at half maximum.

(c) Area Method

The area under an absorption curve is independent of the source line width. Any photon in the arbitrary distribution of recoilless radiation from the source will assume all possible energy values on transmission as the velocity goes through a complete Doppler-shift cycle; every photon, therefore, will contribute equally to the area of absorption curve. The total area of an absorption curve is given by

$$\left(\frac{A}{\pi B}\right) = f_s \gamma_A t e^{-t/2} [I_0(t/2) + I_1(t/2)] \quad (47)$$

The expression for area of Lorentzian distribution is

$$A = \pi \frac{1}{2} W H \quad (48)$$

H is the height of the absorption peak, B is the background and I_0 and I_1 are the Bessel functions with imaginary arguments. Another expression for $t < 6$, is given by Lang⁽⁸⁾ where the absorption area integral for Lorentzian cross section is given by the expression

$$A = \frac{1}{2} f_s \nabla \pi L(t) \quad (49)$$

where ∇ is the natural line width and.

$$L(t) = \sum_{p=1}^{\infty} \frac{(-1)^{p+1} (2p-3)!! t^p}{p! (sp-2)!!} \quad (50)$$

$L(t)$ for various values of t are given in Table 1 as computed by D.W. Hafemeister and E. Brooks Shera (9). Knowing A , t and $L(t)$ from the table, f_s , can be found.

(d) Comparison Method

A method that has been used for finding f_s and f_a involves finding first f_s and t from the equation

$$\frac{N(t, \infty) - N(t, v)}{N(t, \infty) + B} = f_s [1 - e^{-t/2} I_0(t/2)] \quad (51)$$

where $I_0(t)$ is the zero order Bessel function with imaginary argument, $t = nf_a \sigma_a$, $N(t, v)$ is the counting rate of γ - quanta which have passed through the absorber with the source moving at velocity v , and $N(t, \infty)$ is the corresponding rate with the source moving at a sufficiently high velocity so that there is no resonance absorption, and B is the extraneous background. However because the expression $[1 - e^{-t/2} I_0(t/2)]$ does not vary rapidly with t , this procedure does not give very precise values of t for thick absorbers.

4.3 Velocity Calibration

In order to convert the channel number to absolute energy units it is necessary to make a velocity calibration. This was done by running a multilines spectrum with known hyperfine splitting. A source of Co^{57} in palladium and an absorber of

sodium nitropruside ($\text{Na}_2\text{F}(\text{CN})_5\text{NO}\cdot 2\text{H}_2\text{O}$) were used which provided a characteristic two line spectrum of standard separation of 1.726 mm/sec between them.

A least square fitting routine was used to fit the experimental data with Lorentzian distribution. The separation between the two peaks obtained in channels was converted to absolute energy units by the relation

$$\frac{v \cdot N}{128} = 1.726 \text{ mm/sec}$$

$$v = \frac{1.726 \times 128}{N} \frac{\text{mm}}{\text{sec}}$$

where N is the number of channels separating the two peaks and v is the Doppler velocity, which in turn is compared with the velocity shown on the calibration screw of the Doppler shifting mechanism. The procedure was repeated for a number of values. A calibration curve was obtained by plotting v versus the velocity shown on the calibration screw.

4.4 Discussion of Errors

The observed parameters have been determined from the width of the Mossbauer peak. The points of the experimental spectrum are subject to statistical scattering depending on the number of counts per point. Thus the statistical uncertainty in the width and hence the recoilless fraction depends on the number of counts.

Since the peak width depends on the point at half maximum, the error in the width is related to the error in the base line and height of the peak. Because all points equally effect the

base line, the standard error in the baseline is $\sigma_{BL} = \sigma/\sqrt{n}$ where $\sigma = \sqrt{N}$ is the standard deviation in each of the n points in the data. The statistical error in the height of the peak becomes larger as the number of points decreases. The magnitude of error depends on the range and distribution of the data points. The points at the peak contribute most to the error of the height. Thus $\sigma_H = \eta\sigma_{BL}$ where η has a value between 10 and 20 in most cases and can easily be determined from the spectrum.

Since the error in the height is of the order of 5 to 10 times the error in the base line, the base line error can be neglected in estimating the statistical error of the peak width. The error in the width is determined from the error in height assuming the peaks are Lorentzian and using the area, $A = \frac{\pi HW}{2}$, where W is the full width at half maximum and H is the peak height. Thus

$$\delta W = \frac{2A}{\pi H^2}, \quad \delta H = \frac{W}{H} \delta H$$

$$\text{or } \frac{\delta W}{W} = \frac{\delta H}{H}$$

Since $\delta H = \eta\sigma_{BL} = \eta\sqrt{N/n}$

$$\frac{\delta W}{W} = \frac{\eta}{H} \frac{N}{n}$$

Since the velocity range and the density of points is constant, η can be estimated from the Mossbauer spectrum.

For a statistical error in the width, $\frac{\delta W}{W}$, the error in the apparent absorber thickness T_a , and hence in the absorber recoilless fraction f_a is

$$T_a = \sigma_o n f_a$$

$$(T_a + \delta T_a) = \sigma_o n (f_a + \delta f_a)$$

$$\delta T_a = \sigma_o n \delta f_a - T_a - \sigma_o n f_a$$

$$\frac{\delta T_a}{T_a} = \frac{\sigma_o n \delta f_a}{\sigma_o n f_a} = \frac{\delta f_a}{f_a}$$

$$\text{or } \frac{\delta T_a}{T_a} = \frac{\delta f_a}{f_a} = \frac{\delta W}{T_a} \left(\frac{dT_a}{dW} \right)$$

The slope $\frac{dT_a}{dW}$ is calculated from the expression

$$W = W_o + 0.27 \sqrt{\sigma_o n f_a}$$

$$\text{or } W = W_o + 0.27 \sqrt{T_a}$$

$$1 = 0.27 \sqrt{\frac{dT_a}{dW}}$$

$$\frac{dT_a}{dW} = 11.8 \left(\frac{\text{mm}}{\text{sec}} \right)^{-1}$$

for T_a less than about 10.

Thus the statistical error in the recoilless fraction is

$$\frac{\delta f_a}{f_a} = 11.8 \left(\frac{\delta W}{W} \right) \frac{W}{T_a} = \frac{11.8 \eta W}{H \sqrt{nN} T_a} \quad (52)$$

Where H' is normalized by dividing by N . The errors δW and δH are given by the error matrix α , as we have seen in section 4.1.

For a typical spectrum

$$100 \cdot \frac{\Delta f_a}{f_a} \approx 9\%$$

4.5. Background

Probably the largest error introduced in the measurement of f_a for SnO_2 is the non-resonant background radiation, B. It does not only include radioisotope contamination in the source, but also gamma rays which are emitted at angles to its motion and are scattered into the detector.

The predominant origin of B is, however, the Sn X-rays produced during the internal conversion process of 65 keV M4 transition in Sn^{119} . Such an X-ray flux will contain rays of various energies, the most important being 25.27, 25.04 and 28.486 keV. The ratio of the internal conversion coefficient for this transition, $[K/(L+M+N+)]$ M4 is given by [14] as 0.42 ± 0.02 .

Background without the source was 3 counts/min through the single channel analyser window set for the 23.8 keV peak.

A 1.4 mil Palladium metal filter was used to reduce the unwanted X-rays from the Sn^{119} 23.8 kev gamma ray flux. The optimum filter thickness is given by Stöckler and Sano⁽⁹⁾.

t	L(t)	t	L(t)	t	L(t)
0.0	0.000	4.0	2.0951	17.0	4.5823
0.2	0.1905	5.0	2.3831	18.0	4.7193
0.4	0.3637	6.0	2.6390	19.0	4.8524
0.6	0.5220	7.0	2.8714	21.0	5.1082
0.8	0.6673	8.0	3.0860	22.0	5.2313
1.0	0.8015	9.0	3.2864	23.0	5.3517
1.25	0.9555	10.0	3.4751	24.0	5.4694
1.5	1.0966	11.0	3.6540	25.0	5.6980
1.75	1.2267	12.0	3.8245	27.0	5.808
2.0	1.3473	13.0	3.9877	28.0	5.917
2.5	1.5656	14.0	4.1444	29.0	6.026
3.0	1.7594	15.0	4.2954	30.0	6.125
3.5	1.9346	16.0	4.4412		

Table 1

Values of the Lorentzian function $L(t)$ versus absorber thickness t , as given by D.W. Hafemeister.

CHAPTER 5

RESULTS

5.1 Recoilless Fraction

The results in Tables 2 - 6 were obtained from measurements on a set of tin absorbers of different thicknesses varying from 0.5 mg/cm² to 10 mg/cm² using the method of line width dependence on absorber thickness. During the experiment the source was kept at room temperature while the absorber temperature was varied between 78°K and 298°K. The observed spectrum regardless of the absorber temperature, consisted of a single line with unresolved quadropole splitting. This imposed a limit on the use of other methods for calculating the probability of recoilless absorption.

As explained previously, the line width, W_{exp} , was determined by fitting the experimental data with a Lorentzian distribution. The dependence of the experimental line width on absorber thickness is given in Figures 7 to 10.

The slope of the plot between W_{exp} and absorber thickness gave the probability f_a , for recoilless resonance absorption. The value of f_a was found to be between .59 and 0.90 for temperatures ranging from 298°K to 78°K. The intercept gave $\Gamma_s + \Gamma_a = 1.0$ mm/sec at 78°K, where Γ_s and Γ_a are the intrinsic source and absorber line widths. This is not in good agreement with the theoretical value of $2\Gamma_o = 0.624$ mm/sec, Γ_o being the natural line width. This disagreement may be due to unresolved quadropole splitting in SnO₂, which limits the use of equation (36) for the evaluation of f_a .

The dependence of the recoilless fraction on temperature is given in Figure 11.

5.2 Determination of Debye Temperature

Using the experimental values of recoilless fraction, the Debye Temperature, θ , was determined from the Debye equation

$$f_a = \exp \left[- \frac{6E_R}{kT} \left(\frac{1}{4} + \frac{\hbar}{k\theta^2} \int_0^{\theta/T} \frac{x dx}{e^x - 1} \right) \right] \quad (53)$$

where the recoil energy, $E_R = E_{OA}^2 / 2Mc^2 = 2.572 \times 10^{-3}$ eV.

Equation (53) can be written as

$$-\frac{1}{6} \cdot \frac{T}{T_R} \ln f_a = \left(\frac{T}{\theta} \right) \left\{ \frac{1}{4} + (T/\theta)^2 \int_0^{\theta/T} \frac{x dx}{e^x - 1} \right\} \quad (54)$$

$$T_R = \frac{1}{k} \cdot \frac{E_{OA}^2}{2Mc^2}$$

The dependence of f_a with temperature was determined from the Mossbauer spectral lines, as shown in Table (2). For each temperature the corresponding value of θ was determined from equation (54).

To evaluate the integral $\int_0^{\theta/T} [x/(e^x - 1)] dx$ the function $x/(e^x - 1)$ was expanded in series given by Dwight, ⁽¹⁰⁾.

$$\frac{x}{e^x - 1} = 1 - x + \frac{B_1 x^2}{2!} - \frac{B_2 x^4}{4!} + \frac{B_3 x^6}{6!}$$

$$x^2 > 4\pi^2$$

where B_i are Bernoulli numbers. The results are given in Table 2.

A plot of Debye temperature versus temperature is given in Figure 12.

The values thus obtained are in good agreement with those of A. Boyle et al. ⁽¹¹⁾.

$T(^{\circ}\text{k})$	f_a	$\theta(^{\circ}\text{k})$
78	0.90	159
123	0.82	208
191	0.72	244
298	0.59	265

Table 2

Recoilless fraction, f_a , and Debye temperature, θ , as functions of temperature.

Natural Absorber Thickness in mg/cm ²	Line Area in mm/sec	Experimental Line Width in mm/sec
1.50	0.35	1.48
2.50	1.00	1.80
3.50	1.25	2.17
4.00	1.26	2.36
5.00	1.56	2.68
6.00	1.50	2.82
6.50	1.57	3.16

Table 3

Experimentally determined parameters for $T = 78^{\circ}\text{K}$

Natural Absorber Thickness in mg/cm^2	Line Area in mm/sec	Experimental Line width in mm/sec
2.50	0.60	1.77
3.00	0.626	1.83
3.50	0.682	2.08
4.00	0.788	2.108
5.00	0.80	2.39
5.50	0.91	2.295
6.0	1.03	2.66
6.5	1.28	2.75

Table 4

Experimentally determined parameters for $T = 191^\circ\text{K}$

Natural Absorber Thickness in mg/cm^2	Line Area in mm/sec	Line Width in mm/sec
1.0	0.363	1.28
1.5	0.510	1.48
2.5	0.600	1.54
3.5	0.646	1.86
4.0	0.732	2.02
5.0	0.750	2.36
6.0	0.809	2.28
7.5	1.067	2.70

Table 5

Experimentally determined parameters for $T = 298^{\circ}\text{K}$

Natural Absorber Thickness in mg/cm ²	Line Area in mm/sec	Line Width in mm/sec
1.5	0.615	1.73
2.5	0.583	1.75
3.5	0.773	2.06
4.0	0.869	2.22
5.0	0.987	2.52
6.0	1.126	2.76
6.5	1.200	2.94

Table 6. Experimentally determined parameters for
T = 123°K

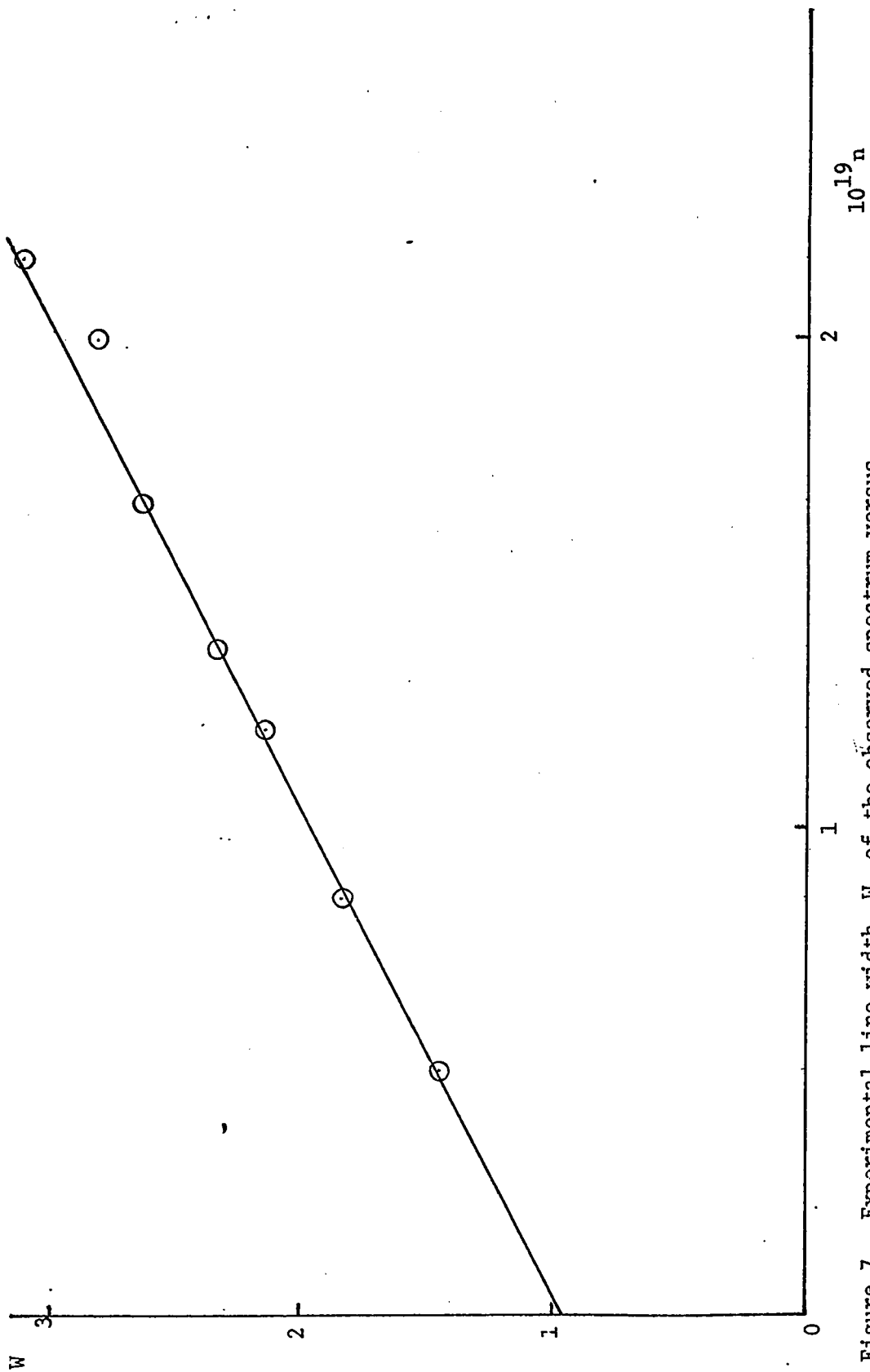


Figure 7. Experimental line width, W, of the observed spectrum versus absorber thickness at 78°K. The abscissa gives the number of atoms containing the Sn¹¹⁹ isotope per cm² of the absorber.

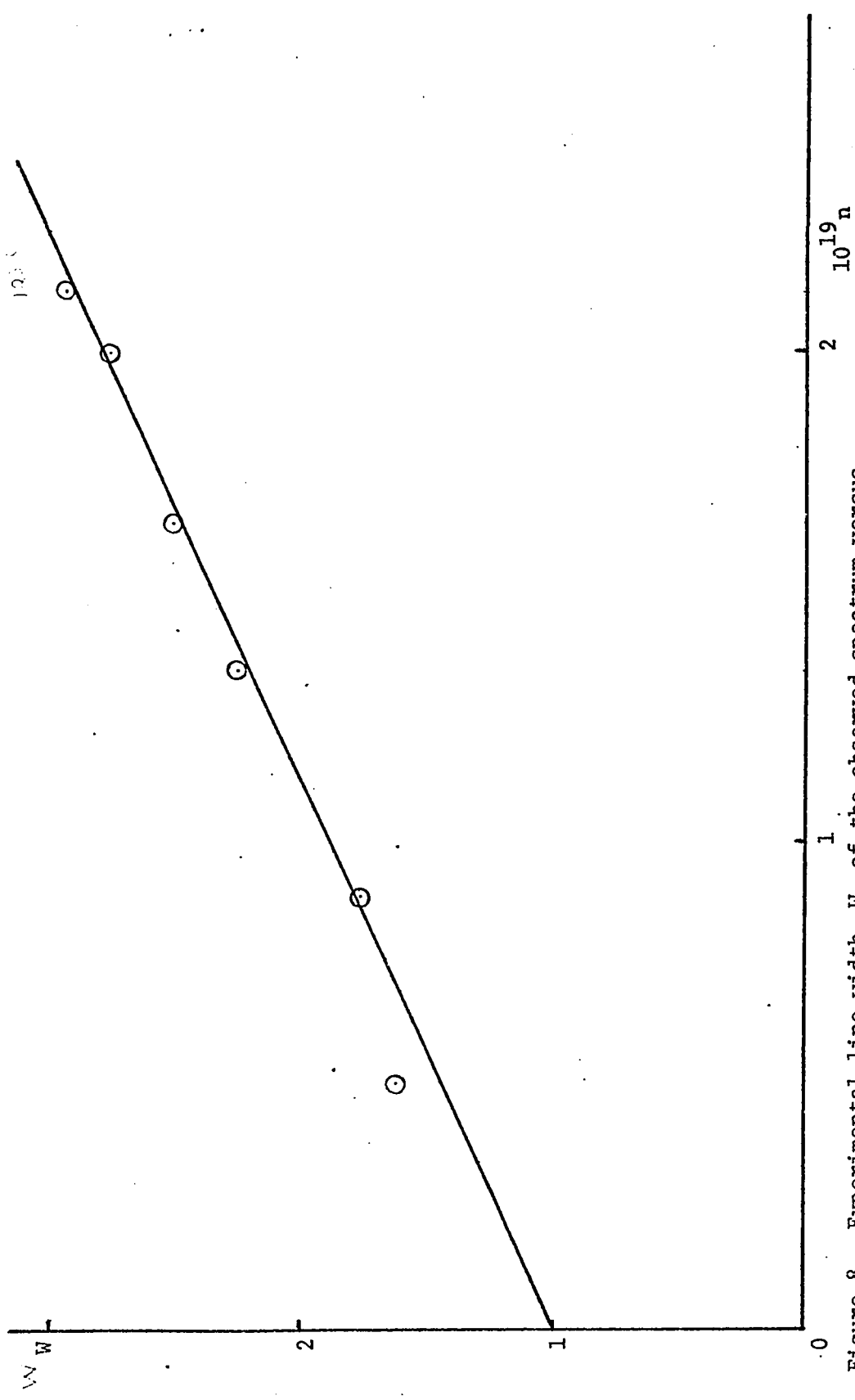


Figure 8. Experimental line width, W , of the observed spectrum versus absorber thickness at 1230K. The abscissa gives the number of atoms containing the Sn¹¹⁹ isotope per cm² of the absorber.

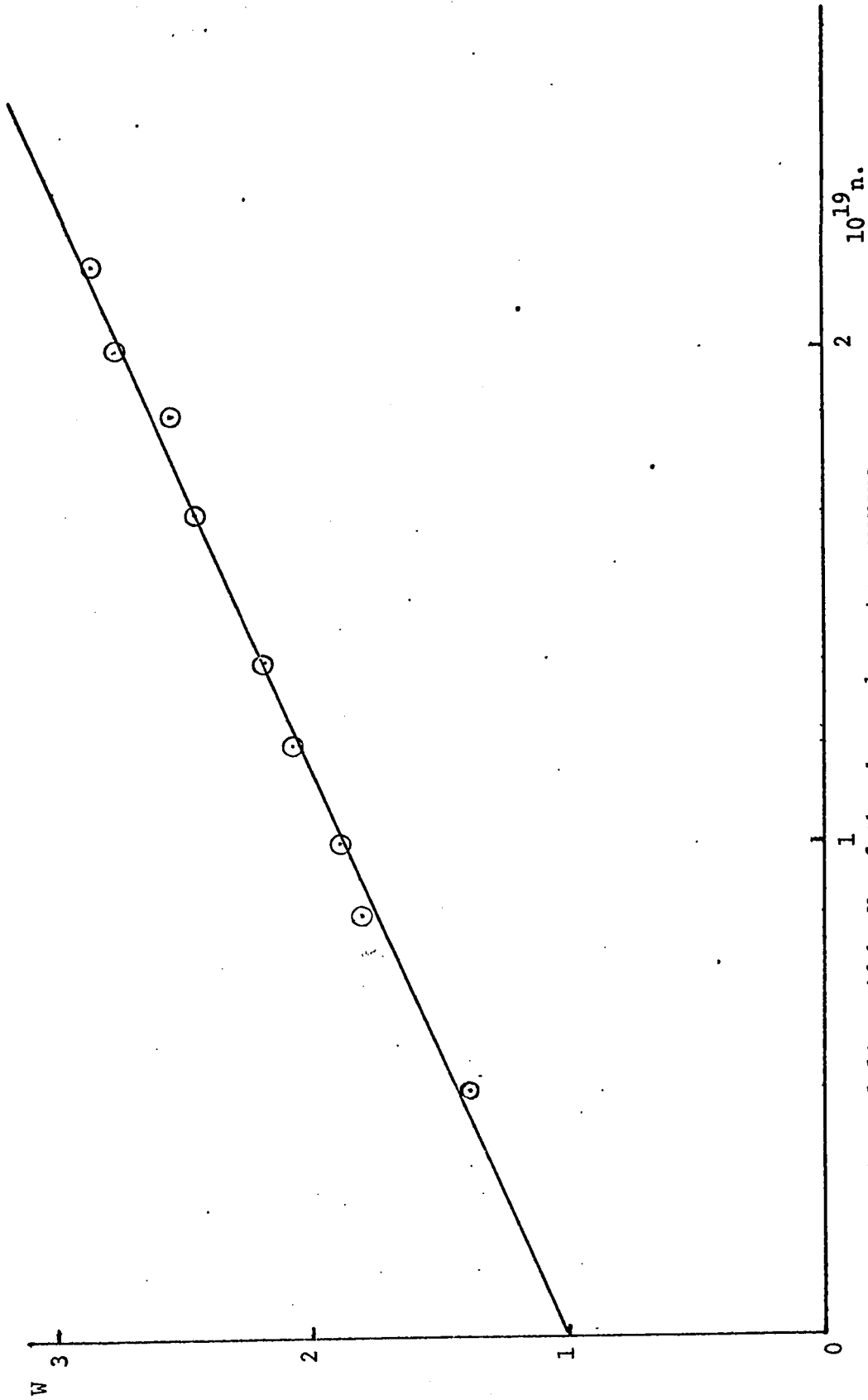


Figure 9. Experimental line width, W , of the observed spectrum versus absorber thickness at 1.91°K . The abscissa gives the number of atoms containing the Sn^{119} isotope per cm^2 of the absorber.

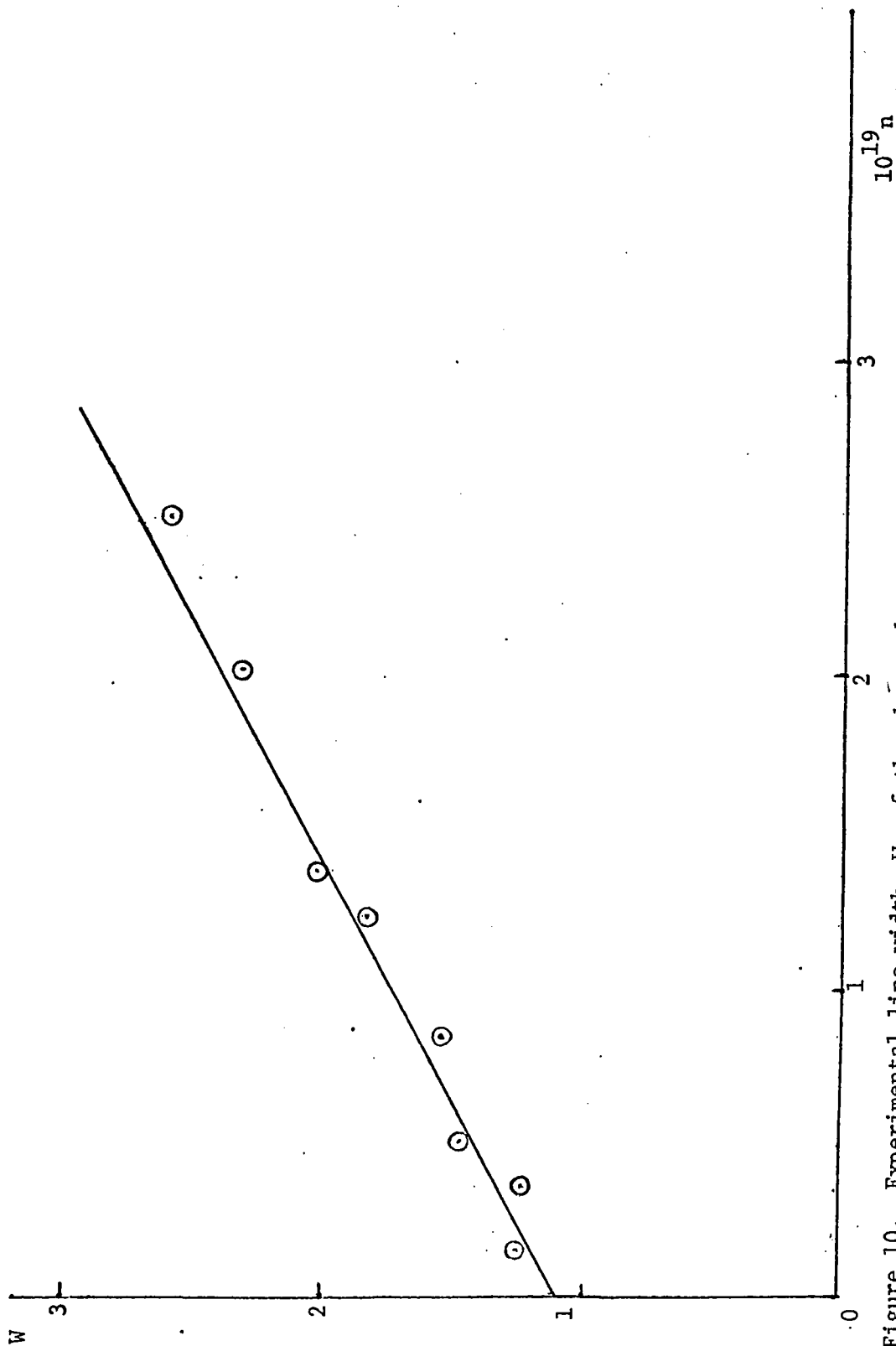


Figure 10. Experimental line width, W , of the observed spectrum versus absorber thickness at 298°K . The abscissa gives the number of atoms containing the Sn^{119} isotope per cm^2 of the absorber.

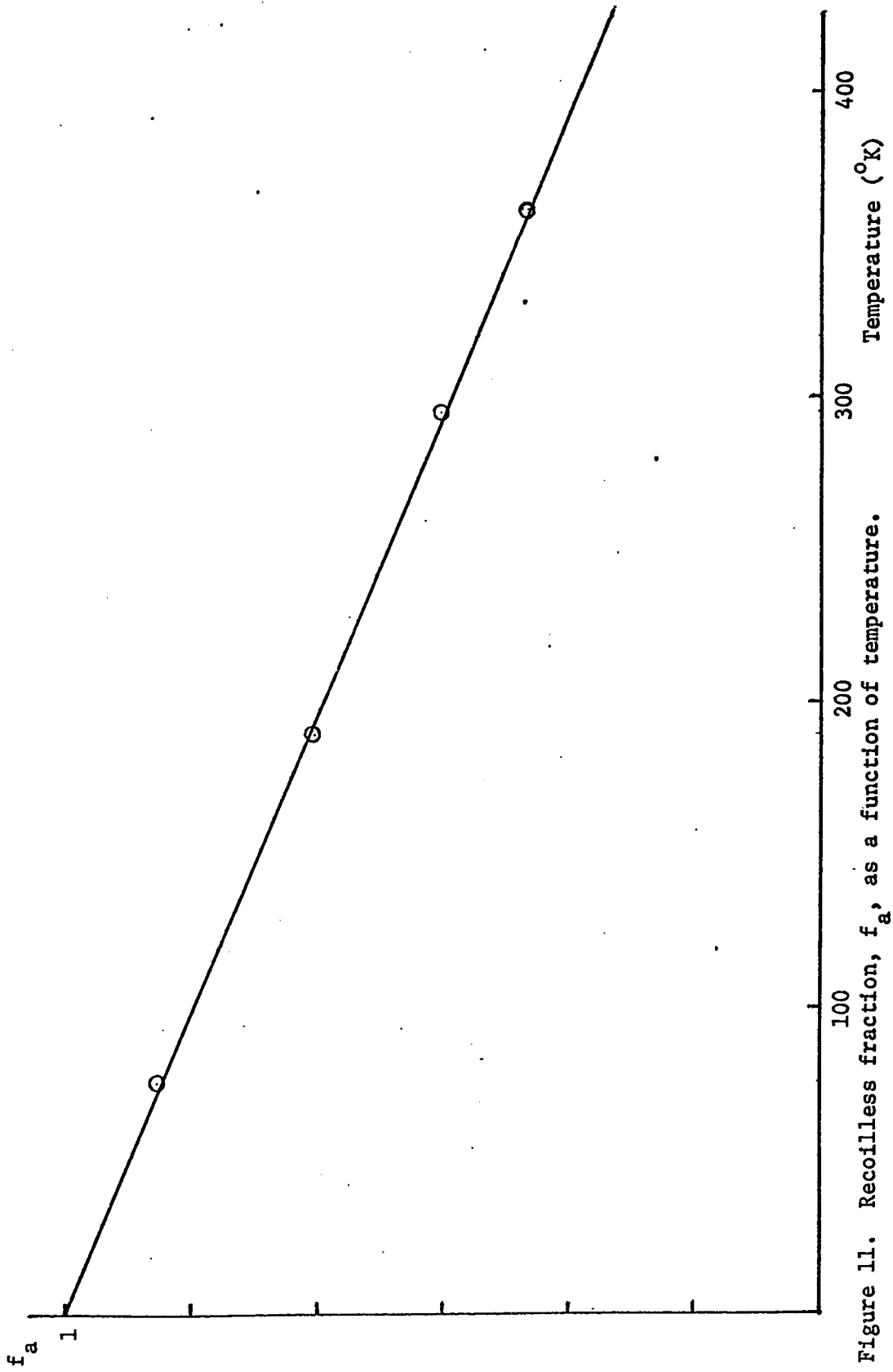


Figure 11. Recoilless fraction, f_a , as a function of temperature.

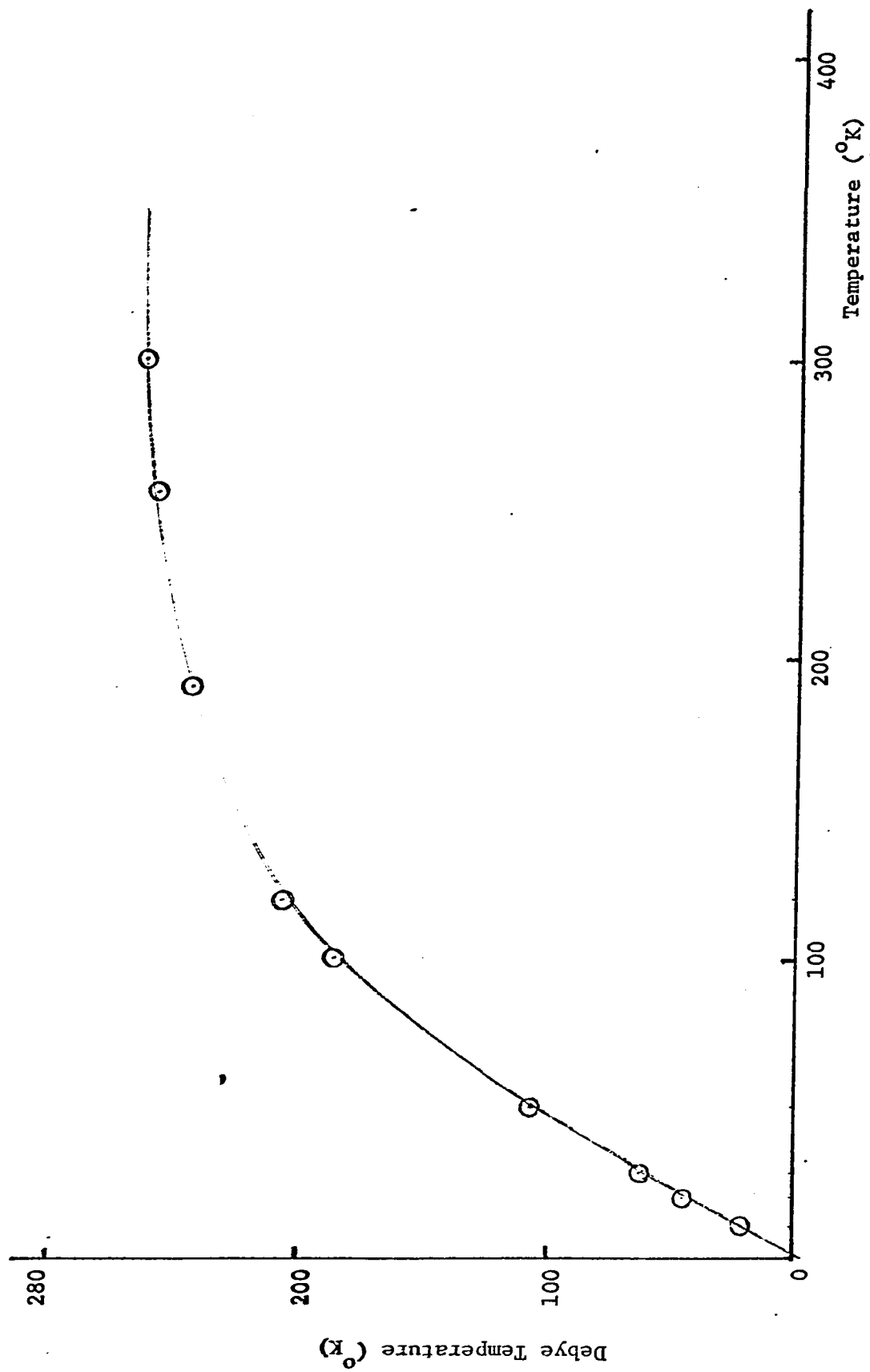


Figure 12. Debye Temperature vs. Temperature

From experimental data on the recoilless fraction

6.3 Conclusion

The conclusions from this investigation are as follows:

- (a) The probability of recoilless absorption for gamma quanta in SnO_2 decreases with increasing temperature. This is because of the excitation of acoustic phonons.
- (b) The apparent line width at zero absorber thickness ($\Gamma_{\text{app}}(0) = \Gamma_s + \Gamma_a$) is not twice the natural line width Γ_o . Γ_s and Γ_a being the intrinsic source and absorber line widths. This imposes some limitations on the methods used for calculating the recoilless fraction.
- (c) Reasonable results were obtained using only the method of line width dependence on absorber thickness. The area method failed to give results of any physical significance. This may be due to unresolved quadropole splitting, of order 0.45 mm/sec measured by Bryukhanov⁽¹⁵⁾ et al.
- (d) The Debye temperatures evaluated from data on Table 2 shows a variation of $\theta = 159^\circ$ at 78°K to $\theta = 265^\circ$ at 298°K .

The scope of this work could be further extended by varying the temperature of absorber above room temperature. This will provide more insight into the SnO_2 structure.

APPENDIX A

DEBYE-WALLER FACTOR

Up to this point the nucleus has been treated as being free, at rest, or in motion. For nuclei bound in a solid, it is necessary to take into account the interaction between the atoms of the lattice. As a result of this there is a probability of sharing the individual recoil energy among the atoms of the lattice.

If M is the effective mass of the recoiling system then the recoil energy is

$$E_R = \frac{(hf)^2}{2Mc^2} \quad (1)$$

From equation (1) it is easily seen that, for large enough M , the recoil energy is small, and therefore it is possible to have $E_R \ll \gamma$. The free-atom recoil energy of a gamma is shared in the excitation of the vibrational states of the lattice, or by phonon propagations through the crystal.

If, as a consequence of the recoil of the nucleus, phonons are created in the lattice with frequencies up to ω_1 , the energy available for the emitted gamma ray will be

$$hf = E_0 - \sum_i \hbar\omega_i \quad (2)$$

Again, if $\sum_i \hbar\omega_i$ is large enough compared with γ , the gamma ray cannot be resonantly absorbed.

Let us now consider the possibility of a zero phonon process.

It was known before the Mossbauer effect, that in elastic scattering of X-rays, there is no lattice excitation. Thus in a beam of scattered X-rays, a fraction of the total intensity will have the same frequency as that of incoming radiation. This fraction is known as the Debye-Waller factor

$$f = e^{-k^2 \langle x^2 \rangle}$$

Where $\langle x^2 \rangle$ is the mean square displacement of an atom under both thermal motion and zero-point motion, and $k = \frac{2\pi}{\lambda}$ is the wavevector of the X-ray.

The close analogy between the X-ray scattering and the recoilless process with gamma radiation suggested an interpretation of the Mossbauer effect in terms of the classical electromagnetic theory (Shapiro, 1960)⁽¹⁶⁾. The Debye Waller factor in the Mossbauer effect can be interpreted as due to the frequency modulation of the gamma radiation by the vibrational motion of the nucleus around its equilibrium position, via Doppler effect.

The vector potential of an electromagnetic wave emitted by a classical oscillator of constant frequency is

$$A(t) = A_0 e^{i\omega_0 t}$$

with

$$|A_0|^2 = 1$$

If ω is a function of time, then

$$A(t) = A_0 \exp \cdot i \cdot \int_0^t \omega(t') \cdot dt' \quad (3)$$

Assuming for simplicity that the emitting oscillator moves in the x direction, with a velocity $v(t)$ which is small compared to the velocity of light c : the Doppler effect changes the frequency of the emitted wave according to the following equation

$$\omega(t') = \omega_0 \left[1 + \frac{v(t')}{c} \right] \quad (4)$$

Substitution in Equation (3) and integration yields

$$A = A_0 \exp[i\omega_0 t] \cdot e^{i\omega_0 x(t)/c} \quad (5)$$

$$A = A_0 \exp(i\omega_0 t) \exp(ix(t)/\lambda)$$

If the oscillator is in harmonic motion with frequency Ω and amplitude x_0 , then

$$x(t) = x_0 \sin\Omega t \quad (6)$$

The vector potential then becomes

$$A = A_0 \exp(i\omega_0 t) \exp(ix_0 \sin\Omega t/\lambda) \quad (7)$$

Expanding the exponential in a Bessel-function series where

$$e^{iysin\theta} = \sum_{n=-\infty}^{+\infty} J_n(y) e^{in\theta}$$

It is found that

$$A(t) = A_0 \cdot \sum_{n=-\infty}^{+\infty} J_n(x_0/\lambda) e^{i(\omega_0 + n\Omega)t} \quad (8)$$

Equation (8) describes an electromagnetic wave which is formed by the superposition of partial waves of frequencies ω_0 , $\omega_0 \pm \Omega$, $\omega_0 + 2\Omega$, $\omega_0 + 3\Omega$.

The amplitude of each wave is given by the bessel function

$$J_n(x_0/\lambda)$$

The unshifted line can be identified with the Mossbauer line.

Its intensity is given by

$$f = |A_n = 0|^2 = J_0^2(x_0/\lambda) \quad (9)$$

Instead of a single frequency of the oscillator which corresponds to the Einstein model of a solid, a continuum of oscillator frequencies is assumed,

$$f = \prod_{m=1}^{3N} J_0^2(x_m/\lambda) \quad (10)$$

where x_m are the amplitudes of the oscillators, since the argument in (11) is small it is possible to write

$$J_0(xm/\lambda) = 1 - \frac{1}{4}(xm/\lambda)^2 + \dots \quad (11)$$

which after substitution in $\ln.f$ from equation (11) gives:

$$\ln.f = -2\sum \frac{1}{4}(x_m^2/\lambda^2) \quad (12)$$

Introducing the mean square deviation of the vibrating nucleus from its equilibrium position

$$\langle x^2 \rangle = \frac{1}{2} \sum x_m^2 \quad (13)$$

Equation (13) is written as

$$\ln.f = -\langle x^2 \rangle / \lambda^2 \quad (14)$$

which for an infinite number of oscillators becomes an exact expression

$$f = e^{-\langle x^2 \rangle / \lambda^2} \quad (15)$$

Where now $\langle x^2 \rangle$ is the mean square displacement of the atom emitting the gamma ray in a given direction, and λ is the wavelength of the gamma ray divided by 2π . In the above derivation a harmonic

motion of the atoms is assumed, which appears to be a good approximation except at temperatures above the Debye temperature.

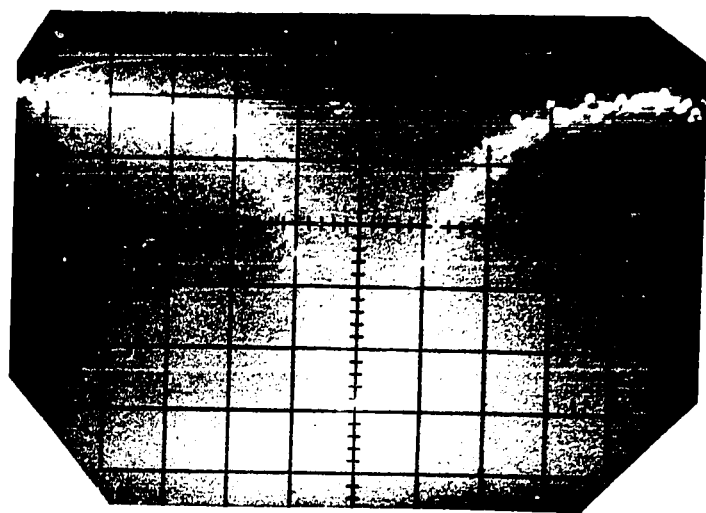
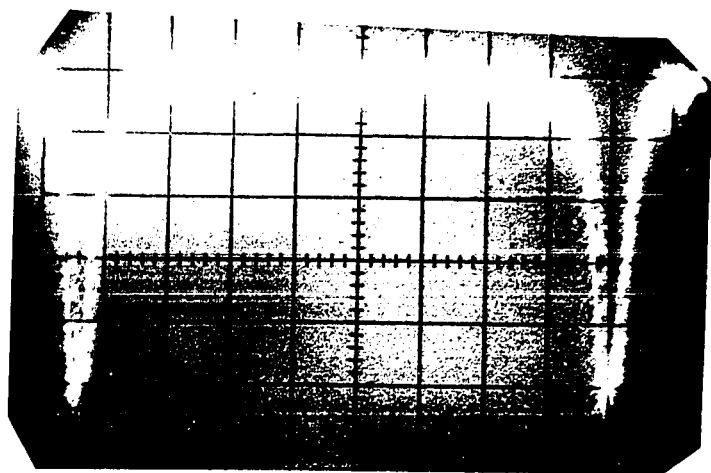


Figure 13. Mössbauer absorption spectrum
of SnO₂ at 290°K.

REFERENCE

1. J.D. Jackson, Canadian Journal of Physics Vol 33, 575 (1955)
2. W. Heitler, "Quantum Theory of Radiation", Clarendon Press, Oxford.
3. G.A. Bykov and Pham Zuy Hien, Soviet Physics, J.E.T.P.,
Vol. 16,646 (1963)
4. S. Margulies and J. Ehram, Nuclear Instruments and Methods
Vol. 12,131 (1961)
5. R.A. Arndt and M.H. MacGregor, Methods in Computational Physics,
Acad. Press Vol. 6,251 (1966)
6. W.M. Visscher (unpublished), Los Alamos Scientific Laboratory
7. J. Heberle, Nuclear Instruments and Methods, Vol. 58,90 (1968).
8. G. Lang, Nuclear Instruments and Methods, Vol. 24,425 (1963)
9. H.A. Stöckler and H. Shano, Nuclear Instruments and Methods,
Vol. 44,103 (1966)
10. H.B. Dwight, Tables of Integrals, The Macmillan Company
11. A.J.F. Boyle et al, Proc. Phys. Soc., Vol. 77,129.
12. A.I. Alikhanov and V.A. Lyubimov, Report to the Tenth Annual
Conference on Nuclear Spectroscopy, Moscow, 1960.
13. Delyagin, Shpinel et al. J.E.T.P., Vol. 14,1256 (1962).
14. D.A. Shirley, M. Kaplan and Axel, Phys. Rev., Vol. 123, No. 3,
816 (1961).
15. Bryukhanov et al., Soviet Physics J.E.T.P., Vol. 16,310 (1962).
16. F.L. Shapero, Soviet Physics Uspekhi, Vol. 3,881 (1961)
17. H. Frauenfelder, "The Mössbauer Effect", W.A. Benjamin, New York, 1962.