because the density of the liquid is lower and hence the Fermi wave length is larger. However, the periods of the curves of Johnson *et al.* appear to be 50% bigger which is a puzzling result.

Our computed curve does not show a minimum at the near-neighbor distance (3.5 Å) as does the Johnson curve. This minimum is not necessary for the stability of the metal since there are volume-dependent terms in the energy in addition to those included in the interaction potential and that potential itself depends upon the volume. However, such a minimum did occur in our treatment of aluminum[4] using the full nonlocal pseudopotential and is absent in the interaction derived from the model potential for aluminum. We conclude that the behavior of the interaction potential at short distances is not well given by the model potential we have used. Perhaps the appropriate behavior could be obtained, however, without use of a nonlocal potential if a different form were used.

VI. CONCLUSIONS

Our experimentation with a model pseudopotential has met with mixed success. We have found it possible to fit the vibration spectrum reasonably well with two parameters and presumably would do better with more. Furthermore, a pseudopotential is found which looks very much like that obtained by consideration of the electronic properties alone. The fact that the scheme seems to work somewhat better for aluminum than for lead might suggest that spin-orbit coupling plays an important role in lead and cannot be accounted for well with the simple pseudopotential. At the same time, a model potential, at least in the form we have chosen, seems not to duplicate all the results of the nonlocal potential. In particular, the effective interaction between ions is not well given.

The most striking finding is the account which the model in lead gives of the observed Kohn anomalies. This account is not sensitive to the detailed form of the model used but only to the form factor for back scattering. The size of the anomalies found would be much the same had we simply extracted the form factor for back scattering from our previously given form factors or if we had attempted an extrapolation of the form factors obtained by Anderson and Gold from studies of the Fermi surface. The model lends additional support in indicating that these form factors are consistent with the general form of the vibration spectrum.

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Measurement of the Mössbauer Recoilless Fraction in β-Sn for 1.3 to 370°K*†

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Doppler-shift measurements of Mössbauer recoilless fractions f in β-Sn show discrepancies of the order of 20 to 30% and sometimes bear quoted errors of ±10%. Such discrepancies can be caused by using incorrect values of α, the internal conversion coefficient; τ_m, the mean life of the excited state; Γ_A and Γ_S, the absorber and source linewidths; and B, the nonresonant background present in the detector at the energy of the Mössbauer γ rays. In the present work, the use of a black resonant absorber and the technique x-γ delayed coincidences combine to eliminate dependence on these parameters in first approximation. In particular, the results $f=0.455\pm0.010$ at 77.3°K and $f=0.72\pm0.01$ at 4.2°K are obtained. The errors are systematic, and are due largely to uncertainties in evaluating the residual resonant transmission of the black absorber, the total magnitude of which is about 5% for $T\leq100$°K. For the experimental temperature range of $1.3\leq T\leq370$°K, f values are obtained at over 300 points for two different source samples. The results are as much as 20% higher than some previously reported values, and also do not agree well with the theoretical calculations of DeWames, Wolfram, and Lehman for $T\leq150$°K. On the other hand, when the data are expressed in terms of a Debye temperature Θ derived at each temperature from the Debye formula for f, the Θ values show remarkably little variation with temperature, and fall on a smooth curve. The results at low temperature help to clarify the data of Wiedemann, Kienle, and Pobell in the superconducting region and immediately above.

1. INTRODUCTION

NUMEROUS authors have reported data on the temperature dependence of the recoilless fraction f in the Mössbauer effect. It is naturally of interest to compare these experimental f values to theoretical predictions, since this can be considered as a direct test of various forms of the phonon frequency spectrum. f is given by the relation

$$f=e^{-2W}, \tag{1}$$

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TABLE I. Summary of previous f values as a function of temperature.[a]

T (°K)	Boyle *et al.*	Barloutaud *et al.*	Experiments Wiedemann *et al.*	Bryukhanov *et al.*	Alekseevskii *et al.*	Theory DeWames *et al.* I	Theory DeWames *et al.* II
4			0.60			0.63	0.61
20			0.59			0.61	0.59
77		0.285±0.015	0.40	0.36±0.04	0.32±0.06	0.40	0.37
90		0.32 ±0.015				0.36	0.32
120	0.30					0.27	0.24
150	0.22					0.22	0.18
200	0.138					0.138	0.107
300	0.035	0.045±0.01		0.06±0.01	0.06±0.01	0.053	0.036
400	0.009					0.019	0.011
500	0.002					0.008	0.004

[a] A value for f at 90°K obtained from resonant scattering by Barloutaud *et al.* has not been given here because in the same paper a more precise value obtained from a transmission experiment is reported. Results by N. N. Delyagin *et al.*, Zh. Eksperim. i Teor. Fiz. **41**, 1347 (1961) [English transl.: Soviet Phys.—JETP **14**, 959 (1962)], have also been omitted because more recent reports by Bryukhanov *et al.* and Gorodinskii *et al.* (footnotes 9, 15) indicate that the large splittings in the velocity spectra of the Delyagin paper are probably instrumental effects which would render f values obtained from them invalid.

where $2W$ is a weighted average over the phonon spectrum, and where the weighting factor is obtained from the theory of the Mössbauer effect.[1]

For β-Sn the data may be compared to two existing calculations. The first uses the Debye approximation for the phonon spectrum and gives f as a unique function of the Debye temperature Θ. Though this allows comparison of Mössbauer Θ values to other Θ values (say specific heat derived), the Debye theory has too many obvious limitations to be taken as an adequate representation of the phonon spectrum. A second calculation recently reported by DeWames, Wolfram, and Lehman (DWL)[2] is perhaps of greater interest. It uses a phonon spectrum that is reconstructed from measured values of the elastic constants of β-Sn. If this method suffers from any fundamental difficulty, it is that the elastic constants were measured at frequencies (10^7 cps) that are many orders of magnitude lower than those frequencies (10^{12} cps) which make the dominant contribution to the average that determines f. (See *Note added in proof.*)

At present, there is rough agreement between *both* theories and the various experimental values. This is possible only because of large discrepancies between different sets of data. It is the purpose of the present paper to sort out some of the possible reasons for the experimental discrepancies, and to report on some new measurements of f.

2. PREVIOUS f MEASUREMENTS

Some previous experiments on the temperature variation of nuclear resonance absorption in β-Sn do not produce absolute values of $f(T)$. For example, in the work of Mitrofanov and Shpinel,[3] the ratio $f^2(T)/f^2(83)$ was obtained for several experimental temperatures T. These data were then compared to a family of curves $f^2(\Theta,T)/f^2(\Theta,83)$ versus T, with Θ as parameter, in which $f(\Theta,T)$ was calculated from the Debye formula

$$f(\Theta,T)=\exp\left\{-\frac{6R}{k\Theta}\left[\frac{1}{4}+\left(\frac{T}{\Theta}\right)^2\int_0^{\Theta/T}\frac{x\,dx}{e^x-1}\right]\right\} \quad (2)$$

(R is the energy of free recoil, k is Boltzmann's constant). Values of Θ obtained from these curves at each temperature T are clearly those that uniquely fit the Debye formula at temperatures T and 83. Since these Θ values vary with temperature, and hence depend on the normalization point $T=83$°K, they cannot be used to find f as a function of T by inversion of Eq. (2). A similar remark applies to some data of Yaqub and Hohenemser.[4]

Experiments in which *absolute* f values were obtained have been reported by Boyle *et al.*,[5] Barloutaud *et al.*,[6,7] Wiedemann *et al.*,[8] Bryukhanov *et al.*,[9] and Alekseevskii *et al.*[10] A tabular summary of these results is given in Table I. Since comparison to the Debye formula is difficult in this form, it is useful to transform to a coordinate system particularly suitable to the Debye theory. Thus, Θ values calculated from f values by inversion of the Debye formula (2) have been plotted in Fig. 1. The plot indicates that with the exception of the two points at 4 and 20°K there is little basis on which to distinguish between a simple fit of Θ=constant and the DWL theory. It is further evident that discrepancies

[1] R. L. Mössbauer, Ann. Rev. Nucl. Sci. **12**, 123 (1962).
[2] R. E. DeWames, T. Wolfram, and G. W. Lehman, Phys. Rev. **131**, 529 (1963).
[3] K. P. Mitrofanov and V. S. Shpinel, Zh. Eksperim. i Teor. Fiz. **40**, 983 (1961) [English transl.: Soviet Phys.—JETP **13**, 686 (1961)].
[4] M. Yaqub and C. Hohenemser, Phys. Rev. **127**, 2028 (1962).
[5] A. J. F. Boyle, D. St. P. Bunbury, and C. Edwards, Proc. Phys. Soc. (London) **77**, 129 (1961).
[6] R. Barloutaud, E. Cotton, J. L. Picou, and J. Quidort, Compt. Rend. **250**, 319 (1960).
[7] R. Barloutaud, J. L. Picou, and C. Tzara, Compt. Rend. **250**, 2705 (1960).
[8] W. H. Wiedemann, P. Kienle, and F. Pobbell, Z. Physik **166**, 109 (1962).
[9] V. A. Bryukhanov, N. N. Delyagin, A. A. Opalenko, and V. S. Shpinel, Zh. Eksperim. i Teor. Fiz. **43**, 432 (1962) [English transl.: Soviet Phys.—JETP **16**, 310 (1963)].
[10] N. E. Alekseevskii, Pham Zuy Hien, V. G. Shapiro, and V. S. Shpinel, Zh. Eksperim. i Teor. Fiz. **43**, 790 (1962) [English transl.: Soviet Phys.—JETP **16**, 559 (1963)].

between sets of points are considerably larger than estimated experimental errors. In the following, sources of systematic error which may account for the discrepancies in reported f values are discussed.

In most cases[5,6,8,9] some form of the usual velocity spectrometer is used, and f values are deduced from the absorption

$$p(v)=[C(\infty)-C(v)]/C(\infty), \tag{3}$$

evaluated at zero velocity [$C(v)$ is the experimental counting rate at velocity v]. The zero-velocity absorption is related to the resonant transmission $\mathcal{T}_A$ by

$$p(0)=f_{\text{eff}}(1-\mathcal{T}_A), \tag{4}$$

where f_{eff} is the effective fraction of resonant quanta emitted by the source in the energy range accepted by the detector. It is not necessarily equal to f, the fraction of nuclear decays resulting in recoilless γ rays since in general the source will be self-absorbing and the nuclear γ rays will be superposed on a background of unresolved K x rays. The resonant transmission is given by

$$\mathcal{T}_A=\int_0^\infty I(E)\exp[-\sigma_0 f' n_A A(E)]dE, \tag{5}$$

where $I(E)$ is the energy spectrum of incident nuclear γ rays, $A(E)$ is the energy dependence of the absorber resonant cross section, f' is the probability of recoilless absorption, n_A is the number of resonant nuclei per cm^2, and σ_0 the maximum resonant cross section for the absorber. For single Lorentzian emission and absorption distributions, $\mathcal{T}_A$ can be expressed in terms of four widths: Γ_S', the width of $I(E)$; Γ_A, the width of $A(E)$; Γ_γ, the natural radiation width; and Γ, the natural total width of the excited state. For a source of finite thickness, Γ_S' is larger than the width Γ_S of the source resonant cross section $S(E)$ [analogous to $A(E)$]. The maximum resonant cross section for the absorber depends on Γ_γ, Γ, and Γ_A by

$$\sigma_0=2\pi\left[\frac{\hbar c}{E_0}\right]^2\frac{2I_e+1}{2I_0+1}\frac{\Gamma_\gamma}{\Gamma}\frac{\Gamma}{\Gamma_A}. \tag{6}$$

For the source, Γ_A is replaced by Γ_S in the above. (E_0 is the transition energy, I_e and I_0 the excited and ground-state spins.) Two of the four widths involved are determined by measured quantities:

$$\Gamma=\hbar/\tau_m, \tag{7a}$$

$$\Gamma_\gamma/\Gamma=1/(1+\alpha). \tag{7b}$$

τ_m is the mean life of the excited state and α is the total internal conversion coefficient of the transition. The other widths are unknown, and must be found or assumed in a resonance absorption experiment.

To extract f or f' from the above equations, one approach[6,8] has been to reduce the x-ray background to negligible proportions, and then to work with source and absorber kept at a common temperature and made of the same material (metallic β Sn). In this case, $f=f'$, $\Gamma_A=\Gamma_S$, and f_{eff} may be expressed in terms of the absorber parameters f' and Γ_A by correcting for self-absorption in the source, if necessary. The most general form for f_{eff} is

$$f_{\text{eff}}=f\int_0^\infty \rho(n)\mathcal{T}(t_S)dn \Big/ \int_0^\infty \rho(n)dn, \tag{8}$$

where $\rho(n)$ is the source density distribution;

$$t_S=f'n\sigma_0 \tag{9}$$

is the source thickness dependent on n, the number of resonant nuclei per cm^2 separating a particular source lamina from the surface; and

$$\mathcal{T}(t)=e^{-t/2}J_0(it/2) \tag{10}$$

is the resonant transmission in the special case when $I(E)$ and $A(E)$ are Lorentzians of equal width and overlap perfectly. Thus, provided that Γ_γ/Γ and Γ/Γ_A are known, experimental values of f' are obtainable by using (8) in (4) and solving for f'. Low x-ray backgrounds required for the above method are achieved with Sn^{118} enriched source material and filtration of the radiation with critical absorbers. These methods are often not wholly successful,[11] and can leave x-ray contaminations of several percent.

It is for this reason that some authors[5,9] have attempted to eliminate f_{eff} from the problem without

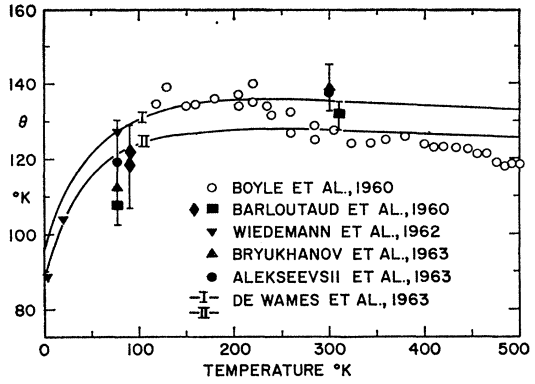

FIG. 1. Previously published experimental f values in β Sn are shown in terms of the Debye temperature Θ obtained by inversion of the Debye formula for f; the theory of DeWames *et al.* based on two sets of elastic-constants data (I, II) is also shown.

[11] The highest enrichment reported in f measurements (88%) still leaves sufficient competing isotopes to produce x-ray intensities comparable to the Sn^{119} γ-ray intensity, especially because the neutron activation cross section is 100 times larger for Sn^{112} than for Sn^{118}. Filtration affects only x rays having energy higher than the γ ray, and thus leaves the In x rays from Sn^{113} untouched. Finally, even if critical absorption is possible, almost every absorption event results in another x ray (characteristic of the critical absorber) which is no better resolved from the γ ray than the original x ray, though it is much reduced in effective solid angle.

TABLE II. Line broadening in β-Sn.[a]

Workers[b]	Source			Absorber			Line broadening Γ_b/Γ[d]		Γ_{exp}/Γ[e]
	Material	T	t_S[c]	Material	T	t_A[c]	$\alpha=5.2$	$\alpha=6.3$	
Boyle *et al.*	β Sn	100	2.67	β Sn	300	1.52	3.10	2.95	4.00
Wiedemann *et al.*	β Sn	77	1.95	β Sn	77	3.9	3.54	3.30	4.40
Bryukhanov *et al.*	SnO_2	300	0.96	β Sn	77	0.72	2.47	2.36	5.1
	SnO_2	300	0.96	β Sn	300	0.61	2.44	2.33	3.9
Alekseevskii *et al.*	SnO_2	300	0.42	β Sn	77	5.2	3.50	3.25	4.85
Gorodinskii *et al.*	β Sn	83	0.17	β Sn	83	2.9	2.96	2.84	5.0

[a] Some linewidth measurements have been omitted because of extremely thick sources and/or absorbers used. They are not very useful for studying line-broadening effects for obvious reasons.
[b] The references for this column are given in footnotes 14, 8, 9, 10, and 15 in that order.
[c] t_S and t_A are dimensionless source and absorber thicknesses. The values given in the table are for $\alpha=5.2$. Values for $\alpha=6.3$ are 15% smaller.
[d] Γ_b is the experimental linewidth, full width at half-maximum (FWHM), expected on basis of the t_S and t_A values given, and on the assumption that $\Gamma_A=\Gamma_B=\Gamma$.
[e] Γ_{exp} is the observed experimental linewidth (FWHM).

reference to the x-ray background. They have measured $p(0)$ for several values of n_A, and by choosing a reference foil of thickness n_{A0}, have obtained experimental ratios $p(0,n_A)/p(0,n_{A0})$ as a function of n_A. By Eq. (4),

$$p(0,n_A)/p(0,n_{A0})=(1-\mathcal{T}_A)/(1-\mathcal{T}_{A0}). \quad (11)$$

The ratio on the right may be calculated as a function of n_A for several values of f'. The calculated curve which best fits the experimental absorption ratios on the left identifies the value of f'.

In both methods described above, the weakest part of the argument is the evaluation of the resonant transmission $\mathcal{T}_A$. As already noted, $\mathcal{T}_A$ depends in general on the four widths Γ_γ, Γ, Γ_A, and Γ_S'. In the reported f measurements which use the zero-velocity transmission method, Γ_γ and Γ are taken from the experimental values of α and τ_m and $\Gamma_A=\Gamma_S'=\Gamma$ is assumed. This allows the use of the simple analytic form (10) for $\mathcal{T}_A$, and Eq. (6) with $\Gamma/\Gamma_A=1$ for σ_0. The f values deduced under these conditions are subject to sizable error for the following reasons:

(1) τ_m carries an experimental error of $\pm 5\%$.[12]

(2) The two reported values of Γ_γ/Γ differ by 20% and are not consistent within experimental error.[12,13]

(3) Γ_A may be 40% larger than Γ, and probably varies with temperature. Even allowing for the experimental errors in Γ and Γ_γ/Γ, observed velocity spectra can not be explained by using emission and absorption widths of magnitude Γ. The evidence for this statement is assembled in Table II, which shows a comparison of experimentally observed linewidths[8–10,14,15] and corresponding linewidths obtained from the numerical calculations of Margulies and Ehrman[16] on the assumption that $\Gamma_A=\Gamma_B=\Gamma$.

(4) For most f measurements, the sources used are thick, and require that the source beam widths Γ_S' be even greater than $\Gamma_S \simeq \Gamma_A$.

It remains to add that Alekseevskii *et al.*,[10] using a SnO_2 source and a β Sn absorber, have deduced f values from the integral absorption,

$$P=\int_{-\infty}^{+\infty} p(v)dv, \quad (12)$$

the properties of which have been analyzed[17–19] for single as well as split line absorption. If the source has a Lorentzian emission cross section of known width, the quadrupole splitting parameter Δ may be explicitly determined by this method, and thus any uncertainty due to Γ_A is eliminated. To carry out the analysis, the source thickness $t_S=n_S\sigma_0 f$ and f_{eff} must be known. Alekseevskii *et al.* assumed $\Gamma_S=\Gamma$, and estimated t_S roughly. (The latter is a valid procedure since t_S was small.) For determining f_{eff}, however, they turned to the zero-velocity absorption method, using SnO_2 absorbers of varying thickness. It is clear, therefore, that all the weaknesses already mentioned for this method are transferred to the integral absorption method in this case. If a net improvement has been made, it is that SnO_2 source and absorbers were employed; for velocity spectra of SnO_2 are more closely explained by natural width emission and absorption than those of β-Sn, so that the assumption $\Gamma_A=\Gamma$ is probably a better approximation.

In summary, then it is likely that all five of the reported f measurements contain undetermined systematic errors from several sources. Some of these may cancel each other, such, as for example, a Γ/Γ_A value that is too large and a Γ_γ/Γ value that is too small. A minimal estimate of the accuracy of reported f values

[12] J. L. Olsen, L. G. Mann, and M. Lindner, Phys. Rev. **106**, 985 (1957).
[13] N. Benzcer-Koller, Phys. Rev. **134**, B1205 (1964).
[14] A. J. Boyle, D. St. P. Bunbury, and C. Edwards, Proc. Phys. Soc. (London) **77**, 1062 (1961).
[15] G. M. Gorodinskii, L. M. Krizhanskii, and E. M. Kruglov, Zh. Eksperim. i Teor. Fiz. **43**, 2050 (1962) [English transl.: Soviet Phys.—JETP **16**, 1449 (1963)].
[16] S. Margulies and J. R. Ehrman, Nucl. Instr. **12**, 131 (1961).
[17] D. A. Shirley and M. Kaplan, Phys. Rev. **123**, 816 (1961).
[18] G. A. Bykov and Pham Zuy Hien, Zh. Eksperim. i Teor. Fiz. **43**, 909 (1962) [English transl.: Soviet Phys.—JETP **16**, 646 (1963)].
[19] G. Lang, Nucl. Instr. **24**, 425 (1963).

would be ±10% with larger deviations certainly not excluded.

3. AN IMPROVED METHOD

In searching for a new experiment that would give precise as well as accurate values of f, one would like to eliminate dependence on the widths Γ_γ, Γ, Γ_A, and Γ_S', as well as the x-ray background. To see how this might be done, consider the total transmission,

$$\mathcal{T}_0(T)=C_e(1-f+f_{\rm eff}\mathcal{T}_A)+B. \quad (13)$$

C_e is the electronic transmission of the nuclear γ rays, B is the transmitted x-ray background, and $f_{\rm eff}$ is the effective fraction of 23.8-keV quanta emitted without recoil by the source (note the difference from the previous usage of $f_{\rm eff}$). The dependence on T indicates that the total transmission is a function of the temperature at least through f and $\mathcal{T}_A$, if not weakly through C_e and B as well. Clearly, if one can manage to eliminate $f_{\rm eff}\mathcal{T}_A$, B, and C_e, (13) may be solved for f, the recoilless fraction of the source.

In the experiment reported here,[20] this was achieved by combining the use of a nearly black resonant absorber with x-γ coincidences and normalization at high temperature. A β-Sn source was clamped in thermal contact with a Sn^{119} enriched resonant absorber, and coincidences between transmitted (25±15)-keV photons were observed by two scintillation counters. The experiment was carried out at the boiling points of nitrogen and water, and thus the ratio of the coincidence rate at 77.3 and 373°K was determined. From (13) it is seen that the most general form of this ratio will be

$$\frac{N_C(77)}{N_C(373)}=\frac{[C_e'(1-f+f_{\rm eff}\mathcal{T}_A)]_{T=77}+B'}{[C_e'(1-f+f_{\rm eff}\mathcal{T}_A)]_{T=373}+B'}. \quad (14)$$

C_e' and B' are equal to C_e and B times the appropriate over-all coincidence efficiency, respectively. To deduce f values from (14), the following simplifications were required:

(a) $C_e'(373)=1.006\,C_e'(77)$ was derived by calculating the effect of thermal expansion on the source and absorber thicknesses. The result was checked experimentally by measuring the atomic absorption of nonresonant 23.7-keV In K x rays obtained from In^{114}.

(b) Since $f\simeq 0.01$ is a good approximation at 373°K, $(1-f+f_{\rm eff}\mathcal{T}_A)_{T=373}=0.99$ was used in the denominator of (14).

(c) $f_{\rm eff}\mathcal{T}_A$ was expressed in terms of a function $g(f)$, dependent in the source recoilless fraction alone.

(d) Critical absorbers were arranged so as to make the non-Sn^{119} coincidence rate B' very nearly zero.

With these steps, (14) reduces to

$$N_C(77)/N_C(373)=0.996[1-f+g(f)]_{T=77}. \quad (15)$$

[20] A preliminary report of this experiment has been given by C. Hohenemser and W. F. Walters, Bull. Am. Phys. Soc. **10**, 64 (1965).

Thus, it is seen that to the extent the indicated simplifications are correct, all five of the causes of uncertainty in the previous f measurements have been eliminated.

Before obtaining f values from (15) it is necessary to explicate the simplifications (c) and (d) further. Simplifications (a) and (b) are sufficiently clear and involve so little possible error that further comment is not needed.

Residual Resonant Transmission

Because the source is thin, the approximation $f_{\rm eff}=f$ was used. By the equality of source and absorber temperatures, $f=f'$ and $\Gamma_A=\Gamma_S$ are very nearly if not exactly correct, and the use of (10) for $\mathcal{T}_A$ is justified. It remains to specify Γ_γ/Γ and Γ/Γ_A. In accordance with the trend in Table II, $\Gamma/\Gamma_A=0.7$ was used, and from the most recent measurement of the internal conversion coefficient,[13] $\Gamma_\gamma/\Gamma=0.16$ was obtained. The resulting form for the residual resonant transmission is therefore

$$f_{\rm eff}\mathcal{T}_A=g(f)\equiv f\exp(-t_A/2)J_0(it_A/2), \quad (16)$$

and for the particular resonant absorber used, $t_A=n_A\sigma_0 f=280f$. While these assumptions are perhaps as poor as any of those made in the previous f measurements, they apply here to a term which contributes less than $0.06f$ to $1-f+g(f)$ for $f>0.4$. Since the experimental f value at 77°K falls into this region, and since the systematic error in the evaluation of $g(f)$ is certainly less than about 20%, it follows that the systematic error in the experimental f value at 77°K will be of the order of 1%. This is about as large as the statistical error of counting in a typical coincidence experiment. It should be added here that the phenomena of resonant re-emission and small-angle Compton scattering, both of which can contribute in principle to the residual transmission of resonant γ rays, were examined and were found to be negligible.

Events not from Sn^{119m}

The problem of contamination of the Sn^{119m} activity by other radio isotopes is a more complex matter. Ideally, one should be able to predict the relative 25±15-keV photon-photon coincidence rate by use of published decay schemes; but since these are incomplete and not always reliable, some self-consistent checks on them were carried out as well. Decay scheme analysis shows that for the 97.15% Sn^{118} source material, the two long-lived activities which may substantially contaminate the Sn^{119m} coincidence rate are 2 year Sb^{125} and >5-year Sn^{121}. The evidence for this is summarized in Table III, in which for all possible long-lived radionuclides the relative source decay rate D and the corresponding coincidence rate N_C are given. They are evaluated at 500 days after source production, a time which corresponds roughly to the time of the experiment. D and N_C were calculated from the isotopic

TABLE III. Possible contaminants of the Sn^{119m} coincidence rate for the 97.15% Sn^{118} source.

Isotope	Stable target a[a]	σ_{nA}[b] (barns)	Radionuclide Isotope	$T_{1/2}$[c]	$D(500)$[d]$\times 10^{28}$	Coincidences ϵ[e]	$N_C(500)$[f]$\times 10^{30}$	x rays[g]
Sn^{112}	<0.0003	1.1	Sn^{113}	118 day	0.005	0.0002	<0.0001	In
Sn^{118}	0.9715	0.02	Sn^{119m}	245 day	7.7	0.04	30	Sn
Sn^{120}	0.0120	0.005	Sn^{121}	>5 year	0.01	$\leq$1.0	$\leq$1.0	Sb
Sn^{122}	0.0010	0.001	Sn^{123}	125 day	0.0002	$\leq$0.0005	$\leq$0.0001	Sb
Sn^{124}	0.0012	0.6	Sb^{125}	2 year	0.25	$\leq$1.0	$\leq$25	Te

[a] Isotopic abundance, Oak Ridge National Laboratory isotopic assay.
[b] Neutron activation cross section, *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington D. C., 1960), NRC 60-2-96, 60-3-144, 60-4-69, 60-4-87, 60-6-78.
[c] Half-life.
[d] Decay rate 500 days after irradiation in units of events per atom per incident neutron (calculated from σ_{nA} and $T_{1/2}$).
[e] Coincidence efficiency, defined as the estimated fraction of decays producing coincidences in the (25$\pm$15)-keV detector [deduced partly from *Nuclear Data Sheets*, compiled by K. Way *et al.* (Printing and Publishing Office, National Academy of Sciences—National Research Council, Washington D. C., 1960), NRC 60-2-106, 60-4-61, 60-4-78, 60-6-66, 60-6-96.
[f] Estimated total coincidence rate 500 days after irradiation in units of events per atom per incident neutron: $N_C(500)=\epsilon D(500)$.
[g] Indicates K x-ray contribution to $N_C(500)$.

content a, the neutron-activation cross section σ_{nA}, the half-life of each radionuclide, and the estimated coincidence efficiency ϵ. All possible mechanisms which, according to the decay schemes, will contribute to coincidences in the 25$\pm$15-keV detector window were considered in the estimation of ϵ. The main sources of coincidences were, as might be expected, K x rays from β decay and internal conversion. Needless to say, the estimate of ϵ was in many cases hindered by insufficient information, such as, for example, experimental values of internal conversion coefficients.

To confirm the decay scheme analysis, relative photon intensities in the 25-keV region were measured directly with a xenon-filled proportional counter. Three sources were used: A 500-day-old Sn^{118}-enriched source similar to the one used in the coincidence experiments (source A); a 500-day-old natural Sn source irradiated in the same container as the enriched source (source B); and a 7-year old natural Sn source (source C).[21] The spectra that were obtained are shown in Fig. 2. By making use of the different ages and isotopic content of the three sources, the principal features of the spectra may be roughly explained. The predominant 24–25-keV activity in source A is attributed to Sn^{119m}, and is in accord with the predictions in Table III. The 27.4- and possibly some 26.0-keV activity in source B is interpreted as K_α x rays from Te and Sb, respectively. Their intensity relative to the 24–25-keV activity is explained by the greater abundance of Sn^{124} and Sn^{120} in source B compared to source A. The absence of 24–25-keV photons in source C follows from its greater age, due to which the 245-day Sn^{119m} has decayed almost completely relative to the 2-year Sb^{125} and the >5-year Sn^{121}. The predominance of the Te x ray over the Sb x ray in source C is consistent with the fact that Sb^{125} was initially produced with about 50 times greater activity than >5-year Sn^{121}.

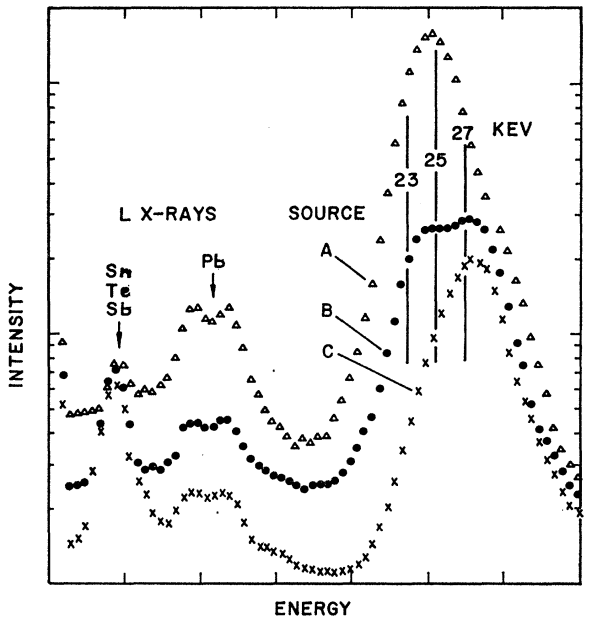

FIG. 2. Relative photon intensities in the 25-keV region are shown. The sources used were A, a 500-day-old Sn^{118} enriched source similar to the one used in the coincidence experiment; B, a 500-day-old natural Sn source; and C, a 7-year-old natural Sn source. The spectra were taken with a xenon-methane-filled proportional counter.

The crucial energy assignments in Fig. 2 were made without reliance on the linearity and proper zeroing of the analyzer, or the use of independent calibration sources. As an example of the procedure used, Fig. 3 shows the spectra obtained by placing Pd, Ag, Cd, and In absorbers (having K absorption edges of 24.34, 25.51, 26.71, and 27.93 keV) between source C and the detector. These spectra not only bracket the 27.4-keV Te x ray between 26.71 and 27.93 keV, but also yield the characteristic x rays of the absorber which, when used as built-in calibration lines, allow the determination of various peak energies to accuracies of $\pm$0.1 keV.

In addition to the general confirmation of the decay scheme picture described above, the spectra in Fig. 2 may be used to obtain estimates of ϵ for Sb^{125} and Sn^{121} without reference to anything but activation cross

[21] This was graciously supplied by the Argonne group.

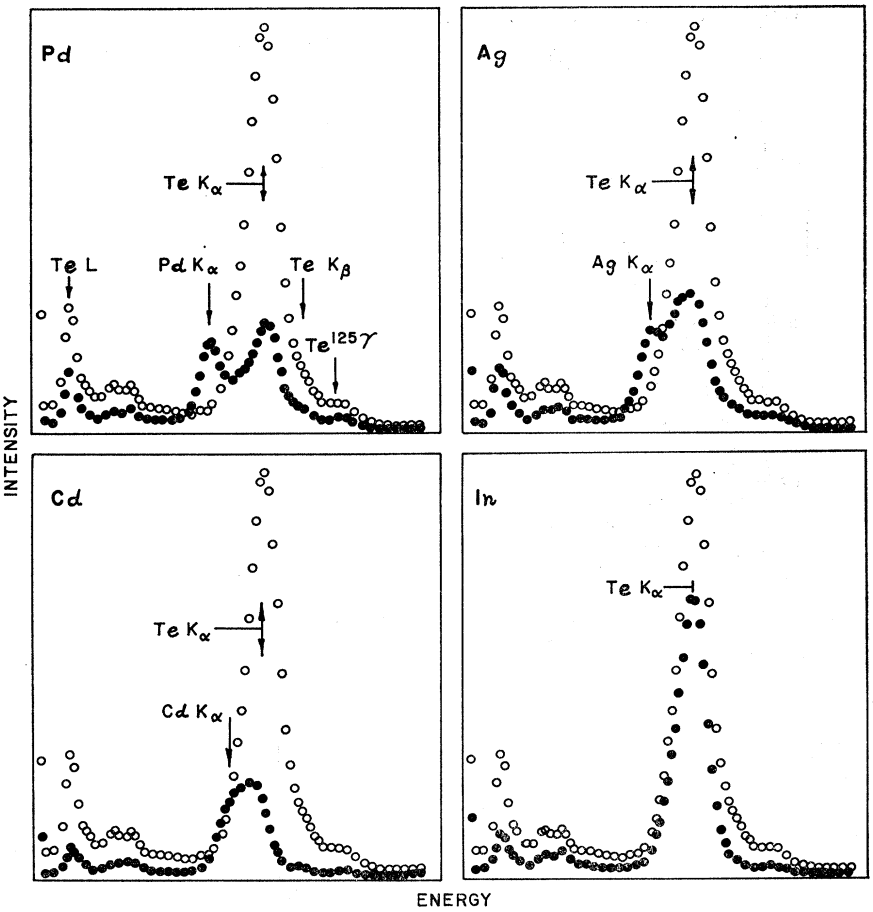

FIG. 3. The effect on the source-C spectrum (see Fig. 2) is shown when Pd, Ag, Cd, and In absorbers are placed in front of the counter. The absorbers were of roughly equal thickness (30 mg/cm²) with the exception of the Cd absorber, which was somewhat thicker (50 mg/cm²). It is, therefore, possible to attribute the acute change of absorption between Cd and In as moving from the high- to the low-energy side of the K edge, and from this one must conclude that the source-C spectrum has a main peak energy between 26.7 and 27.9 keV, the K edges of Cd and In, respectively. Because the absorbers were directly over the detector window, the characteristic K x rays of the first three absorbers are seen as well giving added confirmation to the energy determination of the C-spectrum peak. It is concluded that the C-spectrum peak must be the 27.4-keV Te K_α x ray.

sections and the decay scheme of Sn^{119m}. Since Sn^{119m} is certainly better understood than either of the other radionuclides, this constitutes a valuable independent check on the efficiency estimates in Table III. The result obtained by this argument is that for Sb^{125} and Sn^{121} ϵ is at most 0.55 and 1, respectively.[22] Both of these numbers are consistent with Table III. The conclusion drawn from this is that the Te x rays from Sb^{125} and the Sb x rays from Sn^{121} contribute about 40 and 1%, respectively, of the Sn^{119m} coincidence rate. It should be emphasized that these estimates, though better than those obtained from inspection of the decay schemes, contain sources of error that limit their accuracy to about ±30%.

[22] Since source A differs from source B only in relative isotopic abundance, the observed relative intensities for source B may be used to deduce that there are an average of 0.06 Te x rays per Sn^{119m} γ or x ray in the spectrum of source A. From this it may be shown that the decay of Sb^{125} is accompanied by an average of 1.1 Te x rays. Let δ_{125} and δ_{119} be the average number of ~25-keV photons per decay of Sb^{125} and Sn^{119}, respectively. Let $R_I=0.06$ be the intensity ratio (I_{125}/I_{119}) of ~25-keV photons for source A, and let R_D be the corresponding ratio of decay rates (D_{125}/D_{119}) obtained from Table IV. Then clearly $R_I=\delta_{125}\cdot R_D/\delta_{119}$, and if $\delta_{119}=0.58$ is taken from a recent measurement (Ref. 13), $\delta_{125}=1.1$ is obtained. But this implies that ϵ can be at most half this value, which is the desired result. A similar argument can be applied for the case of Sn^{121}.

The above estimate can be further checked by a delayed coincidence measurement, for the half-lives of the principal contaminants are short compared to the 18.5-nsec half-life of the first excited state of Sn^{119m}. Figure 4 shows the result of such a measurement using the coincidence circuit that was employed in the f measurements. The "prompt" curve is obtained with source C, and serves to define the response of the coincidence circuit at 25 keV, while the "sum" curve was obtained with source A, and shows a half-life consistent with the value reported for Sn^{119m} in the literature. A series of measurements yielded 18.3±0.5-nsec for the half-life. The sum curve does not exclude a prompt contribution. In fact, a detailed study of the two curves indicates that it contains at least 10% and no more than 30% contribution from prompt events,[23]

[23] The prompt curve was folded into an exponential decay, and to the resulting "delayed" curve various fractions of the prompt curve were added to obtain various possible observable sum curves. By appropriate choice of the exponential lifetime and the prompt fraction, a fit to the experimentally observed sum curve was achieved. In the particular case analyzed here, where the prompt slope is only a factor of 2 smaller than the delayed slope, the principal effect of adding prompt events to the delayed curve is not on the slope but the full width at half-maximum (FWHM) of the sum curve. The observed sum curve showed, in fact, a FWHM which was about 6–7% smaller than would be predicted for a pure delayed curve.

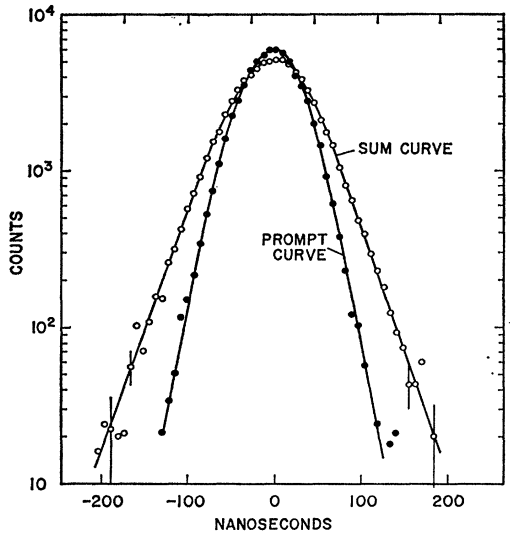

FIG. 4. Delayed coincidence spectra: The prompt curve obtained with a source C contains only Te x-ray events from Sb^{125}, and serves to define the response of the coincidence circuit at 25±15 keV. The sum curve obtained with source A contains predominantly Sn^{119m} coincidences, and shows the characteristic half-life of the first excited state, about 18 nsec. It also contains as much as 30% prompt events, as a detailed analysis of the two curves shows.

which is in good agreement with the decay scheme and proportional counter estimates.

To check on possible contributions to the coincidence rate from unfilterable In K x rays arising in the decay of Sn^{113}, a delayed coincidence spectrum from source A was obtained with Pd absorbers in both channels. The expectation is that the prompt events involving Sb and Te K x rays will be very heavily attenuated, and a prompt contribution from the unfiltered In K x rays will possibly be visible. The observed spectrum showed no such effect, but exhibited only the 18.5-nsec half-life of Sn^{119m} with much reduced intensity. Though the upper limit on Sn^{113} coincidences from this measurement is not as small as the one obtained from the decay schemes, it was possible to conclude that the Sn^{113} contribution is at most 1% of the Sn^{119m} rate.

Thus, the direct observation of half-lives confirms the predictions made earlier from decay schemes and the proportional counter intensity measurements.

With three consistent and independent estimates for the origin and magnitude of the coincidence rate contaminants, it is now easy to devise means of avoiding this contamination. A filter that removes Sb and Te K x rays, but not Sn K x rays and 23.8-keV nuclear γ rays is desired. A foil of Ag, with its K edge at 25.51 keV meets this specification. In fact, if it is placed in such a way that only a few percent of the Ag K x rays produced can enter the counters, it is easy to reduce the relative contribution of prompt coincidences by a factor of 100.

Finally, critical filtration in a coincidence experiment serves as a fourth independent way of confirming the origin and magnitude of the contaminants. The ratio $N_C(77)/N_C(373)$ was measured not only with a Ag critical absorber, but also with no absorber and a Cd absorber. The results were as follows:

Absorber	$N_C(77)/N_C(373)$	B'/C_e'
none	0.74	0.67
Cd	0.58	0.025
Ag	0.57	0

Assuming $B'=0$ for the Ag absorber, the measured values of $N_C(77)/N_C(373)$ by (14) imply the B'/C_e' values indicated. It should be noted that the B'/C_e' values apply here to the case in which the resonant absorber is in place, and not to the case of the bare source which served as the basis of the previous estimates of the coincidence rate contaminations. Hence, B'/C_e' is expected to be higher than before because, compared to the 23.8- and 25.27-keV radiation from Sn^{119m}, the 26.3- and 27.4-keV quanta from the chief contaminants will show a weaker atomic absorption in the resonant absorber. If correction is made for this effect, the above results imply a bare source coincidence contamination of 44%, which is in good agreement with all the other estimates.

Experimental Result at 77.3°K

The geometry of the experiment is shown in Fig. 5, and other important details are summarized in Table IV. The delayed coincidence spectrum was recorded continuously on a multichannel analyzer with sufficient time base to allow the simultaneous determination of the accidental and the true coincidence rate. This arrangement also served as a convenient continuous

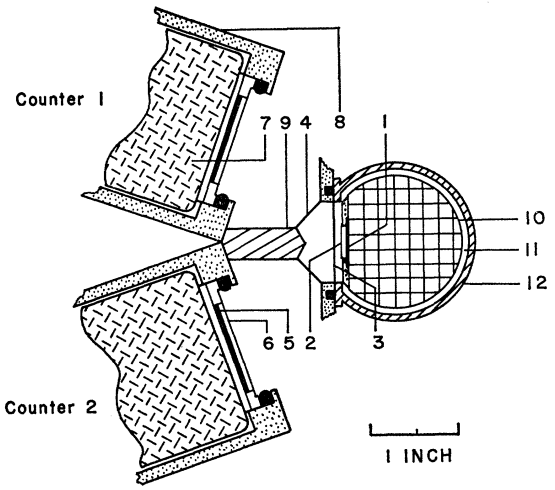

FIG. 5. Source detector geometry: (1) source-resonant absorber sandwich; (2) 0.001 in. Al radiation shield; (3) 0.005-in. Al exit window held by O-ring clamp; (4) Ag critical absorber foil; (5) 1-mm-thick NaI(Tl) crystal; (6) 0.001 in. Al entrance window; (7) 6342A photomultiplier; (8) Al and mumetal shield; (9) brass block to prevent "cross-talk" between the detectors; (10) Cu temperature-controlled block; (11) vacuum; (12) brass vacuum jacket.

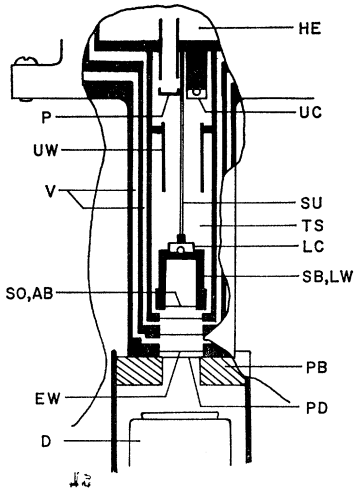

FIG. 6. Schematic cross section of the Dewar tail: The source SO and the absorber AB are shown mounted on the source block SB. On the outside of SB a Cu wire thermometer-heater LW is wound, and on the top of SB, a carbon resistance thermometer is clamped. SB is suspended by a thin inconel tube, SU inside the "thermal switch," TS, whose walls are thick Cu tubing and are an integral part of the He container HE. TS may be filled with exchange gas or alternatively pumped out through the line P. TS is surrounded by two vacuum jackets V as usual. Exit from TS through the jackets is provided by three 0.002-in. Al foils EW. The detector D is covered with a Pd critical absorber PD and a Pb collimator PB. A second Cu wire thermometer UW and a second carbon resistance thermometer UC are attached to HE to aid in liquid transfer and to allow monitoring of HE as the Dewar warms.

check on the linearity of the time base, or equivalently, the time to pulse-height converter. As a stability check on the counters, the singles rate for both counters was printed automatically every six minutes. The source strength was adjusted to give maximal statistical accuracy for the true coincidence rate in a given time interval. Typical runs lasted 24 h, with about half of the time spent at 77.3°K, about one-third the time spent at 373°K, and the remainder spent on warming and cooling and calibrations of the pulse-height analyzers. From some 12 data runs made with the final arrangement of source strength and critical absorbers, 8 were selected because they showed no difficulties in the monitored parameters. Each of these yielded an f value at 77.3°K by (15) after appropriate correction was made for the decay of the source. The results are as follows:

0.451±0.019	0.448±0.016
0.459±0.011	0.441±0.016
0.451±0.016	0.464±0.011
0.448±0.016	0.465±0.014.

The best value derived by simple averaging is 0.455 ±0.005. Since the statistical error of the average is about the same magnitude as the possible remaining systematic error, the final value that can be quoted is

$$f=0.455\pm0.010 \quad \text{at} \quad 77.3°\text{K}. \tag{17}$$

TABLE IV. Details of the coincidence experiment.

Source:	
Activity	5 μ Ci Sn^{119m} produced by 7×10^{13} neutrons/sec/cm² in 2 months.
Material	15 mg/cm², 97.15% Sn^{118}-enriched β Sn, Oak Ridge National Laboratory, Series KN, Sample 1264
Absorbers:	
Resonant	65 mg/cm² 85.7% Sn^{119}-enriched β Sn, Oak Ridge National Laboratory, Series IJ, Sample 1005(a).
Critical	55-mg/cm² Ag metal
Detectors	1-in.×1-mm NaI(Tl) crystal with 0.001-in. Al window mounted on a RCA 6342A photomultiplier; best resolution of 25-keV line was 32%; energy selection was 25±15 keV.
Coincidence resolution	"Prompt" curve with FWHM of 75 nsec; "slope" with half-life of 10 nsec; Delayed curve with FWHM of 98 nsec; slope with half-life of 18.3±0.5 nsec.

4. EXTENSION OF THE TEMPERATURE RANGE

With a single relatively precise f value, it is now possible to use simpler methods for obtaining further f values over a wide range of temperature. As before, independence of the four widths Γ_γ, Γ, Γ_S, and Γ_A and the x ray background is maintained by employing the thin source-black absorber combination in a measurement of the total transmission, but now a single counter set on 25±15 keV is used. Simplifications (a) and (c) are made as before. From (13) and (16) the following equation is written for the singles counting rate at temperature T:

$$C(T)=[1+\delta(T)]\{C_e'[1-f+g(f)]_T+B'\}. \tag{18}$$

The primes on C_e' and B' indicate that the presumed normalized function $\mathcal{T}_0(T)$ in (13) has been multiplied by an appropriate scale factor for conversion to actual counting rates. The term $\delta(T)$ is small and positive, and denotes the deviation of the atomic transmission from its value at $T=0$ due to thermal expansion [see simplification (a)]. Simplification (b), that is $f\sim0.01$ at 370°K, is also used again, and gives

$$C(370)=1.006(0.99C_e'+B'). \tag{19}$$

In place of simplification (d), the known value of f at 77.3°K is used to give

$$C(77.3)=C_e'[1-f+g(f)]_{T=77.3}+B'. \tag{20}$$

It is now possible to solve (19) and (20) for C_e' and B', and thus any set of points $C(T)$ taken in the same geometry as $C(77)$ and $C(370)$ can be converted by (18) to a corresponding set of points $f(T)$.

The advantage of a single-channel experiment is readily apparent. Relatively strong sources may be used, and a large number of counts can be accumulated in a short time. This, in turn, makes possible slow but

continuous warming of a Dewar as a device for temperature variation. For example, in a typical five-day run during which the sample was warmed from 1.3 to 370°K, $C(T)$ values at one-degree intervals were obtained each with better than 0.5% statistical accuracy. In fact, the accuracy of f values obtained from a single run is limited, as in the coincidence experiments by the systematic errors and not statistics.

Preliminary results from this experiment have been reported earlier,[24] and the apparatus and corrections applied are described in detail in the author's dissertation.[25] For the present it will suffice to say that the source-absorber holder consisted of a 65-g Cu block that could be cooled in a metal He^4 Dewar, and then thermally insulated by pumping out a gas-filled thermal switch. From this condition it could be warmed in a controlled manner by electrical heating. Above 40°K, the heater and thermometer were the same winding of fine copper wire and for lower temperatures a carbon resistor was used for temperature measurements. Using standard calibration procedures for the carbon resistor,[26] and the calibration of Dauphinee and Preston[27] for the Cu coil, temperatures were measured with 0.2-deg over-all accuracy in the entire range of $1.3<T<370$°K. Exit from the Dewar was provided by a series of three 0.002-in. Al windows, and a 50-mg/cm² Pd foil was placed between the exit windows and the detector to reduce the unwanted x ray background. The detector was of the same type as used in the coincidence experiment (see Table IV). A schematic diagram of the cryostat tail and source detector geometry is given in Fig. 6.

The counting rates were printed automatically in two channels every 100 sec, one set on 25 ± 15 keV and the other on >55 keV. The >55-keV channel served as a check on electronic stability. Except for temperatures below 40°K (where a Wheatstone bridge was used), the thermometer resistance was printed automatically via a digital voltmeter on the same cycle as the counting rates. The resulting set of simultaneous readings was examined for discontinuities and other defects, and if found trouble-free, the counting rates were summed in groups of 10 to yield a "point" every 1000 sec.

Before $f(T)$ could in fact be obtained from these counting rates and (18), corrections for source decay and temperature variation of the source-counter solid angle were applied. The source decay correction, whose total magnitude was about 5%, was experimentally determined by obtaining counting rates at fixed temperatures at several times during the run. The solid-angle variation, which was due to Dewar structure thermal expansion, was experimentally measured by recording counting rate changes as the Dewar was cooled and the source block kept at constant temperature by supplying electrical heat. The solid-angle variation was maximally a 5% effect. Since both corrections were calculable, and could thus be double checked, it can be stated that they were carried out without affecting the over-all accuracy of the results.

TABLE V. f values.[a]

T	f	T	f
2	0.725	100	0.375
4	0.717	120	0.308
6	0.716	140	0.255
8	0.715	160	0.208
10	0.715	180	0.170
15	0.715	200	0.136
20	0.696	220	0.102
30	0.660	240	0.086
40	0.617	260	0.068
50	0.572	280	0.053
60	0.529	300	0.039
70	0.486	320	0.030
80	0.446	340	0.022
90	0.410	360	0.016

[a] Estimated error is ±0.010.

In addition to runs required for the various corrections, four major data runs of about five days in length were made. Two different source foils were employed to check on possible sample-dependent effects. In three runs temperatures above 300°K were achieved, and in two runs, temperatures below 4.2°K were obtained. Each run consisted of about 300 useful points having on the order of 2×10^6 counts each. No statistically significant differences between samples were observed. Results from two runs on the same source are presented in Fig. 7 as Θ values. The scatter of the points is statistical. The dotted lines enclose the approximate range of systematic error. The low-temperature systematic error is due to the uncertainty in f at 77.3°K. The high-temperature error reflects the fact that f at 370°K was assumed and not measured. The results as f values were plotted as a function of temperature on a large piece of graph paper and French curve fitted with a smooth line. From this, the f values given in Table V were read.

5. SUMMARY AND CONCLUSIONS

(1) With the help of Tables I and V a comparison of previously reported results and the data reported here can be made. It is seen that of all the f values reported previously, those of Boyle *et al.* agree best with those of the present work. For $T<100$°K, the present f values are 10–20% higher than all previously reported values. Comparison to the theory of DeWames *et al.* shows reasonably good agreement only above 150°K. Below this value, the discrepancy between theory and the present data becomes increasingly large with decreasing temperature. (See *Note added in proof.*)

(2) In addition to Θ values derived from the Möss-

[24] C. Hohenemser, Rev. Mod. Phys. **36**, 438 (1964).
[25] C. Hohenemser, Ph.D. dissertation, Washington University, St. Louis, Missouri, 1963 (unpublished).
[26] G. K. White, *Experimental Techniques in Low-temperature Physics* (The Clarendon Press, Oxford, 1959), p. 127.
[27] T. M. Dauphinee and H. Preston-Thomas, Rev. Sci. Instr. **25**, 884 (1954).

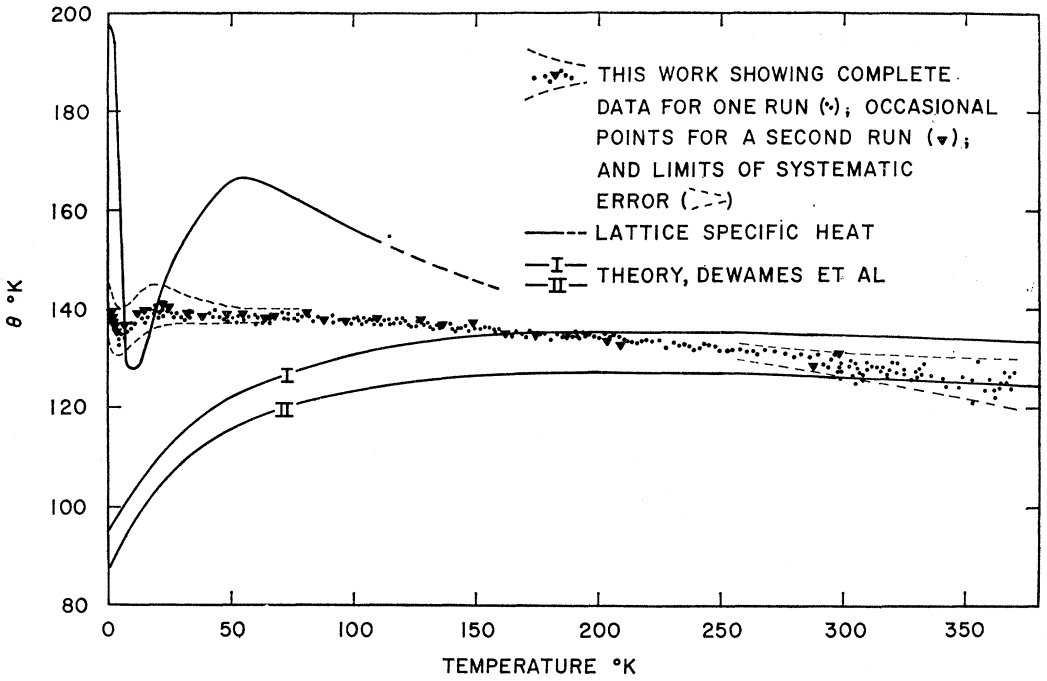

FIG. 7. Experimental results for two runs in terms of Θ values calculated at each temperature from measured f values and the Debye formula for f. For comparison, the theory of DeWames *et al.* and the Θ values derived from lattice specific heats are also indicated.

bauer effect, Fig. 7 shows Θ values derived from C_v, the lattice specific heat at constant volume. The curve that has been drawn represents an empirical fit to the data of several workers,[28–33] and has been corrected in all cases for electronic contribution to the specific heat. The most striking feature of the Mössbauer data is that the Θ values vary remarkably little with temperature, even at low temperature, where the specific-heat Θ values show large and rapid variation. This means, in effect, that the section of the phonon spectrum that is important in determining $2W$ may be (though does not have to be) closely approximated by a Debye-type function, $\rho(\nu)=\text{const}\times\nu^2$. A second general feature of the data is that the specific heat derived low-temperature limit of Θ values differs grossly from the limit of Mössbauer Θ values. Since the Debye spectrum is expected to be valid for specific heats at low temperature,[34] the behavior of the data is as clear an experimental confirmation as any that the Mössbauer effect involves a portion of the phonon spectrum that is totally different than that relevant for low-temperature specific heats. The details of the two observed sets of Θ values must, of course, be explained by the averages over the phonon spectrum[1,34] that determine $2W$ and C_v:

$$2W=\frac{2R}{3N}\int_0^\infty\frac{\rho(\nu)}{h\nu}\left[\frac{1}{2}+\frac{1}{\exp(h\nu/kT)-1}\right]d\nu. \quad (21)$$

$$C_v=\int_0^\infty\rho(\nu)\left[\frac{h\nu}{kT}\right]^2\left\{\frac{k\exp(h\nu/kT)}{[\exp(h\nu/kT)-1]^2}\right\}d\nu. \quad (22)$$

(N is the number of nuclei in the crystal.) Since the behavior of Θ values is so different for $2W$ and C_v, it seems that the two sets of data should provide a sensitive test of any proposed phonon spectrum.

(3) Another conclusion indicated by the general behavior of the present results is that there are no lattice-dependent phase transitions present to within the scatter of the data in Fig. 7. The limit obtainable for this is, in fact, substantially smaller than limits from previous reported Θ values in β-Sn, both specific heat and Mössbauer. While the absence of phase transitions bears no particular significance in this case, it is clear that f measurements of the present precision or better can be useful in phase transition studies.

(4) The results at low temperature also allow a final clarification of the finding of Wiedemann *et al.*[4] that there exists a *discontinuous* change in f, $\Delta f/f=0.4\%$, or $\Delta\Theta=2°\text{K}$, *due to* the onset of superconductivity at 3.7°K. It has already been shown by Yaqub and Hohenemser[8] that any such change, if it exists, is *continuous* and is present in the normal state below the transition as well, and hence is *not due to* superconduc-

[28] F. Lange, Z. Physik Chem. **110**, 343 (1924).
[29] W. H. Keesom and J. van den Ende, Proc. Acad. Sci. Amsterdam **35**, 143 (1932).
[30] W. H. Keesom and P. van Laer, Physica, **5**, 193 (1938).
[31] W. S. Corak and C. B. Satherthwaite, Phys. Rev. **102**, 662 (1956).
[32] Zavaritskii, Zh. Eksperim. i Teor. Fiz. **33**, 1085 (1957) [English transl.: Soviet Phys.—JETP **6**, 837 (1957)].
[33] P. H. Keesom and C. A. Bryant, Phys. Rev. **123**, 491 (1961).
[34] M. Blackman, *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1955), Vol. VII/1, p. 325.

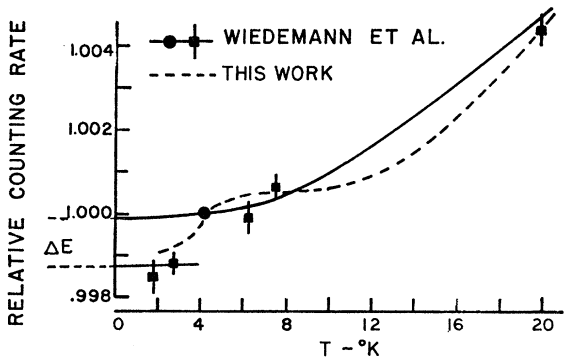

FIG. 8. The data of Wiedemann *et al.* are shown. The solid line was drawn by these workers on the assumption that $\Theta=155°K$ above the superconducting transition at 3.7°K. The dotted lines illustrate that a reasonable fit of the data is possible using the Θ values of the present work. For this fit, a value of the nonresonant background was required which differed somewhat from the value assumed implicitly by Wiedemann *et al.* It should be emphasized that the dotted line is merely illustrative, and can not take into account all possible details of the Wiedemann experiment.

tivity. The present data explicitly contradict the assumption that is essential to the derivation of the superconductivity effect; which is that $\Theta=155°K$ for the region $3.7\leq T\leq 20°K$. The data of Wiedemann *et al.* is reproduced in Fig. 8. The solid line corresponds to $\Theta=155°K$, and the discontinuity ΔE found "at" the transition is obtained from the deviation of the data from the solid curve. The error in this use of Θ is that it is not meaningfully related to f, for with a different choice of the nonresonant background rate, a different temperature dependence of Θ would be required. Wiedemann *et al.* themselves illustrate this when, in the same paper, with the same data, they find independent of their assumption about Θ, that $f(20)=0.59$, $f(4.2)=0.60$ (see Table I). This means by the Debye formula that $\Theta=105$ and 88°K at the temperatures 20 and 4.2°K, respectively. The dotted line in Fig. 8 shows a reasonable fit of the Wiedemann counting rates with the Θ values of the present paper, using a suitable choice of the nonresonant background, the exact value of which is not critical.

(5) It should be added that since the present work was undertaken, f measurements using the delayed coincidence technique combined with black resonant absorbers have been reported by Craig *et al.*[35]; and the use of black-resonant absorber alone has been decribed by Steyert and Taylor,[36] and Housley *et al.*[37,38] All these workers have investigated Fe^{57} dissolved in various host lattices, and all have used certain mixture of Fe compounds which gives a very broad absorption line, and thus allows the achievement of 97% blackness with $n_A\sigma_0 f$ values of ~ 10. It is clear that much in terms of experimental flexibility can be gained in this way, so that it becomes desirable to find a similar absorber for Sn^{119}. It should be equally clear, however, that for Sn^{119} the presence of the large nonresonant K x-ray background necessitates that reliable means of background avoidance be used in precise f measurements. The use of delayed coincidences may be the only way to achieve this.

Note added in proof. Since this paper was submitted, results by DeWames and Lehman of revised lattice dynamical calculations using elastic constants data [R. E. DeWames and G. W. Lehman, Phys. Rev. **135**, A170 (1964)] have come to the author's attention. The revised calculations yield dispersion relations as before, and also give f values for a single crystal in the x direction. DeWames and Lehman expect these to be somewhat low when compared to polycrystalline f values. The new predictions are in much better agreement with the experimental f values of the present paper. However, very recently Rowe *et al.* [J. M. Rowe, B. N. Brockhouse, and E. C. Svensson, Phys. Rev. Letters **14**, 554 (1965)], using the technique of inelastic scattering of slow neutrons, have directly measured the lattice dispersion relations of β-Sn, and have found their results to be in substantial disagreement with both the earlier and the revised dispersion relations of DeWames *et al.* This leaves the theoretical explanation of measured f values far from settled. As a next step, it would be interesting to calculate f values for the polycrystalline case using the neutron data.

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[35] P. P. Craig, O. C. Kistner, B. Mozer, and R. Segnan, Rev. Mod. Phys. **36**, 361 (1964).
[36] W. A. Steyert and R. D. Taylor, Phys. Rev. **134**, A716 (1964).
[37] R. M. Housley, N. E. Erickson, and J. G. Dash, Nucl. Instr. **27**, 29 (1964).
[38] R. M. Housley, J. G. Dash, and R. H. Nussbaum, Phys. Rev. **136**, A464 (1964).