Resonance Absorption of Gamma Rays in Normal and Superconducting Tin*

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A study of the resonant absorption of the 23.8-keV gamma rays in Sn^{119} has been made between 373 and 1.1°K by using the Sb^{119} K-capture parent. The source was prepared by bombarding natural Sn with 10-MeV deuterons in a cyclotron, and was thick enough to absorb resonantly a large portion of the recoilless photons emitted. By studying the self-absorption in this source between 373 and 60°K and taking into account the contributions to the intensity due to resonant scattering, an average value of the Debye temperature θ has been derived. In order to see if the phonon spectrum undergoes a change in passing from the superconducting to the normal phase, the counting rate was measured at several temperatures between 4.2 and 1.1°K, with and without a magnetic field strong enough to destroy the superconductivity of the source. Using the derived value of 140°K for θ, it was concluded that the change in θ between the two phases could not be more than 0.76°K.

1. INTRODUCTION

THE discovery of the isotope effect in superconductors, made independently by Maxwell[1] and by Reynolds, Serin, Wright, and Nesbitt,[2] showed conclusively that the phenomenon of superconductivity is caused by an interaction between the conduction electrons and the lattice vibrations. In their theory, which successfully explains nearly all the thermal and electrodynamic properties of superconductors, Bardeen, Cooper, and Schrieffer have shown that although an electron-phonon interaction causes the appearance of an appropriate gap in the energy spectrum of the electrons, it does not alter the vibrational modes of the lattice to an appreciable extent. This point of view has hitherto been supported by experimental evidence based on specific heat and critical field measurements; indeed, the exponential variation of the electronic specific heat in the superconducting phase was originally deduced by assuming that the lattice contribution remains the same in both phases. Recently, however, Bryant and Keesom[3] and Boorse, Hirshfeld, and Leupold[4] have published data on the low-temperature specific heat of In and Nb which seem to indicate that the lattice part of the specific heat is different in the superconducting and the normal state. A possible explanation of this is that the phonon spectrum undergoes a change. Boorse *et al.*, in an attempt to explain their results on Nb, have, in fact, suggested that the Debye temperature in the normal and superconducting state are 243 and 231°K, respectively.

Specific-heat measurements of superconductors can yield independent values of Debye θ in the normal and the superconducting state only if the electronic contribution can be accurately ascertained in each phase. While this is possible in the normal phase, where the electronic specific heat is known to be proportional to a first power in temperature, it becomes difficult in the superconducting phase, where the exponential law is known to be valid only for a limited temperature region, and where departures are known to exist for several metals at very low temperatures. These deviations can, in all probability, be attributed to the anisotropy of the energy gap,[5] but the possibility of a change in the phonon spectrum cannot be ruled out.

In order to decide this question independently, a more direct experiment can be performed in which a study is made of the change in intensity of the resonant absorption of gamma rays (Mössbauer effect) in a superconductor. Since the intensity of a Mössbauer line is determined by the well-known Debye-Waller factor, any change in Debye θ would manifest itself as a change in the intensity of this line. Lipkin[6] has shown that the high-frequency lattice modes of a crystal play a dominant role in the occurrence of the Mössbauer effect. In superconductors, the most important contribution to the relevant interaction energy comes from phonons of short wavelength. Experimentally this is evident from the fact that the transition temperature of thin films does not change much with film thickness, even if it is reduced to a few atomic layers (Shoenberg).[7] It is therefore possible that the transition affects only a small number of high-frequency modes. Such a change is not likely to influence specific-heat measurements, where the dominant contributions come from the low-frequency end of the phonon spectrum, but might be revealed as a change in the intensity of the Mössbauer line.

2. METHOD AND ITS LIMITATIONS

According to the theory of the Mössbauer effect,[8] if a nucleus of mass M, bound in a crystal lattice, emits

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[1] E. A. Maxwell, Phys. Rev. **78**, 477 (1950).
[2] C. A. Reynolds, B. Serin, W. H. Wright, and L. B. Nesbitt, Phys. Rev. **78**, 487 (1950).
[3] C. A. Bryant and P. H. Keesom, Phys. Rev. Letters **4**, 460 (1960).
[4] H. A. Boorse, A. T. Hirshfeld, and H. Leupold, Phys. Rev. Letters **5**, 246 (1960).
[5] L. N. Cooper, Phys. Rev. Letters **3**, 17 (1959).
[6] H. Lipkin, Ann. Phys. (New York) **9**, 332 (1960).
[7] D. Shoenberg, Nuovo cimento **10**, 459 (1953).
[8] J. van Kranendonk, *Proceedings of the Seventh International Conference on Low-Temperature Physics, Toronto, 1960* (University of Toronto Press, Toronto, 1961), pp. 9–20.

or absorbs a gamma ray of energy E_0, then there exists a finite probability that the transition will take place without any energy transfer to the nucleus. This probability (Debye-Waller factor) is a function of temperature T of the crystal and, in the Debye approximation of the lattice vibrations, is given by

$$f(\theta,T)=\exp\left\{-\frac{E_0^2}{2Mc^2}\frac{6}{k\theta}\left[\frac{1}{4}+\left(\frac{T}{\theta}\right)^2\int_0^{\theta/T}\frac{xdx}{e^x-1}\right]\right\}, \quad (1)$$

where k is the Boltzmann constant, c the velocity of light, and θ the Debye temperature. When the temperature $T\ll\theta$, as is the case for most superconductors, Eq. (1) can be approximated by

$$f(\theta,T)=\exp-\left(\frac{E_0^2}{2Mc^2}\frac{3}{2k\theta}\right). \quad (2)$$

Thus, if f is determined experimentally for a metal in the normal and superconducting phase, a direct comparison of their Debye temperatures can be made.

Such an experiment can be performed only on the very limited class of metals which exhibit both superconductivity and a measurable Mössbauer effect, and of this class the most simply interpretable results will be obtained from specimen in the form of a pure element. We give below a list of suitable isotopes drawn from the table by Craig[9] and their transition temperatures, T_c, as quoted by Shoenberg.[10]

Isotope	T_c—°K	Isotope	T_c—°K
Zn^{67}	0.90	Re^{187}	1.0
Sn^{119}	3.72	Hg^{199}	4.15
Hf^{177}	0.35	Th^{232}	1.4
Ta^{181}	4.40	U^{235}	0.8

Resonant absorption has not been observed so far in Hg, Th, and U. Since the experiment should be conducted over a wide range of reduced temperature T/T_c, the choice becomes even more limited because Zn, Hf, and Re have low transition temperatures and are, therefore, not suitable for study with an ordinary He^4 cryostat. There remains Sn and Ta, whose relative merits are discussed below.

The experiment can be performed either by using a source which is thick enough to remove by self-absorption a large portion of the recoilless photons, or by the more conventional method of destroying the resonance absorption with relative motion between a thin source and thin absorber of identical material. In either case a change in the counting rate C is sought when the superconductivity of the source and absorber is destroyed by a magnetic field. The two methods are not equally sensitive because the quantities measured in the two cases are slightly different, as is seen in the following.

[9] P. P. Craig, *Proceedings of the Seventh International Conference on Low-Temperature Physics, Toronto, 1960* (University of Toronto Press, Toronto, 1961), pp. 22–35.

[10] D. Shoenberg, *Superconductivity* (Cambridge University Press, Cambridge, 1952).

Consider a beam of gamma rays having a recoilless fraction f incident on a resonant lamina of thickness x. Then the transmission of this beam is given by

$$\tau(x,f)=e^{-\mu_e x}\{1-f[1-e^{-\mu_M x/2}J_0(i\mu_M x/2)]\}, \quad (3)$$

if the recoilless portion of the incident gamma rays has an unsplit Lorentzian energy distribution. Here, μ_e is the photoelectric absorption coefficient, and $\mu_M=nf\sigma_0$, where n is the number of resonant nuclei per cc, and $\sigma_0=2\pi\lambda\!\!\!^{-2}[(2I+1)/(2I_0+1)(1+\alpha)]$ is the cross section at resonance. I is the spin of the excited state, I_0 is the spin of the ground state, $\lambda\!\!\!^{-}=\hbar c/E_0$ is the reduced wavelength of the recoilless photons, and α is the internal conversion coefficient.

In a self-absorption experiment using a thick source, the counting rate will be approximately proportional to the value of the transmission for large x:

$$C=C_0(1-f)+B, \quad (4)$$

where C_0 is the counting rate for no Mössbauer effect, and B is the temperature-independent background. Using the expression for f in Eq. (2) and differentiating Eq. (4) with respect to θ, we have the sensitivity

$$S\equiv\frac{1}{C}\frac{dC}{d\theta}=-\frac{3}{2}\frac{E_0^2}{2Mc^2}\frac{1}{k\theta^2}\frac{f}{1-f+B/C_0}. \quad (5)$$

In the moving foil experiment, assuming the foils are thin, the counting rate C_r for no relative motion is proportional to the value of the transmission for small x:

$$C_r=C_0(1-f\mu_M x/2)+B.$$

When source and absorber are set into relative motion and the resonance is completely destroyed, the counting rate is

$$C_m=C_0+B.$$

The quantity which is measured in this experiment is C_m-C_r, and therefore the sensitivity is

$$S\equiv\frac{dC_r}{(C_m-C_r)d\theta}=-\frac{3}{2}\frac{E_0^2}{2Mc^2}\frac{1}{k\theta^2}. \quad (6)$$

Substituting the appropriate values of E_0, M, θ, and f for Sn and Ta in Eqs. (5) and (6), and assuming zero background, we obtain values for the sensitivities as follows:

	Sensitivity $(°K)^{-1}$	
Metal	Self-absorption	Relative motion
Sn	-4.4×10^{-3}	-2.4×10^{-3}
Ta	-1.7×10^{-4}	-3.4×10^{-2}

We, therefore, decided to use the 23.8-keV Mössbauer line in Sn and perform the experiment by the self-absorption method. The self-absorbing source, details of which are given below, was cooled below the transition temperature by means of liquid He^4 and several measurements were made between 3.72 and 1.1°K. At each

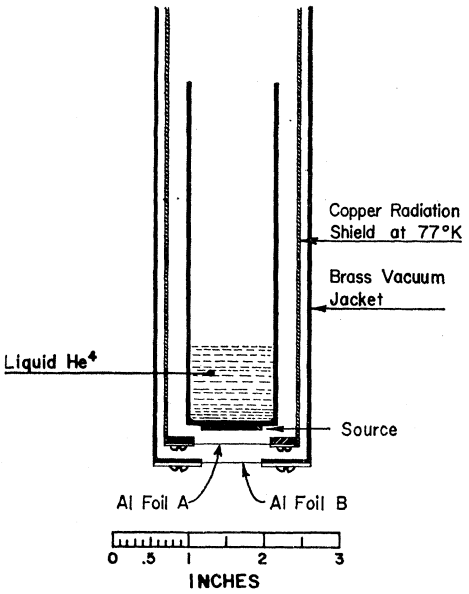

FIG. 1. Cryostat tail.

temperature measurements were repeated by applying a magnetic field of 450 G to destroy the superconductivity. The transition temperature was determined in a separate experiment in which the susceptibility of the metal from which the source was prepared was measured as a function of temperature.

The half-life of the specimen was so short that the time taken to measure its susceptibility before conducting the absorption experiment would have reduced its strength considerably. In addition, the back surface of the bombarded specimen was contaminated with soft solder which becomes a superconductor at a higher temperature than Sn. Its presence, which would have interfered with the determination of the transition temperature, could not be successfully eliminated from the highly radioactive source because of handling difficulties.

However, an estimate of the total number of impurities introduced into the metal by the deuteron bombardment clearly indicated that they were too small to influence the superconductive properties of the source. It was therefore considered safe to assume that the transition temperature of the source would not be appreciably different from that of the pure metal.

The self-absorption experiment, besides being more sensitive in Sn, is attractive also for other reasons. It obviates the necessity of preparing thin films whose superconducting properties may differ from those of the bulk metal. It also avoids the technical difficulties involved in producing relative motion between foils at He^4 temperatures. Any differences which might exist in the physical properties of the two foils can cause spurious effects; these can be avoided in a self-absorbing source where the process of emission and absorption takes place in identical surroundings. In regard to future experiments, it is important to note that the self-absorption method is capable of a wide range of applications. Unlike the moving-foil experiments, it may be used for cases where the lifetime is short and the linewidth too broad to permit reliable mechanical motion.

It must, finally, be pointed out that the sensitivity actually achieved in our experiment was -2.48×10^{-3} per °K. This was obtained by substituting the value of B/C_0 and θ obtained from our data. Background can be largely eliminated by use of coincidence techniques. In Sn, one may employ the gamma-ray parent of the 23.8-keV line which provides a 250-day half-life in a prior gamma ray of 65 keV. Our source utilized the K-capture parent Sb^{119} which directly produces the 23.8-keV state, and, since the accompanying x rays cannot be resolved, precludes the use of coincidence techniques.

3. THE SOURCE AND THE APPARATUS

The source finally adopted for the experiment was a rectangular piece of natural Sn (22.5%Sn^{118}) 1×4×0.1 cm in size. It was bombarded in the Washington University cyclotron with 10-MeV deuterons for 8 h, producing about 150 μC of the required 23.8-keV activity in a total activity of about 1 mC. The primary mechanism of production is given by $Sn^{118}(d,n)Sb^{119}$, followed by K capture (38 h) to the 1.9×10^{-8}-sec level of Sn^{119}. The source was filtered with the Pd K edge in order to reduce the intensity of the 25-keV x rays caused by the K capture and some higher energy nuclear

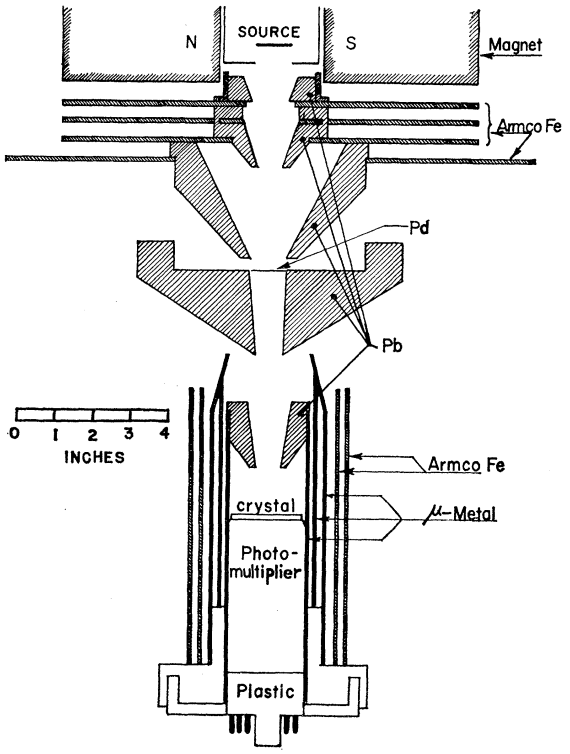

FIG. 2. Experimental counting geometry.

gamma rays. A thickness of 0.0076 cm reduced the relative intensity of these x rays from 85 to 17%.

The cryostat, whose tail is shown in Fig. 1, consisted of a copper He^4 vessel surrounded by a radiation shield at liquid N_2 temperature. The source was soldered to the bottom of this vessel and the gamma rays were made to emerge through two Al foils A and B of thickness 0.0025 and 0.005 cm, respectively. A acted as a radiation shield and was maintained at 77°K, while B formed a part of the outer vacuum jacket. The temperature of the source was determined from the vapor pressure of the He^4 in the cryostat using the T_{55E} scale.[11]

The geometry used for counting can be seen in Fig. 2. A solid angle of about 0.001 sr is subtended by the source at the scintillation counter, giving a good definition of the forward beam. The Pb collimators were essential for obtaining a well-resolved peak in the photomultiplier pulse-height spectrum, and could be manipulated to produce an essentially energy independent background rate on either side of the 23.8-keV maximum. The spectrum obtained by means of the 6342A photomultiplier tube, with the Pd foil and collimators in position, is shown in Fig. 3. In order to suppress the superconductivity of the Sn source, a horseshoe-type electromagnet NS was employed as shown. Shielding of the phototube by means of mu metal and Armco iron was necessary to eliminate the effect of the stray magnetic field, and with the arrangement shown in Fig. 2, the change in gain was less than 3 parts in 10^5 when the field was switched on and off.

The counting rate was measured as a function of temperature with a single-channel differential pulse-height analyzer. The energy window was made sufficiently wide to include some of the flat background on either side of the peak shown in Fig. 3. With the customary precautions, the system was made unusually

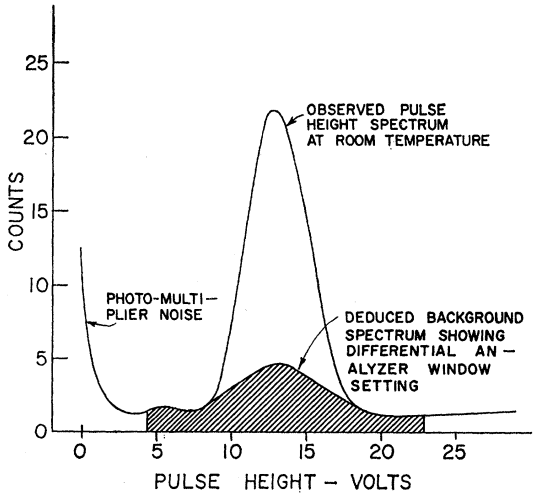

FIG. 3. Pulse-height spectrum for the 23.8-keV gamma rays.

[11] J. R. Clement, J. K. Logan, and J. Gaffney, Phys. Rev. **100**, 743 (1955).

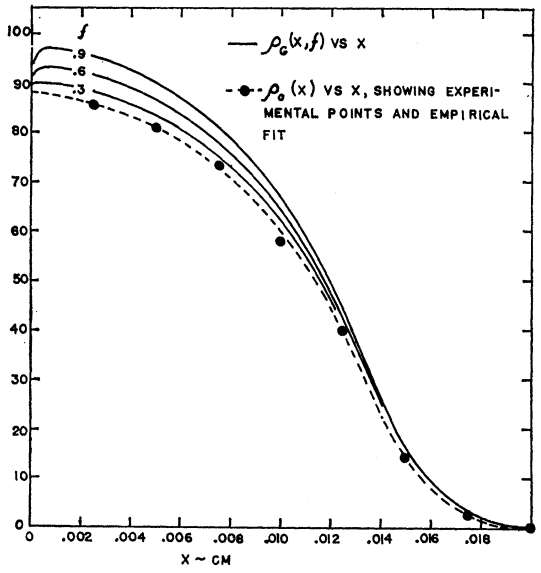

FIG. 4. Source density functions.

stable with respect to change in gain caused by ordinary power fluctuations. Although on account of the rapid decay of the source a high counting rate was desirable, it was kept at a level where pile-up corrections were small.

4. DETERMINATION OF DEBYE θ

In order to obtain a quantitative estimate of a possible change in Debye θ, a knowledge of its value based on the Mössbauer effect was desired. This was obtained from a measurement of the counting rate in the interval $4.2°K \leq T \leq 373°K$. A description of the analysis necessary to interpret the data obtained follows.

The expression for the transmission in Eq. (3) may be used to express the counting rate observed from a source having a resonant fraction f in the case where the source subtends a negligible solid angle at the detector. If $\rho_G(x,f)$ is the generalized density distribution of gamma-emitting nuclei in the source, the total counting rate for an infinitely thick source as a function of f, and by Eq. (1), as a function of T and θ will be given by

$$C(f)=C(T,\theta)=\Omega\int_0^\infty \tau(x,f)\rho_G(x,f)dx, \qquad (7)$$

where Ω is the solid angle subtended by the source at the detector. The generalized density ρ_G is defined exactly in the Appendix. It may be thought of roughly as the sum of density ρ_0, describing the distribution of gamma-emitting nuclei produced by the deuteron bombardment, and the enhancement of this density due to multiple resonant scattering in the source. ρ_G can be calculated if ρ_0 is known.

ρ_0 was found in a separate experiment at room temperature. A stack of 16 foils of natural Sn, each 0.0025 cm thick, was bombarded with the same deuteron beam used in making the source. The radioactivity of each foil was then analyzed by the same differential analyzer operating under conditions of the original experiment. The counting rate was measured first with and then without the Pd filter. Because for each foil the ratio of the two counting rates was approximately constant, it was concluded that the relative density distribution of the 23.8-keV activity for every foil would be given by its counting rate relative to the first foil. The effect of multiple resonant scattering was considered negligible since it is about 1% at room temperature, and affects every foil in the same way.

Figure 4 shows the experimental points for ρ_0, and a smooth curve fitted to them on a scale such that $\Omega\int_0^\infty \rho_0(x)dx=1$. The results of the calculations of ρ_G are also illustrated for three values of f. From this we may conclude that the enhancement of the source density due to multiple scattering is not large. In Fig. 5, the results of the integration of Eq. (7) are illustrated, and again the effect of multiple resonant scattering is seen to be small, though not negligible.

For the purpose of presenting the experimental data for the counting rate independent of the background, the function

$$N(T,\theta)\equiv\frac{C(\infty,\theta)-C(T,\theta)}{C(\infty,\theta)-C(0,\theta)}$$

was calculated and plotted against T for several values of θ in Fig. 6. Experimental values of $N(T,\theta)$ were calculated from the experimental counting rates, and superposed on the plot. $C(\infty,\theta)$ and $C(0,\theta)$ were obtained by extrapolating the experimental counting rate

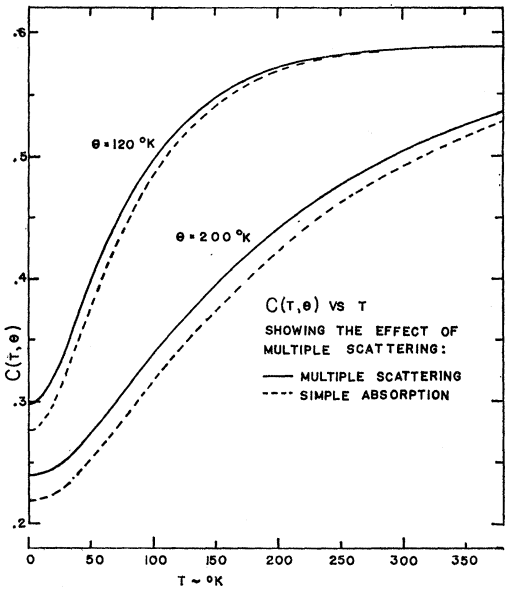

FIG. 5. Calculated 23.8-keV counting rate.

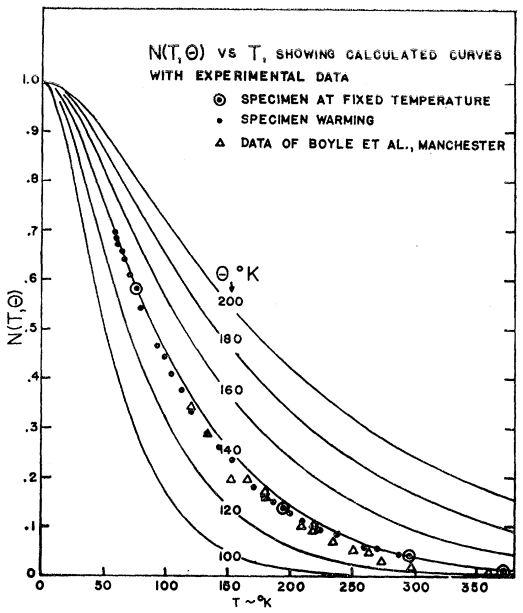

FIG. 6. Normalized counting rates with data above 60°K.

curve to obtain the high-temperature asymptote and the low-temperature limit, respectively.

In order to take into account the decay of the source, three lifetimes of the background (2.8, 5.8, 60 days) in addition to the 38.1-hour half-life of the K-capture parent had to be considered. Our normalization was, therefore, obtained by using an effective decay constant which was fitted to an empirical curve of counting rate against time, with the temperature constant, for a period much longer than the duration of the experiment. It was easy to ascertain that the errors introduced by normalizing in this way were less than the statistical errors of the counting.

It should be recalled that the analysis of the data has been based on the assumption of a Lorentzian line shape. If the line is quadrupole split by an amount larger than the natural linewidth, the apparent value of θ will change. Experimental evidence on the question of quadrupole splitting is not conclusive. Boyle *et al.*,[12] Hanna *et al.*,[13] and Wiedemann *et al.*[14] have reported the absence of quadrupole splitting, while Delyagin *et al.*[15,16] have demonstrated quadrupole splitting and its variation with temperature. It is interesting that the present data agree remarkably well with those of Boyle *et al.*,[17] as has been indicated in Fig. 6.

[12] A. J. F. Boyle, D. St. P. Bunbury, and C. Edwards, Proc. Phys. Soc. (London) **77**, 1062 (1961).
[13] S. S. Hanna, J. Heberle, G. J. Perlow, R. S. Preston, and D. H. Vincent, Phys. Rev. **120**, 2211 (1960).
[14] W. H. Wiedemann, P. Kienle, and F. Pobell, Z. Physik **165**, 109 (1961).
[15] N. N. Delyagin, V. S. Shpinel, V. A. Bryukhanov, and B. Zhvenglinskii, Soviet Phys.—JETP **12**, 159 (1961).
[16] N. N. Delyagin, V. S. Shpinel, V. A. Gryukhanov, Zhur. Eksp. i Teoret. Fiz. **41**, 1347 (1961).
[17] A. J. F. Boyle, D. St. P. Bunbury, C. Edwards, and H. E. Hall, Phys. Soc. (London) **77**, 129 (1961).

If one can assume no significant quadrupole splitting, it should be noted that the experiment of Boyle *et al.* and the present experiment give a value of θ considerably different from that obtained by specific heat measurements. As the calculated cross section for the Mössbauer effect is sensitive to the shape of the phonon spectrum[18] only for moderate and high energies, and since this region does not play a dominant role in low-temperature specific-heat determinations, the observed differences should not be surprising.

For the main purpose of this paper, namely, the superconducting experiment, the actual value of θ is relatively unimportant. In estimating the upper limit on a possible change in θ in the superconducting transition, the value of 140°K was used.

5. DETERMINATION OF θ FOR SUPERCONDUCTING TEMPERATURES

The counting rate measured as a function of temperature from 4.2 to 1.1°K has been plotted in Fig. 7, with an without the magnetic field. In order to show the temperature at which the specimen becomes superconducting, the susceptibility measurements are also indicated on an arbitrary scale.

The continuous curve in Fig. 7 has been calculated to show how the counting rate should change as a function of temperature if θ remains constant at 140°K. In calculating this curve, the value of B/C_0 used was 0.39. This could be computed by using the normalized counting rates for other temperatures.

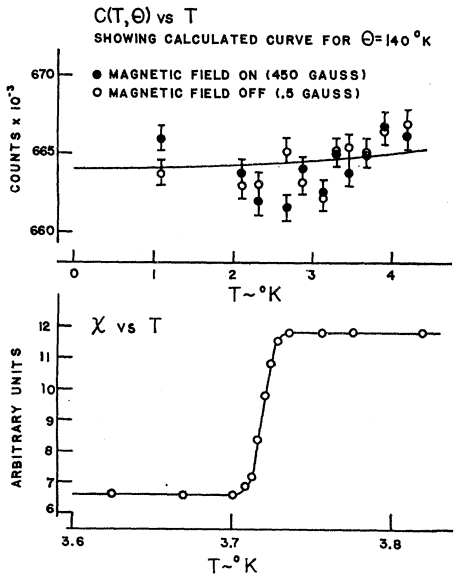

FIG. 7. Top: Counting-rate data in the superconducting region, showing the effect of applying a magnetic field large enough to destroy superconductivity. Bottom: Magnetic susceptibility in the superconducting region.

[18] W. S. Corak and C. B. Satterthwaite, Phys. Rev. **102**, 662 (1956).

As a cursory examination of the plot shows, no clearly defined differences appear between pairs of points taken at the same temperature in the normal and superconducting state. Since one does not know what kind of differences to expect, it is somewhat hazardous to deduce a maximum possible difference $\Delta\theta$ in θ between the two states. In order to make some sort of numerical estimate, however, we define $\Delta\theta$ explicitly in terms of the sensitivity S given in Eq. (5):

$$\Delta\theta \equiv |1/S|\,(\Delta C/C),$$

where $(\Delta C/C)$ is the maximum fractional difference in counting rate between the two states. One rather conservative estimate of this quantity is its average value for the set of eight pairs of points in the normal and superconducting state. Using the value given earlier for the experimental sensitivity, we have

$$\Delta\theta_{\rm av}=0.76°\text{K}.$$

A more realistic estimate would use the root mean squared value of $(\Delta C/C)$ for the eight pairs of points. This yields

$$\Delta\theta_{\rm rms}=0.33°\text{K}.$$

A conclusion which might possibly be drawn from our results is that θ does not remain constant between 4.2 and 1.1°K. A continuous increase of about 2°K, independent of superconductivity, would be consistent with the data.

Finally, it is interesting to compare the present results with those of Wiedemann *et al.*,[14] who interpret their measurements to indicate a discontinuous increase of the Debye-Waller factor of 0.4%, or, using their value of $\theta=155$°K, a discontinuous increase in θ of 2.15°K, as the sample becomes superconducting. Since they did not take measurements in the normal state below the transition, it can not be concluded that their two points below the transition would in the normal state fall back on the curve appropriate for the region above the transition. In fact, since their data are open to an interpretation of a continuous change in θ independent of superconductivity, they are not inconsistent with those of the present experiment.

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APPENDIX. ANALYSIS OF MULTIPLE RESONANT SCATTERING

An expression is desired for the counting rate $C(T,\theta)$ in a geometry where the source subtends a negligible

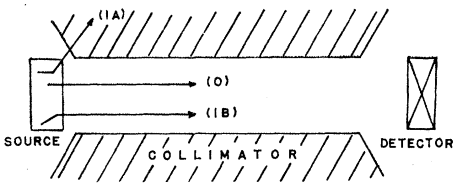

FIG. 8. Classification of events for the multiple scattering analysis.

solid angle at the detector, and where all significant orders of resonant scattering are taken into account. An unsplit Lorentzian energy distribution peaking at E_0 is assumed for the emission and absorption spectra. All nonresonant scattering is ignored.

Figure 8 illustrates the case under consideration. Clearly, a certain portion of the recoilless emissions in the direction of the detector are *scattered out* of the forward beam, while, on the other hand, some recoilless emissions not directed in the forward direction are *scattered into* the forward beam. These two types of events, illustrated by (1A) and (1B) in Fig. 8, and events involving no scattering or absorption in the forward direction, illustrated by (0) in Fig. 8, constitute all types of possible events. We shall refer to the latter as zeroth order events. The former we shall call first order if they originate after a first resonant reabsorption, second order if they originated after a second reabsorption, and so forth. The counting rate at the detector will consist of zeroth order events, first-order events of type (1B), plus higher-order events whose path just prior to exiting the source is equivalent to (1B). It is clear, therefore, that one method of analysis will consist of a series of terms corresponding to successively increasing orders of events, and it is this approach which will be used in the following.

To put the above description into mathematical language, we define the generalized source density as

$$\rho_G(x,f)\equiv\rho_0(x)+\sum_{j=1}^{\infty}\rho_j(x,f), \tag{A1}$$

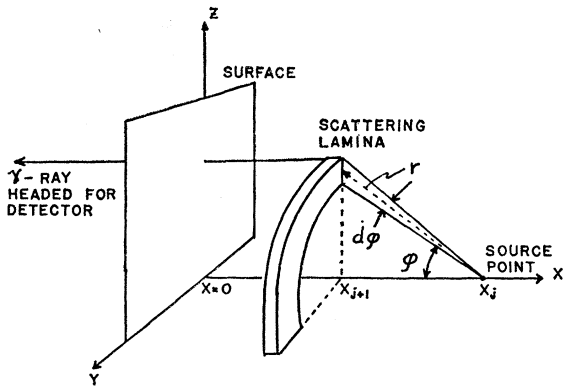

FIG. 9. Internal source geometry defining variables for multiple scattering analysis.

where ρ_j is the density of jth order scattering events. We then require integration according to Eq. (7) of the main text:

$$C(f)=\Omega\int_0^{\infty}\rho_G(x,f)\tau(x,f)dx. \tag{A2}$$

Each ρ_{j+1} is found by an appropriate integral operator L working on ρ_j. Thus, schematically,

$$\rho_{j+1}=L\rho_j=L^j\rho_0;\quad L^0\equiv 1.$$

To find L, it is assumed that the mean free path of the Mössbauer radiation is much less than the area dimensions of the plate-shaped source. This allows the treatment of every point in any particular lamina of the source in the same way as every other point in the lamina. Consider now an emission of order j into an arbitrary direction, resulting in a subsequent resonant reabsorption and gamma-reemission of order $j+1$. As shown in Fig. 9, let x_j define the emission lamina, φ the angle of emission, and x_{j+1} the resonant scattering lamina. Then the increment of $(j+1)$-order resonant scattering centers in the scattering lamina will be given by the following two integrals representing the forward and backward directions:

$$\rho_{j+1}(x_{j+1},f)dx_{j+1}=\int_{x_j=0}^{x_{j+1}-\epsilon}\int_{\varphi=0}^{\pi/2-\delta} f\frac{\rho_j(x_j,f)dx_j}{2}\sin\varphi d\varphi P(r)dr\,\frac{1}{1+\alpha}$$
$$+\int_{x_j=x_{j+1}+\epsilon}^{\infty}\int_{\varphi=\pi/2+\delta}^{\pi} f\frac{\rho_j(x_j,f)dx_j}{2}\sin\varphi d\varphi P(r)dr\,\frac{1}{1+\alpha}, \tag{A3}$$

where

$$r=|(x_{j+1}-x_j)/\cos\varphi|,\quad dr=(dr/dx_{j+1})dx_{j+1}.$$

The terms in the respective integrands may be characterized as follows:

$f\rho_j(x_j,f)dx_j/2$ = recoilless component of the jth order emissions into the forward or backward directions;

$\sin\varphi d\varphi$ = fraction of this radiation going into the direction $(\varphi,\varphi+d\varphi)$;

$P(r)dr$ = probability that this fraction is resonantly reabsorbed in the lamina $(x_{j+1},x_{j+1}+dx_{j+1})$;

$1/(1+\alpha)$ = fraction of subsequent de-excitations that go via gamma emissions.

ϵ and δ are small positive numbers, and serve to indicate

that the integrand is undefined if either is allowed to vanish. The function $P(r)$ is the derivative of the resonant absorption multiplied by the photoelectric attenuation factor:

$$P(r)=e^{-\mu_e r}(d/dr)[1-e^{-\mu_M r/2}J_0(i\mu_M r/2)]$$
$$=(\mu_M/2)e^{-(\mu_e+\mu_M/2)r}[J_0(i\mu_M r/2)+iJ_1(i\mu_M r/2)].$$

The integral operator may be expressed in terms of its kernel $K(\Delta x,f)$ by separating the steps of the integration in Eq. (A3). We, therefore define

$$K(\Delta x,f)=sf^2\int_{\varphi=0}^{\pi/2-\delta}\tan\varphi\left\{\exp\left[-\frac{\Delta x}{\cos\varphi}(\mu_e+\beta)\right]\right\}$$
$$\times\left[J_0\left(\frac{i\beta\Delta x}{\cos\varphi}\right)+iJ_1\left(\frac{i\beta\Delta x}{\cos\varphi}\right)\right]d\varphi, \quad \text{(A4)}$$

where

$$s\equiv n\pi(\hbar c/E_0)^2[(2I+1)/(2I_0+1)],$$
$$\beta=sf(1+\alpha),$$
$$\Delta x_j=|x_{j+1}-x_j|.$$

With this, the $(j+1)$-order density function is given by

$$\rho_{j+1}(x_{j+1},f)=\int_0^{x_{j+1}-\epsilon}\frac{\rho_j(x_j)}{2}K(\Delta x_j,f)dx_j$$
$$+\int_{x_{j+1}+\epsilon}^{\infty}\frac{\rho_j(x_j)}{2}K(\Delta x_j,f)dx_j. \quad \text{(A5)}$$

For the case of Sn^{119} the IBM-704 was programmed to calculate the function $K(\Delta x,f)$ for a grid of Δx and several values of f; then to carry out successive iterations according to Eq. (A5), starting with $\rho_0(x)$, the primary density function in the integrand; then to sum the results to obtain $\rho_G(x,f)$ according to Eq. (A1); and finally to calculate the counting rate as a function of f as given in Eq. (A2). The usefulness of this analysis is dependent on the rapidity of convergence. This in turn depends strongly on f and α. For the case of Sn^{119}, because of the relatively large value of $\alpha=6.3\pm0.4$, convergence of the series for $\rho_G(x,f)$ was rapid for all f. Figure 10 illustrates a particular case, $f=0.7$, by indicating on a semilogarithmic scale the first three terms in the series for $\rho_G(x,f)$.

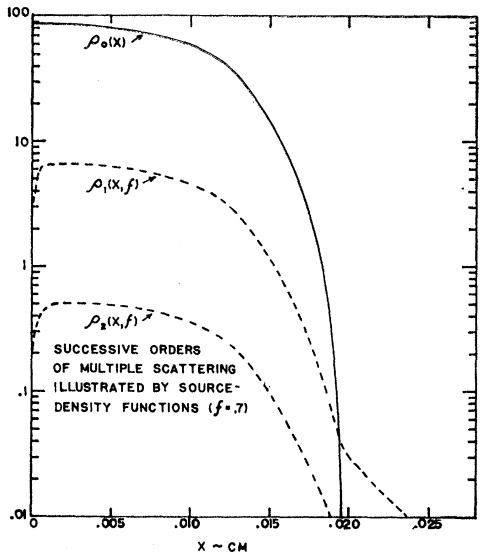

FIG. 10. Illustration of the rapid convergence of the multiple scattering analysis for Sn^{119}.