Figure 9 shows the polarization of neutrons scattered from helium in the direction  $\varphi = \vartheta = 90^{\circ}$ , as a function of incident neutron energy. Polarizations were calculated from Eq. (6) and Eq. (7) using the phase shifts which described the  $n-\alpha$  total cross section and the angular distribution. These polarizations should be quantitatively reliable below 2 Mev. At higher energies the uncertainties in the phase shifts and the influence of *D*-waves make the results of only qualitative value. It may also be possible to use helium to analyze the polarization of fast neutrons, since the scattering of a polarized beam of neutrons from helium will show a left right asymmetry equal to (1+P'P)/(1-P'P), where P' is the polarization of the incoming beam.

I wish to thank Professor H. H. Barschall for his advice on the experimental techniques involved in this work. I also wish to express my appreciation to Professor P. Huber for discussion of his measurements.

PHYSICAL REVIEW

#### VOLUME 86, NUMBER 2

APRIL 15, 1952

# The Isotope Effect in Superconductivity. II. Tin and Lead\*

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The critical magnetic fields, H, of various isotopic mixtures of tin and of lead have been measured as a function of temperature, T.

For tin, the critical temperature  $T_c$  at zero field, is related to the average mass number, M, by the relation  $M^{0.46}T_c = \text{const.}$  The critical magnetic fields,  $H_0$ , at absolute zero are proportional to the critical temperatures. The normalized values of critical field,  $H/H_0$ , are the same function of the variable  $T/T_c$  for all the isotopes. An analytic expression for this function giving the best fit to the experimental data is given. The measurements on lead were made in the temperature range 1.6°K to 4.2°K. The data clearly indicate that the isotope effect is present in this superconductor.

### 1. INTRODUCTION

HIS investigation was a continuation of our work on the dependence of superconducting properties on isotopic mass which was originally discovered in mercury. The details of experimental technique and a bibliography are contained in our first paper,<sup>1</sup> which will be referred to as I. The present work was briefly reported at low temperature conferences<sup>2</sup> and is in

TABLE I. Critical field data for tin. The critical temperatures  $T_c$ , the critical fields at  $0^{\circ}$ K,  $H_o$ , and the slope of the critical field-temperature curves at  $T_c$ ,  $(dH/dT)_{T_c}$ , are tabulated as a function of the average mass number, M. The purities and  $H_0/T_c$  are also listed.

М	Purity (% Sn)	<i>T</i> ¢(°K)	H <sub>0</sub> (oersteds)	$H_0/T_c$	(dH/dT) <sub>T</sub> (oersteds/ °K)
113.6	99.50	3.805	312	82.0	145
118.7	99.995	3.752	304	81.5	144
123.8	99.76	3.659	298	81.4	144

\* This work has been supported by the joint program of the ONR and AEC, the Rutgers University Research Council, and the Radio Corporation of America.

<sup>†</sup> The parts of this paper dealing with the measurements on lead are based on an M.S. thesis submitted by C. Lohman to the Graduate Faculty of Rutgers University. <sup>1</sup> Reynolds, Serin, and Nesbitt, Phys. Rev. 84, 691 (1951).

<sup>2</sup> Serin, Reynolds, and Nesbitt, Proceedings of the Low Temperature Symposium (National Bureau of Standards, March 27 to 29, 1951); C. A. Reynolds and B. Serin, Proceedings of the Inter-national Conference on Low Temperature Physics (Oxford, England, August 22 to 28, 1951).

general agreement with the work of Lock, Pippard, Shoenberg,<sup>3</sup> Maxwell,<sup>4</sup> and Olsen-Bär.<sup>5</sup>

## 2. MEASUREMENTS ON TIN

### (a) Samples

The two samples of tin metal which had isotope distributions differing from the distribution occurring in nature were obtained from the AEC.6 The natural metal (M=118.7) was obtained from the Johnson, Matthey Company. The purities of the samples are listed in Table I.

The metal was cast under vacuum into thin-walled glass capillary tubes. The tin samples were about 0.8 mm in diameter and about 4 cm long. The wall thickness of the capillary tubing was about 0.2 mm.

### (b) Experimental Results

The techniques used to determine the critical magnetic fields as a function of temperature were identical with those described in I. In addition to the two isotope samples, measurements were made on two samples of natural tin. The results for these two samples were the

<sup>4</sup> E. Maxwell, Proceedings of the International Conference on Low Temperature Physics (Oxford, England, August 22 to 28, 1951). <sup>5</sup> M. Olsen-Bär, Nature 168, 245 (1951).

<sup>&</sup>lt;sup>3</sup> Lock, Pippard, and Shoenberg, Proc. Cambridge Phil. Soc. 47, 811 (1951).

<sup>&</sup>lt;sup>6</sup> The isotopes were produced by Carbide and Chemical Division, Oak Ridge National Laboratory, Y-12 Area, Oak Ridge, Tennessee, and were obtained on allocation.



FIG. 1. Curves of detected signal vs current in the Helmholtz coil for three isotopes of tin.

same over the whole range of temperature, so that we give the data only for one of the samples of natural metal.

The shape of the curves of detected signal for the three samples as a function of current through the Helmholtz coil at a given temperature are shown in Fig. 1. The critical magnetic field is given by the field corresponding to the current value at the peak of each curve. The magnetic field in oersteds equals 27.6 times the current value. The large separation and the narrowness of the peaks are noteworthy. The half-widths of all peaks are very close to the ideal width calculated assuming a discontinuous increase in susceptibility at the critical field. The more impure samples, M = 123.8 and M = 113.6 tend to have broad tails on either side of the maximum. The positions of the peaks at any temperature could be determined to an absolute accuracy of about 0.5 per cent.

The critical fields, H, were measured at about 60 values of the temperature, T, in the range 1.27°K to 3.80°K. The critical temperatures,  $T_c$ , obtained by extrapolating the critical field temperature curves to zero magnetic field are given in Table I. Below 2°K the critical magnetic fields were proportional to the square of the temperature. Hence, the critical magnetic fields at 0°K,  $H_0$ , were obtained by plotting H as a function of  $T^2$  and extrapolating to zero temperature. Values for  $H_0$ , as well as the values of the slopes of the critical field-temperature curves at the critical temperatures, are given in Table I.

The values of  $H_0/T_c$  and  $(dH/dT) \tau_c$  given in Table I are in good agreement with these reported by Lock *et al.*<sup>3</sup> for tin isotopes of different average mass numbers.

Fitting the critical temperature data to a relation of the form  $M^{\alpha}T_{c}$ =const, gives  $\alpha$ =0.46±0.02.

In agreement with Lock *et al.*,<sup>3</sup> we find that the normalized critical field  $h=H/H_0$  is the same function of the normalized temperature  $t=T/T_c$  for all the isotopes. This result is illustrated in Fig. 2, where *h* is plotted as a function of  $t^2$ . The dashed line is the extrapolation of the slope of the curve at temperatures below 2°K. The data for temperatures above 2°K clearly deviate appreciably from the parabolic behavior at low temperatures. In order to avoid confusion, less than one-third of the data have been plotted in Fig. 2.

We have 175 values of h in the interval 0.34 < t < 1.0. Because of the large number of experimental points available and the excellent consistency of the data, it seemed worthwhile to derive an analytic expression fitting the data. In order to do this simply, h was assumed to be an even function in the interval -1.0 < h < 1.0; and the function was expanded in Legendre polynomials. This method is equivalent to fitting the data by the method of least squares.<sup>7</sup> Since h was chosen to be an even function, no odd function can be present in the series expansion, so that the expansion may not converge as rapidly as the usual type of least squares expansion. We do not feel that the particular form of the expansion is of great physical significance, so that we preferred the polynomial method because of its great simplicity. The data were thus interpolated to give central differences, and the coefficients of the Legendre polynomials<sup>8</sup> were determined by numerical integration to give

$$\begin{split} h(t) = 0.652P_0 - 0.681P_2 + 0.0213P_4 \\ - 0.00372P_6 - 0.00429P_8 \\ = 1.000 - 1.083t^2 - 0.0659t^4 + 0.349t^6 - 0.216t^8, \end{split}$$

where  $P_n$  is the Legendre polynomial of order *n*. The rms deviation of the data from the expression, h(t), is 0.0025.

#### 3. MEASUREMENTS ON LEAD

# (a) Samples

The lead isotopes were obtained in the form of lead oxide from the AEC.<sup>6</sup> The oxide was reduced to lead



FIG. 2. The normalized critical magnetic fields for the various isotopes of tin are plotted as a function of the square of the normalized temperatures.

<sup>7</sup> See, e.g., A. Sommerfeld, *Partial Differential Equations of Physics* (Academic Press, Inc., New York, 1949), pp. 21–25. <sup>8</sup> We used the excellent tables of Legendre polynomials of H. J. Tallqvist, Acta Societatis Scientiarium Fennicae, NSA 2, No. 4.

 TABLE II. The average mass numbers, M, and the purities of the lead samples.

М	Purity (% Pb)		
206.15	99.88		
207.2	99.998		
207.9	99.7		

metal by heating in a hydrogen atmosphere. The metal was then cast under vacuum into thin-walled capillary tubes. Two samples were also cast from natural metal obtained from the Johnson, Matthey Company. The average mass numbers and the purities of the samples are listed in Table II. The natural metal has an average mass number of 207.2. The data for the two samples of natural metal were the same over the whole range of temperature.

### (b) Experimental Results

The critical magnetic field measurements were made in the temperature range  $1.6^{\circ}$ K to  $4.2^{\circ}$ K, so that the critical fields varied between about 550 and 760 oersteds. The Rutgers precision electromagnet<sup>9</sup> was used to provide these relatively large magnetic fields. Using this magnet, it was possible to determine, with an accuracy of 1 percent, differences of 5 oersteds between magnetic fields having magnitudes of about 500 oersteds.

The measuring techniques were the same as those described in I, except that the samples were mounted horizontally in the liquid helium Dewar flask so that the axes of the samples could be oriented parallel to the field of the electromagnet. The best orientation was determined by rotating the flask into the position that gave minimal widths for the magnetic transitions of the samples.

The half-widths of the magnetic field transitions were about 4 oersteds greater than the ideal width, probably because of the strains introduced in the samples by the tendency of lead to stick to glass. Further it was not

<sup>9</sup> Sommers, Weiss, and Halpern, Rev. Sci. Instr. 22, 612 (1951).

possible to distinguish in any consistent way at a given temperature between the critical fields of M=207.9and M=207.2, probably because of the very poor purity of the former sample. To check the effect of impurities, a sample of natural lead containing 0.2 percent tin was prepared. The tin impurity increased the critical fields at all temperatures by 1.2 oersteds relative to a sample of high purity lead. An impurity effect of this magnitude is large enough and is in the right direction to obscure the magnetic field difference expected on the basis of the isotope effect.

However, the peaks of the transitions of the samples of M=207.2 and M=206.15 were clearly and consistently resolvable. We found the critical field of M=206.15 was  $5.1\pm0.5$  oersteds greater than the critical field of M=207.2. This difference was constant over the whole range of temperature used in these experiments.

The critical field difference of 5.1 oersteds is more than twice as great as would be calculated by assuming that the critical field at 0°K is proportional to  $M^{-\frac{1}{2}}$ . This result is consistent, however, with the results of Olsen-Bär,<sup>5</sup> who on the basis of measurements of the critical temperatures, found that M and  $T_c$  were related as  $M^{0.73}T_c = \text{const.}$  Considering the larger separation of the critical temperatures, our results also lend support to the assumption that the critical magnetic fields at absolute zero are proportional to the critical temperatures.

Despite the poor quality of the data, we are convinced that the isotope effect is present in the superconductor lead, in contradiction to the results of the earlier workers.<sup>10</sup>

#### 4. ACKNOWLEDGMENT

In conclusion we wish to thank Mr. J. Teza and Mr. L. B. Nesbitt for technical assistance, and Dr. M. P. Garfunkel, Mr. W. H. Wright, and Mr. J. Gittleman for help in making the measurements.

<sup>10</sup> H. Kamerlingh Onnes and W. Tuyn, Leiden Communications, No. 160b (1922); E. Justi, Physik. Z. 42, 325 (1941).