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Ginzburg-Landau Parameter of Film Specimens*

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Results of magnetization measurements on thin superconducting films in magnetic fields transverse to the plane of the film are reported. The temperature dependence of the field, $H_\perp$, which causes a transition into the normal phase was determined for specimens of pure indium and tin as well as for alloy films containing these metals as solvents. The data are used to define a Ginzburg-Landau parameter $K_1(T)$, and are compared with various functional forms for the temperature dependence of this parameter that have been proposed. By extrapolating $K_1(T)$ to the transition temperature, the usual Ginzburg-Landau parameter K is obtained. Values of the electron mean free path are also deduced from measurements of the residual resistance ratio in the normal phase. For the indium-based specimens the dependence of K on electron mean free path is in good agreement with the theoretical expression of Gor'kov. It is concluded that no single functional form for the dependence of K_1 on temperature can describe the data. Rather, the form varies continuously as the electron mean free path changes. In particular, in the pure limit the transition field varies appreciably more rapidly than theoretical predicitions based on the microscopic theory of superconductivity.

I. INTRODUCTION

MAGNETIZATION measurements of thin superconducting films in magnetic fields transverse to the plane of the film can be used to find the field at which the transition to the normal phase occurs. The transition field $H_\perp$ is that value for which the magnetic moment vanishes. It has been shown[1,2] that $H_\perp$ is determined by the Ginzburg-Landau parameter, defined as

$$K_1(T)=H_\perp(T)/\sqrt{2}H_c(T). \tag{1}$$

$H_c(T)$ is the thermodynamic critical field, and we denote the Ginzburg-Landau parameter defined by Eq. (1) as K_1. The symbol, K, is reserved for the limiting value of K_1 as the temperature T approaches the transition temperature T_c.

As long as the Ginzburg-Landau theory applies to the experimental conditions, Eq. (1) is exact independent of the magnitude of K, that is independent of whether the superconductor is type I ($K<1/\sqrt{2}$) or type II ($K>1/\sqrt{2}$). The Ginzburg-Landau theory is applicable to any superconductor at temperatures sufficiently close to T_c. By extrapolating the data to the transition temperature, we have determined the Ginzburg-Landau parameter of pure and alloy film specimens with K values between 0.1 and 0.7. Generally speaking, values less than 0.42 are difficult to measure by other methods, since supercooling[3] occurs only in very pure specimens, and the critical field H_{c3} for the destruction of the surface superconducting state[4] exceeds H_c only in specimens for which $K>0.42$.

Gor'kov[5–7] has shown theoretically that for specimens in which the electron mean free path is l, K is given by

$$K=K_0+0.73\lambda_L(0)/l, \tag{2}$$

where

$$K_0=0.96\lambda_L(0)/\xi_0. \tag{3}$$

In these expressions, $\lambda_L(0)$ is the London penetration depth[8] and ξ_0 is the range of coherence[8] of the pure metal. Most of our measurements of K are in reasonable agreement with the values of K and K_0 determined by Eqs. (2) and (3) from independently measured values of $\lambda_L(0)$, ξ_0, and l.

Since we measure $H_\perp$ over an appreciable temperature range, we have assumed that Eq. (1) defines a value of K_1, even at temperatures far removed from T_c. This enables us to investigate the form of the temperature dependence of K_1, a subject of considerable current interest. For alloy type II superconductors it has been generally found[9] that $K_1(T)$ is in reasonable agreement with an expression derived by Gor'kov.[10] However, for relatively pure type II materials,[11,12] type I superconductors,[13,14] and strong coupling superconductors,[15] more rapid variations of $K_1(T)$ than given by the Gor'kov expression have been observed.

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TABLE I. The properties of the specimens. d = thickness, ρ_r = residual resistance ratio; T_c = transition temperature; H_0 = critical field at 0°K; K = Ginzburg-Landau parameter at T_c; $H_\perp(0)$ = transition field at 0°K.

Specimen	Nominal composition	d(Å)	ρ_r	T_c(°K)	H_0(Oe)	KH_0 (Oe)	$H_\perp(0)$ (Oe)
I−1	In	5500	0.012	3.38	280	31	72
I−2	In	2800	0.022	3.37	280	41.5	87
I−3	In	230	0.070	3.37	280	63	125
I−4	In 1.4 at.% Sn	1600	0.073	3.45	286	60	118
I−5	In 1.8 at.% Sn	670	0.075	3.51	291	72.5	143
I−6	In 3.9 at.% Sn	820	0.146	3.59	298	89	180
I−7	In 5.7 at.% Sn	560	0.222	3.78	314	137	258
I−8	In 2.5 at.% Bi	3400	0.450	3.98	330	224.5	396
S−1	Sn	2200	0.063	3.72	305	75	200
S−2	Sn 4 at.% In	2100	0.222	3.66	300	178	366
S−3	Sn 6 at.% In	2400	0.337	3.745	308	221	392

In order to exhibit clearly the change in the form of the temperature dependence among the various specimens, we have adopted a device similar in principle to the one used by Paskin *et al.*,[14] but differing in detail. We write

$$H_\perp = H_0 K f(t)\,, \tag{4}$$

where H_0 is the thermodynamic critical field at 0°K and $t = T/T_c$. If we assume that $H_c = H_0(1-t^2)$, $f(t)$ takes the following forms for the three functions for $K_1(T)$ that have been proposed:

$$f_{\text{G-L}}(t) = 2^{3/2}(1-t^2)/(1+t^2)\,; \tag{5}$$

$$f_B(t) = 2(1-t^2)/(1+t^2)^{1/2}\,; \tag{6}$$

and

$$f_G(t) = 1.77 - 2.20t^2 + 0.50t^4 - 0.07t^6\,. \tag{7}$$

The subscripts in Eqs. (5), (6), and (7), refer to the temperature dependences proposed respectively, by Ginzburg and Landau,[16] Bardeen,[17] and Gor'kov.[10] We take the measured values of $H_\perp(t)$ and at each t form the product $(f^{-1}H_\perp)$. If any one of the foregoing functions describe the data exactly, we expect this product to be independent of t. However, regardless of the fit, as $t \to 1$, this product should approach the same value of KH_0 for all three functions. We can therefore deduce a value for KH_0, by extrapolating the curves of $(f^{-1}H_\perp)$ to the transition temperature. Even though we have assumed a parabolic temperature dependence for $H_c(t)$ in deriving (5), (6), and (7), we can adopt the point of view that they are arbitrary functions to which we are applying an empirical test as to the goodness of fit to the data. We shall show that among our specimens, there is at least one for which each of the foregoing functions seems to apply best.

II. EXPERIMENTAL

A. Specimens

Specimens were prepared by evaporating films on to polished aluminum substrates $\frac{1}{32}$ in. thick. The use of aluminum reduced the stresses on the films[18,19] on cooling to liquid-helium temperatures because of differential contractions between film and substrate. Polishing resulted in a thick oxide layer on the substrate, and there was no evidence of a proximity effect on the transition temperature of the film[20] due to the normal conducting aluminum. After evaporation, a disk $\frac{3}{16}$ in. in diameter was punched out of the center of the substrate, in order to avoid effects associated with the thin edges which appear on films which have been masked.[21] The disk specimens were used for the magnetization measurements.

Nominally pure films were prepared by placing an appropriate charge of metal in molybdenum boats which were heated electrically. During evaporations the pressure in the vacuum system was 2×10^{-6} Torr or better, and the substrates were at room temperature. Alloy films were prepared by dropping many small pellets of the alloy on to the hot boat and evaporating each pellet to completion. A pellet produced a film about 100 Å thick. This procedure was used in an attempt to insure that the specimens were of reasonably homogeneous composition.

During each evaporation films were simultaneously deposited on to glass microscope slides having four electrical terminals painted on them with silver paint. These specimens on glass were used to determine the electrical resistivity of the films by the standard potentiometric procedure. Film thickness was calculated from the room-temperature resistance (corrected for impurity resistance), the geometry of the specimen, and the known resistivity of the pure metal. We estimate that the thickness was determined to a precision of 10%. The resistance ratio, $\rho_r = R(4.2°\text{K})/[R(295°\text{K}) - R(4.2°\text{K})]$, where R is the resistance, was also determined for each film. $R(4.2°\text{K})$ was measured in a large enough magnetic field to insure that the film was completely in the normal conducting phase. Since $R(4.2°\text{K})$ is in the residual-resistance range of the specimens, ρ_r is determined by impurity and boundary scattering, and is inversely proportional to the electron mean free path in the specimens. Because in the alloys films $R(4.2°\text{K})$ in primarily determined by impurity scattering, and even in the pure films there is appreciable boundary and defect scattering, any strains introduced in the films on glass probably has a negligible effect on the resistance.

Values of the thickness and ρ_r for the specimens are listed in Table I.

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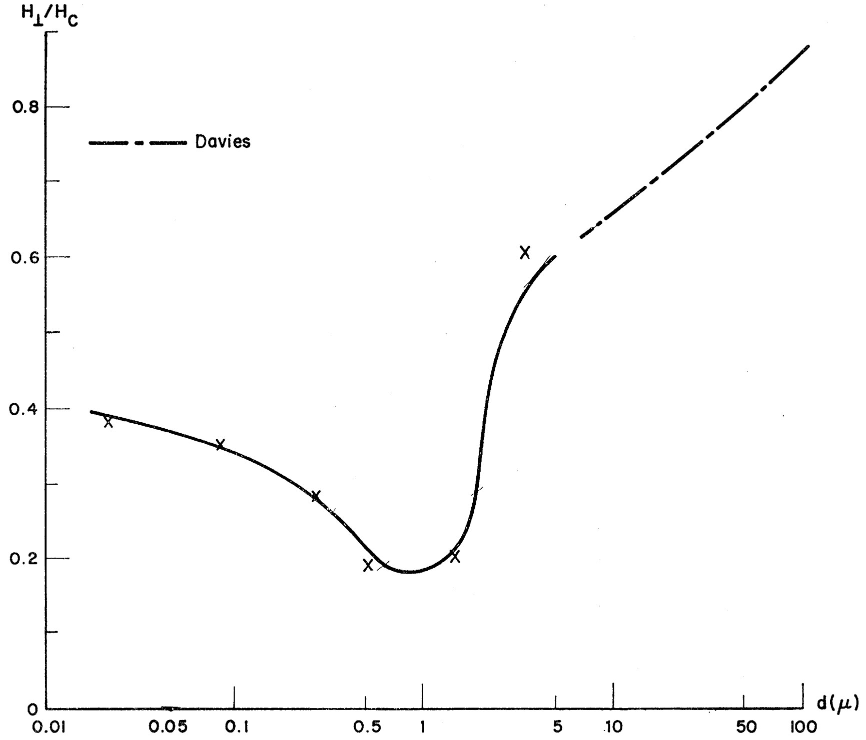

FIG. 1. Dependence of $H_\perp$ on film thickness for indium specimens at $t=0.9$.

B. Magnetization Measurements

Each specimen was mounted in our vibrating sample magnetometer[22] and magnetization curves were recorded at each of several temperatures between 1.4°K and 4.2°K. The general shape of the curves obtained and a discussion of their meaning may be found in our first communication.[13] We state here that measurements were taken in increasing magnetic field, and $H_\perp$ was evaluated by linearly extrapolating the rapidly falling portion of the magnetization curve to zero magnetic moment. After each curve was obtained, the specimen was warmed to above its transition temperature, in order to rid it of trapped flux, before cooling to the temperature of the next magnetization measurement.

The transition temperature of each specimen was determined by extrapolating $H_\perp(T)$ to $H_\perp=0$, and the values obtained are also listed in Table I.

C. Results

i. Dependence of $H_\perp$ on Thickness

As pointed out by Guyon *et al.*,[23] one expects Tinkham's model[1] to apply only to films thinner than some critical thickness. For films thinner than this value, $H_\perp$ decreases with increasing thickness d whereas for films thicker than this value an intermediate state is created with the result that $H_\perp$ increases with d as found by Davies[24] in foil specimens. The dividing line between the two types of behavior occurs at the thickness corresponding to the minimum in the $H_\perp$-versus-d curve. In tin[23,25] this minimum occurs for d about 2500 Å. We show in Fig. 1 the values of $H_\perp$ at $t=0.9$ for indium films of various thicknesses. The thicker films are not listed in Table I because they are of no interest to the subsequent discussion. As can be seen from Fig. 1, the minimum in $H_\perp$ occurs at the rather large value of $d \simeq 8000$ Å. It should be noted that the film data do seem to fit reasonably well on to Davies' data[24] for indium foils at the same temperature, whereas the data obtained for tin[23,25] do not seem to fit together with the foil data as well as those shown in Fig. 1.

All the indium specimens in Table I are less than 8000 Å thick. The three tin-based specimens measured were just slightly less than 2500 Å thick.

ii. Ginzburg-Landau Parameter

In Figs. 2 and 3 we show the results of plotting the data for some of the indium specimens and the three tin specimens as $(f^{-1}H_\perp)$ versus t. It is to be noted that for each specimen it is possible to extrapolate all three curves to a common point at $t=1$, thereby determining the value of KH_0 for each one. The values obtained in this way are also listed in Table I. In order to determine K, we must know H_0. For the film made from pure tin this is no problem since its transition temperature

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is very close to the accepted value for bulk specimens,[26] and we therefore take $H_0=283$ Oe as determined for such specimens. The nominally pure indium specimens have T_c's about 0.03°K lower than the accepted value[26] indicating perhaps some stress in the film. Of course in the alloy films the transition temperatures can differ appreciably from the T_c of the pure metal because of mean free path and valence effects. To estimate H_0 in these cases, we assume that a law of corresponding states exists, i.e., $H_0/T_c=(H_0/T_c)_{\text{pure}}$. Such a law has been demonstrated for bulk indium alloy specimens[19] with concentrations up to about 1.8% tin. At higher concentrations this law may not be exact, and it does not hold quite as well in stressed alloy films[19] as in bulk specimens, but these effects seem small enough not to affect our conclusions. The values of H_0 determined in this way are listed in Table I, and the values of K calculated from them are given in Table II.

The electronic mean free path, l, in the various specimens as calculated from ρ_r are also listed in Table II. For the indium specimens, we use the value of the value $l/\sigma=1.6\times10^{-11}$ Ω cm^2 as determined from the thickness dependence of the electrical resistance of indium films.[19] As pointed out by Toxen *et al.*[19] this value is appreciably larger than the value of l/σ obtained from anomalous-skin-effect measurements on the bulk metal. For tin, we have available only the value of $l/\sigma=1.03\times10^{-11}$ Ω cm^2 determined from measurements on bulk specimens.[27]

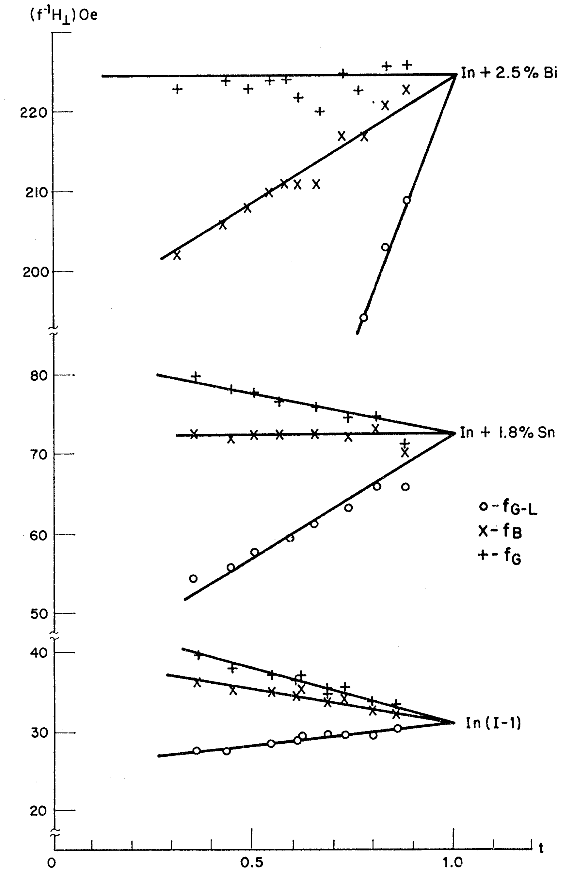

FIG. 2. The functions $(f^{-1}H_\perp)$ are shown as a function of t for indium-based specimens.

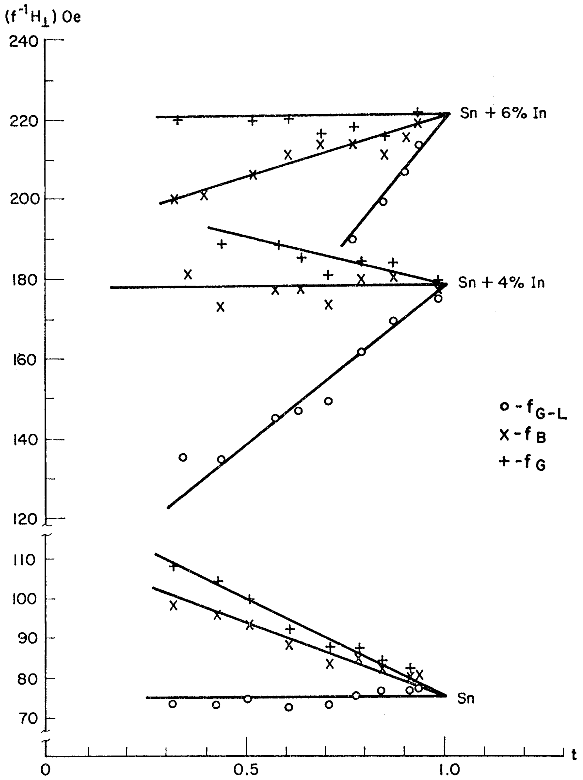

FIG. 3. The functions $(f^{-1}H_\perp)$ are shown as a function of t for tin-based specimens.

To determine K_0 from the data, we plotted K as a function of ρ_r and extrapolated to $\rho_r=0$. This procedure yielded $K_0=0.11$ for indium; and $K_0=0.15$ for tin in fair agreement with values obtained from supercooling experiments[3,4] on bulk specimens. Taking the value[28] $\xi_0/\lambda_L(0)=7.4$ for indium and the value[29] 6.5 for tin, Eq. (3) gives for K_0 the values of 0.13 and 0.15, respectively, in reasonable agreement with the measurements. Equation (2) was used to calculate K from the values of l, and the results are tabulated in Table II. For the indium based specimens, the agreement between the calculated and measured values of K is remarkably

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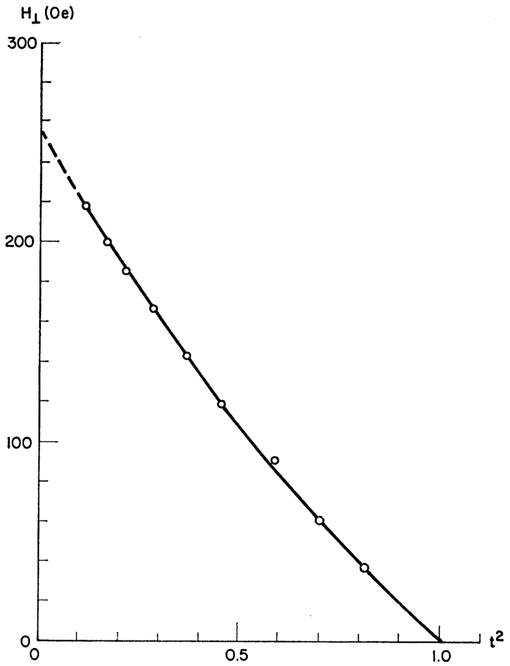

FIG. 4. $H_\perp$ is plotted versus t^2 for the In+5.7% Sn specimen.

good. For the tin specimens the calculated quantities are considerably larger than the measured values of K. This discrepancy could arise because the magnitude of l/σ as determined from the anomalous skin effect is smaller than the value describing film specimens. In fact, if we assume that the value of l/σ appropriate for tin films is equal to the value found in indium films (since indium and tin have similar bulk electrical properties), we obtain the numbers for l and K (calc.) shown in parentheses in Table II. The agreement between the latter values and the measured values of K is now good. Although we cannot be sure that the value of l/σ for tin films should be so modified, the results in Table II make it seem likely that a change would be appropriate.

TABLE II. Parameters derived from the data. l=electronic mean free path; K=Ginzburg-Landau parameter at T_c; $K_1(0)$ =Ginzburg-Landau parameter at 0°K.

Specimen	l(Å)	K(calc.)	K(meas.)	$K_1(0)$	$K_1(0)/K$
I−1	17 200	0.14	0.11	0.18	1.64
I−2	9410	0.16	0.15	0.22	1.47
I−3	2960	0.22	0.22	0.32	1.45
I−4	2840	0.22	0.21	0.295	1.41
I−5	2760	0.22	0.25	0.35	1.40
I−6	1420	0.31	0.30	0.43	1.43
I−7	933	0.40	0.44	0.58	1.31
I−8	460	0.69	0.68	0.85	1.25
S−1	1630 (2540)	0.31 (0.25)	0.25	0.46	1.84
S−2	464 (720)	0.70 (0.51)	0.59	0.86	1.46
S−3	306 (475)	0.99 (0.69)	0.72	0.90	1.25

In connection with the temperature dependence of $K_1(T)$, we note from Figs. 2 and 3 that Ginzburg-Landau function f_{G-L} seems to fit $K_1(T)$ for the Sn specimen very well, and while the fit is not as good for the In specimen, f_{G-L} still gives a slightly better fit than the other two functions. For the In+1.8% Sn and Sn+4% In specimens f_B gives the best fit. Finally, it can be seen that for In+2.5% Bi specimen and the Sn+6% In specimen, the Gor'kov function gives the best fit.

To prevent confusion, not all the data for the indium specimens have been plotted, but the trend indicated above is confirmed by all the data. We conclude that as K increases (or the electron mean free path decreases) the function that best fits the data for $H_\perp(T)$ changes progressively from the Ginzburg-Landau function to the Bardeen function to the Gor'kov function. To check this conclusion we plotted $H_\perp(t)$ for each specimen as a function of t^2 and extrapolated to $t=0$ to find $H_\perp(0)$. A typical curve is shown in Fig. 4. The concave shape makes extrapolation difficult, but we have done it such a way as to underestimate $H_\perp(0)$, and the values obtained are listed in Table I. From these values and those for H_0, Eq. (1) was used to calculate the values for $K_1(0)$ shown in Table II. As can be seen $K_1(0)/K$ decreases reasonably smoothly as K increases. When we recall that $[K_1(0)/K]_{G-L}=2.0$, $[K_1(0)/K]_B=1.4$ and $[K_1(0)/K]_G=1.25$, the trend noted above is substantiated. It is to be noted that the experimentally determined values of this ratio do not depend on the assumption that $H_c(T)$ is a parabolic function of the temperature.

III. DISCUSSION

In considering the conclusion arrived at in the foregoing paragraph, we are very aware of the fact that we are dealing with small differences between the specimens, and that we may have biased the treatment of the data in some way. However, our results taken together with the evidence of other investigations,[11–14] makes it seem very likely that the magnitude of K (of or the electron mean free path) is an important parameter in determining the form of the temperature dependence of K_1. That this is not the only parameter is evident from the slightly different behavior between the Sn and In specimens in this investigation, and from the observations on strong coupling superconductors.[15] We remind the reader that tin and indium are in the class of weak-coupling superconductors.

Although we have chosen to express the results in terms of a Ginzburg-Landau parameter K_1, what we have measured in $H_\perp$ is the temperature dependence of the field value corresponding to the absolute stability limit[10,13] of the normal phase in decreasing magnetic field. In specimens for which $H_\perp(0)<H_c(0)$ the values

of $H_\perp$ correspond to the supercooling field, whereas when the inequality is reversed, $H_\perp$ corresponds to the upper critical field H_{c2} of a Type II superconductor. Of the three functional forms assumed for the temperature dependence of this field, only the Gor'kov function,[10] f_G, has been derived from the microscopic theory of superconductivity. Gor'kov obtained results only near T_c and at 0°K, and arrived at $f_G(T)$ by interpolation. The rigorous extension of the theory to all temperatures has been discussed by Helfand and Werthamer[30] and by Maki and Tsuzuki,[31] who obtained essentially the Gor'kov result. The two other functions f_B and f_{G-L} are semi-empirical, and have no clear fundamental basis. It is also pertinent to remark that function f_G is derived in the pure limit of infinite mean free path.

We must conclude that a clear quantitative discrepancy exists between the theory and the measured values of the fields at which the normal phase becomes unstable in nominally pure indium and tin. It is only when the specimens are reasonably *dirty* (i.e., when the mean free path becomes less than or about equal to the coherence length) that the data agree with the theory for the *pure* limit. The pure indium and tin films we used were type I superconductors, but we remark that data obtained in this laboratory on niobium[12] which is a pure type II superconductor also disagree with the Gor'kov function in the same sense as those for indium and tin. Moreover, it is only the data for reasonably dirty niobium which agree with theory.

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Cesium Ionization Cross Section from Threshold to 50 eV*

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The Cs ionization cross section σ, which was recently measured by Brink at 50 eV and above, has been measured down to threshold in a Tate- and Smith-type apparatus designed for absolute measurements. The density in the collision space was determined by a surface ionization detector which permitted an over-all accuracy on the cross section of 10%. The results are: (1) $\sigma = 5.3$ Å^2 at 50 eV. This is smaller by a factor of 1.8 than the results of the above experiment and the theoretical values of the Gryziński method—the discrepancy has not been explained. (2) The cross section near threshold is linear over a range of 0.8 eV with a slope of 2.2 Å eV^{-1}. Above 0.8 eV, it rises less than linearly. The initial slope is smaller by a factor of 3 than the earlier measurements of Nottingham. (3) Above 12 eV, excitation to the autoionizing states is seen, and above 18 eV, an additional contribution to the total cross section begins. The latter is due to ionization to an excited state of the ion. Cross sections for these two processes have been related to spectroscopic measurements of Beutler *et al.* and have been separated from the basic ionizing channel. The curve for autoionization is resonant in nature with a maximum of 0.8 Å^2. The ionization to the excited ion has an estimated maximum cross section of 1.5 Å^2 at 60 eV.

I. INTRODUCTION

In 1934, Tate and Smith[1] measured the ionization cross section of cesium as a function of the energy of the ionizing electrons. The measured curve included the threshold and showed a structure which was believed to be a result of other ionizing channels. However, no determination of the absolute value of the cross section was made. A magnetically collimated electron beam was passed through a cesium atmosphere of about 5×10^{-5} Torr pressure. A small electric field perpendicular to the magnetic field accelerated and extracted part of the ions produced into the entrance slit of a magnetic mass spectrometer. When tuned to Cs$^+$, the current to the collector of this instrument indicated the signal used to plot the cross-section function. It is assumed that the fraction of extracted ion current to the total number of ions produced remains the same for all energies.

Beutler and Guggenheimer,[2] also in 1934, measured spectroscopically the autoionizing levels starting at 12.3 eV and terminating at two pairs of ionization limits, one at 17.3 eV and one at 19.1 eV. Tate and

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