

Coulomb pseudopotential for superconducting alloys from the normal-state resistivity

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The Coulomb pseudopotential μ^* is estimated from the superconducting T_c and the normal-state resistivity-temperature derivative, $d\rho/dT$. This method is applied to noble-metal alloys. New resistivity measurements for AuAl and AgGa alloys as a function of temperature and concentration are presented. Analysis of these data and published results for T_c and $d\rho/dT$ give $\mu^* = 0.125 \pm 0.009$ for three alloy systems. T_c 's for pure Au and Ag are also estimated. For Au the result is about 10^{-4} K while T_c for Ag is nonzero but exceedingly low. The electron-phonon interaction λ is obtained from $d\rho/dT$ in $n\Omega \text{ cm/K}$ by $\lambda = 0.0312 d\rho/dT$ to an accuracy of 0.01 for noble-metal alloys.

I. INTRODUCTION

The magnitude of the superconducting transition temperature T_c results mainly from the balance between an attractive phonon-mediated interaction λ and a screened repulsive Coulomb interaction μ^* . In the past several years there has been a continued interest in λ . New experimental techniques to estimate λ have been obtained such as point-contact spectroscopy¹ and deconvolution of resistivity data² in the temperature region below about $\Theta/3$. In contrast there is little work on the repulsive interaction and one often handles μ^* by assuming that it is constant and equal to 0.10. This approach is justified, e.g., in strong coupling superconductors where $\mu^* \ll \lambda$. However, the scarcity of experimental methods to obtain μ^* has obviously contributed to this praxis.

In fact there are only two well-known methods to determine μ^* . One is based on the isotope effect³ which for six elements gave values in the range from 0.09 for Mo to 0.17 for Zr. The other method is based on the inversion of tunneling data by Eliashberg's equations.⁴ When these results were renormalized to correct for the high cutoff frequencies used in the numerical data handling⁵ a compilation for about 30 nontransition elements and alloys and for Ta gave results for μ^* in the range 0.09–0.12.

Both these methods have obvious limitations. The isotope effect can be used only when a set of isotopes is available. The tunneling experiments require strong or medium coupling superconductors in order to give sufficient structure in the current-voltage characteristic. Furthermore, the

tunneling technique has sometimes been questioned^{6,7} mainly due to the controversial results for Nb.^{8,9}

In this paper we propose and test a new method to obtain μ^* which is suitable for weakly coupled superconductors. In Sec. II it is shown how μ^* of McMillan's formula³ can be obtained from measurements of T_c and the resistivity for a set of closely related alloys. This method is applied to three noble-metal-based alloy systems. The new experimental results are described in Sec. III. From these data and published results we obtain estimates of μ^* in Sec. IV. In Sec. V it is shown that an essential condition for the analysis is obeyed for these alloys. The various approximations used in the analysis are examined in Sec. VI. In Sec. VII estimates for the T_c 's of Au and Ag are obtained. The main results are summarized in Sec. VIII.

II. DESCRIPTION OF THE METHOD

Our analysis is based on two relations. One is McMillan's formula³ which relates T_c to λ , μ^* , and the Debye temperature Θ ,

$$\left[\ln \frac{\Theta}{1.45T_c} \right]^{-1} = \lambda \frac{1}{1.04(1+\lambda)} - \mu^* \frac{1+0.62\lambda}{1.04(1+\lambda)}. \quad (1)$$

The other relation connects λ with the temperature dependence of the electrical resistivity¹⁰ at a temperature $T \gtrsim \Theta$,

$$\lambda = k \frac{d\rho}{dT} \quad (2)$$

k contains an average over the scattering angle in the transport process and the plasma frequency of the electron gas. Using experimental data for T_c , Θ , and $d\rho/dT$ for several alloys in an alloy system, k and μ^* are obtained from a fit based on Eqs. (1) and (2).

For a set of closely related alloys k is assumed to be a constant independent of alloy concentration. Examples from several alloy systems show¹⁰ that this approach describes empirical data with quantitative precision. The verification of Eq. (2) for the present alloys is discussed in Sec. V. Furthermore, we assume that μ^* is independent of alloy concentration.

The solid solubility limit in the fcc phase restricts the maximum T_c in noble-metal alloys to about¹¹ 0.3 K. As a consequence, the left-hand member of Eq. (1) varies in a relatively small range, and a nonlinear least-squares fit of experimental T_c/Θ and $d\rho/dT$ to Eqs. (1) and (2), with k and μ^* as adjustable parameters, is insensitive to μ^* . A more suitable method in the present case is therefore an approximate linear analysis of Eq. (1). Noble-metal alloys are favorable for such an analysis since λ is relatively small and varies appreciably with solute concentration. Under these conditions the variation of λ is much larger than the variation of $1 + \lambda$ and in Eq. (1) we can replace factors of type $1 + \lambda$ by constants. Inserting Eq. (2) into Eq. (1) one obtains

$$\left[\ln \frac{\Theta}{1.45T_c} \right]^{-1} = k_1 \frac{d\rho}{dT} - k_2, \quad (3)$$

$$k_1 = k [1.04(1 + \lambda)]_{\text{av}}^{-1}, \quad (4)$$

$$k_2 = \mu^* \left[\frac{1 + 0.62\lambda}{1.04(1 + \lambda)} \right]_{\text{av}}, \quad (5)$$

where k_1 and k_2 are constants and $[]_{\text{av}}$ denotes an average for the samples of the alloy system. It will be seen that, for noble-metal alloys, the difference between such an average and its extreme value is at most a few percent.

When λ is less than about 0.3 the difference between k_2 and μ^* in Eq. (5) is less than about 10%. An immediate interpretation of Eq. (3) is, therefore, that plotting $[\ln(\Theta/1.45T_c)]^{-1}$ vs $d\rho/dT$ results in a straight line with an intercept close to $-\mu^*$. This approach was used previously for AuGa alloys.¹² In the present work we improve on this analysis as follows: When experimental data

for T_c/Θ and $d\rho/dT$ are plotted as in Eq. (3), a straight-line fit determines k_1 and k_2 . For a given μ^* and the experimental T_c/Θ one can calculate a set of λ values from Eq. (1). A least-squares fit to Eq. (2) gives k . The averages of the functions of λ in Eqs. (4) and (5) are also obtained with these λ values. The right-hand members of Eqs. (4) and (5) can thus be calculated for a given μ^* . The calculation is repeated for a different μ^* until the results are equal to k_1 and k_2 obtained from Eq. (3). This gives two independent estimates of μ^* for each alloy system.

Since Θ varies little in these alloy systems compared to T_c , we take the simplifying approach that Θ is constant for each alloy system. Experimental data for T_c and $d\rho/dT$ from the literature are used when available. In some cases we had to supplement these with new resistivity measurements. These experiments are described in the next section.

III. EXPERIMENTAL PROCEDURES AND RESULTS

The noble-metal alloy systems studied are AuGa, AuAl, and AgGa. For the AuGa system data for T_c and $d\rho/dT$ were obtained on the same samples.¹² For the resistivity measurements of AuAl and AgGa alloys new alloys were prepared. To avoid interpolation in the T_c data of these alloys,¹¹ those compositions were chosen for which T_c had been measured. For accurate resistivity measurements on metallic samples, thin wires are preferred which combine a well-defined form factor with a conveniently high resistance per unit length. This technique is now described.

TABLE I. Elements used in sample preparation. Sources are as follows: (a), Cominco, Washington, D. C.; (b) and (c), Materials Research Corporation, N. Y.; (d), Johnson and Matthey, England; (e), Koch-Light, England.

Element	Nominal impurity level (ppm)	Source
Au	1	(a)
Au	10	(b)
Ag	1	(c)
Al	10	(d)
Ga	10	(e)

TABLE II. Preparation of AgGa wire samples. T_h is a temperature in the region 600–670°C.

Stage	Treatment
I	Ingot of mass 1–2 g cold-worked, annealed at T_h for 50 h, water-quenched.
II	Cold-worked to approximately $1.2 \times 1.2 \text{ mm}^2$, annealed at T_h for 50 h, water-quenched.
III	Cold-worked to approximately $0.6 \times 0.6 \text{ mm}^2$, annealed at T_h for 50 h, water-quenched.
IV	Cold-drawn to 0.40, annealed at 500°C for 3 h, water-quenched.

A. Preparation of thin-wire samples

The source and nominal purity of the starting materials are given in Table I. Weighed quantities were melted in an evacuated quartz tube on an alumina crucible in an induction furnace for good stirring. Weight losses were negligible and nominal compositions were used throughout. AgGa alloys in the appropriate concentration range of about 10–17 at. % Ga were found to crack easily when subjected to heavy cold-work and several homogenizing heat treatments were required to prepare wire samples. This process is summarized in Table II. T_h is a temperature close to the solidus temperature¹³ and decreases from 670°C for the 10.5 at. % Ga alloys to 605°C for the 17.2 at. % alloy. For AuAl alloys, a less laborious preparation technique was required. After initial cold-working, these samples were homogenized for 15 h at a temperature in the range 670–730°C, water-quenched, cold-drawn to a circular cross section of 0.40 mm diameter and then given a final anneal at 570°C for 3 h followed by water-quenching.

B. Measurements and results

The electrical resistivity was measured with a conventional four-probe dc technique in the temperature range from 230 to 300 K. These temperatures are above the Debye Θ for both alloy systems and the resistivity is expected to be linear in temperature. Regulated temperatures were read from a calibrated Pt resistor thermometer to a relative accuracy of 2 mK. To convert measured resistance values to resistivities, the geometric form factors were determined from the measured length and

mass of the samples and the density calculated from the lattice parameters.¹⁴ Thermal expansion is neglected.

Typical results are illustrated for some AgGa alloys in Fig. 1. The rms deviation of the data from the straight lines shown are about 8×10^{-5} of the measured resistivity for all AgGa alloys and for AuAl alloys with an Al concentration of 7 at. % or less. $d\rho/dT$ for the alloys was obtained from the slope of such straight lines fitted by least squares. The results are given in Table III. The pure Au and Ag samples were also measured and the results are included for comparison. To display all the input data used in the analysis in Sec. IV, data for T_c and $d\rho/dT$ for all samples used have been collected in Table III.

Although the fcc phase boundary¹³ and the T_c measurements¹¹ for AuAl alloys extend up to 14 at. % Al, the results of the resistivity measurements forced us to exclude samples with 8 at. % or more Al from the analysis. In these samples the resistance was found to become a concave function of temperature above about 270 K. For several samples the resistance also showed a weak time dependence. Therefore it seems plausible that the anomaly is connected with a structural change. Possibly such a transformation occurs within the fcc phase since our attempts to observe it by $\text{CuK}\alpha$ radiation at room temperature were unsuccessful. It can be concluded however that other mechanisms than the electron-phonon interaction contribute to $d\rho/dT$ for these samples.

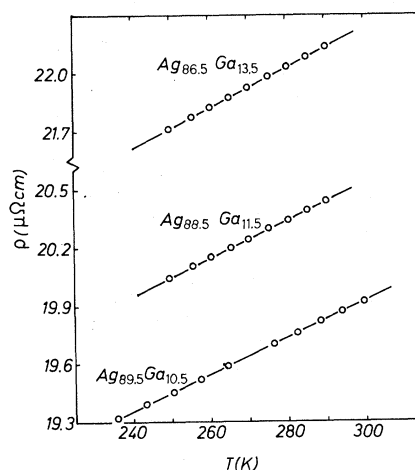


FIG. 1. Resistivity vs temperature for some AgGa alloys.

TABLE III. Data for $d\rho/dT$ and T_c for superconducting alloy systems.

Sample	$d\rho/dT$ ($n\Omega$ cm/K)	T_c (K)
Ag alloys		
Ag	5.832	
Ag _{89.5} Ga _{10.5}	9.524	0.0065 ^a
Ag _{88.5} Ga _{11.5}	9.640	0.0135 ^a
Ag _{86.5} Ga _{13.5}	10.32	0.0310 ^a
Ag _{84.5} Ga _{15.5}	10.55	0.070 ^a
Ag _{82.8} Ga _{17.2}	11.21	0.116 ^a
Au alloys		
Au (Cominco)	8.256	
Au (MRC)	8.064	
Au ₉₆ Al ₄	9.357	0.008 ^a
Au ₉₅ Al ₅	9.627	0.0138 ^a
Au ₉₄ Al ₆	10.10	0.0224, 0.0282 ^a
Au ₉₃ Al ₇	10.53	0.062 ^a
Au ₉₇ Ga ₃	9.45 ^b	0.0078, 0.0094 ^a
Au ₉₆ Ga ₄	9.69 ^b	0.0119, 0.0142, 0.0152 ^a
Au ₉₅ Ga ₅	10.37 ^b	0.032, 0.034, 0.0388 ^a
Au ₉₄ Ga ₆	10.55 ^b	0.070, 0.0755 ^a
Au ₉₂ Ga ₈	11.53 ^b	0.151, 0.168 ^a
Au ₉₀ Ga ₁₀	12.43 ^b	0.264 ^a

^aReference 11.^bReference 12.

IV. ANALYSIS

We take $\Theta = 160$ K for the Au alloys and 220 K for the Ag alloys. Figure 2 shows $[\ln(\Theta/1.45T_c)]^{-1}$ vs $d\rho/dT$ for the AuAl and AgGa systems. A similar graph for the AuGa data was shown previously.¹² Fitting straight lines by linear least squares determined the parameters k_1 and k_2 of Eq. (3). The results are given in Table IV.

μ^* can be obtained by solving graphically either Eq. (4) or Eq. (5). We illustrate this in detail by taking Eq. (4) and AgGa alloys as an example. Suppose $\mu^* = 0.10$. λ for the superconducting alloys is then obtained by Eq. (1) and is found to be in the range 0.24–0.31. The arithmetic mean of

TABLE IV. Results for k_1 and k_2 of Eq. (3).

	AuGa	AuAl	AgGa
k_1 (K/ $n\Omega$ cm)	0.0221 ^a	0.0231	0.0231
k_2	0.102 ^a	0.113	0.117

^aReference 12.

$[1.04(1+\lambda)]^{-1}$ is 0.754. k is determined by fitting values of λ and $d\rho/dT$ to Eq. (2). It is apparent from Sec. V, however, that all three alloy systems studied satisfy Eq. (2) with the same k . Therefore we use all data for λ and $d\rho/dT$ to determine k . For $\mu^* = 0.10$ the result is $k = 0.0276$ K/ $n\Omega$ cm and, from Eq. (4), $k_1 = 0.0208$ K/ $n\Omega$ cm. The point (0.10, 0.0208) lies on the curve shown in Fig. 3 obtained by repeating this calculation for a series

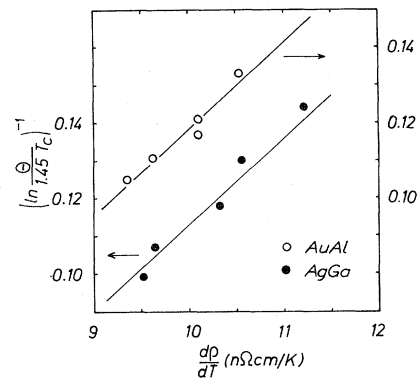


FIG. 2. Determination of k_1 and k_2 for two noble-metal alloy systems by a plot of experimental T_c/Θ and $d\rho/dT$ according to Eq. (3).

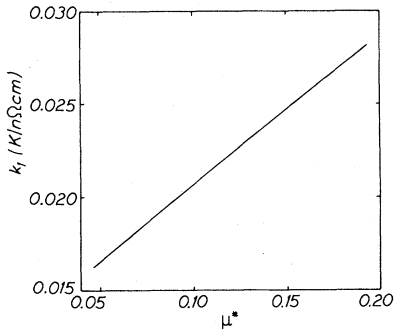


FIG. 3. Solution of Eq. (4) for AgGa alloys. The right-hand member of Eq. (4) is plotted as a function of μ^* . The result for k_1 from Fig. 2 gives μ^* .

of μ^* . With k_1 for AgGa alloys equal to 0.0231 K/n Ω cm from Table IV, the solution to Eq. (4) is found from Fig. 3 to be $\mu^* = 0.127$.

This analysis is performed for the three alloy systems and for both Eq. (4) as well as Eq. (5). The results are collected in Table V. Our estimates of μ^* from Eq. (5) are within 10–15 % from the values of k_2 in agreement with the implication of Eq. (3) that the constant term approximates $-\mu^*$.

Neglecting possible differences between different alloy systems all results in Table V can be summarized as $\mu^* = 0.125 \pm 0.009$. This result contains six independent estimates from three different noble-metal alloy systems and was obtained by different combinations of samples, measurements of T_c and $d\rho/dT$, and experimentalists. These observations give strong empirical evidence for the methods used.

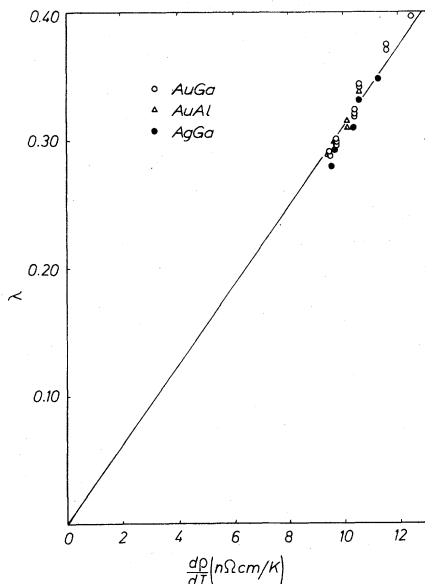


FIG. 4. λ vs $d\rho/dT$ for noble-metal alloys.

V. EMPIRICAL VERIFICATION OF EQUATION (2)

With the result from the preceding section of $\mu^* = 0.125$ for noble-metal alloys we calculate λ from Eq. (1) using the observed T_c 's and $\Theta = 160$ K for the Au alloys and 220 K for the Ag alloys. Figure 4 shows λ vs $d\rho/dT$ for the three alloy systems studied. With $d\rho/dT$ in n Ω cm/K the straight line has the equation

$$\lambda = 0.0312 \frac{d\rho}{dT} . \quad (6)$$

The rms of the deviations is 0.008 in λ . Equation (2) is thus well obeyed for these alloys. The origin is included in Fig. 4 to demonstrate that there is no constant term in Eq. (2).

It should be emphasized that the validity of Eq. (2) is not restricted to any particular result for μ^* . For a given T_c/Θ , λ depends on μ^* by Eq. (1) but the quality of the fit to Eq. (2) does not. Thus, varying μ^* from 0 to 0.30 increases k in Eq. (2) by a factor of 4 but the scatter of the data in a plot such as Fig. 4 remains essentially unaffected.

When any of the three noble-metal alloy systems is considered separately in a fit to Eq. (2) the result in Fig. 4 is not significantly changed. This indicates a somewhat surprising rigid-band behavior and increases the usefulness of Eq. (6). In summary of this section we have found that Eq. (6) is a convenient relation to obtain λ in Au- and Ag-based alloys to an accuracy of about 0.01 in λ . In the next section we shall discuss corrections to this analysis by considering the possibility that k in Eq. (2) depends weakly on solute concentration.

VI. DISCUSSION

A more straightforward method of obtaining μ^* than the one discussed here would use k and μ^* as adjustable parameters in a nonlinear fit of experimental data for T_c/Θ and $d\rho/dT$ to Eqs. (1) and (2). In order to obtain a well-defined result for μ^* by that method, the left-hand member of Eq. (1) should vary appreciably over the set of alloys stud-

TABLE V. Results for μ^* .

	AuGa	AuAl	AgGa
From Eq. (4)	0.122	0.129	0.127
From Eq. (5)	0.116	0.129	0.134

ied implying a variation of T_c from several K down to the mK region. Alloy systems are rare in which such a variation of T_c is observed without accompanying changes in electronic structure overthrowing Eq. (2). Amorphous Zr-based alloys, however, may be promising candidates since it was recently shown¹⁵ that Eq. (2), with negative k and $d\rho/dT$, is obeyed for a set of these alloys where T_c extends from 15 mK to 3 K. Our results indicate that μ^* in these alloys can be determined in such a nonlinear analysis¹⁶ although improvements of the resistivity measurements over those reported in Ref. 15 are apparently required to assess the possibilities of this method.

In the linearized analysis presently employed, the slowly varying functions of λ in Eq. (3) have been replaced by their averages. Furthermore, it was assumed that Θ , k , and μ^* are all constants and independent of alloy concentration c . We now show that these approximations are acceptable at the level of accuracy obtained for the estimates of μ^* .

Low-temperature specific-heat measurements^{17,18} show that Θ decreases with increasing c in noble-metal-based alloys. For both AuGa as well as AgGa alloys $d\Theta/dc$ is about -1 K per at. % and in the absence of other information we can assume a similar value also in AuAl alloys. Such a concentration dependence of Θ has a small influence on the results. We find that this correction increases the estimate of μ^* by 2–3 %.

Inspection of Fig. 4 suggests that the fit to the experimental data could be improved by a weakly curved relation between λ and $d\rho/dT$ including also a term in $(d\rho/dT)^2$. This possibility was examined by allowing for a small concentration dependence of k which is numerically equivalent and physically more reasonable than a higher-order term in $d\rho/dT$. It was then found that an increase of k with c of less than 1% per at. % of solute may represent the best fit to the experimental data.

Comparatively little appears to be known from the literature about the concentration dependence of k in alloy systems. Rough estimates are available, however. When an early estimate of Grimvall¹⁹ is applied to the present alloys, one gets for $d \ln k / dc$ about +0.5% per at. %, and more recently¹² this value was modified to below about 1% per at. % for AuGa alloys from new experimental data. These estimates thus confirm our result for $d \ln k / dc$ of about 1% per at. % of solute.

In Eq. (3) factors of type $1 + \lambda$ were replaced by their average value since the deviations from this average were small. In fact, $d \ln(1 + \lambda) / dc$ is

+ 1.2% per at. % of solute for the Au alloys and + 0.8% per at. % for the AgGa alloys. These values are close to those obtained for $d \ln k / dc$. Therefore, as seen, e.g., from Eq. (4), corrections to the estimate of μ^* which arise from neglecting the small concentration dependence of k almost cancel corrections to replacing $1 + \lambda$ by an average value.

As for the concentration dependence of μ^* we assume that it can be discussed from the Morel-Anderson expression²⁰

$$\mu^* = [\ln(E_F/k_B\Theta) + 1/\mu]^{-1}. \quad (7)$$

Here E_F is the Fermi energy and μ the screened Coulomb repulsion which is expressed in the Fermi momentum k_F and a screening length k_s^{-1} . We assume that k_F increases in a free-electron-like way when solutes are added to the noble-metal hosts and use a Thomas-Fermi model for k_s . For increasing c it is then found that μ decreases, E_F increases, and Θ decreases so that all quantities in Eq. (7) contribute to a negative $d\mu^*/dc$. The result is about $d \ln \mu^* / dc \simeq -0.5\%$ per at. % in all three alloy systems. Such variations of μ^* are small and are also contained within the limits given by the result in Sec. IV.

VII. ESTIMATES OF T_c IN Au AND Ag

The possibility of superconductivity in the noble metals, particularly in Au, continues to be of interest. New methods^{1,2} to obtain λ have been accompanied by estimates of λ for Au. In double-stage nuclear refrigerators recently brought into operation, some of the first experiments^{21,22} have been to search for superconductivity in Au. We compare these results with the estimates of T_c and λ now obtained by the present method.

T_c can be estimated from Eq. (3) without involving any assumption about μ^* except that it is constant in each alloy system. With the measured $d\rho/dT$ for the pure elements and neglecting gap anisotropy we obtain for Au values of T_c in the range 200–400 μ K for the Cominco sample and 100–200 μ K for the Materials Research Corporation (MRC) sample. One can also use the general result in Fig. 4 to obtain λ for the elements. An estimate from the straight line in Fig. 4, however, will overestimate λ for Au by about 0.01 and T_c by a factor of 4 since k depends weakly on solute concentration. We improve on this by taking our result for $d \ln k / dc$ into account and using the Thomas-Fermi estimate of $d \ln \mu^* / dc$. Then it is

found that λ for Au is 0.24 and T_c about 100 μ K.

Our results for T_c of Au are comparable to the estimates from proximity-effect experiments²³ and with the values of T_c obtained by extrapolating λ to zero solute concentration in superconducting alloy systems.^{11,21} Our value of λ , however, is larger since some of these earlier results were based on the assumption that $\mu^* = 0.10$. Theoretical calculations of λ for Au are apparently very sensitive to the choice of pseudopotential.²⁴ Point-contact spectroscopy¹ as well as deconvolution of resistivity integral equations² seem to give too low values of λ .

For Ag the result from Fig. 2 indicates that the net electron-electron interaction is positive and that Ag is thus a superconductor. The estimate of T_c is extremely uncertain. With the value of $d\rho/dT$ from Table III one gets $T_c \simeq 10^{-23}$ K, while a 4% larger value of $d\rho/dT$ obtained²⁵ for Ag at 50°C increases the estimated T_c by 6 orders of magnitude. With our result for μ^* and Eq. (1) both these estimates give $\lambda \simeq 0.16$.

Although some proximity-effect results²⁶ as well as early theoretical estimates²⁷ suggest that Ag is nonsuperconducting, most estimates from proximity-induced superconductivity in Ag give a positive net interaction.²⁸⁻³² A most likely value for T_c from these experiments seems to be in the region of 10^{-5} K although such a figure is again very uncertain since all of these results for T_c span the huge range from 10^{-20} to 10^{-2} K. Extrapolation of λ versus the square of the solute concentration¹¹ for AgGa alloys gave $T_c \simeq 10^{-9}$ K. Our results confirm this general trend that Ag eventually becomes a superconductor albeit T_c is exceedingly low.

VIII. CONCLUDING REMARKS

The main result of the present paper is the demonstration that μ^* can be estimated from measurements of T_c and $d\rho/dT$ for a set of related alloys. This method requires that $\lambda = k(d\rho/dT)$ for these alloys. In fact, three noble-metal alloy systems are found to obey this relation with the same k to an accuracy of about 0.01 in λ . Equation (6) therefore provides a useful way to obtain λ in Au- and Ag-based alloys. As a particular application the T_c 's of the pure noble metals can be estimated.

Since a large range of variation of T_c cannot be attained in the noble-metal alloys due to the phase boundary of the fcc phase, the linearized analysis of Eq. (3) is used to obtain μ^* . We have investigated the approximations in this method and found them to be acceptable at the level of accuracy obtained for μ^* . This conclusion is also empirically supported by the consistent results for three different alloy systems.

In an application of Eq. (2) to superconducting elements under pressure³³ estimates were obtained for the pressure dependence of the plasma frequency ω_p contained in our parameter k . The corresponding application in the present case to obtain the concentration dependence of ω_p in an alloy system would merit further investigation.

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