Superconductivity, Susceptibility, and Specific Heat in the Noble Transition Elements and Alloys. II. Comparison with Theory*

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The results of an experimental study of the superconducting transition temperature T_c, the magnetic susceptibility χ, and the electronic specific heat γT in noble transition metals are discussed in terms of the existing theory for the interactions which affect these parameters. Particular attention is paid to the possible presence of virtual ferromagnetic spin fluctuations in these paramagnetic metals, using the recent theory of Berk and Schrieffer. The existing data do not prove that the spin fluctuations are suppressing the superconducting transition temperature; however, this seems to be the most reasonable explanation for the strong correlation between the decreasing T_c and the increasing ratio of χ/γ. We present calculated values for the variation of the bare density of states, the phonon interaction, the Coulomb interaction, and the spin susceptibility in these metals.

I. INTRODUCTION

IN this paper we would like to make a quantitative study of the behavior of the superconducting transition temperature T_c, the magnetic susceptibility χ, and the electronic specific heat γ in the fcc (noble) transition metals on which we reported in the preceding paper[1] (hereafter referred to as AJ). In particular, we would like to determine whether the presence of the virtual spin fluctuations which appear to play such an important role in palladium[2] is required to explain the rapid decrease of T_c as one alloys past iridium toward rhodium, platinum and palladium (increasing the electron density). As we have already pointed out in AJ, the increasing enhancement of the spin susceptibility with electron density indicates the presence of an increasing Coulomb repulsion between electrons. This repulsion will decrease the pairing interaction, but since the instantaneous Coulomb repulsion is not very effective in reducing the pairing caused by the retarded phonon attraction, we must determine quantitatively whether this reduction is sufficient to account for the observed rapid fall of T_c (see AJ, Fig. 11).

Below in Sec. II, we present the theory which we will use in the analysis. The basic integral equations for the pairing interaction and the mass enhancement including phonon, spin fluctuation, and Coulomb interactions are given in Sec. IIA. In Sec. IIB, we display simple derivations of the expressions for T_c and γ without the presence of spin fluctuations. In Sec. IIC we discuss the Coulomb pseudopotential. In Sec. IID we present the expressions for T_c and γ obtained by Berk and Schrieffer, including spin fluctuations. Finally, in Sec. IIE we discuss the magnetic spin susceptibility.

We then carry out two different analyses of our experimental data. First we investigate what one would conclude if the spin fluctuations were absent. It will be shown in Sec. IIIA that this analysis seems insufficient to explain the experimental results. Then we carry out an analysis in Sec. IIIB of the data using the Berk-Schrieffer theory to account for the effect of the spin fluctuations on the pairing interaction and on the mass renormalization. Although this point of view seems to give the most reasonable qualitative picture with which to understand our experimental data the quantitative agreement is not good. Some possible explanations for this disagreement are discussed. Finally, in Sec. IV, we state the conclusions one can draw from this work.

II. THEORETICAL FORMULATION

A. Basic Equations

Although the metals of interest are weak-coupling superconductors ($T_c/\Theta_D \ll 1/100$), we use the retarded BCS theory of superconductivity developed for strong-coupling superconductors by Schrieffer, Scalapino, and co-workers from the original work of Nambu and Eliashberg.[3] This theory allows one to include the

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[1] K. Andres and M. A. Jensen, preceeding paper, Phys. Rev. 165, 533 (1967), hereafter referred to as AJ.

[2] N. F. Berk and J. R. Schrieffer, Phys. Rev. Letters 17, 433 (1966). S. Doniach, in Proceedings of the Manchester Many-Body Conference, September, 1964 (unpublished).

[3] See, e. g., J. R. Schrieffer, *The Theory of Superconductivity* (W. A. Benjamin, Inc., New York, 1964) p. 180 ff., and references cited therein.

effect of the phonon interactions (and spin-fluctuation interactions) in a self-consistent way on both the superconducting pairing interaction and on the electronic mass. It is found that the mass enhancement from the phonon interaction is typically 50% or more and so represents an important effect.[4] This enhancement or energy renormalization as a function of frequency p_0, is represented by the renormalization parameter $Z(p_0)$ where, if $N_\gamma(0)$[5] is the density of states at the Fermi surface (taken as the zero of energy) obtained from specific-heat experiments and $N(0)$ is the bare density of states (phonon and spin-fluctuation interactions turned off), then

$$Z(0)=N_\gamma(0)/N(0).$$

The integral equations for the gap function $\Delta(p_0)$ and for the renormalization parameter $Z(p_0)$ can be written at temperature T assuming particle-hole symmetry[3,6] as

$$\Delta(p_0)=\frac{N(0)}{Z(p_0)}\int_{\Delta_0}^{\omega_c}dp_0'[K_+(p_0,p_0')-U_c]\times\mathrm{Re}\left[\frac{\Delta(p_0')}{(p_0'^2-\Delta^2(p_0'))^{1/2}}\right]\tanh\frac{p_0'}{2kT}, \quad (1)$$

$$[1-Z(p_0)]p_0=N(0)\int_{\Delta_0}^{\omega_c}dp_0'K_-(p_0,p_0')\times\mathrm{Re}\left[\frac{p_0'}{(p_0'^2-\Delta^2(p_0'))^{1/2}}\right]\tanh\frac{p_0'}{2kT}, \quad (2)$$

where the kernels are given by

$$K_\pm(p_0,p_0')=\int_0^{2k_F}\frac{qdq}{2k_F^2}\int_0^\infty d\omega[\sum_\lambda[\bar{g}_{q\lambda}]^2\mid \mathrm{Im}D_\lambda(q,\omega)\mid\mp\mathrm{Im}t(q,\omega)][(p_0'+p_0+\omega+i\delta)^{-1}\pm(p_0'-p_0+\omega+i\delta)^{-1}]. \quad (3)$$

$\mathrm{Im}D_\lambda(q,\omega)$ is the phonon spectral weight function, $\mathrm{Im}t(q,\omega)$ is the spin-fluctuation spectral weight function, $\bar{g}_{q\lambda}$ is the screened electron-phonon matrix element [q is momentum transfer, ω is frequency ($\hbar\equiv1$), λ is polarization], U_c is the Coulomb pseudopotential (discussed below), k_F is the Fermi momentum, p_0 (and p_0') are frequencies,[7] and ω_c is a cutoff somewhat larger than the phonon frequencies.

We see that $Z(p_0)$ enters the gap equation in the denominator. This can be explained by the following argument. In calculating the matrix elements which enter $K_\pm$, one should not use Bloch waves since the electron-phonon interaction mixes some phonon states with electrons. The actual wave functions are renormalized by $Z^{-1/2}$. Then if one uses the dressed density of states $Z(p_0')N(0)$ and the Bloch matrix elements squared with factors $[Z(p_0)Z(p_0')]^{-1}$ one finds the $Z(p_0)^{-1}$ factor shown in Eq. (1).

Numerical solutions of the above equations have been carried out by MacMillan[8] and by Garland[9] [both with $t(q,\omega)=0$] and also by Berk and Schrieffer[2,10] [$t(q,\omega)\neq0$]. All of these authors use a linearization procedure in which for $T\cong T_c$ one sets the gap to zero in the square root and in the lower limit of integration. One then finds from Eqs. (1) and (2)

$$\Delta(p_0)=\frac{N(0)}{Z(p_0)}\int_0^{\omega_c}\frac{dp_0'}{p_0'}\mid K_+(p_0,p_0')-U_c\mid\Delta(p_0')\times\tanh\frac{p_0'}{2kT_c}, \quad (4)$$

$$[1-Z(p_0)]p_0=N(0)\int_0^{\omega_c}dp_0'K_-(p_0,p_0')\tanh\frac{p_0'}{2kT_c}. \quad (5)$$

B. Model Solutions without Spin Fluctuations

We now study the case with $t(q,\omega)=0$. Inserting Eq. (3) into Eqs. (4) and (5) and reversing the order of integration yields

$$\Delta(p_0)=\frac{N(0)}{Z(p_0)}\left[\int_0^{2k_F}\frac{qdq}{2k_F^2}\int_0^\infty d\omega[\sum_\lambda[\bar{g}_{q\lambda}]^2\mid\mathrm{Im}D_\lambda(q,\omega)\mid]2\int_0^{\omega_c}\frac{dp_0'}{p_0'}\Delta(p_0')\tanh\frac{p_0'}{2kT_c}\left[\frac{p_0'+\omega+i\delta}{(p_0'+\omega+i\delta)^2-p_0^2}\right]-U_c\int_0^{\omega_c}\frac{dp_0'}{p_0'}\Delta(p_0')\tanh\frac{p_0'}{2kT_c}\right], \quad (6)$$

$$1-Z(p_0)=-N(0)\int_0^{2k_F}\frac{qdq}{2k_F^2}\int_0^\infty d\omega\left[\sum_\lambda[\bar{g}_{q\lambda}]^2\mid\mathrm{Im}D_\lambda(q,\omega)\mid\int_0^{\omega_c}\frac{dp_0'2\tanh(p_0'/2kT_c)}{(p_0'+\omega+i\delta)^2-p_0^2}\right]. \quad (7)$$

[4] In Pd the enhancement from the spin fluctuations may be a factor of 2 or 3 (Refs.2 and 25).
[5] That is, $\gamma=\frac{2}{3}\pi^2k_B^2N_\gamma(0)$, where the electronic specific heat at low temperatures is γT.
[6] The assumption of particle-hole symmetry is not necessarily a good approximation in the transition metals, as we will discuss later.
[7] The momentum variables have already been integrated out.
[8] W. L. MacMillan (to be published).
[9] J. W. Garland (to be published).
[10] N. F. Berk and J. R. Schrieffer, in *Proceedings of the Tenth International Conference on Low-Temperature Physics, Moscow, 1967* (Prozvodstrenno-Izdatel'skii Kombinat, VINITI, Moscow, USSR, 1967). See also S. Doniach and S. Englesberg, Phys. Rev. Letters **17**, 750 (1966).

The main contribution to the first p_0' integral in Eq. (6) comes from small p_0'; thus, we neglect p_0' compared to ω ($\cong\omega_{\rm ph}$). The p_0' integration can easily be done for the first iterative solution

$$\Delta(p_0')=\Delta(0) \quad \text{for} \quad p_0'<\omega_{\rm ph}$$
$$=0 \quad \text{for} \quad p_0'>\omega_{\rm ph}$$

yielding

$$\Delta(p_0)\cong\frac{N(0)}{Z(p_0)}\left[\frac{2g^2}{\omega_{\rm ph}}\Delta(0)\ln\frac{\omega_{\rm ph}}{kT_c}\left(\frac{\omega_{\rm ph}^2}{\omega_{\rm ph}^2-p_0^2}\right)-\Delta(0)U_c\ln\frac{\omega_{\rm ph}}{kT_c}\right], \tag{8}$$

where we have assumed $\mathrm{Im}D_\lambda(q,\omega)$ to be independent of q and sharply peaked for $\omega=\omega_{\rm ph}$ with $\bar{g}_{q\lambda}\cong g$. We define $V_{\rm ph}=2g^2/\omega_{\rm ph}$, then solving for T_c (taking $p_0\to 0$ limit) we have

$$kT_c\cong\omega_{\rm ph}\exp(-1/g), \tag{9}$$

$$g\equiv[N(0)V_{\rm ph}-N(0)U_c]/Z(0). \tag{10}$$

We can easily solve Eq. (7) for $Z(0)$ by replacing $\tanh(p_0'/2kT_c)$ by unity since here the large p_0' give the main contribution (more phase space). The integration is then trivial, yielding

$$Z(0)\cong 1+N(0)V_{\rm ph}, \tag{11}$$

where $V_{\rm ph}$ is defined above.

Thus Eq. (10) becomes

$$g=[N(0)V_{\rm ph}-N(0)U_c]/[1+N(0)V_{\rm ph}]. \tag{12}$$

This expression for g is not in agreement with MacMillan's[8] solution because we have taken a simpler approximation for $\Delta(0)$ in Eq. (4). The differences are of no quantitative importance for the metals with which we are concerned.

The presence of $Z(0)[=1+N(0)V_{\rm ph}]$ in the expression for T_c has some experimental support. Bucher *et al.*[11] studied the properties of many superconducting elements, alloys and compounds and found that after estimating the Coulomb repulsion, they obtained a general empirical formula for the phonon interaction of the form

$$K_{\rm ph}=aN_\gamma(0)/[1+bN_\gamma(0)],$$

where a and b were constants. Since from Eq. (12), we expect an expression of the form

$$K_{\rm ph}=N(0)V_{\rm ph}/[1+N(0)V_{\rm ph}].$$

We see that the right-hand sides of these two equations are equal if

$$a=b=N(0)V_{\rm ph}/N_\gamma(0)=V_{\rm ph}/[1+N(0)V_{\rm ph}].$$

We have already shown that $V_{\rm ph}\sim$const[12,13] for much of the region investigated by Bucher *et al.* and since $N(0)V_{\rm ph}$ is generally between 0.3 and 0.7, we see that a and b are not varying very much. Bucher *et al.* did not find $a=b$, but their estimate of the Coulomb interaction was only approximate.

In passing, we note that from Eq. (6) we have an approximate expression for $\Delta(p_0)$, which is

$$\frac{\Delta(p_0)}{\Delta(0)}\cong\frac{Z(0)}{Z(p_0)}\left[\frac{N(0)V_{\rm ph}[1-(p_0/\omega_{\rm ph})^2]^{-1}-N(0)U_c}{N(0)V_{\rm ph}-N(0)U_c}\right]. \tag{13}$$

Similarly, we can determine $Z(p_0)$ by integrating Eq. (7), yielding

$$Z(p_0)\cong 1+N(0)V_{\rm ph}\left[\frac{\omega_{\rm ph}}{2p_0}\ln\left|\frac{1+(p_0/\omega_{\rm ph})}{1-(p_0/\omega_{\rm ph})}\right|\right]. \tag{14}$$

We have plotted $Z(p_0)$ and $\Delta(p_0)$ from Eqs. (13) and (14) in Fig. 1 as a function of $p_0/\omega_{\rm ph}$ for $N(0)V_{\rm ph}=0.5$ and $N(0)U_c=0.2$.

C. Coulomb Pseudopotential

One of the important points in Eq. (1) is the presence of the Coulomb pseudopotential U_c instead of the larger matrix element V_c.[14] This replacement occurs because in obtaining Eq. (1), the momentum coordinates have already been integrated over and the frequency variables p_0' and p_0 are for electrons in the Fermi-surface energy shell (of width $\sim 2\hbar\omega_{\rm ph}$). The phonon interaction is for electrons in this shell but the Coulomb repulsion remains large for interactions far outside the shell. That is, electrons which reside in the shell can virtually scatter outside it and feel a strong Coulomb repulsion. In second-order perturbation theory, one would expect these virtual scatterings to lower the energy of the electrons inside the shell. Physically, the virtual scatterings allow the electrons to stay farther apart in space and so to reduce their net Coulomb repulsion, which is short range in space and time. Since the phonon attraction is long range in time (i.e., retarded) it can still be effective even if the electrons are physically instantaneously not close together. One

[11] E. Bucher, F. Heininger, J. Muller, and J. L. Olsen, in *Proceedings of the Ninth International Conference on Low Temperature Physics* (Plenum Press, Inc., New York, 1965).

[12] M. A. Jensen, Ph.D. thesis, University of California, La Jolla, 1965 (unpublished).

[13] M. A. Jensen and J. P. Maita, Phys. Rev. **149**, 409 (1966).

[14] This was first shown by V. V. Tolmachev in the book by N. N. Bogoliubov, V. V. Tolmachev, and D. V. Shirkov, *A New Method in the Theory of Superconductivity* (Consultants Bureau Inc., New York, 1959), p. 80, and later developed by P. Morel and P. W. Anderson (Ref. 18).

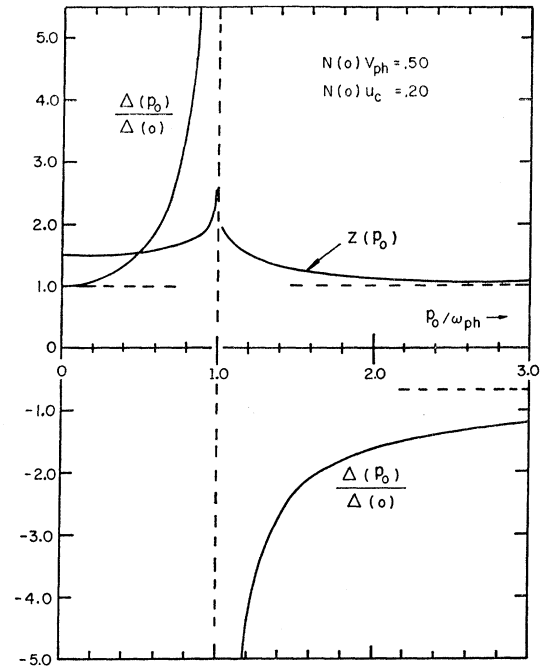

FIG. 1. The variation of the pairing strength $\Delta(p_0)$ and the mass renormalization $Z(p_0)$ for the linearized ($T \cong T_c$) equations given in the text. The first iterative solutions are shown with the parameters $N(0)V_{ph}=0.50$ and $N(0)U_c=0.20$. The dashed horizontal lines indicate the asymptotic values for $Z(\infty)(=1)$ and

$$\Delta(\infty)/\Delta(0)=-\tfrac{2}{3}.$$

electron just "follows" the other sufficiently far behind to not feel the Coulomb repulsion but close enough to make use of the phonon attraction.[15] To estimate the size of U_c, we go back to the original BCS integral equation (where momentum is the variable) with the renormalization included. We then have[12,13] the following integral equation where we have taken

$$\epsilon=\hbar^2k^2/2m,$$

$$\Delta(\epsilon)=[2Z(\epsilon)]^{-1}\int_{-\infty}^{\infty}\frac{N(\epsilon')K(\epsilon,\epsilon')\Delta(\epsilon')}{[\Delta^2(\epsilon')+\epsilon'^2]^{1/2}}\,d\epsilon'. \quad (15)$$

First, we take a square-well model with particle-hole symmetry represented in the following way:

$$\begin{aligned}
&K(\epsilon,\epsilon')=V_{ph}-V_c,\\
&N(\epsilon')=N(0),\\
&\Delta(\epsilon')=\Delta(0), \qquad |\epsilon|,|\epsilon'|<\omega_{ph};\\
&K(\epsilon,\epsilon')=-V_c,\\
&N(\epsilon')=N_1\\
&\Delta(\epsilon')=\Delta_1, \qquad \omega_{ph}<|\epsilon| \text{ and/or } |\epsilon'|<\omega_B;\\
&K(\epsilon,\epsilon')=0, \qquad \text{either } |\epsilon| \text{ or } |\epsilon'|>\omega_B. \quad (16)
\end{aligned}$$

[15] This simple picture needs some qualifications because the electron is not localized at one point but is spread out over a large part of the crystal.

This model is shown schematically in Fig. 2, where ω_{ph} is some characteristic cutoff energy for the phonon interactions and ω_B is the bandwidth $[O(E_F)]$. The solutions to Eq. (15) in this model have been worked out before[12,13,16] and one obtains [assuming $\Delta(0)\cong 1.75kT_c$] the same result as given in Eqs. (9) and (10), with U_c given by

$$U_c=\frac{V_c}{1+N_1V_c\ln(\omega_B/\omega_{ph})}. \quad (17)$$

As one approaches Pd, the bandwidth ω_B for excitations above the Fermi surface should decrease and a decreasing ω_B should increase U_c. For example, if $N(0)V_c$ is 0.5 and $N_1\cong N(0)$ with $\omega_B=50\omega_{ph}$ (1 eV), we find $N(0)U_c=(1/2.9)[N(0)V_c]\cong 0.17$, whereas, if $\omega_B=10\omega_{ph}$ (0.2 eV), then $N(0)U_c=(1/2.2)N(0)V_c\cong 0.23$. However, even for a metal like Pd where $\omega_B\sim 10\omega_{ph}$ for the particle excitations (those above the Fermi surface), the hole excitations have a large ω_B ($\sim 100\omega_{ph}$) and thus we should not assume particle-hole symmetry in solving Eq. (15). In this case we find

$$N(0)U_c=N(0)V_c \times\left[1+\tfrac{1}{2}N_{1,h}V_c\ln\frac{\omega_{B,h}}{\omega_{ph}}+\tfrac{1}{2}N_{1,p}V_c\ln\frac{\omega_{B,p}}{\omega_{ph}}\right]^{-1}, \quad (18)$$

which for $\omega_{B,p}=10\omega_{ph}$, $\omega_{B,h}=100\omega_{ph}$ and $N_{1,h}V_c=N_{1,p}V_c=0.5$ gives $N(0)U_c=N(0)V_c/2.72$; where $\omega_{B,p}$ and $\omega_{B,h}$ are the bandwidth for particle excitations and hole excitations, respectively, and where $N_{1,p}$ and

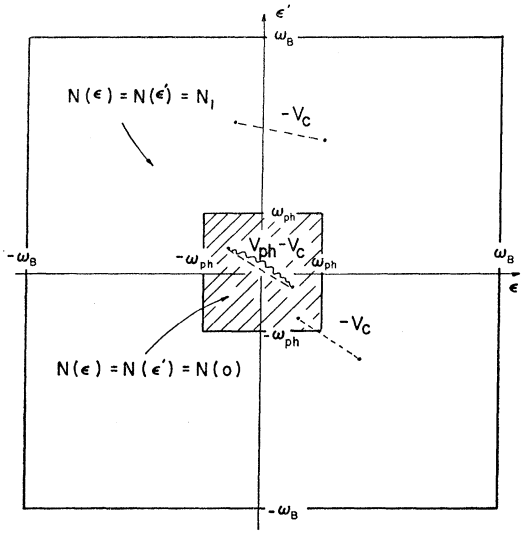

FIG. 2. Schematic of the two-square-well model for the interaction kernel which enters the integral equation for the pairing strength. Only when both electrons are near the energy shell ($|\epsilon|,|\epsilon'|<\omega_{ph}$) does the phonon attraction V_{ph} enter.

[16] G. Rickayzen, *Theory of Superconductivity* (Interscience Publishers, Inc., New York, 1965).

$N_{1,h}$ are the density of states for particle and hole excitations, respectively.

Up to now, we have neglected all but the d-band electrons. We have assumed $N_d(0) \gg N_s(0)$ (which is generally a good approximation), but we can try a simple calculation of $N(0)U_c$ for the following oversimplified model of Pd. We assume all the hole excitations fall in the d band and all the particle excitations fall in the s band. Then Eq. (18) becomes

$$N(0)U_c = N(0)V_c \times \left[1+\tfrac{1}{2}N_{1,d}V_{c,d}\ln\frac{\omega_{B,d}}{\omega_{\rm ph}}+\tfrac{1}{2}N_{1,s}V_{c,s}\ln\frac{\omega_{B,s}}{\omega_{\rm ph}}\right]^{-1} \quad (19)$$

where, e.g., $V_{c,d}$ is the Coulomb interaction between two d-hole excitations. We have had to assume that the Coulomb interaction between a d-hole and a s-particle is $(V_{c,s}V_{c,d})^{1/2}$.[17] Since from Morel and Anderson[18] one expects $N_{1,s}V_{c,s}=0.3$ and taking $\omega_{B,s}\sim 500\omega_{\rm ph}$, $\omega_{B,d}\sim 100\omega_{\rm ph}$, and $N_{1,d}V_{c,d}=0.5$, we have $N(0)U_c \cong (1/3.0)N(0)V$.

Although none of the above calculations could be expected to give more than a *crude* estimate of $N(0)U_c$, we find they all seem to convince one that a reasonable estimate for $N(0)U_c$ is given by

$$N(0)U_c \cong \tfrac{1}{3}N(0)V_c. \quad (20)$$

D. Critical Spin Fluctuations

Up to now we have not included the spin-fluctuation interactions, $t(q, \omega)$ in Eq. (3), in the equations for $\Delta(p_0)$ and $Z(p_0)$. Square-well calculations with spin fluctuations and phonons are possible but since the frequency dependence of $\mathrm{Im}t(q, \omega)$ is not a simple function (it was replaced by a δ function in the phonon case) such a model calculation is not very realistic. Numerical calculations have been carried out by Berk and Schrieffer[2,10] using an expression which they obtain for $t(q, \omega)$ by summing a certain class of diagrams. They assume that the major contribution to $t(q, \omega)$ comes from the following physical process. A singlet-state pair initially occupies the $k\uparrow$ and $-k\downarrow$ states. An exchange scattering occurs in which the $k\uparrow$ electron exchanges places with a $q\uparrow$ electron in the Fermi sea. In the intermediate state (when $q\uparrow$ has scattered to $k\uparrow$ but the $k\uparrow$ electron has not fallen into the $q\uparrow$ hole) the hole in the spin-up sea multiply-scatters with the $-k\downarrow$ electron (particle). In Fig. 3(a), a normal phonon scattering is shown in lowest order and in Fig. 3(b), the spin-fluctuation interaction just described is shown. Thus it is the particle-hole t matrix $t(q, \omega)$ which enters the pairing [just as it was the particle-particle (phonon) t matrix in the usual theory of superconductivity] and we can solve for the particle-hole t matrix from the integral equation shown in Fig. 4. One obtains

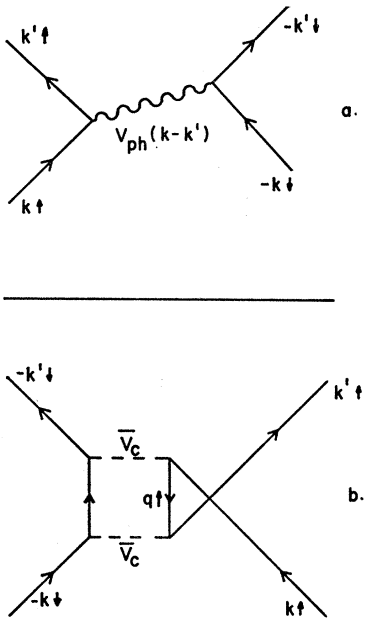

FIG. 3. (a) Lowest-order phonon scattering process and (b) the lowest-order particle-hole exchange scattering which when included to all orders cause the critical spin fluctuations.

$$t_{\rm p-h}(\mathbf{q}, \omega) = \bar{V}_c + \int \frac{d^4p}{(2\pi)^4} t_{\rm p-h}(q, \omega)\bar{V}_c G(\mathbf{p}, p_0)G(\mathbf{p}-\mathbf{q}, p_0-\omega) \quad (21)$$

from which, using

$$\int G(\mathbf{p}, p_0)G(\mathbf{p}-\mathbf{q}, p_0-\omega)\frac{d^4p}{(2\pi)^4} = N(0)U(q, \omega), \quad (22)$$

one obtains

$$t_{\rm p-h}(\mathbf{q}, \omega) = \bar{V}_c/[1-N(0)\bar{V}_c U(\mathbf{q}, \omega)]. \quad (23)$$

$U(q, \omega)$ is the momentum- and frequency-dependent Lindhard dielectric function[19] which approaches unity as q and $\omega \to 0 (\omega/qV_F \ll 1)$. In considering the interactions between the electrons of the Cooper pair one must be careful not to overcount. The pseudopotential U_c already includes certain multiple particle–particle scatterings and thus using $t(q, \omega)$ above overcounts, hence Berk and Schrieffer sum only the terms involving more than single p-h scatterings. Then the $t(q, \omega)$ which

[17] This may be an overestimate due to the orthogonality of s and d wave functions.

[18] P. Morel and P. W. Anderson, Phys. Rev. **125**, 1263 (1962).

[19] J. Lindhard, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **28**, 8 (1954).

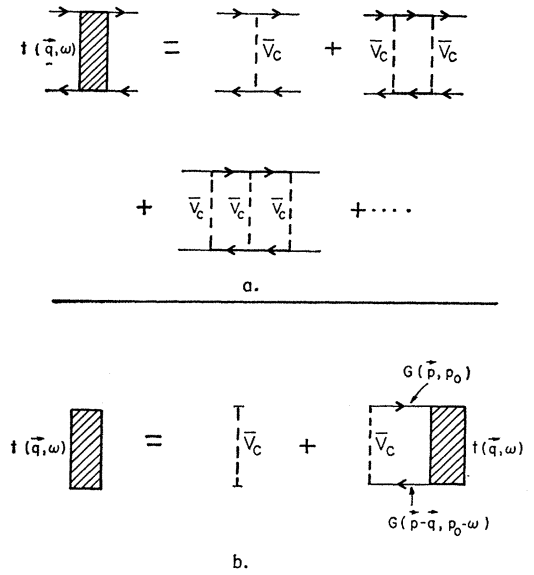

FIG. 4. (a) Definition of the particle-hole t matrix as an infinite sum of multiple scatterings of a particle with a hole (of opposity spin). (b) the integral equation which can be solved for $t(\mathbf{q}, \omega)$.

they put into (3) for $K_{\pm}$ is

$$t_{\mathrm{p-h}}(\mathbf{q}, \omega) = N(0)\bar{V}_c^2 U(\mathbf{q}, \omega)/[1-N(0)\bar{V}_c U(\mathbf{q}, \omega)]. \tag{24}$$

Plugging Eq. (24) into Eqs. (3), (4), and (5), Berk and Schrieffer[20] find after numerical integration that (with $k=\hbar=1$)

$$T_c \cong \omega_c \exp(-1/g), \tag{25}$$

$$g \equiv \frac{N(0)V_{\mathrm{ph}} - N(0)U_c - N(0)V_{\mathrm{spin}}}{1+N(0)V_{\mathrm{ph}} + N(0)V_{\mathrm{spin}}}, \tag{26}$$

$$\begin{aligned} Z(0) &= N_\gamma(0)/N(0) \\ &= (\tfrac{2}{3}\pi^2 K_B^2)^{-1}\gamma/N(0) \\ &= 1+N(0)V_{\mathrm{ph}} + N(0)V_{\mathrm{spin}}, \end{aligned} \tag{27}$$

where

$$\left.\begin{aligned} V_{\mathrm{spin}} &= \alpha \bar{V}_c \ln[1-N(0)\bar{V}_c]^{-1}, \\ \alpha &= 1, \quad N(0)\bar{V}_c \ll 1; \\ \alpha &\cong 2, \quad N(0)\bar{V}_c \sim 1. \end{aligned}\right\} \tag{28}$$

It is found from the numerical integration that $\hbar\omega_c \sim \frac{1}{10}k\Theta_D$,[20] and thus we will, in general, take $\hbar\omega_c = \frac{1}{3}k\Theta_D$ in the following for our analysis, in an attempt for a compromise with the usual BCS-type theory in which $\hbar\omega_c \sim k\Theta_D$. Such an approximation has no important quantitative effects on our results, since we deal only with weak-coupling superconductors.

[20] N. F. Berk, Ph.D. thesis, University of Pennsylvania, 1966 (unpublished).

E. Magnetic Spin Susceptibility

The particle-hole t matrix enhances the spin susceptibility, yielding an integral equation for $\chi(q, \omega)$ which can be solved for constant interaction $\bar{V}_c$, giving[2]

$$\chi(\mathbf{q}, \omega) = \chi_{\mathrm{Pauli}} U(\mathbf{q}, \omega)/[1-N(0)\bar{V}_c U(\mathbf{q}, \omega)], \tag{29}$$

where $\chi_{\mathrm{Pauli}} = 2\mu_B^2 N(0)$ and the other quantities have been defined above. Then the static uniform susceptibility $\chi(0, 0)$ has a contribution from the electron spin paramagnetism given by

$$\chi(0, 0) = 2\mu_B^2 N(0)/[1-N(0)\bar{V}_c]. \tag{30}$$

This is similar to the expression derived by Wolff in the random-phase approximation.[21]

For the same reason that phonons do not affect the spin susceptibility [i.e., $N(0)$ appears in Eq. (30), not $N_\gamma(0)$] the spin fluctuations also do not[20] and thus Eq. (30) does not involve V_{spin} defined in Eq. (28). This result follows when the self-energy is momentum-independent and therefore the integral in Eq. (22) is independent of whether G or G_0 is used.[20] Physically, one sees that because the mass enhancement $N_\gamma(0)/N(0)$ is tied independently to the Fermi surface for each spin direction it does not increase the polarization caused by a magnetic field.[22]

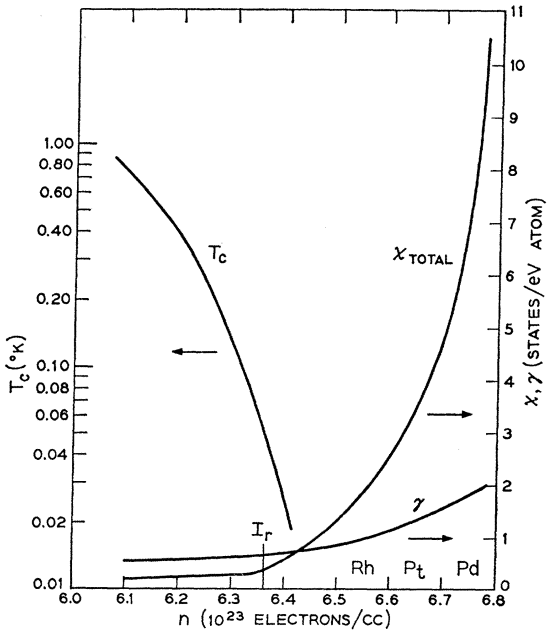

FIG. 5. Superconducting transition temperature T_c, total magnetic susceptibility χ_{total} and electronic specific heat γ. χ and γ are in units of states (eV atom) (i.e., we plot $\chi/2\mu_B^2$ and $3\gamma/2^2 k_B^2$). The data are taken from Ref. 1.

[21] P. A. Wolff, Phys. Rev. **120**, 814 (1960).
[22] M. A. Jensen (to be published).

III. COMPARISON WITH EXPERIMENTS

A. Without Spin Fluctuations

From Eqs. (20), (25)–(28), and (30), we have expressions which relate the experimental parameters T_c, Θ_D, γ, and χ to the parameters $N(0)$, V_c, V_{ph}, and $\bar{V}_c$. We first propose to analyze our data assuming that $V_{spin}=0$ in Eqs. (26) and (27); that is, we use the solutions for g and $Z(0)$ derived above and given in Eqs. (11) and (12). We later study the case where $V_{spin}\neq 0$. The data are shown in Fig. 5.

We take[20] $\omega_{ph}\cong\frac{1}{3}\Theta_D$, $U_c=\bar{V}_c/\beta$ and solve Eqs. (11), (12), and (30) and $N(0)$ obtaining

$$N(0)=\tfrac{1}{2}(1+\beta)N_\chi(0)\times\left\{1-\left[1-\frac{4N_\gamma(0)(L-1)}{N_\chi(0)L\beta[1+(1/\beta)]^2}\right]^{1/2}\right\}, \quad (31)$$

where

$$N_\chi(0)\equiv\chi/2\mu_B^2; \qquad N_\gamma(0)\equiv 3\gamma/2\pi^2k_B^2;$$

$$L\equiv\ln\Theta_D/3T_c.$$

From Eq. (20) we expect $\beta\lesssim 3$, so first we take $\beta=3$, giving

$$N(0)=2N_\chi(0)\{1-[1-\tfrac{3}{4}N_\gamma(0)(L-1)/N_\chi(0)L]^{1/2}\}. \quad (32)$$

Before we solve numerically for $N(0)$, let us discuss the limit when $L\gg 1$ (i.e., $T_c\ll 1°K$) and $N_\gamma(0)\ll N_\chi(0)$, which is appropriate past Rh, then Eq. (32) reduces to

$$N(0)\cong\tfrac{3}{4}N_\gamma(0). \quad (33a)$$

Also from Eqs. (30) and (11) one finds

$$N(0)\bar{V}_c=1-\tfrac{3}{4}[N_\gamma(0)/N_\chi(0)], \quad (33b)$$

$$N(0)V_{ph}=0.33, \quad (33c)$$

$$V_{ph}=\tfrac{4}{9}[N_\gamma(0)]^{-1}. \quad (33d)$$

Thus, since as one nears Rh $N_\chi(0)>N_\gamma(0)$ while both $N_\gamma(0)$ and $N_\chi(0)$ are rapidly increasing [$T_c\to 0$ ($\ll 1°K$)], if we neglect spin fluctuations, we can only explain these results by requiring a rapidly decreasing phonon interaction.

In the alloy region where T_c is large enough to be experimentally observable (see Fig. 6), neither approximation, $L\gg 1$ or $N_\chi(0)\gg N_\gamma(0)$, is a very good one and so we now go back to Eq. (31) and solve for $N(0)$, $\bar{V}_c$, and V_{ph}, using the experimentally determined parameters T_c, Θ_D, $N_\gamma(0)$, and $N_\chi(0)$. However, we have an additional complication in that the susceptibility which should be used [see Eq. (30)] in Eq. (31) is not the total measured susceptibility but only that part which is due to the spin paramagnetism. All metals have diamagnetic contributions to the measured susceptibility both from Landau diamagnetism and from the diamagnetism of the core electrons. Also, in transition metals with unfilled d shells, an "orbital" contribution to the susceptibility χ_{orb} arises because d-band widths are small enough so that excitations can occur in which M_z, the angular momentum component in the field direction, changes without a very large change in the energy. Kubo and Obata[23] have shown for a simple model (in the tight-binding approximation) that the orbital susceptibility χ_{orb} in transition metals should be of the same order as the d-electron Pauli susceptibility [$2\mu_B^2N(0)$]. Unfortunately there is no way at present to make even a reasonable calculation of the actual size of χ_{orb} in real metals. Since χ_{orb} is not expected to be very temperature-dependent[23,24] one can use the temperature dependence of $\chi_{exp}(T)$ and the temperature dependence of the NMR Knight shift to try to sort out the various contributions to the total measured susceptibility. This procedure has been applied to Pt, Pd, and Rh by Clogston, Jaccarino, Yafet, and co-workers,[11] who find that for these elements the diamagnetic and orbital (which is paramagnetic) susceptibilities just about cancel so that the total measured susceptibility is essentially all d-spin susceptibility. The s spin susceptibility is almost negligible since the s density of states is quite small. With this in mind, we will assume that the measured susceptibility of alloys near Rh (see Fig. 8 in AJ) is equal to the spin susceptibility and use the experimental

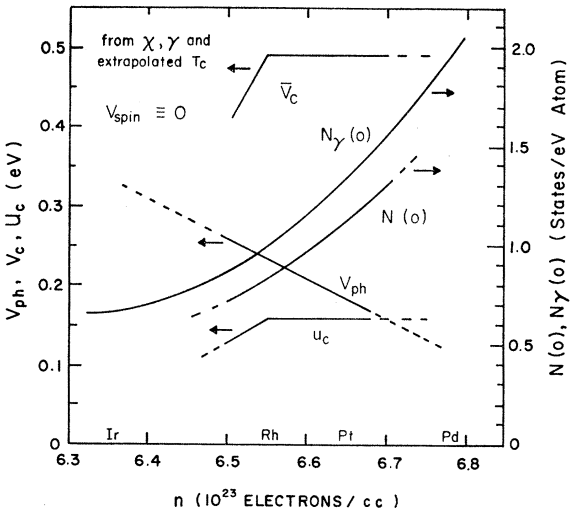

FIG. 6. Behavior of calculated parameters V_{ph}, $\bar{V}_c$, U_c, and $N(0)$ obtained using the theory given in the text without the presence of critical spin fluctuations (i.e., $V_{spin}\equiv 0$). The experimental data for χ, γ, and *extrapolated* T_c data are from a large number of alloys as given in Ref. 1 and described in the text.

[23] R. Kubo and Y. Obata, J. Phys. Soc. Japan **11**, 547 (1956).

[24] Calculations for Ni metal indicate there may be some temperature dependence of $\chi_{orb}[\chi_{orb}(T=0°K)\sim 0.8\chi_{orb}(10^3\ °K)]$; N. Mori, J. Phys. Soc. Japan **20**, 1838 (1965).

TABLE I. Experimental data[a] and the values of theoretically computed parameters calculated using the equations derived in the text with the assumption that $V_{spin}=0$. Some of these results are shown in Fig. 6. The data are taken from the Os–Ir–Pt system (binary alloys only) and extrapolations from these results.

n (10^{23} electrons/cc)	$\ln(\Theta_D/3T_c)$[b]	$N_\gamma(0)$[c] (states/eV atom)	$N_\chi(0)$[c] (states/eV atom)	$N(0)$ (states/eV atom)	$N(0)V_{ph}$	V_{ph} (eV)	$N(0)\bar{V}_c$	$\bar{V}_c$ (eV)	U_c (eV)
6.50	13.19	0.85	1.00	0.718	0.184	0.256	0.282	0.39	0.13
6.55	18.50	0.98	1.35	0.820	0.195	0.238	0.393	0.48	0.16
6.60	29.20	1.15	1.80	0.961	0.197	0.205	0.466	0.48	0.16
6.625	40.57	1.24	2.15	1.03	0.203	0.196	0.520	0.505	0.17
6.65	75.8	1.35	2.60	1.12	0.205	0.183	0.570	0.49	0.16
6.675	24.9	1.45	3.20	1.20	0.208	0.170	0.630	0.48	0.16

[a] Reference 1.
[b] Extrapolated from data for $n=6.1$ to 6.45×10^{23} electrons/cc.
[c] Interpolated from existing data. $N_\gamma(0)=3\gamma/2k^2\pi^2$, $N_\chi(0)=\chi/2\mu_B^2$.

values directly in our calculation of $N(0)$. We *extrapolate* our T_c data into this region, although this extrapolation only estimates R_c for alloys past iridium (see Fig. 5).

Then solving Eq. (32) for $N(0)$ and using Eqs. (11), (20), and (30) we can determine V_{ph}, $\bar{V}_c$, and U_c for the alloys from $n=6.45\times10^{23}$ electrons/cc to $n=6.65\times10^{23}$ electrons/cc (see Fig. 11 in AJ). The input data and the results are given in Table I and are shown in Figs. 6 and 7.[25] The general features agree with the above approximate solutions. The points of special interest here are the behavior of V_{ph} and $\bar{V}_c$. We see that the decrease in T_c has to be attributed to a rapidly decreasing V_{ph}, the size of which ($\sim$0.4 eV for Ir) is not in bad agreement with the values found in other transition metals (0.59 and 0.85 eV) by similar procedures.[8,12,13] While it is true that screening should increase (and so V_{ph} should decrease) when the density of states increases, the screening in transition metals depends on the band structure far from the Fermi surface and hence should not be very sensitive to changes in $N(0)$.[26] For example, the changes in V_{ph} in the range $4<z<6$ were found to be small even though in this region $N(0)$ varies by an order of magnitude.[12,13] Thus, the drop in V_{ph} found past iridium in this analysis would have to be due to some other reason than the increasing $N(0)$. This brings up the main reason why these results do not satisfactorily explain the experiments. We found experimentally (AJ) that there is a strong correlation between the rapid decrease of T_c and the increase of χ, γ, and χ/γ. If the above analysis with $V_{spin}=0$ is correct, then the decrease in T_c is caused by a dropping V_{ph} while the increase in χ/γ is caused by an increasing $N(0)$ ($\bar{V}_c\sim$const), and the strong *correlation* mentioned above would be somewhat fortuitous. If, contrary to the behavior elsewhere in the transition metals, the size of V_{ph} does depend (inversely) on $N(0)$, then it would be the behavior of $N(0)$ alone which causes the drop in T_c and the increase in χ, γ, and χ/γ. However, as we will see, there is a different explanation for the behavior of χ, γ, χ/γ, and R_c which does not raise the above objections.

B. Including Spin Fluctuations

We now include the effect of spin fluctuations and we consider two different regions of our data. First, we study the alloys for which we have T_c data; specifically, below we consider the Os–Ir–Pt system. For these alloys, χ_{spin} is not very much exchange enhanced and so the diamagnetic and orbital contributions to the measured susceptibility are not small compared with χ_{spin}. Since the size of the orbital susceptibility is unknown, we cannot extract from the measured susceptibility the size of χ_{spin} needed in order to make use of Eq. (30). Therefore, for this region, we use the experimental values of T_c, Θ_D ($\cong$410°K) and γ together with Eqs. (20), (25)–(28) to obtain $N(0)$, V_c (assuming $V_c=\bar{V}_c$), and χ_{spin}. If we solve for $N(0)$,

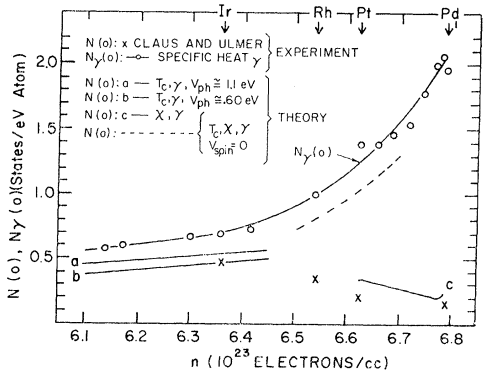

FIG. 7. The bare $[N(0)]$ and enhanced $[N_\gamma(0)]$ density of states both from experiment and from calculations described in the text. The experimental data are given in Ref. 1 (circles) and in Ref. 31 (crosses).

[25] The behavior of $N(0)$ and $\bar{V}_c$ is also shown in Figs. 8 and 9, where it is compared with results of some later calculations.
[26] We have a rigid-band model in mind during this discussion.

TABLE II. Experimental data[a] and the values calculated for several theoretical parameters using the theory of Berk and Schrieffer[b] together with expressions derived in the text. The data are from the Os–Ir–Pt system (binary alloys only).

Z (electrons/atom)	n (10^{23} electrons/cc)	$N_\gamma(0)$[c] (states/eV atom)	g	$N(0)$[d] (states/eV atom)	$N(0)\bar{V}_c$[e]	$N(0)U_c$	$N(0)V_{\text{spin}}$	$N(0)V_{\text{ph}}$	V_{ph} (eV)
8.60	6.107	0.55	0.185	0.373	0.235	0.078	0.065	0.412	1.10
8.70	6.171	0.61	0.175	0.399	0.224	0.091	0.090	0.444	1.10
8.80	6.235	0.63	0.165	0.405	0.294	0.098	0.105	0.454	1.12
8.90	6.299	0.66	0.154	0.416	0.312	0.104	0.120	0.468	1.12
9.00	6.363	0.69	0.144	0.426	0.345	0.115	0.150	0.483	1.13
9.10	6.389	0.71	0.133	0.432	0.357	0.118	0.160	0.491	1.14
9.20	6.415	0.73	0.123	0.437	0.379	0.128	0.180	0.501	1.14
9.30	6.441	(0.77)[f]	(0.112)	0.450	0.392	0.131	0.195	0.515	1.14

[a] Reference 1.
[b] References 2, 10, and 20.
[c] $N_\gamma(0)=\gamma/\frac{2}{3}\pi^2K_B^2$.
[d] $V=2V_{\text{ph}}-U_c=2.0$ eV.
[e] $U_c=\frac{1}{3}\bar{V}_c$.
[f] ( ) means extrapolated value.

eliminating V_{spin} from Eqs. (25) and (26), we find

$$N(0)=\frac{-1+[1+4N_\gamma(0)(1+g)(2V_{\text{ph}}-U_c)]^{1/2}}{2(2V_{\text{ph}}-U_c)}, \quad (34)$$

which can be solved if we fix the parameter $V\equiv(2V_{\text{ph}}-U_c)$. We then calculate $N(0)$ from Eq. (34) with V fixed at reasonable values between the possible extremes of 1.0 and 2.0 eV, which corresponds to $V_{\text{ph}}\cong 0.60$ and 1.1 eV, respectively. The resulting values for $N(0)$ are shown in Fig. 7, curves a and b, for the region $n=6.1$ to 6.4×10^{23} cc^{-1} (which corresponds to $Z=8.6$ to 9.2 in the Os–Ir–Pt alloy series). The data are also given in Table II, for $V=2.0$ eV.

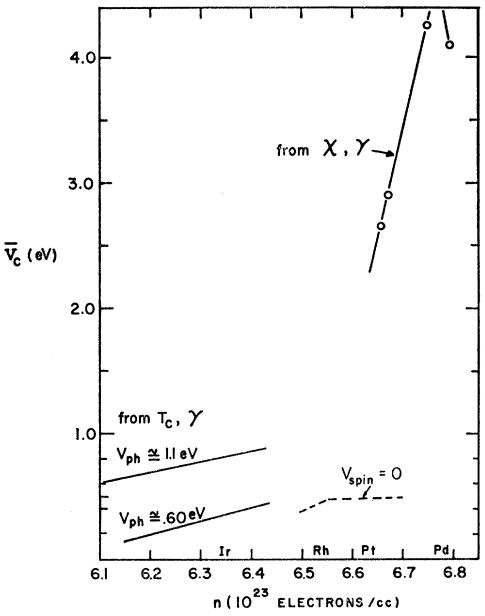

FIG. 8. The irreducible electron-hole Coulomb interaction $\bar{V}_c$ (i.e., with correlation phenomenologically included). The curves labeled with values chosen for V_{ph} (the adjustable parameter) are calculated using experimental T_c, Θ_D, and γ data and the theory described in the text. The curve labeled "from χ, γ" is calculated directly from the Berk–Schrieffer theory, while the curve labeled "$V_{\text{spin}}=0$" is obtained from the theory developed in the text without the inclusion of critical spin fluctuations.

Using the above calculated $N(0)$ values, we can calculate $\bar{V}_c$. We assume that U_c and V_c are related by Eq. (20) and we assume $V_c=\bar{V}_c$, and are then able to solve for $\bar{V}_c$ with only V_{ph} (since $V=2V_{\text{ph}}-\frac{1}{3}\bar{V}_c$) as the adjustable parameter. We have plotted the results of this calculation in Fig. 8 (see also Table II) for values of $2V_{\text{ph}}-\frac{1}{3}V_c=1$ and 2 eV. The curves are labeled by the approximate choice of V_{ph}.

We also wish to study the behavior of the calculated spin susceptibility near iridium. We use the χ_{total} and γ values again from the Os–Ir–Pt system in order to maintain as much accuracy as possible, rather than use some average curves containing the data from all the ternary alloys plotted versus electron density. The results of this calculation are shown in Fig. 9, where we have also plotted γ and χ_{total} (total measured susceptibility). The values calculated using the above theory fall within the hatched region which shows the variation of χ_{spin} with possible choices of V_{ph} (also we have allowed $2U_c<V_c<3U_c$). We notice that the width of the hatched region is small, indicating the relative independence of χ_{spin} on the assumed choice of these parameters. Thus, we have a prediction for the spin susceptibility for the alloys from $Ir_{0.65}Os_{0.35}$ to $Ir_{0.80}Pt_{0.20}$. The measured susceptibility values for the Os–Ir–Pt system are given in AJ and are shown in Fig. 9. We see that the theory predicts a variation of the spin susceptibility in the Os–Ir alloys which is parallel to the total measured susceptibility. This is as we might expect, since the difference due to χ_{orb} and χ_{diam}[27] (as discussed above) should vary only slowly upon alloying. However, the theoretical curve does not rise nearly as abruptly past iridium as does the experimental data.

{*Note added in proof* (1) We have recently pointed

[27] A. M. Clogston, V. Jaccarino, and Y. Yafet, Phys. Rev. **134**, A650 (1964); J. A. Seitchik, A. G. Gossard, and V. Jaccarino, *ibid.* **136**, A1119 (1964); J. A. Seitchik, V. Jaccarino, and J. H. Wernick, *ibid.* **138**, A148 (1965).

TABLE III. Experimental data[a] and the values calculated for several theoretical parameters using the theory of Berk and Schrieffer[b] for the effect of critical spin fluctuations on the electronic specific heat. The data are from the Rh–Pd system.

Z (electrons/atom)	n (10^{23} electrons/cc)	$N_\gamma(0)$[c] (states/eV atom)	$N_\chi(0)$[d] (states/eV atom)	$N_\gamma(0)^*/N(0)$	$N(0)$ (states/eV atom)	$N(0)V_{spin}$	$N(0)\bar{V}_c$	$\bar{V}_c$ (eV)
10.0 (Pd)	6.79	12.1	1.96	8.1	0.24	7.1	0.980	4.1
9.95	6.78	20.9	2.07	10.0	0.21	9.0	0.990	4.7
9.91	6.77	16.9	2.00	9.5	0.21	8.5	0.988	4.7
9.82	6.75	10.4	1.77	7.9	0.23	6.9	0.979	4.25
9.50	6.67	3.94	1.42	4.5	0.32	3.5	0.92	2.90

[a] Reference 1.
[b] References 2, 10, and 20.
[c] $N_\gamma(0)=\gamma/\frac{2}{3}\pi^2K_B^2$.
[d] $N_\chi(0)=\chi/2\mu_B^2$.
[e] From numerical calculations by N. F. Berk and J. R. Schrieffer.

out [M. A. Jensen, in Proceedings of the International Magnetism Conference, Boston, 1967 (to be published)] that the rapid increase in χ past iridium (i.e. as Pt, Rh or Pd is added) is due to a breakdown of the rigid-band model.}

We also wish to study the behavior of the parameters $N(0)$ and $\bar{V}_c$ for the strongly exchange-enhanced Pd-rich alloys ($\chi/\gamma\gg1$). If we neglect V_{ph} compared with V_{spin} (since $V_{spin}\gg1$ eV in this region) then we have from Eqs. (27) and (30)

$$N_\chi(0)/N_\gamma(0)=\{[1-N(0)\bar{V}_c][1+N(0)V_{spin}]\}^{-1}. \tag{35}$$

Since Eq. (28) for V_{spin} is not a very good approximation near $N(0)\bar{V}_c\sim1$, we use the actual numerical results of Berk and Schrieffer[20] and find the values for $N(0)$ shown in Fig. 7 (curve c). The relevant data are given in Table III.

We see that the above analysis (with $V_{spin}\neq0$) predicts $N(0)$ to be a rather slowly varying function of position in the fcc transition metals which decreases from Ir to Pd. The very large value of γ measured for Pd-rich alloys is, according to this theory, caused by many-body effects arising from the critical spin fluctuations. Claus and Ulmer[28] have measured what is claimed to be the bare density of states[20] in Ir, Rh, Pt, and Pd by x-ray emission spectroscopy. They cannot measure an absolute value, but quote the density of states for Rh, Pt, and Pd relative to that for Ir. If we calibrate their results with our calculated value for Ir (using $V_{ph}\cong1.0$ eV), which leads to $N(0)=0.48$ states/(eV atom) then they predict $N(0)=0.33$, 0.21 and 0.15 states/(eV atom) for Rh, Pt, and Pd, respectively. These values are also shown in Fig. 7 and can be seen to agree qualitatively with the results calculated from the Berk-Schrieffer theory (curves a, b, and c). Alternatively, we see that $N(0)$ calculated from T_c,[29] Θ_D, γ, and χ with the assumption that $V_{spin}=0$ (dashed curve) is in strong disagreement with the results of Claus and Ulmer.[30,31] However, recent photoemission studies on Pd by Yu and Spicer[32] do show a peak in the measured density of states near the Fermi surface and thus disagree with the work of Claus and Ulmer. Although the behavior of $N(0)$ found above is consistent with some of the experimental properties of these metals[20] it seems likely for several reasons (some of which we will mention) that the theory overestimates the size of the mass enhancement. The difficulty may lie in

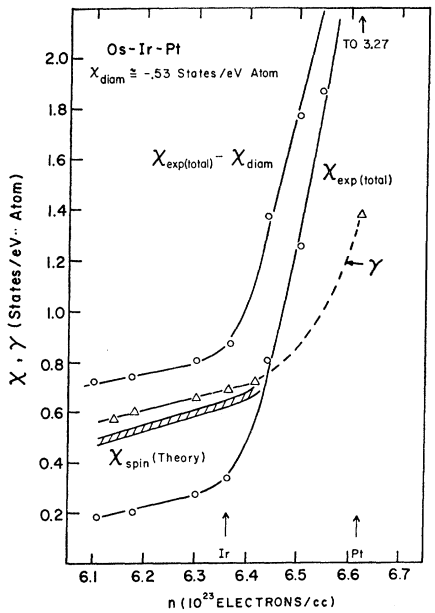

FIG. 9. Magnetic susceptibility $\chi_{exp(total)}$ is the total measured susceptibility. $\chi_{exp(total)}-\chi_{diam}$ is corrected for the diamagnetic contribution estimated from the diamagnetism of Au(−0.53 states/eV atom) (Ref. 27). The triangles are experimental γ values (Ref. 1). The heavy curve labeled χ_{spin} (theory) is calculated from T_c and γ using the Berk–Schrieffer theory as described in the text. The width indicates the probable uncertainty in the calculation, due to the uncertainty in V_{ph}.

[28] χ_{spin} here refers to d-electron paramagnetism; we neglect here the small contribution from the s electrons.
[29] Extrapolated T_c values.
[30] R. Huguenin and D. Baldock, Phys. Rev. Letters **16**, 795 (1966).
[31] H. Claus and K. Ulmer, Z. Phys. **185**, 139 (1965).
[32] A. Y. C. Yu and W. E. Spicer, Phys. Rev. Letters **17**, 1171 (1960).

the neglect of the details (e.g., anisotropy) of the Fermi surface of the metals near Pd. {*Note added in proof* (2) Experiments on the Rh–Ni system by Bucher *et al.* [E. Bucher, W. F. Brinkman, H. J. Williams, and J. D. Maita, Phys. Rev. Letters **18,** 1125 (1967)] indicate that the model calculations which give Eq. (28) somewhat overestimate the size of V_{spin}. From the work of Doniach [S. Doniach, Phys. Rev. Letters **18,** 554 (1967)] and Schrieffer [J. R. Schrieffer, in Proceedings of the International Magnetism Conference, Boston, 1967 (to be published)] it appears that the overestimation of V_{spin} may be mainly due to the use of a contact potential which underestimates the fall of $t(\mathbf{q}, \omega)$ with $\mathbf{q}$.}

For the region near Pd where $\chi \gg \gamma$, we can also determine $\bar{V}_c$. Using Eqs. (27), (28), and (30) we find the results shown in Fig. 8 (open circles). We see that $\bar{V}_c$ (we did not assume $V_c = \bar{V}_c$) near Pd is $\sim$4 eV, which is almost an order of magnitude larger than had been thought.[27] A value this large, however, is consistent with the large value for the $\bar{V}_c$ found for Ni in **Be**–Ni alloys by Klein and Heeger.[33,34] We see that the results for $\bar{V}_c$ from the calculation with $V_{\text{spin}}=0$ (dashed curve in Fig. 8) are much smaller than the values found using the Berk-Schrieffer theory. Also, $\bar{V}_c$ apparently increases abruptly as one moves past iridium which is probably due to the fact that $\bar{V}_c$ being intra-atomic depends more on the atomic properties (and also on the lattice spacing as discussed in AJ) than on the band properties. Thus, in Ir–Pt alloys, one might consider Pt as a kind of virtual magnetic impurity which can be polarized for a short time by the multiple scattering of electrons. [*Note added in proof* (3) The fact that the more rapid increase in χ past iridium is not accompanied by a more rapid fall of T_c past iridium may be due to the fact that local spin fluctuations, e.g., on a Pt site, are large momentum and hence higher frequency excitations and hence have short lifetimes compared with the long wavelength low-frequency spin fluctuations which are most disruptive of the superconducting pairing.]

[33] A. Klein and A. J. Heeger, Phys. Rev. **144,** 458 (1966).
[34] S. Doniach, Phys. Rev. Letters **18,** 554 (1967).

IV. SUMMARY AND CONCLUSIONS

In the two preceding sections we have studied the behavior of the parameters $N(0)$, V_{ph}, V_c, $\bar{V}_c$, and U_c, which one determines from the experimental data for T_c, Θ_D, γ, and χ, using two different theoretical models. In the first model we included only the retarded phonon interaction and the instantaneous Coulomb interaction in the pairing equations. In the second model we added the dynamic interaction caused by the presence of ferromagnetic spin fluctuations following the theory of Berk and Schrieffer.

Using the first model we can explain the abrupt decrease in the superconducting transition temperature as one fills up the d band only by a decreasing phonon interaction. We therefore find no simple explanation for the strong empirical correlation between the decreasing transition temperature and the increasing exchange enhancement of the spin susceptibility. If, however, the screening which reduces V_{ph} is very sensitive to the density of states at the Fermi surface (unlike the behavior observed in most transition metals) then the increase in the (bare) density of states as one fills up the d band could explain the correlation between decreasing T_c and increasing χ.

On the other hand when the critical spin fluctuations are included we find a very natural explanation for the strong correlation between the decreasing transition temperatures and the increase of χ, γ, and χ/γ as one fills up the d band. However, the rapid increase of χ past iridium (e.g., in Ir–Pt alloys) would lead one to predict an even more rapid decrease of the superconducting transition temperature than is observed. [See note added in proof (3)]. This theory also predicts an unexpectedly large mass enhancement near Pd which may indicate that it overestimates the importance of the spin fluctuations. [See note added in proof (2).]

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