THE RIGID MUFFIN-TIN APPROXIMATION FOR THE ELECTRON-PHONON INTERACTION IN TRANSITION METALS*

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1.   Introduction

Substantial progress has been achieved in recent years in our ability to calculate the electron-phonon parameters of transition metals. This progress has been based primarily on one or the other of two *ad hoc* prescriptions for the electron-phonon matrix elements, the "rigid muffin-tin approximation" (RMTA) *or* the "fitted modified tight-binding approximation" (FMTBA). Here, I shall mainly be concerned with the RMTA. Comparisons between RMTA calculations and several types of experiments show that the RMTA is remarkably accurate for average electron-phonon properties such as the electron-phonon mass enhancement λ. There are indications, however, that the RMTA matrix elements may be too small at low momentum transfers.

In this paper I shall attempt to demonstrate these assertions concerning the accuracy of the RMTA and shall offer some simple observations which may help to explain why they are true. I shall also try to place the numerous electron-phonon calculations in a broader perspective by showing how they can be used to explain the trends in the strength of the electron-phonon coupling among the transition metals and the A-15 compounds.

2.   Average Electron-Phonon Properties in the RMTA

The basic problem in the theory of the electron-phonon interaction is the calculation of the transition rate between Bloch states ψ_k and $\psi_{k'}$ due to an infinitesimal atomic displacement δR. The matrix element for this transition is

$$I^{\alpha}_{k'k} = \langle \psi_{k'} \mid \delta V/\delta R_{\alpha} \mid \psi_k \rangle , \qquad (1)$$

where $\delta V/\delta R_{\alpha}$ is the change in (self-consistent) crystal potential per unit displacement due to the displacement of a single atom in direction α. In the RMTA, $\delta V(r)/\delta R_{\alpha}$ is replaced by the gradient of the usual muffin-tin type potential which would enter an augmented plane wave or Korringa-Kohn-Rostocker band calculation.

The RMTA has been used to calculate a host of electron-phonon interaction dependent properties for a wide variety of materials. Calculations of $\langle I^2 \rangle$, the Fermi surface average of $\Sigma_{\alpha} \mid I^{\alpha}_{k'k} \mid^2$ have been extremely popular because Gaspari and Gyorffy (1972) showed how it could be obtained rather simply in the course of a typical band structure calculation.

The electron phonon matrix element $g^j_{k'k}$ is defined in terms of $I^\alpha_{k'k}$ by

$$g^j_{k'k} = \sum_\alpha \hat{\epsilon}^j_\alpha(k' - k) I^\alpha_{k'k} \sqrt{\frac{\hbar}{2M_a \omega^j_{k'-k}}} \quad , \qquad (2)$$

where $\hat{\epsilon}^j(q)$ is a phonon polarization vector, ω^j_q is a phonon frequency and M_a is the atomic mass. The absolute square of $g^j_{k'k}$ determines the transition rate between Bloch state ψ_k and $\psi_{k'}$ due to the presence of a phonon of wave vector $k' - k$ and polarization j. In principle, the energy of the Bloch state ψ_k should differ from that of state $\psi_{k'}$ by $\omega^j_{k'-k}$, but phonon energies are usually small on an electronic structure scale so in practice both of the states, k and k' can be taken to be at the Fermi energy for most transition metal systems.

Most of the experimentally observable quantities related to the electron-phonon interaction can be viewed as averages over the Fermi surface of $|g^j_{k'k}|^2$. The mass enhancement parameter which gives the electron-phonon enhancement of the specific heat and which enters the McMillan (1968) equation for the superconducting transition temperature is given by

$$\lambda = \frac{2}{N(0)} \sum_{kk'j} \delta(\epsilon_k)\delta(\epsilon_{k'}) \frac{|g^j_{k'k}|^2}{\hbar \omega^j_{k'k}} \quad , \qquad (3)$$

where $N(0)$ is the (single-spin) Fermi energy density of states. A quantity very similar to λ determines the high temperature electrical resistivity. For temperatures greater than the Debye temperature, the resistivity is given rather accurately by a relaxation time approximation

$$\rho = \frac{3}{2e^2 N(0)\langle v^2 \rangle \tau_{ep}} \quad , \qquad (4)$$

where $\langle v^2 \rangle$ is the mean square Fermi velocity and where the electron-phonon lifetime τ_{ep} is determined by the transport version of λ

$$\hbar/\tau_{ep} = 2\pi k_B T \lambda_{tr} \quad , \qquad (5)$$

$$\lambda_{tr} = \frac{1}{N(0)\langle v^2 \rangle} \sum_{kk'j} \delta(\epsilon_k)\delta(\epsilon_{k'})(v_k - v_{k'})^2 \frac{|g^j_{k'k}|^2}{\hbar \omega^j_{k'k}} \quad . \qquad (6)$$

Other electron-phonon properties can be written in terms of more restricted averages over $|g^j_{k'k}|^2$ as will be shown in Sect. 3.

The question "How well does the RMTA work?" is answered in part by Table I which shows values of λ as calculated using the RMTA and as deduced from experiment for various transition metals. Overall the

Table 1. Values of λ Calculated Using the Rigid Muffin-Tin or
Rigid Atomic Sphere Approximations and
Deduced from Experiment*

Material	Reference	$\lambda_{calc.}$	$\lambda_{emp.}$
V	Glötzel et al. (1979)	1.2^a	0.83^t, $\geq 0.8^r$
Nb	Butler et al. (1977)	1.2	1.2^t, 1.0^r, 0.96^c
	Butler et al. (1979)	1.1	
	Glötzel et al. (1979)	1.3^a	
Ta	Glötzel et al. (1979)	0.9^a	0.7^t $(0.7 - 0.9)^r$
Mo	Pinski et al. (1978a)	0.40	0.44^c
	Glötzel et al. (1979)	0.4^a	
W	Glötzel et al. (1979)	0.25^a	0.24^c
Pd	Pinski et al. (1978b)	0.41	0.38^r
	Glötzel et al. (1979)	0.5^a	
Pt	Glötzel et al. (1979)	0.7^a	

*(a) Rigid atomic sphere approximation, (t) estimate from
tunneling data, V (Zasadzinski et al., 1980), Nb (Bostock et al.,
1980), Ta (Shen, 1972), (r) estimate from resistivity, (c)
estimate from T_c.

agreement between calculation and experiment is quite impressive. While
considering these results one should remember that neither the calcula-
tion nor the experiments are completely free from ambiguity. In the
calculations, the phonon frequencies and polarization vectors were
obtained from Born-von Kármán fits to the experimental phonon curves but
the electronic structures were determined from energy band calculations
with their consequent uncertainties. In particular, it should be noted
that the calculations of Glötzel et al., are based on a linearized ver-
sion of KKR theory which employs overlapping atomic spheres rather than
the more usual muffin-tin spheres. Their approximation for the
electron-phonon matrix element might more properly be called the "rigid
atomic sphere approximation".

The ambiguities in the experimental values for λ arise from various
sources. Specific heat data can be used with a band calculation to
deduce λ from

$$1 + \lambda = N^*(0)/N_B(0) , \qquad (7)$$

where $N^*(0)$ is the enhanced density of states and $N_B(0)$ is the band
structure density of states. This method of obtaining λ, however,
requires an accurate $N_B(0)$ and begs the question of the existence of
other many-body enhancements (Rietschel and Winter, 1979).

The most common method of estimating λ is via the superconducting
transition temperature using an equation originally due to McMillan
(1968) but modified by Allen and Dynes (1972)

$$T_c = \frac{\hbar\langle\omega\rangle_{\log}}{1.2k_B}\exp\left[-\frac{1.04(1+\lambda)}{\lambda - \mu^* - 0.62\,\lambda\mu^*}\right] . \tag{8}$$

Once T_c has been measured λ can be determined from Eq. (8) if $\langle\omega\rangle_{\log}$ (a particular average of the phonon frequencies) and μ^* (the residual Coulomb repulsion) are known. Unfortunately, they (μ^* especially) never are known accurately for transition metals. In principle λ, $\langle\omega\rangle_{\log}$, and μ^* can all be determined from an analysis of superconducting tunneling data, but there are difficult experimental problems and the analysis of the data is not nearly so straightforward for transition metals as for the simple metals.

There are few experimental difficulties in measuring the room temperature resistivity so a comparison of the calculated and experimental resisitivity should provide a significant test for the RMTA. Calculated and experimental resistivities for Nb and Pd (Pinski et al., 1980a) are shown in Fig. 1. If we assume that $N(0)\langle v^2\rangle$ has been calculated accurately and that the resistivity indeed varies in a manner described by Eqs. (4) and (5) at 300 K we can conclude that the RMTA values of λ for both of these metals are about 10% too large. There is a possible ambiguity even in this test of the RMTA since the resistivities of both Nb and Pd show substantial negative deviations from linearity at temperatures above 300 K. These deviations may be due to "Mott Fermi smearing" or to "non-classical conduction channels" (Allen, 1980) neither of which are included in Eq. (4). If these effects are influencing the resistivity at 300 K, the RMTA values for λ might be even more accurate than Fig. . suggests.

Improving upon the RMTA in the sense of calculating more accurate values of λ for transition metals may prove to be quite difficult. It is
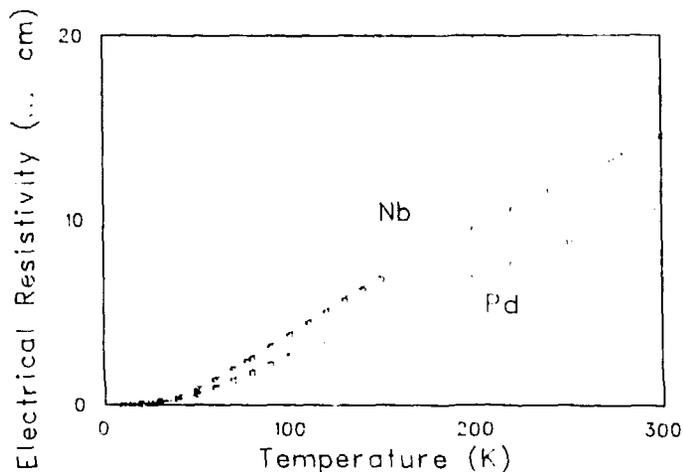

Fig. 1. The electrical resistivities of Nb and Pd. The solid lines are from solutions to the Blotzmann equation which use RMTA matrix elements and take into account both the wave-vector and energy dependence of the distribution function. The experimental results are from Webb (1969) for Nb and from White and Woods (1958) for Pd.

possible (certainly in principle and probably in practice, see e.g. Winter, 1978) to evaluate $\delta V(x)/\delta R_{\ell\alpha}$ numerically within local density theory. The matrix elements calculated from such a function using Eq. (1) would certainly have a firmer theoretical foundation than rigid muffin-tin matrix elements but they would not be exact because they neglect Coulomb vertex convections. These corrections have never been evaluated for a transition metal, but for Al they reduce the high temperature resistivity by about 10% (Rasolt and Devlin, 1976).

3. A Sum Rule and Problems For Small Momentum Transfer

A useful sum rule which helps to make the success of the RMTA more plausible can be derived from the translational invariance of the crystal. The self-consistent potential in the crystal $V(x)$ depends implicitly on the positions of all the atoms $\{R_\ell\}$. Moving from x to $x + \delta$ with the crystal fixed in space is equivalent to translating the crystal through $-\delta$, thus

$$V(x + \delta, \{R_\ell\}) = V(x, \{R_\ell - \delta\}) \ . \tag{9}$$

Expanding Eq. (9) to first order in δ yields

$$-\nabla_\alpha V(x) = \sum_\ell \frac{\delta V(x)}{\delta R_{\ell\alpha}} \ . \tag{10}$$

This sum rule will be satisfied by any approximation (such as the RMTA) in which the crystal potential is written as a sum of "atomic" contributions

$$V(x) = \sum_\ell V_a(x - R_\ell) \ , \tag{11}$$

which are assumed to move rigidly when an atom is displaced. Although Eq. (10) is easily satisfied it is often violated by schemes which attempt to "improve" on the RMTA. The RMTA amounts (essentially) to approximating $\delta V(x)/\delta R_{o\alpha}$ by $-\nabla_\alpha V(x)$ for all points x in the cell at the origin so that Eq. 10 can be written

$$[\delta V(x)/\delta R_{o\alpha}]_{RMTA} = [\delta V(x)/\delta R_{o\alpha}]_{exact} + \sum_{\ell \neq 0} \delta V(x)/\delta R_{\ell\alpha} \ . \tag{12}$$

The RMTA is justified if (for x in the cell at the origin) the second term on the right hand side of Eq. (12) can be neglected. This requires that the perturbation in the potential due to the displacement of an atom not extend outside the cell in which that atom is located. This approximation may not be bad in a transition metal with its localized d-orbitals and its high density of states which can screen out charge fluctuations within a short distance.

These assumptions (and the RMTA) clearly fail for the simple metals. For the limiting case of very weak potentials we have a rigorous theory (Bardeen, 1937) which tells us that it is the screened ionic (pseudo) potential which moves rigidly when an atom moves. Such a potential is much stronger and of much longer range than a muffin-tin potential. The

two types of potential correspond to two very different ways of writing the total crystal potential as a sum of atomic contributions (Lee and Heine, 1972).

The difference between the two types of potential is most apparent at small momentum transfers. For a nearly free electron system $I^{\alpha}_{k'k}$ = $(k' - k)_{\alpha} v(k'-k)$ where $v(q)$ is the fourier transform of whatever potential moves rigidly with an atom. The low q limit of a screened pseudopotential for an ion of valence Z in a system with Fermi energy density of states $N(0)$ is $v(0) = -Z/N(0)$. The low q limit for scattering off of a muffin-tin potential which gives the same band structure will be much smaller.

There is some evidence that the RMTA matrix elements for transition metals are also too weak at low q. Figure 2 shows the same Nb resistivity data shown in Fig. 1 but here it is displayed as the ratio of the calculated to the experimental values. The ratio is very close to unity above 30 K but it drops significantly at lower temperatures. The experimental resistivity exceeds the calculated value by a factor of two at 10 K. The RMTA results at this temperature depend upon the low q, low ω matrix elements (ħω < 3 meV) and upon how well one solves the Boltzmann equation. Our solution to the Boltzmann equation is not perfect but a better solution allowing even more variational freedom to the distribution function could only lower the resistivity.

Further evidence for weak low q matrix elements may be seen in the electron-phonon spectral function $\alpha^2(\omega)F(\omega)$. This function is a more detailed measure of the electron-phonon interaction than λ since it picks out only those contributions to λ which are associated with frequency ω

$$\alpha^2(\omega)F(\omega) = \frac{1}{N(0)} \sum_{kk'j} \delta(\varepsilon_k)\delta(\varepsilon_{k'}) \left| g^j_{k'k} \right|^2 \delta(\hbar\omega - \hbar\omega^j_{k'k}) . \quad (13)$$
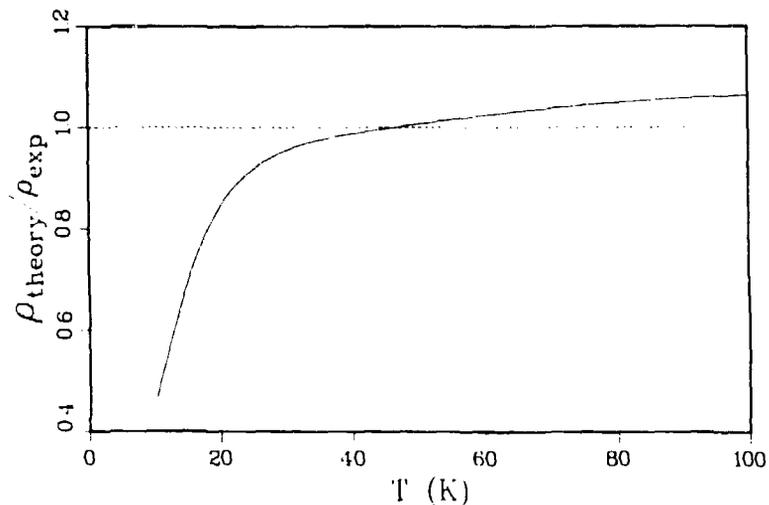

Fig. 2. The ratio of the resistivity calculated in the RMTA to the experimental values for Nb.

The low frequency $\alpha^2(\omega)F(\omega)$ inferred by Arnold et al. (1979) from tunneling data for Nb exceeds that calculated in the RMTA (Butler, et al., 1979) by an amount which is consistent with the discrepancy between the experimental and RMTA resistivities.

The phonon linewidth $\gamma_j(q)$ can in principle be used to measure the contributions to λ which arise from phonons of a particular wave vector and momentum transfer since

$$\gamma_j(q) = \pi \sum_{kk'} \delta(\varepsilon_k)\delta(\varepsilon_{k'}) \mid g^j_{k'k} \mid {}^2\omega^j_{k'k}\delta(k - k' - q) \ . \qquad (14)$$

The RMTA has been very successful in predicting structure in the linewidth (Butler, et al. 1977, Pinski and Butler, 1979); however, there is one accurate measurement of a low q transverse phonon linewidth in Nb at q = (0.07, 0.07, 0.00)π/a (Shapiro et al., 1975) for which the experimental value exceeds the RMTA result by a factor of approximately 1.5.

The too small low q matrix elements may be responsible for a possible inaccuracy of the RMTA in predicting the anisotropy of the electron-phonon mass enhancement. The mass enhancement is not truly a constant but varies from point to point on the Fermi surface. The electron-phonon contribution to the k dependent mass enhancement is given by

$$\lambda_k = 2 \sum_{k'} \delta(\varepsilon_{k'}) \mid g^j_{k'k} \mid {}^2/\hbar\omega^j_{k'k} \ . \qquad (15)$$

The average of λ_k over the Fermi energy is just λ. The wave vector dependent mass enhancement can also be written in the form

$$\lambda_k = 2 \int \frac{d\omega}{\omega}\alpha^2_k(\omega)F(\omega) \ , \qquad (16)$$

where $\alpha^2_k(\omega)F(\omega)$ is a k dependent spectral function obtained by eliminating the sum over k in Eq. (13). Now it is known from de Haas-van Alphen measurements of electron lifetimes (Nowak, 1972) that the low frequency part of $\alpha^2_k(\omega)F(\omega)$ is quite anisotropic. It is much more anisotropic than the average over all frequencies λ_k. It might be expected therefore that the RMTA would underestimate the anisotropy in λ_k. Such an effect has apparently been observed by Crabtree et al. (1979) who obtained 1 + λ_k by dividing calculated band velocities by experimental quasiparticle velocities. The relative anisotropy in λ_k inferred in this way, although small in comparison with that of some simple metals, was substantially greater than that calculated in the RMTA (Harmon and Sinha, 1977, Pinski et al., 1980b). It should be remembered however, that there is some uncertainty in the interpretation of the experiments since small errors in the band velocities would cause substantial error in the anisotropy of λ_k and since other many-body enhancements may also be anisotropic.

4. The Variation of the Superconducting T_c Among Transition Metal Systems

Despite the possible problems with small momentum transfers discussed in the preceeding section, the RMTA appears to give a good account of the

overall strength of the electron-phonon coupling in transition metals.
It can also be used to rationalize the empirical rules which have been
found to govern the occurrence of high T_c superconductivity in transition
metal based systems. The following set of rules are based on trends
observed primarily in the transition metals and the A-15 compounds.

(a) The rule of the half-filled d-band. High T_c's tend to be found
     only in those transition metal elements and compounds with approxi-
     mately half-filled d-bands.
(b) The rule of e/a. Within a given crystal structure T_c correlates
     with e/a, the average number of valence electrons per atom
     (Matthias, 1971).
(c) The rule of 4-d superiority. For a given crystal structure and e/a
     the compounds based on the 4-d transition metal elements (e.g., Nb)
     tend to have higher T_c's than those based on corresponding 3-d
     (e.g., V) or 5-d (e.g., Ta) elements.
(d) The rule of togetherness. For a given crystal structure, e/a, and
     period, electron-phonon coupling is enhanced if the transition metal
     atoms are closer together.
(e) The rule of the perversity of nature. For a given class of
     materials those with the highest T_c's tend to be less stable and
     more difficult to make.

Insights gained from RMTA and tight-binding calculations can explain
rules (a), (b) and (d) in a qualitative fashion. If, following
McMillan (1968), we write λ as

$$\lambda = N(0)\langle I^2 \rangle / M \langle \omega^2 \rangle , \qquad (17)$$

we see that λ depends on three factors $N(0)$, $\langle I^2 \rangle$, and $M\langle \omega^2 \rangle$. The last
of these factors is essentially a measure of the lattice stiffness and
we know qualitatively how it should vary. It should peak near the
center of the transition metal series for the strongly bonded metals
like Mo. This variation by itself would lead us to expect little or no
superconductivity for half-filled d-bands.

The RMTA calculations (Butler, 1977, Pettifor, 1977, and
Papaconstantopoulos et al., 1977) indicate however that $\langle I^2 \rangle$ is also
largest near the center of the transition metal series. In fact, the
variation in $\langle I^2 \rangle$ with e/a is stronger than that of $M\langle \omega^2 \rangle$ so the ratio
$\langle I^2 \rangle / M\langle \omega^2 \rangle$ is largest for an approximately half-filled d-band.

This result explains the absence of superconductivity near the ends of
the transition metal series. In the RMTA, $\langle I^2 \rangle$ can be written approxi-
mately for transition metals as

$$\langle I^2 \rangle \sim f_2 f_3 / V^2 , \qquad (18)$$

where f_2 and f_3 are the fractions of the Fermi energy density of states
which are associated with the d and f character respectively of the KKR
wave functions, and V is the volume per atom. f_3 of course has nothing
to do with atomic f orbitals but arises because d orbitals on neigh-
boring sites have tails which hang over into the site at the origin.
These tails yield a non d-like density of states when expanded about the
origin. At the low e/a side of the series (e.g., Y) $\langle I^2 \rangle$ is small pri-
marily because V is large. At the opposite end of the series (e.g., Pd)
f_3 and hence $\langle I^2 \rangle$ are small because the d-orbitals are concentrated near
the nucleus.

The observed variations of T_c with e/a (Matthias, 1971) result primarily from the rapid variation of the Fermi energy density of states with e/a superimposed on the variation of $<I^2>/M<\omega^2>$. A high density of states in the vicinity of the Fermi energy increases λ both directly because it is a factor in Eq. (17) and indirectly through a renormalization of the phonon frequencies which lower $M<\omega^2>$.

The rule of "4-d superiority" and the rule of "togetherness" are illustrated in Fig. 3 for the A-15 compounds with e/a's of 4.50 and 4.75. The former rule is difficult to explain on general grounds. When one descends a column in the periodic table (e.g., V, Nb, Ta), $<I^2>$ and $M<\omega^2>$ both increase whereas N(0) typically decreases. T_c may also be affected by variations in the Coulomb repulsion which is likely to be greatest for the 3d's. It is difficult to predict *a-priori* the net effect of these conflicting treads.

The rule of "togetherness" can be understood in terms of the overlapping of d-orbitals between transition metal atoms. This is necessary to obtain a large $<I^2>$ and it is clearly facilitated by a small lattice parameter. The variation of T_c with lattice parameter seen in Fig. 3 can be understood therefore in terms of a simplistic picture in which the simple metal atoms act as relatively inert "spacers" which determine the magnitude of the lattice parameter. This picture is supported by the RMTA calculations for the A-15's of Klein et al., (1979) who found that $<I^2>$ is dominated by contributions coming from the transition metal site.

Much attention has been given in recent years to the lattice instabilities that are fairly common in high T_c materials. In the final rule, I am also suggesting, however, a phenomenon which may be difficult to detect experimentally, namely, a tendency for high T_c phases to have a higher free energy than competing phases with lower T_c's. It is likely, for example, that phases with a high Fermi energy density of states are at a disadvantage when the Fermi energy falls near the center of the d-band complex. Finding ways to violate or circumvent this rule seems to be one of the keys to raising T_c.
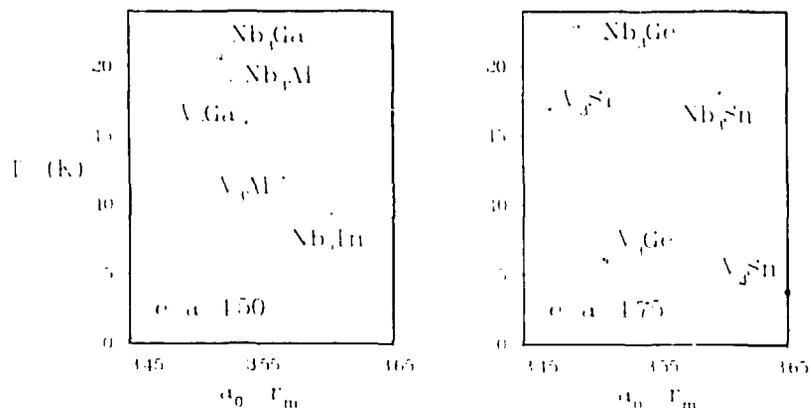

Fig. 3. T_c vs lattice parameter for the A-15's with electron per atom ratios of 4.50 and 4.75. The lattice parameters have been nor-malized by the metallic radius of the transition metal atom.

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