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Calculations of the superconducting properties of 32 metals with $Z \leq 49$

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The results of self-consistent band-structure calculations and the measured Debye temperatures are used in conjunction with the theory of Gaspari and Gyorffy to determine the electron-phonon interaction η, and the mass-enhancement factor λ for 32 metals with $Z = 1$ to $Z = 49$. A correction to η which accounts for the overestimated d-f contribution is included in a manner consistent with the work of Boyer *et al.*, and of Pettifor. The Coulomb pseudopotential μ^* is found from the empirical formula of Bennemann and Garland, and is used with our calculated λ's in the McMillan expression to calculate the superconducting transition temperature T_c for these materials. The calculations of η are based on the self-consistent potentials generated by Morruzzi *et al.* in which they have treated those metals that are noncubic in a fcc or bcc structure having the same density as the real material. Agreement between our calculations and experiment are generally good, with the exception of some of the non-fcc or -bcc materials. In particular our calculations are in excellent agreement with the Matthias rules. To the extent that one can generalize to compounds, our results show that this computational approach provides a reliable guide in the search for higher-T_c materials.

I. INTRODUCTION

A theory of electron-phonon interaction was formulated by Gaspari and Gyorffy[1] (GG) within the rigid-muffin-tin approximation, in terms of quantities obtainable from energy-band-structure calculations. Since the appearance of this first paper,[1] the GG theory has been applied to calculating the mass-enhancement factor λ and the superconducting transition temperature T_c for several transition metals[2-5] and transition-metal compounds.[3,6,7] These calculations have led to the conclusion that band-theory-based evaluation of some of the parameters that control superconductivity is reliable, and that there is reasonable hope to expect theoretical predictions of higher-T_c materials.

With this thought in mind we present here a systematic study of the variation of the electron-phonon interaction η, the mass enhancement λ and T_c, as a function of the atomic number Z, for 32 metals with $Z = 1$–49.

II. CALCULATIONS

We have used self-consistent potentials generated by three of us [Morruzzi, Janak, and Williams (MJW)] in the course of total-energy calculations by the Korringa-Kohn-Rostocker method which are reported elsewhere.[8] These potentials were found using the Hedin-Lundqvist[9] treatment of exchange and correlation. The calculations for those materials which have the hcp structure were done in either the fcc or bcc structure, with the lattice constant determined by using the same density as in the real hcp structure.

MJW have calculated the equilibrium lattice parameters a_0 of the elements under consideration to better than 4% accuracy, by minimizing the total energy as a function of a_0. Since a 1% change in lattice constant corresponds to an approximate 5% change in the electron-phonon interaction, and since we use the experimental Debye temperature, measured for the true value of a_0, in our calculations of λ (see below), some fairly substantial inaccuracies are possible. Fortunately, in the course of the minimum energy determination, MJW have determined self-consistent potentials for lattice constants which differ by less than 2% from the true a_0, and it is these potentials that we have used. The lattice constants and crystal structures used in our calculations are given in Table I. Using the MJW potentials, we performed a final run of an augmented-plane-wave (APW) calculation, and obtained eigenvalues and eigenfunctions on a mesh of 285 $\vec{k}$ points in the $\frac{1}{48}$th of a bcc Brillouin zone, or 240 $\vec{k}$ points in the $\frac{1}{48}$th of an fcc Brillouin zone, as applicable.

The APW results were interpolated by using a modification of the "Quad" scheme,[10] to give the total and angular momentum components of the densities of states (DOS), n and n_l. The fitted

TABLE I. Lattice constants and crystal structures used.

Element	Latt. const. (Bohr units)	Structure	Element	Latt. const. (Bohr units)	Structure
H	4.30	fcc	Zn	7.25	fcc
Li	6.60	bcc	Ga	7.83	fcc
Be	6.06	fcc	Rb	10.60	bcc
Na	7.70	bcc	Sr	11.00	fcc
Mg	8.58	fcc	Y	9.34	fcc
Al	7.60	fcc	Zr	6.60	bcc
K	9.90	bcc	Nb	6.20	bcc
Ca	10.40	fcc	Mo	5.89	bcc
Sc	6.70	bcc	Tc	7.25	fcc
Ti	7.74	fcc	Ru	7.20	fcc
V	5.72	bcc	Rh	7.18	fcc
Cr	5.38	bcc	Pd	7.36	fcc
Mn	6.80	fcc	Ag	7.68	fcc
Fe	5.30	bcc	Cd	8.20	fcc
Co	6.54	fcc	In	8.95	fcc
Ni	6.55	fcc			
Cu	6.83	fcc			

energies had an rms deviation of 3 mRy from the APW values, in the vicinity of the Fermi level. The Monte Carlo sampling used 512 000 *k* points for the bcc structure and 432 000 *k* points for the fcc structure, resulting in negligible statistical error.

Using the DOS and the scattering-phase shifts, also found from the self-consistent muffin-tin potentials, we applied the GG theory and obtained the electron-phonon interaction η. In order to derive the parameter λ from these calculations we used the expression[11]

$$\lambda = \eta / M\langle\omega^2\rangle , \qquad (1)$$

with $\langle\omega^2\rangle$ approximated by $\frac{1}{2}\Theta_D^2$, where Θ_D is the Debye temperature, obtained from specific-heat measurements.[12] Since our approach here is to study the trends which the quantities η and λ display across the Periodic Table, we have not attempted to obtain a more accurate $M\langle\omega^2\rangle$ from neutron-scattering measurements as we have done previously.[3,5-7] In the same spirit we have neglected the small nonspherical corrections to the GG formula discussed by Butler *et al.*[4]

T_c was calculated from the well-known McMillan formula,[11] also utilizing the known Debye temperatures, and with the value of the Coulomb pseudopotential μ^* calculated from the following empirical formula due to Bennemann and Garland[13]:

$$\mu^* = 0.26 N(0) / [1 + N(0)] , \qquad (2)$$

where $N(0)$ is the total DOS at the Fermi level expressed in units of states/eV atom. The results are analyzed in Sec. III.

III. DISCUSSION

According to the theory of Gaspari and Gyorffy, the electron-phonon interaction η depends mainly on the product of the following quantities evaluated at the Fermi energy E_F: (a) the trigonometric functions $\sin^2(\delta_{l+1} - \delta_l)$, where δ_l are the scattering phase shifts, and (b) the ratios n_l/N_l where n_l is the l component of the density of states at the Fermi level, and N_l is the so-called free-scatterer DOS (the DOS from a single muffin-tin potential in a zero-potential background[4]). Hence, large values of η can be obtained when E_F is near resonance ($\delta_l \simeq \frac{1}{2}\pi$) and $\delta_l \pm 1 \simeq 0$, and when n_l/N_l are also large. In Table II we have tabulated the phase shifts δ_l and the ratios n_l/N_l. We note that (a) δ_s is near resonance for hydrogen, (b) δ_d is near resonance for the 3*d* and 4*d* transition metals; (c) for the simple and noble metals, none of the δ_l are near resonance, and (d) n_p/N_p and n_f/N_f are large for the transition metals.

In Table III we list the values of η and λ, and also their separate contributions from the *s*-*p*, *p*-*d*, and *d*-*f* electron-phonon scattering. We also list "corrected" values of η and λ as discussed below. We note here that all high-η materials (except H) have their largest contribution coming from the *d*-*f* scattering.

It should be noted that large η's due to *d*-*f* scattering rest on the most uncertain aspect of the rigid-muffin-tin (RMT) model. In contrast to the *s*-*p* and *d*-*p* scattering, which are fundamentally intra-atomic effects, the *d*-*f* scattering probes the electron-ion interaction in the outer portions of

TABLE II. Phase shifts and n_l/N_l ratios.

Element	δ_s	δ_p	δ_d	δ_f	n_s/N_s	n_p/N_p	n_d/N_d	n_f/N_f
H	0.906	0.027	0.001	0.000	1.182	0.831	3.049	4.574
Li	−0.215	0.148	0.004	0.000	0.955	1.751	1.450	3.167
Be	−0.083	0.445	0.016	0.001	0.502	0.089	0.626	0.083
Na	−0.095	0.035	0.009	0.000	1.005	1.137	1.075	1.120
Mg	0.191	0.207	0.030	0.001	0.905	0.988	1.515	1.738
Al	0.344	0.436	0.065	0.003	0.960	0.591	1.296	1.762
K	−0.165	−0.026	0.033	0.001	1.200	1.178	1.037	0.934
Ca	−0.247	−0.020	0.157	0.002	0.230	3.881	1.888	3.942
Sc	−0.648	−0.172	0.440	0.003	0.121	4.449	1.843	4.943
Ti	−0.641	−0.157	0.754	0.003	0.414	3.101	0.788	6.382
V	−0.691	−0.173	1.030	0.003	0.550	2.842	0.634	6.935
Cr	−0.728	−0.184	−1.121	0.003	0.106	0.833	0.313	2.791
Mn	−0.523	−0.082	−0.798	0.002	0.239	0.978	0.964	5.856
Fe	−0.497	−0.068	−0.564	0.002	0.223	0.422	4.035	5.574
Co	−0.397	−0.022	−0.416	0.002	0.210	0.382	3.396	4.451
Ni	−0.280	0.020	−0.324	0.001	0.162	0.000	8.375	2.821
Cu	−0.059	0.096	−0.145	0.001	0.564	0.862	0.928	0.842
Zn	0.249	0.258	−0.038	0.002	1.262	0.733	1.462	1.417
Ga	0.751	0.579	0.013	0.002	1.239	0.444	0.769	1.938
Rb	−0.204	−0.073	0.047	0.001	1.441	1.141	1.159	0.848
Sr	−0.448	−0.153	0.218	0.003	0.000	0.001	0.001	0.002
Y	−0.703	−0.261	0.468	0.005	0.316	2.100	1.396	3.851
Zr	−0.919	−0.368	0.786	0.007	0.539	2.393	0.771	2.858
Nb	−0.932	−0.363	1.142	0.007	0.807	2.927	0.681	3.845
Mo	−0.971	−0.381	−1.274	0.010	0.122	1.016	0.455	2.092
Tc	−0.840	−0.294	−0.976	0.008	0.419	1.207	1.050	3.391
Ru	−0.722	−0.222	−0.682	0.006	0.296	0.798	1.586	3.288
Rh	−0.611	−0.160	−0.484	0.005	0.314	0.511	3.079	2.881
Pd	−0.393	−0.064	−0.311	0.002	0.314	0.332	7.188	2.661
Ag	−0.189	0.025	−0.149	0.003	0.602	0.951	0.776	0.529
Cd	0.174	0.224	−0.060	0.004	1.336	0.718	1.381	1.146
In	0.726	0.570	0.001	0.003	1.299	0.434	0.739	1.880

the atomic cell where non-muffin-tin effects and screening are most important. In an earlier paper[5] we found that the approximate inclusion of non-muffin-tin effects significantly reduces the d-f contribution to η. Pettifor[14] has also shown that an approximate treatment of the screening ignored in the RMT model causes a similar reduction. Taking these two effects into account our best estimate is that the d-f contribution should be reduced by approximately a factor of 2 from the RMT value. To explore the effect of such a reduction on the trends in η, λ, and T_c with atomic number we present in Tables III and IV a second set of these quantities (labeled II) in which the d-f contribution has been arbitrarily halved. The significantly closer correspondence of these "corrected" results to the measured values of T_c confirms our expectation that the RMT model substantially overestimates d-f scattering.

It should also be mentioned here that η, which is an implicit average over the Fermi surface in the GG theory, is not very sensitive to the different currently accepted treatments of exchange.[4,15] This is in contrast to the Fermi surface areas, which in the cases of V and Nb,[15] as well as Cu,[16] have been found to be significantly different, for different exchange approximations.

In Table IV we give the measured values of the Θ_D, T_c, and the calculated values of E_F, $n(E_F)$, μ^*, T_c(I), and T_c(II). We note that (a) vanadium and niobium are found with high T_c's as is known experimentally, (b) all other elements have much smaller values of both λ and T_c, and (c) we confirm previous theoretical predictions[17] that metallic hydrogen would be a high-temperature superconductor. The present results for hydrogen are slightly different from those given in Ref. 17, due to the difference between the exchange parameters and the lattice constants used in the respective calculations. According to the calculations scandium, contrary to the experiment, is a superconductor. We offer the explanation that this result may be due to the fact that we have performed our calculation in the bcc structure, while this metal has the hcp structure.

This may also be the reason that technetium, a relatively high T_c material, showed a substantially lower T_c from our calculations. In addition, since technetium has soft optic phonon modes,[18] the Debye approximation considerably overestimates $\langle\omega^2\rangle$. Lowering of Θ_D to about 300 °K will result in a higher λ and a T_c close to the measured value. This along with the fact that the real crystal structure of technetium is hcp can readily account for the discrepancy.

For the simple metals, aluminum, gallium, and indium, which have one p electron in the outer shell, we find that our values of λ are too small to reproduce the measured T_c. This may be due to the approximation made in using Eq. (1) in which the electronic and phonon contributions to λ are separated. Allen and Cohen[19] have also done calculations of λ and T_c for several simple metals by the pseudopotential method and showed that Eq. (1) seriously underestimates λ. Another source of error for Ga and In is our performing these calculations in the fcc structure instead of the orthorombic and tetragonal structure, respectively. It may also be that these discrepancies are simply a demonstration of the fact that our calculations of T_c are not accurate to better than ±3 °K. We may be able to clarify these points by performing calculations on the free-electron-like metal Pb which is a high-temperature superconductor.

Finally, we note that using the Bennemann-Garland formula we obtain the expected higher values of μ^* for the magnetic materials. It should also be stressed here that using $\mu^* = 0.13$ for V or Nb would not change our T_c results by more than 2 °K.

The results of Tables III and IV are displayed in Figs. 1–4, where the quantities $n(E_F)$, η, λ, and T_c are plotted versus Z. Comparing Figs. 1 and 4 we observe that a high DOS at the Fermi energy is not always a valid criterion for identifying

TABLE III. Electron-phonon interaction η and mass-enhancement factor λ, and their *s-p*, *p-d*, and *d-f* components. The index II refers to the corrected values obtained by reducing the *d-f* contribution by a factor of 2.

Element	η_{sp}	η_{pd}	η_{df}	η_{tot}(I)	η_{tot}(II)	λ_{sp}	λ_{pd}	λ_{df}	λ_{tot}(I)	λ_{tot}(II)
		(eV/Å^2)								
H	6.438	0.040	0.000	6.478	6.478	2.089	0.013	0.000	2.102	2.102
Li	0.177	0.088	0.000	0.265	0.265	0.243	0.120	0.000	0.363	0.363
Be	0.207	0.352	0.001	0.560	0.560	0.012	0.022	0.000	0.034	0.034
Na	0.015	0.001	0.000	0.017	0.017	0.030	0.003	0.000	0.033	0.033
Mg	0.000	0.115	0.008	0.123	0.119	0.000	0.033	0.003	0.036	0.035
Al	0.012	0.510	0.068	0.590	0.558	0.003	0.116	0.016	0.135	0.127
K	0.008	0.003	0.000	0.011	0.011	0.028	0.009	0.003	0.040	0.039
Ca	0.010	0.105	0.123	0.239	0.178	0.006	0.056	0.066	0.128	0.096
Sc	0.036	1.719	1.554	3.309	2.530	0.007	0.332	0.300	0.639	0.489
Ti	0.130	1.427	3.283	4.840	3.200	0.017	0.190	0.438	0.645	0.426
V	0.200	1.641	5.053	6.894	4.370	0.031	0.251	0.773	1.055	0.669
Cr	0.040	0.570	3.592	4.202	2.400	0.002	0.031	0.196	0.229	0.131
Mn	0.024	0.465	4.993	5.482	2.980	0.003	0.057	0.608	0.668	0.364
Fe	0.005	0.215	5.404	5.623	2.920	0.000	0.020	0.500	0.520	0.270
Co	0.005	0.189	3.705	3.900	2.050	0.000	0.018	0.358	0.376	0.197
Ni	0.000	0.000	1.762	1.762	0.880	0.000	0.000	0.167	0.167	0.084
Cu	0.035	0.272	0.149	0.456	0.380	0.005	0.041	0.023	0.069	0.058
Zn	0.000	0.516	0.028	0.544	0.530	0.000	0.083	0.005	0.088	0.086
Ga	0.045	0.549	0.001	0.595	0.590	0.007	0.087	0.000	0.094	0.094
Rb	0.006	0.009	0.001	0.016	0.016	0.026	0.036	0.006	0.068	0.065
Sr	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Y	0.048	1.024	1.266	2.337	1.700	0.008	0.165	0.205	0.378	0.276
Zr	0.246	2.144	2.268	4.658	3.520	0.036	0.313	0.330	0.679	0.514
Nb	0.470	2.726	4.431	7.627	5.410	0.075	0.437	0.710	1.222	0.867
Mo	0.069	1.008	4.720	5.797	3.440	0.004	0.058	0.274	0.336	0.199
Tc	0.129	0.954	7.010	8.094	4.590	0.009	0.065	0.476	0.550	0.312
Ru	0.052	0.474	5.985	6.510	3.520	0.002	0.015	0.184	0.201	0.109
Rh	0.023	0.243	4.456	4.722	2.490	0.001	0.011	0.212	0.224	0.118
Pd	0.004	0.099	1.889	1.992	1.050	0.001	0.014	0.266	0.281	0.148
Ag	0.076	0.130	0.083	0.288	0.250	0.016	0.027	0.016	0.059	0.051
Cd	0.005	0.316	0.039	0.359	0.340	0.001	0.072	0.009	0.082	0.077
In	0.024	0.331	0.000	0.355	0.350	0.020	0.278	0.000	0.298	0.298

TABLE IV. Measured values of Θ_D and T_c. Calculated values of E_F, $n(E_F)$, μ^*, and T_c. The index II refers to the corrected results.

Element	Θ_D(°K)	E_F(Ry)	$n(E_F)$ (states/Ry atom spin)	μ *	T_c(I)(°K)	T_c(II)(°K)	$T_{c\,\mathrm{exp}}$(°K)
H	1861 [a]	0.732	0.65	0.023	252.00	252.00	...
Li	344	0.290	3.40	0.087	0.65	0.65	0.0
Be	1440	0.814	0.44	0.016	0.0	0.0	0.026
Na	158	0.247	3.04	0.080	0.0	0.0	0.0
Mg	400	0.402	3.18	0.083	0.0	0.0	0.0
Al	428	0.618	2.39	0.068	0.0	0.0	1.175
K	91	0.159	5.26	0.113	0.0	0.0	0.0
Ca	230	0.286	12.26	0.167	0.0	0.0	0.0
Sc	360	0.515	16.02	0.183	2.92	0.51	0.0
Ti	420	0.588	12.38	0.168	4.46	0.28	0.39
V	380	0.675	12.70	0.169	16.65	4.62	5.43
Cr	630	0.811	4.74	0.107	0.0	0.0	0.0
Mn	410	0.699	12.02	0.166	5.16	0.04	0.0
Fe	467	0.739	26.11	0.206	0.54	0.0	0.0
Co	445	0.722	14.34	0.176	0.04	0.0	0.0
Ni	450	0.671	27.18	0.208	0.0	0.0	0.0
Cu	343	0.598	1.96	0.058	0.0	0.0	0.0
Zn	327	0.611	2.14	0.062	0.0	0.0	0.375
Ga	320	0.525	1.85	0.056	0.0	0.0	1.080
Rb	56	0.146	6.44	0.126	0.0	0.0	0.0
Sr	147	0.288	0.01	0.0	0.0	0.0	0.0
Y	280	0.437	10.94	0.160	0.07	0.0	0.0
Zr	291	0.605	8.58	0.145	5.00	1.53	0.53
Nb	275	0.676	9.71	0.153	16.79	8.77	9.25
Mo	450	0.806	4.42	0.102	0.45	0.00	0.916
Tc	411	0.756	7.86	0.139	3.37	0.03	7.73
Ru	600	0.713	7.39	0.135	0.0	0.0	0.493
Rh	480	0.670	8.68	0.146	0.0	0.0	0.0
Pd	274	0.523	14.85	0.179	0.0	0.0	0.0
Ag	225	0.517	1.74	0.053	0.0	0.0	0.0
Cd	209	0.512	2.48	0.069	0.0	0.0	0.52
In	108	0.409	2.27	0.065	0.04	0.04	3.40

[a] See Ref. 17.

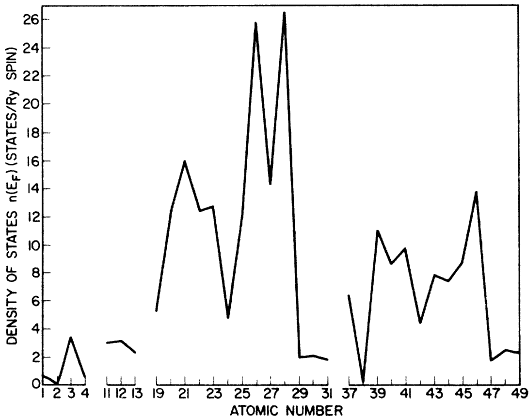

FIG. 1. Total density of states per Rydberg per spin as a function of the atomic number Z.

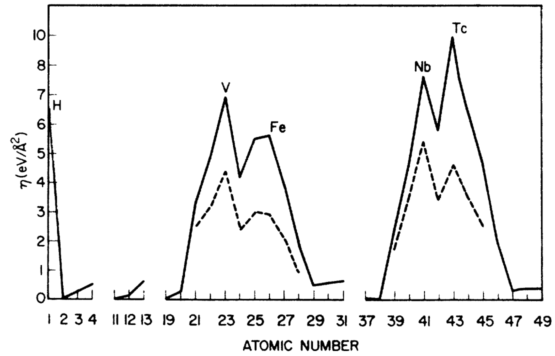

FIG. 2. Electron-phonon interaction η plotted vs the atomic number Z. The dotted line refers to the "corrected" values.

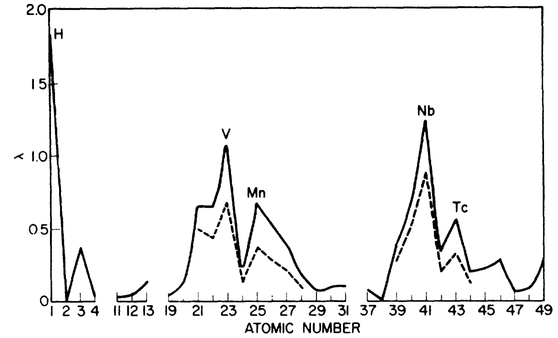

FIG. 3. Mass enhancement factor λ plotted vs the atomic number Z. The dotted line refers to the "corrected" values.

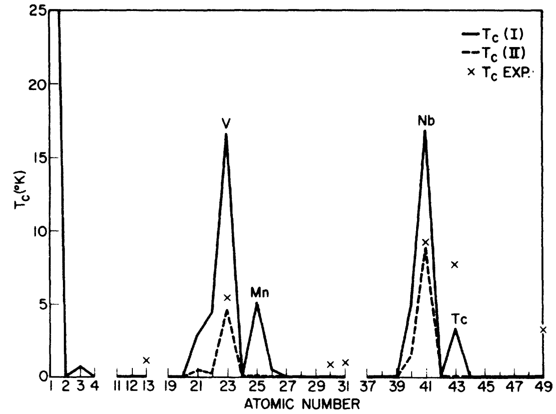

FIG. 4. Superconducting transition temperature T_c plotted vs the atomic number Z. The dotted line refers to the "corrected" values.

superconductivity. The most appropriate criteria are the quantities η and λ shown in Figs. 2 and 3. We note that the $n(E_F)$ for Fe and Ni are almost a factor of 2 larger than those of V and Nb, while almost the reverse is true of η and λ. In this connection, an inspection of our figures reveals that the suggestion of Dynes and Varma[20] that λ is proportional to $n(E_F)$ through the $3d$ or $4d$ series of transition metals, is a rather poor approximation even if we exclude the magnetic materials.

It is also important to note, when looking at Fig. 4, that our calculations are fully in agreement with the Matthias rule which correlates maximum T_c for electron-per-atom ratio of 4.5 and 6.5 and a minimum T_c at 5.5.

In conclusion, despite the possible discrepancies for the non-fcc or -bcc materials, we believe that by using the Gaspari-Gyorffy theory, the results of self-consistent energy-band calculations, and the measured Θ_D we can reliably account for all the high-temperature superconductors in the first half of the Periodic Table. We view this as a promising step in the direction of predicting new superconductors in more complex materials.

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