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THEORETICAL PREDICTION OF MoN AS A HIGH T_c SUPERCONDUCTOR

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A re-examination of several recent band structure calculations for transition metal compounds has suggested that Mo-based B1 structure compounds should be good superconductors. Using augmented-plane-wave band calculations and the Gaspari-Gyorffy approach for the electron-phonon interaction, we predict that MoC and MoN should have significantly higher T_c than their Nb-based counterparts NbC (T_c=11K) and NbN (T_c=17K).

The rigid muffin-tin approximation (RMTA) of Gaspari and Gyorffy[1] has been used widely in the past decade to understand or predict superconducting transition temperatures (T_c) in metals. The RMTA leads to an expression for $\eta = N(E_F)\langle I^2\rangle$ (in standard notation[1]) which can easily be evaluated using results of band structure calculations. Recently a re-interpretation of the RMTA has been given by one of the authors,[2] and the extensive calculations by the NRL group on bcc Nb and V,[3] Nb- and V-based A15 compounds[4] and on the B1 structure compounds NbC[5] and NbN[6] were re-examined. The conclusions of this study[2] included (i) $\langle I^2\rangle_X$ for atom X varies little within a given crystal structure, and (ii) $\langle I^2\rangle_{Nb}$ is 40-50% larger in NbC and NbN than in bcc Nb and Nb_3X A15 compounds.

These results suggest an investigation of T_c in B1 NbX compounds other than NbC and NbN (T_c=11K and 17K, respectively[7]). The obvious extension is to NbO; however, it is known to be a very poor superconductor[8] (T_c=1.2K). A related approach which is available, however, is to replace Nb with Mo, in which case $\langle I^2\rangle_{Mo}$ should be similar to (if not larger than[9]) $\langle I^2\rangle_{Nb}$. In addition, noting that Nb and Mo compounds often form good rigid-band systems, we have found that a rigid band prediction based on the band structures of NbC and NbN suggests an increase by 20-50% in $N(E_F)$ in their Mo counterparts. {This is not the case in systems dominated by d-d bonding, such as the bcc elements and A15 compounds, where replacement of Nb by Mo results in a sharp decrease in $N(E_F)$.} Since we expect similar values of $\langle I^2\rangle$ and similar, if not softer, lattice frequencies, noticeably higher values of T_c should occur for MoC and MoN compared to their Nb counterparts. Indeed, a survey of the literature reveals that B1 structure $MoC_{0.69}$ and $MoC_{\sim 1.0}$ have been reported[7,10] to superconduct at 12.1K and 14.3K, respectively. {Note that T_c(MoC) $\approx T_c$(NbN) if rigid band behavior holds on both sublattices, and this relation is roughly satisfied by the experimental values of T_c quoted above.}

Although essentially rigid band behavior of T_c has been verified[6] in the $NbC_{1-x}N_x$ system, it remains unclear to what extent rigid band behavior will occur on the metal sublattice. Therefore we have carried out self-consistent augmented-plane-wave band calculations for MoC and MoN identical to those described previously[6,11] for NbC and NbN. The lattice constant a=4.343Å which was used for MoC was obtained by linear extrapolation from the values measured[12] for $MoC_{0.69}$ and $MoC_{0.75}$, since a was reported to be linear in this range. For MoN we have estimated a=4.250Å, which follows by assuming the difference a(MoC)-a(MoN) to be the same as a(NbC)-a(NbN).

The resulting density of states of MoC and MoN are shown in Fig. 1, and qualitatively they are similar to their Nb counterparts. However, the values of $N(E_F)$ (see Table I) are 2.5 times larger than in the Nb-based compounds, and therefore they are considerably larger than the rigid band model predicted. To calculate T_c we use the RMTA generalized to compounds[5] to calculate η for each constituent, the modified[6] Bennemann-

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Garland formula for μ^*, and the Allen-Dynes modification[13] of the McMillan equation for T_c. For the phonon spectra there is limited data available for MoC_x (from specific heat[10]), and the phonon moments for MoN are obtained by assuming they scale as in the $NbC_{1-x}N_x$ system. The resulting quantities entering into the determination of T_c are presented in Table I.

The surprisingly high predicted value of T_c (MoN)=29K should be taken to indicate that MoN should be a considerably better superconductor than is NbN, rather than as a precise prediction. This high T_c results primarily from a large value of λ_{Mo}, due in turn to the large value of $N(E_F)$. This increase in T_c (relative to NbN) persists in spite of a larger value of μ^* and harder phonon frequencies, both of which are detrimental to T_c. In fact, the large values of $N(E_F)$ and λ obtained for MoN suggests the phonon moments will be softer than those we have used, which implies that λ and T_c will actually be larger than obtained here. On a more sobering note, the large values of λ and T_c suggest that MoN may lie beyond the phase boundary of stability for these B1 structure compounds, such as occurs in large λ regions of many phases. However, recent successes in preparing other metastable phases with high T_c lend some optimism here, and we encourage experimental tests of our prediction.

REFERENCES:

(1) Gaspari, G.D. and Gyorffy, B.L., Phys. Rev. Lett. 28, (1972) 801.
(2) Pickett, W.E., to be published.
(3) Papaconstantopoulos, D.A., Boyer, L.L., Klein, B.M., Williams, A.R., Moruzzi, V.L. and Janak, J.R., Phys. Rev. B15 (1977) 4221.
(4) Klein, B.M., Boyer, L.L. and Papaconstantopoulos, D.A., Phys. Rev. Lett. 42 (1979) 530.
(5) Klein, B.M., Papaconstantopoulos, D.A. and Boyer, L.L., in Superconductivity in d- and f-Band Metals, edited by D.H. Douglass (Plenum, New York, 1976) 339; Klein, B.M. and Papaconstantopoulos, D.A., J. Phys. F 6 (1976) 1135.
(6) Papaconstantopoulos, D.A., in Physics of Transition Metals 1980, edited by P. Rhodes (Inst. of Phys. Conf. Ser. No 55, London, 1981 563.
(7) Toth, L.E., Transition Metal Carbides and Nitrides, (Academic, New York, 1971) Ch. 7.
(8) Khan, H.R., Raub, Ch. J., Gardner, W.E., Fertig, W.A., Johnson, D.C. and Maple, M.B., Mat. Res. Bull. 9 (1974) 1129.
(9) Butler, W.H., Phys. Rev. B15 (1977) 5267.
(10) Toth, L.E. and Zbasnik, J., Acta Met. 16 (1968) 1177.
(11) Klein, B.M., Papaconstantopoulos, D.A. and Boyer, L.L., Phys. Rev. B22 (1980) 1946.
(12) Storms, E.K., The Refractory Carbides (Academic, New York, 1967) 132.
(13) Allen, P.B. and Dynes, R.C., Phys. Rev. B12 (1975) 905.

Table I. Quantities entering T_c: $N(E_F)$ in (eV-spin-cell)$^{-1}$, η, $M\langle\omega^2\rangle$ in (eV/Å^2), 1, 2 denote atoms 1, 2.

	NbC	NbN	MoC	MoN
$N(E_F)$	0.31	0.48	0.78	1.14
ω_{log}(K)	372	248	329	276
$(M\langle\omega^2\rangle)_1$	12.7	9.0	15.2	10.9
$(M\langle\omega^2\rangle)_2$	14.3	16.7	14.3	16.7
η_1	4.8	6.9	10.7	13.8
η_2	3.0	2.7	2.5	5.1
λ	0.59	0.93	0.88	1.58
T_c(K)	9.8	15.4	16.2	29.4

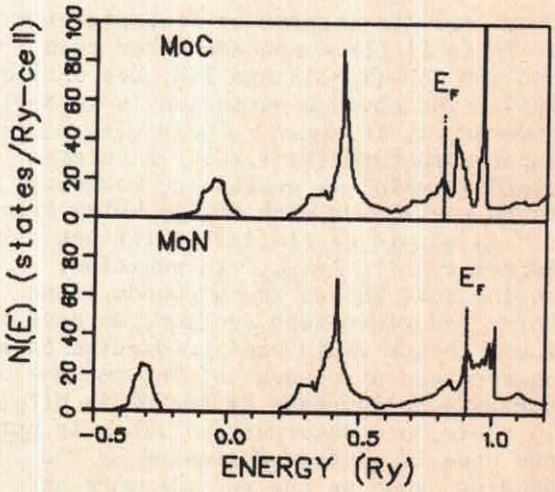

Figure 1. N(E) for MoC and MoN.