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Theoretical calculation of T_c for lead (*) (**)

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Résumé. — Nous calculons la température de transition supraconductrice du Pb avec la théorie de Gaspari et Gyorffy, en utilisant nos calculs *ab initio* de structure de bande. La valeur calculée ainsi est la moitié de la valeur expérimentale. Cet écart est attribué à l'approximation du potentiel *muffin tin* rigide. Pour tenir compte de cette approximation nous présentons une méthode simple, sans paramètre ajustable. La nouvelle valeur calculée de la température de transition est en accord excellent avec la valeur expérimentale.

Abstract. — Using our *ab initio* band structure results, we calculate the transition temperature of Pb according to the Gaspari-Gyorffy theory. The calculated value is half the experimental one. The discrepancy is attributed to the rigid muffin tin approximation. A simple method with no adjustable parameters is developed to account for this approximation. The new transition temperature is in excellent agreement with experiment.

Following the work of McMillan [1], which relates in a practical way the superconducting transition temperature T_c to microscopic quantities, a large amount of research has been reported along those lines. In particular, the theory of Gaspari and Gyorffy [2, 3], which we employ here, was designed especially for transition metals. However, as we shall illustrate below, this theory properly corrected can give very good estimates of the electron-phonon interaction η for normal metals like lead as well. The parameter η, which together with the average phonon frequency $\langle \omega^2 \rangle$, [1, 4] defines T_c, can be expressed in the Gaspari-Gyorffy theory in terms of partial densities of states and phase shifts readily available from traditional band structure calculations.

In the first stage of this work we have determined, within the Rigid Muffin Tin Approximation (RMTA), the quantity η using our self-consistent *ab initio* band calculation. The latter includes relativistic effects, such as the mass velocity and Darwin corrections, which for a heavy metal such as Pb are important [5, 6]. The spin orbit interaction has been neglected because, although it is very important for the structure of the Fermi surface, is not expected to be very significant for the electron-phonon interaction, and consequently for T_c, which is our primary concern here. Details of our band structure calculation, together with comparisons with previous calculations and experimental data are published elsewhere [7].

The total and partial densities of states n_t and n_l

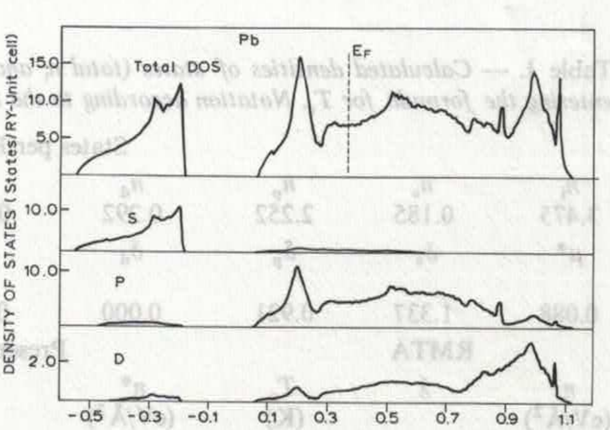

Fig. 1. — Electronic density of states (DOS) of Pb and its s-, p- and d-like components.

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respectively, which are calculated by our modified QUAD method [8], are shown in figure 1. The same quantities evaluated at the Fermi level E_F, together with the scattering phase shift δ_l, the mass enhancement λ and the free scatterer partial densities of states $n_l^{(1)}$ are listed in table I. We have calculated T_c according to the Allen and Dynes equation [4] with $M \langle \omega^2 \rangle$ the phononic contribution to λ, taken from their paper. The Coulomb potential μ^* was evaluated by the Bennemann and Garland formula [9]. The experimental value of λ, measured through tunnelling experiments (see refs. [4, 10]) is twice as large as our *ab initio* value calculated here.

This discrepancy must be due to the inherent approximations of the Gaspari-Gyorffy (GG) theory; namely the spherical, the local and the rigid muffin tin approximation [2, 3, 11]. The first two approximations are not expected to be important for lead. The spherical approximation is known to be well satisfied for cubic crystals and $l = 0, 1, 2$ [12].

The local approximation for lead is also expected to be well obeyed due to the large contribution of Umklapp electron-phonon scattering [11, 13-15]. It has been demonstrated [11-13, 14] that the local approximation is better satisfied for large Umklapp scattering. Furthermore, the validity of the local approximation can be also verified from the similarity of the phonon density of states curve, $F(\omega)$ and the $\alpha^2(\omega) F(\omega)$ profile [11].

The Eliashberg function $\alpha^2(\omega) F(\omega)$ defined in ref. [1] has been determined experimentally by superconducting tunnelling measurements (see refs. [4, 10]). Thus, the discrepancy between the observed and calculated values of λ and T_c must be mainly attributed to the rigid muffin tin approximation (RMTA). The validity of RMTA depends on the importance of the tails of the self consistent potential about the edge of the muffin-tin sphere. For transition metals, where the scattering of electrons is dominated by the atomic potential inside the muffin tin sphere, this approximation is expected to be quite good.

For lead, however, one must go beyond this approximation by adding a correction term to the RMTA potential. On physical grounds this correction term $V_c(r)$ is expected to have a strong influence near the muffin-tin radius, r_{MT}, and decrease rapidly away from the muffin-tin sphere. Therefore a plausible choice for $V_c(r)$ is

$$V_c(r) = \frac{V_0}{e^{q_{TF}(r - r_{MT})} + 1} \tag{1}$$

where q_{TF} is the inverse of the Thomas-Fermi screening length and V_0 is an as yet unspecified parameter which denotes the depth of the correction potential.

To determine V_0 without any resort to experimental information, we have adapted the equivalent RMTA electron-phonon pseudopotential of Lee and Heine [16] which has been expressed in terms of the same quantities that appear in the GG formula [11]. This pseudopotential in q-space has the form [11]

$$\begin{aligned} \tilde{V}_{RMTA}(q) = \frac{1}{\pi k_F\, n_t(E_F)} \left(\frac{2\, m E_F}{\hbar^2}\right)^{1/2} \times \\ \times \sum_l \left(\frac{n_l\, n_{l+1}}{n_l^{(1)}\, n_{l+1}^{(1)}}\right)^{1/2} \sin\,(\delta_l - \delta_{l+1}) \\ \times \sum_{l'}^{l} (2\, l' + 1)\, P_{l'}\left(1 - \frac{q^2}{2\, k_F^2}\right) \end{aligned} \tag{2}$$

where k_F is the free electron radius of the Fermi sphere and P_l is the Legendre polynomial of order l. The rest of the symbols have been defined before in the calculation of η.

The sine term in the above expression and equivalently in the GG formula was obtained by evaluating the radial matrix element of the derivative of the RMTA potential [3]. Obviously the addition of the potential (1) into the RMTA potential will result in replacing the sine terms above by

$$\sin\,(\delta_l - \delta_{l+1}) + \Delta V_{l,l+1}\,.$$

Table I. — *Calculated densities of states (total n_t and partial n_l) at the Fermi level and other basic quantities entering the formula for T_c. Notation according to the text and references* [1-4].

States per Ry.-atom-spin								
n_t	n_s	n_p	n_d	n_f	$n_s^{(1)}$	$n_p^{(1)}$	$n_d^{(1)}$	$n_f^{(1)}$
3.475	0.185	2.252	0.292	0.060	0.159	3.226	0.252	0.031
μ^*	δ_s	δ_p	δ_d	δ_f		E_F (Ry.)	$M \langle \omega^2 \rangle$ (eV/Å^2)	
0.088	1.337	0.921	0.000	0.004		0.365	1.434	
RMTA			Present Method			Experimental		
η (eV/Å^2)	λ	T_c (K)	η^* (eV/Å^2)	λ^*	T_c^* (^{a}) (K)	η (eV/Å^2)	λ	T_c (K)
1.199	0.836	3.3	2.216	1.545	7.24	2.223	1.55	7.19

(^{a}) To obtain T_c^* we used the Coulomb pseudopotential of ref. [4], $\mu^* = 0.105$.

The terms $\Delta V_{l,l+1}$, being the radial matrix elements of the derivative of (1), are expressed in terms of V_0. The depth V_0 can be determined then by demanding that the new pseudopotential $\tilde{V}_{ep}(q)$ has the correct limit [17, 18] : $-Z/2\,n_t(E_F)$ as q approaches zero.

In figure 2 we have plotted both $\tilde{V}_{RMTA}(q)$ and $\tilde{V}_{ep}(q)$ as a function of $q/2\,k_F$. The deficiency of the RMTA can be seen from the fact that the RMTA pseudopotential reaches about half the correct value as q approaches zero.

The new renormalized values of n^*, λ^* and T_c^* are listed in table I. The agreement with the experimentally determined value of λ is excellent.

Good agreement with experiment have been obtained before [11, 15, 19-22] using empirical pseudopotentials. In the present approach the pseudopotential was determined from our first principles band structure calculation and its limiting value as $q \to 0$, without any resort to experimental data.

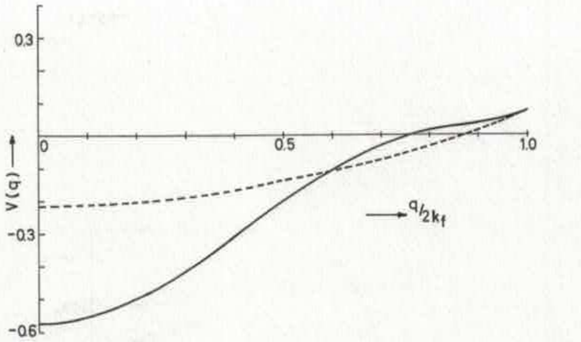

Fig. 2. — The RMTA (broken lines) and the corrected, $\tilde{V}_{ep}(q)$, (solid lines) pseudopotentials for lead.

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