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Diamagnetic Susceptibility of Lithium[*]

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(Received 31 July 1972)

We have calculated the diamagnetic susceptibility of lithium using an expression derived by us recently, since no reliable theoretical result is available. We compare our result with available experimental and theoretical results.

The diamagnetic susceptibility of lithium was first calculated by Kjeldaas and Kohn.[1] They used a generalized effective-mass theory and took into account fourth-order terms in the expansion of $\vec{k} - \vec{k}_0$. However, their result is only of limited usefulness, since they considered the very special case of electrons confined to the vicinity of the top or bottom of an energy band. Furthermore, the ratio of the second to the first term in the expansion in powers of k_0 is 0.6 for lithium, so that higher-order terms may also be important in their calculation.

Fletcher and Larson[2] calculated the diamagnetic susceptibility of lithium by applying the Landau–Peierls[3] formula to the equilibrium quasiparticle energy calculated in the Bohm–Pines[4] theory of electron correlation, and by modifying their Coulomb-gas calculations by using a band effective mass. However, their result is not reliable since there are other contributions to the diamagnetic susceptibility which are of the same order of magnitude as the Landau–Peierls term[5] and further, as evident from Kjeldaas and Kohn's result,[1] the effective-mass formalism does not yield correct results for lithium. Glasser[6] calculated the diamagnetic susceptibility of lithium by subtracting from his result for the magnetic susceptibility the result for the paramagnetic susceptibility calculated from Abe's expression.[7] However, it has been shown[5] that the difference between Glasser's expression for magnetic susceptibility and Abe's expression for paramagnetic susceptibility is identically equal to Misra and Roth's[5] expression for diamagnetic susceptibility obtained by using nondegenerate perturbation theory. Therefore, Glasser's result is not reliable, since, because of the strong pseudopotential, the nondegenerate perturbation theory is not valid for lithium.

Misra and Roth[5] calculated the diamagnetic susceptibility of the alkali metals from their general expression for diamagnetic susceptibility of simple metals, which was derived through the use of a pseudopotential formalism and degenerate perturbation theory. However, we have recently shown[8] that there is a difficulty in using Misra and Roth's expression for metals for which either the Fourier components of the pseudopotential are strong, or for which there is a neck in the Fermi surface. From the model-potential form factors calculated by Animalu and Heine and tabulated by Harrison,[9] we note that the pseudopotential is strong for the (110) and (200) G shells of lithium while the pseudopotential for the other G shells of lithium, as well as the pseudopotentials of all the other alkali met-

TABLE I. Diamagnetic susceptibility of lithium (χ in 10^{-6} cgs volume units).

χ_0	$\sum_G D_G$	χ_d
−0.266	−0.6503	−0.093

als, are weak. Therefore, while Misra and Roth's[5] calculations of diamagnetic susceptibility are accurate for the other alkali metals, they are not accurate for lithium.

Recently, the diamagnetic susceptibility of lithium has been calculated by many other authors. Timbie and White[10] and Brown *et al.*[11] have calculated the diamagnetic susceptibility of lithium from Misra and Roth's expression obtained using nondegenerate perturbation theory. Pappadopoulas and Jones[12] derived an expression for χ using a propagator method, but their result, up to the second order in the potential, is identical to the Misra–Roth nondegenerate calculation.[5] Therefore, it is not surprising that the results of Timbie and White,[10] Brown *et al.*,[11] and Pappadopulas and Jones[12] are in good agreement with each other. Unfortunately, these results are not reliable, since, because of the strong pseudopotential, the nondegenerate perturbation theory is not valid for lithium. Isihara and Tsai[13] have obtained an expression for the diamagnetic susceptibility of an interacting electron gas, and then replaced the free-electron mass by the effective mass to calculate the diamagnetic susceptibility of lithium. However, as evident from Kjeldaas and Kohn's result,[1] the effective-mass formalism does not yield correct results for lithium. Therefore, since no reliable theoretical result is available, we have calculated the diamagnetic susceptibility of lithium using the expression recently derived by us.[8] We also compare our result with the other theoretical results and the available experimental results.

The expression for the diamagnetic susceptibility χ_d derived by us[8] is

$$\chi_d = \chi_0 \left\{ 1 + \sum_G \left[\frac{\gamma_+ - \gamma_-}{2a_0^{1/2}} - 1 - \frac{3G_\perp^2 v^2 I}{a_0^{1/2} G^2} - \frac{1}{3}\left(\frac{N}{N_0} - 1 \right) \right]_G \right\}, \qquad (1)$$

where χ_0 is the diamagnetic susceptibility of free electrons appropriate to the density of conduction electrons in the metal, $v, \gamma_\pm, N, a_0$, and I are functions of the pseudopotential, the chemical potential, and the reciprocal-lattice vector $\vec{G}$, the expressions for which are given in Eqs. (13), (17), (18), (20), and (36), respectively (with a replaced by a_0) in Ref. 8. Also, N_0 is the total number of electrons per unit volume and $G_\perp$ is the magnitude of the component $\vec{G}$ which is perpendicular to the magnetic field. We have used the model-potential form factors for lithium obtained by Animalu and Heine and tabulated by Harrison.[9] We have calculated χ_d from Eq. (1) which we may write as

$$\chi_d = \chi_0 \Big(1 + \sum_G D_G \Big). \qquad (2)$$

We have evaluated the D_G's and these were summed over the neighbor shells until distinct convergence was obtained. In Table I, the result for the diamagnetic susceptibility of lithium is tabulated. In Table II, we compare our result with the available experimental and theoretical results. We note that our result, which depends sensitively on the choice of the pseudopotential, is in good agreement with the available experimental results.[5,14] Unfortunately, there is a large uncertainty in the experimental results.

Finally, we shall discuss the role of many-body effects on the diamagnetic susceptibility of lithium. There have been a large number of calculations[15] of many-body effects on the diamagnetism of Coulomb gas in the limits of very high or very low densities. However, these results are not valid for metallic densities. Further, there is the problem of departure of an actual system from a uniform electron gas, a consequence of the lattice potential which leads to the band structure. Recently, Philippas and McClure[14] have calculated the effect of the electron-electron interaction on the diamagnetic susceptibility of Bloch electrons using the Green's-function formalism in the Hartree–Fock approximation. They have shown that the result for the diamagnetism can be divided into two terms. The first term, which they call the quasiparticle term, is the diamagnetism calculated treating the self-energy as a one-particle nonlocal pseudopotential, and is easily evaluated by our theory.[8] The second term is an explicit many-body correction to the orbital paramagnetism which has the same form as, but opposite sign from, the

TABLE II. Comparison of χ_d with available results (χ in 10^{-6} cgs volume units).

χ^{expt}		χ^{theoret}						
(Ref. 5)	(Ref. 14)	(Ref. 1)	(Ref. 2)	(Ref. 6)	(Ref. 5)	(Ref. 11)	(Ref. 13)	Present calc.
−0.14 ± 0.15	−0.06 ± 0.16	−0.074	−0.31	−0.13	−0.233	−0.174	−0.150	−0.093

many-body correction to the spin paramagnetism. This term is, in general, difficult to evaluate but they estimate that, for lithium, this term is about 15% of the diamagnetic susceptibility and is of the same sign. Therefore, we estimate that the orbital paramagnetism of lithium is approximately equal to -0.014 cgs volume units.

One of the authors (P.K.M.) is grateful to Professor Leonard Kleinman for his kind hospitality at the University of Texas.

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*Research sponsored by the U. S. Air Force Office of Scientific Research (AFSC) under Grant No. AFOSR 72-2308.

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Concept of the Second-Nearest-Neighbor-Electron Interaction

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(Received 16 October 1972)

The second-nearest-neighbor-electron (SNE) interaction is discussed in comparison with the nearest-neighbor-electron interaction. In tetrahedral crystals, the bond length of the nearest-neighbor electrons is $0.82d_0$, where d_0 is the distance between two atoms. Using this value and the assumption of the Morse-type potential for the SNE interaction, the conclusion is reached that the interaction length l_e for SNE interaction is in the proximity of the inflection point of the Morse-type curve.

In a recent paper[1] concerning the theory of the piezoelectricity of zinc-blende-type and wurtzite-type crystals, the author introduced the concept of the second-nearest-neighbor-electron (π electronic) interaction in tetrahedral crystals. The main results were that the second-nearest-neighbor-electron (SNE) interaction energy has the Morse-type potential, and that, in tetrahedral crystals, the interaction length l_e for SNE interaction is in the proximity of the inflection point of the Morse-type curve. But a clear discussion for these situations was not given. This note attempts to answer the question why the SNE interaction in tetrahedral crystals is in the proximity of the inflection point of the Morse-type potential.

Figure 1 shows the idealized electron model for the ionicity $f_i = 0$ tetrahedral crystals (only one pair of second-nearest-neighbor electrons is shown). $f_i = 0$ means that the covalent electron stands on the center of the two atoms.[2] In this case, the bond length l_e of the SNE interaction is

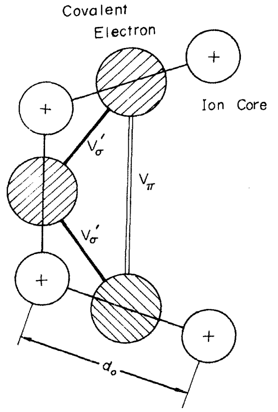

FIG. 1. Electron-structure model for $f_i = 0$ crystals (homopolar crystals). Open circles show the ion core and cross-hatched circles show the covalent electrons. V_σ' shows the ordinary covalent-bonding energy and V_π shows the second-nearest-neighbor electron bonding.