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Theoretical Magnetic Susceptibilities of Metallic Lithium and Sodium

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The magnetic susceptibilities of metallic lithium and sodium are computed by using the best available theoretical information. Bardeen's effective masses and exchange and correlation terms are included. This procedure appreciably affects the theoretical paramagnetism for free electrons, giving results considerably different from those obtained from Pauli's equation with the assumption that the electrons are perfectly free and that exchange and correlation terms may be neglected. The final computed susceptibility for sodium is close to the observed value, but that for lithium is about twice the highest experimental one. The possible sources of error are investigated and it is concluded that the presence of a quartic term in the expansion of the one-electron energy parameter in terms of a power series in the electron wave number would be sufficient to account for the deviation. The level density curves obtained from x-ray emission curves are discussed and it is concluded that all details of these curves cannot be explained on the basis of the simple picture ordinarily used.

1. Introduction

THE purpose of this paper is to apply our present knowledge of the electronic structure of metallic lithium and sodium to a computation of their magnetic susceptibilities. It is convenient to regard the computation from the standpoint of the following four stages of approximation:

1. Paramagnetism of valence electron, neglecting the influence of exchange and correlation energy.
2. Paramagnetism of valence electrons including exchange and correlation effects.
3. Inclusion of diamagnetism of valence electron on the basis of the Landau-Peierls-Wilson theory.
4. Inclusion of ion-core diamagnetism.

The first approximation corresponds to the original Pauli[1] theory of paramagnetism, in which the change in energy arising from the interaction of spin and magnetic field is considered on the basis of the simple band picture. In this approximation, the volume susceptibility is

$$\chi = 2\beta^2 g_s / V, \tag{1}$$

where β is the Bohr magneton, g_s is the density of electronic levels of one spin at the top of the filled region, and V is the volume of the specimen. This term is practically temperature independent.

The next approximation is that in which the spin-aligning exchange forces and opposing correlation forces are taken into account. The qualitative importance of exchange for the susceptibilities of metals was first pointed out by Bloch[2] in connection with his discussion of free electron ferromagnetism, whereas the importance of correlation was pointed out by Wigner.[3] These terms cannot be treated independently of the Pauli term, as we shall see in Section 3. We shall also see that they have a very great influence on the susceptibility of lithium.

The valence electron diamagnetism was first treated by Landau,[4] who showed that for perfectly free electrons the diamagnetic term is

$$-4\pi m\beta^2 h^{-2}(N/3\pi V)^{\frac{1}{3}} \tag{2}$$

where N is the total number of atoms and V the volume of the crystal. The theory was subsequently extended by Peierls[5] and by Wilson[6] for the case in which the one-electron wave functions have the form

$$\psi_{\mathbf{k}}(r) = \chi_{\mathbf{k}}(\mathbf{r}) \exp\left[2\pi i\mathbf{k}\cdot\mathbf{r}\right], \tag{3}$$

in which $\mathbf{k}$ is the electronic wave number vector and $\chi_{\mathbf{k}}$ has the same periodicity as the lattice. In this approximation there are the following three

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[1] W. Pauli, Zeits. f. Physik **41**, 81 (1927).

[2] F. Bloch, Zeits. f. Physik **61**, 545 (1929).

[3] E. Wigner, Phys. Rev. **46**, 1002 (1934).

[4] L. Landau, Zeits. f. Physik **64**, 629 (1930).

[5] R. Peierls, Zeits. f. Physik **80**, 763 (1933).

[6] A. H. Wilson, *The Theory of Metals* (Cambridge University Press, 1936), p. 108.

essentially diamagnetic contributions to the susceptibility:

1. The expression

$$\chi_1=\frac{e^2}{24\pi^2h^2c^2}\int\frac{df}{d\epsilon}\left[\frac{\partial^2\epsilon(\mathbf{k})}{\partial k_x{}^2}\frac{\partial^2\epsilon(\mathbf{k})}{\partial k_y{}^2}-\left(\frac{\partial^2\epsilon(\mathbf{k})}{\partial k_x\partial k_y}\right)^2\right]d\tau(\mathbf{k}),\quad (4)$$

in which

$$f=\frac{1}{\exp\left[\{\epsilon(\mathbf{k})-\epsilon'\}/kT\right]+1}$$

and $\epsilon(\mathbf{k})$ is the energy of the electron having wave number $\mathbf{k}$. ϵ' is a constant. It is assumed in this expression and the following one that the field is in the z direction. Expression (4) reduces to Landau's result (2) when $\epsilon(\mathbf{k})=h^2\mathbf{k}^2/2m$. It may be easily seen that the integrand in (4) is proportional to the Gaussian curvature in $\mathbf{k}$ space of the energy contour associated with the boundary of the filled region.[7] Regions where this curvature is positive add a diamagnetic contribution to the susceptibility, whereas regions of negative curvature add a paramagnetic contribution.

2.

$$\chi_2=-\frac{e^2}{\pi^2mc^2}\int\frac{1}{\exp\left[\{\epsilon(\mathbf{k})-\epsilon'\}/kT\right]+1}d\tau(\mathbf{k})\times\int\left\{\left|\frac{\partial\chi_k}{\partial k_x}\right|^2+\left|\frac{\partial\chi_k}{\partial k_y}\right|^2\right\}d\tau(\mathbf{r}),\quad (5)$$

in which the first integral extends over the spatial coordinates of the electrons and the second extends over all occupied values of $\mathbf{k}$. This quantity is zero for perfectly free electrons and is equal to the atomic diamagnetic susceptibility when the atoms are widely separated. Thus it effectively provides a measure of the amount by which the electrons are "bound."

3. A third term, having no simple analog, which is zero both for perfectly free electrons and perfectly bound electrons and is estimated to be small in intermediate cases.

In previous computations of the susceptibility, the exchange and correlation terms have not been explicitly included, and, in addition, it has usually been assumed that the electrons are perfectly free. We shall include the effects of exchange and correlation as well as may be done at the present time, and shall also employ the approximation in which $\epsilon(\mathbf{k})$ has the form

$$\epsilon(\mathbf{k})=h^2\mathbf{k}^2/2m^*,\quad (6)$$

where m^* is the theoretically determined effective mass, which varies from metal to metal.

2. The Free Electron Paramagnetism Neglecting Exchange and Correlation

By using Eq. (6), Pauli's Eq. (1) may be placed in either of the forms

$$\chi=\tfrac{3}{2}n\beta^2/\epsilon(k_0)=3nm^*\beta^2/h^2k_0{}^2,$$

where k_0 is the value of k at the top of the filled region, and n is the number of electrons per unit volume. This term is practically temperature independent as far as we are concerned.

The value of m^*/m is almost exactly unity for sodium, whereas that for lithium is considerably larger. The most reliable values in these two cases have been determined by Bardeen[8] and are 0.93 and 1.53, respectively. One[9] of us obtained the somewhat smaller value 1.35 in the second case, using a less reliable method.

The values of the volume susceptibilities obtained when Bardeen's values of the effective mass are substituted into Pauli's equation are given in Table I. It should be noted that in this approximation the susceptibility is proportional to the effective electron mass.

3. Inclusion of Exchange and Correlation Terms

The effect of exchange and correlation terms is, in essence, to change the effective electron mass as the electrons are redistributed among the energy levels under the influence of the magnetic field. For this reason, these terms and the Pauli term must be discussed simultaneously.

Let us assume that the metal possesses n electrons per unit volume and that in a given

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[7] Transforming to spherical coordinates in k space, the integral may be written as

$$\int_0^\infty\frac{\partial f}{\partial\epsilon}\frac{1}{3k}\frac{d\epsilon}{dk}\left(2\frac{d^2\epsilon}{dk^2}+\frac{1}{k}\frac{d\epsilon}{dk}\right)4\pi k^2dk,$$

when ϵ is a function of the absolute value k alone.

[8] J. Bardeen, J. Chem. Phys. **6**, 367 (1938).

[9] F. Seitz, Phys. Rev. **47**, 400 (1935).

TABLE I. *The volume susceptibilities given by Pauli's equation. (The values are expressed in c.g.s. units times 10^{-6}.)*

	m^*/m	χ, CALCULATED
Li	1.53	1.22
Na	0.93	0.59

state of magnetization $\frac{1}{2}n-p$ of the electrons occupy the lowest levels of the band associated with one spin and $\frac{1}{2}n+p$ occupy the lowest levels of the band corresponding to opposite spin. If $\epsilon(r)$ is the energy of the state occupied by the rth electron, counting from bottom of the band, in the approximation in which exchange and correlation are neglected (Hartree approximation), the energy in the presence of a field and for the same approximation is

$$E(p, H)=\sum_{r=1}^{\frac{1}{2}n-p}\left[\epsilon(r)+\frac{eh}{4\pi mc}H\right]+\sum_{r=1}^{\frac{1}{2}n+p}\left[\epsilon(r)-\frac{eh}{4\pi mc}H\right]. \tag{7}$$

Pauli's equation may be derived from this by computing the value $p'(H)$ for which this is a minimum, by determining the magnetic moment per unit volume M from the relation

$$M(p'(H))=2p'(H)eH/4\pi mc$$

and by using the equation

$$\chi=\partial M(H)/\partial H. \tag{8}$$

We shall carry through the equivalent procedure after adding exchange and correlation terms.

The exchange energy is a result of interaction between electrons of parallel spin and its average is

$$-0.458e^2/r_s, \tag{9}$$

when p is zero, where r_s is related to n by the equation

$$(4\pi/3)r_s^3=1/n. \tag{10}$$

When p is not zero, the average value for the electrons in the band containing $\frac{1}{2}n+p$ electrons is

$$-0.458xe^2/r_s, \tag{11}$$

where

$$x=[(n+2p)/n]^{\frac{1}{3}};$$

and the average value in the other band is

$$-0.458ye^2/r_s, \tag{12}$$

where

$$y=[(n-2p)/n]^{\frac{1}{3}}.$$

Thus the total exchange energy is

$$-0.458e^2(x^4+y^4)n/2r_s. \tag{13}$$

The correlation energy is principally dependent upon the number of electrons having opposite spins. Its average value when p is zero is

$$-e^2f(r_s)=-0.288e^2/(r_s+5.1a_h) \tag{14}$$

where a_h is the Bohr radius. When p is not zero, the total value is

$$-e^2[(\tfrac{1}{2}n+p)f(r_s/y)+(\tfrac{1}{2}n-p)f(r_s/x)]. \tag{15}$$

Adding Eqs. (7), (13), and (15), and minimizing the result with the assumption that p/n is small, we find

$$p=n\beta H/4\alpha, \tag{16}$$

where

$$\alpha=\frac{n}{4}\left(\frac{d\epsilon}{dr}\right)_{r=\frac{1}{2}n}-\frac{2}{9}\frac{0.458e^2}{r_s}-\frac{5}{9}f'(r_s)r_se^2-\frac{1}{18}f''(r_s)r_s^2e^2. \tag{17}$$

Using Eq. (8), we obtain

$$\chi=n\beta^2/2\alpha. \tag{18}$$

It is easy to see that the exchange term tends to decrease α and hence increase the susceptibility. The first correlation term, containing f', is positive, and hence increases α, whereas the second has opposite sign. Since the second is usually no more than one-tenth of the first in absolute magnitude, the net effect of the correlation term is opposite to that of the exchange term.

The contributions to α are listed in Table II along with the susceptibility computed from Eq. (18). It may be seen that α is reduced by a factor of nearly three in the case of lithium as a result of the influence of exchange and correlation terms. The reduction is not as large for sodium, but is still appreciable. The susceptibilities obtained with and without the correction terms are compared in Table V.

It may be mentioned at this point that the susceptibility determined in the present approximation, and also in the final one, is very sensitive to the value of the effective mass because of the close competition between the first term in α and the other two. Thus a decrease in m^* of the order of ten percent decreases the susceptibility by about twenty-five percent in the case of lithium and about fifteen percent in the case of sodium.

4. THE FREE ELECTRON DIAMAGNETISM

The only appreciable contribution to the susceptibility from the three free electron diamagnetic terms is that arising from Eq. (4). The values obtained from this by using Eq. (6), that is, by neglecting any influence of exchange and correlation on the Gaussian curvature, are given in Table III. We shall discuss the influence of exchange below.

The second term was computed for lithium, by using the wave functions derived by one of us,[9] and was found to be less than 0.02×10^{-6}. The value for sodium should be even smaller, of course, so the term is practically negligible in both cases.

The third term, which also vanishes for perfectly free electrons is of the same order of magnitude as the second, and hence is also negligible.

The question of the extent to which the occupied levels of these metals satisfy the equation

$$\epsilon(\mathbf{k})=h^2\mathbf{k}^2/2m^* \tag{19}$$

will be discussed in more detail in Section 5.

TABLE III. *The free electron diamagnetism obtained from the first term of the Peierls-Wilson theory (neglecting effects of exchange and correlation terms).*

	m^*/m	$\chi\cdot10^6$ (C.G.S.)
Li	1.53	-0.17
Na	0.93	-0.23

TABLE II. *Contributions to α (Eq. (17)), and the volume susceptibility computed from Eq. (18). (The values of terms in α are expressed in ev.)*

	$\frac{1}{4}n(d\epsilon/dr)_{r=\frac{1}{2}n}=\frac{1}{3}\epsilon(k_0)$	EXCHANGE	CORRELATION	TOTAL	$\chi\cdot10^6$(C.G.S.)
Li	1.02	-0.86	0.19	0.35	3.54
Na	1.12	-0.70	0.19	0.61	1.11

Finally, we must consider the effect of exchange and correlation upon the free-electron diamagnetism. Bardeen has pointed out that at the absolute zero of temperature the slope of $\epsilon(\mathbf{k})$ is infinite at the top of the filled region in the approximation including exchange. An examination of the function[10] giving exchange as a function of k at absolute zero of temperature, namely

$$-Ck_0\left(1-\frac{k_0^2-k^2}{2k_0k}\log\left|\frac{k_0-k}{k_0+k}\right|\right), \tag{20}$$

where $C=2e^2$, and k_0 is the value of k at the top of the filled region, shows that the mean value of the second derivative is also infinite (negatively) thereby indicating that the Gaussian curvature is infinite. In order to investigate this point further, we carried through an extensive solution of Bardeen's integral equation[11] for $\epsilon(k)$ for perfectly free electrons, namely,

$$\epsilon(k)=\frac{h^2k^2}{2m}+C\int\frac{1}{\exp\left[\{\epsilon(k')-\epsilon'\}/kT\right]+1}\frac{k'}{k}\times\log\left|\frac{k'-k}{k'+k}\right|dk', \tag{21}$$

where ϵ' is a constant. This was solved to an approximation in which the form of $\epsilon(k)$ near the top of the filled region is given by

$$\epsilon(k)=\epsilon(k_0)+a(k-k_0)+b(k-k_0)^2, \tag{22}$$

where a and b are constants. The solution was obtained by substituting (22) into the integral term, expanding the Fermi-Dirac distribution function to quadratic terms in $(k'-k_0)$, by integrating the result, and then determining implicit equations for a and b by differentiation. The resulting equations are

$$\begin{aligned} a&=\frac{h^2k_0}{m}+C\log\frac{k_0a}{kT},\\ b&=\frac{h^2}{m}-\frac{C}{k_0}\log\frac{Ck_0}{kT}+\frac{3}{2}\frac{C}{k_0}+C\frac{b}{a}. \end{aligned} \tag{23}$$

The solution of these for the vicinity of room

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[10] J. Bardeen, Phys. Rev. **49**, 656 (1936).

[11] E. P. Wigner, Trans. Faraday Soc. **34**, 678 (1938); J. Bardeen, Phys. Rev. **50**, 1098 (1936).

TABLE IV. *The diamagnetic contribution to the magnetic susceptibility from the ion-core terms (after Van Vleck).*

	$\chi \cdot 10^6$
Li	-0.05
Na	-0.18

temperature shows that b is negative, so that, in this approximation, an electron near the top of the filled region behaves like an electron having positive charge. The absolute magnitude of the effective electron mass derived from b varies between 2.5 and 3.0 times the actual mass for lithium and sodium. These results do not agree with experiment, for if they did the Hall coefficient would be positive (opposite to that of bismuth), whereas the observed coefficients are negative. Since the influence of correlations on the density of levels of the entire solid is opposite to that of exchange, it seems likely that the sign of the effective mass is changed back to a positive value when correlations are included.

The Gaussian curvature at the top of the filled region computed from (22) is very close to zero, being only a few tenths the value for perfectly free electrons. If our supposition concerning the influence of correlations is correct, this result would also be altered in a final approximation, presumably in such a direction as to make the Gaussian curvature larger and positive. For this reason, we have employed the free electron diamagnetic terms given in Table III in the final result. Fortunately, the free electron diamagnetism is small near room temperature in any case, so that it seems unlikely that an error in order of magnitude is introduced into the computed susceptibility by this procedure.

It is easy to see from Eqs. (22) and (23) that the reciprocal of the effective electron mass and the integrand of Eq. (4) become negatively infinite as $\log (kT/Ck_0)$ when T approaches zero. It does not seem reasonable to suppose that the influence of correlations will exactly compensate for this. As a result, we may expect a rather mild singularity in the electrical and magnetic properties of the monovalent metals near absolute zero. In particular, the susceptibility should remain paramagnetic and become logarithmically infinite if the influence of exchange is predominant. The variation in conductivity is more difficult to predict, for the mean free path as well as the effective mass and density of states enters into this quantity, and we may expect all three factors to be influenced by exchange. De Haas, Casimir, and van den Berg[12] have found that the resistivity of gold rises with decreasing temperature in the immediate vicinity of absolute zero. It certainly would be unsafe to attempt to identify this anomaly with the influence of exchange at the present time, but the possibility should not be overlooked.

5. THE ION-CORE TERMS

The ion-core terms are small in both lithium and sodium. The values taken from Van Vleck's book[13] are given in Table IV.

6. SUMMARY

The various terms entering the total susceptibility are summarized in Table V and their resultant is compared with the available experimental results.

It may be seen that the agreement between the theoretical values for sodium and the closely correlated experimental ones is as good as may be reasonably expected. This supports the already well-founded belief that the electrons in sodium

TABLE V. *The contributions to the volume susceptibility of lithium and sodium. (The theoretical values are taken from previous tables; all results are expressed in terms of 10^{-6} times the c.g.s. unit).*

	SPIN PARAMAG.	FREE ELECTRON DIAMAG.	ION-CORE DIAMAG.	TOTAL	EXPERIMENTAL VALUES
Li	3.54	-0.17	-0.05	3.32	0.27 to 1.6[14] 2.0[15] 1.4[16]
Na	1.11	-0.23	-0.18	0.70	0.51[14] 0.59[17] 0.59[18] 0.63[19]

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[12] W. J. de Haas, H. B. G. Casimir and G. J. van den Berg, Physica **5**, 225 (1938).

[13] J. H. Van Vleck, *The Theory of Electric and Magnetic Susceptibilities* (Oxford University Press, 1932), p. 359.

[14] K. Honda, Ann. d. Physik **32**, 1027 (1910); M. Owen, *ibid.* **37**, 657 (1912).

[15] F. Bitter, Phys. Rev. **36**, 978 (1930).

[16] S. R. Rao and S. Sriraman, Proc. Ind. Acad. Sci. **5A**, 343 (1937).

[17] W. Sucksmith, Phil. Mag. **2**, 21 (1926).

[18] J. C. McLennan, R. Ruedy and E. Cohen, Proc. Roy. Soc. **116**, 468 (1927).

[19] C. Lane, Phil. Mag. **8**, 354 (1929).

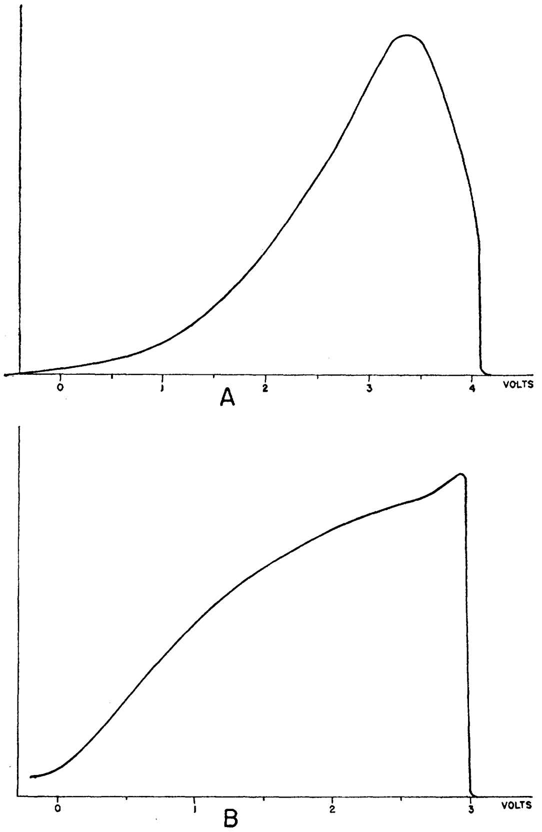

FIG. 1. The relative densities of conduction levels in lithium and sodium as determined from x-ray emission spectra (after Skinner). Curve A is for lithium and is determined from the K spectrum, whereas curve B is for sodium and is determined from the L spectrum.

behave as though they were almost perfectly free. The situation is not nearly so good in lithium, however. In the first place, the experimental values are much less certain, ranging from 0.27×10^{-6} to 2.0×10^{-6}. The most recently determined values[20] are those of Bitter[15] and of Rao and Sriraman,[16] which are, respectively, 2.0×10^{-6} and 1.4×10^{-6}, and presumably are the most reliable. The theoretical value of 3.32×10^{-6} is definitely larger than either of these, a fact indicating that the free electron model favors paramagnetism more than it should.

In this connection, it is interesting to raise the question of the extent to which the occupied levels of these metals satisfy the equation

$$\epsilon(k)=h^2k^2/2m^*.$$

The x-ray emission spectra of lithium and sodium, obtained by O'Bryan and Skinner[21] (Fig. 1) seem to imply that there is a fundamental difference in the behavior of the levels at the top of the filled regions in these two metals. The curve for lithium seems to decrease smoothly from a maximum before dropping abruptly, whereas the curve for sodium does not exhibit the same behavior. If it is assumed that the effect is genuine, as is probably beyond question, there would seem to be three explanations:

1. The difference is intimately connected with the fact that the emission curve for lithium corresponds to K transitions, whereas that for sodium corresponds to L transitions. In the elementary theory developed by Jones, Mott and Skinner,[22] the only difference arising from this source would be that the sodium curve should vary as the three-halves power of the energy before dropping abruptly, whereas the lithium curve should vary as the square root of the energy. It is possible, however, that the emission problem is much more complicated than is assumed in the simple theory. In this event, measurement of the K-emission spectrum for sodium should serve a very useful purpose in giving us a chance to compare curves of the same emission type

2. The difference is connected with a difference in the influence of the zone boundaries in the two cases. This possibility cannot be discarded from consideration because the top of the filled region is very near the zone boundary in the (110) direction in the monovalent body-centered metals. All work on sodium indicates that the electrons in this metal are very nearly perfectly free, so that an influence of the zone boundary would seem more likely in the case of lithium. A somewhat superficial investigation of this problem has led us to conclude that a dip of the type observed in lithium could arise in this way only if the filled region extended beyond the zone boundary in the (110) direction. According to unpublished work of Herring and Huntington,[23]

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[20] C. Starr has kindly communicated to us his as yet unpublished result for the observed volume susceptibility of lithium which is 1.9×10^{-6} c.g.s.

[21] See H. W. B. Skinner, *Reports in Progress in Physics* (Cambridge University Press, 1939), Vol. 5, p. 257.

[22] H. Jones, N. F. Mott and H. W. B. Skinner, Phys. Rev. **45**, 370 (1934).

[23] We are indebted to Dr. Herring and Mr. Huntington for discussions of their work.

this is extremely unlikely and should not be taken as a serious possibility at present.

3. Bardeen[11] has pointed out that the absolute value of exchange energy decreases very rapidly near the top of the filled region, and gives rise to a decrease in the density of levels in this region. In fact, the density of levels would be zero at the top of this region for an ideal gas at the absolute zero of temperature. The problem of electron-electron correlations has not been solved with sufficient completeness to say accurately to what extent this minimum would be influenced by their effects. With the use of Wigner's expression (14) for the average correlation energy, it is possible to compute the change in correlation energy of the system of free electrons when an electron is added to or subtracted from the top of the filled region. This computation shows that in absolute value the change in correlation energy is only slightly greater than half the average value, thereby indicating that the function corresponding to the absolute change of correlation energy when an electron of arbitrary wave number is subtracted from the distribution also drops rapidly near the top of the filled region. Hence correlation does not tend to compensate for the effect of variation in exchange but augments it instead. If, however, we assume that the resultant effect of the variations in exchange and correlation energy for electrons near the top of the filled region is sufficient to explain the decrease in density for lithium, it is difficult to understand why a similar dip does not occur in sodium.

On the whole then, it seems that the problem of the x-ray determined densities does not have a simple solution in terms of the present knowledge of the behavior of levels in the alkali metals.

Although, for the reasons discussed in paragraph 2 above, the influence of the zone boundary in lithium probably is not sufficient to account for a minimum in electron density at the top of the filled region, it is not negligible as far as the susceptibility is concerned. The factors appear to be the following:

1. There should be a slight tendency for the $\epsilon(\mathbf{k})$ curves to bend over in the (110) direction. As far as the diamagnetic terms are concerned, this should decrease the integral (4) since the change in Gaussian curvature (cf. Section 1) would be negative. Moreover, the Pauli paramagnetism should also be increased, for the density of levels is increased. Thus the net effect of the bending in the (110) direction should be an increase in paramagnetism.

2. The $\epsilon(\mathbf{k})$ curve for lithium lies beneath the free electron parabola for small values of k, but should eventually approach this curve for sufficiently large values of $\mathbf{k}$. From the standpoint of Eq. (19), this means that the more accurate $\epsilon(\mathbf{k})$ curve should contain quartic and higher even-powered terms in $\mathbf{k}$ of essentially positive sign. We shall refer to these as quartic terms in the following discussion. Evidently the influence of quartic terms is a diamagnetic one, for they increase the Gaussian curvature at the edge of the filled region, thus increasing (4), and decrease the density of levels, thereby decreasing the spin paramagnetism.

It appears that the most likely source of error in treating lithium is the neglect of the quartic terms in the $\epsilon(\mathbf{k})$ curve. Not only will they operate to reduce the susceptibility, but preliminary calculations by Herring indicate that they will affect the value of $\epsilon(k_0)$ by an amount comparable with that required to obtain closer agreement between the computed and observed susceptibilities. Fortunately a relatively small increase in the contribution to α (Eq. (17)) arising from the Pauli term is required to lower the susceptibility by the proper amount, for this term is almost exactly balanced by the exchange and correlation terms (see Section 3).

We are indebted to Professor J. H. Van Vleck for extensive criticism of this work.