

Brief Reports

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Anisotropic electron-impurity scattering in dilute AlLi: (Small) corrections to the local-density approximation

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We have examined several improvements to a previously reported calculation of Dingle temperatures [L. Cole *et al.*, Phys. Rev. Lett. **42**, 1174 (1979)] for dilute AlLi, including self-interaction corrections and other nonlocal effects in the exchange and correlation potential. The scattering rates (or Dingle temperatures) are changed only slightly from the values obtained previously using the Kohn-Sham density-functional method with the local-density approximation. In particular, the calculated anisotropy remains strong, and the discrepancy with observed isotropy is unresolved.

I. INTRODUCTION

Recently we and our co-workers reported de Hass-van Alphen data, together with theoretical calculations of orbitally averaged electron-impurity scattering rates on the Fermi surface in dilute AlLi.^{1,2} The original calculation¹ used Sorbello's multiple-plane-wave (MPW) formalism³ to account for band-structure-induced anisotropy. Thus the initial and final states were Bloch waves, while the intermediate states were approximated as single plane waves. The scattering of these plane-wave components was calculated using the density-functional formalism,^{4,5} within the local density approximation.⁵ The resulting self-consistent perturbation may be characterized by phase shifts δ_l , so that the scattering rate has the form

$$\tau^{-1}(\vec{k}) = \sum_l \gamma_l(\vec{k}) \sin \delta_l, \tag{1}$$

where the anisotropic coefficients $\gamma_l(\vec{k})$ incorporate the Bloch character of the state \vec{k} . The de Hass-van Alphen

effect measures orbital averages, which are parametrized as Dingle temperatures X_Γ :

$$2\pi k_B X_\Gamma = \hbar \langle \tau^{-1}(\vec{k}) \rangle_\Gamma / \langle 1 + \lambda(\vec{k}) \rangle_\Gamma. \tag{2}$$

Electron-phonon renormalization $\lambda(\vec{k})$ is discussed in Ref. 1. The calculation just described¹ predicted a large anisotropy of X_Γ for orbits on the third zone arm of the Fermi surface (Table I and Fig. 1). This anisotropy mirrors that of the *s*-wave coefficient $\gamma_0(\vec{k})$ and must be expected³ whenever δ_0 dominates the scattering as it does in our case (Table II).

By contrast, very little anisotropy was found experimentally² (Table I), and this has motivated further theoretical work. Reference 2 describes an improved self-consistent treatment that incorporates the Bloch character of the intermediate states (the main effect is lattice backscattering). This requires the use of the Korringa-Kohn-Rostoker (KKR) formalism, as recently discussed in this context in Ref. 6. The improved calculation still produces a large *s*-wave phase shift, and the predicted anisotropy of the X_Γ is

TABLE I. Dingle temperatures as inferred experimentally from de Hass-van Alphen amplitudes (Ref. 2), and as calculated originally in Ref. 1, shown with corrections calculated in Ref. 2 and in the present work. Orbits are identified in Fig. 1.

Orbit	$\psi[110]$	$\psi[111]$	$\alpha[100]$	$\beta[100]$	$\gamma[110]$	$\xi[100]$
<i>X</i> (K/at. %)						
Experiment (Ref. 2)			(42 ± 5)	(35 ± 8)	(40 ± 5)	
Calculation (Ref. 1)	45	40	54	33	81	58
Corrections ΔX						
(Ref. 2)	7	10	1	5	-5	6
Present	-5	-6	-4	-4	-3	-5

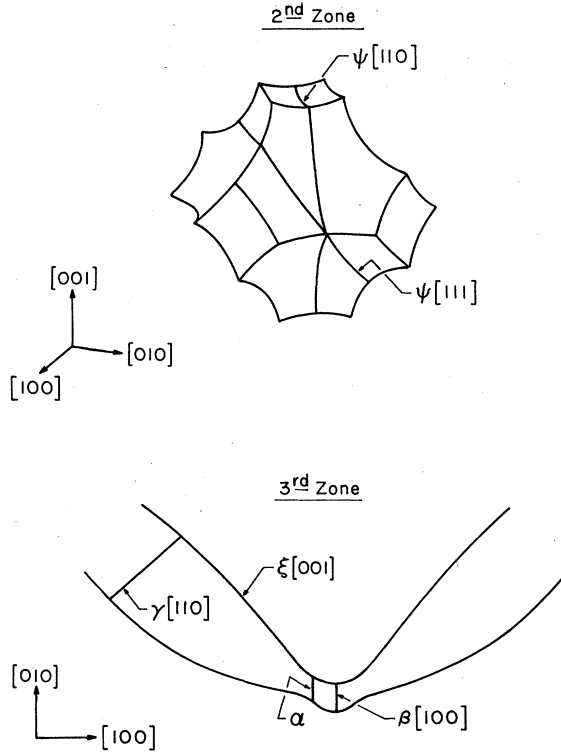


FIG. 1. Extremal orbits on the Fermi surface corresponding to the Dingle temperatures listed on Table I.

reduced only slightly (Table I).

We have also investigated a number of improvements to the local-density approximation, within the MPW formalism. In this Brief Report we describe these calculations and show their separate effects. Taken together, they result in small and rather uniform reduction in the X_{Γ} . The following brief review will be a useful introduction.

In Ref. 1 the Kohn-Sham equations⁵ were solved to find the two Li bound states as well as the scattering states. The effective potential in these equations is

$$v_{\text{eff}}(\vec{r}) = v_0(\vec{r}) + e^2 \int d^3 r' \frac{n(\vec{r}')}{|\vec{r} - \vec{r}'|} + v_{\text{xc}}[n; \vec{r}], \quad (3)$$

where $v_0(\vec{r})$ is the “externally applied” potential of the impurity, and the exchange-correlation (xc) potential

$$v_{\text{xc}}[n; \vec{r}] = \delta E_{\text{xc}} / \delta n(\vec{r}) \quad (4)$$

is obtained from the xc energy functional

$$E_{\text{xc}}[n] = \int d^3 r n(\vec{r}) \epsilon_{\text{xc}}[n; \vec{r}]. \quad (5)$$

The local-density approximation (LDA) is made by letting ϵ_{xc} be the xc energy per particle in a uniform electron gas of density $n(\vec{r})$, i.e., $\epsilon_{\text{xc}}^{\text{LDA}}[n; \vec{r}] = \epsilon_{\text{xc}}(n(\vec{r}))$. The resulting potential is related to the chemical potential

$$v_{\text{xc}}^{\text{LDA}}(\vec{r}) = \frac{d}{dn(\vec{r})} [n(\vec{r}) \epsilon_{\text{xc}}(n(\vec{r}))] = \mu_{\text{xc}}(n(\vec{r})) \quad (6)$$

of a uniform electron gas of density $n(\vec{r})$.

We now discuss various refinements.

II. CORE CORRECTIONS

A. Self-interaction correction (SIC) for Li

The second and third terms of Eq. (3) depend on the total electron density, and thus each contains self-interaction contributions. In principle these cancel, but when E_{xc} is approximated, cancellation may be incomplete. As a result, the LDA predicts the wrong $r \rightarrow \infty$ behavior of $v_{\text{eff}}(\vec{r})$ and $n(\vec{r})$ for the ground state of an atom.^{7,8} A number of schemes⁷⁻¹⁰ for correcting this have appeared in the literature, and we have used a similar one that may be described as follows: subtract from $E_{\text{xc}}^{\text{LDA}}$ the self-interaction of each occupied orbital $\Psi_{\alpha}(\vec{r})$ that would remain if all the other electrons were taken away. Minimization of the resulting total energy functional E^{SIC} produces a state-dependent xc potential

$$v_{\text{xc}}^{\text{SIC}}(\alpha; r) = \mu_{\text{xc}}(n(\vec{r})) - e^2 \int d^3 r' \frac{n_{\alpha}(\vec{r}')}{|\vec{r} - \vec{r}'|} - \mu_{\text{xc}}(n_{\alpha}(\vec{r})), \quad (7)$$

where $n_{\alpha}(\vec{r}) = |\psi_{\alpha}(\vec{r})|^2$. A detailed discussion of the SIC as formulated within spin-density functional theory is given by Perdew and Zunger.¹⁰

The SIC improves the charge distribution of the two bound electrons, and this in turn affects the scattering of the conduction electrons. Table II shows the changes in the Fermi-level phase shifts that result from SIC corrected bound states. δ_0 is decreased because the bound states are pulled closer to the Li nucleus, making it less attractive to the scattering electrons. Since $\delta_0 < 0$ to begin with, the scattering rates are increased but only slightly.¹¹ This effect is outweighed by the others to be discussed.

TABLE II. Phase shifts δ_l computed in Ref. 1, and various corrections computed in the present work.

l	0	1	2	3
δ_l (Ref. 1)	-1.214	-0.341	-0.148	-0.033
Corrections $\Delta\delta_l$				
SIC in Li	-0.026	-0.004	0.004	0.001
V_{ps} core in Al	0.040	0.047	0.002	-0.013
ADA	0.034	0.035	-0.014	-0.007
Total Correction	0.048	0.078	-0.008	-0.019

B. Pseudopotential core correction for the Al vacancy

The applied potential is the difference between those due to the Li and Al ions,

$$v_0(\vec{r}) = -3\frac{e^2}{r} - v_{\text{Al}}(\vec{r}) \quad (8)$$

The Al vacancy was characterized in Ref. 1 as a hole in jellium, and accordingly the pseudopotential core contribution was volume averaged. This procedure had proven to be effective in the calculation of vacancy formation energies.¹² In the present context we consider the scattering of Bloch waves, for which the localized Al ions present no perturbation. This suggests an alternative approach in which jellium is not introduced, and we simply use $v_{\text{Al}}(\vec{r}) = -3e^2\Theta(r - R_c)/r$, with the Ashcroft core parameter $R_c = 0.59 \text{ \AA}$. This tends to localize the perturbation without changing its average. As a result, δ_0 is increased and correspondingly the X_{Γ} are decreased.

III. NONLOCALITY IN E_{xc}

Methods to account for nonlocal dependence of E_{xc} on $n(\vec{r})$ have been proposed recently by Gunnarsson, Jonson, and Lundqvist¹³ and by Alonso and Girifalco.¹⁴ In Ref. 13, for example, one approximates $\epsilon_{\text{xc}}[n; \vec{r}]$ by $\epsilon_{\text{xc}}(\bar{n}(\vec{r}))$ similarly as in the LDA, but with the xc energy per particle evaluated at an averaged density

$$\bar{n}(\vec{r}) = \int d^3r' w(\vec{r} - \vec{r}'; \bar{n}(\vec{r})) n(\vec{r}') \quad (9)$$

This is called the average density approximation (ADA). The weighting function w is chosen in Ref. 13 so that $E_{\text{xc}}^{\text{ADA}}$ reproduces the exact leading correction $\sim [n(\vec{r}) - n(\vec{r}')]^2$ to E_{xc} for the almost-homogeneous electron gas. Their results are tabulated in the form of the Fourier transform $\tilde{w}(q, r_s)$. To facilitate our self-consistent calculation, we follow Cole and Harmon¹⁵ and fit the Ref. 13 results by simply taking an average (in real space) over a sphere with radius $R = k_F^{-1}(n(\vec{r}))$, which fits the more exact \tilde{w} quite well (Fig. 2) for $r_s = 2$. (For aluminum, $r_s = 2.07$; the local r_s value differs appreciably from this only within a small volume near the origin, where the density is larger and the LDA should be relatively good.) The changes in the phase shifts are again small (Table II), and contribute to a net fairly isotropic decrease in the Dingle temperatures.

In principle, one should go further and consider the nonlocality of v_{xc} itself. The present formalism provides an exact description of ground-state properties, but not of the quasiparticles:¹ Sham and Kohn¹⁶ pointed out that the latter are described by Schrödinger-like equations with a nonlocal, energy-dependent self-energy in place of $v_{\text{eff}}(\vec{r})$ [Eq. (3)]. Because of the increased difficulty inherent in using a nonlocal self-energy here, we have not attempted it.

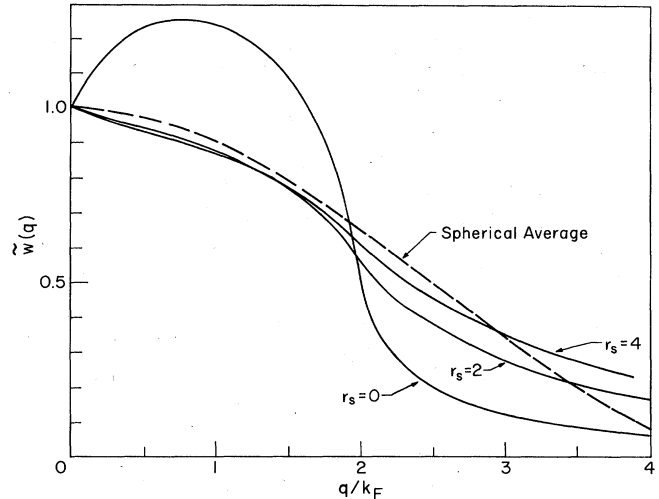


FIG. 2. Fourier transform $\tilde{w}(q)$ of the average density approximation weight function $w(\vec{r} - \vec{r}')$, as calculated in Ref. 13 for various r_s values (solid lines), and as calculated using the spherical average described in the text (dashed line).

IV. CONCLUSIONS

Improvements to the LDA considered here appear to be more important for bound states, whose effective potential is changed qualitatively (i.e., $-1/r \rightarrow -2/r$ at intermediate distances $a_0 < r < 1/k_F$), than for the scattering states. We conclude that the LDA provides an accurate $v_{\text{eff}}(\vec{r})$ appropriate for the ground-state properties of the conduction electrons. We leave open the question whether nonlocal self-energy is important for the treatment of quasiparticle scattering.

Of the many factors we have considered, only one weakens s -wave scattering sufficiently compared with p wave to remove the predicted anisotropy: if the Li is assumed to be interstitial, the X_{Γ} are predicted in the range 27–40 K. However, the neutron scattering work of Solt and Werner¹⁷ indicates that the Li is substitutional.

Finally, the corrections found here and in the lattice backscattering study of Ref. 2 are both small in magnitude. In the latter case, however, they do reduce the anisotropy. We anticipate that further development of the KKR method will be useful for problems of this kind.

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¹¹The four phase shifts shown in Table II are not constrained to satisfy the Friedel sum rule; we retain eight phase shifts in the self-consistent calculation, and these obey it automatically to within less than 1%.

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