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Theory of the de Haas–van Alphen effect for heavy-fermion alloys

A Wasserman†‡, M Springford† and A C Hewson§

† School of Mathematical and Physical Sciences, University of Sussex, Brighton BN1 9QH, UK

§ Department of Mathematics, Imperial College, London SW2 2B2, UK

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Abstract. Luttinger's approach for a theory of the de Haas–van Alphen effect is applied to mean-field theory of the SU(N) Anderson lattice model. Using a single-channel approximation we obtain an expression for the fermion mass enhancement and its magnetic field dependence. The results are applied to the heavy-fermion compound CeB_6 where the field dependence of the mass is compared with measurements from de Haas–van Alphen and specific heat experiments.

The de Haas–van Alphen (DHVA) effect is primarily used to investigate quasi-particle aspects of metals and alloys such as their Fermi surfaces and effective masses. Recently the class of materials referred to as heavy-fermion (HF) compounds has been studied by this technique. When interpreted in terms of standard Lifshitz–Kosevich (LK) (1955) theory for the DHVA amplitude, the results appear to be generally similar to results for conventional materials except for highly enhanced quasi-particle masses, ranging up to several hundred times the expected band masses (Reinders *et al* 1986). Band-structure calculations, which have long been the primary theoretical tool for interpreting DHVA results, cannot readily explain these enormous mass enhancements even though in several instances the calculated and DHVA measured Fermi surfaces are in good agreement. As a consequence it is generally thought that the mass enhancements arise from many-body interactions, in particular through hybridisation between conduction electrons and localised but highly correlated 4f or 5f electrons on the rare-earth sites of the HF alloy.

In view of the apparently strong local many-body character in HF materials it is initially surprising that LK theory with greatly enhanced electron masses seems adequate for expressing observed field and temperature dependences of DHVA amplitudes. Even though small departures from the LK quasiparticle expression due to many-body effects have been observed in the case of electron–phonon interactions (Elliot *et al* 1980, Khalid *et al* 1988), it is well known that many-body effects appear primarily as a renormalisation of the LK quasi-particle mass, and the Fermi surfaces of several HF materials have recently been mapped out on this basis (Reinders *et al* 1986, Joss *et al* 1987, Taillefer *et al* 1987).

In the course of these DHVA investigations a magnetic-field-dependent quenching of the quasi-particle mass has also been observed in some HF compounds (Joss *et al* 1987,

‡ Permanent address: Department of Physics, Oregon State University, Corvallis, OR 97331, USA.

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Onuki *et al* 1988) with a similar effect observed in specific heat measurements (Bredl 1987, Amato *et al* 1987, Stewart *et al* 1988). An extension of LK theory which allows for the effect of strong on-site correlations while preserving its observed simplicity seems then an appropriate and helpful objective for gaining better understanding of the various DHVA results in HF compounds.

An approach to a DHVA theory which allows for inclusion of some kinds of interaction was developed by Luttinger (1960, 1961), Fowler and Prange (1965) and Engelsberg and Simpson (ES) (1970). This approach relies on a conventional perturbation expansion of the thermodynamic potential in terms of interacting fermions. However, the strong local correlations which are important in HF materials suggest a different and less familiar description in which the unperturbed state is already a many-body state. In this case the standard techniques of many-body theory, which are basic to the ES method, are not obviously applicable.

The locally correlated HF system is usually described in terms of the Anderson model (see, e.g., Fulde 1988). Among the methods for taking account of strong local correlations in this model is one that introduces auxiliary boson fields and constraint fields to restore some familiar aspects of many-body methods (Coleman 1984). These novel 'slave boson' methods have been used within the framework of less conventional perturbation theories (for a recent review see Bickers (1987)) and with functional integral methods (Read and Newns 1983, Read 1985) to illuminate several aspects of HF system behaviour. An especially appealing and physically intuitive result of the 'slave boson' approach is the mean-field approximation. This approximation, which is exact for the case of the dilute rare-earth alloys in the limit of large orbital degeneracy N, has also been applied to the Anderson lattice of rare-earth atoms (for a recent review see Newns and Read (1987)). The lattice mean-field theory has the attractive consequence of furnishing quasi-particle 'band' states that incorporate strong on-site correlations, so that it becomes possible, within the framework of Luttinger's approach, to provide a straightforward theory of the DHVA effect which resembles LK theory in a form reminiscent of the ES treatment of electron–phonon effects but with a contribution characteristic of the HF model instead.

The mean-field limit has recently been applied to the DHVA effect by Rasul (1989) using functional integration. Rasul also considered Gaussian fluctuations about the mean field to find the corrections of order $1/N$ and found them to be of little consequence for the DHVA effect. The method used here is the direct application of Luttinger's approach to HF mean-field theory. By noting that only one angular momentum channel contributes any significant amplitude, we make a single-channel approximation and obtain results which correspond, generally, to Rasul's but provide an extension of that work in describing both the mass enhancement and its magnetic field dependence.

Our starting point is the SU(N) Anderson lattice model into which 'slave bosons' b_i on the ith site have been introduced to take account of strong (infinite) on-site correlations:

$$H = \sum_{k,m} \varepsilon_{k,m} c^{\dagger}_{k,m} c_{k,m} + \sum_{i,m} (E_{\mathrm{F}} - mh) f^{\dagger}_{i,m} f_{i,m} + V \sum_{i,m} (f^{\dagger}_{i,m} c_{i,m} b_i + \mathrm{HC}) + \lambda \left(\sum_{i,m} f^{\dagger}_{i,m} f_{i,m} + \sum_i b^{\dagger}_i b_i - \sum_i 1 \right). \tag{1}$$

In the SU(N) model the conduction electrons ($c_{k,m}$), of wavevector $\boldsymbol{k}$ and angular momentum m, are considered to be N-fold degenerate, corresponding to local f-state

(f_{im}) degeneracy. For the present a magnetic field lifts only the 'spin' degeneracy of the local f levels and conduction electrons, with $h = g\mu_B H$ the reduced magnetic field, g the electron g-factor, μ_B the Bohr magneton and H the magnetic field. The scalar Lagrange multiplier λ constrains the combined occupancy of the ith site singlet bosons (b_i) and local f electrons (f_{im}) to unity.

The mean field is taken in equation (1) with $\langle b_i \rangle = z$ to give

$$H = \sum_{k,m} \varepsilon_{k,m} c^\dagger_{k,m} c_{k,m} + \sum_{i,m} (E_F - mh) f^\dagger_{i,m} f_{i,m} + Vz \sum_{i,m} (f^\dagger_{i,m} c_{i,m} + \text{HC}) + \lambda \left(\sum_{i,m} f^\dagger_{i,m} f_{i,m} + z^2 \sum_i 1 - \sum_i 1 \right). \tag{2}$$

Using

$$c_{i,m} = \frac{1}{\sqrt{N_s}} \sum_k \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{R}_i) c_{k,m} \qquad f_{i,m} = \frac{1}{\sqrt{N_s}} \sum_k \exp(\mathrm{i}\boldsymbol{k} \cdot \boldsymbol{R}_i) f_{k,m}$$

where N_s is the number of sites and $\boldsymbol{R}_j$ is the location of the jth site, the Hamiltonian can be fully transformed into the $\boldsymbol{k}$-space of the Brillouin zone to give

$$H = \sum_{k,m} \varepsilon_{k,m} c^\dagger_{k,m} c_{k,m} + \sum_{k,m} (E_F + \lambda - mh) f^\dagger_{k,m} f_{k,m} + Vz \sum_{k,m} (f^\dagger_{k,m} c_{k,m} + \text{HC}) + \lambda(z^2 - 1). \tag{3}$$

Redefining $E_F + \lambda = \mu + T_A$, with μ the chemical potential and T_A the Kondo temperature, one has in the magnetic moment limit $E_F \ll 0$

$$T_A \simeq D \exp(-2D|E_F|/NV^2).$$

This description is thought to be appropriate for the mixed valence states of Ce rare-earth ions in materials such as $CeCu_6$, $CeAl_2$ or CeB_6.

Within this simplest quasi-particle model the thermodynamic potential can be written

$$\Omega = -T \operatorname{Tr} \left(\sum_n \log[-\mathbf{G}^{-1}(\zeta_n)] \right) \tag{4}$$

where Tr is a trace and $\mathbf{G}^{-1}$ is the inverse of the Green function matrix

$$\mathbf{G} = \begin{pmatrix} G_{ff}(\zeta_n) & G_{fc}(\zeta_n) \\ G_{cf}(\zeta_n) & G_{cc}(\zeta_n) \end{pmatrix}$$

with $\zeta_n = \mathrm{i}(2n+1)\pi T + \mu$, T the temperature and n the set of positive and negative integers. $G_{ff}(\zeta_n)$, $G_{cc}(\zeta_n)$, $G_{fc}(\zeta_n)$ and $G_{cf}(\zeta_n)$ are the Fourier-transformed time-ordered fermion Green functions continued to the imaginary energy axis. As a result of the simplicity of the Hamiltonian in the mean-field approximation, we easily find these to be

$$G_{ff}(\zeta_n) = [\zeta_n - (E_F + \lambda - mh) - V^2 z^2/(\zeta_n - \varepsilon_{k,m})]^{-1} \tag{5}$$

$$G_{cc}(\zeta_n) = \{\zeta_n - \varepsilon_{k,m} - V^2 z^2/[\zeta_n - (E_F + \lambda - mh)]\}^{-1} \tag{6}$$

$$G_{fc}(\zeta_n) = G_{cf}(\zeta_n) = Vz/(\zeta_n - \varepsilon_{k,m})[\zeta_n - (E_F + \lambda - mh) - V^2 z^2/(\zeta_n - \varepsilon_{k,m})]. \tag{7}$$

The $\varepsilon_{k,m}$ refer to the band states with angular momentum channels m which correspond to those of the f-electron sites. Using the relation

$$\mathrm{Tr}(\log \mathbf{A}) = \mathrm{Tr}[\log(\det \mathbf{A})]$$

equation (4) gives for the thermodynamic potential

$$\Omega = -T\,\mathrm{Tr}\left\{\sum_{n,m=-J}^{m=J} \log\left[\left(\zeta_n - \varepsilon_{k,m} - \frac{V^2 z^2}{\zeta_n - (E_\mathrm{F} + \lambda - mh)}\right) \times [\zeta_n - (E_\mathrm{F} + \lambda - mh)]\right]\right\} \tag{8}$$

where the sum over $2J + 1$ magnetic sublevels contained in the trace operation is explicitly indicated. Following Luttinger, to obtain the DHVA effect to leading order in ω_c/μ we replace the conduction band levels $\varepsilon_{k,m}$ with Landau levels according to

$$\varepsilon_{k,m} \rightarrow \varepsilon(p, k_z, m) = k_z^2/2m_\mathrm{B} + \omega_c(p + \tfrac{1}{2}) - mh \tag{9}$$

with $\omega_c = eH/m_\mathrm{B}$, where m_B is the band effective mass, and we assume that any magnetic field dependence introduced into the hybridisation matrix element may be neglected. (Apart from the case $N = 2$ it is not clear how to interpret these N-fold Zeeman-split Landau levels, but formally it does not affect the following argument.) The trace, which now covers the Landau levels, contributes

$$\mathrm{Tr} = \frac{m\omega_c}{2\pi^2}\int_{-\infty}^{\infty} \mathrm{d}k_z \sum_{p=0}^{\infty}.$$

The thermodynamic potential (equation (8)) consists of a term that will eventually contribute oscillations in magnetic field H, as well as a non-oscillatory term. Ignoring the non-oscillatory term, we have

$$\Omega_\mathrm{osc} = -T\,\mathrm{Tr}\left[\sum_{\substack{n \\ m=-J}}^{m=J} \log\left(\frac{k_z^2}{2m} + \omega_c(p + \tfrac{1}{2}) - mh - \zeta_n + \frac{V^2 z^2}{\zeta_n - (E_\mathrm{F} + \lambda - mh)}\right)\right] \tag{10}$$

which is the starting form of ES theory for electron–phonon interactions (Engelsberg and Simpson 1970). The subsequent analysis can be done in several ways. A convenient method is to use a complex integral representation for the logarithm (Wasserman and Bharatiya 1979) and following that scheme the trace, contour integral and remaining manipulations are easily performed to find the oscillatory contribution to the thermodynamic potential:

$$\Omega_\mathrm{osc} = -\frac{2(m_\mathrm{B}\omega_c)^{3/2}T}{\pi^2}\sum_{\substack{n \\ m=-J}}^{m=J}\sum_{r=1}\frac{(-1)^r}{r^{3/2}}\cos\left(\frac{2\pi r\{\mu + mh - \mathrm{Re}[X(\zeta_n, m)]\}}{\omega_c} - \frac{\pi}{4}\right)$$
$$\times \exp\left(-\frac{2\pi r}{\omega_c}[\zeta_n - \mathrm{Im}[X(\zeta_n, m)]]\right)\exp\left(-\frac{2\pi^2 r T_\mathrm{D}}{\omega_c}\right) \tag{11}$$

where only ζ_n in the upper half-plane are summed, and where

$$\mathrm{Re}[X(\zeta_n, m)] = -V^2 z^2 (T_\mathrm{A} - mh)/[\zeta_n^2 + (T_\mathrm{A} - mh)^2] \tag{12}$$

$$\mathrm{Im}[X(\zeta_n, m)] = -\zeta_n V^2 z^2/[\zeta_n^2 + (T_\mathrm{A} - mh)^2]. \tag{13}$$

The conduction electron 'spin sum' is absorbed into the DHVA phase and, for completeness, the conventional impurity scattering factor containing the Dingle temperature T_D, which is assumed channel independent, has also been included. This is, essentially, the ES result for a HF system in the mean-field approximation. Substituting the mean-field value $V^2z^2 = n_f T_A/N\rho_0$, where n_f is the mean occupancy of the f level and ρ_0 is the unperturbed conduction band density of states, we find that, at low temperatures $T \ll T_A$, and ignoring ζ_n in the denominators of $\mathrm{Re}[X(\zeta_n, m)]$ and $\mathrm{Im}[X(\zeta_n, m)]$,

$$\Omega_{osc} = -\frac{2(m_B\omega_c)^{3/2}T}{\pi^2}\sum_{m=-J}^{J}\sum_{r=1}\frac{(-1)^r}{r^{3/2}}\cos\left[\frac{2\pi r(\mu + 2Dn_f/N)}{\omega_c} + g\frac{m_B}{m_0}\pi rm\left(1+\frac{2Dn_f}{NT_A}\right) - \frac{\pi}{4}\right] \times \sum_n \exp\left[-\frac{2\pi^2 r}{\omega_c}\zeta_n\left(1+\frac{2Dn_fT_A}{N(T_A - mh)^2}\right)\right]\exp\left(-\frac{2\pi^2 rT_D}{\omega_c}\right) \tag{14}$$

where $\rho_0 = 1/2D$ is the density of conduction band states taken to be uniform in energy, with $2D$ the conduction band width.

When $\mathrm{Re}[X(\zeta_n, m)]$ is expanded to first order in the magnetic field, equation (14) gives an expression which describes the DHVA effect as arising, additively, from $2J + 1$ sets of quasi-particle Fermi surface, each with the same frequency but with different amplitudes and different phases. The amplitude contribution from each of these $2J + 1$ channels arises from quasi-particles of different enhanced masses. But, for finite fields and temperatures, only the lightest of these quasi-particles, i.e. $(m = -J)$, contributes a significant DHVA amplitude. It is assumed then that this will be the observed DHVA channel and the measured quasi-particle mass and phase will be associated with this channel.

When only a single channel m is considered, the sum over ζ_n is easily done to give

$$\Omega_{osc} = -\frac{2(m_B\omega_c)^{3/2}T}{2\pi^2}\sum_{r=1}\frac{(-1)^r}{r^{3/2}}\cos\left[\frac{2\pi r(\mu + 2Dn_f/N)}{\omega_c} + g\frac{m_B}{m_0}\pi rm\left(1+\frac{2Dn_f}{NT_A}\right) - \frac{\pi}{4}\right] \times \left\{\sinh\left[\frac{2\pi^2 rT}{\omega_c}\left(1+\frac{2Dn_fT_A}{N(T_A - mh)^2}\right)\right]\right\}^{-1}\exp\left(-\frac{2\pi^2 rT_D}{\omega_c}\right) \tag{15}$$

and, with $m = -J$, the observable DHVA amplitude $A(r, -J)$ can be written

$$A(r, -J) = [\![\sinh\{(2\pi^2 rT/\omega_c)[1 + 2Dn_fT_A/N(T_A + Jh)^2]\}]\!]^{-1} \tag{16}$$

where the effective mass ratio m^*/m_B for this channel is

$$m^*/m_B = 1 + 2Dn_fT_A/\,N(T_A + Jh)^2. \tag{17}$$

In weak fields $(Jh \ll T_A)$

$$m^*/m_B = 1 + 2Dn_f/NT_A \tag{18}$$

which is the weak-field magnetic susceptibility and specific heat enhancement in the mean-field approximation (Newns and Read 1987). This is also the DHVA quasi-particle

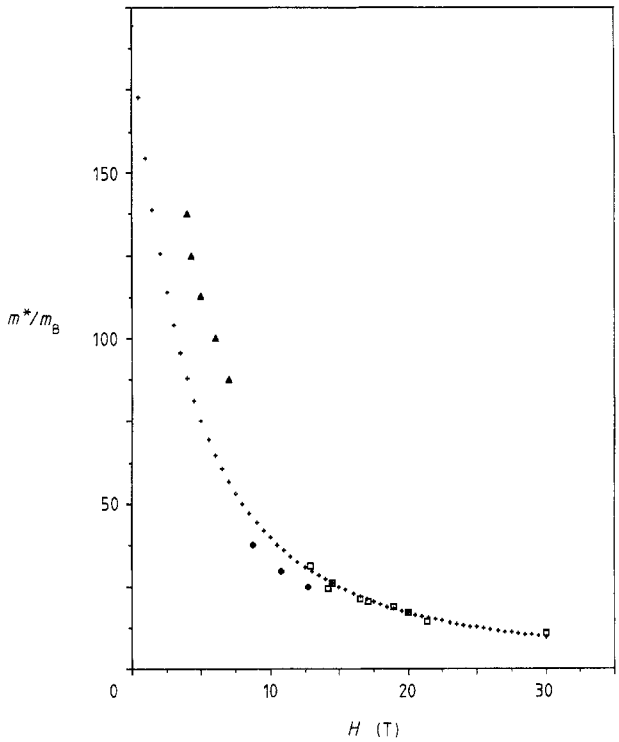

Figure 1. Enhanced quasi-particle mass ratio m^*/m_B as a function of the magnetic field: □, DHVA results from Joss *et al* (1987); ◆, DHVA results from Onuki *et al* (1988) (band mass ratios are obtained by normalisation of the experimental DHVA mass ratios to a calculated band mass of 0.56 (Norman and Min 1988)); ▲, ratio of the measured specific heat (Bredl 1987) to a calculated band density of states of 1.8 mJ mol^{-1} K^{-2} (Norman and Min 1988); +, calculated from equation (17) with parameters $T_A = 15$ K, $g = 1.1$, $2Dn_f = 1.5$ eV and $J = \frac{5}{2}$ (with these parameters the value of m^*/m_B at $H = 0$ is 194).

mass result of Rasul (1989). Within this Landau level description, $\mathrm{Re}[X(\zeta_n, m)]$ gives the shifted chemical potential per channel, which at $T = 0$ and small fields is

$$\tilde{\mu}(m) = (\mu + n_f/N\rho_0) + mh(n_f/N\rho_0 T_A + 1)$$

from which the large-N magnetisation M can be found:

$$M = \rho_0 \sum_m m\left(1 + \frac{n_f}{N\rho_0 T_A}\right)mh = \tfrac{1}{3}\mu_{\mathrm{eff}}^2 H\left(\frac{N}{2D} + \frac{n_f}{T_A}\right)$$

where

$$\mu_{\mathrm{eff}}^2 = g^2\mu_B^2 J(J+1).$$

The single-channel expression, equation (17), describes a magnetic-field-dependent DHVA mass with high-field effective-mass quenching. Mean-field theories are generally considered to be weak-field theories requiring $mh \ll T_A$, but this restriction roughly coincides with the region in which the $m = -J$ amplitude dominates the DHVA effect. However, in keeping with the spirit of a high-field DHVA theory and noting that when $mh \simeq T_A$ the mth-channel DHVA amplitude vanishes, we conjecture that a high-field continuation of this result accounts for the general features of an effective-mass quenching which according to this description approaches the band mass. Mass quenching has been seen in the specific heats of $CeCu_6$ and CeB_6 (Bredl 1987, Stewart *et al* 1988, Amato *et al* 1987). But, in the DHVA effect it has only been seen thus far in CeB_6 so that comparative experimental data are limited. The combined experimental masses measured for CeB_6 by the DHVA effect by Joss *et al* (1987) and Onuki *et al* (1988) are shown in figure 1, together with several low field m^*/m_B values estimated by Norman

and Min (1988) based on the specific heat measurements of Bredl (1987). Equation (17) with parameters shown in figure 1 gives a reasonable account of the experimental values of m^*/m_B. No attempt has been made to optimise the fit. Norman and Min fit the limited high-field data using a quite different expression, obtained form the resonance level Kondo model of Schotte and Schotte (1975), which seems unable to account of the low-field data. The result found from the present work seems consistent with both high- and low-field mass enhancements obtained from DHVA and specific heats, respectively.

The field dependence of the effective mass in $CeCu_6$ observed in specific heat measurements (Amato *et al* 1987, Stewart *et al* 1988) was not observed in DHVA measurements over the range 6–10 T (Springford and Reinders 1988). This result seems inconsistent with equation (17) which infers a discernible magnetic field mass quenching.

From equation (15), it may be seen that the DHVA oscillation frequency of each channel is shifted from its unenhanced value by $2Dn_f/N$. This small T_A-independent term suggests that the Fermi surface should not be strongly affected by hybridisation, a result which may help to understand why the Fermi surfaces of $LaAl_2$ and LaB_6 bear a strong resemblance to those of $CeAl_2$ and CeB_6, respectively (Springford and Reinders 1988). However, a similar correspondence between $LaCu_6$ and $CeCu_6$ is not found (Onuki *et al* 1987, Springford and Reinders 1988).

In assessing models for the HF lattice it is not clear how artificial the SU(N) model is for describing the angular momentum states of conduction electrons other than $m = \pm\frac{1}{2}$. In the case of conventional metals and alloys where the DHVA amplitudes are not spin dependent the $N = 2$ model gives a normal 'spin-splitting' contribution to the DHVA amplitude $C_r = \cos(\pi g m_B r/2m_0)$ where, as before, m_B is the band mass and m_0 is the bare electron mass. In the HF system for the case $N = 2$, Rasul (1988) uses $C_r = \cos(\pi g m_H r/2m_0)$ where m_H is the zero-field HF enhanced mass. Because of our single-channel approximation the 'spin-splitting' results given here take a different form from Rasul's with a somewhat different interpretation.

In the expression for the DHVA thermodynamic potential of equations (14) and (15) a channel-dependent contribution generated by the SU(N) model appears in the argument Φ_r of the cosine term:

$$\Phi_r = 2\pi rF/H - \psi_r \tag{19}$$

where F is the DHVA frequency for an extremal orbit. Incorporating the harmonic factor $(-1)^r$ into ψ_r, we have the J-dependent phase

$$\psi_r = g(m_B/m_0)\pi rJ(n_f/N\rho_0 T_A + 1) + \pi r + \pi/4 \tag{20}$$

so Φ_r remains finite in the notional limit of infinite magnetic field. In some cases this infinite-field phase can be measured, modulo π. Its value could indicate some further constraints on lattice models for HF materials.

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