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LETTER TO THE EDITOR

On the de Haas–van Alphen effect in inhomogeneous alloys

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Abstract

We show that Landau-level broadening in alloys occurs naturally as a consequence of random variations in the local quasiparticle density, without the need to consider a relaxation time. This approach predicts Lorentzian-broadened Landau levels similar to those derived by Dingle using the relaxation-time approximation. However, rather than being determined by a finite relaxation time τ, the Landau-level widths instead depend directly on the rate at which the de Haas–van Alphen frequency changes with alloy composition. The results are in good agreement with recent data from three very different alloy systems.

The relaxation-time approximation (RTA), in which quasiparticles scatter in random events at a characteristic rate τ^{-1}, has proved invaluable in understanding the electrical resistivity and thermal conductivity of metals [1]. It has also been used to treat the de Haas–van Alphen (dHvA) effect, i.e. magnetic quantum oscillations of the magnetization [2, 3]. Dingle showed that the Lorentzian broadening of Landau levels in metals, observed using the dHvA, is qualitatively explained by the RTA for elastic scattering [4]. The predicted exponential damping of quantum oscillations due to impurities, with a characteristic scaling temperature known as the Dingle temperature $T_D = \hbar/2\pi k_B \tau$, was subsequently verified experimentally for the vast majority of known metals [2, 3].

In spite of this apparent success, the RTA proves inadequate when considering thermodynamic functions of state, such as the electronic specific heat γT or magnetic susceptibility χ, because of the formation of bound states associated with impurities [5]. More generally, impurities, surfaces or defects of any kind will modify the lattice periodicity and the local potential of the crystal, so the quasiparticle wavefunctions (i.e. the eigenstates of a perfect, periodic, infinite crystal [1]) will no longer be eigenstates of the modified crystal [1, 6, 7][1]. The assumption that the consequent alteration of, for example, the Landau-level structure can be parametrized merely by a relaxation rate τ^{-1} [4] seems to be a considerable oversimplification.

[1] The CPA (coherent potential approximation—a mean-field-theory approach) [6, 7] is often used to calculate the 'smearing' (in energy and wave-vector, $\boldsymbol{k}$) of the electronic states in metallic solid solutions.

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A deeper problem arises in the alloy $Hs_{1-x}Do_x$, where Hs represents the host material and Do the dopant impurity. When x becomes ~ 0.5, one should be concerned as to how far the distinction between 'host' and 'dopant' can be stretched. As numerous experiments have shown (see e.g. references [8–12]), the quasiparticle wavefunctions are no longer those of the host material, weakly perturbed by enhanced impurity scattering. By contrast, Dingle's model only considers the statistical broadening of Landau levels, without considering how the underlying band structure is being modified by the alloying [4].

In view of the difficulty in separating effects due to the variation of the band structure with x and effects due to the changes in the actual quasiparticle lifetime, we have chosen to abandon the RTA and instead adopt a semi-empirical approach based on statistical variations of the local quasiparticle density in an alloy. The essential advantage of this approach is that the sensitivity of the electronic structure to x is considered as the starting point. Our model is therefore related to the work of Woltjer [13]. Using a spatially varying electron density, Woltjer was able to provide convincing simulations of Shubnikov–de Haas oscillations and the quantum Hall effect in two-dimensional semiconductor systems without the need to invoke localization [13][2]. However, in contrast to Woltjer's work, which involved numerical simulations of experimental data, in the current letter we provide an analytical solution which predicts the damping of dHvA oscillations.

Our model is applicable to systems for which the lattice parameters are only weakly dependent on x and for dHvA oscillations arising from circular Fermi-surface cross-sections[3]. Encouragingly, it predicts Lorentzian-broadened Landau levels similar to those described by Dingle [4][4]. However, rather than being determined by a finite relaxation time τ, the Landau-level widths instead depend directly on the rate at which the dHvA frequency $F(x)$ changes with x, enabling estimates to be made that compare very favourably with experiment.

The starting point of the model is the fact that x, the local concentration of Do in Hs, is subject to statistical variations about the mean value $\bar{x}$. This will lead to a spread of Fermi-surface cross-sections $A(x)$ about the mean $A(\bar{x})$. As the dHvA frequency is given by $F = (\hbar/2\pi e)A$ [2], the variation in A in turn leads to a spread of dHvA frequencies $F(x)$ and to phase-smearing effects [2, 4]. In this letter we restrict ourselves to simple circular Fermi-surface cross-sections of k-space area A that are easy to relate to the local quasiparticle concentration $N(x)$ [1]. However, rather than assuming that $N(\bar{x})$ is linearly dependent on $\bar{x}$, we choose a semi-empirical approach whereby $A'(\bar{x})$ and $F'(\bar{x})$, the derivatives respectively of the mean cross-section $A(\bar{x})$ and mean dHvA frequency $F(\bar{x})$ with respect to $\bar{x}$, are those obtained from experiment. The dHvA frequency $F(x)$ corresponding to a particular value of x can then be obtained from

$$F(x) \approx F(\bar{x}) + [x - \bar{x}]F'(\bar{x}). \tag{1}$$

Given the finite separation, a, between ions[5], the average number of Hs and Do ions encountered in a quasiparticle path comprising p orbits of the circular cyclotron trajectory is determined by binomial statistics; the path will be of length $2\pi p l_c(\bar{x}) = 2\pi p\sqrt{2\hbar F(\bar{x})/eB^2}$. For such a path, $m \approx xn$ of these will be of the Do type and $n - m \approx [1 - x]n$ of these will be

[2] By analogy with the conclusions of the present letter, Woltjer's work might also explain why Landau levels appear to have Lorentzian line shapes in two-dimensional systems [14] for which models based on the RTA predict otherwise [15, 16].

[3] It remains to be tested whether our approach can adequately accommodate large lattice mismatches between Hs and Do.

[4] In this context, it is interesting to note that the CPA predicts Lorentzian broadening of the so-called 'Bloch spectral function' in the presence of substitutional disorder [7].

[5] For a simple cubic system, a is the lattice parameter, while more generally, $a = N^{1/3}$, where N is the total ion concentration inclusive of both Hs and Do ions.

of the Hs type. The probability that m of these ions are of type Do, corresponding to a local dopant concentration $x = m/n$ (and local dHvA frequency $F(x)$), is therefore

$$p(m,n) = \frac{\bar{x}^m[1-\bar{x}]^{n-m}n!}{m![n-m]!}. \tag{2}$$

Under standard experimental conditions, n will always be a large number for metals. Taking the 'necks' of Au as one example of a Fermi-surface cross-section ($F_N \approx 1530$ T) that is not especially large [3], $a \sim 2.6$ Å while $l_c \sim 1400$ Å in a magnetic field of $B \sim 10$ T, implying that $n \sim 540$. Clearly, it would be impractical to work with such a large number of terms in calculations. It is well known, however, that the skew factor $\eta = 1/\sqrt{6\bar{x}[1-\bar{x}]n}$ for the binomial distribution vanishes for large n, causing the binomial distribution to become well approximated by the normal distribution [17]. In implementing this approximation, the mean becomes $\mu_m = n\bar{x}$ and the variance becomes $\sigma_m^2 = n\bar{x}[1-\bar{x}]$. Following the established idea that a variation in F results in a 'phase smearing' which produces a damping of the dHvA effect [2,4] the damping factor becomes the result of the Fourier transformation:

$$R_i \approx \int_{-\infty}^{+\infty} \frac{n}{\sigma_m} \exp\left(\frac{-[x-\bar{x}]^2n^2}{2\sigma_m^2}\right) \cos\left(\frac{2\pi p[x-\bar{x}]F'(\bar{x})}{B}\right) \mathrm{d}x. \tag{3}$$

Making the substitution $\phi \equiv 2\pi p[x-\bar{x}]F'(\bar{x})/B$, and performing the integration in the ϕ-domain, we obtain

$$R_i \approx \int_{-\infty}^{+\infty} \frac{1}{\sigma_\phi} \exp\left(\frac{-\phi^2}{2\sigma_\phi^2}\right) \cos(\phi)\, \mathrm{d}\phi \approx \exp\left(\frac{-\sigma_\phi^2}{2}\right) \tag{4}$$

where

$$\sigma_\phi^2 = \frac{2\pi p\bar{x}[1-\bar{x}]F'(\bar{x})^2 a}{B}\sqrt{\frac{e}{2\hbar F}} \tag{5}$$

is now the phase variance.

The expected symmetry between Hs and Do is immediately seen in the presence of terms in both $[1-\bar{x}]$ and $\bar{x}$ in equation (5), enabling this model to be applied across an entire alloy series $0 < x < 1$. More satisfyingly, since the exponent is linear in both the harmonic index p and $1/B$, the functional form of R_i is exactly that obtained by Dingle using the RTA [4]. After inverse Fourier transformation and the parabolic band substitution $F = m^*E/\hbar e$, a Lorentzian Landau-level line shape

$$f(E) = \frac{\Gamma}{\pi[E^2+\Gamma^2]} \tag{6}$$

is obtained, with the level width Γ given by

$$\Gamma = \frac{\bar{x}[1-\bar{x}]F'(\bar{x})^2 a}{m^*}\sqrt{\frac{\hbar e^3}{8F}}. \tag{7}$$

This latter result implies that a Lorentzian broadening of the Landau levels results naturally from frequency-smearing effects caused by the substitution of dopants without needing to consider the concept of a relaxation time.

We now consider whether this model can account for a significant amount of the Landau-level broadening observed in well known alloy systems. Three experiments involving approximately circular Fermi-surface cross-sections are considered.

Ag impurities in Au. Dilute alloys of the form Ag_xAu_{1-x} provide a useful test case, since the lattice parameters of Au and Ag are very similar and the neck orbit, giving rise to a dHvA frequency of $F_N \approx 1530$ T in Au, is thought to be very nearly circular [3]. To facilitate a

comparison with existing experimental data, the results of our model are stated in terms of an effective Dingle temperature

$$T_{\mathrm{D}} = \frac{\bar{x}[1-\bar{x}]F'(\bar{x})^2 a}{\pi k_{\mathrm{B}} m^*}\sqrt{\frac{\hbar e^3}{2F}}. \tag{8}$$

On inserting the appropriate values for $Ag_{0.01}Au_{0.99}$, of $F'_{\mathrm{N}} \sim 650$ T, $m^* \sim 0.29\,m_{\mathrm{e}}$ and $a \sim 2.7$ Å for the face-centred cubic lattice [1–3], we obtain $T_{\mathrm{D}} \approx 1.1$ K for each per cent of Ag, which compares favourably with the experimentally obtained value of 0.8 K for each per cent of Ag [8, 11]. Thus our model for Landau-level broadening appears to be able to predict reasonable values for the observed T_{D} for Ag_xAu_{1-x}.

Kondo alloys. Our model can also be applied to the $Ce_xLa_{1-x}B_6$ series. Because this is a Kondo system in which the effective mass varies with the magnetic field, it is more meaningful to present the results in terms of a mean free path, since this quantity is not renormalized by the interactions that give rise to Kondo behaviour [18]. In having abandoned the RTA, however, we have in effect also abandoned the concept of a mean free path. Nevertheless, by making a comparison with the formulae of Dingle, we can define an *effective mean free path*,

$$l_{\mathrm{eff}} = \frac{2\hbar F}{e\bar{x}[1-\bar{x}]F'(\bar{x})^2 a}. \tag{9}$$

On inserting the appropriate values of $F_{\alpha,3} \sim 7970$ T, $F'_{\alpha,3} \sim 550$ T and $a \sim 4.0$ Å, for the worst-case alloy $x = 0.5$ [12], we obtain $l_{\mathrm{eff}} \approx 350$ nm, which is within experimental error of the maximum value obtained experimentally [12]. Thus, this model can explain why the broadening of the Landau levels in the $Ce_xLa_{1-x}B_6$ intermetallic compounds was observed to be unexpectedly low [12].

A doped insulator. Finally, it is interesting to consider the case of a doped insulator, to investigate whether one should expect to observe the dHvA effect in such systems. Low-density, weakly ferromagnetic electron-gas systems, which have been of recent interest, certainly fall into the category, and dHvA oscillations have been observed [10]. The simplest model is that of a variable density, N, where $N(x) = xN'$. On deriving $F(x)$ and $F'(x)$ for a spherical Fermi surface, we obtain

$$\frac{-\sigma_\phi^2}{2} = \frac{p}{24}[1-x]\frac{\hbar a}{eB}N'. \tag{10}$$

Interestingly, at very low concentrations, $x \to 0$, the extent to which the quantum oscillations are damped does not depend on x. Since no estimates of T_{D} or l_{eff} have been published [10], the best we can do is estimate the lowest field at which we should expect quantum oscillations to be observed. According to Dingle [4], the threshold field is given approximately by the inequality $\omega_{\mathrm{c}}\tau \gtrsim 1$. The equivalent inequality according to our model is $\phi^2 \gtrsim 2\pi$. Upon substituting $a \sim 4.0$ Å and $N' \sim 5 \times 10^{27}$ m^{-3} [10], we obtain $B \gtrsim 9$ T. This is in very good agreement with experiment; Goodrich [19] reports that dHvA oscillations are only observed at fields of 10 T and greater.

In summary, we have shown that Lorenzian broadening of Landau levels in alloys can be derived statistically by considering a distribution of Fermi-surface cross-sections, without invoking the relaxation-time concept. We have considered three alloy systems, and shown that the extent to which the Landau levels are broadened, or the extent to which the quantum oscillations are damped, compares favourably with experiment. In view of the current experimental interest in alloy systems, we hope that this letter stimulates further consideration of, for example, more general Fermi-surface shapes and the effect of lattice mismatch between host and impurity.

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