# Quasiparticle picture for the de Haas–van Alphen effect

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(Received 17 October 1977)

Physical arguments are given for the quasiparticle behavior observed in the de Haas–van Alphen effect. An upper limit to the maximum deviation from quasiparticle behavior which may be observed is given and the asymptotic behavior as $T \to 0$ is derived. These results indicate why the deviations from quasiparticle behavior might appear small, even for large magnetic fields. Misinterpretations of earlier work are clarified and an application of the picture to experiments on nearly ferromagnetic palladium-nickel alloys is made.

In our work, Simpson and I found[1] that the de Haas–van Alphen (dHvA) oscillations are affected by electron-phonon interactions only to the extent that the cyclotron frequency $\omega_c^* = eH/m^*c$, which occurs in the amplitude of the oscillations, has the electron band mass $m$ renormalized to $m^*$. There are no further temperature dependencies due to $m^*$ or due to the electron lifetime $\tau_{ep}(T)$ as seen in transport properties. This quasiparticle result was shown[1] to be valid as long as the cyclotron frequency $\omega_c^*$ is much less than any characteristic phonon frequency $\omega_{ph}$. $\omega_{ph}$ is defined as the lowest frequency peak in the phonon density of states. If there are no peaks at lower frequencies, then $\omega_{ph}$ is the Debye frequency. In almost all physical circumstances we have $\omega_c^* \ll \omega_{ph}$. In our model, the mass enhancement $m^*/m$ was precisely the same as that occurring in the low-temperature electronic specific heat. This was due to the fact that we considered all electron orbits to be spherical with identical isotropic coupling to the phonons. In most metals this is not the case. Tomlinson and Carbotte[2] have calculated the anisotropy of the coupling for lead. From their work we would expect $m^*/m$ for most orbits to be close to the average seen in the specific heat.

Since the results we found were presented rather mathematically and they are of importance to experimentalists in the field, I should like to give some more physical interpretations to our results.

In Ref. 1, it was shown that it is *not* the real or imaginary part of the self-energy evaluated at the Fermi energy which contributes to the dHvA effect. It is the analytic continuation of the full self-energy evaluated at the poles of the Fermi function $\omega_n = (2n+1)\pi kT$. The same sum over poles appears in all oscillatory thermodynamic effects, even without interactions. It arises from an integral which weights an oscillatory function $e^{i\omega\epsilon}$ with the Fermi function $f(\epsilon)$. The oscillatory function brings in the discrete electron thermal energies $\omega_n$ above the Fermi energy separated by $2\pi k_B T$. The theorem we proved in I, Eq. (3.4), the "generalized Fowler-Prange" theorem, may be restated as follows. If $(\omega_n/\omega_{ph})^2 \ll 1$ for all $\omega_n$ which contribute significantly to the oscillatory behavior, then the renormalization effects due to electron-phonon interactions enter through

$$\zeta(\omega_n, T) \simeq \omega_n \int_0^\infty d\nu \frac{2\alpha^2(\nu)F(\nu)}{\nu} = \omega_n \left(\frac{m^*}{m} - 1\right).$$

Note that the function $\zeta(\omega_n, T)$, which represents the entire effect of the phonon interaction, is real. Restating this in yet another way, we may say that for $\omega_c^* \ll \omega_{ph}$, those electron states which contribute to the dHvA amplitude ($\epsilon < \omega_n \lesssim \omega_c^*/\pi$), do not probe any detailed phonon structure. They see only one of the moments of the phonon spectrum, the one that gives $m^*/m$.

I should like to review some facts about the dHvA effect which go into the proof of the above statements. Temperatures such that $\pi kT \geq \hbar\omega_c/\pi$, represent the high-temperature limit for the dHvA effect. In this case only the first pole at $\omega_0 = \pi kT$ makes a significant contribution. The lowest thermal level precisely probes the $m^*/m$ moment of the phonon spectrum. As usually stated[3] one needs a fairly sharp Fermi surface on the scale of $\omega_c$ in order for oscillations to appear. The oscillations are due to electrons being dumped out of a quantized Landau level as the level moves through the Fermi energy with the variation of the magnetic field. If $kT \gg \hbar\omega_c$, the electrons are thermally spread over many Landau levels above the Fermi level and no change in occupation numbers occur as a Landau level moves through the Fermi energy.

For $kT \ll \hbar\omega_c$, the dHvA effect includes the sum over many electron thermal levels $\omega_n$, up until $\omega_n \approx \omega_c/\pi$. For higher thermal levels the amplitude is negligible. Still, as long as $\omega_c \ll \omega_{ph}$, the detailed phonon structure is not probed by the electrons, they see only the average which leads to an $m^*/m$ renormalization of the mass. This is the

 

case for most orbits in most metals. However, if one can find situations where $\omega_c$ approaches $\omega_{ph}$, deviations from undamped quasiparticle behavior should occur. In order to make a somewhat realistic calculation Simpson and I chose to use the measured phonon density of states of mercury ($\omega_{ph} \approx 21\,^{\circ}\mathrm{K}$), rather than a Debye or Einstein model. To get a large cyclotron frequency $\omega_c^*$ we chose a large magnetic field, 100 kG, and an electron orbit with a low band mass, $m^*/m = 0.183$. With the realistic density of states chosen, a computer calculation was used to obtain the deviations. The numerical work showed that in this case deviations of the amplitude from quasiparticle behavior as large as 17% were possible. The experiments done by Palin[4] on mercury in a slightly smaller field (82 kG) were consistent with our predicted deviations. The way this worked out was that a (2 to 3)% change in slope of the $\ln[A(T)/T]$-vs-$T$ plot could account for the predicted deviations. Since the thermal expansion of mercury seems to be changing the band mass by this amount, the experiments[4] to see the predicted deviations in the most favorable case were exceedingly difficult.

I should like to emphasize that there is a simple and direct method for evaluating the deviations from quasiparticle behavior. Given the model for the phonon spectrum and the coupling to electrons ($\alpha^2F$) for a particular orbit, the following equations [(3.4) and (4.1) of Ref. 1] will give the detailed predictions for the amplitude in the dHvA effect:

$$\zeta(\omega_n) = \pi k_B T \int_0^\infty d\nu \frac{2\alpha^2(\nu)F(\nu)}{\nu} \times \left\{1 + 2\sum_{m=1}^{n}\left[1+\left(\frac{2\pi m k_B T}{\nu}\right)^2\right]^{-1}\right\},$$

$$A = \sum_{n=0}^{\infty} \exp\left[-\left(\frac{2\pi}{\omega_c}\right)[\omega_n + \zeta(\omega_n)]\right].$$

The thermal levels which exist along the imaginary energy axis should become as useful in describing equilibrium thermodynamic effects as the imaginary part of the energy is useful for describing lifetime effects in transport processes.

We may do some further analysis using this approach to help elucidate why the deviations from quasiparticle behavior grew at most to 17% in the example of Ref. 1 where the magnetic field was chosen to be 100 kG, $m^*/m = 0.183$, and the electron-phonon mass enhancement $1+\lambda = 2.6$. The amplitude was shown in Eq. (4.6) of Ref. 1 to have a lower limit

$$A \geq [2\sinh(2\pi^2 k_B T/\hbar\omega_c^*)]^{-1} \equiv A^0 .$$

As we argued earlier, for $k_B T/\hbar\omega_c^* \gtrsim 1$, $A$ approaches this lower limit, which is the quasiparticle result. The largest deviations from quasiparticle behavior occur in the amplitude for $k_B T/\hbar\omega_c^* \ll 1$. In this limit the amplitude must be handled quite carefully. A large number of thermal levels $\omega_n$ must be summed over. This point is emphasized by Mueller and Myron.[5] In the limit $\omega_c^*/\omega_{ph} \gg 1$, $kT/\hbar\omega_c^* \ll 1$, the self-energy that enters the amplitude may be approximated by $\zeta(\omega_n) \gtrsim \pi k_B T\lambda$. Thus in the low-temperature very-high-field limit the greatest deviations from quasiparticle behavior will occur. An upper limit for the amplitude may be estimated using the lower limit on the self-energy:

$$A < [\exp(-2\pi^2\lambda k_B T/\hbar\omega_c)]/[2\sinh(2\pi^2 k_B T/\hbar\omega_c)] .$$

The ln of the deviations are measured by

$$\ln\frac{A}{A^0} < \ln\frac{[\exp(-2\pi^2\lambda k_B T/\hbar\omega_c)]\sinh(2\pi^2 k_B T/\hbar\omega_c^*)}{\sinh(2\pi^2 k_B T/\hbar\omega_c)} .$$

In the limit $kT/\hbar\omega_c^* \ll 1$, we have

$$\ln(A/A_0) < \ln(1+\lambda) = \ln(m^*/m) .$$

Thus for fields such that $\omega_c/\omega_{ph} \gg 1$, the upper limit for the maximum deviation we might expect is $\ln(1+\lambda)$, equal to 0.96 for mercury. In this light, it is perhaps not too surprising that for $\omega_c^*/\omega_{ph} \gtrsim 1$, our previous numerical work has shown deviations only as large as $\ln(A/A_0)$ equal to 0.17. The work of Mueller and Myron[5] shows larger deviations for fields larger than 100 kG but is limited by the upper limit given above.

Our previous numerical work indicated the difficulty in obtaining a numerically convergent series for $kT/\hbar\omega_c^* \ll 1$. The details of this slow convergence were emphasized in Ref. 5. Their analysis may be used to show that in the very-low-temperature limit

$$\lim_{T\to 0}\frac{\partial}{\partial T}\ln(A/A^0) = 0 .$$

Thus the extrapolation of the amplitude to zero temperature is simple once the asymptotically flat region is reached for $\ln(A/A^0)$. This would then imply that the curve drawn in Fig. 4 of Ref. 1 has reached its maximum value and may be extended to $T=0$ with zero slope.

With the above analysis one may gain some more insight as to why deviations from the quasiparticle result are not readily observable. An upper limit to the deviations from quasiparticle behavior is the ln of the electron-phonon mass enhancement. This deviation will occur only for fields much greater than the highest characteristic phonon frequency. The most a magnetic field can do to the mass enhancement is remove all of it. The deviation will

not appear in the high-temperature limit $k_B T \gtrsim \hbar\omega_c^*$ as we argued earlier, but will grow as the temperature is lowered to be much less than $\hbar\omega_c^*/k_B$. Thus, the change in slope of a $\ln(A/A^0)$ plot as a function of temperature will be small except under extreme experimental conditions. Mueller and Myron[5] have calculated explicit values for the deviations in the high-field limit.

Now I would like to clarify a misunderstanding in the work of Gantmakher.[6] He claims that the results of our work apply only to mercury since we used numerical methods in the last stage of our calculations. From our original work and the restatement above it should be clear that as long as $\omega_c^* \ll \omega_{ph}$, an undamped quasiparticle behavior should describe the dHvA effect. For $\omega_c^* \lesssim \omega_{ph}$, the deviations will be small and will depend on the detailed orbit in the particular metal we are considering. Because of its strong-coupling low-lying, phonon modes and low-mass electron orbit, mercury is just about the best metal to use to see any deviation from undamped quasiparticle behavior in the dHvA effect. In the largest fields then available, with the smallest mass orbits, the effects of deviations are theoretically predicted to be of the order of 2% in the measured slope which is just about the accuracy of the experiments.[4]

Gantmakher also interprets our work physically by the statement that the influence of the variations of the scattering probability with temperature offsets the changes in effective mass. In fact, the way the problem was formulated $\tau_{ep}$ does not appear at all. We could, if we wished, follow a suggestion of Hopfield,[7] and rewrite the mass enhancement term at a very *high temperature* as follows:

$$A = \exp\left(-\frac{2\pi^2 k_B T}{\hbar\omega_c}\frac{m^*}{m}\right)$$

$$= \exp\left[-\frac{2\pi^2 k_B T}{\hbar\omega_c}\left(\frac{m^*}{m}-1\right)\right]\exp\left(-\frac{2\pi^2 k_B T}{\hbar\omega_c}\right)$$

$$= \exp\left(-\frac{2\pi}{\omega_c\tau_{ep}}\right)\exp\left(-\frac{2\pi^2 k_B T}{\hbar\omega_c}\right),$$

where at high temperatures[1] we have (for the electron-phonon problem $kT \gg \hbar\omega_{ph}$)

$$\frac{1}{\tau_{ep}} = \mathrm{Im}\Sigma(\omega_{n=0}, T) \simeq \frac{\pi}{\hbar} k_B T \int_0^\infty d\nu \frac{2\alpha^2(\nu)F(\nu)}{\nu},$$

$\exp(-2\pi^2 k_B T/\hbar\omega_c)$ is the amplitude without mass enhancement, and $\exp(-2\pi/\omega_c\tau_{ep})$ is the electron-phonon lifetime effect. Here we have used the high-temperature limit of the phonon occupation function $n(\nu) \approx k_B T/h\nu$, and the identity

$$\frac{m^*}{m} - 1 = \int_0^\infty d\nu \frac{2\alpha^2(\nu)F(\nu)}{\nu}.$$

I do not believe that this helps elucidate the physics. First, because it is not true in general but only when $k_B T \gg \hbar\omega_{ph}$. Second, the dHvA oscillations are observable at temperatures up to but not much greater than $\hbar\omega_c^*/k_B$ [due to the amplitude factor $\exp(-2\pi^2 k_B T/\hbar\omega_c^*)$]. Thus the dHvA high-temperature regime is almost always the low-temperature regime for electron-phonon interactions.

Next I would like to question the conclusions in Ref. 6. Gantmakher has approached the problem on the basis of "classical" calculations of the electron lifetime $\tau_{ep}$ ($1/\nu_{e,ph}$ in Gantmakher's notation) in the presence of the electron-phonon interaction. The conclusion that the calculated $\tau_{ep}$ is consistent with experiment seems to be based on the following. The experimental results of Palin[4] for $\ln(A/T)$ vs $T$ are used and the effect of $1/\tau_{ep}$ $[\alpha T^{3/2} X_{e,ph}(4^0)]$ is subtracted. The new points found are shown to fit a straight line as well as the line drawn through the original experimental points. That would be just fine if there were no importance to the slope of the line. Palin very carefully determined what the slope of the line should be by doing auxiliary experiments at a series of fixed temperatures. He plotted $\ln(A/H^{1/2})$ vs $1/H$ to get the effective mass. The results were consistent to within 2% with the effective mass determined by the slope of $\ln(A/T)$ vs $T$. When we use the figure shown in Gantmakher's work,[6a] we find that the change in slope made by his $\tau_{ep}$ correction is much larger than 2% (~30% for the 82-kG case).

This predicted deviation is yet another indication that a simple interpretation of the electron-phonon interaction as producing a lifetime effect, which may be represented as a Dingle temperature, is invalid. There may be another formulation of the dHvA effect which makes use of the real and imaginary parts of the self-energy along the real energy axis. The formulation we have used[1] is valid and is the most direct one available.

In their work, Mueller and Myron[5] suggest higher-field low-temperature experiments to discern the difference between the predictions of our work and Gantmakher's.

The last subject I would like to discuss is the application of our work to nearly ferromagnetic systems. In a recent experimental work Hörnfeldt, Dronjak, and Nordborg[8] examined the dHvA effect in the nearly ferromagnetic alloy PdNi (0.35-at.% Ni). They observed a rather large shift in the position of the spin-split zero, which was interpreted as an exchange enhancement of the spin-splitting factor due to the Ni impurity. They also find that the cyclotron mass is unenhanced for the particular orbit studied. This observation contradicts the prediction we made,[1] since the specific heat is found to be enhanced by 6% at this alloy composi-

tion. There are several possible explanations. The most likely is that the electrons in this orbit are too weakly coupled to spin fluctuations to see the effect of this amount of Ni. In fact Hörnfeldt *et al.* analyze a particular orbit (on the $\Gamma 6$ sheet) and estimated a 6% change in the susceptibility enhancement factor ($\Delta S_c'/S_c = 0.25/4$) upon the addition of 0.35-at.% Ni to Pd. Now in the original PdNi alloy experiments[9] it was found that the enhancement of specific heat is much less than the enhancement of the susceptibility, in qualitative agreement with theory.[10] If we use the experimental ratio $(\Delta\gamma/\gamma_{Pd})/(\Delta\chi_F/\chi_{Pd}) \approx 0.23$ given by Chouteau *et al.*[9] for these low Ni concentrations we would predict a change in mass enhancement of the orbit considered by Hörnfeldt *et al.*[8] of 1.4%. This estimate is in agreement with their measurement that $m^*$ is unchanged to within the experimental accuracy of (3–4)%.[11]

Possibly one of the most significant discoveries found in the work of Hörnfeldt *et al.*[8] is the rather low value of the Dingle temperature ($T_D \sim 1$ °K) for 0.35-at.% Ni impurity. As they point out, ordinarily it takes only 100 ppm magnetic impurities in noble metals to reach this value for the Dingle temperature. Experimentally, Ni behaves more as a host atom than an impurity scatterer when dissolved in Pd. This fact opens the field of nearly ferromagnetic alloys to study by the dHvA effect. The results on other alloy compositions of PdNi should be quite interesting. In addition, the RhNi nearly ferromagnetic alloys offer another interesting system to examine.

ACKNOWLEDGMENTS

I would like to thank C. Gould and Dr. W.F. Brinkman for inquiries that prompted this investigation. I would also like to thank Dr. D. Shoenberg, Professor F. M. Mueller, and Professor S. Hörnfeldt for interesting correspondence. This work was supported in part by NSF Grant No. DMR 74-18030.

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