

*INFLUENCE OF QUASILOCAL STATES ON THE DE HAAS-VAN ALPHEN EFFECT IN METALS  
OF THE BISMUTH TYPE*

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We obtained the positions and widths of the quasilocal energy levels of an electron in a non-ideal metal situated in a magnetic field. We show that such levels lead to an oscillating addition to the free energy, with a frequency differing slightly from the fundamental frequency. The laws governing the experimentally observed beats are explained.

**1. INTRODUCTION**

THE influence of impurities on the de Haas-van Alphen effect have been discussed already many times. Dingle<sup>[1]</sup> was the first to assume that the collisions between the electrons and the impurities lead to a broadening of the Landau levels. As a result, each harmonic of the oscillating addition to the thermodynamic quantities contains an additional factor  $\exp(-2\pi p/\omega_c \tau)$ , known as the Dingle factor. Here  $p$  is the number of the harmonic,  $\omega_c = eH/mc$  is the cyclotron frequency, and  $\tau$  is a relaxation parameter formally introduced by Dingle.

A paper by Bychkov<sup>[2]</sup> is devoted to a proof of Dingle's assumption. By summing an important class of diagrams for the electron Green's functions, he showed that  $\tau$  is proportional to the free path time in the absence of the magnetic field. Dingle's assumption turns out to be valid if  $\omega_c \tau \ll \xi/\hbar \omega_c$  ( $\xi$ —chemical potential). In the opposite limiting case, the influence of the impurities reduces to the Dingle factor only in the regions  $|\Delta| \gg (\hbar \omega_c)^2/\xi$ , where  $\Delta$  is the energy reckoned from the Landau level<sup>[2]</sup>. Williamson, Foner, and Smith<sup>[3]</sup>, using the method of I. M. Lifshitz and Kosevich<sup>[4,5]</sup>, generalized Dingle's theory to the case of an arbitrary Fermi surface. They showed that in this case, too, a Dingle factor appears, in which  $m$  should be taken to mean the effective mass of the electron moving on the extremal section of the Fermi surface in the magnetic field. Brailsford<sup>[6]</sup>, also considering the case of an arbitrary shape of the Fermi surface, found that the impurities lead not only to a change in the amplitude of the oscillations, but also of their period. These changes are due to the broadening and shift of the Fermi level under the influence of the impurities<sup>[7]</sup>.

The purpose of this article is to ascertain how the de Haas-van Alphen effect is influenced by the quasilocal electron states that arise when a non-ideal metal is placed in a magnetic field.

**2. IMPURITY OF AN ELECTRON IN A MAGNETIC FIELD**

We confine ourselves to a consideration of metals with a small number of carriers, in which a small amount of atoms of another element is dissolved. In this case the dispersion of the electron in the ideal metal

can be regarded as quadratic, and the impurity potential can be regarded as  $\delta$ -like:

$$U(\mathbf{r}) = -U_0 \sum_i \delta(\mathbf{r} - \mathbf{R}_i).$$

Here  $\mathbf{R}_i$  is the radius vector of the  $i$ -th impurity ion. We shall assume that  $U_0 > 0$ , i.e., the impurities lead, by assumption, to the occurrence of  $\delta$ -like potential wells that are randomly distributed over the lattice. Actually we are considering the case of a shallow potential well of small radius, when the well itself does not lead to the formation of bound states. The magnetic field, limiting the motion of the electron in two degrees of freedom, contributes to the occurrence of a bound state<sup>[8-10]</sup>. In the interval  $(0, \hbar \omega_c/2)$  there appears a local level due to the existence of the bound state of the electron in the magnetic field<sup>[8,9]</sup>. In addition, at the points alternating with the Landau levels, under certain conditions, an appreciable increase in the spectral density, typical of quasilocal levels takes place.

Calculations similar to those used for a zero magnetic field<sup>[11,12]</sup> yield an equation for the local and quasilocal levels:

$$1 + U_0 F_0(E) = 0, \quad (1)$$

where

$$F_0(E) = \lim_{\epsilon \rightarrow 0} G_0(0, 0; E + i\epsilon) + \pi i \nu_0(E);$$

$G_0(\mathbf{r}, \mathbf{r}'; E)$  is the Green's function of the electron in an ideal metal situated in the magnetic field, written in the coordinate representation;  $\nu_0(E)$  is the corresponding density of states per unit volume. The solution of (1) is of the form  $E = E' + iE''$ , with  $E'' \sim \nu_0(E')$ , so that we can speak of quasilocal levels only if  $\nu_0(E')$  is small. This shows (see Fig. 1) that the quasilocal levels can be located to the left of the Landau levels. In this region we have<sup>[8,9]</sup>:

$$F_0 = -\pi \frac{\sqrt{2m^3/\omega_c}}{(2\pi\hbar)^2} \frac{1}{\gamma\Delta}.$$

It is seen from Fig. 1 that when  $U_0 > 0$  (attraction of the electron to the impurity) and for arbitrarily small  $U_0$ , there actually appear on the left of the Landau levels quasilocal levels which are roots of Eq. (1). Substituting  $F_0$  in (1), we get

$$E_1 = 1/2 \hbar \omega_c - \Delta_0,$$

$$E_{ql}^{(n)} = \hbar \omega_c (n + 1/2) - \Delta_0,$$

where  $\Delta_0 = (1/2) m f^2 \omega_c^2$ ,  $f = m U_0 / 2 \pi \hbar^2$  is the scattering length, and  $n = 1, 2, \dots$

The level  $E_1$  is local, since it is located in the forbidden energy region. It corresponds to the bound state of an electron moving in the field of an impurity atom. Its position was obtained by Skobov<sup>[8]</sup> and by Bychkov<sup>[9]</sup> by another method. The levels  $E_{ql}^{(n)}$  fall in the region where the spectral density of the ideal metal differs from zero, and have therefore a finite width. The quantum states with energies  $E_{ql}^{(n)}$  correspond to the motion of an electron near an impurity atom with rare transitions to one of the neighboring impurity atoms.

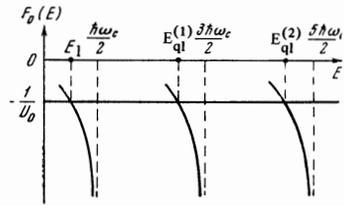


FIG. 1. Graphic solution of Eq. (1).

Confining ourselves to the quasiclassical case  $\hbar \omega_c \ll \zeta$ , we obtain for the width of the  $n$ -th quasilocal level the expression

$$\Gamma_n = 4 \Delta_0 (\Delta_0 / \hbar \omega_c)^{1/2} \sqrt{n}.$$

Owing to the small parameter  $\Delta_0 / \hbar \omega_c$  which enters in  $\Gamma_n$ , those values of  $n$ , at which  $\Gamma_n \ll \hbar \omega_c$ , turn out to be larger than unity. If  $\omega_c \tau_F \gg 1$ , where  $\tau_F = \hbar / \Gamma_F$  is the lifetime of the quasilocal state with the Fermi energy, then the levels located at the Fermi boundary (which play the principal role in the de Haas-van Alphen effect) have a width much smaller than the distance between them. This leads a very substantial increase of the spectral density near the quasilocal levels. The condition  $\omega_c \tau_F \gg 1$  denotes that  $H \ll H_c = c \hbar \lambda / e f^3$ , where  $\lambda$  is the wavelength of the electron with Fermi energy. If  $H > H_c$ , then the electrons are not captured by the impurity atoms. The quasilocal levels spread out. On the other hand, the magnetic field should not be weaker than a certain value, since  $\Delta_0 \sim H^2$  and in a weak field the quasilocal levels fall in the region where the Landau levels are smeared out. However, inasmuch as this smearing is of second order in the impurity concentration<sup>[11]</sup>, the minimal field will be so weak, that the picture of the oscillations will be destroyed by the thermal motion before the quasilocal levels fall in the zone where the Landau levels are smeared.

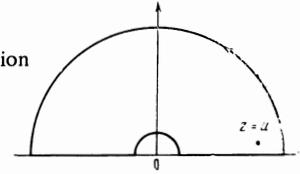
### 3. CONTRIBUTION OF QUASILOCAL LEVELS TO THE STATE DENSITY

The quasilocal levels give rise to an increment of the state density; this increment can be written, in the approximation linear in the impurity density, in the form<sup>[12]</sup>

$$\Delta v_{gl}(E) = \frac{N}{\pi} \sum_n \frac{\Gamma_n}{(E - E_{ql}^{(n)})^2 + \Gamma_n^2}, \quad (2)$$

where  $N$  is the volume concentration of the impurities. Using the Poisson formula for the calculation of this

FIG. 2. Contour for the calculation of the integral (3).



sum, we arrive at the integral

$$I_p(u) = \int_0^\infty dx \frac{\sqrt{x} \exp(2\pi i p x)}{(x-u)(x-u^*)}, \quad (3)$$

where

$$u = \left( \frac{E + \Delta_0}{\hbar \omega_c} \right)^{1/2} \left[ \left( \frac{E + \Delta_0}{\hbar \omega_c} \right)^{1/2} + 4i \left( \frac{\Delta_0}{\hbar \omega_c} \right)^{3/2} \right].$$

In order to find the approximate value of this integral in the case when  $\Delta_0 \ll \hbar \omega_c$ , we integrate the analytic branch of the function

$$\frac{\sqrt{z} \exp(2\pi i p z)}{(z-u)(z-u^*)}$$

along the contour shown in Fig. 2 (the cut is drawn along the positive real semi-axis). The residue of the integrand at the point  $z = u$  makes the main contribution to the oscillating part of the state density. Integration along the negative semi-axis gives a monotonic term, which contains the factor  $(\Delta_0 / \hbar \omega_c)^{3/2}$  and in our case can be discarded. Thus, in the case  $\omega_c \tau_F \gg 1$  the contribution of the quasilocal levels to the constant diamagnetism is negligibly small. This is due to the fact that when the quasilocal levels have a small width, the sum of the Lorentz peaks in formula (2) can be replaced by a sum of  $\delta$ -functions. Such a sum arises in the state density of the two-dimensional electron gas which, as is well known, has no constant diamagnetic susceptibility. The contribution due to the residue turns out to be

$$\Delta v(E) = 2 \frac{N}{\hbar \omega_c} \sum_{p=1}^\infty (-1)^p \exp \left[ -\frac{2\pi p}{\omega_c \tau(E)} \right] \cos 2\pi p \frac{E + \Delta_0}{\hbar \omega_c} \quad (4)$$

where

$$\tau(E) = \hbar^2 \omega_c / 4 \Delta_0^{3/2} E^{1/2}.$$

Thus, the oscillating addition to the state density has an exponential factor which recalls the Dingle factor, but now the "relaxation parameter" is the lifetime and not the free path time  $\tau$ . We note that  $\tau_F / \tau \sim N \rho^4 / f$ , where  $\rho = \sqrt{c \hbar / e H}$  is the minimum magnetic length. If we let  $\Delta_0$  approach zero in (4), we obtain a result that follows directly from (2) when all the  $\Gamma_n$  vanish.

### 4. OSCILLATING PART OF THE FREE ENERGY

The quasilocal levels, the distances of which to the corresponding Landau levels depend nonlinearly on the magnetic field, lead to an oscillating addition to the thermodynamic quantities; the frequency of this addition differs from the fundamental frequency<sup>[13]</sup>. Indeed, assuming that  $kT \ll \zeta$  and using (4), we obtain the contribution of the quasilocal levels to the oscillating part of the free energy

$$\Delta F = 2NkT \sum_{p=1}^\infty \frac{(-1)^{1+p}}{p} \exp \left( -\frac{2\pi p}{\omega_c \tau_F} \right) \times \frac{\cos \{2\pi p (\zeta + \Delta_0) / \hbar \omega_c\}}{\text{sh} (2\pi^2 p k T / \hbar \omega_c)}. \quad (5)$$

Thus, the quasilocal levels lead to a relative change of the oscillating part of the free energy, which is equal to  $N\rho^3$  in order of magnitude.

It is seen from formula (5), that the frequency of the oscillations due to the quasilocal levels is larger by a factor

$$\delta\Omega = \Omega_0\Delta_0/\zeta \quad (6)$$

than the frequency of the fundamental oscillations  $\Omega_0 = mc\zeta/e\hbar$ . Formula (6) holds for any form of the Fermi surface and the scattering potential<sup>[13]</sup>. Only  $\Delta_0$  and  $\Omega_0$  will, of course, be different.

The addition to the free energy (5), when added to the fundamental oscillations, causes beats on the plot of the magnetic moment against the reciprocal field<sup>[13]</sup>. Similar beats were observed also in the study of the influence of tellurium and selenium impurities on the de Haas-van Alphen effect in bismuth<sup>[14]</sup>. The beat frequency  $\delta\Omega$ , in accord with experiment, is proportional to the oscillation frequency. Brandt and Lyubutina<sup>[14]</sup> indicate that the beats arise only in the case of impurities of the donor type. This corresponds to the explanation presented here for the beats, since only in the case of donor-type impurities do quasilocal levels arise, since the potential of the impurity after its ionization corresponds to attraction (in our case  $U_0 > 0$ ). Naturally, the theory constructed here of the "beat effects" explains also one more experimental fact, namely the stability of the beats with respect to the technology of sample preparation. The beats are the consequence of the unique energy structure of a homogeneous metal with impurities, and not of its spatial inhomogeneity (as might be assumed).

In conclusion I am sincerely grateful to M. I. Kaganov for directing the work, and I. M. Lifshitz, A. M. Kosevich, and S. A. Gredeskul for valuable discussions.

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