On the anomalous *H* dependence of the amplitude of de Haas–van Alphen oscillations in $CeCu_2Si_2$

V. V. Val'kov[a)] and D. M. Dzebisashvili

Institute of Physics, Siberian Branch of the Russian Academy of Sciences, 660036 Krasnoyarsk, Russia; Krasnoyarsk State University, 660036 Krasnoyarsk, Russia

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The de Haas–van Alphen effect is studied in heavy-fermion antiferromagnets near the spin-flip transition is studied. It is shown that the strong increase occurring in the amplitude of oscillations near the spin-flip point in an increasing magnetic field, as observed experimentally in $CeCu_2Si_2$, can be explained by strong single-site correlations and magnetic ordering in the subsystem of localized electronic states. © *1998 American Institute of Physics.* [S0021-3640(98)00904-9]

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The electronic structure of heavy-fermion compounds remains one of the most intriguing objects in the physics of strongly correlated systems. The appearance of new scenarios of the low-temperature behavior of such substances has stimulated experimental studies directed toward clarifying the structure of their ground state. In this respect the de Haas–van Alphen (dHvA) effect, which makes it possible to determine directly the characteristics of Fermi quasiparticles, plays a special role.

The experimental observation[1–3] of sharp oscillations of the magnetization (which will be referred to below as dHvA oscillations) in a strong magnetic field in heavy-fermion systems has confirmed the existence in them of a Fermi surface and quasi-Fermi excitations at low temperatures. However, the appearance of a number of characteristic features in the oscillatory dependence of the magnetization makes it difficult to analyze the experimental data directly on the basis of the classical Lifshitz–Kosevich theory.[4] For example, the study of the dHvA effect in the heavy-fermion superconductor $CeCu_2Si_2$ has revealed two nontrivial features.[3] First, it was discovered that the period of the dHvA oscillations changes sharply upon transition through the spin-flip point in an increasing magnetic field. The second feature is that the dependence of the amplitude of the dHvA oscillations on the magnetic field H is anomalously strong near the spin-flip transition. In the conventional dHvA effect the amplitude increase becomes appreciable only after a large number of dHvA oscillations, whereas in $CeCu_2Si_2$ a substantial increase in amplitude was observed even for neighboring spikes of the magnetization. This behavior occurred both to the left and right of the spin-flip phase transition point.

$CeCu_2Si_2$ is a heavy-fermion antiferromagnetic superconductor. Long-range antiferromagnetic order is established at low temperatures ($T<T_N\cong 0.5$ K). The dHvA effect is observed in magnetic fields above the second critical field H_{c2}, where $CeCu_2Si_2$ was in

the normal state. The low Néel temperature indicates that the exchange-interaction integrals are relatively small. For this reason a quantizing magnetic field gives rise to strong canting of the antiferromagnetic sublattices, while in magnetic fields $H \sim 7$ T a phase transition occurs from the canted antiferromagnetic phase into the collinear ferromagnetic phase. Thus, the dHvA effect in $CeCu_2Si_2$ was observed under conditions of coexistence with a spin-flip phase transition. This fact made it possible to explain the sharp change in the period of dHvA oscillations at a transition through the spin-flip point in conducting antiferromagnets.[5,6] The key factor in the interpretation of the period change was to take into account the strong field dependence of the antiferromagnetic order parameter near the spin-flip point as well as the influence of this parameter on the electronic structure.

In the present letter we report the results of a theoretical study of the dHvA effect in an antiferromagnetic heavy-fermion compound near the spin-flip transition. The main result is an explanation of the anomalously strong increase observed experimentally in $CeCu_2Si_2$ of the dHvA amplitude in an increasing external magnetic field. It is shown that the physical mechanism responsible for this anomaly is the result of the combined effect of strong single-site correlations and long-range magnetic order.

To describe the anomalous increase in the amplitude of the oscillations in $CeCu_2Si_2$ we shall examine a periodic Anderson model that includes, besides the standard terms in the Hamiltonian for this model, interactions leading to the formation of long-range antiferromagnetic order. In this case the Hamiltonian of the system can be written in the form

$$H=\sum_{k\sigma}(\varepsilon_{k\sigma}-\mu)c^{+}_{k\sigma}c_{k\sigma}+\sum_{f\sigma}(E_{0\sigma}-\mu)X^{\sigma\sigma}_{f}+\frac{1}{\sqrt{N}}\sum_{fk\sigma}(V_k\exp(-ikR_f)c^{+}_{k\sigma}X^{0\sigma}_{f} + \text{h.c.})-\frac{1}{2}\sum_{ff'}\mathrm{I}_{ff'}(\mathbf{S}_f\cdot\mathbf{S}_{f'})-J\sum_{f}(\mathbf{S}_f\cdot\vec{\sigma}_f), \quad (1)$$

where the first term describes a system of itinerant electrons with energy $\varepsilon_{k\sigma}=\varepsilon_k - 2\sigma\mu_B H$, $\sigma=\pm 1/2$, and chemical potential μ in an external magnetic field H. The second term in the Hamiltonian describes the subsystem of localized electronic states in the atomic representation: $X^{\sigma\sigma}_f$ and $X^{0\sigma}_f$ are Hubbard operators, $E_{0\sigma}=E_0-\sigma g\mu_B H$, and E_0 is the energy of a localized level in the paramagnetic phase. Here, to simplify the exposition we have confined ourselves to the simplest case, where the basis of localized states includes states without electrons and states with one electron and projection of the spin angular momentum on the z axis equal to σ. The third term in Eq. (1) takes account of the hybridization processes between the collectivized and localized subsystems. The fourth term in the Hamiltonian represents the interaction that gives rise to antiferromagnetic order in the subsystem of localized electrons. $\mathbf{S}_f$ are spin angular momentum vector operators referring to site f in the crystal lattice. Finally, the last term in Eq. (1) takes account of $s-f$ exchange coupling between two groups of electrons.

We shall confine our analysis to magnetic fields in the range to the right of the spin-flip transition point. In this case ferromagnetic ordering of the spin moments is established in the subsystem of localized electronic states. To derive the self-consistency equations describing the low-temperature thermodynamics of the system we employed the Matsubara Green's functions constructed in terms of both conventional Fermi second-quantization operators and Hubbard operators. Besides the standard Fermi diagrammatic technique, the diagrammatic technique for Hubbard operators was also used. Without

going into computational details, we present the system of equations for the thermodynamic averages, leading to renormalizations of the parameters of the electronic structure in the ferromagnetic phase:

$$\langle X^{\sigma\sigma}\rangle = \exp(-(\tilde{E}_\sigma - \mu)/T)/Z + \frac{1}{N}\sum_k (\psi_{k\sigma} - 2\sigma\phi_k), \tag{2}$$

where

$$Z = 1 + \exp(-(\tilde{E}_\uparrow - \mu)/T) + \exp(-(\tilde{E}_\downarrow - \mu)/T),$$

$$\psi_{k\sigma} = \frac{E^+_{k\sigma} - \tilde{\varepsilon}_{k\sigma}}{2\nu_{k\sigma}} f\left(\frac{E^+_{k\sigma} - \mu}{T}\right) + \frac{\tilde{\varepsilon}_{k\sigma} - E^-_{k\sigma}}{2\nu_{k\sigma}} f\left(\frac{E^-_{k\sigma} - \mu}{T}\right) - f\left(\frac{\tilde{E}_\sigma - \mu}{T}\right),$$

$$\phi_k = n_k - \left(\exp\left(\frac{\overline{H}}{T}\right) - 1\right)^{-1}, \quad n_k = (\exp(\omega_k/T) - 1)^{-1}, \quad f(x) = (\exp(x) + 1)^{-1}.$$

The renormalized energy parameters appearing in these expressions have the form $\tilde{E}_\sigma = E_0 - \sigma\overline{H}$, $\tilde{\varepsilon}_{k\sigma} = \varepsilon_{k\sigma} - \sigma JR$, and $\overline{H} = g\mu_B H + I_0 R + J\langle\sigma^z\rangle$. The mixon spectrum associated with the hybridization interaction is described by the formulas

$$E^\pm_{k\sigma} = (\tilde{E}_\sigma + \tilde{\varepsilon}_{k\sigma})/2 \pm \nu_{k\sigma}, \quad \nu_{k\sigma} = ((\tilde{E}_\sigma - \tilde{\varepsilon}_{k\sigma})^2/4 + K_\sigma V_k^2)^{1/2}.$$

For what follows it is important that strong single-site correlations renormalize the effective hybridization interaction constant. Formally, this appears as the factor $K_\sigma = \langle X^{\sigma\sigma}\rangle + \langle X^{00}\rangle$ in the radicand in the expression for $\nu_{k\sigma}$. This renormalization is strongest in the saturated ferromagnetic state, where $K_\uparrow = 1$ while $K_\downarrow = 0$. In this case the hybridization channel remains completely open for electrons with spin moment oriented in the direction of the field, while for electrons with the opposite spin polarization the hybridization processes are completely suppressed. However, since hybridization mixing strongly influences the effective mass of a Fermi quasiparticle, the above-noted renormalization of the hybridization constant strongly influences the amplitude of the dHvA effect. Specifically, a change in the degree of ordering in the subsystem of localized electronic states will affect the dHvA amplitude.

To calculate the dHvA effect in a right-hand neighborhood of the spin-flip transition under conditions of hybridization mixing of itinerant and localized electronic states we shall employ the technique of summing over the Matsubara frequencies with the electron propagators represented in the form of contour integrals in the complex region.[7] This technique for magnetically ordered semiconductor systems was used in Ref. 8. In our case the oscillatory part of the magnetization of itinerant electrons can be written in the form

$$M_\sim = -\sum_\sigma \sum_{k=1}^{\infty} \sum_{\omega_n > 0} \frac{(-1)^k}{\sqrt{k}} A_{k\sigma}(\omega_n) \sin\left(2\pi k \frac{\tilde{\mu}_{n\sigma}}{\hbar\omega_c} + \varphi_\sigma\right), \tag{3}$$

where the ''partial'' amplitudes are determined by the expression

$$A_{k\sigma}(\omega_n) = \left(\frac{TVe\tilde{\mu}_{n\sigma}}{\pi\hbar^2 c}\right)\left(\frac{m_\parallel}{\hbar\omega_c}\right)^{1/2} \exp\left(-\frac{2\pi k\omega_n\alpha_{n\sigma}}{\hbar\omega_c}\right), \quad \alpha_{n\sigma} = 1 + \Gamma_{n\sigma},$$

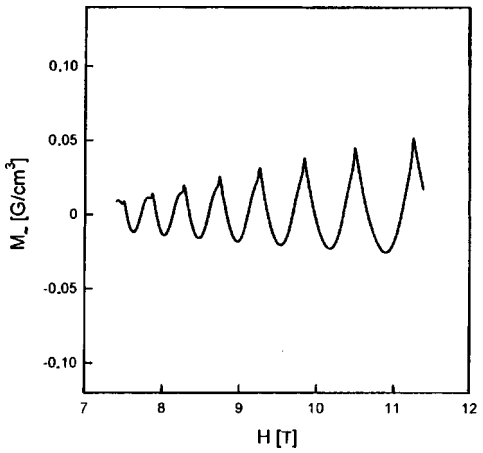

FIG. 1. Oscillatory part of the magnetization in a right-hand neighborhood of the spin-flip transition in a heavy-fermion antiferromagnet.

$$\Gamma_{n\sigma}=K_{\sigma}|v|^2\{\omega_n^2+(\widetilde{E}_{\sigma}-\mu)^2\}^{-1}, \quad \widetilde{\mu}_{n\sigma}=\mu+\sigma JR+(\widetilde{E}_{\sigma}-\mu)\Gamma_{n\sigma}. \tag{4}$$

The results obtained by solving the self-consistent equations for the averages $\langle X^{\sigma\sigma}\rangle$, the chemical potential μ, and $\langle\sigma^z\rangle$ numerically enabled us to analyze on the basis of Eqs. (3) and (4) the dHvA effect in a right-hand neighborhood of the spin-flip transition. Figure 1 shows the oscillatory part of the magnetization of itinerant electrons calculated in this manner. The following values of the model parameters were used in the calculations: $J=0.5$ eV, $V=0.05$ eV, $T=0.1$ K, $m=0.1m_0$, $I_0=-8.1\times10^{-4}$ eV, and $E_0=0.3$ eV. The choice of values for some of the parameters was dictated, specifically, by experimental data on $CeCu_2Si_2$. Thus, for example, $H_c\cong 7$ T, while the period of the oscillations was matched by varying the parameters to the value in the experiment of Ref. 3. The position of the localized level was fixed so as to obtain a high electronic specific heat. The oscillations displayed in Fig. 1 are due to only one spin subband, corresponding to the characteristic electron magnetic moment being oriented antiparallel to the magnetic field. The amplitude of the magnetization oscillations due to the other spin subband is very small, and the corresponding contribution to the dHvA effect is absent. One can see that as the magnetic field increases, strong growth of the amplitude of the dHvA oscillations occurs with neighboring spikes in the magnetization differing in amplitude by a substantial amount.

The physical mechanism of the anomalous growth of the amplitude involves the following. At the point of the spin-flip transition the gap in the spin-wave excitations spectrum equals zero. For this reason thermal excitation of spin waves occurs relatively easily. This results in an appreciable decrease of the magnetization. As the magnetic field increases, the gap in the spin-wave excitation spectrum increases. Such an increase in the activation energy greatly decreases the density of thermally excited magnons. For this reason the magnetization tends toward its maximum value.

Magnetic ordering in the subsystem of localized electronic states lifts the degeneracy of these states in the direction of the spin moment. Since the energy splitting $\widetilde{E}_{\downarrow} - \widetilde{E}_{\uparrow}$ is much higher than the temperature, only levels corresponding to an orientation of the localized magnetic moment in a direction collinear with the magnetic field are occupied. This circumstance together with the presence of strong single-site electronic correlations determine the different intensities of the hybridization processes for itinerant and localized electrons. Thus, the hybridization scattering channel for electrons spin-polarized in the direction of the field remains open. For this reason, the electronic states of the corresponding spin subbands are strongly hybridized and have a large effective mass (the chemical potential lies near the value of $\widetilde{E}_{\uparrow}$). This results in a high electronic specific heat at low temperatures.

A different situation arises for electronic states with the opposite orientation of the spin moments. For these states the renormalization of the hybridization constant on account of Hubbard correlations becomes so large that substantial (in the limiting case complete) suppression of the hybridization scattering channel occurs. Electronic states with such (negative in what follows) polarization of the spin moment remain weakly hybridized. The amplitude of the dHvA effect for them remains high enough so that sharp oscillations of magnetization in an increasing magnetic field can be recorded experimentally. The thermal excitation of magnons gives rise to an increase in the average $\langle X^{\downarrow\downarrow} \rangle$, which in turn increases the effective hybridization constant for electrons with ''down'' spin. For this reason, as temperature increases, the amplitude of the dHvA oscillations decreases rapidly, as is observed experimentally in all heavy-fermion compounds. The picture is reversed if $\langle X^{\downarrow\downarrow} \rangle$ decreases for any reason. This can be achieved, for example, by increasing the external magnetic field. Such an effect of a magnetic field is especially effective near a spin-flip transition. Indeed, for a relatively small change in field H from $H = H_c$ to a value of H such that $H - H_c \gg T$ the activation energy of magnons increases to such an extent that there are no spin-wave excitations in the system and $\langle S^z \rangle$ approaches its maximum value, while the effective hybridization constant for electrons with negative spin polarization approaches zero. This is the reason for the anomalously rapid increase of the amplitude of dHvA oscillations.

As temperature increases, the above-discussed anomaly of the H dependence of the dHvA amplitude will become larger. This is due to the increasing number of thermally excited magnons. For this reason, as H increases from $H = H_c$ to $H - H_c \gg T$ the magnetization will change by a large amount. As a result, the relative change in amplitude of the oscillations will also become large.

In conclusion, we note that the above-examined mechanism of the anomalous H dependence of the amplitude of the oscillations will operate in all antiferromagnetic heavy-fermion compounds with low Néel temperatures. It is important only that the field H_c at which the spin-flip transition occurs lie in the range where the dHvA effect is observed. Significantly, investigation of this anomaly will yield, besides the standard information about the properties of a Fermi system, additional information about the magnetically ordered subsystem of localized states.

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[a)]e-mail: vvv@iph.krasnoyarsk.su

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