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Evolution of Spin and Field Dependences of the Effective Mass with Pressure in $CeIn_3$

M. Endo,[1] N. Kimura,[1] H. Aoki,[1] T. Terashima,[2] S. Uji,[2] T. Matsumoto,[2] and T. Ebihara[3]

[1]*Center for Low Temperature Science, Tohoku University, Aramaki aza Aoba, Sendai 980-8578, Japan*
[2]*National Institute for Materials Science, Tsukuba, Ibaraki 305-0047, Japan*
[3]*Department of Physics, Shizuoka University, Shizuoka 422-8529, Japan*

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We have studied the field and spin dependences of the effective masses in $CeIn_3$ as a function of pressure via the de Haas–van Alphen (dHvA) effect. The effective mass increases with the field at pressures up to about 10 kbar and then decreases with the field. The spin direction of the dominant dHvA oscillation is likely to be reversed across the same pressure. The dHvA frequency changes significantly at the pressure P_c for the antiferromagnetic quantum critical point and two neighboring pressures P_2 and P_4 below and above P_c. The spin and field dependences rapidly diminish across P_c and finally disappear above P_4. These observations are discussed in conjunction with relevant observations and theories.

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The strongly correlated f-electron systems exhibit various interesting phenomena under magnetic fields like the metamagnetic transition in $CeRu_2Si_2$. The f-electron nature seems to change from itinerant to localized across the metamagnetic transition field (H_m). The effective masses (EM's) of conduction electrons increase with the field up to H_m and then decrease with the field [1]. They start to depend on the orientation of spin near H_m and seem to depend strongly on it above H_m [2]. On the other hand, it is reported in some compounds that the EM's depend strongly on magnetic field and spin although the up and down spin Fermi surfaces are nearly the same [3–7]. These dependences as well as the metamagnetic transition reflect the competition among magnetic field and the magnetic interactions such as the Kondo effect and the RKKY interaction. However, experimentally as well as theoretically it is not clarified in what condition and how the EM depends on spin and field.

Pressure is a useful tool to change the relative strengths of magnetic interactions and therefore to study how the dependences change with the magnetic interactions. The following two pressure regions will be particularly interesting in this respect: the pressure above which the strength of the Kondo effect dominates that of the RKKY interaction and the critical pressure (P_c) for the quantum critical point (QCP) where magnetic ordering is suppressed. In the former case the competition and the resultant dependences will be investigated. In the latter case two competing scenarios are proposed. In one scenario the Kondo effect is strong enough to quench local moments and spin-density wave (SDW) is formed at low temperatures below P_c. In the other one the effective Kondo temperature decreases to zero upon approaching P_c from high pressure side and the composite fermion breaks up at P_c to give rise to local moments. It would be an interesting question to the competing scenarios why and how the dependences are observed.

$CeIn_3$ is a very suitable system for this study. It crystallizes in the high symmetry cubic structure of the $AuCu_3$. The Ce moments order antiferromagnetically at about 10 K. The previous transport [8] and nuclear quadrupole resonance (NQR) [9] studies report that the Kondo effect overcomes the RKKY interaction at pressures around 10–15 kbar. The QCP is reached by applying hydrostatic pressure about 26 kbar and superconductivity is observed at pressures around P_c [10].

The single crystals were grown by the In self-flux method. Hydrostatic pressures up to 30.6 kbar were produced by clamped piston cylinder cells. We mostly used an equal mixture of n-propanol and i-propanol as a pressure transmitting medium. For a few pressures we also used an equal mixture of i-pentane and n-pentane to confirm that the results do not depend on the medium. The pressures at low temperatures were determined by the resistivity of manganin wire which was calibrated against the superconducting transition of Sn. The dHvA oscillations were detected in a ^{3}He cryostat under pressure and in a dilution refrigerator at ambient pressure using the field modulation technique [11,12]. At ambient pressure we have observed several new frequency branches in addition to those reported previously [13]. With fields parallel to [001], the frequencies (EM's) of the new branches are 2460 T (22 m_0), 770 T (28 m_0), 740 T (37 m_0), and 520 T (11 m_0). A new frequency of 2920 T with the largest mass of 53 m_0 is observed in the vicinity of the [001] direction but not along [001]. The details of the angle resolved study will be reported in a separate paper. As described below, strong field and spin dependences are observed for [001]. We chose this direction for pressure study. de Haas–van Alphen (dHvA) oscillations with frequencies of 3210 T and 520 T can be observed with the pressure cell. Only the former frequency oscillation can be detected at all pressures. It is denoted by the symbol d and is attributed to a hole surface centered at the Γ point [13].

We will first demonstrate that the EM's of the d oscillation depend on magnetic field and spin. The upper panel of Fig. 1 shows the d oscillation at three different temperatures. At the lowest temperature of 30 mK there is a shallow minimum around 6.5 T which becomes less obvious at higher temperatures. The decrease of the signal amplitude at high field side arises from the experimental condition of the Bessel factor [11] and is not an intrinsic effect. This observation indicates that the d oscillation consists of two component oscillations whose frequencies are nearly equal but whose EM's are significantly different. It can be shown that the minimum or the beatlike behavior does not arise from a bi-crystal structure or a Fermi surface topology by examining the angular dependence of the oscillations [14].

We will use the following phenomenological formula to interpret the present observations. If we assume the physical quantities to depend on spin and field, the conventional Lifshitz-Kosevich (LK) formula for the dHvA fundamental frequency oscillation can be modified to give [2,11],

$$\tilde{M}(H,T) = A_\uparrow(H,T)\sin\left[\frac{2\pi F_\uparrow(H)}{H} + \xi_\uparrow(H) + \xi_0\right] + A_\downarrow(H,T)\sin\left[\frac{2\pi F_\downarrow(H)}{H} + \xi_\downarrow(H) + \xi_0\right]. \tag{1}$$

Here, A_σ and F_σ denote the amplitude and frequency of the oscillation from up ($\sigma = \uparrow$) or down ($\sigma = \downarrow$) spin electrons, respectively. The spin and field dependent phase in Eq. (1) is given by $\xi_\sigma = \mp\pi g m^*{}_\sigma(H)/(2\ \mathrm{m_0})$. Here, $m^*{}_\sigma(H)$ denotes the effective mass, g denotes the g factor which is here assumed to be independent of spin and field, and $-$ and $+$ correspond to $\uparrow$ and $\downarrow$, respectively. The frequency can be also assumed to be independent of spin and field, because no splitting of the frequency is observed in the fast Fourier transform (FFT) analysis over a long field range. Equation (1) can be also expressed in the following form which is suitable when $A_\uparrow \geq A_\downarrow$.

$$\tilde{M}(H,T) = [A_\uparrow^2 + A_\downarrow^2 + 2A_\uparrow A_\downarrow\cos(\xi_\downarrow - \xi_\uparrow)]^{1/2} \times \sin\left[\frac{2\pi F(H)}{H} + \xi_\uparrow(H) + \xi_0 + \alpha(H,T)\right], \tag{2}$$

$$\alpha(H,T) = \tan^{-1}\frac{[\frac{A_\downarrow}{A_\uparrow}\sin(\xi_\downarrow - \xi_\uparrow)]}{[1 + \frac{A_\downarrow}{A_\uparrow}\cos(\xi_\downarrow - \xi_\uparrow)]}. \tag{3}$$

Here α denotes the phase shift of the observed oscillation from the phase of the dominant up spin oscillation. By exchanging up and down, we obtain an equivalent formula which is suitable when $A_\downarrow \geq A_\uparrow$. Equation (1) has been successfully applied to analyze the anomalous field dependence of EM [2,5,6].

The amplitude of the oscillation depends on the phase difference $\xi_\downarrow - \xi_\uparrow$. When the phase difference is $2n\pi((2n+1)\pi)$, the observed dHvA amplitude becomes $A_\uparrow + A_\downarrow$, i.e., maximum ($|A_\uparrow - A_\downarrow|$, i.e., minimum). When the EM's change monotonously with field, the waveform of the oscillation becomes a beat pattern making maxima and minima. At high temperatures the amplitude of the oscillation with the heavier mass is small, and therefore the minimum is less obvious. It is noted that the minima and maxima of the second harmonic frequency oscillation take place in a definite relation to those of the fundamental frequency oscillation. This relation has been used to confirm the assumption of field and spin dependences.

The lower panel of Fig. 1 shows the EM determined by the conventional method as a function of magnetic field, i.e., by neglecting the spin dependence. The shallow minimum of the EM around 6.5 T and large error bars are the artifacts that arise from the minimum of the oscillation amplitude. The EM increases with increasing field. It is found from measurements with a pulsed magnet that the EM becomes smaller around 20–35 T and then increases to a slightly larger value at 60 T than that at 15 T, implying that the beatlike behavior persists up to higher fields [15]. We have not observed any obvious metamagneticlike anomaly in the ac susceptibility in fields up to 17 T and to pressures up to 30.6 kbar.

The top panel of Fig. 2 shows the frequency of the d oscillation as a function of pressure. The relative frequency change $[F(P) - F(0)]/F(0)$ is shown for the d oscillation to show the changes more precisely. The X_1 and X_2 oscillations with EM's of about 10 $\mathrm{m_0}$ are observed at high pressures. Since they have no obvious corresponding frequencies at ambient pressure, the values of the frequencies are shown. Significant changes of the

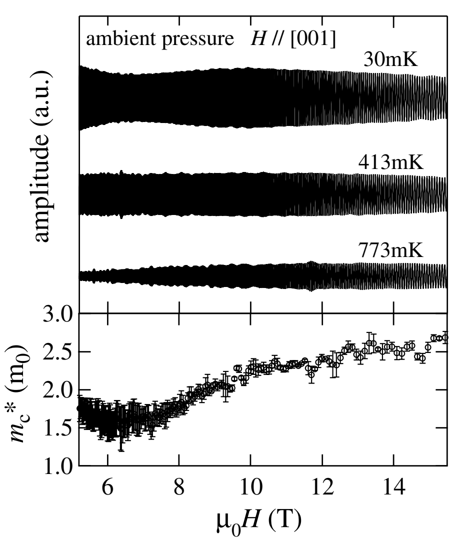

FIG. 1. The d oscillation at various temperatures (upper panel) and the effective mass (lower panel) as a function of magnetic field.

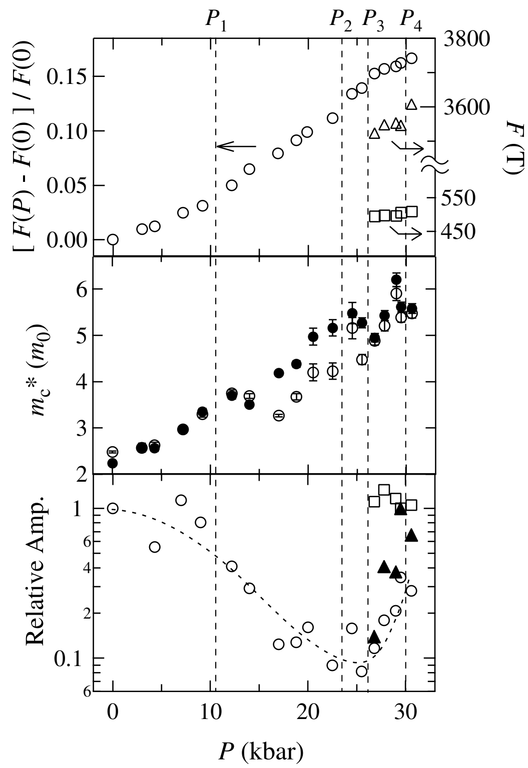

FIG. 2. Top panel: frequencies of d (open circle), X_1 (open triangle), and X_2 (open square). Middle panel: low (closed circle) and high field (open circle) EM's of d. Bottom panel: normalized signal amplitudes of d (open circle), X_1 (closed triangle), and X_2 (open square) as a function of pressure. The broken line is a guide to the eye.

frequencies are observed at pressures denoted by P_1, P_2, P_3, and P_4. The frequency of d increases with pressure at a rate of about 3.4×10^{-3}/kbar up to about P_1. Then it starts to increase more rapidly up to P_2. A rapid increase of the frequency is observed across P_2. The new frequency oscillations X_1 and X_2 start to be observed above P_3 which nearly corresponds to the reported P_c's. A change of dHvA frequencies has been reported for [111] at P_c [16]. The frequency of X_1 is found to change appreciably across P_4 although no significant changes are noted for d and X_2.

The middle panel shows the EM's of d as a function of pressure. The closed and open circles show the EM's determined from the FFT analysis between 9.8–11.6 T and 13.3–16.8 T, respectively. The long field range was used to make the artifact from the beatlike behavior smaller. The measured mass at the low fields approximately gives the lighter one because its signal amplitude is dominantly large there. However, the oscillation amplitude with the heavier mass is not negligible at the high fields. To estimate the effect from the spin dependent mass on the field dependence, we examined the waveforms and the EM's of the fundamental and second harmonic frequency oscillations as a function of field at each pressure. It has been found that the high field mass does not exactly correspond to the lighter mass, but that the field dependence in Fig. 2 shows semiquantitatively that of the lighter mass except for a few pressures near 10 kbar like 14 kbar.

The low field EM increases with pressure and seems to have a shoulder around P_1, and again a shoulder or a maximum around P_2, and finally another maximum just below P_4. It is noted that the largest maximum of the EM does not reside around the QCP. The field dependence of the EM also changes with pressure. It almost disappears around P_1 and then the EM starts to decrease with field above P_1. Although the EM decreases with field between P_3 and P_4, the field dependence becomes less obvious again. At the highest pressure beyond P_4, the field dependence almost disappears. The present pressure dependence of the EM is consistent with the behavior of the prefactor A of T_2 term of resistivity [8]. The value of A decreases largely with field below the QCP but is almost constant above the QCP. The observation that the largest maximum of the EM moves to a higher pressure than P_c is consistent with the difference of the field dependence across P_c. The coincidence between the field dependences of the lighter mass and the value of A indicates that the heavier mass is likely to have a similar field dependence to that of the lighter mass.

The bottom panel shows the FFT amplitudes of d normalized to that of ambient pressure. Also shown are the amplitudes of X_1 and X_2 normalized to those at 29.5 kbar. Although the data points scatter due to the beatlike behavior as well as experimental error, it is obvious that the minimum of the amplitude of d is somewhere around P_3, but not around 29 kbar where the EM becomes maximum and consequently the amplitude is expected to be minimum. The amplitude of X_1 seems to increase rapidly with pressure from P_3, whereas X_2 has a sizable amplitude from P_3 which changes moderately with pressure.

Figure 3 shows the dHvA oscillations as a function of inverse field at 0, 16.9, and 30.6 kbar, respectively. At 0 and some pressures like 16.9 kbar between P_1 and P_2, there are obvious minima in the fundamental or second harmonic frequency oscillations indicating that the EM's depend on spin. The shallow minimum indicates that the amplitudes or the EM's of up and down spin oscillations are considerably different. A simulation based on Eq. (1) gives an estimate that the difference in the EM's is about a factor of 2 both for ambient pressure and 16.9 kbar.

As the field dependence of the EM is reversed across P_1, the spin orientation of the dominant oscillation is likely to change together. The insets in Figs. 3(a) and 3(b) show the oscillations at higher and lower field sides of the minimum at three different temperatures. The peak positions or the phases shift towards the minimum position with increasing temperature. Equations (2) and (3) tell us that if the EM's of the up and down spin electrons are different, the ratio $A_\downarrow/A_\uparrow$ changes with temperature, and consequently the phase of the oscillation changes

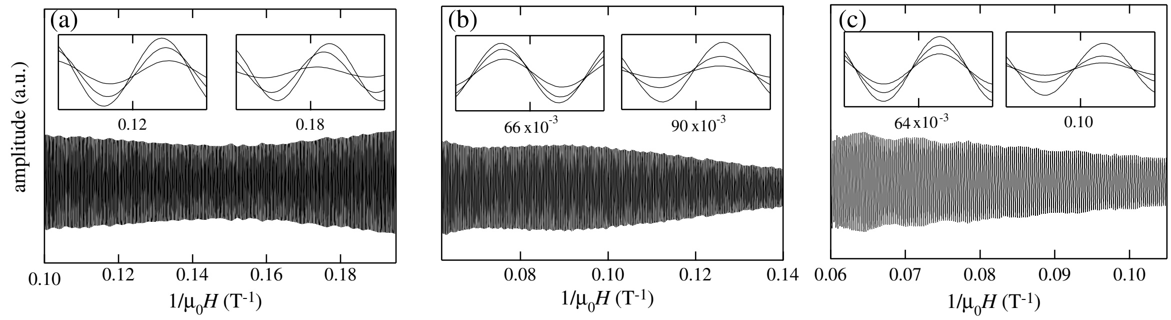

FIG. 3. The dHvA oscillations observed at (a) 0 kbar and 30 mK, (b) 16.9 kbar and 0.3 K, and (c) 30.6 kbar and 0.3 K. The insets show the dHvA waveforms at three different temperatures. The short beat of the oscillations in (c) arises from the interference between d and X_1.

with temperature. Particularly, the peak shifts towards the minimum with increasing temperature when the dominant oscillation is the down (up) spin state and the EM increases (decreases) with field [2]. The present observation implies that the spin direction of the dominant oscillation is opposite for 0 and 16.9 kbar because the field dependence of the EM is opposite. On the other hand, Fig. 3(c) indicates that the phase of the oscillation does not change at any fields observed. Since the EM does not depend on field and the spin splitting behavior is observed in the harmonic content [4], this observation indicates that the ratio $A_\downarrow/A_\uparrow$ is temperature independent; i.e., the EM's are the same.

In summary, in the both pressure ranges below and above P_1 the EM depends on field and spin but the dependences are likely to be reversed. The dependences have been theoretically discussed by three groups [17–19] to predict that the EM for one spin direction decreases with field. The behavior of the lighter EM above P_1 is consistent with the predictions. However, the heavier EM is likely to decrease with field too in the present experiments. This is consistent with the observations in $PrPb_3$ [5], $CePd_2Si_2$ [6], and CeTe [7], but is inconsistent with the predictions. The reversed dependences may arise from the interchange of the dominant magnetic interaction across P_1.

We have observed a complicated electronic structure change around the QCP. The electronic structure changes at P_2 and P_4, in addition to P_3 which is assumed to be P_c. The large frequency change across P_2 probably corresponds to the rapid drop of T_N at about 24.5 kbar reported by the NQR measurements [20]. The electronic structure change across P_3 could be attributed to the change of magnetic structure from antiferromagnetic to paramagnetic. However, the appreciable change of the X_1 frequency across P_4 might imply that another change in the magnetic structure takes place. The spin and field dependences diminishes rapidly across P_3 and finally disappears above P_3, giving the largest maximum of the EM not at P_3, but near P_4. In accord with this observation, the signal amplitude of d is strongly damped near P_3, implying that a magnetic fluctuation responsible for the mass enhancement may be also responsible for the damping. If the observation that the effect of the fluctuation is selectively effective on particular orbits like X_1 is not accidental, the fluctuation could be related with a particular Q vector of magnetic order. On the other hand, the large effective mass above P_4 may raise a question whether the mass enhancement can be solely attributed to the magnetic fluctuation near P_3 that is suppressed by field.

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