High-field de Haas–van Alphen measurements in Pd

J. J. Vuillemin
Department of Physics, University of Arizona, Tucson, Arizona 85721

N. Harrison
National High Magnetic Field Laboratory, Los Alamos National Laboratory, LANL, Los Alamos, New Mexico 87545

R. G. Goodrich
Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803

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The de Haas–van Alphen (dHvA) effect in 99.999% pure palladium has been observed in pulsed fields up to 60 T directed along [100]. We report a dHvA frequency of 73.5 kT with a cyclotron effective mass $=12.5m_0$. Such a frequency is not reported previously but is predicted by band theory for the open hole sheet of the Fermi surface. We also observe strong harmonic content near 50 T for the electron sheet and this is interpreted in terms of a field-dependent g factor. [S0163-1829(99)05707-0]

Among the transition metals, Pd continues to attract interest for several reasons. It has a relatively high density of electronic states, but is not superconducting or magnetic.[1] Its susceptibility is strongly enhanced and its response to magnetic impurities suggests that pure Pd is bordering on ferromagnetism.[2] Spin fluctuations are thought to contribute to the characteristic properties of this metal but a quantitative understanding of this contribution is hindered by the lack of data in high magnetic fields. Previous de Haas–van Alphen (dHvA) studies in Pd are extensive, but they do not cover the field range currently available and they do not directly determine all of the major Fermi-surface (FS) parameters predicted by theory.[3,4]

In this paper, we describe dHvA measurements in Pd that extend the field to 60 T for the [100] direction. Our results yield important information about the FS parameters. A new dHvA frequency is reported that is much larger than any of the previously observed fundamental frequencies. This oscillation is attributed to the open hole sheet of the FS. We also have observed strong harmonic content near 50 T for the electron sheet. This suggests that the g factor is field dependent even though there are less than 0.001% magnetic impurities present.

The dHvA oscillations in Pd were observed over the temperature range 1.6–2.0 K in pulsed magnetic fields supplied by the National High Magnetic Field Laboratory, Los Alamos. The specimen raw material came from Johnson-Matthey Co. in the form of 99.999% Pd wire, 1 mm in diameter. An oriented single-crystal rod was grown from this material by the rf floating zone technique starting from a [100] seed. The rod was cut to a length of 3 mm by spark erosion and etched until the diameter was 0.2 mm. This specimen was immersed in superfluid He to avoid eddy-current heating during the 7-ms rise time of the applied field. The data were collected at the rate of 500 kHz. The oscillatory susceptibility was measured from the voltage induced in a balanced pickup coil arrangement coupled to the specimen and the field was determined by integrating the induced emf in a nearby second coil. The field was calibrated using the known dHvA frequencies of Cu and other materials.

The frequency and amplitude for many orbits in Pd were determined from the complex oscillatory data by performing a discrete Fourier transform (DFT) of the signal versus reciprocal field. The cyclotron effective mass was obtained from the temperature variation of the amplitude and the cross sectional area A (in a.u.) of the FS was found from the well-known relation

$$A=(2\pi e/\hbar c)F=2.673\times10^{-5}F \quad (1)$$

where F is the dHvA frequency (in T).[5]

A previously unobserved dHvA frequency is clearly seen in our Pd data with the field above 55 T. A typical DFT over the frequency range of interest is plotted in Fig. 1 showing the new frequency at 73.5 kT. All of the known fundamental

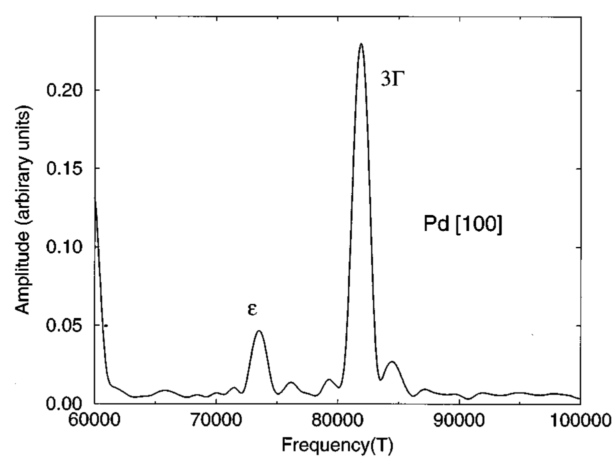

FIG. 1. A portion of the Fourier spectrum in Pd with the field directed along [100] showing the ε dHvA frequency at 73.5 kT. The field is in the range 55–57 T and the temperature is 1.6 K. The large peak at 83.6 kT is the third harmonic of the Γ-centered electron sheet.

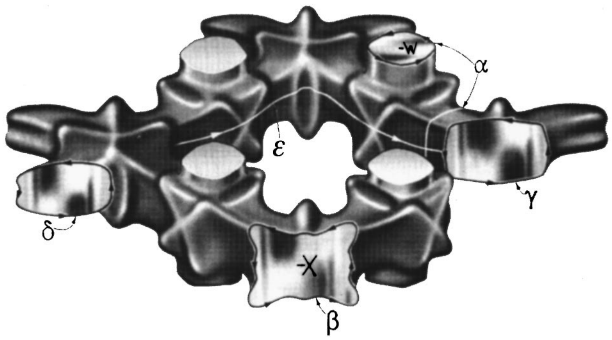

FIG. 2. The open hole sheet of the FS in Pd and the various extremal orbits it supports. The arms are directed along [100]. After Dye *et al.* (Ref. 3).

frequencies in Pd are much less than this and none of their harmonics or reasonable combination frequencies match this value. Such a frequency is expected, however, for the relatively large ε orbit on the open hole sheet of the Pd FS shown in Fig. 2. Table I shows the FS cross-sectional area and effective mass of the new oscillation along with the values predicted for the ε orbit by a Korringa-Kohn-Rostoker (KKR) phase shift parameterization of the FS.[3] The agreement of the predicted area with the observed value is remarkably good. The predicted cyclotron effective mass is also reasonably close to the observed value. We conclude, therefore, that the dHvA frequency we observe at 73.5 kT is due to the ε orbit on the open hole sheet of the FS in Pd.

The existence of this orbit provides evidence to confirm that the open hole sheet is indeed topologically equivalent to a network of ⟨100⟩ cylinders intersecting at the X point in the Brillouin zone. The unenhanced effective mass of the ε orbit is estimated from band theory to be $7.3m_0$.[1] Our effective-mass measurement of $12.5m_0$ yields a total enhancement factor of 1.7 for this orbit including electron-phonon and electron-electron contributions. This factor is somewhat higher than that for other orbits, which have a nearly constant enhancement of 1.3–1.5.[1] The calculations of Pinski and Butler give $\lambda_{e\text{-ph}}=0.4-0.5$ and this suggests that $\lambda_{e-e}=0.2-0.3$ for the ε orbit in Pd.[6]

We also have observed unusual harmonic content for the electron sheet of the FS in Pd above 40 T. This is interpreted in terms of a field-dependent g factor in the spin splitting of the Landau levels. In the Lifshitz-Kosevich model, the amplitude of the pth dHvA harmonic is proportional to $\cos(p\pi g m^*/2)$, where g is the conduction-electron g factor, and m^* the cyclotron effective mass without electron-phonon enhancement in units of the free-electron mass.[5] The first harmonic amplitude of the [100] electron oscillation in our specimen passes through a node near 50 T. This requires

TABLE I. The ε orbit on the open hole sheet of the FS in Pd.

Source	Area (a.u.)	Cyclotron effective mass[a]
This work	1.96	12.5
KKR fit[b]	1.97	11.5[c]

[a]In units of the free-electron mass.
[b]Reference 3.
[c]Includes many-body enhancement.

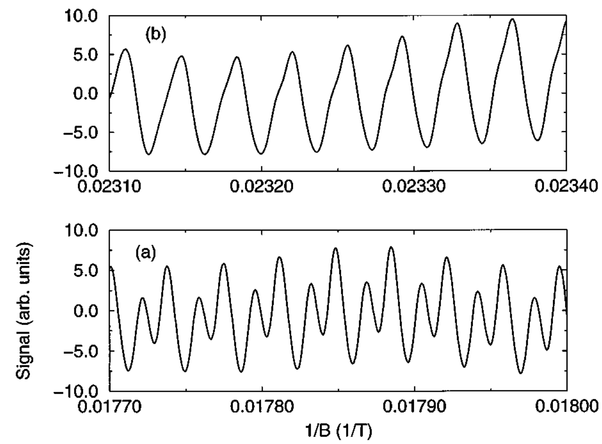

FIG. 3. The dHvA oscillations due to the electron sheet in Pd at the temperature of 1.6 K with the field directed along [100]. (a) The second harmonic is dominant above 50 T but is rather weak at slightly lower fields (b).

$g(50)m^*=2n+1$, where $g(B)$ is the g factor at the field B in T and n is an integer. Low-field measurements indicate that $g(0)m^*\approx 2n+0.5$ so the change $\Delta(gm^*)\approx 0.5$ between 0 and 50 T corresponding to a relative change in gm^* of about 0.03 since n is estimated to be 8 from the magnetization.[7,8] We attribute this change to the g factor because there is no evidence for a field dependence in m^*.[4]

The change in the spin splitting of the Landau levels is also evident in the harmonic content of the electron oscillations. Figure 3(a) shows the strong second harmonic content above 50 T that is absent at slightly lower fields [Fig. 3(b)]. The ratio $A(2)/A(1)$ of the second to first harmonic dHvA amplitude is shown in Fig. 4(a) as a function of reciprocal field. This ratio yields the magnitude of the change in gm^* shown in Fig. 4(b) and the wave form suggests that g increases with the field.[5] This change is consistent with magnetization measurements in pure Pd to 35 T, which shows a

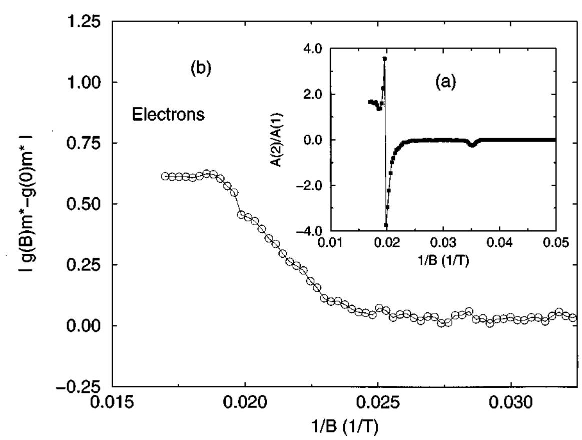

FIG. 4. (a) The ratio $A(2)/A(1)$ of the second harmonic amplitude to the first versus reciprocal field for the [100] electron oscillation in Pd at the temperature of 1.6 K. (b) The magnitude of the corresponding change in the product $g(B)m^*$ versus reciprocal field where $g(B)$ and m^* are defined in the text.

positive curvature.[9] The change we observe in gm^* includes contributions from spin fluctuations and field-dependent changes in the band structure.

The g factor in Pd is also known to be sensitive to magnetic impurities.[9,10] It is conceivable that the very small magnetic impurity content in our sample is responsible for the change in the g factor we observe near 50 T. This is unlikely because measurements of g in dilute alloys of Pd (Fe) at lower fields predict that such a change would require a substantially higher magnetic impurity content ($\sim$100 ppm).[10]

In summary, our high-field amplitude measurements suggest that the [100] electron g factor changes by about 3% over the field range 0–50 T in Pd with less than about 10 ppm magnetic impurities. It is not clear whether this change is due to a field-dependent band structure or a dynamical response (e.g., spin fluctuations). Our dHvA frequency measurements yield the first objective determination of the largest [100] extremal cross section of the Pd FS, the area enclosed by the ε orbit. This area measurement provides a stringent test for the predictive ability of single-electron band-structure calculations. The corresponding effective-mass measurement is useful for estimating the corrections for the rather strong many-body effects in this element. The close agreement between the KKR parameterized area of the ε orbit and experiment indicates that single-electron band calculations are capable of predicting the FS cross sections in Pd rather well.

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[1] W. Joss and G. W. Crabtree, Phys. Rev. B **30**, 5646 (1984).
[2] G. G. Low and T. M. Holden, Proc. Phys. Soc. **89**, 119 (1966).
[3] D. H. Dye, S. A. Campbell, J. B. Ketterson, N. B. Sandesara, and J. J. Vuillemin, Phys. Rev. B **23**, 462 (1981).
[4] W. Joss, L. N. Hall, G. W. Crabtree, and J. J. Vuillemin, Phys. Rev. B **30**, 5637 (1984).
[5] See, for example, D. Shoenberg, *Magnetic Oscillations in Metals* (Cambridge University Press, Cambridge, 1984).
[6] F. J. Pinski and W. J. Butler, Phys. Rev. B **16**, 6010 (1979).
[7] H. Ohlsén, P. Gustafsson, L. Nordborg, and S. P. Hörnfeldt, Phys. Rev. B **29**, 3022 (1984).
[8] J. Hjelm, Int. J. Mod. Phys. B **7**, 271 (1993).
[9] A. Tal, L. W. Roeland, M. Springford, P. Wise, and R. G. Jordan, J. Phys. F **16**, 893 (1985).
[10] P. T. Coleridge, P. Wise, P. H. P. Reinders, and M. Springford, J. Phys. F **16**, 2027 (1986).