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Title:

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$\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$: a quasicrystal showing the de Haas-van Alphen effect

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ABSTRACT

We have measured the de Haas-van Alphen effect in the icosahedral quasicrystal $\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$. We have found two well-defined frequencies with the magnetic field parallel to a five-fold axis, and two different ones with the field parallel to a two-fold axis. On increasing the temperature, the amplitude of the oscillations substantially decreased, suggesting that the carriers have large masses.

Quasicrystals¹ have rotational symmetries prohibited by real-space translational invariance. Therefore, Bloch's theorem does not apply to these materials. Measurements of the resistivity and specific heat of quasicrystals show results similar to results for disordered materials.^{2,3} On the other hand, quasicrystals have long-range order, because their X-ray diffraction spots are as sharp as those of any crystal. An $\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$ quasicrystal has even shown the Borrmann effect⁴, the anomalous transmission of X-rays due to multiple scattering effects. This property had previously only been observed in crystals with high structural perfection. Under these circumstances, what is the meaning of the Fermi surface of a quasicrystal? Can we probe it experimentally? We approach these questions by studying the de Haas-van Alphen effect, the oscillatory component of the magnetization periodic in the inverse of the magnetic field.⁵

We have measured the de Haas-van Alphen (dHvA) effect in an $\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$ quasicrystal. The effect measures the extremal Fermi-surface cross section normal to the direction of the magnetic field. It provides information about the size and topology of the Fermi surface of a metal, and on the effective mass and scattering rates of the charge carriers. Requirements for the observation of the dHvA effect are: low temperature, high magnetic field, and high sample quality. An additional difficulty for quasicrystals is their high resistivity. This circumstance is unfavorable for the observation of the dHvA effect. Furthermore, the dense presence of Bragg planes everywhere fractures the Fermi surface into a multiplicity of tiny pieces. Magnetic

breakdown in high fields provides a mechanism for reconnecting the fractured Fermi surface, yielding a continuous cyclotron orbit and an observable dHvA effect.

To qualify as the dHvA effect, an oscillatory signal has to satisfy three criteria: periodicity in the inverse magnetic field, decreasing amplitude with either increasing temperature or decreasing magnetic field, and reproducibility. Moreover, the signal should vanish for an empty sample holder.

$\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$ is a thermodynamically stable, face-centered icosahedral quasicrystal (I^* in the notation of Rokhsar *et al.*⁶). Our samples were cut from the same quasicrystal in which Kycia *et al.*⁴ observed the Borrmann effect. This observation confirmed the high degree of structural perfection of the samples. X-ray diffraction measurements revealed that all diffraction spots were consistent with those of a face-centered icosahedral quasicrystalline structure, implying the absence of second phases or inclusions of, *e.g.*, small regions of pure aluminum or approximants.

We have carried out the experiments at the Pulsed Field Facility of the National High Magnetic Field Laboratory in Los Alamos. We used a 50 T pulsed magnet and a ^3He bath cryostat (temperatures down to 400 mK) inside the bore of the magnet. The ^3He bath temperature was measured using a silicon diode and by monitoring the vapor pressure above the liquid.

We used two quasicrystal samples. One sample ($3\text{ mm} \times 0.75\text{ mm} \times 0.50\text{ mm}$) had its long direction aligned along a five-fold axis. The other sample was a compact assembly of four smaller samples, $3\text{ mm} \times 0.25\text{ mm} \times 0.25\text{ mm}$ each, with the long directions along a two-fold axis. The four individual samples were electrically insulated from each other, with all two-fold axes parallel. This construction reduced eddy-current heating due to the magnetic field pulse, a concern in this type of experiment. The magnetic field was applied parallel to the long direction of the samples.

A balanced pair of pickup coils measured the dHvA signal inductively. One coil contained the sample; the second coil remained empty. An electronic bridge circuit detected the differential signal. The areas of the two coils were equal to within one part in 300, so the contributions of the applied field in each coil very nearly canceled; passive electronics in the bridge circuit decreased the residual difference to less than one part in 10^5 . The magnetic field was measured by integrating the voltage induced in a third pickup coil placed around the sample.

As a check of our measurement system, we have performed separate experiments on a $\text{Au}[111]$ single crystal, observing the expected neck and belly frequencies, as well as harmonics up to eighth order. This also served to calibrate the field pickup coil to an accuracy of better than 0.1 % over the entire field range.

High quality quasicrystals in the Al-Pd-Mn system exist for a range of compositions.⁷ We assume, however, that a perfect quasicrystal should exist only at an isolated, unique stoichiometry. A similar view is held for quasicrystals in the Al-Cu-Fe system.⁸ Measurements of the dHvA effect may provide a way of picking out that unique stoichiometry.

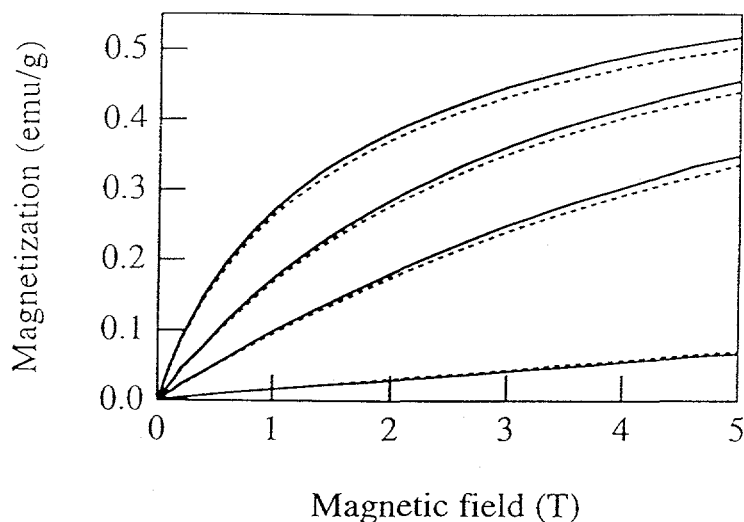


Fig. 1. Magnetization of the quasicrystal, measured using a SQUID magnetometer. The magnetic field is applied parallel to a two-fold axis (solid lines) or to a five-fold axis (broken lines). Temperatures are 2 K (top curve), 5 K, 10 K and 50 K (bottom curve).

Slight differences in composition among the various Al-Pd-Mn quasicrystals result in different room temperature resistivities, magnetizations and temperature dependences of the resistivity. Our quasicrystals had a room temperature resistivity of $1250 \mu\Omega \text{ cm}$. The resistivity was nearly temperature independent between room and helium temperatures, decreasing by only 4%. This suggests that the dominant scattering mechanism in quasicrystals is not phonon scattering but rather scattering due to the quasicrystalline structure of the material itself.

In Fig. 1 we show the magnetization of our samples for temperatures between 2 K and 50 K, and for magnetic fields up to 5 T. The measurements were carried out using a SQUID magnetometer. The magnetization of the quasicrystal is small, saturating around 0.5 emu/g. The anisotropy of the magnetization between the two field directions is 3%. Surprisingly, there is no evidence in the data of any magnetic phase transition in steady fields below 5 T, nor was such evidence for ordering observed in the 50 T pulsed field experiments themselves. This suggests that $\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$ is a weak paramagnet in which the expected strong Mn moments are quenched or play only a modest role in fields up to 50 T and temperatures down to 400 mK.

In Fig. 2, we show the power spectral density of two signals obtained under identical conditions. For both spectra, the magnetic field was parallel to a five-fold axis, the temperature was 400 mK, and the magnetic field was between 50 T and 37 T. Note the high degree of agreement between the two spectra. The two main frequencies in both spectra have values of 0.96 kT and 3.55 kT. Not only are the values of the frequencies equal, but also the peak heights are identical within 2%. The degree of agreement between these data is evidence of the reproducibility of these experiments.

With increasing temperature, the amplitude of the oscillations decreased. Measurements at 470 mK showed oscillations whose amplitudes were reduced by at least

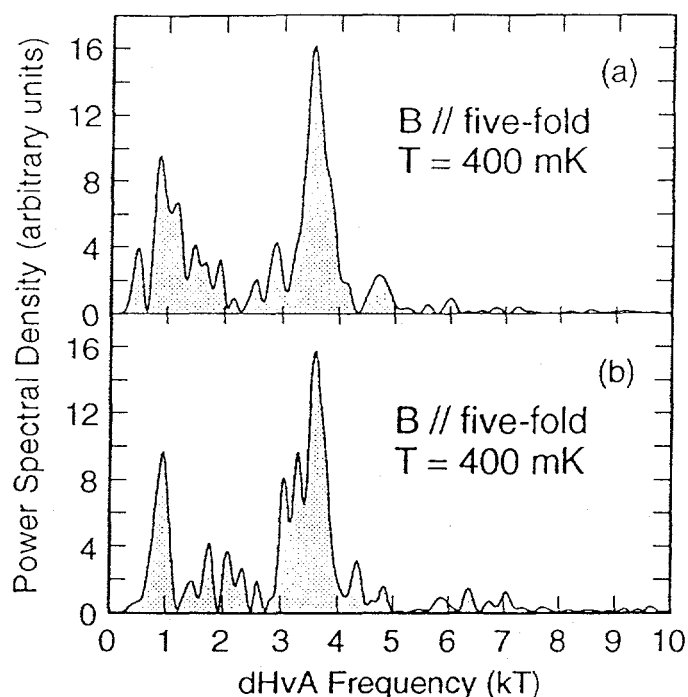


Fig. 2. Power spectral density of the dHvA oscillations observed in $\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$, with the magnetic field along a five-fold axis. In (a) and (b) results are shown of two separate measurements. In both measurements the sample temperature was 400 mK. Both spectra have peaks at frequency values of 0.96 kT and 3.55 kT. The height and position of the peaks are nearly identical; this shows the reproducibility of these experiments.

a factor 16. This large reduction for a small temperature increase indicates that the charge carriers in the quasicrystal have large effective masses. When we interpret this amplitude decrease within the Lifshitz-Kosevich theory of the dHvA effect,⁵ we find that the 3.55 kT orbit, having the larger amplitude, has a mass of about $37 m_e$, where m_e is the free electron mass.

At temperatures higher than 470 mK, no reproducible oscillations were observed with the magnetic field parallel to a five-fold axis.

Spectra of subsequent measurements without a quasicrystal sample in the coils did not show the same oscillatory signals as the low temperature data. Power spectral densities of empty coil measurements showed small peaks at several frequencies. None of these peaks coincided with 0.96 kT or 3.55 kT. The amplitude of the highest peak in each spectrum was smaller by a factor of at least 30 than the peaks shown in Fig. 2. We conclude that these power spectral densities are those of background noise due to vibrations, incomplete screening, or induced motion of the coils or magnet.

Next, we show the power spectral density with the field along the two-fold axis at an initial temperature of 450 mK in Fig. 3a and at 500 mK in Fig. 3b. The magnetic field again ranged between 50 T and 37 T. The two largest peaks in both spectra have frequencies of 0.36 kT and 1.21 kT, different from the values with the

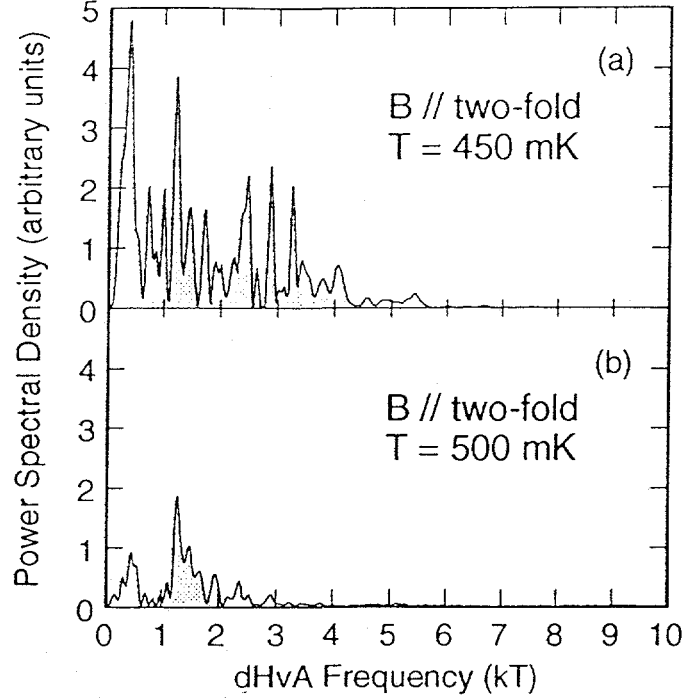


Fig. 3. Power spectral density of the dHvA oscillations observed in $\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$ with the magnetic field parallel to a two-fold axis. In (a) we shown data at a temperature of 450 mK. The temperature in (b) is 500 mK. Peaks occur in both spectra with frequencies of 0.36 kT and 1.21 kT, but with significantly different heights. The lowest temperature has the highest amplitude. As discussed in the text, the rapid decrease of the amplitude suggests that the charge carriers have large masses of about $25 m_e$.

field parallel to a five-fold axis. Peaks at the same frequencies are present at the higher temperature, but with much reduced amplitudes. From the reduction with temperature, we calculate that the charge carriers giving rise to the 1.21 kT orbit have effective masses of about $25 m_e$.

Table 1 summarizes these results.

We have compared the dHvA frequencies of the quasicrystal with those of Al, Pd, and Mn.⁹ We find that, with the possible exception of the 0.36 kT frequency, which is close to an Al frequency (the [100] γ monster orbit, with a frequency of 391.4 T), none of the observed $\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$ frequencies is coincident with any of the frequencies

Table 1: dHvA frequencies and masses for the quasicrystal. We have only included the masses for the largest amplitude orbits for each direction.

direction	F_1 (kT)	m_1^*/m_e	F_2 (kT)	m_2^*/m_e
five-fold	0.96	—	3.55	37
two-fold	0.36	—	1.21	25

of their elemental constituents.^a We therefore conclude that the frequencies shown in Figures 2 and 3, and summarized in Table 1, are representative of the quasicrystal $\text{Al}_{70}\text{Pd}_{21.5}\text{Mn}_{8.5}$ and are not due to second-phase inclusions.

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^aBinary line compound of these elements include MnAl_6 , MnAl_4 , MnAl_3 , Pd_2Al_3 , PdAl_3 , PdAl , and Pd_2Al . We have not found any dHvA results on these compounds in the literature, but we believe it is unlikely that one of these causes the dHvA effect we have observed.