Hysteresis in the de Haas-van Alphen Effect

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A hysteresis loop is observed for the first time in the de Haas-van Alphen (dHvA) effect of beryllium at low temperatures and quantizing magnetic field applied parallel to the hexagonal axis of the single crystal. The irreversible behavior of the magnetization occurs at the paramagnetic part of the dHvA period in conditions of Condon domain formation arising by strong enough dHvA amplitude. The resulting extremely nonlinear response to a very small modulation field offers the possibility to find in a simple way the Condon domain phase diagram. From a harmonic analysis, the shape and size of the hysteresis loop is constructed.

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It is well known that the irreversibility of the magnetization process by domain wall motion is due to energy barriers arising from a variety of defects inside magnetic material\textsuperscript{[1]}. The resulting hysteresis effects in usual magnetic substances, i.e. in substances where the atomic magnetic moments, due largely to the electron spin, are the reason of magnetism, have been investigated in detail in the past. However, in an applied magnetic field, the orbital motion of free electrons in metals leads also to magnetization. For Landau quantization\textsuperscript{[2]} of the electronic system, the oscillating magnetization known as the de Haas-van Alphen (dHvA) effect has been extensively studied in single crystals at low temperatures\textsuperscript{[3, 5]}.

There is no experimental data reporting hysteresis in the dHvA effect apart from the trivial case caused by eddy currents when the applied magnetic field is varied fast enough\textsuperscript{[4]}. If the dHvA amplitude becomes very large and comparable to the oscillation period, i.e. $\mu_0 \partial M/\partial B > 1$ for magnetization $M$ and magnetic induction $B$, a thermodynamic instability arises at the paramagnetic ($\chi = \partial M/\partial H > 0$ for susceptibility $\chi$ and applied magnetic field $H$) part of every dHvA period, according to the Pippard-Shoenberg concept of high magnetic interaction\textsuperscript{[3, 6]}. For long samples with the applied field parallel to the long axis (demagnetization factor $n \approx 0$) the instability is avoided by a jump of the magnetic induction $B$ between the stable states $B_1$ and $B_2$ at a critical field $H_c$. For plate-like samples perpendicular to $H$ ($n \approx 1$) $B = \mu_0 H$ is required so that the induction can not jump. In this case Condon domains arise with neighboring regions of respective inductions $B_1$ and $B_2$ in the applied field interval $B_1 < \mu_0 H < B_2$\textsuperscript{[6]}. For samples with $0 < n < 1$ this interval decreases proportionally to $n$. For samples of arbitrary shape there is an inhomogeneous state - Condon domain state (CDS) with the same phases $B_1$ and $B_2$.

After the discovery of Condon domains\textsuperscript{[6, 7]} hysteresis in the dHvA effect needs to be considered again. Indeed, the CDS consists of two phases of different induction values with a magnetization current in the domain walls. This needs usually extra energy. It was shown that the transition from the homogeneous state to the CDS is of first order\textsuperscript{[8]}. At this phase transition one could expect, in principle, all phenomena like irreversibility, supercooling and hysteresis that exist at first order phase transitions, e.g. the liquid - gas transition. Naturally, Condon discussed these problems in his first paper on domains\textsuperscript{[6]} concluding that neither supercooling nor hysteresis had been observed in all reported data. Since then, the above phenomena were discussed in several papers\textsuperscript{[3, 9, 10]}. Despite Condon domains themselves have been observed and investigated experimentally\textsuperscript{[11, 12, 13, 14]}, there is nevertheless up to now no experimental data proving hysteresis.

In this Letter we present the first experimental observation of hysteresis in beryllium single crystals. The hysteresis itself turns out to be very small. Therefore, several methods were used to prove the existence of hysteresis. First the hysteresis effect is measured directly by Hall probes in DC fields, then a standard AC method is used with various modulation levels, frequencies, and magnetic field ramp rates. Finally, the DC hysteresis loop is reconstructed by assembling several higher harmonics of the AC pickup voltage. Moreover, it is shown that the Condon domain phase diagram can be measured directly from the response to sufficiently small AC fields. Finally, Plummer’s\textsuperscript{[15]} strange and up to now only incompletely understood data is explained.

Beryllium is to our knowledge the best metal to investigate hysteresis effects related to Condon domain formation. First, due to its cigar-like Fermi surface (small curvature at the maximal cross sections) the dHvA amplitude is very high for $H \parallel [0001]$. Secondly, in this configuration two rather close dHvA frequencies 970 T and 940 T coexist leading to a beat in $M$ and $\chi$. Because of this beat there is the unique possibility to change the dHvA...
amplitude and the critical parameter $\chi$ by a factor three at constant $T$ by varying the magnetic field only very little. Thus, experimental conditions can be adjusted so that the transition to the inhomogeneous Condon domain state occurs in a part of each beat period. The sample was cut from the same piece that was used earlier for the preparation of the plate-like sample in which Condon domains were first observed by $\mu$SR [12]. The results shown here were measured on a rod-like sample of size $8 \times 2 \times 1$ mm$^3$ with the long axis parallel to [0001]. The Dingle temperature was $T_D = 2.0$ K. The measurements were made either directly by a micro Hall probe placed close to one end of the sample or by a compensated pickup coil using low frequencies of about 21 Hz and small modulation level ($< 6$ G). The experiments were carried out in a 10 T superconducting coil with homogeneity better than 10 ppm in 1 cm$^3$.

Fig. 1 shows Hall probe traces for an up and down sweep of the applied magnetic field around a dHvA antinode at 3.6 T and at $T = 1.3$ K. The hysteresis loop is very small and only visible by a zoom. A periodically arising induction difference $\delta B$ of about 3 G between the up and down sweep is measured at the steeper part, i.e. the paramagnetic part, of each dHvA period. The signal is about ten times higher than the noise level. The applied field is measured here by another Hall probe, placed sufficiently far from the sample, as the superconducting solenoid has its own small hysteresis when the current is swept. The hysteresis effect is clearly observed in fig. 1 using DC Hall probes but rather under the most favorable conditions at 1.3 K around a maximum of a magnetization beat. The DC Hall technique is not sensitive enough to study hysteresis as a function of temperature and field.

A standard AC modulation method with a compensated coil system used at very low modulation level $h$ is much more sensitive to detect nonreversible magnetization or hysteresis. This method is used to determine exactly the point of its appearance. If the modulation amplitude $h$ is much smaller than the oscillation period $\Delta H$, the measured response corresponds in good approximation to the derivative, i.e. the susceptibility $\chi(H) = \partial M/\partial H$. If the modulation is further decreased, the result should not change, the measurements should only be more precise.

Fig. 2 shows a different behavior as a function of modulation field amplitude $h$. All $\chi(H)$ curves of Fig. 2(a) and (b) are measured at low temperature where in the regions of each antinode hysteresis exists like it was shown on Fig. 1. Fig. 2(a) shows the normalized pickup voltage, i.e. response divided by the modulation level, measured by the AC method with 6 and 0.5 G modulation amplitude. In both cases $h \ll \Delta H$ so that one could expect identical curves. In fact, both curves are completely congruent except at the regions near the antinodes. Here, only the high modulation (6 G) level gives the expected result, which is the well known dHvA oscillation beat of beryllium. For comparison fig. 2(b) shows $\chi(H)$, calculated from $B(H)$ curves measured with Hall probes like in fig. 1 without field modulation. For small modulation amplitude deep "notches" in the dHvA oscillation envelope are observed. The notches occur at magnetic fields where the dHvA amplitude is big enough that the above described instability arises. Instead of a further increase, after having crossed the critical point, the dHvA amplitude decreases at the paramagnetic ($\chi > 0$) part of the dHvA period only. The diamagnetic part does not change. One would expect that both $M(H)$ and $\chi(H)$ oscillate around zero. This shows that the small modulation measurements can not correspond to the real $\chi(H)$ wave form. The amplitude decrease is absent at temperatures $T > 3.4$ K and at the nodes of the dHvA beat where the single crystal is in the homogeneous state.

The decrease of the dHvA amplitude at low modulation level is schematically shown on fig. 3(a). If the modulation is much bigger than the width of the hysteresis $\delta H$ the AC response corresponds in good agreement to the slope, i.e. $\chi(H)$. As soon as the modulation becomes comparable to the hysteresis loop size the response decreases [11]. The effect was measured for several modulation levels at different temperatures and at different magnetic fields with respect to the dHvA beat phase. The results are shown on fig. 3(b) and (c). The modulation level where the dHvA amplitude decreases gives the hysteresis width $\delta H$ which agrees with the above observed values of $\delta B$. No decrease is observed at temperatures $T > 3.4$ K and near the beat nodes.

This explanation is checked in a simple way. Usually the applied magnetic field $H$ ramps much slower than the AC modulation field $h$, i.e. the full magnetic field $H + h$. FIG. 1: Hysteresis loop observed by Hall probes in the paramagnetic part of the dHvA oscillations of beryllium. $B(H)$ traces for an up and down sweep of the applied magnetic field around a dHvA antinode at 3.6 T and $T = 1.3$ K (scale on the left). The hysteresis loop is visible in the insert. The induction difference $\delta B$ between these curves shows the value of the hysteresis (scale on the right).
FIG. 2: (a) Pickup voltages divided by the modulation level for low and high modulation amplitude at 1.3 K. (b) DC susceptibility derived from magnetization measurements with Hall probes without field modulation. Graphs to the right show respective zooms.

FIG. 3: (a) Schematic representation of the hysteresis loop showing that the response to an applied AC modulation field is nonlinear. (b) Modulation level dependence of the normalized pickup voltage for characteristic magnetic fields at 1.3 K. (c) The same dependence at the antinode at $T = 2.2$ K and $T = 4.2$ K. Nonlinearities arise at a critical temperature of 3.4 K.

always oscillates around the quasi static offset field $H$ and $B(H + h)$ makes a loop in presence of hysteresis. If the ramp rates are changed in a way that $dh/dt$ and $dH/dt$ are the same, then the magnetic field sweeps only forward with small steps in the direction of the ramp. Under this condition $B(H + h)$ never makes a loop as we go always along the hysteresis loop boundary and never inside. In this regime, which is usually not used, the lock-in amplifier does not measure the correct amplitude. We observed however in this regime the usual dHvA beat signal without notches.

The notches in the envelope of the first harmonic in-phase AC response are "compensated" by steeply rising higher harmonics at the same critical point. At the same field a phase shift appears in the pickup voltage. This means that the response to an AC modulation becomes extremely nonlinear in the presence of hysteresis. Fig. 4 shows the third harmonic (a) and the imaginary part (b) of the pickup voltage in a wide region of magnetic field at 1.3 K. The amplitude is big in both curves only around the beat antinodes. In the regions of nodes the signal is about zero. The insert of fig. 4(a) shows the 3rd harmonic in the same field interval as in fig. 2. The comparison shows that the signal appears and disappears with a threshold character at the critical points of the transition to the CDS. The insert in fig. 4(b) shows schematically that the response to an initially sinusoidal modulation field becomes highly distorted in the presence of a hysteresis loop. The response becomes more rectangular shaped like a window-function. This function can be composed, as it is well known, of odd harmonics of a sine. This explains that the third harmonic content of the pickup voltage is very high in the CDS. At magnetic fields without hysteresis the 3rd harmonic is very small as the modulation level $h = 2.5$ G is much less than the dHvA period of about 130 G at 3.6 T.

The hysteresis shape and size is reconstructed in fig. 5 like e.g. in [17]. The response to the sinusoidal modulation of 2.5 G amplitude is calculated by adding all in and out of phase contributions up to the fifth harmonic. The

FIG. 4: (a) Third harmonic of the pickup voltage. The insert shows a zoom to the above discussed field range. (b) Out of phase part of the pickup voltage. (both at 2.5 G modulation level and 1.3 K). The insert shows a schematic representation of the hysteresis showing that the response to a sinusoidal field modulation becomes window shaped and is slightly phase shifted with respect to the input.
same procedure was applied at different positions along the dHvA oscillations. At the diamagnetic part ($\chi < 0$) of every dHvA period all harmonics vanish and only the in phase response persists. Hence, a line with negative slope is calculated. The same behavior is found for $\chi > 0$ around a node with a line with positive slope. Whereas at the paramagnetic part in the region of the notches hysteresis arises and its size is maximal at the dHvA beat antinode.

The observed threshold behavior of the third harmonic and of the out of phase signal does not essentially change when the modulation level is varied providing $h \ll \Delta H$. This behavior was observed in the frequency range from 8 Hz up to about 200 Hz. Thus, these measurements offer a simple way to determine the Condon domain phase diagram.

Moreover, hysteresis may explain the data measured by Plummer \cite{Plummer1977} with an AC mutual inductance method at low modulation level. First Plummer thought that a new dHvA frequency was discovered. As the Fermi surface of beryllium was not consistent with this frequency, eddy currents at the rather high frequency of 100 Hz were invoked \cite{Plummer1977}. A comparison of Plummer’s data with our results (see Fig. 4a), black curve) shows the similarity. The deep notches in the dHvA oscillation envelope seem indeed to be the result of a new frequency. We repeated the measurements at higher frequencies and found the same behavior.

Hysteresis accompanies without doubt the appearance of the CDS resulting from the Pippard-Shoenberg instability. For any sample shape, not only in plate-like samples, in the CDS coexist two phases $B_1$ and $B_2$ with domain walls between them. The wall motion and pinning might, in principle, depend on the sample shape. Therefore, a plate-like sample was measured in the same $T$ and $H$ range. We found the same phase diagram for hysteresis formation in both samples. The hysteresis size, however, needs to be investigated more precisely as function of temperature, magnetic field and for various $n$.

Hysteresis itself is a result of interaction or pinning of domain walls with defects, impurities and the surface of the crystal. Moreover, the question of domain wall motion can not be neglected in many phenomena like e.g. acoustic wave propagation and absorption, and helicon waves \cite{Solt1977}. Recently, the question of Condon domain wall motion was considered in detail theoretically \cite{Egorov2002}. Unfortunately, an idealized model of a domain wall was used without taking into account the direct link with lattice deformation \cite{Egorov2002} and the metastable behavior due to hysteresis.

In conclusion we have shown that hysteresis occurs in the dHvA effect under the conditions of the CDS domain state. The observed hysteresis loop width is rather small, only a few Gauss. We have shown that the out of phase part and the third harmonic of the pickup voltage rise steeply when the magnetization becomes irreversible. This threshold character offers a simple and robust possibility to measure a Condon domain phase diagram.

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