Direct Observation of Condon Domains in Silver by Hall Probes

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(Dated: November 6, 2018)

Using a set of micro Hall probes for the detection of the local induction, the inhomogeneous Condon domain structure has been directly observed at the surface of a pure silver single crystal under strong Landau quantization in magnetic fields up to 10 T. The inhomogeneous induction occurs in the theoretically predicted part of the $H-T$ Condon domain phase diagram. Information about size, shape and orientation of the domains is obtained by analyzing Hall probes placed along and across the long sample axis and by tilting the sample. On a beryllium surface the induction inhomogeneity is almost absent although the expected induction splitting here is at least ten times higher than in silver.

PACS numbers: 75.45.+j, 71.70.Di, 75.60.-d

Periodic formation and disappearance of a phase with diamagnetic and paramagnetic domains was predicted by Condon\textsuperscript{1} to occur in a normally nonmagnetic metal in strong magnetic fields ($H$) at low temperatures ($T$). The domains arise under the condition $\chi = \mu_0 \partial M / \partial B > 1$, where $M$ is the oscillating magnetization of the electrons due to Landau quantization and $B = \mu_0 (H + M)$ the total induction inside the sample. In this case $\mu_0 \partial H / \partial B = 1 - \chi < 0$ which implies thermodynamically unstable sections and the multivaluedness of the induction $B(H)$ within some part of each dHvA oscillation period\textsuperscript{2,3}. For a long rod-like sample oriented along $H$, the instability is avoided by a discontinuous jump $\delta B = B_2 - B_1$ between two stable values $B_1$ and $B_2$ at a given critical field $H_c$. For a plate-like sample perpendicular to $H$, the boundary condition $B = \mu_0 H$ for a uniformly magnetized state leads to domain formation with alternating regions of diamagnetic and paramagnetic magnetization for $H$ in the range $B_1 < \mu_0 H < B_2$\textsuperscript{4}. The proportion of the domains varies with $H$ so that $B = \mu_0 H$ is fulfilled as an average over the sample\textsuperscript{5,6}. The $H-B$ diagram is similar to the $p-V$ diagram of a van der Waals gas, only with more than one discontinuity interval $\delta B$, situated periodically on the $B$ axis.

The existence of domains has been firstly discovered by Condon and Walstedt on a single crystal of silver\textsuperscript{7}. The domains were revealed by a local field splitting of the NMR line, corresponding to a local field difference of about 12 G between the paramagnetic and diamagnetic regions in fields of about 9 T. This pioneering result remained the only reference work in the next decades. New experimental possibilities appeared with the development of muon spin rotation. Condon domains were observed in beryllium, white tin, aluminum, lead, and indium\textsuperscript{8,9}. By now, no doubt, Condon domains are expected to appear in pure single crystals of all metals. Thermodynamic aspects of the Condon domain phase transition have been recently treated theoretically\textsuperscript{10}. While the state of art in this field has been recently reviewed\textsuperscript{11}, some important questions however, concerning domain size and topology, domain wall energy, and pinning properties can only be solved with a detailed knowledge of the domain structure.

The state with Condon domains can be considered as physically similar to the intermediate state of type I superconductors, where superconducting and normal regions form in an applied magnetic field. Therefore, domain structures resulting of such different phenomena as superconductivity and dHvA effect may be rather similar. Unfortunately, the magnetic contrast, that is the ratio of $\delta B$ to $B_2$, is not more than 0.1 % for Condon domains (compared to 100% for the intermediate state). Besides, the magnetic field itself is here hundred times higher. Thus, methods like magnetic decoration or magneto-optical detection used for intermediate state imaging\textsuperscript{12} can not be used for Condon domains.

In this Letter we present the first experimental results for direct observation of Condon domain structures in silver by a system of ten micro Hall probes being close to the single crystal surface. In the homogeneous state, without domains, all probes show the same dHvA signal $B_1(H)$, i. e. all $B_i = B(H)$, where $i = 1, 2...10$ are the Hall probe numbers. In the domain state, the Hall voltages differ between the different probes in the paramagnetic part of the dHvA period. This implies an inhomogeneous magnetic field distribution due to Condon domains at the sample surface. In our measurements the surface of the crystal was either normal to $H$ direction or slightly tilted ($13^\circ$). By comparing the data of neighboring Hall probes, new information about the domain structure has been extracted.

Fig. 1 shows the Hall probe set-up made of a 1 $\mu$m thick Si doped GaAs layer sandwiched between two 10 nm
thick undoped GaAs layers. Two arrays of five Hall probes \((10 \times 10 \text{ \(\mu\)m}^2 \text{ at 40 \(\mu\)m distance})\) are placed at a distance of \(b = 1 \text{ mm}\). One array, \(L\), is oriented along the long axis of the sample; the other, \(T\), transverse to this axis. A DC Hall current of 100 \(\mu\)A was applied in series to all five Hall probes of an array. The Hall voltages were read out simultaneously by 5 Keithley multimeters; the arrays \(L\) and \(T\) were measured one after another. Due to the 3D conducting layer the \(V_i(B)\) characteristics of the Hall probes were in good approximation linear up to 10 T even at 1.3 K. The correct calibration of the Hall probes was tested at temperatures between 4.2-3.6 K where all Hall probes showed exactly the same dHvA oscillations even at 1.3 K. The correct calibration of the Hall probes was tested at temperatures between 4.2-3.6 K where all Hall probes showed exactly the same dHvA oscillations of the homogenous silver sample. The detection limit of the Hall probes was smaller than 1 G. A high homogeneity (better than 10 ppm in 1 cm\(^3\)) 10 T superconducting magnet was used to set a fixed offset magnetic field \(H_0\). The slowly varying superimposed field \(H_V\) (\(\pm 15 \text{ mT}\)) was made by a watercooled resistive coil. Thus the total applied magnetic field was \(H = H_0 + H_V\).

The measurements were performed on a high quality silver single crystal of \(2.4 \times 1.6 \times 1.0 \text{ mm}^3\). The largest surface of the sample was normal to the [100]-axis of the crystal. The sample was prepared in the same way as in experiments on radio frequency size effect and time of flight effect (see references in [11]). The very good quality of the sample results in a very low Dingle temperature, which was estimated from our measurements to be about \(T_D = 0.2 \text{ K}\). The sample was annealed in \(O_2\) \((10^{-2} \text{ Pa})\) at 750°C during 10 hours. It has a residual resistance ratio \(RRR = R_{300K}/R_{4.2K} = 1.6 \times 10^4\), measured by the contactless Zernov-Sharvin method [12]. For a mirror-like surface, the crystal was slightly repolished by 0.1 \(\mu\)m diamond paste after annealing. The surface before polishing had a roughness of about 20-30 \(\mu\m\) and no induction splitting due to Condon domains could be observed. The sample was glued by narrow strips of cigarette-paper to the set-up frame to fix the crystal on the Hall probes in order to avoid damage or strain of the single crystal upon cooling down. Moreover, the sample was slightly pressed by a cotton tampon to hold it reliably in high magnetic field.

Fig. 2 shows typical \(B(H)\) traces for Hall probes \(B_1\) and \(B_3\) of the L-array over three dHvA periods at 10 T and 1.3 K. In each paramagnetic part of the dHvA period two different inductions are measured at the surface of the sample whereas the induction is homogeneous in the diamagnetic part. The measured traces are reversible for increasing and decreasing magnetic field. We ascribe the measured difference between the induction of neighboring Hall probes to the existence of Condon domains.

The maximal induction splitting \(\delta B\) in a dHvA period was measured as a function of temperature at 10 T (see Fig. 3a) and as a function of field at 1.3 K (see Fig. 3b). At 10 T, the phase boundary is crossed at about 3 K. At 1.3 K, the crossing occurs at about 5 T. The field and temperature range for the occurrence of the induction splitting is in agreement with the \(T-H\) phase diagram for the Condon domain state in Ag, as shown in the inset of Fig. 3a, for the theoretically calculated phase-diagram of Ag with Dingle temperature 0.2 K [13]. The solid line in Fig. 3a is the calculated induction splitting \(\delta B\) [13] with the phase transition temperature (3 K) and the maximum splitting in silver (12 G) measured by Condon and Walstedt [11] as parameters.

An anomalous alternating transition order of the Hall probes between the diamagnetic and paramagnetic phase is shown in Fig. 4. Although reproducible, the observed order depends strongly on the experimental configuration. A basically different behavior can be seen between the T- and L-probes in, respectively, Figs. 4 and 5. No regular transition order was observed for T-probes. Sometimes, they transit in ascending (1-5) or in descending order, as if the domain laminae were slightly tilted to the long axis of the sample. Sometimes, as shown in

![FIG. 1: Experimental configuration with the longitudinal (L) and transverse (T) arrays of Hall probes at distance \(b = 1 \text{ mm}\) \((s^2 = 10 \times 10 \text{ \(\mu\)m}^2; d = 40 \text{ \(\mu\)m})\).](image1)

![FIG. 2: \(B(H)\) trace for silver single crystal with \(H \parallel [100]\) showing the splitting \(\delta B\) of \(B_1\) and \(B_3\) of the L-array for three dHvA periods separated by dotted lines. The calibration of \(B\) is with respect to the offset field \(\mu_0H_0 = 10 \text{ T}\).](image2)
Fig. 3: Maximum induction difference $\delta B$ as a function of temperature (a) and magnetic field (b). Solid line in (a) is calculated from [13]. Inset shows the theoretical phase diagram [13] with dotted lines (a) and (b) indicating the two measuring tracks.

FIG. 4: Example of successive transitions for five T-probes between diamagnetic and paramagnetic phase. The sweep rate was 0.5 mT/min.

$$\delta B = B_1 - B_5$$

Fig. 4, a middle T-probe transits the last or the first, as if the laminae are bent. In contrast, the order of the L-probes is always 1,2,3,4,5 or reversed as it is shown in Fig. 5a. This implies that the domain structure is approximately laminar with the laminae mainly oriented transverse to the long axis of the sample. However, we found that the L-probe sequence changes alternately between dHvA periods which implies that the domain-wall movement changes direction along the long sample axis between successive dHvA periods. $\delta B = B_1 - B_5$, shown in Fig. 5a, changes sign alternately during four or five periods (see Fig. 5a). Then the, what we will call, "pendulum" effect breaks down during two periods where the transition order is not clear. After this the pendulum effect repeats.

On a slightly tilted sample the pendulum effect disappears completely. Fig. 5a shows the change of the situation after tilting the sample. After rotation around the long sample axis by 13° domains draw up to a regular laminar structure oriented always transverse to the long axis. A similar behavior was observed in white tin in the intermediate state [9, 10] indicating the preference of domain walls to align along the sample surface. Furthermore, the transition order is now the same for all dHvA periods (see Fig. 6b). The rotation of the silver single crystal with respect to the magnetic field affects the dHvA frequency spectrum. Only one dHvA frequency ("belly") remains for the 13° tilted sample. The beating pattern in the oscillatory dHvA signal of the magnetization for the perpendicular field orientation might play a role in the occurrence of the pendulum effect. In this respect we...
note that the dHvA frequencies (belly orbit at 47300 T and rosette orbit at 19000 T) for the perpendicular field orientation would be compatible with the observed pendulum effect of Fig. 6a. The transitions of the individual Hall probes are very sharp compared to the whole field range of the domain state in neighboring Hall probes (see Fig. 4). This means that the thickness of the wall is much smaller than the period of the domain structure. We have never seen more than one transition of a Hall probe in a period. This implies that we saw always only one boundary between para- and diamagnetic phases in an array of 5 successive Hall probes meaning that the period of the domain structure is certainly larger than the distance of \( \approx 150 \, \mu m \) between the edge L-probes. This under limit for the domain period \( (p) \) should be compared with the value obtained from the square-root averaged expression \( p \propto \sqrt{w \ell} \) for a sample with thickness \( (t) \) and domain wall thickness \( (w) \) \[2\]. With the proposed cyclotron radius for the wall thickness \( 1 \, \mu m \) at 10 T in Ag, one obtains at least a 5 times smaller value \( (\approx 30 \, \mu m) \) \[4\]. Therefore, from our experiments we find a wall thickness of at least 20 \( \mu m \). This is in agreement with the observation that two neighboring middle L-probes at a distance of 40 \( \mu m \) show often intermediate but different induction values. Therefore, the thickness of a domain wall can not be much smaller than 20 \( \mu m \).

As the real domain pattern turns out to be somewhat bigger than expected, we need either a new set-up with better adapted Hall-probe distances or a scanning Hall probe for more detailed measurements of the domain structure.

Exactly the same measurements as presented above were performed on a beryllium sample cut from the same single crystal where Condon domain formation was observed using muon spectroscopy \[5\]. The sample was prepared with a surface quality comparable to the Ag crystal. Even though the expected \( \delta B \) inside the crystal is ten times higher than in silver, we did not find \( \delta B > 2 \, G \) on the sample surface. The attempt of Condon and Walstedt to find domains in beryllium by NMR was not successful, either \[6\]. The authors gave explanations related to the quadrupole broadening and the long nuclear thermalization time in beryllium. However, now we believe that the main reason is the absence of induction splitting \( \delta B \) at the sample surface. This could be an intrinsic property of beryllium related to its anisotropic magnetostriction \[14\].

In conclusion, Condon domains in silver with induction splitting up to 10 G were observed by micro Hall probes at fields and temperatures which are in agreement with the theoretically estimated phase diagram. A laminar domain structure was found with the orientation mainly transverse to the long sample axis. The domain transitions are always reversible for increasing and decreasing magnet field. For a slightly tilted sample the strange pendulum effect disappears, and the transitions occur in the same order for all dHvA periods. The domain period is not smaller than 150 \( \mu m \) and the domain wall thickness must be about 20 \( \mu m \). Condon domains in beryllium do not emerge to the surface.

We are grateful indebted to M. Schlenker for his support and continuous interest to this work, to I. Sheikin and V. Mineev for fruitful discussions, and to J. Marcus for his help in sample surface preparation. F. Schartner is acknowledged for the preparation of the Hall probes.

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1. J.H. Condon, Phys. Rev. 145, 526 (1966).
2. D. Shoenberg, Magnetic oscillations in metals, (Cambridge University Press, Cambridge, 1984).
3. G. Solt et al., Phys. Rev. B 59, 6834 (1999).
4. J.H. Condon and R.E. Walstedt, Phys. Rev. Lett. 21, 612 (1968).
5. G. Solt et al., Phys. Rev. Lett. 76, 2575 (1996).
6. G. Solt and V.S. Egorov, Physica B 318, 231 (2002).
7. A. Gordon et al., Phys. Rev. Lett. 81, 2787 (1998).
8. A. Gordon et al., Adv. in Physics 52, 385 (2003).
9. J.D. Livingstone and W. DeSorbo, in Superconductivity, edited by R.D. Parks (Dekker, New York, 1969), Vol. 2.
10. Yu.V. Sharvin, Soviet Phys. JETP, 6, 1031 (1958).
11. V.A. Gasparov and R. Huguenin, Adv. in Physics, 42, 393 (1993).
12. V.B. Zernov and Yu.V. Sharvin, Zh. Eksp. Teor. Fiz., 36, 1038 (1959).
13. A. Gordon et al., Phys. Rev. B 59, 10864 (1999).
14. V.S. Egorov and P.V. Lykov, Soviet Phys. JETP, 94, 162 (2002).