DIRECT EVIDENCE FOR MAGNETIC DOMAINS IN SILVER

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Pulsed NMR studies of ^{109}Ag nuclei in a single-crystal specimen of Ag metal at helium temperatures with an applied field $H_0 \cong 90$ kOe reveal the presence of magnetic domains due to de Haas–van Alphen magnetization through the associated splitting of the ^{109}Ag NMR line. Domain formation is found to have the expected de Haas–van Alphen periodicity, and the domains behave in generally good accord with theory.

Some time ago it was demonstrated[1] that under conditions of strong de Haas–van Alphen (H-A) magnetization intensity, I, relative to the H-A period, such magnetization can achieve a state of lower free energy by breaking up into domains having equal and opposite magnetizations, $\pm I_D$. It was further shown[1,2] that the resulting domains are regions of uniform magnetization intensity I and magnetic induction B, which are wide (typically 30 μ) relative to the wall regions separating them (typically 2 μ) as a consequence of the positive wall energy.[2] Although this phenomenon was found to be consistent with a number of experimental effects,[1] there has been up to the present no direct observation of the domains themselves.

Here we report direct evidence for H-A domain formation in a single-crystal slab of silver metal, based on results from a pulsed-NMR study of the ^{109}Ag nuclei contained in an rf skin depth at the surface of the slab. The experiments were carried out at helium temperatures with an applied field $H_0 \cong 90$ kOe normal to the plane of the slab, giving a ^{109}Ag nuclear Larmor frequency $f \cong 18$ MHz. With this arrangement the domain or mixed-phase state of magnetization gives rise to two NMR lines corresponding to regions of parallel ($+I_D$) and antiparallel ($-I_D$) magnetization intensity relative to H_0. These lines are separated in frequency by an amount corresponding to the splitting $\Delta B = 8\pi I_D$ of magnetic induction between the two types of domain. This "two-frequency" effect is clearly observed, and our study demonstrates that the domain effect has the following properties predicted in Refs. 1 and 2: (1) The two-frequency effect appears and disappears with the H-A periodicity as H_0 is swept, in accord with the expectation that domains form only during the paramagnetic ($dI/dB > 0$) portion of the H-A cycle; (2) the two components of the domain-region NMR spectrum are sharply defined indicating a reasonably large ratio of domain dimensions to wall thickness; (3) the local values of B in the domains are independent of applied field H_0, continuity of the average value of B at the surface being satisfied by adjustment of the relative areas of parallel and antiparallel domains; (4) the splitting ΔB is slightly smaller than the H-A period as expected.

Silver was chosen for these experiments because of a number of favorable properties. It shows the strongest tendency toward domain formation of any metal.[3] With the applied field along the $\langle 100 \rangle$ crystal axis there are two periodic H-A magnetizations. The dominant one is due to the belly oscillations and has a period of about 16.7 G at 90 kG; the weaker is due to the rosette orbits and has a period ~2.5 times longer. Further, silver possesses two ~50% abundant species with nuclear spins $I = \frac{1}{2}$ (^{107}Ag and ^{109}Ag), so that quadrupole effects are excluded from the NMR spectra. The natural linewidths of these resonances are of the order of tenths of a gauss and allow good resolution of the spectrum of B fields in the domains.

The silver specimen used was a plate ~8 mm square × 0.8 mm thick along a [100] direction. This piece was spark machined from a single-crystal boule of silver having a residual resistance ratio of ~3000.[4] The specimen was chemically etched and heavily tin-plated on five sides, so that the silver nuclei on those surfaces could not contribute to the resonance signal. The remaining face was then mechanically and chemically polished. The two copper coils of a crossed-coil NMR head were wound directly onto the specimen, which was mounted with its plane surface perpendicular to the axis of a 100-kG superconducting solenoid. A pulsed-NMR setup of the coherent type was employed, so that the ^{109}Ag free-precession signals observed were mixed with an rf reference signal from the same oscillator used to drive the transmitter. The 90-kOe applied field was sufficiently homogeneous over the specimen surface to give a transverse ^{109}Ag decay time $T_2^* \sim 1.5$ msec, which allowed a de-

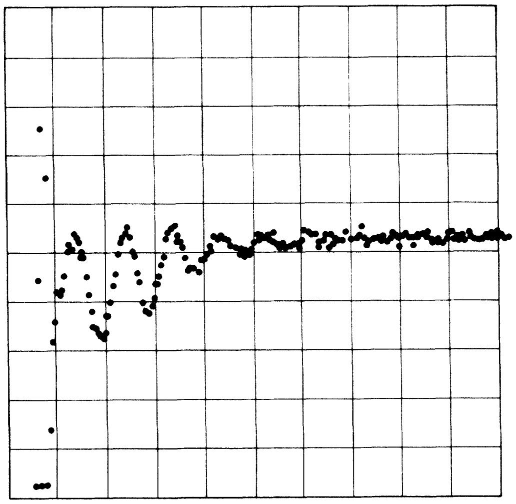

FIG. 1. ^{109}Ag free-precession signal showing the presence of two well-defined nuclear Larmor frequencies. The horizontal scale is 0.5 msec per division.

termination of magnetic induction B to within ~0.5 G. The free-induction signals from the ^{109}Ag nuclear spins were clearly visible on an oscilloscope at 4.2°K, though the signal-to-noise ratio was rather poor. A multichannel analyzer was employed to accumulate 10 or so free induction signals and improve the quality of the data.

The composition of the ^{109}Ag NMR signal with H_0 adjusted to the center of a domain region is shown in Fig. 1. Two distinct NMR frequencies are apparent here, one at very nearly "zero beat" with the reference oscillator giving the exponentially decaying baseline; the other, at a difference frequency of ~2.2 kHz, is seen as an exponentially decaying oscillation superimposed on the baseline. Upon adjusting the reference oscillator to zero beat with the second signal, a picture similar to Fig. 1 is obtained, the roles of the two lines of the NMR spectrum simply being reversed. This result is by itself strong evidence for magnetic domains. A splitting $\Delta B \cong 11$ G is obtained from these measurements. In the diamagnetic portion of the H-A cycle only a single NMR frequency is found corresponding to the expected disappearance of the domains.

The detailed behavior of the induction B inside the specimen as a function of H_0 has been monitored by stepping H_0 in small increments with a constant reference oscillator frequency. Values of the ^{109}Ag Larmor frequency (interpreted as B) are then obtained from photographs such as in Fig. 1. Figure 2(a) shows the values of B measured in this way over a range of H_0 values somewhat greater than one H-A period, using two different reference frequencies. At the center of the plot the domain region is clearly in evidence with two branches of B which level off and become independent of H_0 as the theory requires. As noted above, the average B field in the specimen is then maintained equal to B_0 through adjustment of the relative areas occupied by the two types of domain. This may be checked by measuring the nuclear signal amplitudes for the two branches of B vs H_0, these amplitudes being simply proportional to the total sample area occupied by their respective domain types in the region of constant B values. The signal amplitudes are plotted in Fig. 2(b) and show the expected complementary behavior. Outside the region of constant B values the signal amplitudes

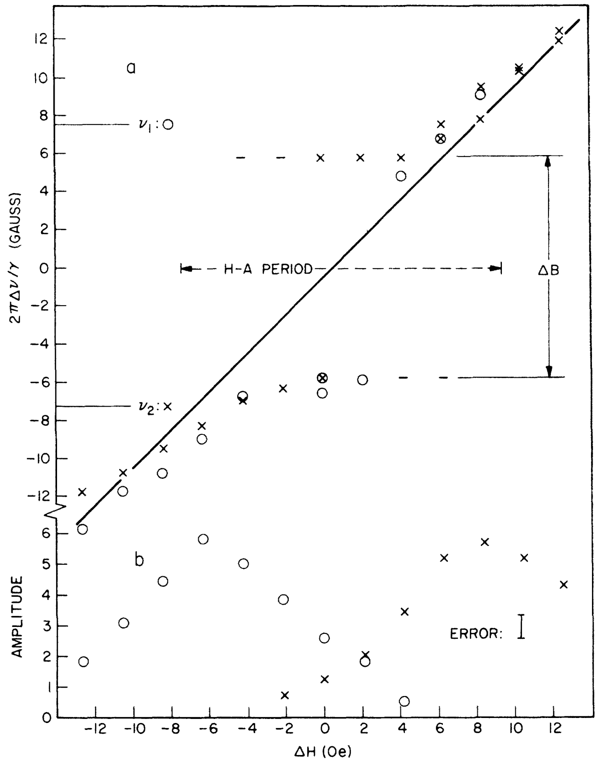

FIG. 2. (a) Values of magnetic induction in the rf skin-depth layer obtained from measured ^{109}Ag Larmor frequencies, plotted as a function of applied field H_0. Two reference-oscillator frequencies were used, $\nu_1 = 17\,913.7$ kHz and $\nu_2 = 17\,916.7$ kHz. The $\Delta H_0 = 0$ point on the horizontal scale corresponds to approximately 89 970 Oe. The dashes indicate the presence of NMR signals too weak for accurate frequency measurements. (b) Free-precession signal amplitude (in arbitrary units) for the two branches of B vs H_0, with the same correspondence of circles (ν_1) and crosses (ν_2) to reference-oscillator frequencies as in (a).

drop because the Larmor frequencies become increasingly detuned from the driving oscillator. Other studies in which the field and oscillator frequency are stepped in synchronism show that the signal amplitude is substantially constant in the single-mode region.

Figure 2(a) shows that the B field in the sample roughly follows H_0 outside the region of domain formation. A solid line of slope unity is drawn in for comparison. We note that a slight discrepancy exists between the data points and the solid line at the extrema of the H-A period indicated in Fig. 2(a), i.e., the change of $2\pi\Delta\nu/\gamma$ over the H-A period appears to be ~10% greater than the change in H_0. This effect is thought to be real and has been observed in other studies of Larmor frequency versus field H_0 over several H-A periods. It is not consistent with the model[1] of strictly periodic H-A magnetization, since at the center of the diamagnetic ($dI/dB < 0$) region one then has $I = 0$ and $B = H_0$ regardless of sample geometry. We tentatively attribute this effect to the H-A magnetization and possible oscillatory Knight-shift contribution from the rosette orbit. Rosette-orbit effects were in evidence in the domain-formation phenomena, the NMR results being much clearer in some regions of H_0 than in others removed by several belly-orbit periods.

The strength of the H-A magnetization which is responsible for the domains is customarily characterized by a parameter $a \equiv 8\pi^2 I_0/\Delta H_0$, where I_0 is the peak value of the sinusoidal function $I(B)$ and ΔH_0 is the H-A period.[5] It was previously shown[1] that domain formation is only possible for $a > 1$. Here we may calculate a from the measured splitting ΔB: $a = (\pi\Delta B/\Delta H_0)/[\sin(\pi\Delta B/\Delta H_0)]$. The data of Fig. 2(a) give $\Delta B/\Delta H_0 \cong 0.69$ yielding $a \cong 2.6$ at $T = 1.4$°K, a value which is nearly that obtained from other work.[6] However, the temperature dependence of ΔB (and therefore of a) is smaller than expected. If there were an oscillatory Knight shift[7] it might change the quantitative interpretation of ΔB.

During the temperature dependence studies it was noted that there was no splitting ΔB due to domains above about 2.5°K and that the existence of a splitting at 2.2°K depended on whether the sample has been heated or cooled to that temperature, consistent with a supercooling effect.

We also studied a "sandwich" sample on the belief that there might be less domain branching. The sample consisted of two of the type described previously, mounted with the silver faces adjacent with a gap of a few microns. The results showed that there was more broadening of the NMR in the sandwich even though the two silver pieces had the same relative positions that they had in the original boule. Otherwise the results were equivalent, suggesting that the observed ΔB splitting is a bulk effect in accordance with the theory, rather than a purely surface effect.

Similar experiments have been attempted on beryllium at 19 kG but the nuclear thermalization time of $\sim\frac{1}{2}$ h and the inherent nuclear quadrupole splitting made the data collection and interpretation difficult. A periodic line broadening was observed but its explanation in terms of a ΔB splitting was not clear.

In conclusion, this experiment confirms the prediction of magnetic domains for this system. The sharp NMR splitting indicates a large ratio of domain dimensions to wall thickness and confirms the calculated positive wall energy.

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[1] J. H. Condon, Phys. Rev. 145, 526 (1966).

[2] J. H. Condon, in Proceedings of the Tenth International Conference on Low Temperature Physics, Moscow, U. S. S. R., 31 August-6 September 1966 (Vimiti Publishing House, Moscow, U. S. S. R., 1967).

[3] D. Shoenberg, Phil. Trans. Roy. Soc. A255, 85 (1962).

[4] We thank P. H. Schmidt and C. C. Grimes for the silver sample and residual resistance ratio determination. We also thank F. S. L. Hsu and H. J. Levinstein for spark machining and polishing the samples.

[5] A. B. Pippard, Proc. Roy. Soc. (London), Ser. A 272, 192 (1963).

[6] G. Seidel, private communication; at $T = 1.4$°K, $H_0 = 78$ kG, $a = 1.5$, and $T_D = 1.0$° for a sample with residual resistance ratio ~700. Extrapolation to 90 kG and use of $T_D = 0.8$°K for our purer sample yields $a = 2.6$.

[7] J. M. Reynolds et al., Phys. Rev. Letters 16, 609 (1966), and references therein.