Flux Quantization in Magnetic Nanowires Imaged by Electron Holography

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Off-axis electron holography has been employed to visualize and measure the magnetic flux leaking from cobalt wires having a radius of 40 nm and a length of a few μm. The magnetic flux of a single nanowire can be understood as the flux of a superposition of magnetic dipoles oriented along the nanowire axis at locations revealed by the phase as well as the amplitude image reconstructed from the hologram. Surprisingly, the reconstructed phase image showed that the total flux of the individual dipoles was always h/e, defined as a flux quantum Φ_0 and corresponding to a phase change of 2π. We were able to image single nanowires with a flux of 1Φ, 2Φ, and $3\Phi_0$, measured with a precision of $\Phi_0/32$.

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Typically, the electron microscopic observation of magnetic structures is made by the so-called Lorentz microscopy technique [1]. However, off-axis electron holography allows one to measure quantitatively the flux leaking from a magnetic specimen [2], and has been successfully applied to ferromagnetic microprobes [3] and micrometer-sized barium ferrite particles [4]. On the other hand, electrodeposition in nanoporous membranes has been shown to be a useful technique to make submicrometric particles of controlled morphology [5]. In this paper, we report the study of cobalt cylindrical nanowires, with a radius of 35–45 nm and a length of 1–6 μm. They were obtained by electrodeposition in track-etched polycarbonate foils [6], typically 6 μm in thickness, produced by irradiation with heavy ions and etching along the tracks left by the ions, resulting in cylindrical pores with radius ranging from 15 nm to a few micrometers. This technique of template synthesis by electrodeposition has been recently reviewed [7] and magnetic properties in Co and Ni wires were reported [8]. Cobalt wires were grown using a bath containing cobalt sulfate and a buffer acid (boric acid). In a standard three-electrode configuration, we applied a constant potential to reduce the Co ions in the membrane. A saturating external field was applied perpendicular to the membrane. Magnetic measurements performed on a 3 mm disk of membrane indicated that 90% of the magnetization was kept in the remanent state. Keeping the sample in this state, the membrane was dissolved and the nanowires in the suspension were subsequently deposited on carbon foils without holes supported by a microscopic grid.

Electron holography of magnetic specimens requires that the current in the objective lens is switched off to keep the magnetization state of the sample. The condenser lens was used to obtain a parallel illumination of the specimen. In our field emission transmission electron microscope (TEM) Hitachi HF-2000 FEG, the biprism is inserted near the back focal plane of the objective lens above the first intermediate lens. Since the objective lens was switched off, the specimen had to be focused with the first intermediate lens, while the remaining three lenses were fully excited to reach the maximum possible magnification in this configuration [1100 times on the charge-coupled device (CCD) camera]. A negative voltage is applied to the biprism fiber in order to separate the reference wave and the object wave. With the first intermediate lens the two waves are overlapped and the specimen is focused. The width of the interference region depends on the voltage applied to the biprism fiber. Typically, for -18 V we obtain an interference field of 6.5 μm width and interference fringes with a spacing of approximately 80 nm. The contrast $(I_{\max} - I_{\min})/(I_{\max} + I_{\min})$ of the interference fringes corresponds to 10%, which gives still reliable phase images. Because of the low magnification, one interference fringe period corresponds to only 4 to 5.5 pixels on the CCD camera depending on the orientation of the fringes with respect to the CCD camera. This limits the contrast of the interference fringes as well as the maximum magnitude of the voltage which can be applied to the biprism fiber. The holograms were recorded with a Gatan slow scan CCD camera with 1024×1024 pixels and a dynamic range of 14 bits in recording times of 4–8 s. The images were reconstructed with the help of HOLOWORKS 1.0 [9]. The phase images have a maximum spatial resolution of twice the fringe spacing, i.e., 160 nm. The noise level in the phase images was measured in flat phase regions (i.e., far from the magnetic nanowire) as the standard deviation of the phase in several square regions of 160 nm edge length; a value of $2\pi/40$ was found. In order to decrease the disturbing influence of the noise on the appearance of phase maps of Figs. 2 and 3, a slightly smaller aperture was selected in Fourier space that limits the spatial resolution to 200 nm. Additionally, the streaks present in the sideband due to diffraction at the biprism fiber were partially eliminated by filtering them out. The phase images were n times amplified by representing the

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phase modulo $2\pi/n$ instead of modulo 2π. This technique, called phase difference amplification [2], is also amplifying the noise.

According to the Aharonov-Bohm effect the phase difference between the object and reference wave measured by electron holography corresponds to $\Delta\phi = (e/h)\int\int \mathbf{B} \cdot \mathbf{n}\, dS$, where the integral is taken over the surface enclosed between the two electron trajectories [2,3]. It follows from this formula that a phase change of 2π corresponds to a magnetic flux of h/e. Since the reference wave passes the object at a distance which corresponds to the width of the interference field (i.e., typically 6.5 μm) it is perturbed by the far field of the magnetic specimen. We have checked that the magnetic field already at a distance of 3 μm from a nanowire would perturb only slightly the reference wave. We must keep in mind that the phase image is different from the two-dimensional distribution of the magnetic flux in the specimen plane, because the phase difference is integrated along the direction of the optical axis of the microscope [3].

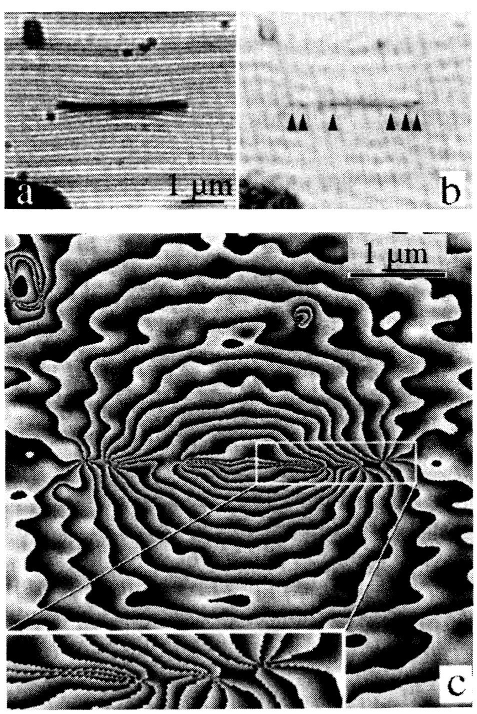

FIG. 1. (a) Electron hologram of a cylindrical cobalt nanowire 3 μm in length, 40 nm in radius. Note the small fragments of polycarbonate membrane as well as pieces of broken nanowires. (b) Reconstructed amplitude image in a logarithmic scale. Black spots (marked by arrows) are indicating the ends of three dipoles located on the nanowire. (c) Reconstructed phase image, 8 times phase amplified. Three parallel dipoles, each with a flux of $\phi_0 = h/e$ are located on the nanowire. The region marked is shown 2 times magnified in the inset.

Figure 1(a) shows a part of a hologram with a cobalt nanowire of 3 μm length near the center of the image. The curving of the interference fringes due to the magnetic field can directly be observed. The opposite curvature of the interference fringes above and below the nanowire reflects the opposite gradient of the magnetic flux on the two sides. The reconstructed images of the amplitude and the phase are shown in Figs. 1(b) and 1(c), respectively. The phase image in Fig. 1(c) is a phase difference image, amplified 8 times, showing the magnetic flux lines. The phase difference from one bright line to the next corresponds to $2\pi/8$. Obviously, three parallel dispoles are located on this nanowire; the poles can be recognized easily, since eight bright lines always join together at their positions. The phase changes rapidly in the vicinity of the poles (compare 2 times magnified inset) and consequently the interference fringes of the hologram are strongly bent near the poles. As a result the amplitude image shows a strong (black) contrast at the position of the poles. It should be realized that this is actually an artifact due to the limited resolution in the hologram. Notice that no Fourier filtering has been applied to obtain these two images. The phase image, Fig. 1(c), reveals that the phase changes by $+2\pi$ on a circular path in the positive sense, on the left hand side of the image and by -2π on the right hand side. Since a phase difference of 2π corresponds to a flux of h/e, this implies that each of the three dipoles has a magnetic flux of $\phi_0 = h/e$, i.e., the magnetic flux of the nanowire is quantified in units of ϕ_0.

Figures 2 and 3 show similar phase amplified images of two different nanowires of length 6 and 3 μm, respectively. Two dipoles, each with a flux of ϕ_0, can be recognized in the first case (Fig. 2) and a single dipole with a flux of ϕ_0 in the second case (Fig. 3). Localized disturbances of the phase images caused by polycarbonate membrane residues as well as by small pieces of broken nanowires can be noticed in both cases. We checked that apart from these disturbances the phase change was exclusively due to the magnetic flux by taking an additional hologram of the same nanowire after reversing the specimen upside down

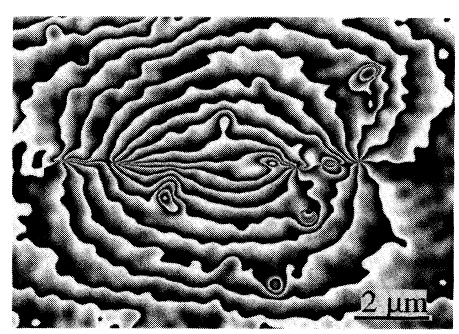

FIG. 2. Reconstructed phase image of a 6 μm long nanowire, 8 times phase amplified. Two parallel dipoles, each with a flux of $\phi_0 = h/e$ are located on the nanowire.

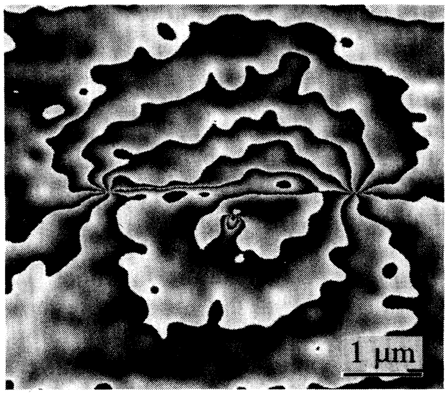

FIG. 3. Reconstructed phase image of a 3 μm long nanowire, 8 times phase amplified. A single dipole with a flux of $\phi_0 = h/e$ can be recognized.

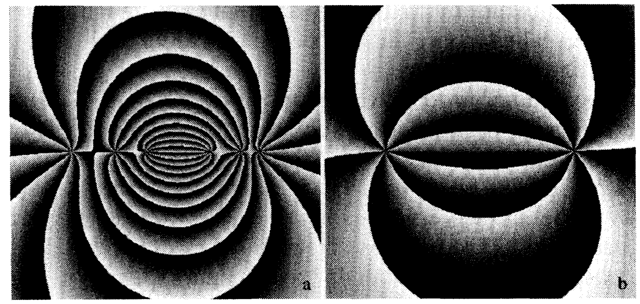

FIG. 4. (a) Simulated phase image (8 times phase amplified) of three parallel dipoles each with a flux of $\phi_0 = h/e$. The lengths have been selected to correspond to Fig. 1(c). (b) Simulated phase image (8 times phase amplified) of a dipole with a flux of $\phi_0\sqrt{3}/2$. Instead of eight equiphase lines only seven meet at the poles indicating a flux of approximately $7\phi_0/8$.

[10]. The phase image of the reversed hologram indicted just a reversed magnetization, excluding in this way effects by possible charging of the specimen. For the three samples we have verified with 32 times phase amplified images that the flux around each pole was quantified with a value of $\phi_0 = h/e$. We thus consider that the quantization is verified up to the precision of our phase determination.

The images presented allow us to estimate the average magnetization of the three nanowires if we simply add the dipole moments. Taking the lengths of the three dipoles in Fig. 1(c) 3, 2.4, and 1.3 μm and a wire radius of 45 nm (measured by conventional transmission electron microscopy) we estimated a magnetization of 1.2×10^6 A/m. The second nanowire (radius 40 nm), with dipole lengths of 6 and 3.6 μm, has an average magnetization of 1.0×10^6 A/m, and the third nanowire (radius 40 nm) of 0.6×10^6 A/m. All these values are below the saturation magnetization of bulk cobalt 1.42×10^6 A/m and are compatible with magnetic measurements on a macroscopic scale.

We simulated the observed phase images using an approach similar to Matteucci *et al.* [3], using the software package MATHEMATICA to calculate the phase images of a superposition of dipoles. A factor of 8 has been used for the phase difference amplification in the simulations. Since the centering of the sideband during reconstruction of the holograms is limited in precision, a weak linear phase wedge was added to account for the slight asymmetry of the observed phase images. Figure 4(a) presents a simulation for three superposed dipoles with lengths corresponding to Fig. 1(c), reproducing all important features of the experimental phase image.

Up to now, to our knowledge, no experiment allowed a complete visualization of the magnetization state of such small magnetic particles. The magnetic state we observed in Figs. 1 and 2 is unexpected for the remanent state. A superposition of collinear dipoles as shown in our figures is in contrast with the common belief of uniform magnetization in small particles. In Figs. 1 and 2 the poles of the superposed magnetic dipoles are separated and between two poles of the same "magnetic charge" we can recognize the presence of a neutral point. The magnetic charges shown in our figures are equal to the quantified magnetic charge g predicted by Dirac [11]:

$$g = \frac{h}{e\mu_0}.$$

Our determination of the phase difference modulo 2π along a closed curve is directly related to Dirac's method of derivation of the "magnetic charge" quantization. In order to check that the quantization we observed was not an artifact of the measurement method, we simulated the phase image of a fractional flux of $\phi_0\sqrt{3}/2$, positioning poles of magnetic charge $\pm g\sqrt{3}/2$ at the ends of a wire [Fig. 4(b)]. Along the dipole line joining the poles (nodal line) the calculated phase is not continuous, but we can evaluate the total flux, on a closed curve avoiding the nodal line, by counting the number of equiphase lines joining at a specific pole. For the fractional flux there are seven equiphase lines in the 8 times phase amplified image Fig. 4(b). This indicates that it is possible to differentiate between an experimental phase image of a dipole with a flux which corresponds to an integer multiple of ϕ_0 and one of a dipole with a fraction of ϕ_0.

We were able to obtain images of nanowires with a total flux, measured on a surface excluding the singularities (i.e., the nanowire), having quantified values, reproducibly evidenced for $n = 1$, 2, and 3 dipoles, thus confirming Dirac's prediction of magnetic charge quantization. The possibility of an unexplained experimental limitation is still open. If the observations are due to a limitation of the experimental technique, the quantization effect will give us the limit of resolution of electron holography applied to magnetic structures. If not, electron holography is able to image the spatial distribution of quantized "magnetic charges" and to measure their value.

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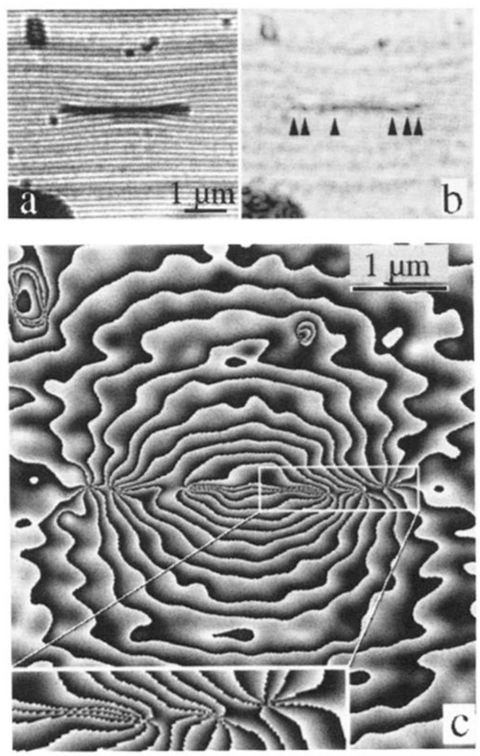

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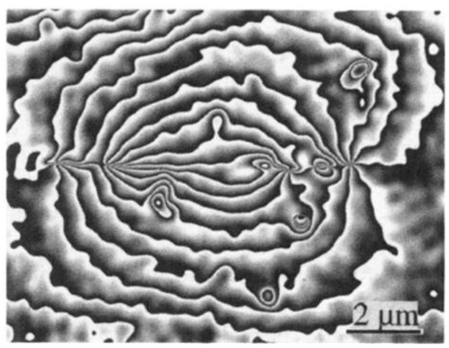

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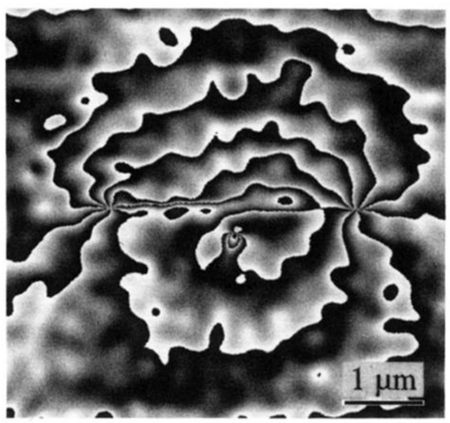

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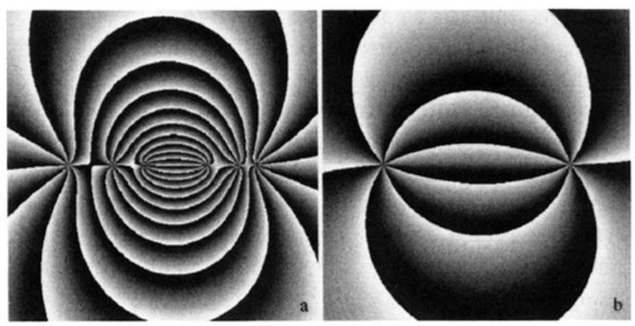

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