Magnetic ordering in 45 nm-diameter multisegmented FeGa/Cu nanowires: single nanowires and arrays

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Off-axis electron holography procedure.

As the phase image obtained contains information of not only the magnetic potential but also mean inner potential, is important to separate the components to obtain reliable quantitative measurements. The way to achieve this procedure is: i) turning the specimen over after acquiring a hologram and acquiring a second hologram from the same region, ii) performing a magnetization reversal experiment in situ in the microscope and then selecting pairs of holograms that differ only in the magnetization direction, iii) acquire holograms at two different microscope accelerating voltages, or iv) subtract with a phase image acquired above the Curie or Néel temperature of the specimen. On this work, we performed the magnetization reversal and the different voltages approaches.

Figure S1 presents the in-situ magnetization reversal procedure performed on a single GaFe/Cu nanowire. After tilting the microscope grid 25 degrees on X and 5 on Y we measure the change in length of the nanowire, as this results on the projection in X towards the CCD detector we estimate a tilt on the nanowire of 22.4 degrees. On this position (tilted 22 degrees) the OL is turned ON to its maximum value to induce a direction towards the bottom of the wire. The lens is turned off and the wire is tilted back to zero so the holograms can be recorded (Figure S2c), the wire is tilted now to -22 degrees and the procedure is repeated (Figure S2d).The change in magnetization can be observed on the vicinity of the nanowire as contrast change towards the opposite direction.



Figure S1. In situ magnetization reversal procedure to obtain to holograms with different magnetization direction.



Figure S2. Comparison of the original state of the nanowire and the resultant magnetic contribution after the in-situ magnetization process was performed.

Figure S2 let us compare the difference between the original state of the nanowire (before the in situ magnetization reversal procedure) and the corresponding magnetic contribution after we subtracted half the unwrapped phase images obtained after tilting and magnetizing. The contrast between regions (above and below the wire) is evident and a preferential value of the magnetic induction on the sides of the nanowires is observed.

Finally, a quantitative evaluation of the magnetic flux can be obtained from the reconstructed phase maps. The total phase shift across ($\Delta \phi$) the nanowire in the unwrapped phase image, is proportional to the total flux by:

$$M = \frac{\Delta \varphi \cdot \phi_0}{2\pi \cdot \mu_0 \cdot S}$$

Thus the magnetization can be evaluated where S, is the cross-section area of the nanowire, μ_0 is the magnetic permeability of the vacuum and ϕ_0 is the magnetic flux quantum (\hbar/e). The phase profile plotted in Figure 7e, is from the region marked in the unwrapped phase image of Figure 7c yielding a value of approximately 5.3 radians. Then, the magnetic phase shift across the nanowire is proportional to the magnetic flux enclosed by the interfering trajectories of the electron beam. Therefore, the magnetic flux density (B) was evaluated using the equation:

$$B_{\perp(x,y)} = \frac{\overline{h}}{et} \frac{\delta \phi_{M(x,y)}}{\delta x}$$

where, ħ is the reduced Planck constant, e the electron charge and t the local thickness. The measured phase gradient can be converted to magnetization (in Teslas).

Micromagnetic modelling

A cylindrical model representing a multisegmented nanowire of 40 nm diameter and 300 nm total length is shown in **Figure S3.** This model includes FeGa / Cu alternated sections of 11.0 and 6.0 nm length, respectively.



Figure S3. Cylindrical model for a FeGa/Cu multisegmented nanowire.

Micromagnetic calculations for determining the magnetization distribution were performed based on the dynamic magnetization description given by the Landau-Lifshitz Gilbert equation of motion and its time integration technique by means of finite/boundary element method [1,2]. Magnetic parameters for $Fe_{80}Ga_{20}$ alloy were used for micromagnetic calculations as follows: magnetocrystalline anisotropy constant, $K_1 = 0$, saturation magnetization, M_s , of 1.63 T [3] and exchange constant, A, of 1.69 x10⁻¹¹ J/m (estimated within the frame of mean field approximation). Complementary, Cu layers were considered diamagnetic. The magnetization reversal was simulated as follows: An initial magnetizing field H of +1.0 T was applied along the longitudinal nanowire axis in order to reach magnetic saturation towards +H; then H was progressively decreased until H = 0.0 T, followed by a gradual negative decrease up to H = -1.0 T.

The demagnetizing process beginning at the remanence state is illustrated in **Figure S4** as a series of snapshots of longitudinal profiles obtained at the middle section of the model, for variable *H* field values. Fig.S5a corresponds to the remanence magnetization (*H*=0.0 T). The onset of the coherent rotation towards –*H* direction occurs for *H* = --0.025 T, as indicated in red circles in Fig.S5b.This rotation occurs at the interface of FeGa/Cu segments, in a similar process resembling a nucleation controlled mechanism [4]. Areas with perpendicular orientation preserve their direction for decreasing –*H* up to -0.125 T (see below). For *H* = --0.075 T (Fig.S5c), most of the FeGa portions with previous axial direction are changing their direction towards –*H* (see areas circled in red), except at the very center of the model, where the original direction pointing to +*H* is still present. For *H* = --0.1 T (Fig.S5d), realignment of the magnetization within these FeGa segments in the direction of –*H* is almost completed (see areas circled in red) and, at the same time, the axially oriented magnetic moments begins to rotate towards –*H*. For *H* = --0.125 T (Fig.S5e), most of magnetization within the cylinder has rotated in the direction of –*H*, including the FeGa portions with perpendicular orientation. Further decrease of -*H* (up to -0.15 T) causes the beginning of gradual rotation of the vortex configuration towards –*H* (up to -0.375T), for which a majority axial orientation was observed for *H*= -0.575 T (Fig.S5g).



Figure S4. Simulated magnetization reversal process for the cylindrical FeGa/Cu micromagnetic model: a) Remnant state, b) H = -0.025 T, c) H = -0.075 T, d) H = -0.1 T, e) H = -0.125 T, f) H = -0.375 T, g) H = -0.575 T

Figure S5 presents the work flow to retrieve the phase information from the holograms using HoloWorks 5.0: a) Region of interest on TEM. b) Electron hologram, the field of view is 980 nm. c) Reference hologram. d) Complex image obtained from the inverse of the Fourier transform of the electron hologram where a circular mask is performed on one of the sidebands. e) Modulo 2π phase image from the complex image. f) Phase unwrapping algorithms are used to remove the 2π phase discontinuities. g) A script is used to correct tilting on the image to avoid any phase discontinuities. h) Gauss filtering performed to remove interference fringes.



Figure S5. Workflow to obtain unwrapped phase images from the electron holograms acquired. Two different zones were acquired at two different accelerating voltages (200 and 120KV).



Figure S6. Room temperature uncorrected magnetization parallel (blue) and perpendicular (green) to the nanowire axis vs. applied field hysteresis loops measured using vibrating sample magnetometry of the FeGa/Cu - AAO ensemble.

Figure S6. Displays the magnetization loops measured using vibrating sample magnetometry of the FeGa/Cu - AAO ensemble. As the alumina matrix is diamagnetic and the nanowire arrays may have large demagnetizing dipolar interactions which contribute to the negative slope of the magnetization at high fields in the figure.

References

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