Spin-Wave Quantization in Ferromagnetic Nickel Nanowires

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The dynamical properties of uniform two-dimensional arrays of nickel nanowires have been investigated by inelastic light scattering. Multiple spin waves are observed that are in accordance with dipole-exchange theory predictions for the quantization of bulk spin waves. This first study of the spin-wave dynamics in ferromagnetic nanowire arrays reveals strong mode quantization effects and indications of a subtle magnetic interplay between nanowires. The results show that it is important to take proper account of these effects for the fundamental physics and future technological developments of magnetic nanowires.

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Regular arrays of magnetic submicrometer-size dots and wires are currently attracting great interest due to their interesting physics and potential applications in magnetic sensors and memory devices [1,2], and their static properties have been studied extensively [3–7]. Their dynamical properties are of fundamental importance in evaluating the time scale of the magnetization reversal process, which governs the writing time in magnetic storage devices [8–10]. The low-energy magnetic excitations or spin waves (SWs) that define the time scale are best investigated via Brillouin (inelastic) light scattering (BLS) [11]. Only surface SWs have been observed so far by BLS in deep-etched micron-size magnetic circular dots and rectangular wires [12–15]. Here we employ BLS to study SWs in highly ordered arrays of ferromagnetic nickel nanowires, fabricated by self-assembly in Al_2O_3 templates following Nielsch *et al.* [16,17], and analyze the results by adapting a recent dipole-exchange theory for cylindrical nanowires [18]. Our novel results reveal three SWs whose frequencies sharply increase with decreasing diameter. The theory shows that these modes are caused by the radial quantization of bulk SWs. Besides its fundamental significance for the physics of nanomagnets, such quantization and also interwire coupling are factors that will strongly influence nanomagnetic device performance.

The two-dimensional arrays of nickel nanowires were prepared in the following way. Hexagonally ordered porous Al_2O_3 templates were first fabricated on aluminum substrates using a two-step electrochemical anodization process [19]. By means of pulsed electrodeposition, the resulting high-aspect-ratio porous alumina templates were homogeneously filled with nickel [16]. Scanning electron micrographs of the nanowire structures [17] revealed that they have a length of about 1 μm and a periodic wire spacing of 100 nm, with respective nanowire diameters of 30, 40, and 55 nm. Samples of nanowires with a 25 nm diameter and interwire separation of 65 nm were also obtained. Using superconducting quantum interference device magnetometry [17], the saturation magnetization, M_s, of the nickel nanowires was found to be 0.0480 T.

Brillouin spectra at room temperature were recorded in the 180° backscattering geometry using a (3 + 3)-pass tandem Fabry-Pérot interferometer equipped with a silicon avalanche diode detector, and the 514.5 nm line of an argon-ion laser. The laser light was incident on the top surface of each alumina membrane such that the surface normal, which is parallel to the symmetry axes of the nanowires, lies in the scattering plane. Measurements were made in *p-s* polarization configurations, with a typical data acquisition time of 8 h. Figure 1 shows a typical *p-s* polarized anti-Stokes Brillouin spectrum (which is similar to the Stokes spectrum) in zero applied field, recorded at an incident angle of 45°, of a 30 nm diameter nickel nanowire sample, which features three Brillouin peaks. Since they appear symmetrically, in the respective Stokes and anti-Stokes spectra, about the elastic peak and their positions shift on application of a magnetic field (see inset of Fig. 1), they are attributed to bulk SWs. The spectral peaks were fitted with a Lorentzian function, as shown in Fig. 1, and the variation of the fitted SW frequencies with the nanowire radius is displayed in Fig. 2. The inset of Fig. 1 shows the variation of the SW frequencies when a magnetic field is applied transverse to the axis of the nanowires. This field causes a canting of the magnetization

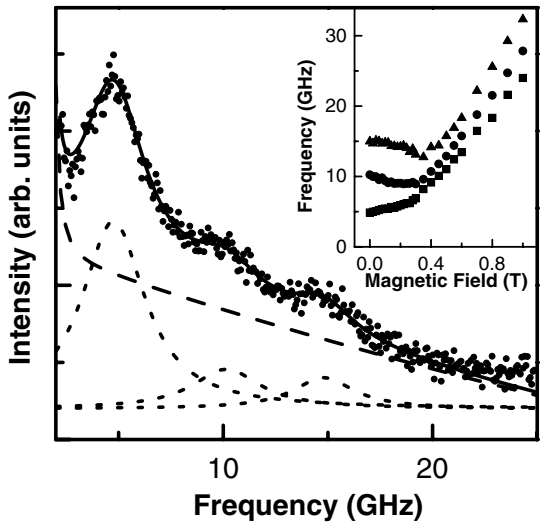

FIG. 1. Brillouin anti-Stokes spectrum of the 30 nm diameter nickel nanowire sample in zero applied magnetic field. Experimental data are denoted by dots. The peaks are due to the three bulk spin waves. The spectrum is fitted with Lorentzian functions (dotted curves) and a background (dashed curve); the full fitted spectrum is shown as a solid curve. The inset shows the bulk spin-wave frequencies versus applied magnetic field for the same sample.

direction, which acquires a component in the transverse direction (for which the demagnetizing field is $2\pi M_s \approx 0.30$ T). It is noticeable that there is a different behavior for the SW frequencies above and below this approximate field value.

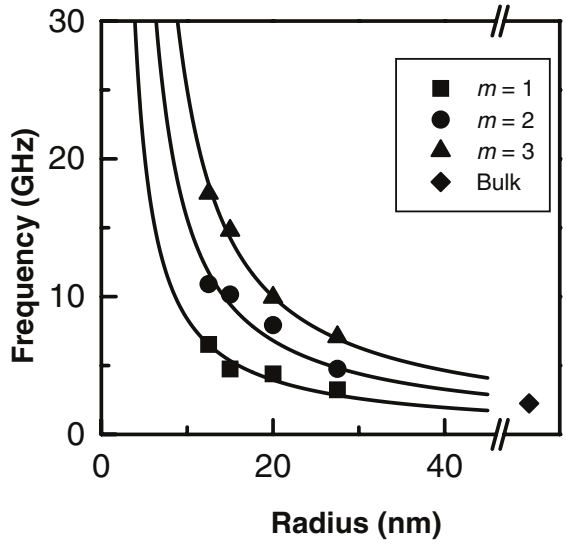

FIG. 2. Variation of bulk spin-wave frequencies with nanowire radius in zero applied magnetic field. The experimental data points, for each radius, correspond to the three values of the azimuthal quantum number m. The solid curves represent the respective best fits of the experimental data with Eq. (4). The diamond denotes the frequency of the bulk spin wave for bulk nickel [20].

The theory of Arias and Mills [18] was formulated for dipole-exchange SW excitations in ferromagnetic nanowires of circular cross section, where the magnetization is parallel to the axis of the wire. In this theory the general form of the magnetic scalar potential within the nanowire, as obtained from the Bloch equation of motion with dipole field terms and exchange terms included, can be expressed in cylindrical coordinates as

$$\Phi_M(r, \phi, z) = J_m(qr)\exp(im\phi)\exp(ikz). \quad (1)$$

Here k is the component of the SW wave vector along the z direction (parallel to the axis of a nanowire), and $J_m(qr)$ is a Bessel function of order m ($= 1, 2, 3, \ldots$). The complex variable q is found from the roots of a sixth-order polynomial [18]. In the case of zero applied magnetic field, we find that this can be factorized as

$$(q^2 + k^2)[Dq^2(Dq^2 + 4\pi M_s + 2Dk^2) + (Dk^2)^2 - \omega^2] = 0, \quad (2)$$

where D is the exchange stiffness, M_s is the saturation magnetization, and ω is the SW frequency. The bulk standing modes of the nanowires correspond to real values of q (which has the role of a SW radial wave vector component) and thus correspond to SW frequencies that are given by

$$\omega = [Dq^2(Dq^2 + 4\pi M_s + 2Dk^2) + (Dk^2)^2]^{1/2}. \quad (3)$$

The full theory [18] requires taking into account numerically the "mixing" of the bulk SWs with the surface SWs, which correspond to imaginary values of q, together with boundary conditions. Here we modify this approach to obtain an approximate analytic theory applicable for small and large pinning [11], for which the radial function $J_m(qr)$ will have antinodes and nodes, respectively, at $r = R$ (the radius of a nanowire). Denoting $a_m = q_m R$, the lowest three values are $a_1 = 1.84$, $a_2 = 3.05$, and $a_3 = 4.20$ for $m = 1$, 2, and 3, respectively, in the case of small pinning. The corresponding numbers for large pinning are $a_1 = 3.83$, $a_2 = 5.14$, and $a_3 = 6.38$.

The expression for ω can be further simplified if $k^2 \ll (a_m/R)^2$ (see discussion below):

$$\omega = \left\{D\left(\frac{a_m}{R}\right)^2\left[D\left(\frac{a_m}{R}\right)^2 + 4\pi M_s\right]\right\}^{1/2}. \quad (4)$$

The above equation is used to fit our results with D as a parameter and M_s fixed at the measured value of 0.0480 T [17]. Figure 2 shows that the fit thus obtained is good, in the case of small pinning, and yields $D = 3.13 \times 10^{-14}$ T cm^2, which is some 11 times smaller than that obtained by Sandercock and Wettling [20] in a BLS study of SWs in bulk nickel. The reason for this discrepancy may be associated with the different sample geometry and direction of the SW wave vector (compared to Ref. [20]) or, more likely, due to the long-range dipolar fields of the surrounding nanowires in the array. The fit to the experimental data is poor for large pinning, thus implying that the surface anisotropies in the nanowire samples are small.

Our $k = 0.041\ \mathrm{nm}^{-1}$ and therefore $k^2 = 1.6 \times 10^{-3}\ \mathrm{nm}^{-2}$. Now $(a_m/R)^2$ ranges from 4.48×10^{-3} to $0.11\ \mathrm{nm}^{-2}$ and therefore $k^2 \ll (a_m/R)^2$, thus justifying our use of Eq. (4). A measure of the relative importance of the exchange to dipolar coupling on a SW at wave number k is given by $\rho \equiv (Dk^2/4\pi M_s)^{1/2}$ (see, e.g., Ref. [21]). In the case of our nickel nanowires $\rho = 0.093$, which is small, but not sufficiently small that exchange effects may be neglected. Another measure is the exchange length, defined as ρ/k, which has the value of 2.27 nm for the nickel nanowires. The range of radii R studied, 12.5 to 27.5 nm, is larger than the exchange length, but not by a very large factor. This clearly indicates that a dipole-exchange theory [18] is necessary for the analysis of SWs in small-size nickel nanowires, and that the earlier magnetostatic theories [21,22] are inadequate because exchange effects are ignored.

In conclusion, we report the first observation by BLS of multiple SWs in uniform arrays of nickel nanowires. Our analysis, on the basis of a dipole-exchange theory, indicates that the discrete modes observed are a consequence of the quantization of bulk SWs due to confinement by the small cross section of the nanowires. Previously, BLS has only been employed to study surface SW confinement in surfaces patterned with micrometer-size magnetic platelets and strips [12]. By contrast, interesting new bulk effects arise in the present case due to the extremely anisotropic geometry of the long nanometer-size cylinders. Such a geometry is envisaged for the next generation of perpendicular magnetic storage media based on metallic nanowire arrays. Besides the spin reversal dynamics in single magnetic nanowires, the interactions between the nanowires indirectly evidenced here are also of great importance at high packing density. Future developments in quantum nanomagnetic data storage technology will depend on a detailed understanding of the physical nature and magnitude of intra- and interwire magnetic interactions.

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