Self-similar Magnetoresistance of Fibonacci Ultra-Thin Magnetic Films

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We study numerically the magnetic properties (magnetization and magnetoresistance) of ultrathin magnetic films (Fe/Cr) grown following the Fibonacci sequence. We use a phenomenological model which includes Zeeman, cubic anisotropy, bilinear and biquadratic exchange energies. Our physical parameters are based on experimental data recently reported, which contain biquadratic exchange coupling with magnitude comparable to the bilinear exchange coupling. When biquadratic exchange coupling is sufficiently large a striking self-similar pattern emerges.

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The discovery of quasicrystals in 1984 [1] aroused a great interest, both theoretically and experimentally, in quasiperiodic systems. One of the most important reasons for that is because they can be defined as an intermediate state between an ordered crystal (their definition and construction follow purely deterministic rules) and a disordered solid (many of their physical properties exhibit an erratic-like appearance) [2]. On the theoretical side, a wide variety of particles, namely electrons [3], phonons [4], plasmon-polaritons [5], spin waves [6], etc, have been studied. A quite complex fractal energy spectrum, which can be considered as their basic signature, is a common feature of these systems. On the experimental side, the procedure to grow quasiperiodic superlattices became standard after Merlin et al. [7], who reported the realization of the first quasiperiodic superlattice following the Fibonacci sequence by means of molecular beam epitaxy (MBE).

Parallel to these developments in the field of quasicrystals, the properties of magnetic exchange interactions between ferromagnetic films separated by non-magnetic spacers have been also widely investigated [8]. The discovery of physical properties like antiferromagnetic exchange coupling [9], giant magnetoresistance (GMR) [10], oscillatory behavior of the exchange coupling [11], and biquadratic exchange coupling (BEC) [12], made these films excellent options for technological applications and attractive objects of research. For example, GMR in magnetic films has been widely considered for applications in information storage technology [13].

It is known that GMR also occurs in non-periodic granular systems, like Cu-Co alloys consisting of ultrafine Co-rich precipitate particles in Cu-rich matrix [14]. Due to the fact that precipitate particles of these heterogeneous alloys have an average diameter and an average spacing similar to magnetic films, the origin of GMR in granular systems is also similar to the one found in magnetic films [15]. Therefore, quasiperiodic systems which present magnetoresistive properties can be a first step for a better understanding of magnetoresistance in granular systems. On the other hand, from a technological point of view (as we will show later in this letter) the BEC associated with quasiperiodicity permit us to control magnetic field regions, where magnetoresistance remains almost constant before saturation.

The aim of this work is to investigate the influence of quasiperiodicity on the magnetic properties of ultrathin magnetic films. In particular, we are interested in Fe/Cr(100) structures, which follow a Fibonacci sequence, whose experimental magnetic parameters were recently reported by Rezende et al. [16].

A Fibonacci structure can be grown experimentally by juxtaposing two building blocks A and B following a Fibonacci sequence. In our specific case we choose Fe as the building block A and Cr as the building block B. A Fibonacci sequence $S_N$ is generated by appending the $N−2$ sequence to the $N−1$ one, i.e., $S_N = S_{N−1}S_{N−2}$ ($N ≥ 2$). This construction algorithm requires initial conditions which are chosen to be $S_0 = B$ and $S_1 = A$. The Fibonacci generations are: $S_0 = [B]$, $S_1 = [A]$, $S_2 = [AB]$, $S_3 = [ABA]$, etc. Therefore, the well known trilayer Fe/Cr/Fe is the magnetic counterpart of the third Fibonacci generation (A/B/A). We remark that only odd Fibonacci generations have a magnetic counterpart, because they start and finish with an A (Fe) building block. Fig. 1 shows schematically the third and fifth Fibonacci generations and their magnetic counterpart, where $t$ is the thickness of a single Fe layer (single Cr layer). It is important to note a double Fe layer whose thickness is $2t$ in the fifth generation corresponding to a double letter A. It is easy to show that the quasiperiodic magnetic films, for any Fibonacci generation, will be composed by single Cr layers, single Fe layers and double Fe layers.

We consider the ferromagnetic films with magnetization in the plane $xy$ and take the $z$ axis as the growth
Electrons form a free-electron gas, which has been shown that the GMR varies linearly with \( \cos(\Delta \theta) \), and that it is responsible for the GMR effect [15]. It has been accepted as responsible for the GMR effect [16].

The thickness and the angular orientation of the magnetic layers are critical in determining the magnetic properties. The very strong demagnetization field, generated by tipping the magnetization out of plane, will suppress any tendency for the magnetization to tilt out of plane. The global behavior of the system is well described by a simple theory in terms of the magnetic energy per unit area [13], i.e.,

\[
E_T = E_Z + E_{bl} + E_{bq} + E_a,
\]

where \( E_Z \) is the Zeeman energy, \( E_{bl} \) is the bilinear energy, \( E_{bq} \) is the biquadratic energy and \( E_a \) is the cubic anisotropy energy. It is usual to write the total magnetic energy in terms of experimental parameters (or effective fields) of each interaction,

\[
\frac{E_T}{tM_S} = \sum_{i=1}^{n} \left( \frac{t_i}{t} \right) \left\{ -H_0 \cos(\theta_i - \theta_H) + \frac{1}{8} H_a \sin^2(2\theta_i) \right\} + \sum_{i=1}^{n-1} \left\{ -H_{bl} \cos(\theta_i - \theta_{i+1}) + H_{bq} \cos^2(\theta_i - \theta_{i+1}) \right\},
\]

where \( t \) is the thickness of a single Fe layer and we assume \( M_i = M_S \). \( H_{bl} \) is the conventional bilinear exchange coupling field which favors antiferromagnetic alignment (ferromagnetic alignment) if negative (positive). We are concerned here to the case \( H_{bl} < 0 \) because magnetoresistive effects occur only for this case. \( H_{bq} \) is the BEC field, which is responsible for a 90° alignment between two adjacent magnetizations and is experimentally found to be positive [12]. \( H_a \) is the cubic anisotropy field which renders the (100) direction an easy direction. \( H_0 \) is the external in-plane magnetic field and \( \theta_H \) is its angular orientation. From now on we consider \( \theta_H = 0 \), which means that the magnetic field is applied along the easy axis. The thickness and the angular orientation of the \( i \)-th Fe layer are given by \( t_i \) and \( \theta_i \), respectively.

The equilibrium positions of the magnetizations \( \{\theta_i\} \) are numerically calculated by minimizing the magnetic energy given by (2). It should be remarked that it has proved difficult for us to generate accurately configurations for larger structures, mainly when the BEC is strong [13]. However, we got results in sufficiently large generations to infer important informations about the effect of the quasiperiodicity.

Theoretically, the spin-dependent scattering is accepted as responsible for the GMR effect [13]. It has been shown that GMR varies linearly with \( \cos(\Delta \theta) \) when electrons form a free-electrons gas, i.e., there is no barriers between adjacent films [13]. Here, \( \Delta \theta \) is the angular difference between adjacent magnetizations. In metallic systems like Fe/Cr this angular dependence is valid and once the set \( \{\theta_i\} \) is found, we obtain normalized values for magnetoresistance, i.e,

\[
R(H_0)/R(0) = \sum_{i=1}^{n-1} \left[ 1 - \cos(\theta_i - \theta_{i+1}) \right]/2(n-1),
\]

where \( R(0) \) is the resistance at zero field.

Now we present numerical calculations for the magnetization and the magnetoresistance curves for Fibonacci ultra-thin magnetic films. The physical motivation for that is because the Fibonacci quasiperiodic structure can exhibit magnetic properties not found in the periodic case [13], and the long range correlations induced by the construction of this system are expected to be reflected somehow in the magnetoresistance curves. We have considered physical parameters based on realistic values of the magnetic coupling fields, whose experimental data were recently reported [13]. We assume the cubic anisotropy field \( H_a = 0.5 \) kOe, corresponding to Fe(100) with \( t > 30 \) Å growth by sputtering [8]. We choose the bilinear and the biquadratic fields, \( H_{bl} \) and \( H_{bq} \), such that their values lie in three regions of interest: i) close to the region of first antiferromagnetic-ferromagnetic transition where \( H_{bl} \) is moderate [13]; ii) near to the maximum of first antiferromagnetic peak, where \( H_{bl} \) reach its maximum value [13]; and iii) in the second antiferromagnetic peak, where \( H_{bl} \) is small and equal to \( H_{bq} [13] \).

In Fig. 2 we show the curves of the normalized magnetization and magnetoresistance versus the magnetic field, for the third Fibonacci generation (corresponding to the Fe/Cr/Fe trilayer). We assumed \( H_{bl} = -0.15 \) kOe and \( H_{bq} = 0.05 \) kOe (\( |H_{bl}| > H_{bq} \)). These parameters correspond to a realistic sample with Cr thickness equal to 15 Å. From there one can identify two first order phase transitions at \( H_1 \sim 100 \) Oe and \( H_2 \sim 220 \) Oe. Also there are three magnetic phases presented: i) an antiferromagnetic phase \( (H_0 < 100 \) Oe); ii) a 90° phase \( (100 \) Oe \( < H_0 < 220 \) Oe); iii) and a saturated phase \( (H_0 > 220 \) Oe). We remark that our numerical calculations indicate that a first order phase transition occurs when \( H_a > 2(|H_{bl}| + 2H_{bq}) \). Since the transition magnetic fields are the same for both the magnetization and the magnetoresistance, from now on we concentrate our discussion on the magnetoresistance curves, because it is easier to investigate their self-similar pattern.

Fig. 3 shows the normalized magnetoresistance curves for the fifth(a) and seventh(b) Fibonacci generations with the same experimental parameters considered in Fig. 2. In Fig. 3(a) we can identify four first order phase transitions, where each one is due to a 90° jump of magnetization. This behavior is always displayed when the BEC is present in the magnetic energy. Previous works on phase diagrams have looked carefully the origin and features of the so-called 90° phase [17][14]. For the seventh generation there are eight first order phase transitions and nine magnetic phases are present from the antiferromagnetic phase \( (H_0 < 38 \) Oe) to the saturated one \( (H_0 > 440 \) Oe). Note a clear self-similar pattern of magnetoresistance curves by comparing Fig. 2 and Fig. 3, i.e., the pattern of the trilayer Fe/Cr/Fe is always present in the next generations. On the contrary, when \( H_{bl} = -1.0 \) kOe and \( H_{bq} = 0.1 \) kOe (\( |H_{bl}| > H_{bq} \)), which correspond to...
to a sample with Cr thickness equal to 10 Å, the self-similarity is not observed, as it is shown in Fig. 4. For this set of parameters, the majority of phase transitions are of second order and we have found numerically that this occurs when $H_d < 2(|H_{bl}| + 2H_{bq})$. However, when the ratio between $H_{bq}$ and $H_{bl}$ is increased ($|H_{bl}| = H_{bq} = 35$ Oe), we observe again a striking self-similar pattern (see Fig. 5), where each new transition occurs for a value of magnetic field which is about a half of the previous one. For this set of parameters the magnetoresistance is approximately 1/2 its value at zero magnetic field, because the magnetizations of the adjacent Fe films are nearly perpendicular to each other due to the strong biquadratic field. For the third generation, Fig. 5(a), there is only one transition at $H_1 \sim 70$ Oe and two magnetic phases: a 90° phase at $H_0 < 70$ Oe and a saturated phase at $H_0 > 70$ Oe [19]. In the fifth generation, Fig. 5(b), there are two transitions at $H_1 \sim 70$ Oe and $H_2 \sim 140$ Oe, respectively. For the seventh generation, as one can see from Fig. 5(c), there are three transitions at $H_1 \sim 35$ Oe, $H_2 \sim 70$ Oe and $H_3 \sim 140$ Oe.

From the numerical results above discussed, we can infer that the magnetoresistance exhibits a self-similar behavior when: (a) $H_{bq}$ is comparable to $H_{bl}$, and (b) there is a first order phase transition (see Fig. 3 and Fig. 5). A possible explanation for that is because the BEC reinforces the quasiperiodic order, which is responsible by the self-similarity in quasiperiodic systems. This is an unexpected effect of this unusual exchange coupling and, as far as we know, this is the first system which presents magnetoresistance with self-similar properties. Besides, from a technological point of view, magnetoresistance with almost constant regions (Fig. 3 and Fig. 5) opens new perspectives in information storage technology by the possibility of a recording system with more than two states. Certainly, Fibonacci ultra-thin magnetic films can be realized experimentally following the procedures of ref.[12] or [20] to grow the samples.

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FIG. 1. The third and fifth Fibonacci generations and their magnetic counterpart.

FIG. 2. Magnetization (a) and magnetoresistance (b) versus magnetic field for the third Fibonacci generation with $H_{bl} = -150$ Oe and $H_{bq} = 50$ Oe. In Fig. 2(a) the arrows indicate the relative positions of the magnetizations in each phase.

FIG. 3. Magnetoresistance for the fifth (a) and seventh (b) Fibonacci generations with the same parameters of Fig. 2. In Fig. 3(a) the relative positions of magnetizations are indicated by the arrows, and the Fe double layer is indicated by the bigger arrow.

FIG. 4. Magnetoresistance for the third (a), fifth (b) and seventh (c) Fibonacci generations with $H_{bl} = -1.0$ kOe and $H_{bq} = 0.1$ kOe.
FIG. 5. Magnetoresistance for the third (a), fifth (b) and seventh (c) Fibonacci generations with $|H_{bl}| = H_{bq} = 35$ Oe, which correspond to a sample with Cr thickness equal to 25 Å. Note a striking self-similar pattern. In Fig. 5(a) and Fig. 5(b) the arrows indicate the relative positions of the magnetizations in each phase.