Influence of the biquadratic interlayer coupling in the specific heat of Fibonacci magnetic multilayers

C.G. Bezerra\textsuperscript{a}, E. L. Albuquerque \textsuperscript{b} * and M.G. Cottam\textsuperscript{a}

(a) Department of Physics and Astronomy, University of Western Ontario, London, Ontario N6A 3K7, Canada.
(b) Departamento de Física, Universidade Federal do Rio Grande do Norte, 59072-970, Natal-RN, Brazil.

Abstract

A theoretical study of the specific heat $C(T)$ as a function of temperature in Fibonacci magnetic superlattices is presented. We consider quasiperiodic structures composed of ferromagnetic films, each described by the Heisenberg model, with biquadratic and bilinear coupling between them. We have taken the ratios between the biquadratic and bilinear exchange terms according to experimental data recently measured for different regions of their regime. Although some previous properties of the spin wave specific heat are also reproduced here, new features appear in this case, the most important of them being an interesting broken-symmetry related to the interlayer biquadratic term.

PACS: 05.20-y; 61.43.Hv; 61.44.Br; 75.30.Ds

Keywords: Quasicrystals; Spin waves; Fractal behavior; Thermodynamical properties.

*Corresponding author, e-mail: eudenilson@dfte.ufrn.br
I. INTRODUCTION

The study of the properties of magnetic multilayers has been one of the most active fields in the last decade. The understanding of a number of new and intriguing results, such as the biquadratic exchange term in the free magnetic energy of the system, became an exciting challenge from both theoretical and experimental point of view. Until recently, it was found that the biquadratic exchange coupling was too small when compared to the bilinear term, but recent works have proved that it can play a remarkable role in the properties of magnetic multilayers.

It is known that the magnetic properties can depend strongly on the stacking pattern of the layers. In this respect, the physical properties of a new class of artificial material, the so-called quasiperiodic structures, became recently an attractive field of research. Quasiperiodic structures, which can be idealized as the experimental realization of a one-dimensional quasicrystal, are composed from the superposition of two (or more) building blocks that are arranged in a desired manner. They can be defined as an intermediate state between an ordered system (a periodic crystal) and a disordered one (an amorphous solid), despite the purely deterministic rules used to generate them. They also share a general property, which is perhaps their most characteristic one, namely a complex fractal spectra of energy, which can be considered as their basic signature. Their first experimental realization (in quasiperiodic GaAs-AlAs heterostructures) was carry out by Merlin and collaborators, and since then they have become a rapidly expanding object of theoretical and experimental research.

Recently, Tsallis and collaborators presented a model, based on the most well-known and simple deterministic fractal geometry (the triadic Cantor set), to study the thermodynamic properties. This set is obtained through the repetition of a simple rule: divide a given segment into three equal parts, and then eliminate the central one. They showed that the specific heat of such a system exhibits a very particular behavior: it oscillates log-periodically around a mean value that equals the fractal dimension of the spectrum.
Connections with natural spectra showing multifractal properties were also demonstrated afterwards. Furthermore, in a recent publication, we extended the model described by Tsallis et al., to study the specific heat, of real spin waves modes that propagate in quasiperiodic structures, with new features of the specific heat behavior. Throughout these papers, the classical Maxwell-Boltzmann statistics were used.

It is the aim of this work to investigate these properties even further, by studying the influence of the biquadratic exchange term in the specific heat spectra of spin waves in a magnetic quasiperiodic structure obeying the Fibonacci sequence. Spin-wave studies of magnetic systems, where there are contributions of bilinear and biquadratic exchange interactions, are quite recent. Such investigations renewed the interest in spin-wave analysis, particularly for non-collinear configurations, since it can give important information about the coupling parameters. The biquadratic coupling in real systems usually favors perpendicular alignment of the film magnetization, whereas the bilinear one favors both parallel or antiparallel configurations. The inclusion of both exchange terms leads to interesting physical properties.

The plan of this work is as follows. In Section II we present our general theoretical model, based on ferromagnetic films described by the Heisenberg Hamiltonian with a biquadratic exchange term, which are stacked following a Fibonacci sequence. The multifractal spectra then obtained will be used to determine the specific heat spectra for the Fibonacci quasiperiodic magnetic arrangement described in Section III. The numerical results and the discussion of their main features are presented in section IV.

II. THE MODEL

In this section we follow the spin-wave model of Ref. 20 for quasiperiodic magnetic superlattices with biquadratic exchange coupling. Specifically, we consider magnetic superlattices composed of $n_A$ layers of material $A$ and $n_B$ layers of material $B$. Materials $A$ and $B$ are simple cubic Heisenberg ferromagnets with bulk bilinear exchange constants $J_A$ and $J_B$, and
The spin quantum numbers of the magnetic moments in each material are $S_A$ and $S_B$, respectively. In our model we consider that within the bulk of materials $A$ and $B$ the magnetic moments interact only by the bilinear exchange couplings $J_A$ and $J_B$. However, across the interfaces, the magnetic moments interact by interfacial bilinear ($J_{bl}^I$) and biquadratic ($J_{bq}$) exchange couplings. The Hamiltonian for the bulk of each component is

$$H_\alpha = (-1/2)\sum_{i,j} J_\alpha \vec{S}_i \cdot \vec{S}_j - g\mu_B H_0 \sum_i S_i^z,$$  
(1)

where the sum in the first term is over nearest neighbors $j$, $H_0$ is the applied magnetic field in the $z$-direction, and $\alpha$ is equal to $A$ or $B$. Also, $g$ is the usual Landé factor and $\mu_B$ is the Bohr magneton. The spins of sites that are at the interfaces have a Hamiltonian which includes an additional biquadratic exchange coupling term, namely:

$$H_{bq} = \sum_{i,j} J_{bq} (\vec{S}_i \cdot \vec{S}_j)^2.$$  
(2)

It should be remarked that this term is responsible for the new effects found in the specific heat reported in this paper.

The spin wave dispersion relation in a magnetic superlattice is found by considering the wave solution inside each material and applying appropriate boundary conditions at the interfaces. The solutions for each material can be written as a linear combination of the positive- and negative-going solutions of the bulk case, i.e.,

$$S_i^+ = \{A_l \exp[i \vec{k}_A \cdot (\vec{r} - \vec{r}_{lA})] 
+A_l' \exp[-i \vec{k}_A \cdot (\vec{r} - \vec{r}_{lA})]\} \exp(-i\omega t)$$  
(3)

in component A, cell $l$, and

$$S_i^+ = \{B_l \exp[i \vec{k}_B \cdot (\vec{r} - \vec{r}_{lB})] 
+B_l' \exp[-i \vec{k}_B \cdot (\vec{r} - \vec{r}_{lB})]\} \exp(-i\omega t)$$  
(4)

in component B, cell $l$. 

4
These solutions are linked together using the equation of motion for the operator $S_i^+ = S_i^x + iS_i^y$, which for a site $i$ at an interface and after using the RPA approximation, is (see Ref. 20 for details)

$$
\hbar \frac{\partial}{\partial t} S_i^+ = g\mu_B H_0 S_i^+ + \sum_{n.n.} J_\alpha (S_j S_i^+ - S_i S_j^+) \\
+ \sum_{n.n.} J_{bq} \{ [S_i^2 (2S_j - 1) - S_i (S_j - 1)] S_j^+ \\
- [S_j^2 (2S_i - 1) - S_j (S_i - 1)] S_i^+ \}.
$$

(5)

The boundary conditions, after a tedious but straightforward calculation, can be written in a matrix form like

$$
M_A \begin{bmatrix} A_l \\ A'_l \end{bmatrix} = N_B \begin{bmatrix} B_l \\ B'_l \end{bmatrix},
$$

(6)

and

$$
M_B \begin{bmatrix} B_l \\ B'_l \end{bmatrix} = N_A \begin{bmatrix} A_{l+1} \\ A'_{l+1} \end{bmatrix}.
$$

(7)

The explicit form of the above matrices can be found elsewhere. Therefore, the explicit relation between the $lth$ and $(l+1)th$ unit cell amplitudes is

$$
\begin{bmatrix} A_{l+1} \\ A'_{l+1} \end{bmatrix} = N_A^{-1} M_B N_B^{-1} M_A \begin{bmatrix} A_l \\ A'_l \end{bmatrix},
$$

(8)

Here $T = N_A^{-1} M_B N_B^{-1} M_A$ is a transfer matrix. This equation, combined with the Bloch ansatz yields

$$
[T - \exp(iQD)] \begin{bmatrix} A_l \\ A'_l \end{bmatrix} = 0.
$$

(9)
The analogous relation between \((A_{l-1}, A'_{l-1})\) and \((A_l, A'_l)\) combined with equation (9) yields

\[
\cos(QD) = (1/2) \text{Tr}[T].
\]

(10)

Here \(Q\) is the Bloch wavevector of the collective excitation, and \(D\) is the size of the superlattice unit cell. Equation (10) follows from the fact that \(T\) is an unimodular 2x2 matrix, and it describes the bulk modes of spin waves in a magnetic superlattice. Once we know the form of the transfer matrix \(T\), the bulk spin wave spectrum is determined.

Let us briefly review the quasiperiodic sequence considered in this work. First we recall the definition of a substitution sequence. Take a finite set \(\xi\) (here \(\xi = \{A, B\}\)) called an alphabet and denote by \(\xi^*\) the set of all words of finite length that can be written in this alphabet. Now let \(\zeta\) be a map from \(\xi\) to \(\xi^*\) by specifying that \(\zeta\) acts on a word by substituting each letter (e.g. A) of this word by its corresponding image \(\zeta(A)\). A sequence is then called a substitution sequence, if it is a fixpoint of \(\zeta\), i.e. if it remains invariant when each letter in the sequence is replaced by its image under \(\zeta\). One of the most famous substitution sequences is the so-called Fibonacci quasiperiodic sequence, whose substitution rule is \(A \rightarrow \zeta(A) = AB, B \rightarrow \zeta(B) = A\). This sequence can also be constructed by appending the \(n-2\)th generation to the \(n-1\)th one, i.e., \(S_n = S_{n-1}S_{n-2} (n \geq 2)\). This algorithm requires the initial conditions \(S_0 = B\) and \(S_1 = A\). Some of the first Fibonacci generations are,

\[
S_2 = [AB],\ S_3 = [ABA],\ S_4 = [ABAAB],\ etc.
\]

In a given generation \(S_n\), the total number of building blocks is given by the Fibonacci number \(F_n\), which is obtained by the relation \(F_n = F_{n-1} + F_{n-2}\), with \(F_0 = F_1 = 1\). Also, \(F_{n-1}\) and \(F_{n-2}\) are the numbers of building blocks \(A\) and \(B\), respectively. As the generation order increases \((n >> 1)\), the ratio \(F_n/F_{n-1}\) approaches \(\tau = (1 + \sqrt{5})/2\), an irrational number which is known as the golden mean. From an experimental perspective, a Fibonacci superlattice is grown juxtaposing the two magnetic materials \(A\) and \(B\) according to the above described sequence of letters. It can also be shown that the transfer matrices of consecutive Fibonacci generations are related by a convenient recursive relation.
\[ T_{s_n} = T_{s_{n-2}} \cdot T_{s_{n-1}}, \quad n \geq 2. \] (11)

It is easy to see that, from the knowledge of the transfer matrices \( T_{S_0} \) and \( T_{S_1} \), we can determine the transfer matrix for any generation and consequently the spin-wave dispersion relation. It should be remarked that the matrix \( T_{S_2} \) recovers the periodic case.

### III. SPECIFIC HEAT SPECTRA

The spin wave fractal spectra for the Fibonacci superlattices with biquadratic exchange coupling is shown in Fig. 1 for a fixed value of the in-plane dimensionless wavevector, namely \( k_x a = 2.0 \). We can clearly see the forbidden and allowed energies versus the Fibonacci generation number \( n \), up to their 10th generation, whose unit cell is composed of \( F_9 = 55 \) A’s and \( F_8 = 34 \) B’s building blocks. The number of allowed bands is equal to three times the Fibonacci number \( F_n \) of the correspondent generation. Notice that, as expected, for large \( n \) the allowed band regions get narrower and narrower and they have a typical Cantor set structure.

We address now the specific heat of the spectra shown in Fig. 1. The description below, which follows the lines of Ref. 17, is general and has been successfully applied to many other banded spectra (see also Refs. 23 and 24). In Fig. 1 each spectrum, for a fixed generation number \( n \), has \( m \) allowed continuous bands. We consider the level density within each band to be constant. The partition function for the \( n \)th generation is then given by:

\[ Z_n = \int_0^\infty \rho(\epsilon)e^{-\beta\epsilon}d\epsilon, \] (12)

Here \( \beta = 1/T \) (by choosing the Boltzmann’s constant \( k_B = 1 \)), and we take the density of states \( \rho(\epsilon) = 1 \). After a straightforward calculation we can write \( Z_n \) as

\[ Z_n = \frac{1}{\beta} \sum_{i=1,3,\ldots}^{2m-1} e^{-\beta \epsilon_i} [1 - e^{-\beta \Delta_i}]. \] (13)

Here the subscript \( n \) is the generation number, \( m \) is the number of allowed bands and \( \Delta_i = \epsilon_{i+1} - \epsilon_i \) is the difference between the top and bottom energy levels of each band.
The specific heat is then given by

\[ C_n(T) = \frac{\partial}{\partial T} [T^2 \frac{\partial \ln Z_n}{\partial T}], \tag{14} \]

which can be written as

\[ C_n(T) = 1 + \frac{\beta f_n}{Z_n} - \frac{g_n^2}{Z_n^2}, \tag{15} \]

with

\[ f_n = \sum_{i=1,3,...}^{2m-1} [\epsilon_i e^{-\beta \epsilon_i} - \epsilon_{i+1} e^{-\beta \epsilon_{i+1}}]. \tag{16} \]

and

\[ g_n = \sum_{i=1,3,...}^{2m-1} [\epsilon_i e^{-\beta \epsilon_i} - \epsilon_{i+1} e^{-\beta \epsilon_{i+1}}]. \tag{17} \]

Therefore, once we know the energy spectra of the spin waves which propagates in a given sequence’s generation of a quasiperiodic structure, we can determine the associated specific heat by using (15).

**IV. NUMERICAL RESULTS AND DISCUSSIONS**

In this section we present numerical results obtained for the specific heat of Fibonacci magnetic multilayers. In our calculations we have assumed the spin quantum numbers \( S_A = 1.0 \) and \( S_B = 1.5 \), and have taken values of the ratio between the interlayer biquadratic and bilinear exchange terms (\( R = J_{bq}/J_{bl} \)) in accordance with experimental data recently measured for different regions of the biquadratic and bilinear exchange couplings’ regime\[^{12}\].

Fig. 2 shows the log-log plot of the spin wave specific heat as a function of temperature in the low temperature regime and in the absence of the biquadratic term (\( R = 0 \)). We have taken the dimensionless common in-plane wavevector to be \( k_x a = 2.0 \). As we can see, there is an interesting harmonic oscillation of the specific heat, which is a log-periodic function of the temperature, i.e., \( C_n(T) = \alpha C_n(aT) \), where \( \alpha \) is a constant, and \( a \) an arbitrary number. These log-periodic oscillations can be traced back to the log-periodicity.
of the density of state’s spectral staircase. They resemble very much the shape of the devil’s staircase obtained from idealized Cantor sets. The number of oscillations depends on the generation number of the Fibonacci sequence: the bigger the system, the greater the number of oscillations. Most important, however, is the well defined even and odd parity spectra related to the generation number of the Fibonacci structure, with the amplitudes of the latter being bigger than the amplitudes of the former. They are due to the parity of the set of eigenvalues \( \epsilon_i \) used to calculate the partition function given by (13). These harmonic oscillations can be identified as the signature of the Fibonacci structure, and it has no counterpart in the idealized triadic Cantor set.

There is an important modification, however, when the biquadratic coupling is present \( (R \neq 0) \). By contrast with the spectra found in the previous work \(^{17}\) (but with other similarities remaining), the two different symmetrical profiles of oscillations are broken for different ratios between the biquadratic and bilinear terms. To reinforce this intriguing behavior, we show, in Figs. 3 and 4, the evolution of this broken-symmetry for \( R = 0.2 \) (where the even-mode symmetry starts to be broken) and 0.4 (where the odd-mode symmetry also starts to be broken), respectively. A possible explanation is that these different behaviors arise because the biquadratic exchange term reduces the effective coupling between the adjacent magnetic layers at the interface, e.g., as may be deduced by examining the overall coefficients of \( S_i^+ \) or \( S_j^+ \) in (5) (see also Ref. 20). Also the biquadratic exchange induces long range correlations that emphasize the quasiperiodicity of the system. In other words, it looks like these effects make the whole structure see better its quasiperiodicity, in effect increasing its degree of disorder as \( R \) increases! It turns out that the spin wave energy band structure is more disordered and, as a consequence, this is reflected in the specific heat curves, which do not have the well-defined standard profiles (actually the oscillations are non-harmonic!) found in the absence of the biquadratic term, as well as presented for other excitations.\(^{23,24}\) This argument is reinforced by previous works on the correlation lengths of magnetic systems exhibiting biquadratic exchange coupling (see, for example, Sørensen and Young\(^ {25}\)).
To summarize, we have proposed in this paper a realistic model to study the specific heat contribution from the energy spectra of real spin waves in quasicrystals of the Fibonacci type, stressing the important role played by the biquadratic exchange term. Certainly the theoretical predictions shown here can be tested experimentally, and we encourage experimentalists to carry out such studies.

Further investigations for the case of the specific heat due to propagation of mixed modes is currently under consideration, and we hope to present these results in a later publication.

Acknowledgements: We would like to thank the Brazilian Research Council CNPq and NSERC of Canada for financial support. One of us (ELA) also thanks the hospitality of the Department of Physics and Astronomy, University of Western Ontario, where his contribution to this work was made.
REFERENCES

1 A. Azevedo, C. Chesman, S.M. Rezende, F.M. de Aguiar, X. Bian and S.S.P. Parkin, Phys. Rev. Lett. 76 (1996) 4837.

2 C. Chesman, M.A. Lucena, M.C. de Moura, A. Azevedo, F.M. de Aguiar and S.M. Rezende, Phys. Rev. B 58 (1998) 101.

3 C.G. Bezerra, J.M. de Araújo, C. Chesman and E.L. Albuquerque, Phys. Rev. B 60 (1999) 9264.

4 C.G. Bezerra, J.M. de Araújo, C. Chesman and E.L. Albuquerque, J. Appl. Phys 89 (2001) 2286.

5 C. Janot, Quasicrystals, a primer (Oxford University Press, Oxford, 1993).

6 J.M. Luck, Phys. Rev. B 39 (1989) 5834.

7 F. Axel and H. Terauchi, Phys. Rev. Lett. 66 (1991) 2223.

8 M. Quilichini and T. Janssen, Rev. Mod. Phys. 69, 277 (1997).

9 C. G. Bezerra and E. L. Albuquerque, Physica A 245 (1997) 379.

10 M.S. Vasconcelos and E.L. Albuquerque, Phys. Rev. B 57 (1998) 2826.

11 D.H.A.L. Anselmo, M.G. Cottam and E.L. Albuquerque, J. Phys.: Condens. Matter 12 (2000) 1041.

12 M.S. Vasconcelos and E.L. Albuquerque, Solid State Commun. 117 (2001) 495.

13 R. Merlin, K. Bajema, R. Clarke, K.M. Mohanty and J.D. Axe, Phys. Rev. Lett. 57 (1986) 1157.

14 C. Tsallis, L.R. da Silva, R.S. Mendes, R.O. Vallejos and A.M. Mariz, Phys. Rev. E 56 (1997) R4922.

15 P. Carpena, A.V. Coronado and P.B-Galvan, Phys. Rev. E 61 (2000) 2281.
16 P. Carpena, A.V. Coronado and P.B-Galvan, Physica A 287 (2000) 37.

17 C.G. Bezerra, E.L. Albuquerque, A.M. Mariz, L.R. da Silva and C. Tsallis, Physica A 294 (2001) 415.

18 M. Maccio, M.G. Pini, P.Politi and A. Rettori, Phys. Rev. B 49 (1994) 3283.

19 J. Barnaš, Phys. Stat. Sol (b) 203 (1997) 221.

20 C.G. Bezerra and M.G. Cottam, Phys. Rev. B, to be published (2001).

21 S.M. Rezende, C. Chesman, M.A. Lucena, A. Azevedo, F.M. de Aguiar and S.S.P. Parkin, J. Appl. Phys. 84 (1998) 958.

22 N.S. Almeida and D.L. Mills, Phys. Rev. B 52 (1995) 13504.

23 P.W. Mauriz, E.L. Albuquerque and M.S. Vasconcelos, Phys. Rev. B, 63 (2001) 184203.

24 P.W. Mauriz, E.L. Albuquerque and M.S. Vasconcelos, Physica A 294 (2001) 402.

25 E.S. Sørensen and A.P. Young, Phys Rev. B 42 (1990) 754.
**Figure Captions**

1. Energy spectra of spin waves for the quasiperiodic Fibonacci structure when a biquadratic interlayer exchange term is present. Here we consider the dimensionless in-plane wavevector \( k_x a = 2.0 \).

2. Log-log plot of the specific heat versus temperature for the generation numbers of the Fibonacci quasiperiodic sequence in the absence of the biquadratic term: (a) even generation numbers; (b) odd generation numbers.

3. Same as Fig. 2, but for the ratio between the interlayer biquadratic and bilinear exchange terms \( R = 0.2 \). From this value the even symmetry starts to be broken.

4. Same as Fig. 2, but for the ratio between the interlayer biquadratic and bilinear exchange terms \( R = 0.4 \). From this value the odd symmetry also starts to be broken.
