Evidence of extended orientational order in amorphous Fe/Sm thin films

J. M. Ruiz* and X. X. Zhang
Departament de Fisica Fonamental, Facultat de Fisica, Universitat de Barcelona, Diagonal 647, 08028 Barcelona, Spain

C. Ferrater
Departament de Fisica Aplicada, Facultat de Fisica, Universitat de Barcelona, Diagonal 647, 08028 Barcelona, Spain

J. Tejada
Departament de Fisica Fonamental, Facultat de Fisica, Universitat de Barcelona, Diagonal 647, 08028 Barcelona, Spain
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Amorphous thin films of Fe/Sm, prepared by evaporation methods, have been magnetically characterized and the results were interpreted in terms of the random magnets theory. The samples behave as 2D and 3D random magnets depending on the total thickness of the film. From our data the existence of orientational order, which greatly influences the magnetic behavior of the films, is also clear.

INTRODUCTION

The purpose of this work is to study the properties of amorphous thin films of Fe/Sm under the random magnet theory and to investigate the orientational order in these materials. The term "random magnet" is used to define amorphous materials with random magetic anisotropy. The theory used to describe it was initiated by Imry and Ma[1] and later was developed mainly by Chudnovsky and co-workers[2–5] and Sellmyer and Nafis.[6] This theory has been successfully applied to other systems.[7–10]

The concept of amorphous material related to these random magnet systems needs to be made precise. We have to distinguish between the positional and the orientational order in a solid. The positional order is referred to the local arrangement of the atoms and is due to local interactions among them. We can characterize the range of the positional order by r_0, which corresponds to the average size of the perfect lattice. So, we can consider r_0 as the positional correlation length. Regions separated by $r > r_0$ must be approximated by different pieces of the perfect lattice. Each piece has a certain orientation in the space. The length at which these orientations are correlated determines the range of the orientational order R_o. In a perfect crystal, positional and orientational order are long range and related to each other. However, for a disordered system we can imagine a solid which has only short-range positional order but long-range correlations in the orientation of locally defined crystallographic axes $R_0 \gg r_0$. This idea was theoretically studied by Halperin and Nelson[11] and reported experimentally by Chudnovsky and Tejada[12] and others.[13]

The positional order may be determined by x-ray or electron diffraction methods. There is no experimental method, however, to observe directly the orientational order in an amorphous solid. A possibility to detect orientational order follows from the theory of random magnets.

In a crystalline ferromagnet, the strong exchange interaction between atomic magnetic moments tends to align them, but the possible directions of the resulting magnetization are determined by the anisotropy axes of the crystal. In an amorphous solid, however, one can talk only about local anisotropy axes. As a result of this random anisotropy the ferromagnetic correlation length, R_f, remains finite for $d < 4$, d being the dimensionality of the system.[1] The ferromagnetic correlation length R_f depends on the exchange, the strength of the random anisotropy, and the length R_o at which anisotropy axes are correlated. From the random magnet theory we can compute the effective exchange field $H_{ex} = 2A/M_o R_o$, M_o being the saturation magnetization and A the exchange constant (erg/cm). Using this parameter and taking into account that the field acting on individual moments is $H_{int} = 2A/M_o a^2$, a being the average distance between magnetic atoms, we can obtain the value of R_o comparing these two fields, as we will see below.

EXPERIMENTAL

Compositionally modulated thin films of samarium and iron were prepared by using two electron guns under high vacuum conditions. The pressure during the evaporation was always lower than 5×10^{-7} Torr. Kapton was the substate for the modulated structure. The samples correspond to the following composition:

Sample I: Ag(100 Å)[Fe(3 Å)/Sm(2 Å)]$_{x400}$ Ag(100 Å)

Sample II:

Cu(100 Å)[[Fe(3 Å)/Sm(2 Å)]$_{x5}$/Cu(100 Å)]$_{x16}$

Sample III: [Fe(18 Å)/Sm(6 Å)]$_{x45}$.

The substrate temperature during the growth process was 150, 260, and 25 °C, respectively. The evaporation rate of the materials was 0.5 Å/s, measured by a quartz crystal

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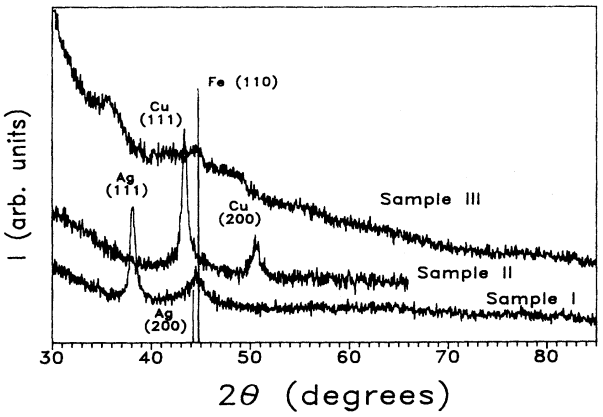

FIG. 1. X-ray diffraction patterns for the three samples.

oscillator. The structure of the samples was examined by x-ray diffraction and by electron diffraction. Magnetic properties were measured by a superconducting quantum interference device in the temperature range from 5 to 300 K at applied fields up to 5.4 T.

The structure of the samples was analyzed by x-ray diffraction (Cu $k\alpha$). The diffraction patterns are shown in Fig. 1. It was found that sample I is formed by amorphous SmFe but, nevertheless, if we change the substrate temperature during the growth process we observe the formation of polycrystalline α-Fe.[18] The diffraction pattern for sample II only shows the peaks corresponding to the crystalline Cu (used as a buffer and an isolating layer). Sample III does not show evidence of the presence of any well-defined crystalline component [only a broad and small peak in the x-ray pattern near the α-Fe (110) peak]. This sample was also observed by electron microscope (scanning electron microscope and transmission electron microscope) and tested by electron diffraction. The corresponding diffraction patterns show that the resultant structure is effectively amorphous. From these data, it is difficult to calculate the size of the nanocrystals which may form the samples, but we can estimate an upper limit for this size of about 40 Å. The presence of nanocrystals of higher size in the samples would be evidenced by sharper peaks in the position of the α-Fe (110) peak and, also, by the presence of diffraction spots in the nanodiffraction pattern.

RESULTS AND DISCUSSION

Magnetic properties of some amorphous ferromagnets are well described within the random anisotropy real-space model proposed by Chudnovsky and others.[2–6] The model assumes a ferromagnetic exchange interaction among the atoms of the alloy and a local random anisotropy which fluctuates from one magnetic atom to another one. This theory has been already tested for amorphous alloys of rare-earth–ferromagnetic materials with different preparation methods.[8,9,14,15] Following the notation of Chudnovsky and co-workers,[4,5] two characteristic fields can be defined: the anisotropy field $H_r = 2K/M_o$ (which coincides with the anisotropy field of a crystalline ferromagnet), and the exchange field $H_{ex} = 2A/M_o R_o^2$, where K is the anisotropy constant (erg/cm^3), A the exchange constant, and R_o corresponds to the distance over which the local anisotropy axes are correlated. As we will see, R_o could be much larger than the positional correlation length, and this will be a crucial characteristic of these materials.[16]

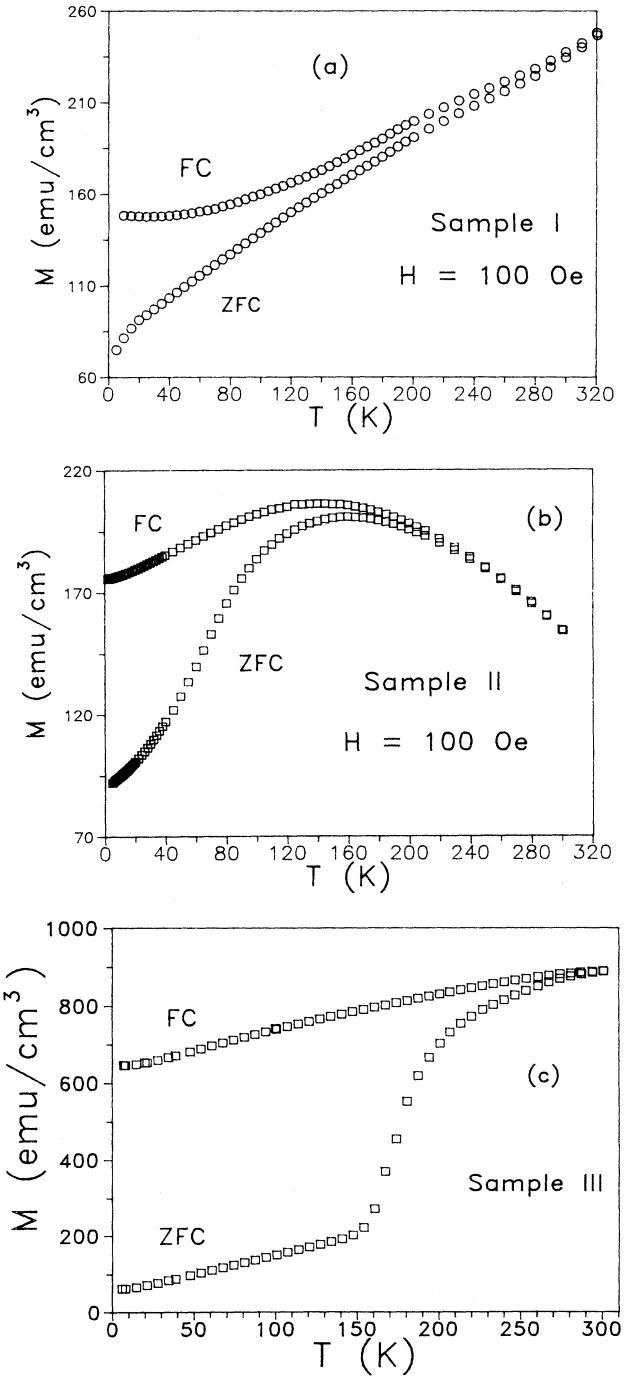

FIG. 2. (a)–(c) ZFC-FC processes. The applied field was 100 Oe for all the samples.

A. Low-field regime

When the samples are cooling down in zero field (ZFC process), the magnetic moments lay in a metastable disordered state, which has no net magnetization and which cannot be removed if we apply a low field. This state has been called correlated spin glass[2] (CSG). If the temperature is increased the magnetic moments tend to align towards the field (the stable state). When only slight deviations from the direction of the field are allowed we arrive at the reversible zone, which corresponds to the ferromagnet of wandering axes state. If the temperature is decreased keeping the field applied (FC process) then the magnetization freezes in a new direction due to the competition between the anisotropy, the external field, and the exchange interaction. In this case all the magnetic moments are frozen in their random directions but with a preferred orientation pointing to the field direction. This magnetic structure is called asperomagnet[17] and presents a nonzero net magnetization. So we can conclude that in the ZFC process our samples are in a CSG or correlated speromagnet state and, after the FC process, in an asperomagnetic state (see Fig. 2).

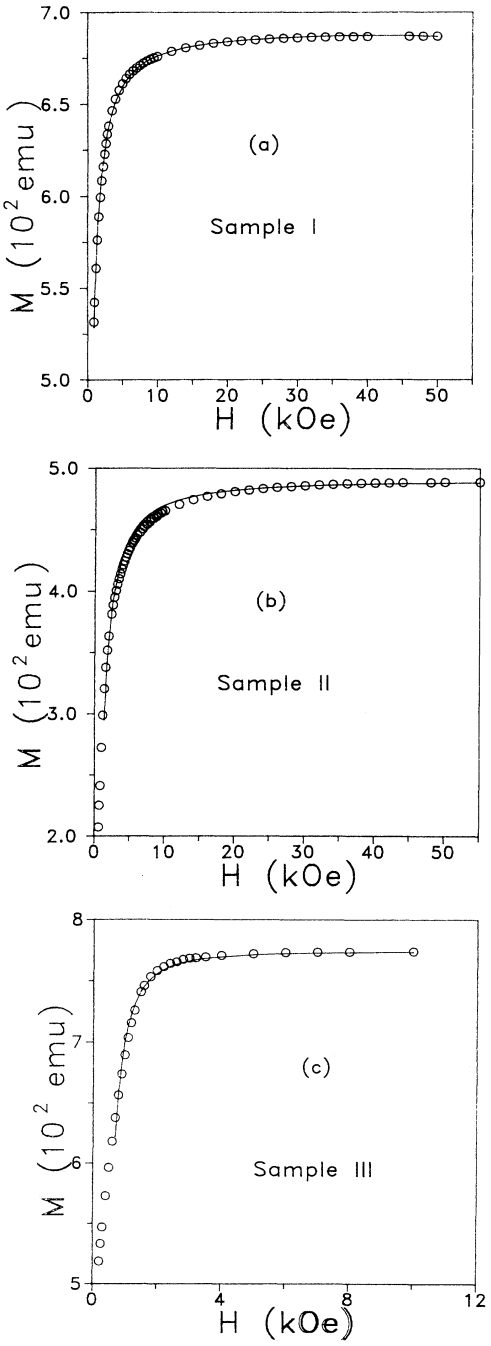

FIG. 3. (a)–(c) $M(H)$ curves at 5 K. The solid line corresponds to the fitting of the experimental values according to Eqs. (1) and (2). See text for detailed explanation.

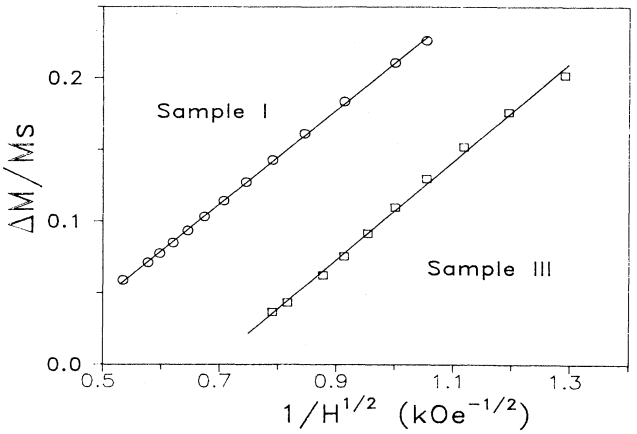

FIG. 4. Asymptotic behavior at $H \ll H_{ex}$ for samples I and III [see Eq. (3) in the text].

B. High-field regime

The magnetization process of a three-dimensional (3D) and a 2D random magnet follows the equations.[5,8]

$$\frac{\delta M}{M_o} = \frac{\Lambda^2}{60p} \int_0^\infty dx\, x^2 \exp(-px) C(x), \quad \text{(3D case)}, \qquad (1)$$

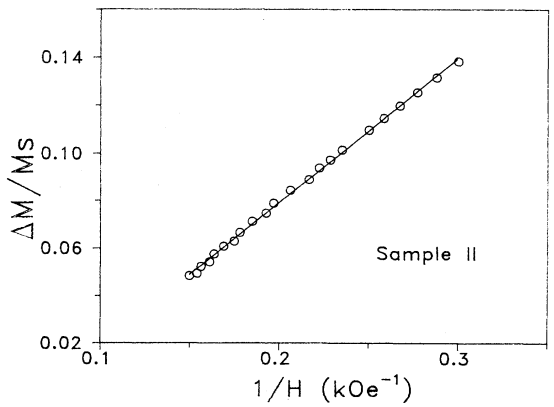

FIG. 5. Asymptotic behavior at $H \ll H_{ex}$ for sample II [see Eq. (3) in the text].

TABLE I. Calculated values of the parameters from the fitting of the $M(H)$ and $M(T)$ curves.

Sample	H_r (kOe)	H_{ex} (kOe)	Λ	D (meV Å^2)	Ra (Å)
I	7.5±0.5	9.4±0.8	0.80	116±20	60±8
II	11.5±0.7	20±2	0.56	64±12	30±5
III	1.5±0.1	3.0±0.1	0.50	250±45	150±20

$$\frac{\delta M}{M_o}=\frac{\Lambda^2}{32p}\int_0^\infty dx\, xK_1(px)C(x), \quad \text{(2D case)}, \tag{2}$$

where $\Lambda=H_r/H_{ex}$, $p^2=H/H_{ex}$, $C(x)$ is the correlation function of the anisotropy axes, $K_1(x)$ is the modified Hankel function of first order, and x is expressed in units of R_o. For these formulas we can deduce the asymptotic behaviors:

$$H \ll H_{ex}: \ \delta M \propto 1/\sqrt{H} \ \text{(3D case)}, \quad \delta M \propto 1/H \ \text{(2D case)}, \tag{3}$$

$$H \gg H_{ex}: \ \delta M \propto 1/H^2, \ \text{(3D and 2D cases)}. \tag{4}$$

Note that the qualitative difference between the 2D and 3D cases is only detectable in the low-field regime $H \ll H_{ex}$.

Analyzing the $M(H)$ curves for the three samples (see Fig. 3), we observe the asymptotic behaviors described above. Samples I and III exhibit a 3D behavior (Fig. 4) and sample II exhibits a 2D behavior (Fig. 5) in the low-field regime. It is also observed in the high-field regime $1/H^2$ for the three samples (Fig. 6). From the slopes of these adjustments (Fig. 6) we can estimate the anisotropy

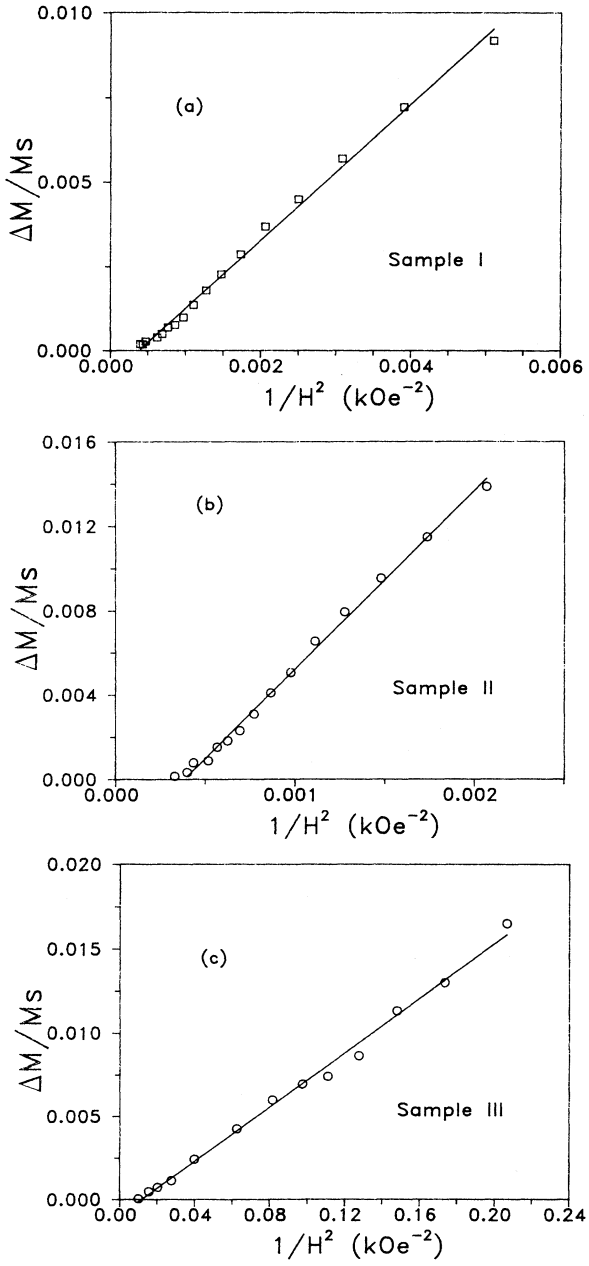

FIG. 6. (a)–(c) Asymptotic behavior at $H > H_{ex}$ for all three samples [see Eq. (4)].

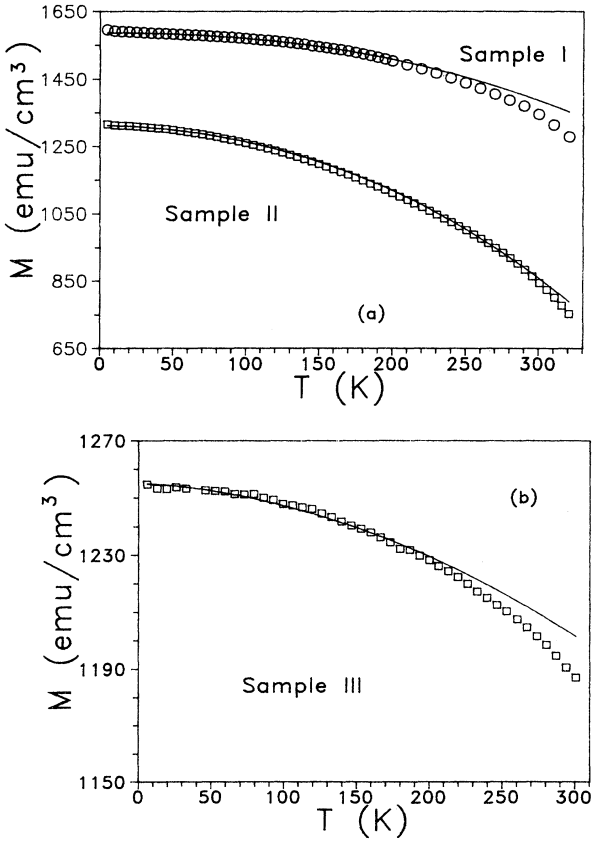

FIG. 7. (a) and (b) Variation of the saturation magnetization with temperature, $M(T)$ curves. The solid lines correspond to the spin-wave law [Eq. (5)]. The values of the calculated stiffness constants, D (meV Å^2), are listed in Table I.

fields H_r.[5,8] With the calculated values of H_r we fitted the $M(H)$ magnetization curves according to Eqs. (1) and (2). These equations are valid in the whole region of approach to saturation. Our fit procedure is similar to that used by Tejada *et al.* in Ref. 14 and we have also used a Gaussian correlation function $C(x)$ to compute the numeric integral. The values of H_{ex} with the corresponding errors are calculated from these fittings and they are listed in Table I. Note that the condition $H \gg H_{ex}$ to observe the $1/H^2$ term must be changed to $H > H_{ex}$ because we are not in the weak anisotropy limit $\Lambda = H_r/H_{ex} \ll 1$.

C. Orientational order

As we have said in the Introduction, in these random materials we have to distinguish between the positional order and the orientational order.

The range of the positional order (positional correlation length), r_0 can be determined by x-ray and electron diffraction methods. For our samples, the results of these techniques give us a maximum value for r_0 of about 40 Å. In spite of this, the correlation length of the orientational order, R_o, cannot be determined by direct methods. Fortunately, this parameter greatly influences the magnetic behavior of the samples and, as we will explain, it can be deduced from magnetic measurements. From the fitting of the magnetization curves of the samples, we have obtained the effective exchange field $H_{ex} = 2A/M_o R_o^2$ (see Table I), which depends on the R_o parameter. This field is different from the field acting on individual moments $H_{int} = 2A/M_o a^2$. If we know the value of H_{int} we will be able to calculate R_o. H_{int} can be obtained either from Mössbauer measurements or from the spin-wave spectra. Mössbauer spectroscopy is not suitable for these samples due to the low concentration of Fe atoms in front of the substrate, cover layers, and Sm atoms. The other way to obtain H_{int}, i.e., from the spin-wave spectra, is more adequate in this case. The variation of the saturation magnetization with temperature follows the spin-wave law (Fig. 7)

$$M(T) = M_o(1 - BT^{3/2} - AT^{5/2}) , \tag{5}$$

$$B = 2.612 \frac{g\mu_B}{M_o} \left[\frac{K_B}{4\pi D} \right]^{3/2} . \tag{5'}$$

From these data we can calculate the stiffness constant D of the samples. This parameter allows us to estimate the magnitude of the correlation length of the anisotropy axes R_o, which can be obtained from the formula[12]

$$R_o^2 = D/\mu_B H_{ex} , \tag{6}$$

where μ_B is the Bohr magneton. The values of D and the calculated R_o for all the samples are listed in Table I. It is important to notice that for these samples we have found that no positional order, i.e., crystalline order, were present over distances larger than ~40 Å. But now we find that the correlation length of the anisotropy axis R_o is about 60 Å for sample I, 30 Å for sample II, and 150 Å for sample III. Now we can conclude that in samples I and III, randomness produces the following hierarchy of scales:

$$a < r_0 < R_o < R_f , \tag{7}$$

where R_f is the ferromagnetic correlation length.

It is remarkable that for sample II we get a value of R_o of only about 30 Å which is of the same order as that estimated for r_o. This is due to the preparation method of the sample. The thicker Cu layers separating thin layers of Fe/Sm make impossible the formation of Fe/Sm blocks of a size higher than a few tenths of angstroms. In this sense, sample II can be considered as a control sample, in order to test the degree of validity of the mathematical calculations of the theory.

*Author to whom correspondence should be sent.

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