Partial phase transition and quantum effects in helimagnetic films under an applied magnetic field

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We study the phase transition in a helimagnetic film with Heisenberg spins under an applied magnetic field in the c direction perpendicular to the film. The helical structure is due to the antiferromagnetic interaction between next-nearest neighbors in the c direction. Helimagnetic films in zero field are known to have a strong modification of the in-plane helical angle near the film surfaces. We show that spins react to a moderate applied magnetic field by creating a particular spin configuration along the c axis. With increasing temperature (T), using Monte Carlo simulations we show that the system undergoes a phase transition triggered by the destruction of the ordering of a number of layers. This partial phase transition is shown to be intimately related to the ground-state spin structure. We show why some layers undergo a phase transition while others do not. The Green’s function method for non collinear magnets is also carried out to investigate effects of quantum fluctuations. Non-uniform zero-point spin contractions and a crossover of layer magnetizations at low T are shown and discussed.

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I. INTRODUCTION

Helimagnets have been subject of intensive investigations over the last four decades since the discovery of its ordering [1, 2]: in the bulk, a spin in a space direction turns an angle θ with respect to the orientation of its previous nearest neighbor (see Fig. 1). This helical structure can take place in several directions simultaneously with different helical angles. The helical structure shown in Fig. 1 is due to the competition between the interaction between nearest neighbors (NN) and the antiferromagnetic interaction between next-nearest neighbors (NNN). Other helimagnetic structures have also been very early investigated [3, 4]. Spin-wave properties in bulk helimagnets have been investigated by spin-wave theories [6, 7] and Green’s function method [8]. Heat capacity in bulk MnSi has been experimentally investigated [9].

We confine ourselves to the case of a Heisenberg helical film in an applied magnetic field. Helimagnets are special cases of a large family of periodic non collinear spin structures called frustrated systems of XY and Heisenberg spins. The frustration has several origins: (i) it can be due to the geometry of the lattice such as the triangular lattice, the face-centered cubic (FCC) and hexagonal-close-packed (HCP) lattices, with antiferromagnetic NN interaction [11, 12] (ii) it can be due to competing interactions between NN and NNN such as the case of helimagnets [11, 12] shown in Fig. 1 (iii) it can be due to the competition between the exchange interaction which favors collinear spin configurations and the Dzyaloshinskii-Moriya (DM) interaction which favors perpendicular spin arrangements.

Effects of the frustration have been extensively studied in various systems during the last 30 years. The reader is referred to recent reviews on bulk frustrated systems given in Ref. 14. When frustration effects are coupled with surface effects, the situation is often complicated. Let us mention our previous works on a frustrated surface [15] and on a frustrated FCC antiferromagnetic film [16] where surface spin rearrangements and surface phase transitions have been found. We have also recently shown results in zero field of thin films of body-centered cubic (BCC) and simple cubic (SC) structures [17, 18]. The helical angle along the c axis perpendicular to the film surface was found to strongly vary in the vicinity of the surface. The phase transition and quantum fluctuations have been presented.

In this paper we are interested in the effect of an external magnetic field applied along the c axis perpendicular to the film surface of a helimagnet with both classical and quantum Heisenberg spins. Note that without an applied field, the spins lie in the xy planes: spins in the same plane are parallel while two NN in the adjacent planes form an angle α which varies with the position of the planes [13], unlike in the bulk. As will be seen below, the applied magnetic field gives a very complex spin configuration across the film thickness. We determine this ground state (GS) by the numerical steepest descent method. We will show by Monte Carlo (MC) simulation that the phase transition in the field is due to the disordering of a number of layers inside the film.

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We identify the condition under which a layer becomes disordered. This partial phase transition is not usual in thin films where one observes more often the disordering of the surface layer, not an interior layer. At low temperatures, we investigate effects of quantum fluctuations using a Green’s function (GF) method for non-collinear spin configurations.

The paper is organized as follows. Section II is devoted to the description of the model and the determination of the classical GS. The structure of the GS spin configuration is shown as a function of the applied field. Section III is used to show the MC results at finite temperatures where a partial phase transition is observed. Effects of the magnetic field strength and the film thickness are displayed and discussed in terms of quantum fluctuations. Section V is devoted to concluding remarks.

We consider a thin film of SC lattice of $N_z$ layers stacked in the $c$ direction. Each lattice site is occupied by a Heisenberg spin. For the GS determination, the spins are supposed to be classical spins in this section. The Hamiltonian is given by

$$H = -\sum_{\langle i,j \rangle} J_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j - \sum_i \mathbf{H} \cdot \mathbf{S}_i \tag{1}$$

where $J_{i,j}$ is the interaction between two spins $\mathbf{S}_i$ and $\mathbf{S}_j$ occupying the lattice sites $i$ and $j$ and $\mathbf{H}$ denotes an external magnetic field applied along the $c$ axis. To generate helical angles in the $c$ direction, we suppose an antiferromagnetic interaction $J_2$ between NNN in the $c$ direction in addition to the ferromagnetic interaction $J_1$ between NN in all directions. For simplicity, we suppose that $J_1$ is the same everywhere. For this section we shall suppose $J_2$ is the same everywhere for the presentation clarity. Note that in the bulk in zero field, the helical angle along the $c$ axis is given by $\cos \alpha = \frac{J_1}{|J_2|}$ for a SC lattice [13] with $|J_2| > 0.25 J_1$. Below this value, the ferromagnetic ordering is stable.

In this paper we will study physical properties as functions of $J_2/J_1$, $H/J_1$ and $k_B T/J_1$. Hereafter, for notation simplicity we will take $J_1 = 1$ and $k_B = 1$. The temperature is thus in unit of $J_1/k_B$, the field and the energy are in unit of $J_1$.

In a film, the angles between NN in adjacent planes are not uniform across the film: a strong variation is observed near the surfaces. An exact determination can be done by energy minimization [17] or by numerical steepest descent method [15, 16]. The latter is particularly efficient for complex situations such as the present case where the spins are no longer in the $xy$ planes in an applied field: a spin in the $i-\text{th}$ layer is determined by two parameters which are the angle with its NN in the adjacent plane, say $\alpha_{i,i+1}$, and the azimuthal angle $\beta_i$ formed with the $c$ axis. Since there is no competing interaction in the $xy$ planes, spins in each plane are parallel. In this paper we use the steepest descent method which consists in calculating the local field at each site and aligning the spin in its local field to minimize its energy. The reader is referred to Ref. 18 for a detailed description. In so doing for all sites and repeating many times until a convergence to the lowest energy is obtained with a desired precision (usually at the $6$-th digit, namely at $\simeq 10^{-6}$ per cents), one obtains the GS configuration. Note that we have used several thousands of different initial conditions to check the convergence to a single GS for each set of parameters.

Figures 2 and 3 show the spin components $S^z$, $S^y$ and $S^x$ for all layers. The spin lengths in the $xy$ planes are shown in Fig 2. Since the spin structure in a field is complicated and plays an important role in the partial phase transition shown in the next section, let us describe it in details and explain the physical reason lying behind:

- Several planes have negative $z$ spin components. This can be understood by examining the competition between the magnetic field which tends to align spins in the $c$ direction, and the antiferromagnetic interaction $J_2$ which tries to preserve the antiferromagnetic ordering. This is very similar to the case of collinear antiferromagnets: in a weak magnetic field the spins remain antiparallel, and in a moderate field, the so-called “spin flop” occurs: the neighboring spins stay antiparallel with each other but turn themselves perpendicular to the field direction to reduce the field effect 19.

- Due to the symmetry of the two surfaces, one observes the following symmetry with respect to the middle of the film:

  (i) $S^x_1 = S^x_{N_z}$, $S^x_2 = S^x_{N_z-1}$, $S^x_3 = S^x_{N_z-2}$ etc.
FIG. 2: Spin components across the film in the case where $H = 0$. The horizontal axis $Z$ represents plane $Z$ ($Z = 1$ is the first plane etc.): (a) $S^z$; (b) $S^x$ (red) and $S^y$ (blue); (c) Modulus $S^{xy}$ of the projection of the spins on the $xy$ plane. See text for comments.

(ii) $S^y_1 = -S^y_{N_z}$, $S^y_2 = -S^y_{N_z-1}$, $S^y_3 = -S^y_{N_z-2}$ etc.

(iii) $S^z_1 = -S^z_{N_z}$, $S^z_2 = -S^z_{N_z-1}$, $S^z_3 = -S^z_{N_z-2}$ etc.

Note that while the $z$ components are equal, the $x$ and $y$ components are antiparallel (Fig. 2b): the spins preserve their antiferromagnetic interaction for the transverse components. This is similar to the case of spin flop in the bulk (see p. 86 of Ref. [19]). Only at a very strong field that all spins turn into the field direction.

- The GS spin configuration depends on the field magnitude $H$. If $H$ increases, we observe an interesting phenomenon: Figure 4 shows the spin configurations projected on the $xy$ plane (top view) for increasing magnetic field. We see that the spins of each chain tend progressively to lie in a same plane perpendicular to the $xy$ planes (Figs. 4a-b-c). The "planar zone" observed in Fig. 4c occurs between $H \approx 0.35$ and 0.5. For stronger fields they are no more planar (Fig. 4d-e-f). Note that the larger the $xy$ component is, the smaller the $z$ component becomes: for example in Fig. 4a the spins are in the $xy$ plane without field ($H = 0$) and in Fig. 4f they are almost parallel to the $c$ axis because of a high field.

A full view of the "chain" of $N_z$ spins along the $c$ axis between the two surfaces is shown in Fig. 3.

Note that the angle in the $xy$ plane is determined by the NNN interaction $J_2$. Without field, the symmetry is about the $c$ axis, so $x$ and $y$ spin components are equivalent (see Fig. 1). Under the field, due to the surface effect, the spins make different angles with the $c$ axis giving rise to different $z$ components for the layers across the film as shown in Fig. 2a. Of course, the symmetry axis is still the $c$ axis, so all $S^x$ and $S^y$ are invariant under a rotation around the $c$ axis. Figure 2b shows the symmetry of $S^x$ as that of $S^y$ across the film as outlined in remark (iii). Fig. 2b is thus an instantaneous configuration between $S^x$ and $S^y$ for each layer across the film. As the simulation time is going on these components rotate about the $c$ axis but their symmetry outlined in remark (iii) is valid at any time. The $xy$ spin modulus $S^{xy}$ shown in Fig. 2c, on the other hand, is time-invariant. The phase transition occurring for layers with large $S^{xy}$ ($xy$ disordering) is shown in the next section.
FIG. 4: Top view of $S^{xy}$ (projection of spins on $xy$ plane) across the film for several values of $H$: (a) 0, (b) 0.03, (c) 0.2, (d) 0.4, (e) 0.7, (f) 1.7. The radius of the circle, equal to 1, is the spin full length: for high fields, spins are strongly aligned along the $c$ axis, $S^{xy}$ is therefore much smaller than 1.

III. PHASE TRANSITION

We recall that for bulk materials, in spite of their long history, the nature of the phase transition in non-collinear magnets such as stacked triangular XY and Heisenberg antiferromagnets has been elucidated only recently [20–22]. On the other hand, surface effects in thin films have been intensively studied during the last three decades [19, 23, 24]. Most of theoretical studies were limited to collinear magnetic orderings. Phase transitions in thin films with non-collinear ground states have been only recently studied [15–18, 25]. MC simulations of a helimagnetic thin film [26] and a few experiments in helimagnets [27, 28] have also been carried out. These investigations were motivated by the fact that helical magnets present a great potential of applications in spintronics with spin-dependent electron transport [29–31].

As described in the previous section, the planar helical spin configuration in zero field becomes non planar in a perpendicular field. In order to interpret the phase transition shown below, let us mention that a layer having a large $z$ spin-component parallel to the field cannot have a phase transition because its magnetization will never become zero. This is similar to a ferromagnet in a field. However, layers having large negative $z$ spin-components (antiparallel to the field) can undergo a transition due to the magnetization reversal at a higher temperature similarly to an antiferromagnet in a field. In addition, the $xy$ spin-components whose $xy$ fluctuations are not affected by the perpendicular field can make a transition. Having mentioned these, we expect that some layers will undergo a phase transition, while others will not. This is indeed what we observed in MC simulations shown in the following.

For MC simulations, we use the Metropolis algorithm (see chapter 8 of Ref. [19]) and a sample size $N \times N \times N_z$ with $N = 20, 40, 60, 100$ for detecting lateral-size effects and $N_z = 8, 12, 16$ for thickness effects. The equilibrium time is $10^5$ MC steps/spin and the thermal average is performed with the following $10^5$ MC steps/spin.

A. Results: example of 12-layer film

In order to appreciate the effect of the applied field, let us show first the case where $H = 0$ in Fig. 5. We see there that all layers undergo a phase transition within a narrow region of $T$.

FIG. 5: (a) Layer magnetization and (b) layer magnetic susceptibility versus $T$ for $H = 0, J_2 = -1, N_z = 12$. Dark olive green void squares for the first layer, maroon void triangles for the second, red circles for the third, indigo triangles for the fourth, dark blue squares for the fifth, dark green void circles for the sixth layer.

In an applied field, as seen earlier, in the GS all layers do not have the same characteristics so one expects different behaviors. Figure 6 shows the layer magnetizations and the layer susceptibilities as functions of $T$ for $H = 0.2$ with $J_2 = -1, N_z = 12$ (only the first six layers are shown, the other six are symmetric). Several remarks are listed below:

- Only layer 3 and layer 5 have a phase transition: their magnetizations strongly fall down at the transition temperature. This can be understood from what we have anticipated above: these layers have the largest $xy$ components (see Fig. 2). Since the correlation between $xy$ components do not depend on the applied field, the temperature destroys the in-plane ferromagnetic ordering causing the transition. It is not the case for the $z$ components which are kept non zero by the field. Of course, symmetr-
FIG. 6: (a) Layer magnetization and (b) layer magnetic susceptibility versus $T$ for $H = 0.2$, $J_2 = -1$, $N_z = 12$. Dark olive green void squares for the first layer, maroon void triangles for the second, red circles for the third, indigo triangles for the fourth, dark blue squares for the fifth, dark green void circles for the sixth layer.

ric layers 8 and 10 have the same transition (not shown).

• Layers with small amplitudes of $xy$ components do not have a strong transverse ordering at finite $T$: the absence of pronounced peaks in the susceptibility indicates that they do not make a transition (see Fig. 6).

• Note that the $xy$ spin components of layers 3 and 5 are disordered at $T_c \approx 1.275$ indicated by pronounced peaks of the susceptibility.

What we learn from the example shown above is that under an applied magnetic field the film can have a partial transition: some layers with large $xy$ spin components undergo a phase transition (destruction of their transverse $xy$ correlation). This picture is confirmed by several simulations for various field strengths. Another example is shown in the case of a strong field $H = 0.7$: the GS is shown in Fig. 7 where we observe large $xy$ spin components of layers 3, 4, and 5 (and symmetric layers 7, 8 and 9). We should expect a transition for each of these layers. This is indeed the case: we show these transitions in Fig. 8 where sharp peaks of the susceptibilities of these layers are observed.

We close this section by showing some size effects. Figure 9 shows the effect of lateral size ($xy$ planes) on the layer susceptibility. As expected in a continuous tran-
sition, the peaks of the susceptibilities of the layers undergoing a transition grow strongly with the layer lattice size.

B. Effects of the film thickness

As for the thickness effects, we note that changing the thickness (odd or even number of layers) will change the GS spin configuration so that the layers with largest $xy$ components are not the same. As a consequence, the layers which undergo the transition are not the same for different thicknesses. We show in Fig. 10 the layer susceptibilities for $N_z = 8$ and 16. For $N_z = 8$, the layers which undergo a transition are the first, third and fourth layers with pronounced peaks, while for $N = 16$, the layers which undergo a transition are the third, fifth, seventh and eighth layers.

![Fig. 10: Magnetic susceptibility versus $T$ for two thicknesses with $H = 0.2, J_2 = -1$: (a) $N_z = 8$, (b) $N_z = 16$. Dark olive green void squares are for the first layer, maroon void triangles for the second, red circles for the third, indigo triangles for the fourth, dark blue squares for the fifth, dark green void circles for the sixth, black diamonds for the seventh, dark brown void diamonds for the eighth. See text for comments.](image)

IV. QUANTUM FLUCTUATIONS, LAYER MAGNETIZATIONS AND SPIN-WAVE SPECTRUM

We shall extend here the method used in Ref. 17 for zero field to the case where an applied magnetic field is present. The method remains essentially the same except for the fact that each spin is defined not only by its angles with the NN in the adjacent layers but also by its azimuthal angle formed with the $c$ axis as seen in section II.

We use in the following the Hamiltonian (1) but with quantum Heisenberg spins $S_i$ of magnitude $1/2$. In addition, it is known that in two dimensions there is no long-range order at finite temperature for isotropic spin models with short-range interaction. Since our films have small thickness, it is useful to add an anisotropic interaction to stabilize the long-range ordering at finite temperatures. Let us use the following anisotropy between $S_i$ and $S_j$ which stabilizes the angle between their local quantization axes $S_{iz}^i$ and $S_{iz}^j$:

$$\mathcal{H}_a = -I_1 \sum_{<i,j>} S_i^z S_j^z \cos \theta_{ij}$$

where $I_1$ is supposed to be positive, small compared to $J_1$, and limited to NN.

The general method has been recently described in details in Refs. 17, 18. To save space, let us give the results for the simple cubic helimagnetic film in a field. We define the following two double-time Green’s functions in...
writing coupled equations

\[D_n g_{n-2,n'} + E_n f_{n-2,n'} + B_n g_{n-1,n'} + C_n f_{n-1,n'} + (\omega + A_n) g_{n,n'} + B_n^+ g_{n+1,n'} + C_n^+ f_{n+1,n'} + D_n^+ g_{n+2,n'} + E_n^+ f_{n+2,n'} = 2 \langle S_n^z \rangle \delta_{n,n'}\]

\[-E_n g_{n-2,n'} - D_n f_{n-2,n'} - C_n g_{n-1,n'} - B_n f_{n-1,n'} + (\omega - A_n) f_{n,n'} - C_n^+ g_{n+1,n'} - B_n^+ f_{n+1,n'} + E_n^+ f_{n+2,n'} = 0\]

where \( n = 1, 2, ..., N_z \), \( d_n = J_1/J_2^0 \), \( \gamma = (\cos k_x a + \cos k_y b)/2 \). The coefficients are given by

\[A_n = -8J_1^0 < S_n^z > (1 + d_n - \gamma)\]

\[-2 < S_{n+1}^z > \cos \theta_{n,n+1}(d_n + J_1^+)\]

\[-2 < S_{n-1}^z > \cos \theta_{n,n-1}(d_n + J_1^+)\]

\[-2J_2 < S_{n+2}^z > \cos \theta_{n,n+2}\]

\[-2J_2 < S_{n-2}^z > \cos \theta_{n,n-2} - H \cos \zeta_n\]

\[B_n^\pm = 2J_1^0 \langle S_n^z \rangle (\cos \theta_{n,n\pm1} + 1)\]

\[C_n^\pm = 2J_1^0 \langle S_n^z \rangle (\cos \theta_{n,n\pm1} - 1)\]

\[E_n^\pm = J_2 \langle S_n^z \rangle (\cos \theta_{n,n\pm2} - 1)\]

\[D_n^\pm = J_2 \langle S_n^z \rangle (\cos \theta_{n,n\pm2} + 1)\]

\( \omega \) is the spin-wave frequency, \( k_x \) and \( k_y \) denote the wavevector components in the \( xy \) planes, \( n \) is the index of the layer along the c axis with \( n = 1 \) being the surface layer, \( n = 2 \) the second layer and so on. The angle \( \zeta_n \) is the azimuthal angle formed by a spin in the layer \( n \) with the c axis. Note that (i) if \( n = 1 \) then there are no \( n - 1 \) and \( n - 2 \) terms in the matrix coefficients, (ii) if \( n = 2 \) then there are no \( n - 2 \) terms, (iii) if \( n = N_z \) then there are no \( n + 1 \) and \( n + 2 \) terms, (iv) if \( n = N_z - 1 \) then there are no \( n + 2 \) terms. Besides, we have distinguished the in-plane NN interaction \( J_1^0 \) from the inter-plane NN one \( J_2^0 \). If we write all equations explicitly for \( n = 1, ..., N_z \) we can put these equations under a matrix of dimension \( 2N_z \times 2N_z \). Solving this matrix equation, one gets the spin-wave frequencies \( \omega \) at a given wave vector and a given \( T \).

The layer magnetizations can be calculated at finite temperatures self-consistently. The numerical method to carry out this task has been described in details in Ref. 17. It is noted that in bulk antiferromagnets and helimagnets the spin length is contracted at \( T = 0 \) due to quantum fluctuations 18. Therefore, we also calculate the layer magnetization at \( T = 0 \) 17, 32. It is interesting to note that due to the difference of the local field acting on a spin near the surface, the spin contraction is expected to be different for different layers.

We show in Fig. 12 the spin length of different layers at \( T = 0 \) for \( N = 12 \) and \( J_2 = -1 \) as functions of \( H \). All spin contractions are not sensitive for \( H \) lower than 0.4, but rapidly become smaller for further increasing \( H \). They spin lengths are all saturated at the same value for \( H > 2 \). Figure 13 shows the spin length as a function of \( J_2 \). When \( J_2 \geq -0.4 \), the spin configuration becomes ferromagnetic, and as a consequence the contraction tends
to 0. Note that in zero field, the critical value of $J_2$ is -0.25. In both figures 12 and 13 the surface layer and the third layer have smaller contractions than the other layers. This can be understood by examining the anti-ferromagnetic contribution to the GS energy of a spin in these layers: they are smaller than those of the other layers.

We show in Fig. 14 the layer magnetizations versus $T$ for the case where $J_2 = -1$ and $N_z = 12$: (a) $J_2 = -1$, (b) $J_2 = -0.5$, (c) $J_2 = -2$. Dark olive green void squares correspond to the magnetization of the first layer, maroon void triangles to the second, red circles to the third, indigo triangles to the fourth, dark blue squares to the fifth, dark green void circles to the sixth layer. The inset in the top figure shows an enlarged region at low $T$. See text for comments.

Before closing this section, let us discuss about the spin-wave spectrum. Let us remind that to solve self-consistently Eqs. (5)-(6) at each $T$, we use as inputs $\langle S_1^z \rangle, \langle S_2^z \rangle, ..., \langle S_{N_z}^z \rangle$ to search for the eigenvalues $\omega$ for each vector $(k_x, k_y)$ and then calculate the out-
puts $< S^z_1 >, < S^z_2 >, \ldots, < S^z_N >$. The self-consistent solution is obtained when the outputs are equal to the inputs at a desired convergence precision fixed at the fifth digit, namely $10^{-5}$ (see other details in Ref. 17). Figure 15 shows the spin-wave spectrum in the direction $k_x = k_y$ of the Brillouin zone at $T = 0.353$ and $T = 1.212$ for comparison. As seen, as $T$ increases the spin-wave frequency decreases. Near the transition (not shown) it tends to zero. Figure 16 shows the spin-wave spectrum at $T = 0.353$ for $J_2 = 0.5$ and $J_2 = -1$, for comparison. Examining them closely, we see that the distribution of the spin-wave modes (positions of the branches in the spectrum) are quite different for the two cases. When summed up for calculating the layer magnetizations, they give rise to the difference observed for the two cases shown in Fig. 14.

V. CONCLUDING REMARKS

In this paper we have shown (i) the GS spin configuration of a Heisenberg helimagnetic thin film in a magnetic field applied along the $c$ axis perpendicular to the film, (ii) the phase transition occurring in the film at a finite temperature, (iii) quantum effects at low $T$ and the temperature dependence of the layer magnetizations as well as the spin-wave spectrum.

Synthetically, we can summarize that under the applied magnetic field, the spins in the GS make different angles between them across the film. When the temperature increases, the layers with large $xy$ spin-components undergo a phase transition where the transverse (in-plane) $xy$ ordering is destroyed. This "transverse" transition is possible because the $xy$ spin-components do not depend on the field. Other layers with small $xy$ spin-components, namely large $z$ components, do not make a transition because the ordering in $S^z$ is maintained by the applied field. The transition of a number of layers with large $xy$ spin-components, not all layers, is a new phenomenon discovered here with our present model.

We have also investigated the quantum version of the model by using the Green’s function method. The results show that the zero-point spin contraction is different from layer to layer. We also find a crossover of layer magnetizations which depends on $J_2$, namely on the magnitude of helical angles.

Experiments are often performed on materials with helical structures often more complicated than the model considered in this paper. However, the clear physical pictures given in our present analysis are believed to be useful in the search for the interpretation of experimental data.

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