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CHAOS IN MAGNETIC RESONANCE EXPERIMENTS

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Abstract. – Non linear antiferromagnetic resonance was observed by parallel pumping (PP) with continuous waves in (NH₃CH₂)₂CuCl₄. This system is unique in its low threshold for PP and its strong anisotropy. The absorbed microwave power displays self spiking. As a function of power the irregularity of periods between the spikes changes. Reproducible transitions from nearly regular to irregular spiking are found (RI-transitions) with information dimensions $D_I$ in the range $2.5 \leq D_I \leq 5$. For certain parameter settings a hierarchy of RI transitions with different amplitudes is observed. The measurements are compared to the results of the 2n-dimensional stroboscopic model (SM), which describes the dynamics of n non-linear coupled spin wave modes. The SM simulates the mentioned behaviour for $n = 2$ and for strong anisotropy. Certain measurements with large $D_I$ were more readily modeled by the three mode SM.

1. Introduction

After the discovery of spin wave instabilities in magnetic resonance experiments it became clear that the magnetization of the sample in these processes doesn’t necessarily approach a steady state [1]. Subsequently experiments were performed of the dynamics in the instabilities of subsidiary absorption, premature saturation and parallel pumping (PP) [2-3]. Recently such measurements have been described in the framework of chaos.

The feature common to chaotic phenomena is that as some external parameter is varied the dynamical behaviour of the system may change in a universal way. Different systems follow the same routes from periodic to chaotic motion. Chaotic scenarios were then observed experimentally [5-8].

An example of behaviour within the chaotic regime is the transition from regular pulsing to irregular self pulsing (RI-transitions) [9-10].

In this paper we will report on PP experimental results in (CH₂NH₃)₂CuCl₄. The main experimental findings are: regular relaxation oscillations, RI transitions and for certain parameter settings a hierarchical superposition of RI transitions. These experimental findings are simulated with the use of the classically derived stroboscopic model (SM) [11]. Some of the important results of this paper are presented in figure 1 as a comparison of experimental and theoretical results.

2. Experiment and results

Single crystals of antiferromagnetic (CH₂NH₃)₂CuCl₄ were placed at the center of an X-band cavity with the easy axis of magnetization parallel of the static $H₀$ and the microwave field $h$. The time dependence of the microwave absorption was recorded by an ADC and the power was changed between runs with a step motor which adjusted a precision attenuator.

By increasing the power well above the threshold for PP auto oscillations with irregular periods and amplitudes with the shape of relaxation oscillations were found in the kHz region. Once the oscillations were set up the dynamical behaviour of the system was studied as a function of the power $P$ as control parameter. The main findings are: plain periodic relaxation oscillations of the type shown in figure 1a. The width of the spikes (full width at half the peak maximum), and the mean period between spikes increase with power as shown in figure 2 over an extremely narrow power range of $\Delta h = 2 \mu T$. In a different region of parameter space and on a larger scale of control parameter range $\Delta h = 20 \mu T$ one observes RI transitions. In this paper three different measurements (#1, #2, #3) of RI transitions are described which were recorded at the fields 0.8830 T, 0.8835 T and 0.8840 T. In figure 3, a typical...

Fig. 1. – Comparison of typical experimental time series measured by PP in (CH₂NH₃)₂CuCl₄ with numerical results from the SM. Left, theory and right experiment: a) plain relaxation oscillations, b) relaxation oscillations with irregular periods and c) high dimensional chaos.
sequence of runs from measurement #2 is presented. By increasing \( H_0 \) by 2 \( \mu \)T one observes a superposition of irregular spiking at different fundamental frequencies (Fig. 4). Starting at the highest power level (\( P = 8.9 \) dBm) one observes spikes at irregular time intervals. The spikes all have the same shape: a fast rise (40 \( \mu \)s) and an average width of 1.4 ms. As the power is decreased the mean period between spikes, the width of the spikes and their amplitudes decrease. By decreasing the power further a second, very low frequency spiking is superimposed on the spikes already present. This second level of spiking goes through the same transition as the first level of spikes. A further decrease in \( P \) reveals a third level of dynamics. The widths of these spikes are presented in figure 5.

3. Data analysis

The dimension function \( D(\gamma) \) was calculated for the runs presented in figure 3 by evaluating the inverse slope of \( \log <\delta(n)> \) vs. \( \log n \) from the embedded time series [12]. First the signals consisting of 5120 data points were embedded in an \( E \)-dimensional space by using the sampling time as time delay. Then 1100 reference points were chosen at random the embedded points. Each reference point was compared to \( n \) other randomly chosen points and the generalized distance \( \delta_{j,k,E}(n) \) to the \( k \)-th nearest neighbour is calculated. Now the generalized average distance over all \( m \) reference points is calculated

\[
\langle \delta^\gamma \rangle = \frac{1}{m} \sum_{j=1}^{m} \delta_{j,k,E}^\gamma (n) .
\]
The average \( \langle \delta^7 \rangle \) was calculated for \( 45 \leq n \leq 4000 \) and for embedding dimensions \( E = 1, ..., 10 \). The order of nearest neighbours was \( k = 20 \). A typical result is shown in figure 6. The slope of \( \log \langle \delta^7 \rangle \) decreases with higher embedding dimension and with higher values of \( n \) and becomes nearly constant. This saturation value for the slope determines the dimension. The slopes were computed from a linear regression in a certain range of \( n \), (Fig. 7). As a function of embedding dimension the slope saturates in a rather large range of \( 5 \leq E_{sat} \leq 8 \) but \( D (\gamma = 0) \) stays well below \( E \), which indicates that the data is of deterministic and not of random origin. In figure 8 the dimension is plotted for the whole measurement #2.

As a function of \( n \) no true saturation occurred. This fact is indicated by the two significantly different values of \( D (\gamma = 0) \) in figure 8 derived from the two regression intervals \( 40 \leq \log_5 n \leq 45 \) and \( 45 \leq \log_5 n \leq 50 \). Starting at high microwave field \( h = 0.061 \) mT the dimension takes on the value \( D (\gamma = 0) = 2.1 \). As the power is reduced the dimension initially starts to increase to a value of \( D (\gamma = 0) = 2.6 \). Then there is a sudden jump to a value of 1.5 at \( h = 0.0545 \) mT. A further increase in power leads to a gradual increase in dimension.

Due to the rather restricted number of data points the determination of the dimensions for these data sets is not unambiguous. Because the dimension function is defined only in the limit \( n \rightarrow \infty \) one has to use very high values of \( n \) to be sure that the asymptotic slope of \( \log \langle \delta^7 \rangle \) vs. \( \log n \) is reached. Furthermore the sampling time should be of the order of about one quarter of the minimum period present in the data which can’t be fulfilled for our data without missing spikes. Despite these difficulties the calculation indicates high values of dimension spread over the range \( 2.5 \leq D (\gamma = 0) \leq D_1 \leq 5 \).

Considering the difficulty in obtaining dimensions for our data one might just make a statistical analysis of the periods \( T_k \) between the spikes at times \( t_k \) and \( t_{k+1} \). Instead of calculating the average deviation

\[
\alpha = (1/\tau p) \sum_{k=1}^{p} |\tau - T_k|
\]

of the \( p \) periods from the mean period \( \tau \) one can replace \( T_k \) with \( T_k \mod \tau \) and adjust \( \tau' \) to obtain a minimum value for \( \alpha \) as

\[
I = (1/\tau' p) \sum_{k=1}^{p} |\tau' - T_k \mod \tau'|.
\]

For a sequence of equal periods \( I = 0 \) and for a random distribution of periods \( I = 0.58 \) [10]. This modified definition of the dispersion of periods from a certain \( \tau \) is probably better suited to analyse RI transitions as e.g. a sequence of period doublings would also give
$I = 0$. In figure 9 $I$ is shown for the three runs #1, #2 and #3.

For all three magnetic field settings the system starts off is an irregular state with $I \approx 0.4$ (far from randomness, $I = 0.58$), then follows a route to nearly regular motion with $I \approx 0.2$, the system then jumps suddenly back to irregular spiking. This is followad by another very similar route to nearly regular spiking of which only the beginning was recorded in the measurements #1 and #2. Note that the power level where the most regular spiking occurs increases with increasing field $H_0$, i.e. with increasing wave vector of the pumped magnons.

Also the transition rate from irregular to regular spiking increases with increasing wave vector and the window of regular spiking becomes narrower.

![Graph showing $I$ vs. driving field $h$ for the three measurements described in the text.](image)

**Fig. 9.** $I$ vs. driving field $h$ for the three measurements described in the text. In all three cases the curve is an exponential of the form $I = I_0 \left(1 - e^{(h_h - h_0)/h_1}\right)$ where $I_0 = 0.4$, $h_0 = 0.049$ mT, $h_2 = 0.054$ mT, $h_3 = 0.055$ mT; $b_1 = 0.0021$ mT, $b_2 = 0.015$ mT, and $b_3 = 0.0011$ mT.

### 4. The stroboscopic model SM

The SM [11] is derived from the classical equation of motion $ds/dt = s \times H_{eff} +$ damping. A standing spin wave mode which is equivalent to two magnons of opposite wave vector sign is represented by one classical precessing about a normalized strong static field. Only the slow motion of the precessing spins is treated by analytically integrating the spin precession over two pump periods using the approximation that the static field is much larger than all other fields. In all calculations presented here only the PP case is considered which leads to the set of equations: $(\Delta \phi = \phi_{1,2} - \phi_{2,1})$

$$\phi_{1,2} = \cos \theta_{1,2} A_{1,2} \cos 2\phi_{1,2} + d_{1,2} (1 - \cos \theta_{1,2}) + \frac{B}{4} \cos \theta_{1,2} \sin^2 \theta_{2,1} \sin 2\Delta \phi + \omega_{1,2}.$$  (5)

Here $A_k$ are proportional to the pumping fields, $d_k$ are the self detuning terms, $\omega_k$ are the detuning terms, $\tau_k$ are the damping terms of the two modes and $B$ describes the interaction between the two modes. The following function is considered to be an expression which can be compared to the absorption signal of the experiment: $signal = \sum_{i=1}^{2} A_i \sin^2 \theta_i \cos 2\phi_i$. Figures 10 and 11 show the phase space for the two mode calculations presented in figures 1a and b. Both modes are driven above their threshold values ($A_k > \tau_k$). In the first example the trajectories fall onto a limit cycle and the signal has the form of relaxation oscillations. In the next example the periods between the relaxation oscillations become irregular.

![Graph showing phase space for the two mode calculation with parameter values: $r_{1,2} = 0.1$, $A_1 = 0.5$, $A_2 = 0.39$, $b_1 = 0$, $b_2 = -0.4$, $d_1 = -0.5$, $d_2 = 0.5$, $B = -32$. Note that both modes are driven above their threshold values. The signal consists of relaxation oscillations as seen from the shape of the limit cycle.](image)

**Fig. 10.** SM calculation with parameter values: $r_{1,2} = 0.1$, $A_1 = 0.5$, $A_2 = 0.39$, $b_1 = 0$, $b_2 = -0.4$, $d_1 = -0.5$, $d_2 = 0.5$, $B = -32$. Note that both modes are driven above their threshold values. The signal consists of relaxation oscillations as seen from the shape of the limit cycle.

![Graph showing phase space for the two mode calculation with parameter values: $r_1 = 0.1$, $r_2 = 0.085$, $A_1 = 0.5$, $A_2 = 0.39$, $b_1 = 0$, $b_2 = -0.4$, $d_{1,2} = 0.5$, $B = -32$. Note that both modes are driven above their threshold values. The signal consists of relaxation oscillations with irregular periods.](image)

**Fig. 11.** SM calculation with parameter values: $r_1 = 0.1$, $r_2 = 0.085$, $A_1 = 0.5$, $A_2 = 0.39$, $b_1 = 0$, $b_2 = -0.4$, $d_{1,2} = 0.5$, $B = -32$. Note that both modes are driven above their threshold values. The signal consists of relaxation oscillations with irregular periods.

The two mode SM already predicts irregular spiking which can be compared to certain experimental time series measured in antiferromagnetic (CH$_2$NH$_3$)$_2$CuCl$_4$. Also calculations performed so far, indicate that this type of behaviour only exists in a very narrow range of parameters. This feature is in good agreement with experimental observations. On the other hand more complex types of relaxation oscil-
lations have been measured, see figure 1c. An attempt to model the observed dynamics is made with the three mode SM. For antiferromagnetic spin waves it seems to be physically justified to consider higher dimensional models than for example for ferromagnetic YIG because in the antiferromagnets the density of spin wave modes in the magnon band is very high due to the small dipolar interaction.

Furthermore, the calculations of the dimension function for certain runs of the PP data performed in section three suggest the presence of comparatively high dimensional attractors \((D_t \approx 5)\). This indicates that the minimum number of independent variables needed to model the experimental behaviour is five. A measurement of highly irregular relaxation oscillations is compared in figure 1c to a three mode simulation with the SM. (The parameters are: \(r_1 = 0.1, r_2 = 0.042, r_3 = 0.01, A_1 = 0.4, A_2 = 0.08, A_3 = 0.05, d_{1,2,3} = 0.5\). Mode one is driven at resonance and the detuning increases from mode two to mode three \(\omega_1 = 0, \omega_2 = -0.4, \omega_3 = -1\). Mode one and two are coupled strongly to mode two and three, but the coupling between mode one and three is weak \(B_{1,2} = -30, B_{1,3} = 2\).)

5. Conclusions

Nonlinear antiferromagnetic resonance was observed by the parallel pumping technique with continuous waves in \((\text{NH}_3\text{CH}_2)_2\text{CuCl}_4\). The main results are: with all external parameters held constant self spiking of the absorbed microwave power is observed. As the microwave power is changed the irregularity of periods between the spikes changes. Reproducible transitions from regular to irregular spiking are observed (RI-transitions) at different main field settings. The dimension functions for RI-transitions are: \(2.5 \leq D_t \leq 5\).

The classically derived two mode stroboscopic model qualitatively predicts the observed behaviour. Certain high dimensional measurements were more readily modeled by the six dimensional three mode SM.

For certain parameter settings a hierarchy of RI-transitions is observed.

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