Eur. Phys. J. B **10**, 99–103 (1999)

THE EUROPEAN PHYSICAL JOURNAL B

EDP Sciences

Route to and from the NMR chaos in diamagnets

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Received 19 August 1998 and Received in final form 15 December 1998

Abstract. The route to and from the chaos *via* period doubling bifurcations in nuclear spin system with dipole-dipole interactions is investigated. The transition points are found. It is shown that route from the chaos proceeds according the Feigenbaum scenario.

PACS. 05.45.Pq Numerical simulations of chaotic models – 76.60.-k Nuclear magnetic resonance and relaxation

1 Introduction

In our recent paper [1] we have studied the second order Suhl instability [2] in nuclear spin-system (NSS) with dipole-dipole interactions. The corresponding thresholds were calculated and the parametrical unstable nonuniform modes were found for various detuning ranges. This study was undertaken in order to investigate other than quasi-equilibrium regime [3,4] of NSS development. The present article theoretically studies the route to the chaos in mentioned spin-system. The article also suggests the experimental conditions for verification of the results obtained. In case of their good agreement it will be possible to investigate the different chaotic regimes [5–7] both theoretically and experimentally in such a medium.

Number of papers are devoted to the subject of onset of chaos caused by the excitation of parametrical unstable modes in magnetic materials [8–11]. Detailed studies of chaotic behavior of spin systems in ferromagnets [12–15], antiferromagnets [16,17], *etc.*, have been conducted previously. However, according to my best knowledge the NMR chaos in NSS with DDI has not yet been considered.

On the other hand the dipolar coupled spin-system is a very attractive object for theoretical examination due to the clarity of the character of spin-spin interactions. The different properties of NSS with DDI, such as its quasi-nonequilibrium dynamics [3] and onset of nuclear magnetic ordering [18] caused by NMR saturation, possible Bose condensation [19], etc., are being studied. Exact knowledge of dipole-dipole forces simplifies calculation of various coupling constants, while in electron spin-systems considered above [8–17] calculation of coupling constants between spin-wave modes is difficult and consequently theoretical prediction of excited parametrically unstable modes is almost impossible. Additionally it should be mentioned that in connection with the development of dynamical polarization methods [20] it is easy to handle NSS at low spin temperatures.

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2 Nonlinear equations for NSS

Let us consider a NSS consisting of nuclear spins ^{19}F ($I = 1/2$) in CaF_2 spherical sample. The sample is placed in a strong static magnetic field H_0 and undergoes the transverse pumping. H_0 is applied along the crystallographic axis z. High initial polarization of NSS and its further contact with paramagnetic impurities is provided by the dynamical polarization. The secular part of the NSS Hamiltonian has the form [21]:

$$\mathcal{H}_0 = -\sum_f^N \omega_0 I_f^z - \sum_{fg}^N \varepsilon_{fg}(2I_f^z I_g^z - I_f^+ I_g^-) - \omega_1 \sum_f^N \left(I_f^+ \mathrm{e}^{i\omega t} + I_f^- \mathrm{e}^{-i\omega t} \right), \tag{1}$$

$$\varepsilon_{fg} = -\frac{g^2(1-3\cos^2\theta_{fg})}{4|\mathbf{r}_f - \mathbf{r}_g|^3},$$

where $\omega_0 = gH_0$ ($\hbar = k_\mathrm{B} = 1$), g is the gyromagnetic ratio for nuclei, $\mathbf{I}_f$ is the operator of the spin located in the lattice site f, $I_f^\pm = I_f^x \pm i I_f^y$, N is the number of spins in a lattice, ω_1 and ω are the amplitude (in frequency units) and frequency of the pumping magnetic field, $\mathbf{r}_f$ is the radius-vector of the spin situated in the site f, θ_{fg} is the angle between $\mathbf{r}_f - \mathbf{r}_g$ and z.

The nonsecular terms in Hamiltonian (1) are neglected because the gap in the spectrum of linear excitations is assumed to be much larger than the width of the spectrum zone (in presence of a strong static magnetic field). Thus the first order Suhl instability [2] does not take place and the probability of three magnon processes is minute.

Let us use the momentum presentation

$$I_{\mathbf{k}}^{z}=\frac{1}{N}\sum_{f}^{N} I_{f}^{z}\mathrm{e}^{-i\mathbf{k}\mathbf{r}_{f}},$$
$$I_{\mathbf{k}}^{\pm}=\frac{1}{N}\sum_{f}^{N} I_{f}^{\pm}\mathrm{e}^{\mp i\mathbf{k}\mathbf{r}_{i}},$$
$$\varepsilon_{\mathbf{k}}=\sum_{g}^{N}\varepsilon_{fg}\mathrm{e}^{-i\mathbf{k}(\mathbf{r}_{f}-\mathbf{r}_{g})}.$$

Thus from (1) we get the equation of motion in the rotating (with frequency ω) frame:

$$\mathrm{d}M_{\mathbf{k}}^{+}/\mathrm{d}t=-\left(i\Delta+\gamma_{\mathbf{k}}\right)M_{\mathbf{k}}^{+} - 2i\sum_{\mathbf{q}}\varepsilon_{\mathbf{q}}\left(2M_{\mathbf{k}-\mathbf{q}}^{+}M_{\mathbf{q}}^{z}+M_{\mathbf{q}}^{+}M_{\mathbf{k}-\mathbf{q}}^{z}\right)+2i\omega_{1}M_{\mathbf{k}}^{z}, \quad (2)$$

$$\mathrm{d}M_{\mathbf{k}}^{z}/\mathrm{d}t=-\gamma_{\mathbf{k}}^{\prime}\left(M_{\mathbf{k}}^{z}-\delta_{\mathbf{k0}}M_{\mathrm{st}}\right) + i\sum_{\mathbf{q}}\varepsilon_{\mathbf{q}}\left(2M_{\mathbf{q}}^{+}M_{\mathbf{q}-\mathbf{k}}^{-}-M_{\mathbf{q}}^{-}M_{\mathbf{q}+\mathbf{k}}^{+}\right)+i\omega_{1}\left(M_{\mathbf{k}}^{+}-M_{-\mathbf{k}}^{-}\right),$$

where the Weiss field approximation [21] has been used; $\Delta=\omega_0-\omega$; $\mathbf{M_k}\equiv\langle\mathbf{I_k}\rangle$, $\langle\cdots\rangle$ denotes quantum-statistical averaging; $\delta_{\mathbf{k0}}$ is Kronecker's symbol and $M_{\mathrm{st}}\simeq 1/2$ is a static value of M_0^z in absence of transverse pumping (low and positive nuclear spin temperatures are considered). The damping constants $\gamma_{\mathbf{k}}$, $\gamma_{\mathbf{k}}^{\prime}$ generally differ from each other. For simplicity one can consider the case when a relaxation rate caused by the dynamical polarization is much more than the one caused by the interaction with thermal excitations of nuclear spins. Then the following equality takes place [22,23] $\gamma_{\mathbf{k}}=1/2\gamma_{\mathbf{k}}^{\prime}\equiv\gamma$.

Neglecting the nonlinear terms in the set of equations (2) one obtains the solution in the form of linear spin waves [4] characterized by the dispersion law $\omega_{\mathbf{k}}=\omega_0+\varepsilon_{\mathbf{k}}$ [24]. The spectrum of linear spin excitations in CaF_2 (H_0 is applied along the direction [001]) is localized between the boundaries (see Ref. [21]) $\varepsilon_{\mathbf{k}_{\mathrm{I}}}=\varepsilon_{\min}=-9,687g^2/(4a^3)$ and $\varepsilon_{\mathbf{k}_{\mathrm{II}}}=\varepsilon_{\max}=5,352g^2/(4a^3)$ (a is the lattice parameter), where $\mathbf{k}_{\mathrm{I}}=(0,0,\pi/a)$ and $\mathbf{k}_{\mathrm{II}}=(\pi/a,\pi/a,0)$.

If the amplitude of pumping field is small, only the uniform mode is excited and the stationary solution of the set of equations (2) has the form $M_0^{+}=\left(M_0^{-}\right)^{*}=\omega_1/(\Delta-i\gamma)$, $M_{\mathbf{k}}^{+}=M_{\mathbf{k}}^{-}=0$. If the amplitude of the transverse pumping field exceeds the critical value the above solution become unstable and another stationary solution [2,25] with $M_{\mathbf{k}}^{+}=\left(M_{\mathbf{k}}^{-}\right)^{*}\neq 0$ appears. Let us consider the resonant case $\Delta=0$. Then only the points $\mathbf{k}=0$ and $\mathbf{k}_{\mathrm{I}}=(0,0,\pi/a)$ in $\mathbf{k}$ space are excited *i.e.* the mode from the bottom of the spectrum zone is unstable [1]. Thus taking into account the identities $\mathbf{k}_{\mathrm{I}}=-\mathbf{k}_{\mathrm{I}}$, $2\mathbf{k}_{\mathrm{I}}=(0,0,0)$ one obtains from (2) the following system of equations:

$$\begin{aligned}
\mathrm{d}m_{\mathbf{k}_{\mathrm{I}}}^{+}/\mathrm{d}\tau&=-m_{\mathbf{k}_{\mathrm{I}}}^{+}-2ic\left(2m_0^{+}m_{\mathbf{k}_{\mathrm{I}}}^{z}+m_{\mathbf{k}_{\mathrm{I}}}^{+}m_0^{z}\right)+2ibm_{\mathbf{k}_{\mathrm{I}}}^{z},\\
\mathrm{d}m_0^{+}/\mathrm{d}\tau&=-m_0^{+}-6icm_{\mathbf{k}_{\mathrm{I}}}^{z}m_{\mathbf{k}_{\mathrm{I}}}^{+}+2ibm_0^{z},\\
\mathrm{d}m_{\mathbf{k}_{\mathrm{I}}}^{z}/\mathrm{d}\tau&=-2m_{\mathbf{k}_{\mathrm{I}}}^{z}+ic\left(m_0^{-}m_{\mathbf{k}_{\mathrm{I}}}^{+}-m_{\mathbf{k}_{\mathrm{I}}}^{-}m_0^{+}\right)\\
&\quad+ib\left(m_{\mathbf{k}_{\mathrm{I}}}^{+}-m_{\mathbf{k}_{\mathrm{I}}}^{-}\right),\\
\mathrm{d}m_0^{z}/\mathrm{d}\tau&=-2\left(m_0^{z}-1\right)+ib\left(m_0^{+}-m_0^{-}\right),
\end{aligned}\qquad(3)$$

where the following dimensionalness quantities are introduced $\tau=\gamma t$, $\mathbf{m_k}=\mathbf{M_k}/M_{\mathrm{st}}$, $b=\omega_1/\gamma$, $c=\varepsilon_{\min}M_{\mathrm{st}}/\gamma$. So one gets the system of six nonlinear equations for the variables $\mathrm{Re}\left(m_{\mathbf{k}_{\mathrm{I}}}^{+}\right)$, $\mathrm{Im}\left(m_{\mathbf{k}_{\mathrm{I}}}^{+}\right)$, $\mathrm{Re}\left(m_0^{+}\right)$, $\mathrm{Im}\left(m_0^{+}\right)$, $m_{\mathbf{k}_{\mathrm{I}}}^{z}$ and m_0^z.

3 Results

The calculations are made for the parameter $c=10$. The chaotic regimes have not been found for the parameter c, which sufficiently differs from the mentioned value.

Computer calculations give the following results: At $b_0=r=0.317$ the parametrical instability occurs and the solution in the form of fixed point with $m_{\mathbf{k}_{\mathrm{I}}}^{+}\neq 0$ appears; at $b_1=5.67r$ the fixed point become unstable and the auto-oscillations (limit cycle) appears; at $b_2=7.72r$ the period doubling and at $b_\infty=7.98r$ the onset of chaos occurs. These results are represented in Figure 1 as the time series for longitudinal polarization $p_{\mathrm{l}}=m_0^z$ (Figs. 1a, b, c) and phase portraits for transverse polarization in the rotating frame $p_{\mathrm{t}}=\left|m_0^{+}\right|$ *versus* p_{l} (Figs. 1d, e, f).

The chaos continues till $b_\infty^{\prime}=9.90r$, when the system returns to the periodical motion. In particular, at $b_3=10.05r$ and at $b_4=10.80r$ the transitions $4T-2T$ and $2T-1T$ take place, respectively. The Analysis of quantities $b_\infty^{\prime}$, b_3 and b_4 gives the value for Feigenbaum constant [26] $\delta\approx\left(b_4-b_\infty^{\prime}\right)/\left(b_3-b_\infty^{\prime}\right)=6$ instead of the real one $\delta=4.6692...$. Further, after b reaches the value $b_5=14.5r$ one gets again the solution in the form of fixed point with $m_{\mathbf{k}_{\mathrm{I}}}^{+}=0$. This behavior of NSS is represented in Figure 2. $4T$, $2T$ and $1T$ period limit cycles are shown as time series of p_{l} (Figs. 2a, b, c) and phase portraits p_{l} *versus* p_{t} (Figs. 2d, e, f).

Let us consider the conditions for experimental verification. The static magnetic field should have the value $\omega_0\gg\omega_{\mathrm{d}}\sim g^2/(4a^3)=0.88\times10^4\ \mathrm{s}^{-1}$ (ω_{d} is the characteristic scale of DDI). After the initial polarization of NSS $p_{\mathrm{l}}\sim 0.99$ is achieved the dynamical polarization should be fixed at the level providing the relaxation rate for nuclear spins $\gamma=\left|\varepsilon_{\min}\right|/20=0.42\times10^4\ \mathrm{s}^{-1}$ ($c=10$). Then applying transverse resonant ($\omega=\omega_0$) magnetic field the parametrical instability should be observed (see also Ref. [1]) at the amplitude $\omega_1/\gamma=r=0.317$, *i.e.* $\omega_1=0.13\times10^4\ \mathrm{s}^{-1}$. Further, increasing ω_1 one can observe the periodic motion (with frequency $f\approx2\gamma=0.85\times10^4 s^{-1}$), period doubling, onset of the chaos and route from the chaos at the amplitudes given in the previous paragraphs.

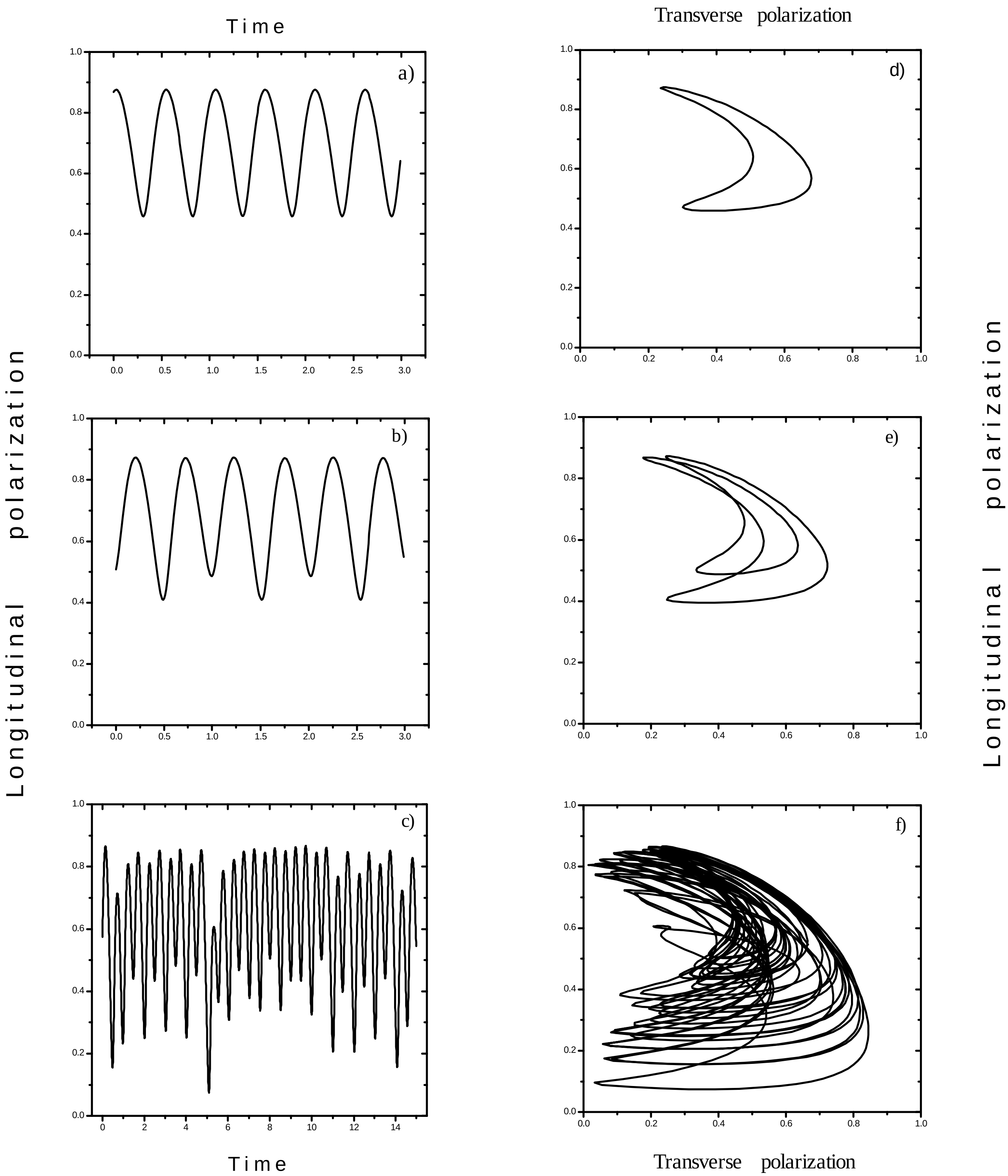

Fig. 1. The route to the chaos. a), b), c) time series for longitudinal ($p_{\rm l}$) polarization. d), e), f) phase portraits for $p_{\rm l}$ *versus* transverse ($p_{\rm t}$) polarization. a), d) $1T$ period limit cycle at $b = \omega_1/\gamma = 7.65r = 2.42$. b), e) $2T$ period limit cycle at $b = 7.85r = 2.49$. c), f) chaotic behavior at $b = 8.5r = 2.69$. Time is scaled by γ.

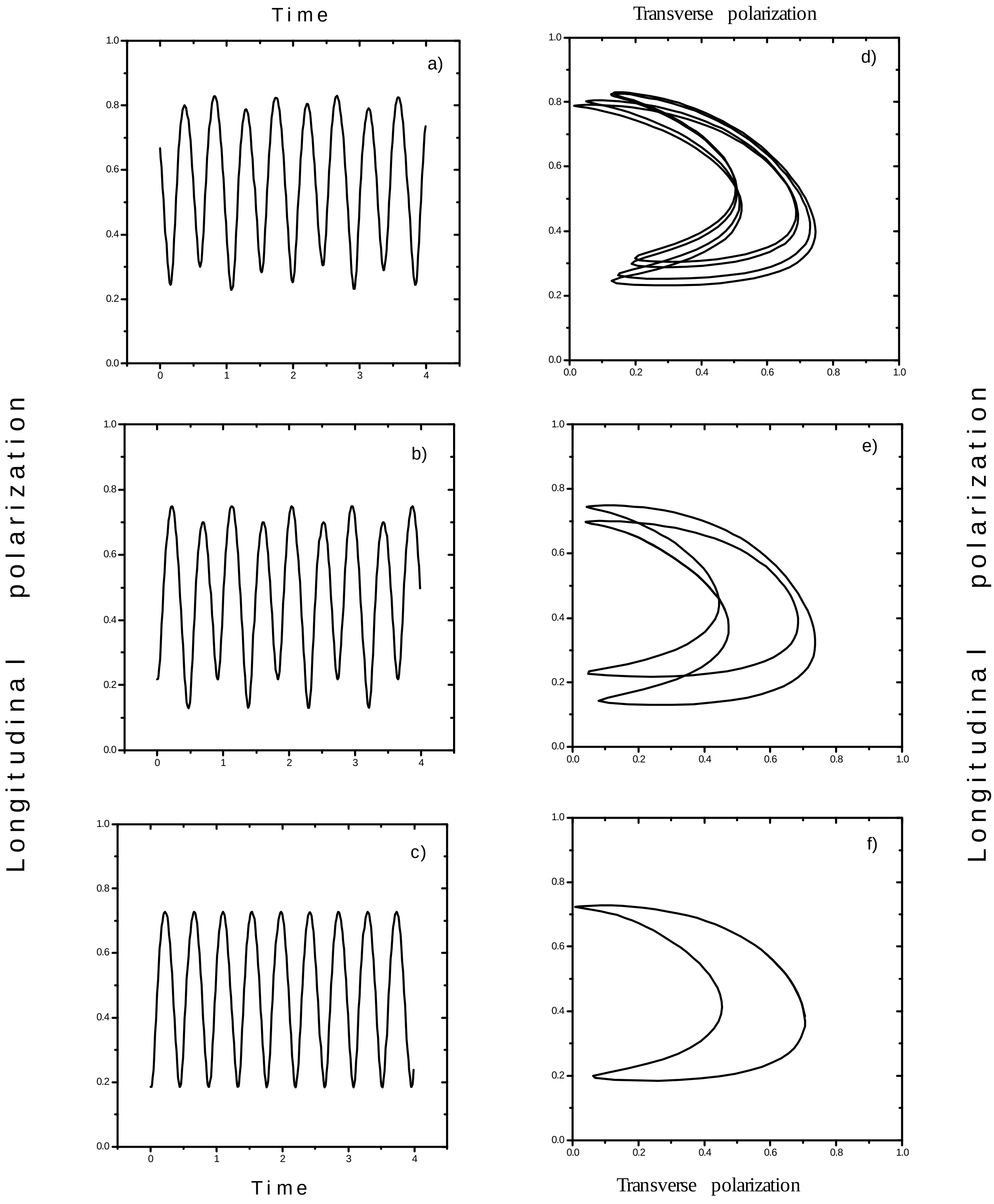

Fig. 2. The route from the chaos. a), b), c) p_{l} *vs.* τ and d), e), f) p_{l} *vs.* p_{t}. a), d) $4T$ period limit cycle at $b = 10r = 3.17$. b), e) $2T$ period limit cycle at $b = 10.6r = 3.36$. c), f) $1T$ period limit cycle at $b = 11r = 3.49$. τ is scaled by γ.

Author is very thankful to Sh. Revishvili and Dr. A.D. Rogava for their useful suggestions concerning computational procedures. Author is also very obliged to Mr. and Mrs. Abeywickramas for the help in manuscript preparation process.

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