## Small-signal amplification in the electrical conductivity of barium sodium niobate crystals

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We observe a resonance behavior of oscillations prior to Hopf bifurcations in an autonomous system. The system studied is a barium sodium niobate crystal in which voltage oscillations are induced by a periodically modulated current density. The resonance behavior is measured as a function of the dc-offset current density. The shape of the resonance peak and its dependence on the dc offset are found to be in fair quantitative agreement with recent theoretical studies.

The amplification of small signals in nonlinear dynamical systems prior to the onset of period-doubling bifurcations was first studied by Heldstab, Thomas, Geisel, and Radons.<sup>1</sup> The theory has been extended by Wiesenfeld and McNamara<sup>2,3</sup> and by Hackenbracht and Höck.<sup>4</sup> The concept predicts an enhanced sensitivity of nonlinear dynamical systems to small external excitations as a result of a coupling between the signal and internal fluctuations. The magnitude of amplification should grow substantially as the critical parameter approaches the instability point. So far, experiments with nonautonomous systems have confirmed the theoretical predictions. The systems studied<br>are analog simulations,<sup>2,5</sup> voltage driven p-n junctions,<sup>6</sup> and parametrically modulated NMR lasers.<sup>7</sup> In this Rapid Communication we present experimental results obtained in an autonomous system. We study the smallsignal amplification near a Hopf instability in the electrical conductivity of barium sodium niobate (BSN, stoichiometric composition  $Ba_2NaNb_5O_{15}$  single crystals.

The experimental setup and procedure for observing electrical instabilities in BSN crystals are described elseelectrical instabilities in BSN crystals are described els<br>where.<sup>8-11</sup> The control parameters are sample temperatu and oxygen partial pressure at the sample and current density. In the experiments described here we set the sample temperature at 500'C and the oxygen partial pressure at 1000 mbar. A constant current density is applied and the voltage across the crystal is measured. At a critical value of the current density the voltage begins to oscillate, indicating the first Hopf bifurcation, and at a higher critical current density we observe a second Hopt bifurcation. For the chosen experimental conditions, the first critical current density is  $j_{c1} = 0.566$  mA/cm<sup>2</sup>, the second is  $j_{c2} = 3.1$  mA/cm<sup>2</sup>. The internal oscillation frequencies here are  $f_1 = 216$  mHz and  $f_2 = 365$  mHz, respectively. The oscillation amplitudes are in the order of few millivolts superimposed on a dc offset of about 65 V at the first bifurcation and about 90 V at the second bifurcation.

In addition to the dc offset, we now apply an ac current density with a small amplitude  $j_{ac}$  and with the frequency  $f_{ac}$ . This leads to driven voltage oscillations with the same frequency. We measure the amplitude of these oscillations Using a fast-Fourier-transform signal analyzer in order to enhance the signal-to-noise ratio. The dc offset is set below, but close to, either the first or the second critical value and the frequency  $f_{ac}$  is varied over a region around the corresponding internal frequency.

In Fig. <sup>1</sup> the measured amplitude of the voltage oscillation is plotted as a function of the driving frequency. The measured data for three different dc-offset values are indicated by circles, squares, and triangles, respectively. Figure 1(a) shows the results obtained from measurements



FIG. l. The squared amplitude of the measured voltage oscillation is shown as a function of the difference between the frequency of the external modulation signal  $f_{ac}$  and the internal critical frequency  $f_1$  (a) and  $f_2$  (b). The symbols indicate measured data, the curves are obtained by Lorentzian fits. For the three curves in each figure  $\Delta j_{\text{dc}} = (j_{ci} - j_{\text{dc}})$ ,  $(i = 1,2)$  is de creased from  $\Delta j_{\text{dc}} = 27.4 \mu\text{A/cm}^2$  (triangles) to  $\Delta j_{\text{dc}} = 17.1$  $\mu$ A/cm<sup>2</sup> (squares) to  $\Delta j_{\text{dc}} = 10.3 \mu$ A/cm<sup>2</sup> (circles). The amplitude of the modulation signal is  $j_{ac} = 0.034 \mu A/cm^2$  for all curves. (a) Results from measurement below, but close to, the first Hopf bifurcation with  $j_{c1} = 0.566$  mA/cm<sup>2</sup> and  $f_1 = 216$ mHZ. (b) Results from measurements below, but close to, the second Hopf bifurcation with  $j_{c2} = 3.1$  mA/cm<sup>2</sup> and  $f_2 = 365$ mHz.

near the first Hopf bifurcation and Fig. 1(b) the results from near the second bifurcation. We note here that in both cases the chosen value of the amplitude  $j_{ac}$  is much both cases the chosen value of the amplitude  $f_{ac}$  is much smaller than the smallest deviation  $\Delta f_{dc} = f_{ci} - f_{dc}$  $(i = 1,2)$  of the dc offset from the critical value. This means that the ac and dc components of the current density combined are by themselves not sufficient to induce the internal oscillation. Rather, the system reacts to the periodic excitation by exhibiting a resonance bebavior. The heights of the resonance peaks increase as  $j_{dc}$  approaches  $j_{ci}$ , indicating an increase in the amplification of the input signal. Simultaneously the widths decrease, which means that the system's frequency selectivity is enhanced.

In order to study the resonance behavior quantitatively, we try to fit Lorentzians<sup>1-3</sup> to the squared oscillation amplitude values. The curves in Fig. <sup>1</sup> are Lorentzians of the form

$$
S(\Delta) = \frac{K}{\Delta^2 + \varepsilon^2} ,
$$

fitted by a least-square method. Here  $S$  is the squared voltage amplitude,  $\Delta = (f_{ac} - f_{ci})$  is the frequency detuning, and K and  $\varepsilon$  are fit parameters. For each curve we obtain a K value to within an error of less than  $1\%$ . For all three curves in Figs. 1(a) and 1(b), respectively, the K values are nearly the same to within an error of 12%. The constancy of  $K$  is expected for Lorentzian-type resonances as the current density amplitude is kept constant. The determination of  $\varepsilon$  for each curve yields errors of about 8%-10%. The errors in  $\varepsilon$  are largest when  $i_{\text{dc}}$  is nearest to  $j_{c1}$  or  $j_{c2}$ . For the three different curves within Figs. 1(a) and 1(b) we get different  $\varepsilon$  values.

The quantity  $\varepsilon$  has the property of a bifurcation parameter, $2$  being related to the deviation of the control parameter from its critical value.  $\varepsilon$  is zero at the bifurcation and should increase with the deviation. We compare the  $\varepsilon$ values obtained from the Lorentzian fits with the corresponding values of the deviation  $\Delta j_{\text{dc}}$  and find a definite dependence of  $\varepsilon$  on  $\Delta j_{\text{dc}}$ . At the first bifurcation  $\varepsilon$  increases linearly with  $\Delta j_{\text{dc}}$  to within an error of less than 1%. At the second bifurcation a linear plot yields errors of more than 10%; a better fit with an error of only 2.5% is obtained for a scaling relation of the form  $\varepsilon \propto \Delta t \, \mathrm{d} \varepsilon^3$ .

Increasing the excitation amplitude we find a saturation of the small-signal amplification. We attribute this to the influence of nonlinear effects, as has been recently investigated generally by Bryant and Wiesenfeld.<sup>5</sup> On the other hand, a suppression of the instability or a shift of the bifurcation point due to the external excitation are not found in our case. More detailed experimental results will be published elsewhere.<sup>12</sup>

We have shown that in the driven electrical conductivity of BSN crystals the sensitivity to periodic small-signal excitations is enhanced near the first and the second Hopf bifurcation. The shape of the resonance peaks can be approximated by Lorentzians in both cases. We obtain a clear dependence of the height and width of the resonance curve on the deviation of the control parameter from the critical point.

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