

# An investigation into the equilibrium behaviour of iodine and chlorine dioxide

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## Abstract

## 1 Introduction

The concept of equilibrium was introduced in 1803 when French chemist Claude Berthollet found that chemical reactions are reversible [1]. To be at equilibrium, each reactant and product within a chemical reaction must be at equilibrium and the rate of the forward reaction must equal the rate of the reverse reaction. An important feature of a chemical reaction is how it behaves over time and whether or not it reaches equilibrium, but it is often difficult to experimentally determine this. Instead, it is possible to model chemical reactions using mathematical techniques which can more easily handle a process having numerous parameters and very large or small time requirements.

In 1906, William Bray first reported the relationship between iodine and chlorine dioxide when he used the oxidation of iodide ions by chlorine dioxide to assist in the analytical determination of chlorine dioxide [2]. This reaction can be modelled by the following ODE system which will be studied in this investigation:

$$\begin{aligned}\frac{dX}{dt} &= K_1 - K_2X - \frac{4K_3XY}{U + X^2} \\ \frac{dY}{dt} &= K_2X - \frac{K_3XY}{U + X^2}\end{aligned}$$

where  $X$  and  $Y$  are the concentrations of iodine and chlorine dioxide, respectively,  $t$  is time, and  $K_1, K_2, K_3$  and  $U$  are positive constants.

In this report we will analyse this system using techniques from bifurcation theory and numerical analysis in order to investigate the above relationship between iodine and chlorine dioxide over time. This system is derived from the chlorine dioxide-iodine-malonic acid reaction, which has been heavily studied due to its oscillatory properties and its demonstration of symmetry-breaking, reaction-diffusion Turing patterns [4]. Our primary focus is to investigate the hypothesis that this reaction tends monotonically towards equilibrium. As this reaction is known to be oscillatory in nature, we will utilise sensitivity analysis to determine whether there are points at which the reaction switches from oscillatory behaviour to reach equilibrium.

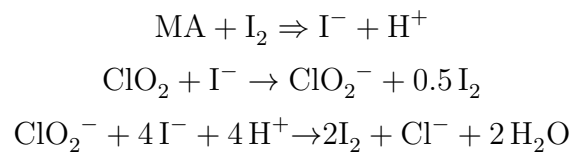
## 2 Background

### 2.1 Bifurcation Theory

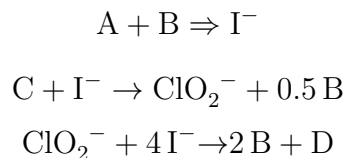
Bifurcations are defined as *qualitative* change in behaviour of the solutions to a system of ODE's, as parameters are varied. In this scenario, the parameters to be varied are the positive constants  $K_1, K_2, K_3$  and  $U$ . We use bifurcation theory in order to study both the behaviour of chemical reactions as they tend towards their equilibrium, as well as determining the equilibrium points of the chemical reactions themselves. Consequently, we can apply bifurcation theory to the iodine - chlorine dioxide chemical reaction; determine the equilibrium points of the reaction, and explore the hypothesis that chemical reactions tend monotonically towards equilibrium.

### 2.2 Model Description

The chlorine dioxide-iodine-malonic acid reaction can be represented with the following model, known as the Lengyel-Epstein model [4]:



To turn this system into a system of differential equations, it is assumed that not all of the reactants react at the same rate and so the reactants that react comparatively slower can be treated as constants. In these systems,  $\text{MA}, \text{ClO}_2$  and  $\text{I}_2$  are treated as constants and as a result (as well as ignoring  $\text{H}^+, \text{Cl}^-$  and  $\text{H}_2\text{O}$ ) we focus on  $\text{ClO}_2^-$  and  $\text{I}^-$ . The resulting system can then be written as:



From this, the change in concentration of  $\text{ClO}_2^-$  and  $\text{I}^-$  can be found which gives us the resultant ODE system, which is unusual because it is substrate-inhibited<sup>1</sup> and autocatalytic<sup>2</sup>, as well as being oscillatory in

<sup>1</sup>Substrate-inhibited: The product of an enzyme reaction inhibits the enzyme's activity

<sup>2</sup>Autocatalytic: One of the reaction products is also a catalyst for the same or a coupled reaction

nature and producing Turing structures [3, 2].

## 3 Mathematical Methods

### 3.1 Visualisation of Process

In order to garner a higher intuition for the behaviour of our system it is valuable to visualise the equation dependence on the parameters involved. As such MATLAB graphing software is employed to provide this insight, results are displayed in section 4.

### 3.2 Non-dimensionalisation

The ODE system can be non-dimensionalised in order to reduce the number of parameters in the system in hopes of making analysis easier as it is preferable to see how a solution changes in response to fewer parameters. The method for non-dimensionalising is in Appendix section 6.1. This process produces the following system:

$$\begin{aligned}\frac{d\hat{X}}{d\hat{t}} &= m - \hat{X} - \frac{4\hat{X}\hat{Y}}{1 + \hat{X}^2} \\ \frac{d\hat{Y}}{d\hat{t}} &= n \left( \hat{X} - \frac{\hat{X}\hat{Y}}{1 + \hat{X}^2} \right)\end{aligned}$$

Where

$$m = \frac{K_1}{K_2\sqrt{U}}, n = \frac{K_3}{K_2\sqrt{U}}, X = \sqrt{U}\hat{X}, Y = \frac{K_2U}{K_3}\hat{Y}, t = \frac{1}{K_2}\hat{t}$$

### 3.3 Fixed points

The fixed points of the system can be found using the non-dimensionalised system. The fixed points are the points where  $\frac{d\hat{X}}{d\hat{t}} = 0$  and  $\frac{d\hat{Y}}{d\hat{t}} = 0$ . This process is in Appendix section 6.2 and results in:

$$(\hat{X}_*, \hat{Y}_*) = \left( \frac{m}{5}, 1 + \frac{m^2}{25} \right)$$

This is equivalent to

$$(X_*, Y_*) = \left( \frac{K_1}{5\sqrt{U}K_2}, 1 + \frac{K_1^2}{25UK_2^2} \right)$$

### 3.4 Jacobian

The Jacobian of the non-dimensionalised system was found in Appendix section 6.3. The general Jacobian is  $J(x) =$

$$\begin{bmatrix} -1 - 4\hat{Y} \left( \frac{1-\hat{X}^2}{(1-\hat{X}^2)^2} \right) & \frac{-4\hat{X}}{1+\hat{X}^2} \\ n - n\hat{Y} \left( \frac{1-\hat{X}^2}{(1-\hat{X}^2)^2} \right) & \frac{-n\hat{X}}{1+\hat{X}^2} \end{bmatrix}$$

The Jacobian at the value of the fixed point is  $J\left(\frac{m}{5}, 1 + \frac{m^2}{25}\right) =$

$$\begin{bmatrix} \frac{3m^2-125}{25+m^2} & \frac{-20m}{25+m^2} \\ \frac{2nm^2}{25+m^2} & \frac{-5nm}{25+m^2} \end{bmatrix}$$

### 3.5 Eigenvalues of the Jacobian

The eigenvalues of this problem can be found by taking the determinant of the above Jacobian. This process is in Appendix section 6.4 and produces the eigenvalues:

$$\lambda_{1,2} = \frac{3m^2 - 5nm - 125 \pm \sqrt{(5nm - 3m^2 + 125)^2 - 4(25nm^3 + 625nm)}}{50 + 2m^2}$$

Eigenvalues play an important part in determining the stability of a steady state. A steady state  $(\hat{X}_*, \hat{Y}_*)$  is hyperbolic if all the eigenvalues of the Jacobian  $J(\hat{X}_*, \hat{Y}_*)$  have non-zero real part. Given that a steady state is hyperbolic, the Hartman-Grobman Theorem (HG Thm) applies. The HG Thm states that given that a steady state is hyperbolic, then in a small neighbourhood of the steady state, the phase portrait of the non-linear system is equivalent to the linearised system.

For the non-dimensionalised system, both  $m, n$  are positive given that  $K_1, K_2, K_3, U$  are positive. It is easy to observe that  $\lambda_{1,2} \neq 0$  which implies that the steady state  $\left(\frac{m}{5}, 1 + \frac{m^2}{25}\right)$  is hyperbolic. This non-linear system can be analysed using the linearised system.

In general, the eigenvalues of the Jacobian are complex [5]. The complex part of the eigenvalues determine the oscillatory behaviour, while the real part determines the stability. The stability depends on whether the real part of the eigenvalues are negative or positive. Let the real part of the eigenvalue for this system be  $\alpha$ .

$$Re(\lambda_{1,2}) = \alpha = \frac{3m^2 - 5nm - 125}{50 + 2m^2}$$

If  $\alpha$  is positive, the steady state is unstable.  $\alpha$  is positive when,

$$\frac{3m^2 - 5nm - 125}{50 + 2m^2} > 0 \Rightarrow n < \frac{3m}{5} - \frac{25}{m}$$

If  $\alpha$  is negative, the steady state is stable.  $\alpha$  is negative when,

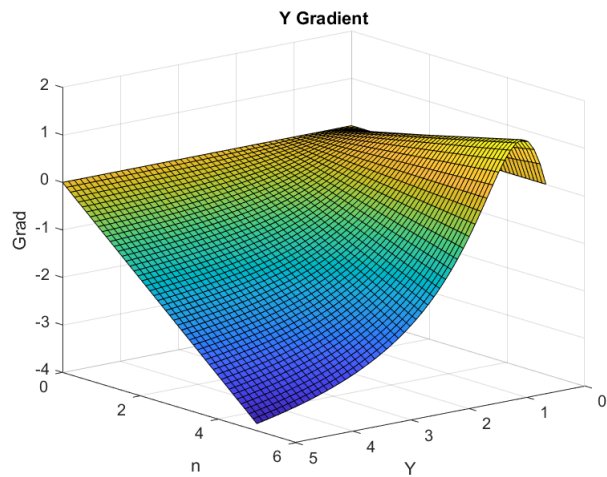
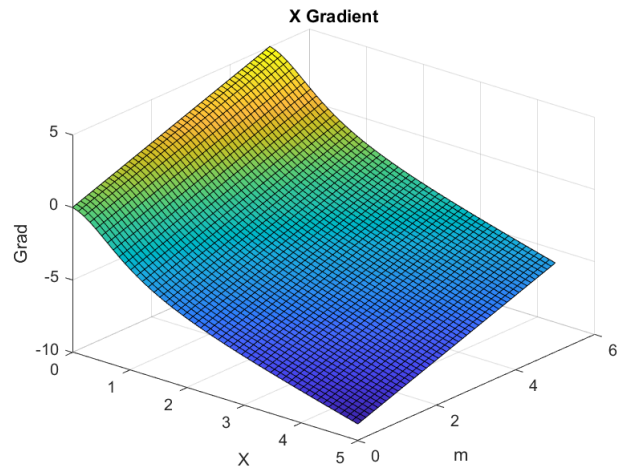
$$\frac{3m^2 - 5nm - 125}{50 + 2m^2} < 0 \Rightarrow n > \frac{3m}{5} - \frac{25}{m}$$

We will now consider values of  $m, n$  that will result in stable and unstable behaviour. This will be discussed in the Section 4.

## 4 Results

### 4.1 Visualization

In plotting the non-dimensional chemical population gradient dependence on the non-dimensional parameters  $m$  and  $n$  the following gradient plots were created:





Phase portraits for various values of  $m$  and  $n$  in the non-dimensionalised system are plotted below. The steady state for each plot is denoted by a red star.

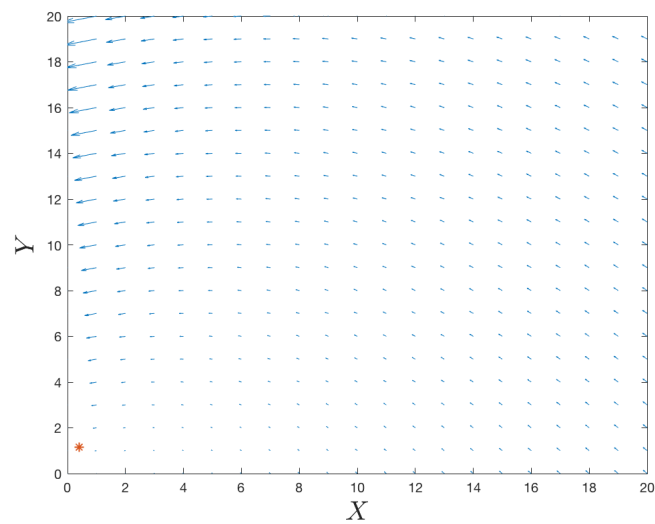


Figure 1:  $m = 2, n = 5$

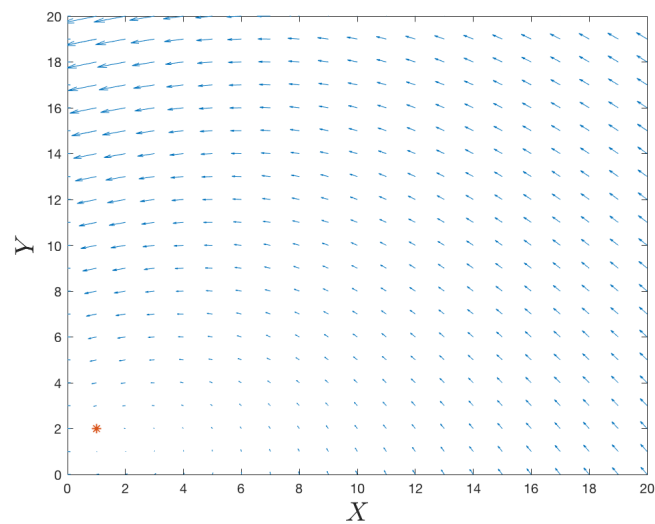
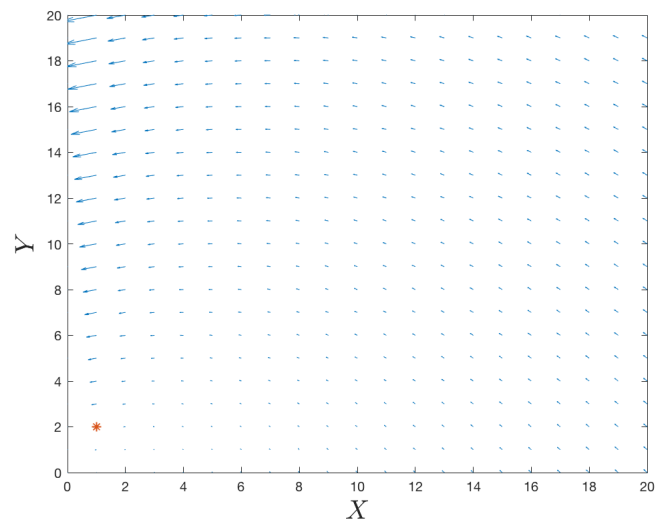
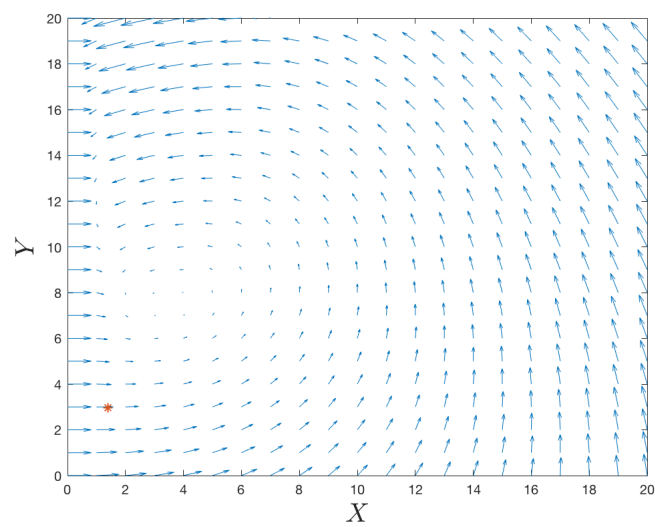


Figure 2:  $m = 5, n = 2$

Figure 3:  $m = 5, n = 7$ Figure 4:  $m = 7, n = 1$ 

## 5 Conclusions

## 6 Appendix

### 6.1 Non-dimensionalisation

$$\frac{dX}{dt} = K_1 - K_2X - \frac{4K_3XY}{U + X^2} \quad (1)$$

$$\frac{dY}{dt} = K_2X - \frac{K_3XY}{U + X^2} \quad (2)$$

Let  $X = X_c\hat{X}$ ,  $Y = Y_c\hat{Y}$ ,  $t = t_c\hat{t}$  ( $\star$ ) and note that  $\frac{d\hat{t}}{dt} = \frac{1}{t_c}$   
Substitute ( $\star$ ) into (1):

The LHS is:

$$\frac{dX}{dt} = \frac{X_c}{t_c} \frac{d\hat{X}}{d\hat{t}}$$

We combine this with the RHS and get

$$\frac{d\hat{X}}{d\hat{t}} = \frac{t_c}{X_c} \left[ K_1 - K_2X_c\hat{X} - \frac{4K_3X_c\hat{X}Y_c\hat{Y}}{U + X_c^2\hat{X}^2} \right]$$

$$\frac{d\hat{X}}{d\hat{t}} = \frac{t_c}{X_c} K_1 - t_c K_2 \hat{X} - \frac{t_c K_3 Y_c 4\hat{X}\hat{Y}}{U(1 + \frac{X_c^2}{U}\hat{X}^2)}$$

Now let

$$\frac{X_c^2}{U} = 1 \rightarrow X_c = \sqrt{U}$$

$$t_c K_2 = 1 \rightarrow t_c = \frac{1}{K_2}$$

$$\frac{t_c K_3 Y_c}{U} = 1 \rightarrow Y_c = \frac{U}{t_c K_3} = \frac{K_2 U}{K_3}$$

Then let the constant m be

$$m = \frac{t_c}{X_c} K_1 = \frac{K_1}{K_2 \sqrt{U}}$$

Thus, the non-dimensional version of (1) is

$$\frac{d\hat{X}}{d\hat{t}} = m - \hat{X} - \frac{4\hat{X}\hat{Y}}{1 + \hat{X}^2}$$

Solving similarly for (2):

$$\frac{d\hat{Y}}{d\hat{t}} = \frac{t_c}{Y_c} \left[ K_2 X_c \hat{X} - \frac{k_3 X_c \hat{X} Y_c \hat{Y}}{U + X_c^2 \hat{X}^2} \right]$$

$$\frac{d\hat{Y}}{d\hat{t}} = \frac{t_c}{Y_c} K_2 X_c \hat{X} - \frac{K_3 X_c t_c \hat{X} \hat{Y}}{U(1 + \frac{X_c^2}{U} \hat{X}^2)}$$

Substituting in the values of  $X_c = \sqrt{U}$ ,  $t_c = \frac{1}{K_2}$ ,  $Y_c = \frac{K_2 U}{K_3}$  we get

$$\frac{d\hat{Y}}{d\hat{t}} = \frac{K_3 K_2 \sqrt{U}}{K_2 K_2 U} \hat{X} - \frac{K_3 \sqrt{U} \hat{X} \hat{Y}}{K_2 U(1 + \hat{X}^2)}$$

$$\frac{d\hat{Y}}{d\hat{t}} = \frac{K_3}{K_2 \sqrt{U}} \hat{X} - \frac{K_3 \hat{X} \hat{Y}}{K_2 \sqrt{U}(1 + \hat{X}^2)}$$

Let the constant  $n$  be

$$n = \frac{K_3}{K_2 \sqrt{U}}$$

Then the non-dimensional version of (2) is

$$\frac{d\hat{Y}}{d\hat{t}} = n \left( \hat{X} - \frac{\hat{X} \hat{Y}}{1 + \hat{X}^2} \right)$$

The dimensionless version of the original equation is thus:

$$\frac{d\hat{X}}{d\hat{t}} = m - \hat{X} - \frac{4\hat{X}\hat{Y}}{1 + \hat{X}^2}$$

$$\frac{d\hat{Y}}{d\hat{t}} = n \left( \hat{X} - \frac{\hat{X} \hat{Y}}{1 + \hat{X}^2} \right)$$

Where

$$m = \frac{K_1}{K_2 \sqrt{U}}, n = \frac{K_3}{K_2 \sqrt{U}}, X = \sqrt{U} \hat{X}, y = \frac{K_2 U}{K_3} \hat{Y}, t = \frac{1}{K_2} \hat{t}$$

## 6.2 Fixed points

To find the fixed points we set  $\frac{d\hat{X}}{d\hat{t}} = 0$  and  $\frac{d\hat{Y}}{d\hat{t}} = 0$

So

$$\frac{d\hat{Y}}{d\hat{t}} = 0 \rightarrow n \left( \hat{X} - \frac{\hat{X} \hat{Y}}{1 + \hat{X}^2} \right) = 0$$

$$n\hat{X} = \frac{n\hat{X}\hat{Y}}{1 + \hat{X}^2}$$

$$1 = \frac{\hat{Y}}{1 + \hat{X}^2}$$

And so  $\hat{Y}_* = 1 + \hat{X}^2$

And also

$$\frac{d\hat{X}}{d\hat{t}} = 0 \rightarrow m - \hat{X} - \frac{4\hat{X}\hat{Y}}{1 + \hat{X}^2}$$

We substitute in  $\hat{Y}_* = 1 + \hat{X}^2$  to get

$$m - \hat{X} - \frac{4\hat{X}(1 + \hat{X}^2)}{1 + \hat{X}^2} = 0$$

$$m - \hat{X} - 4\hat{X} = 0$$

$$m - 5\hat{X} = 0 \rightarrow \hat{X}_* = \frac{m}{5}$$

And so then  $\hat{Y}_* = 1 + \frac{m^2}{25}$

So thus we have a fixed point at

$$(\hat{X}_*, \hat{Y}_*) = \left( \frac{m}{5}, 1 + \frac{m^2}{25} \right)$$

As we have previously let  $m = \frac{K_1}{\sqrt{UK_2}}$ , this is equivalent to

$$(X_*, Y_*) = \left( \frac{\frac{K_1}{\sqrt{UK_2}}}{5}, 1 + \frac{\left(\frac{K_1}{\sqrt{UK_2}}\right)^2}{25} \right)$$

$$(X_*, Y_*) = \left( \frac{K_1}{5\sqrt{UK_2}}, 1 + \frac{K_1^2}{25UK_2^2} \right)$$

### 6.3 Jacobian

Our non-dimensionalised system is:

$$\frac{d\hat{X}}{d\hat{t}} = m - \hat{X} - \frac{4\hat{X}\hat{Y}}{1 + \hat{X}^2} = m - \hat{X} - 4\hat{Y} \left( \frac{\hat{X}}{1 + \hat{X}^2} \right) = f(\hat{X}, \hat{Y})$$

$$\frac{d\hat{Y}}{d\hat{t}} = n \left( \hat{X} - \frac{\hat{X}\hat{Y}}{1 + \hat{X}^2} \right) = n\hat{X} - n\hat{Y} \left( \frac{\hat{X}}{1 + \hat{X}^2} \right) = g(\hat{X}, \hat{Y})$$

The Jacobian takes the form  $J(\underline{x}) =$

$$\begin{bmatrix} f_X(\underline{x}) & f_Y(\underline{x}) \\ g_X(\underline{x}) & g_Y(\underline{x}) \end{bmatrix}$$

For this system the derivatives are

$$f_{\hat{X}} = -1 - 4\hat{Y} \left( \frac{(1 + \hat{X}^2)(1) - \hat{X}(2\hat{X})}{(1 + \hat{X}^2)^2} \right)$$

$$\begin{aligned}
&= -1 - 4\hat{Y} \left( \frac{1 + \hat{X}^2 - 2\hat{X}^2}{(1 + \hat{X}^2)^2} \right) \\
&= -1 - 4\hat{Y} \left( \frac{1 - \hat{X}^2}{(1 - \hat{X}^2)^2} \right)
\end{aligned}$$

Therefore,

$$f_{\hat{X}} = -1 - 4\hat{Y} \left( \frac{1 - \hat{X}^2}{(1 - \hat{X}^2)^2} \right)$$

and

$$f_{\hat{Y}} = \frac{-4\hat{X}}{1 + \hat{X}^2}$$

and

$$\begin{aligned}
g_{\hat{X}} &= n - n\hat{Y} \left( \frac{(1 + \hat{X}^2)(1) - (\hat{X})(2\hat{X})}{(1 - \hat{X}^2)^2} \right) \\
&= n - n\hat{Y} \left( \frac{1 - \hat{X}^2}{(1 - \hat{X}^2)^2} \right)
\end{aligned}$$

Therefore,

$$g_{\hat{X}} = n - n\hat{Y} \left( \frac{1 - \hat{X}^2}{(1 - \hat{X}^2)^2} \right)$$

and

$$g_{\hat{Y}} = \frac{-n\hat{X}}{1 + \hat{X}^2}$$

Therefore the Jacobian is:  $J(\underline{x}) =$

$$\begin{bmatrix}
-1 - 4\hat{Y} \left( \frac{1 - \hat{X}^2}{(1 - \hat{X}^2)^2} \right) & \frac{-4\hat{X}}{1 + \hat{X}^2} \\
n - n\hat{Y} \left( \frac{1 - \hat{X}^2}{(1 - \hat{X}^2)^2} \right) & \frac{-n\hat{X}}{1 + \hat{X}^2}
\end{bmatrix}$$

Then  $J\left(\frac{m}{5}, 1 + \frac{m^2}{25}\right) =$

$$\begin{aligned}
&\begin{bmatrix}
-1 - 4\left(1 + \frac{m^2}{25}\right) \left( \frac{1 - \frac{m^2}{5}}{\left(1 - \frac{m^2}{5}\right)^2} \right) & \frac{-4\left(\frac{m}{5}\right)}{1 + \frac{m^2}{25}} \\
n - n\left(1 + \frac{m^2}{25}\right) \left( \frac{1 - \frac{m^2}{5}}{\left(1 - \frac{m^2}{5}\right)^2} \right) & \frac{-n\left(\frac{m}{5}\right)}{1 + \frac{m^2}{25}}
\end{bmatrix} \\
&= \begin{bmatrix}
\frac{-(1 + \frac{m^2}{25}) - 4\left(1 - \frac{m^2}{5}\right)}{\left(1 + \frac{m^2}{25}\right)} & \frac{-4m}{\frac{25 + m^2}{25}} \\
\frac{n\left(1 + \frac{m^2}{25}\right) - n\left(1 - \frac{m^2}{5}\right)}{\left(1 + \frac{m^2}{25}\right)} & \frac{-nm}{\frac{25 + m^2}{25}}
\end{bmatrix}
\end{aligned}$$

$$\begin{aligned}
&= \begin{bmatrix} \frac{-1 - \frac{m^2}{25} - 4 + \frac{4m^2}{25}}{25+m^2} & \frac{-4m}{5} \left( \frac{25}{25+m^2} \right) \\ \frac{n + \frac{nm^2}{25} - n + \frac{nm^2}{25}}{25+m^2} & \frac{-nm}{5} \left( \frac{25}{25+m^2} \right) \end{bmatrix} \\
&= \begin{bmatrix} \frac{-5 + \frac{3m^2}{25}}{25+m^2} & \frac{-20m}{25+m^2} \\ \frac{\frac{2nm^2}{25}}{25+m^2} & \frac{-5nm}{25+m^2} \end{bmatrix} \\
&= \begin{bmatrix} \left( \frac{-125+3m^2}{25} \right) \left( \frac{25}{25+m^2} \right) & \frac{-20m}{25+m^2} \\ \left( \frac{2nm^2}{25} \right) \left( \frac{25}{25+m^2} \right) & \frac{-5nm}{25+m^2} \end{bmatrix}
\end{aligned}$$

So then  $J\left(\frac{m}{5}, 1 + \frac{m^2}{25}\right) =$

$$\begin{bmatrix} \frac{3m^2-125}{25+m^2} & \frac{-20m}{25+m^2} \\ \frac{2nm^2}{25+m^2} & \frac{-5nm}{25+m^2} \end{bmatrix}$$

## 6.4 Eigenvalues of the Jacobian

We find

$$\begin{aligned}
\det J &= \left| \begin{bmatrix} \left( \frac{3m^2-125}{25+m^2} - \lambda \right) & \frac{-20m}{25+m^2} \\ \frac{2nm^2}{25+m^2} & \left( \frac{-5nm}{25+m^2} - \lambda \right) \end{bmatrix} \right| \\
&= \left( \frac{3m^2-125}{25+m^2} - \lambda \right) \left( \frac{-5nm}{25+m^2} - \lambda \right) - \left( \frac{-20nm}{25+m^2} \right) \left( \frac{2nm^2}{25+m^2} \right) = 0 \\
&= \frac{(3m^2-125)(-5nm)}{(25+m^2)^2} - \left( \frac{3m^2-125}{25+m^2} \right) \lambda + \left( \frac{5nm}{25+m^2} \right) \lambda + \lambda^2 + \frac{40nm^3}{(25+m^2)^2} = 0 \\
&= \frac{-15nm^3 + 625nm}{(25+m^2)^2} + \frac{-3m^2 + 125 + 5nm}{25+m^2} \lambda + \lambda^2 + \frac{40nm^3}{(25+m^2)^2} = 0 \\
&= \lambda^2 + \frac{5nm - 3m^2 + 125}{25+m^2} \lambda + \frac{40nm^3 - 15nm^3 + 625nm}{(25+m^2)^2} = 0 \\
&= \lambda^2 + \frac{5nm - 3m^2 + 125}{25+m^2} \lambda + \frac{25nm^3 + 625nm}{(25+m^2)^2} = 0
\end{aligned}$$

Hence then

$$\begin{aligned}
\lambda_{1,2} &= \frac{-\frac{5nm-3m^2+125}{25+m^2} \pm \sqrt{\left( \frac{5nm-3m^2+125}{25+m^2} \right)^2 - 4(1) \left( \frac{25nm^3+625nm}{(25+m^2)^2} \right)}}{2} \\
&= \frac{\frac{3m^2-5nm-125}{25+m^2} \pm \sqrt{(5nm-3m^2+125)^2 - 4(25nm^3+625nm)}}{(25+m^2)^2}}{2}
\end{aligned}$$

$$= \frac{3m^2 - 5nm - 125 \pm \sqrt{(5nm - 3m^2 + 125)^2 - 4(25nm^3 + 625nm)}}{2(25 + m^2)}$$

And hence

$$\lambda_{1,2} = \frac{3m^2 - 5nm - 125 \pm \sqrt{(5nm - 3m^2 + 125)^2 - 4(25nm^3 + 625nm)}}{50 + 2m^2}$$

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