Stochastische Resonanz
in gekoppelten Systemen
Stochastic Resonance
in Coupled Systems

Diplomarbeit
zur Erlangung des Grades eines Diplom-Physikers

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$A_i$ amplitude of driving force for particle $i$
$D$ diffusion constant
$F$ force
$H$ Hessian of $V$
$HLA$ hysteresis loop area
$K(t)$ transition matrix derived from Kramers rates
$P_{res}^i(t)$ residence time distribution of particle $i$
$R$ particle radius
$RES$ first peak area of residence time distribution
$T$ kinetic temperature
$U(x)$ double well potential
$V(x_1, x_2, t)$ complete potential
$V_i$ potential energy of state $i$
$X$ transformation matrix, $p(t) \rightarrow \langle x(t) \rangle$

$d$ distance between separate double wells
$e$ elementary charge
$f_i(t)$ harmonic driving force of particle $i$
$k$ Kramers rate
$k_B$ Boltzmann constant
$k_{ij}$ Kramers rate for transitions from state $j$ to state $i$
$m$ mass of particle
$p(t)$ probability distribution
$p_i(t)$ probability to find the system at time $t$ in state $i$
$r$ absolute distance between particles
$r_{ij}$ absolute distance between particles $i$ and $j$
$t$ continuous time
$t_n$ discrete time, $t_n = n \Delta t$
$x(t)$ system variable, $(x_1, x_2)(t)$
$x_0$ distance between well and barrier
$x_i(t)$ $x$-position of particle $i$
$x^i_n$ $x$-position of particle $i$ at time $t_n$
$x_{ij}(t)$ $x_i$ coordinate of state $j$

$\Delta t$ width of discrete time step
$\Delta U$ barrier height of double well potential
List of Symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>$\Delta x$</td>
<td>distance in $x$-direction, $\Delta x = x_1 - x_2$</td>
</tr>
<tr>
<td>$\Phi(</td>
<td>x_1 - x_2</td>
</tr>
<tr>
<td>$\Omega$</td>
<td>angular frequency of the driving force</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>radius of integration for $RES$</td>
</tr>
<tr>
<td>$\alpha_i$</td>
<td>correction of system coordinate assigned to state $i$</td>
</tr>
<tr>
<td>$\beta$</td>
<td>inverse temperature, $\beta = (k_B T)^{-1}$</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>dampening constant</td>
</tr>
<tr>
<td>$\delta_{ij}$</td>
<td>Kronecker symbol</td>
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<tr>
<td>$\delta(t-s)$</td>
<td>delta distribution</td>
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<tr>
<td>$\epsilon$</td>
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<tr>
<td>$\eta$</td>
<td>friction coefficient, $\eta = m \gamma$</td>
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<td>$\kappa$</td>
<td>screening constant, inverse Debye length</td>
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<tr>
<td>$\lambda_B$</td>
<td>Bjerrum length</td>
</tr>
<tr>
<td>$\nu$</td>
<td>dynamical viscosity</td>
</tr>
<tr>
<td>$\xi(t)$</td>
<td>stochastic force, white noise</td>
</tr>
<tr>
<td>$\dot{\xi}_n$</td>
<td>stochastic velocity increment of particle $i$ at time $t_n$</td>
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<tr>
<td>$\sigma^2$</td>
<td>variance</td>
</tr>
<tr>
<td>$\tau$</td>
<td>period of driving force</td>
</tr>
<tr>
<td>$\phi$</td>
<td>coupling constant, strength of screened Coulomb potential</td>
</tr>
<tr>
<td>$\varphi_i$</td>
<td>phase of driving force of particle $i$</td>
</tr>
<tr>
<td>$\omega_\alpha$</td>
<td>angular frequency of the metastable state</td>
</tr>
<tr>
<td>$\omega^a_i$</td>
<td>angular frequencies of the eigenmodes of the metastable state</td>
</tr>
<tr>
<td>$\omega_b$</td>
<td>positive-valued angular frequency of the instable state/at the barrier/at the saddle point</td>
</tr>
<tr>
<td>$\omega^s_i$</td>
<td>angular frequencies of the stable eigenmodes of the saddle point</td>
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1. Introduction

1.1. Stochastic resonance—when noise is not a nuisance

Generations of scientists and engineers have worked hard on the problem of noise reduction. Wherever encoded information (termed signal) is processed, e.g. at a radio-station or in a measuring instrument, there is an inevitable loss of quality due to external and internal disturbances (termed noise). The primary technical solutions would be either to amplify the input (e.g. maximal transmitting power) or to improve the screening from external disturbances. The presence of internal noise cannot be totally suppressed as long as temperature is a non-vanishing quantity. This is important especially for all systems where the signal power input is weak relative to the noise of the environment which itself is related to the thermal energy of its constituents. This thermal energy is the main source of internal disturbance, as it provides an energy reservoir that allows interaction between signal and system leading to phenomena known as fluctuation and dissipation. If these interactions must be taken into account explicitly this is physically done by modelling a heat bath as an idealised environment in contact to the pure system.

In contrast to these annoying side-effects there are circumstances when noise takes over a substantial and supporting role, if not a leading one. Not only is noise responsible for effects like diffusion and Brownian motion, it also is an indispensable part of reaction rate theory where thermal noise is the agent of thermally activated escape over a potential barrier—noise is a trigger of chemical reactions! The last point may also be seen in analogy to the physical problem of decay of metastable states and leads to the following Gedankenexperiment: Imagine a dynamical system with some input signal that is so weak that in the absence of noise there will be no detectable signal output at all. If noise is added to the system in an amount which exceeds the signal power by far, there will be much output noise effectively covering any signal information. But could it be possible that a specific noise level exists, strong enough to provide thermal energy for the system to be activated by the signal, and still weak enough not to bury the signal completely?

Similar thoughts might have guided Benzi et al. in the early 1980s when looking for an applicable mechanism that would serve as a trigger for the switching in the Earth’s climate. With a period of about 100,000 years the change between warm climate and ice ages correlates with the variation in the eccentricity of the Earth’s orbit. However, the direct influence of the variation in eccentricity is too weak to produce such dramatic effects like ice ages. Only if it was possible to find conditions which allow for a cooperative amplification of these effects, then such a triggered system could be a valid model. The next step was, of course, to present
1. Introduction

Figure 1.1.: Stochastic resonance as a trial explanation for ice ages; a weak input signal (eccentricity) leading to strong variations in the output signal (temperature).

a model system according to the required conditions. It concerned a bistable “climatic potential”, two sources of input (the variation in eccentricity as a signal and random fluctuations as noise) and a clear-cut output signal against the noisy background that actually showed the hoped for strong periodicity (cf. fig. 1.1). This mechanism was further on referred to as stochastic resonance. Because of the complexity of modelling climatic processes the question whether the quasi-periodic occurrence of ice ages can be explained by stochastic resonance is still under discussion and far from being decided.

Today, stochastic resonance is a well established concept on its own. A comprehensive description in form of a “Theory of stochastic resonance” was given by McNamara and Wiesenfeld in 1989 [1] for two basic kinds of one-dimensional dynamical systems considering a continuous as well as a discrete variable. Early experimental realisations had been a Schmitt trigger electronic circuit and a bidirectional ring laser system. Stochastic resonance was even found applicable to biological systems like single neurones (nerve cells) or to quantum systems like superconducting quantum interference devices (SQUIDs) [2].

For statistical physics, experiments involving colloidal particles trapped by optical tweezers are of particular interest for several reasons:

- Micro beads suspended in an aqueous solution perform Brownian motion due to uncountable random collisions with the surrounding water molecules. The system size is optimal in the sense of a compromise: small enough to give a measurable response (dislocation) to an appropriate amount of molecular collisions, and large enough to detect the resulting motion by standard means (optical microscope plus digital camera).

- By use of refraction, laser beams generate a punctiform attractive force field acting on the micro beads. In combination with acoustooptic deflectors they represent a highly flexible device for creating a potential landscape in every desired shape (as long as it is sufficiently smooth and bounded).

- Therefore such a system includes all the premises for experimental verification of theoretical findings: it has a manageable size; all relevant parameters
are conveniently accessible; the underlying statistical effects are not artificially designed—they originate directly from atomic/molecular motion.

And last but not least, under suitable conditions, such a system exhibits stochastic resonance. This was experimentally shown for a single colloidal particle in a double well potential by the group of Clemens Bechinger at 2. Physikalisches Institut, Universität Stuttgart [3].

1.2. Stochastic resonance in coupled systems

For a start, single particle systems are absolutely fine. They permit an elementary approach under exclusion of further external influences. But the need for more complex models is indisputable: Nature has taught us that in her realm the richest complexity evolves from the simplest rules. Even within plain biological systems, the single neurones are never isolated but partake in a multitude of interactions via electrical sensory stimuli or biochemical processes, e.g. receptor bonds, exchange of messenger proteins, differences in ion concentration, etc. A more realistic model should therefore incorporate an explicit internal interaction term for more than a single particle. And this is the task of the present diploma thesis.

As a matter of fact, the micro beads used as colloidal particles are electrically charged. For this reason a system consisting of two or more micro beads automatically contains an intrinsic interaction potential, unless the charges are not totally screened or otherwise “switched off”. This provides an opportunity for a realistic model system comprising exactly two interacting (and thereby coupled) particles, in a manner of speaking, a hydrogen atom for stochastic resonance.

Admittedly, the last remark should not suggest that the present thesis impudently aims at a complete and self-consistent theory of stochastic resonance in coupled systems. Rather, it is a humble theoretical investigation of specific elementary aspects of stochastic resonance in coupled systems. Those aspects are, in few words:

- dependence of stochastic resonance on the coupling strength,
- dependence of stochastic resonance on the phase between driving forces,
- combined dependence on coupling strength and driving frequency, and
- reliability of different quantifiers.

The means of investigation applied to the problem are numerical integrations of two different kinds which are derived from:

- a Langevin equation,
- a model based on Kramers rates.
1. Introduction

Since there exists a variety of bona fide quantifiers for stochastic resonance without any axiomatic predetermination, each data analysis should be tested by several quantifiers in order to distinguish between more or less appropriate measures. A prominent member of the quantifier family is the signal-to-noise ratio which compares the output power at the signal frequency to the total power of the background noise. This method often requires a cutoff frequency, in particular if the stochasticity of the process relies on white noise—as it does in most theoretical cases. Then the noise spectral density does not vanish at high frequencies and the integral inevitably becomes infinite.

Keeping in mind the additional value of a system of coupled colloidal particles as a model system for more general problems of statistical physics, quantifiers which are closer related to directly measurable quantities and thereby more intuitive are utilised throughout this thesis. These quantifiers are:

- hysteresis loop area,
- residence time distribution.

A direct connection of the hysteresis loop area with the signal-to-noise ratio and the spectral power density (another quantifier) has been shown for weak, purly harmonic driving [4]. Under these conditions (i.e. within the regime of linear response) each of the three quantifiers emanates from the other two. In order to properly estimate the reliability of the applied quantifiers it is certainly not a bad idea to choose a second quantifier not belonging to this trio. This applies for the selected residence time distribution.
2. The Models

2.1. The experimental setup

Although this thesis is a work in theoretical physics, it is nevertheless closely related to experiment by collaboration with the group of Clemens Bechinger at Universität Stuttgart. An experimental realisation of a system of coupled colloidal particles is in progress as main part of the diploma thesis of Tobias Sawetzki. Here, I will present a brief outline of the experimental setup at Stuttgart [3, 5].

![Experimental Setup Diagram]

**Figure 2.1.** Core of experimental setup; the deflected laser beam enters from above and is focussed within the sample cell; the trap is occupied by a silica micro bead (drawn in disproportionate size); the lower lens is for observation and the laser beam is filtered out.

The setup is composed of three main parts:

1. a pair of optical tweezers,
2. the sample cell as shown in fig. 2.1, and
3. a data acquisition system.

First I will describe the core of the experiment: the sample cell. The aqueous suspension is fed in by a standard deionising circuit. The particles consist of silica with \((1.57 \pm 0.06)\) μm diameter. Within the sample cell they sink until the electrostatic repulsion of the lower wall stops them. The photonic pressure of the laser beam pushes them further down to a distance from the bottom of approximately 500 nm.

For spatially extended operation of the optical tweezers, laser light of wavelength 532 nm is deflected by two acoustooptical deflectors (AOD). The AODs can be
operated with a switching rate of 50kHz. This rate is significantly larger than the inverse of the typical relaxation time of our system so that the effect of the tweezers on the particles is quasi-static. The laser beam is guided into an optical microscope where it is focussed into the cuvette that contains liquid and particles.

The optical tweezers can operate within a plane of $150 \mu m \times 150 \mu m$, with nanometer resolution and repeatability on the order of 100 nm. To build a double well trap, two pairs of tweezers are needed in an appropriate distance. Problems with interference of laser beams do not occur because only one time split beam is used so that at all times the light is focussed to one single spot. The intensity of each tweezer is varied in 20 steps which allows for a quasi-harmonic modulation of the generated potential.

Experimental data is recorded by a CCD camera at a speed of 20 frames per second and further processed by a PC that also controls the AODs. Experiments are performed at room temperature with stability of about $\pm 1K$.

### 2.2. The potential landscape

In vertical direction ($z$-axis) the particle position is always balanced between four forces: gravity and light pressure push it down whereas electrostatic repulsion and buoyancy push it up. The gradient force of the refracted laser beam acts merely in the lateral directions ($x,y$)-plane), so the whole arrangement can easily be regarded as a two-dimensional setup within the focal plane.

![Figure 2.2.](image)

**Figure 2.2.** Lateral section of sample cell: a quartet of optical traps (large spots) forms two double wells (dashed circles).

In a setup for two particles where each particle is confined to a double well, we need four overlapping point traps as shown in fig. 2.2. Two point traps define an axis through the centres of each well. The axes are denoted $x_1$ and $x_2$, respectively. The well to well distance is two times the distance from a well minimum to the saddle point, i.e. $2x_0$. The perpendicular spacing between both axes is denoted
with $d$. We refer to this quantity whenever we speak of the “distance between double wells”.

Yet another reduction of dimension results from the assumption that the trajectory of each particle almost always stays close to the most probable path. Therefore intrawell dynamics may be described by a motion that mirrors exclusively the $x$-coordinate of the particle contrary to the $y$-coordinate which is supposed to fluctuate independently around its relative maximum. As a consequence we can neglect these fluctuations and resort to a mere one-dimensional model for each particle.

![Figure 2.3: Approximation of two Gaussian wells ($g_+$ and $g_-$) and their sum at the region of mutual overlapping by a polynomial of degree four ($f$).](image)

The standard means of modelling a bistable potential is a quartic function. But may such a handy polynomial be an adequate approximation for two overlapping laser traps? The intensity of a laser beam across its diameter varies according to a Gaussian distribution. In fig. 2.3, two Gaussian distribution functions with variance $\sigma^2 = 1/2$ are depicted, $g_\pm(x) = -1/\sqrt{\pi} \exp[-(x \mp 1)^2]$, and their sum, $g_+(x) + g_-(x)$, too. The Gaussian $g_+$ is an even function with respect to $x - 1$, and $g_-$ is an even function with respect to $x + 1$. So the minima of both functions are quadratic in $x \mp 1$ plus terms of order $(x \mp 1)^4$ and higher. The same is true for the relative maximum of $g_+(x) + g_-(x)$ at $x = 0$.

Now the quartic function $f(x) = 1/\sqrt{\pi} \left[ (1 - 2/e) (x^2 - 1)^2 - 1 \right]$ is an exquisite approximation in the broader sense of an interpolation between the single Gaussian functions $g_+$ and $g_-$ at their minima and their sum, $g_+ + g_-$, at its relative maximum. It can be adjusted to the position of the extremal points as well as to their second derivatives, as the enlargement on the right hand side of fig. 2.3 demonstrates. The divergence of $f$ for larger values of $|x|$ is irrelevant because the energy necessary for the particle to “see” those high values exceeds the thermal energy (which is of the same order as the barrier between the wells) by far.
2. The Models

So, from now on, we will no longer concern ourselves with Gaussians but simply adopt a quartic potential of the form

$$ U(x_i) = \Delta U \left[ \left( \frac{x_i}{x_0} \right)^2 - 1 \right]^2 $$

for particle \( i \), where \( \Delta U \) means the height of the potential barrier between both wells, and \( x_0 \) is the (symmetric) distance of each well bottom from the barrier top. This is the standard form of a double well potential with minima at \( \pm x_0 \). An example with \( x_0 \) and \( \Delta U \) set equal to 1 is depicted in fig. 2.4. Sketched as a three-dimensional potential landscape, \( V(x_1, x_2, t) \) looks familiar, comparable to an egg carton with compartments for four eggs. But one must not confuse the \( (x_1, x_2) \)-plane with the \( (x, y) \)-plane of fig. 2.2! Two single particle variables, each performing a motion parallel to the \( x \)-axis, are transformed into a combined system variable moving within two perpendicular directions.

The next ingredient to our model is the driving force that induces asymmetry into the potential. It shall be a spatially constant force which is modulated harmonically:

$$ f_i(t) = A_i \cos (\Omega t + \varphi_i). $$

The force \( f_i \) acts on particle \( i \) with strength (amplitude) \( A_i \) and oscillates with angular frequency \( \Omega \) plus an additional phase \( \varphi_i \).

The third term to be brought into account must combine both particles, for otherwise there would not be any coupling and therefore no coupled system. This is done by the surface charges residing on the micro beads which evoke a repulsive force between the particles according to a screened Coulomb potential,

$$ \Phi(|x_1 - x_2|) = \phi \frac{\exp(-\kappa |r_{12}|)}{r_{12}}. $$

Figure 2.4.: Potential \( U(x_1, x_2) \) with \( x_0 = \Delta U = 1 \).
2.2. The potential landscape

The distance \( r_{12} \) is matched in dimension by the Bjerrum length \( \lambda_B \) that is part of the coupling constant \( \phi \) and denotes the distance between two elementary charges at which the Coulomb interaction within pure water equals thermal energy. The screening depends on the screening constant \( \kappa \) which is the inverse of the Debye length \( 1/\kappa \). The exact form of \( \phi \),

\[
\phi = \left( \frac{Z^* \exp(\kappa R)}{1 + \kappa R} \right)^2 \lambda_B k_B T, \tag{2.4}
\]

is derived from the Derjaguin-Landau-Verwey-Overbeek linearized mean-field model for colloidal electrostatic interactions [6]. \( Z^* \) is an effective charge number assigned to the average micro bead, and \( R \) denotes the particle radius. The distance function,

\[
r_{12} = \sqrt{(x_1 - x_2)^2 + d^2}, \tag{2.5}
\]

provides the distance between both particles wherein the distance (in y-direction) between two double wells, \( d \), enters together with the relative distance (in x-direction) between both particles, \( |x_1 - x_2| \).

![Diagram](image)

**Figure 2.5.** Potential \( \Phi (|x_1 - x_2|) \) with \( \phi = \kappa = d = 1 \).

The effect of the screened Coulomb potential can be described with fig. 2.5. Its repulsive effect is strongest parallel to the \( x_1 = x_2 \) diagonal where both beads are closest to one another. The negative gradient will either direct into the \( (1, -1) \)-direction or into the \( (-1, 1) \)-direction, i.e. into the direction of largest separation between the particles.

All pieces put together, the total time dependent scalar potential for our two-dimensional configuration space reads:

\[
V(x_1, x_2, t) = \sum_i \left( U(x_i) - x_i f_i(t) \right) + \Phi (|x_1 - x_2|). \tag{2.6}
\]
2. The Models

The situation is sketched in fig. 2.6. As opposed to fig. 2.4, the “egg carton” is somewhat distorted due to the influence of the repulsive Φ-potential. This means that the potential wells of the first and the third quadrant are less deep than the wells of the second and the fourth quadrant. In other words, the potential barriers as seen from the wells of quadrants 2 and 4 stay nearly the same, whereas the barriers as seen from the wells of quadrants 1 and 3 are reduced in height. This is done by the symmetry breaking nature of the screened Coulomb potential. The term “barrier height” needs a brief explanation as to what it means within a potential landscape. It refers to the potential difference between a minimum (well) and a saddle point (barrier) that determines the energetically most favourable connection between neighbouring wells.

2.3. Langevin dynamics

A system consisting of two suspended colloidal particles and some millilitres of water including some ions possesses about $10^{23}$ degrees of freedom. But we are merely interested in two of them. These are the positions of the colloidal particles with respect to the double well axis (the straight line through both potential minima, parallel to the $x$-axis). Fortunately the many variables of the water molecules are comparatively fast variables that on the average mostly add up to zero. Therefore the dynamics for each of our particles can be modelled as Brownian motion, i.e. as a fluctuating stochastic force $\xi(t)$ and a dissipative braking force. The latter is implemented via a dampening constant $\gamma$ leading to the braking force $-m\gamma v$. Combining mass and dampening into an effective friction coefficient, $\eta = m\gamma$, 

![Figure 2.6: Potential $V(x_1, x_2, t)$ with $x_0 = \Delta U = \phi = \kappa = d = 1$ and $A_1 = A_2 = 0$.](image)
which is an inverse mobility, Newton’s equation of motion reads

\[ m\ddot{x}(t) = -\eta\dot{x}(t) + \xi(t) \]  

(2.7)

and is termed Langevin equation.

If we apply an external force \( F(x, t) \) acting on the particle that shall not influence the interaction with the surrounding solvent so that \( \eta \) and \( \xi \) remain unchanged, we simply add it to our Langevin equation:

\[ m\ddot{x}(t) = -\eta\dot{x}(t) + F(x, t) + \xi(t). \]  

(2.8)

This expression simplifies if we assume high dampening (large \( \eta \)). Provided that \( \eta \dot{x} \gg m\ddot{x} \), we may safely neglect inertial forces and thereby infer

\[ \eta\dot{x}(t) = F(x, t) + \xi(t). \]  

(2.9)

For our system, the assumption of the overdamped limit is completely justified because the permanently exchanged amounts of energy and momentum due to the enormous number of molecular collisions are huge and fast relative to the momentum and inertia of the Brownian particle [7].

Now we take into account that we observe two particles, each with its own position variable \( x_i, i \in \{1, 2\} \). We replace the force by the gradient field of the complete potential \( V \) as derived in eqn. (2.6), so we finally obtain two coupled one-dimensional stochastic differential equations:

\[ \eta\dot{x}_i(t) = -\frac{\partial}{\partial x_i} V(x_1, x_2, t) + \xi_i(t). \]  

(2.10)

Concerning the stochastic forces \( \xi_i(t) \) we take the standard approach and realise them as Gaussian white noise with zero mean,

\[ \langle \xi_i(t) \rangle = 0, \]  

(2.11)

and delta correlation,

\[ \langle \xi_i(t) \xi_j(s) \rangle = 2\eta k_B T \delta_{ij} \delta(t - s). \]  

(2.12)

The latter is related to the diffusion constant via \( D = k_B T / \eta \).

The friction coefficient might be derived from the Stokes formula, \( \eta = 6\pi \nu r \), with particle radius \( r \) and viscosity \( \nu \). But experiment has shown that this approximation does not hold because of the strong influence of the containing walls onto the micro beads [4]. Therefore we apply a value for \( \eta \) which was found by adapting a numerical simulation to the experimental findings.

For numerical simulation it is necessary to transform eqn. (2.10) to discrete time steps. Apparently this is an easy index manipulation:

\[ x_{n+1}^i = x_n^i - \frac{\Delta t}{\eta} \frac{\partial}{\partial x_i} V_n(x_1^n, x_2^n) + \xi_n^i. \]  

(2.13)
2. The Models

Here the particle number (1 or 2) is denoted by the upper index, \( i \), the discrete time steps of width \( \Delta t \) are counted by the lower index, \( n \). The connection between continuous and discrete variables is established via \( t_n := n \Delta t \) and \( x_n^i := x_i(t_n) \), respectively.

Only the transformation of the stochastic force is marginally more delicate: we must keep in mind that the noise intensity must be invariant with respect to the chosen time step length \( \Delta t \). Since \( \xi \) is a kind of time integral of \( \xi \) over a domain of length \( \Delta t \) (Wiener process), the correlation must be proportional to the time step length \([8]\). Finally, mean and correlation of \( \xi \) read:

\[
\langle \xi_n \rangle = 0 \tag{2.14}
\]

and

\[
\langle \xi_n \xi_m \rangle = 2 \frac{k_B T}{\eta} \delta_{ij} \delta_{nm} \Delta t. \tag{2.15}
\]

Now, eqn. (2.13) together with properties (2.14) and (2.15) permits numerical simulation of the particle dynamics.

2.4. Kramers rates

In a seminal paper from 1940, Hendrik Anthony Kramers found a general solution of the problem of escape from a metastable state that is of high interest to many fields of science like physical chemistry or statistical physics. Here is only enough space for a brief reminder of his findings as far as we make use of them. For a comprehensive review of Kramers’ results and their influences until today, cf. [9], for their application in statistical physics and chemistry, cf. [10].

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{kramers_escape.png}
\caption{Kramers’ escape problem; a metastable state \((x_a)\) is separated from a free region \((x_c)\) by a potential barrier \((x_b)\) of height \(\Delta U\).}
\end{figure}

The title of Kramers’ paper, “Brownian Motion in a Field of Force and the Diffusion Model of Chemical Reactions”, reveals much of his understanding an escape process as governed by Brownian motion driven by thermal forces. A
2.4. Kramers rates

The metastable state corresponding to the reaction variable \( x = x_a \) as depicted in fig. 2.7 decays (in the case of overdamped dynamics, i.e. in the large \( \eta \) limit) according to the Kramers rate

\[
k = \frac{\omega_a \omega_b}{2\pi \gamma} \exp \left( -\beta \Delta U \right).
\]

The angular frequencies, \( \omega_a \) and \( \omega_b \), are derived from the curvature of the underlying potential at the metastable state, \( U(x_a) \), and at the unstable state, \( U(x_b) \), via \( \omega_a = \sqrt{U''(x_a)/m} \) and \( \omega_b = \sqrt{|U''(x_a)|/m} \), respectively. By inserting these relations, eqn. (2.16) yields

\[
k = \frac{\sqrt{U''(x_a)} |U''(x_b)|}{2\pi \gamma} \exp \left( -\beta \Delta U \right).
\]

The most astounding quality of this result is its independence of any detail of the potential except for the curvature in two points plus the barrier height.

Kramers’ result can be generalised to many dimensions wherein the dynamics are governed by an \( N \)-dimensional reaction coordinate in a potential landscape. To this end we adopt the results derived in [9] and readjust them to our own setup which is a two-dimensional one. Instead of the curvature of the potential we have to consider second derivatives in tensorial form.

Ordered in matrix form with elements \( H_{ij} = \partial^2 V / (\partial x_i \partial x_j) \), this matrix \( H \) is known as the Hessian of \( V \) which in our case is a \((2 \times 2)\)-matrix. Its eigenvalues correspond to the angular frequencies of the one-dimensional model according to \( \Pi_i \omega_i = \sqrt{\det (H)/m} \), the Hessian being calculated at the position of either a minimum or a saddle point. The only negative eigenvalue of \( H \) at a saddle point which represents the growth rate of small deviations into the unstable direction, plays a significant role, and its positive value is denoted by \( \omega_b \), in analogy to its role in eqn. (2.16).

The general form of the Kramers rate for a multi-dimensional potential landscape and overdamped dynamics reads as follows:

\[
k = \frac{\omega_b}{2\pi \gamma} \frac{\Pi_j \omega_j^a}{\Pi_j \omega_j^b} \exp \left( -\beta \Delta V \right).
\]

Therein \( \omega_i^a \) are the angular frequencies of the stable modes at the saddle point, \( \omega_i^b \) are those of the metastable point, and the notation \( \Pi' \) indicates the exclusion of \( \omega_b \) from the product.

Eqn. (2.18) enables us to calculate the Kramers rates for transitions of the system variable from one potential well of \( V \) to an other. As there are four wells, each connected with two neighbouring wells via saddle points, there are up to eight different rates \( k_{ij} \). The indices are attached to each rate according to fig. 2.8: \( k_{ij} \) means the rate for transitions starting in well \( j \) and ending in well \( i \).

Let \( p_i(t) \) be a probability distribution for the system to be found in state \( i \) which is equivalent to its system variable to be located in the \( i \)-th quadrant. Then
we know already: \( \sum_i p_i(t) = 1 \) for all times and thereby \( \sum_i \dot{p}_i(t) = 0 \). Any future distribution can easily be computed if the Kramers rates are known, we simply must balance the probability flows into and out of state \( i \). The inward flow depends on the decay rates of neighbouring states times the present value of their respective probabilities: \( \sum_j k_{ij}(t) p_j(t) \) with \( j \) being a neighbour of \( i \). The outward flow follows from exchange of indices and sign: \( \sum_j -k_{ji}(t) p_i(t) \) with \( j \) being a neighbour of \( i \). If we require \( k_{ii} = 0 \) and \( k_{ij} = 0 \) whenever \( i \) and \( j \) are not neighbours, then we obtain the master equation

\[
\dot{p}_i(t) = \sum_j \left( k_{ij}(t) p_j(t) - k_{ji}(t) p_i(t) \right).
\] (2.19)

But if we require \( k_{ij} = 0 \) whenever \( i \) and \( j \) are not neighbours and set \( k_{ii} = -\sum_{j \neq i} k_{ji} \) and let \( K \) be a matrix with \( K_{ij} = k_{ij} \), \( p \) a vector with components \( p_i \), then we obtain the master equation in plain tensorial form:

\[
\dot{p}(t) = K(t)p(t).
\] (2.20)

In this form, a numerical integration can be implemented on a PC without any difficulty, particularly with regard to the periodic time dependance of \( K \).

### 2.5. Quantifiers and observed quantities

The two preceding sections presented two different models that may be evaluated either by computer simulation or by numerical integration. The former applies to Langevin dynamics and results in a system trajectory \( x(t) = (x_1, x_2)(t) \). The latter applies to an approach via Kramers rates and results in a time dependent
2.5. Quantiﬁers and observed quantities

probability distribution, \( p(t) = (p_1, p_2, p_3, p_4)(t) \), associated with four discrete system states. A connection between both approaches may be established if we make use of two properties:

1. the equivalence of long time average and ensemble average (ergodicity), and
2. the inherent periodicity which is due to the harmonic driving.

Point number 1 means that there must be a correspondence between time averages based upon \( x(t) \) and the probability distribution \( p(t) \) which actually represents an ensemble average for the same system. Point number 2 gives a hint as to what kind of averages should be taken into consideration, namely those mirroring the inherent periodicity.

At fi rst, the nature of solutions of eqn. (2.20) needs to be elucidated a little. In the absence of a modulated driving (i.e. \( A_i = 0 \) or \( \Omega = 0 \)) \( K \) is constant and \( p(t) \) converges exponentially to an equilibrium distribution with only minor discrepancies from a Boltzmann distribution:

\[
\lim_{t \to \infty} p_i(t) = \frac{\alpha_i \exp(-\beta V_i)}{\sum_j \alpha_j \exp(-\beta V_j)}.
\]

where the corrections \( \alpha_j \) take the differences in the eigenmodes into account, and the \( V_j \) denote the potential energy of state \( j \). For very slow driving, \( p(t) \) follows close to the present equilibrium distribution, and for faster driving its evolution will become more and more retarded and quasi-damped until the modulation will become so fast that the system effectively feels little more than the average force—which, of course, is zero. It is clear that in the intermediate regime, where we hope to fi nd a substantial response to the modulating force, the evolution of \( p(t) \) will converge onto a limit cycle that must obey the same periodicity as the driving.

Because this last result must also hold for \( x(t) \) in a suitable time average, we investigated a periodical average of the kind

\[
\langle x_i(t) \rangle := \frac{1}{N} \sum_{n=1}^{N} x_i(t + n\tau), \quad t_0 \leq t < t_0 + \tau,
\]

where \( \tau = 2\pi/\Omega \) is the period of the driving force. This average permits the evaluation of our fi rst quantiﬁer for stochastic resonance, the hysteresis loop area, which is deﬁned as the integral over one full period of the driving force:

\[
HLA_i := \int_{\tau} \langle x_i(t) \rangle \, d(f_i(t)).
\]

The probability distribution can now be linked to the average \( \langle x_i(t) \rangle \) by use of the present \( (x_1, x_2) \)-pair of coordinates of each state \( j \).

Let \( x_{ij}(t) \) denote the \( x_i \)-coordinate of the well bottom of state \( j \) at time \( t \). Then we can calculate an expectation value of the \( x_i \)-coordinate of the system by using
\[ \langle x_i(t) \rangle = \sum_j x_{ij}(t) p_j(t) \] Written as a \((2 \times 4)\)-matrix \(X\) with \(X_{ij} = x_{ij}\), the preceding relation leads to the transformation equation
\[ \langle x(t) \rangle = X(t) p(t) \] (2.24)
that allows to compare the results of the Langevin and the Kramers approach. This equation is but an approximation that is correct for quasi-discrete system states. In other words, if there is a non-vanishing probability for the system to stay at places far off the potential wells, and if this probability is furthermore “smeared out” asymmetrically, then there will be deviations from eqn. (2.24).
Assuming the system behaves properly and stays at the well bottoms for most of the time, we infer from the last two equations the hysteresis loop area of the probability distribution \(p(t)\):
\[ HLA_i = \int \ldots \int \left( e_i^T X(t) p(t) \right) p_i(t) \, dt. \] (2.25)
The transposed unity vector \(e_i^T\) is either the row vector \((1, 0)\) for \(i = 1\) or \((0, 1)\) for \(i = 2\). Alternatively, \(HLA\) could be defined as a vector function.

The second quantifier we consider here, the residence time distribution, does only apply to the actual system trajectory acquired by simulation of the Langevin equation. The reason is readily understood because residence times can only be measured in a specific system by observing \(x(t)\) but not in an ensemble average.

By virtue of the quantity \(-1/\tilde{p}_i(t)\) which is the average lifetime of the population of state \(i\) at time \(t\), provided that \(\tilde{p}_i(t)\) is negative, one cannot infer how long an individual system might or might not reside within this state. Even if, in the ensemble average, the state decays rather fast at a specific instance, an individual system might even enter it and reside for a long time. It is not even possible to calculate the probability of such an event by virtue of decay rates all on their own.

Nevertheless, as a bona fide quantifier, the residence time distribution is helpful even if we can only apply it to our simulation data. This is done via the area under the first peak at \(t = \tau/2\) of the residence time distribution \(P^i_{res}(t)\):
\[ RES_i := \int_{(1/2-\alpha)\tau}^{(1/2+\alpha)\tau} P^i_{res}(t) \, dt. \] (2.26)
The number \(0 < \alpha \leq 1/2\) can be chosen at will from this interval and determines the sensitivity of the integration. Throughout this thesis, the figure
\[ \alpha := 0.2 \] (2.27)
will be kept constant.

Now that the “measures” are defined, a physical explanation of their meaning and relevance seems appropriate. The two other quantifiers, signal-to-noise ratio and spectral power amplification, have in common that they are calculated by sophisticated mathematical tools involving autocorrelation functions, Fourier
transforms, the Wiener-Khinchin-Theorem, etc. To someone who is not really endowed with higher mathematical education I would explain this with a person that sits in front of a hi-fi-system and gazes at the up-and-down bouncing equaliser LED-bars—but does not hear the music at all! In a similar way, those quantifiers are a mighty tool for frequency and power analysis, but on an abstract metalevel. In this thesis, a more direct approach to the things we really can see and manipulate is preferred, and be it for the sake of simplicity.

What does eqn. (2.25) actually mean? Via differentiation of the r.h.s. we get

\[ \frac{d}{dt} f_i(t) = A_i \cos(\Omega t + \varphi_i) = -A_i \Omega \sin(\Omega t + \varphi_i) \, dt, \]

which is, of course, the temporal derivative of the driving force acting on particle \( i \).

Together with the average particle coordinate, \( \langle x_i(t) \rangle \), it denotes the force acting on the particle along its way that is parametrised by time \( t \). This average way is, in a system like this with periodically modulated driving force, a closed loop. If some kind of resistance is added, a hysteresis loop evolves, similar to those we know from ferro magnets. To be precise, there will be no remanence as long as we do not shut down the thermal noise. Nevertheless, as dimensions coincide, the area enclosed by such a loop denotes the work that is gained from the external driving, provided that the other relevant forces are all conservative. The notion of stochastic resonance may then be linked to the maximal attainable amount of work/energy that appears as a maximum in the HLA-curve.

Understanding the residence time distribution is even more facile: one merely needs to stop the time until a particle successfully leaves its well. Afterwards, all times close to a half-period are counted and divided by the number of all jumps that occurred during observation. If all jumps happen to occur after a timespan around \( \tau/2 \), one gets a 1, if nothing happens, there will be a 0, everything else lies in between. This is plausible even for the layman. Why should this help? If for nothing else, then for communicating physical ideas to the public in a way that does not put the public to rout. And, as we will see later in this thesis, valuable results may even be achieved by this innocuous and unsophisticated method.

### 2.6. Units and system parameters

In a colloidal system, most parameters have very small values due to the minuscule dimension of the experimental setup, and we would rather not bother with all those annoying negativ exponents in our results and plots. So, for convenience, all quantities are adjusted to “natural” units that are derived from system parameters. To this aim, the unit of length will be \( [l] = 1 \mu m \), and masses will be scaled to \( [m] = 1 \text{mg} \). Time, \( [t] = 1 \text{s} \), and temperature, \( [T] = 1 \text{K} \), stay unchanged. Important composed units are then: force, \( [F] = 1 \frac{\text{mg} \cdot \mu \text{m}}{\text{s}^2} = 10^{-15} \text{N} = 1 \text{pN} \), and energy, \( [E] = 1 \frac{\text{mg} \cdot \mu \text{m}^2}{\text{s}^2} = 10^{-18} \text{J} = 1 \text{aJ} \).
2. The Models

Most system parameters stay constant throughout all simulations. We have a constant temperature and thus constant thermal energy:

\[ T = 292 \text{K}, \quad (2.29) \]
\[ k_B T = 4.033 \text{aJ}. \quad (2.30) \]

The double well potential is specified by the well to well distance, \( 2x_0 \), and by the barrier height, \( \Delta U \):

\[ x_0 = 0.8 \mu m, \quad (2.31) \]
\[ \Delta U = 2.9 k_B T. \quad (2.32) \]

The driving amplitude was also kept constant and equal for all particles:

\[ A_i x_0 = 1.9 k_B T, \quad (2.33) \]
\[ \varphi_1 = 0. \quad (2.34) \]

The parameter values concerning the screened Coulomb potential are comparatively insecure as they are not directly measurable. In experiment, the ion concentration within the aqueous solution did not stay constant so that the screening “constant” and thus the coupling strength varied in time. Nevertheless, for any calculation, definite values must be chosen, beginning with the average charge per micro bead:

\[ Z^* = (10000 \pm 5000) \]
\[ \text{chosen as } \rightarrow Z^* := 10000, \quad (2.35) \]

the Debye length:

\[ 1/\kappa = (0.25 \ldots 0.40) \mu m \]
\[ \text{chosen as } \rightarrow 1/\kappa := 0.33 \mu m, \quad (2.36) \]

the particle radius:

\[ 2R = (1.57 \pm 0.06) \mu m \]
\[ \text{chosen as } \rightarrow R := 0.75 \mu m. \quad (2.37) \]

The Bjerrum length is defined by natural constants according to

\[ \lambda_B := \frac{e^2}{\epsilon k_B T} = 0.72 \cdot 10^{-3} \mu m, \quad (2.38) \]

where \( e \) is the elementary charge and \( \epsilon \approx 80 \) is the dielectric constant of pure water. According to eqn.(2.5), the coupling constant \( \phi \) may take values in between
\[ \beta \phi_{\text{min}} = 9.3 \cdot 10^4 \mu m \quad \text{and} \quad \beta \phi_{\text{max}} = 4.1 \cdot 10^6 \mu m. \]

Indeed, the uncertainties of \( Z^* \) and \( \kappa \) in each case produce one order of magnitude of variation in \( \phi \). The positive aspect of this is: we can simply pick up a convenient value without the usual problem of finding the exact match to experimental findings. For the two-particle simulations and the Kramers rates, a value of

\[ \beta \phi := 6.1 \cdot 10^5 \mu m \tag{2.39} \]

was calculated from the arbitrarily chosen values of \( R, Z^*, \lambda_B, \) and \( \kappa \).

The effective coupling strength can overall be varied by three parameters, but for simplicity only one parameter may be varied and the other two are kept constant. On the one hand, the parameter \( d \), the distance between two double wells, is difficult to control in experiment. The reasons are inhomogenities of the surface charge on the walls of the sample cell. On the other hand, during experiment, the Debye length is decreased by the permanently increasing ion concentration within the suspension, as stated before. This effect also is nearly uncontrollable, but it has the advantage to be homogeneous in space and steady in time. Therefore a variation of the coupling strength is experimentally realised by the automatic change in the screening constant, \( \kappa \).

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig2.9}
\caption{Contour lines of Coulomb force; \( Z^* \) is fixed at 10 000.}
\end{figure}

In this thesis, the coupling strength is varied dependent on the variable \( d \) because the theoretical modeling started from a more geometrical point of view involving various distances between the potential wells. The decisive figure is, of course, the negative gradient of the screened Coulomb potential, the resulting Coulomb force acting between the particles:

\[ -\frac{\partial}{\partial x_i} \Phi (|\Delta x|) = (-1)^{i+1} \Delta x (\kappa r + 1) \phi \frac{\exp (-\kappa r)}{r^3}, \tag{2.40} \]
where $\Delta x = x_1 - x_2$ and $r = r_{12}$. In fig. 2.9 we can see that within the relevant ranges there is nearly a linear dependency between all those values of $d$ and $1/\kappa$ yielding the same resulting force. The diagram shows the situation for particle 1 where the particle separation $\Delta x$ is equal to $x_0$, e.g. particle 1 at a saddle point ($x_1 = 0$) and the other particle at the well bottom (e.g. $x_2 = -x_0$). The positive sign means that the force acts on particle 1 into the positive $x$-direction. For some values of the resulting force, contour lines in the $(d, \kappa)$-plane are shown. The value of $Z^*$ is kept constant at 10000.

The diagram can easily be used for different values of $Z^*$ because the force is proportional to $(Z^*)^2$. For example, if $Z^*$ is equal to $Z^*_{\text{min}} = 5000$ instead of the original 10000, every force isoline in fig. 2.9 is shifted to the position of the next lower force isoline. As a consequence, each choice of setting the coupling strength, either by $d$ or by $\kappa$, is equally justified since the same resulting force and thus the same dynamical effect may be attained by every parameter pair belonging to the same force isoline.

The last parameter to be specified is the friction coefficient $\eta$. By experiment, a diffusion constant of $D \approx 0.24 \text{µm}^2/\text{s}$ was found, thus yielding

$$\eta := \frac{k_B T}{D} = 16.8 \frac{\text{mg}}{\text{s}}.$$  \hspace{1cm} (2.41)

Indeed, this value is somewhat larger than the estimate after the Stokes formula, as shown in [4].
3. Developing the Programs

3.1. Langevin simulation for two coupled particles

The development of the first simulation program took place gradually. As a matter of fact it was a start from scratch—and it made up a fine occasion to begin as thorough as possible. Based on eqn. (2.13), the consecutive stations were:

1. a single particle in a double well with noise and harmonic driving,
2. two particles in two separate double wells, still without coupling,
3. two particles in two separate double wells with coupling.

The first station was ideal to test the implementation of different random number generators as far as velocity and reliability were concerned. Another advantage was to compare first single particle simulation results to prior results in [4] which were received by numerical integration of a Fokker-Planck-equation. The second station was mainly a transformation from one-dimensional variables to two-dimensional arrays. This led to the implementation of the NAG random number generator G05FDF [11] that is capable of producing vectors of normally distributed random numbers. So by now there was a pair of Gaussian random numbers at hand for each simulation step. The third and final station introduced the core of the whole model: the coupling generated by the screened Coulomb potential.

Parallel to these model upgrades the aspect of data gathering and data analysis was advanced, too:

- Each simulation step was recorded without any further analysis. This was only practical for short time simulations and merely of demonstrative use.
- Periodical averages of the trajectories in compliance with eqn. (2.22) were calculated on-line whereby long time simulations became feasible. Since the value of $\tau/\Delta t$ could become quite large, i.e. each period of the driving force lasted many simulation steps, the resulting graph of $\langle x_i(t) \rangle$ was rather rough and its resolution exceedingly high. Therefore a number of 500 time intervals was fixed and all simulation steps belonging into a span of $t' \leq t_n < t'+\tau/500$ were summed up to one value of of $\langle x_i(t') \rangle$.
- Numerical integration of $HLA$ according to eqn. (2.23) was implemented. For this purpose the 500 values of $\langle x_i(t) \rangle$ were pairwise linked to trapezes without using any sophisticated interpolation schemes.
Jumps of each subsystem from one well to the other were counted and the waiting time between each jump was recorded on-line yielding the residence time distribution $P_{\text{res}}(t)$. In order to make use of $P_{\text{res}}$ in form of a histogram, the recording of waiting times was done in classes of frequency corresponding to integer multiples of $\tau/20$. From this histogram, the determination of $\text{RES}$ via eqn. (2.26) was simply the sum over all classes beginning with the class of $t = \tau \left[ 10 - 20\alpha \right] /20$ up to $t = \tau \left[ 10 + 20\alpha \right] /20$ with $0 < \alpha \leq 1/2$.

Data records were written to plain ASCII-files for later generation of diagrams.

### 3.2. Kramers rates for two coupled particles

The main effort of this part was not the numerical integration of eqn. (2.20) but the computation of the single Kramers rates for different states of the potential landscape according to eqn. (2.18). Because harmonic driving and coupling make each local extremum of the potential landscape to change position, curvatures and height, the first step must be to compute the Kramers rates comprised in $K(t)$. This is done for all time steps of one period $\tau$ of the driving, for they are needed for numerical integration of $\dot{p}(t)$.

In order to determine the location of extremal points of the potential landscape at discrete time steps an approximation scheme is needed. Therefore a quasi-Newton method involving the inverse of the Hessian to render a good approximation of the direction to the next evaluation point is utilised. Details can be found in [12]. As there were merely four minima and four saddle points to be determined, no special effort was made with respect to computation efficiency. Furthermore, after the first run, every change of the potential landscape due to the driving was both smooth and small, so every following determination of extremal points based on the previous run and thus became fast.

A severe problem to be considered is the choice of the starting point for the search of extremal points. The more distorted the potential landscape gets as a consequence of large coupling or strong driving, the less appropriate are the preset values $(x_0, x_0), (x_0, -x_0), \ldots$ for the minima and $(0, x_0), (x_0, 0), \ldots$ for the saddle points. In particular the local maximum near the origin of the coordinate system grows in influence and distracts the quasi-Newton algorithm. This could be dealt with by starting with small coupling and weak driving and gradually increasing the values while updating the starting points.

When the necessary extremal positions are found, $K_n$ can be calculated without any difficulty. In discrete time, eqn. (2.20) reads

$$p_{n+1} = (\Delta t \ K_n + \text{id}) \ p_n$$

and is integrated straightforward in accordance with a plain Euler-algorithm. The periodicity of the driving force leads alternately to convex and concave segments of the solution whereby the shortcoming of the Euler-algorithm is reduced. The
evolution of the probability distribution $p$ is carried forward until the difference between succeeding periods, $|p_i(t) - p_i(t - \tau)|$, falls below a threshold value for all $i = 1, \ldots, 4$. Then we assume a stable periodic solution reached.

Further on, the resulting data is treated as in the Langevin simulation except for the residence time distribution, which is not obtainable from Kramers rates.

### 3.3. Langevin simulation for four coupled particles

The four particle system is an extension of the Langevin simulation described in section 3.1. Since a realistic surface charge distribution on the containing walls of the sample cell is far from homogeneous, a configuration built of four pairs of optical traps equivalent to four double wells was preferred in experiment. In particular with regard to experimental verification of any theoretical findings, it did not seem futile to convey the results from a two particle system to a four particle system.

![Figure 3.1](image)

**Figure 3.1.**: Arrangement of four double wells in a square. Each of the short arrows points into the direction of the positiv $x_1$ coordinate.

Besides an increase in dimension from 2 to 4, the new system geometry as sketched in fig. 3.1 required a new definition of the coupling potential. As every particle kept its own one-dimensional dynamic, introducing a two-dimensional coordinate system solely for the purpose of distance calculation via vector differences would have meant superfluous effort. Four particles make up six pairs and thereby 12 partial forces to be added because each pair of particles generates an absolute force that is multiplied by the cosines of two directions in which it affects each partner. This is done by the coordinates $x_1$ to $x_4$ that are deliberately chosen to conserve a rotational symmetry. In form of summed pair potentials, the screened
Coulomb potential eqn. (2.3) then reads as follows:

\[ \Phi(x) = \frac{1}{2} \sum_{i,j} \Phi_{ij} = \frac{1}{2} \sum_{i,j} \phi \lambda_B \frac{\exp(-kr_{ij})}{r_{ij}}, \quad (3.2) \]

but the distances are defined different from eqn. (2.5):

\[ r_{ij} = \sqrt{(d/2 - x_i)^2 + (d/2 + x_j)^2} \quad \text{for } j = i + 1 \mod 4, \quad (3.3) \]

\[ r_{ij} = \sqrt{(x_i + x_j)^2 + d^2} \quad \text{for } j = i + 2 \mod 4, \quad (3.4) \]

\[ r_{ij} = 0 \quad \text{for } i = j, \quad (3.5) \]

\[ r_{ij} = r_{ji} \quad \text{for all } i, j. \quad (3.6) \]

The cosines mentioned above result naturally from differentiating eqn. (3.2) with respect to each coordinate, \(x_1, \ldots, x_4\).

Another consequence of the increased number of particles is the number of relative phases between the driving forces. For two particles this parameter is a single quantity and is specified by \(\varphi_1 - \varphi_2\). Four driving forces, of course, can be distinguished by three relative phases so that a wholistic investigation of the phase dependance of stochastic resonance would require the sampling of a three-dimensional parameter manifold. Because the arrangement is highly symmetric, this would include large redundances, so that it is economically advisable to look for significant configurations of phase shifts in advance.

**Figure 3.2.**: Phase shift symbols. A: particle is driven to the right, force at maximum; B: both wells are balanced, zero-crossing of force; C: particle is driven to the left.

I selected four different setups which vary in the prevalence of either in-phase or out-of-phase junctions between neighbouring particles. For better understanding the four configurations shall be described by suggestive sketches that are in accordance with the legend presented in fig. 3.2. Each configuration is represented by sketches at four equidistant times corresponding to (from left to right) \(\Omega t = 0, \pi/2, \pi, \text{ and } 3\pi/2\). Each sketch shows the instantaneous bias of the driving for every particle in accordance with the arrangement depicted in fig. 3.1.

- **Configuration #1** presents a maximum of synchronisation: all drives are in phase, i.e. \(\varphi_i = 0\), for \(i = 1, \ldots, 4\). This configurations is invariant under rotations of half-integer multiples of \(\pi\). All particles are equivalent.

- **Configuration #2**, all pairs of neighbours are in relative antiphase: \(\varphi_1 = \varphi_2 + \pi = \varphi_3 = \varphi_4 + \pi\). There is invariance under rotations of integer multiples of \(\pi\), so particles 1 and 3 are equivalent as well as particles 2
3.3. Langevin simulation for four coupled particles

Figure 3.3.: Phase shift #1: all drives synchronised.

Figure 3.4.: Phase shift #2: drives are pairwise in antiphase.

Figure 3.5.: Phase shift #3: a configuration initially realised in Stuttgart.

Figure 3.6.: Phase shift #4: a variation of #3.
and 4. An additional symmetry is implied by pairity: The whole setup is invariant under mirroring with respect to both the diagonal axes through the centre. Therefore particles 1 and 3 are equivalent to particles 2 and 4 by pairity.

- A cyclic increasing phase shift of $\pi/2$ ($\varphi_{i+1} = \varphi_i + \pi/2$ for $i = 1, 2, 3$) leads to configuration #3 that was initially realised at Universität Stuttgart by Tobias Sawetzky, Carmen Schmitt, and Clemens Bechinger [5]. Here, the driving of the two nearest neighbours of each particle is synchronised, not in terms of the phase difference (which equals $\pi$) but as “seen” by the particle in between. By means of symmetry transformations, now also involving time shifts, one finds again that all particles are equivalent.

- The last configuration comprises a small variation of configuration #4: opposing particles have phase difference zero instead of $\pi$. Even for this setup the equivalence of all particles holds. For all configurations, the notion of equivalence means that, in case of deterministic motion, the resulting dynamics would be identical if properly transformed by means of symmetry operations like time shift or change of pairity ($x_i \rightarrow -x_i$).

A final remark on further properties of the four-particle simulation: the random numbers are produced by the same random number generator as in the two particle simulation (G05FDF, cf. [11]), and data analysis is performed on-line, as described in section 3.1.
4. Testing the Programs

4.1. Reproducibility of known results

The first objective during the final phase of programming was to reach comparability to previous results from an identical one-particle system without coupling [4]. To switch off the coupling is easily done by setting $\phi = 0$ or (better) by preventing the program from calling the respective subroutine—in the Langevin simulation the most time consuming part. All parameter values were set as listed in the preceding section. The choice of the time step $\Delta t$ was not yet optimised since this will be done after the programs demonstrated their reliability. For the time being, everything worked fine with

$$\Delta t = 0.01 \text{s.}$$

The figures 4.1 to 4.5 present the results of both, Langevin simulation and Kramers rates, for harmonic driving periods of $\tau = 10\text{s}, 20\text{s}, 50\text{s}, 100\text{s},$ and $200\text{s}$. The accord with [4] is excellent! Of course, it is no surprise, for Langevin equation and Fokker-Planck equation are closely related and both equally well applicable for Brownian dynamics. But nevertheless, the good agreement of Langevin and Kramers approach could not be expected 

Moreover, the series of figures exemplifies the typical behaviour of the average system coordinate $\langle x(t) \rangle$ with respect to different driving frequencies. At fast driving, the particle scarcely can follow in any direction and, on the average, stays close to zero. That must not be misinterpreted, since it merely means that in each phase of the drive the particle stays with nearly equal probability in anyone of

\[\text{Figure 4.1.: Test \#1: } \tau = 10\text{s.}\]
4. Testing the Programs

Figure 4.2.: Test #2: $\tau = 20\text{s}$.

Figure 4.3.: Test #3: $\tau = 50\text{s}$.

Figure 4.4.: Test #4: $\tau = 100\text{s}$.
4.1. Reproducibility of known results

both wells. Furthermore, a dynamical system that persistently “lingers” around an unstable fixed point although being kicked all the time is fairly un-physical.

For longer times (i.e. slower driving, \( \tau = 50 \text{s} \)) an optimum is reached with respect to the hysteresis loop area. Still longer times lead to the previously described behaviour of closing in to an equilibrium distribution. An interesting discrepancy arises in the last test example, fig. 4.5: at the turning points of the driving force, the Kramers rates lead to an averaged position which lies beyond \( \pm x_0 = \pm 0.8 \mu m \), thereby mirroring the effect of the averaged position of the present positions of the minima of the potential landscape. The Langevin simulation, however, does not reach that far. A plausible explanation would be that the barrier height of \( 2.9 k_B T \) does not lead to a perfect concentration of the sojourn probability density within each potential well. In fact, the thermal energy of the particles is high enough that they “feel” the asymmetry of the quartic well which is more strictly cut off to the outward and smoother to the inward direction. The average position therefore is closer to zero.

The next series of diagrams, figures 4.6 to 4.10, reflects the time dependent effects already mentioned. Beginning with fast driving, the first peak at \( \tau/2 = 5 \text{s} \) is vanishingly small, whereas the bulk probability is denoted by the vertical bar
4. Testing the Programs

Figure 4.7.: Residence time distribution #2: $\tau = 20$ s.

Figure 4.8.: Residence time distribution #3: $\tau = 50$ s.

Figure 4.9.: Residence time distribution #4: $\tau = 100$ s.

Figure 4.10.: Residence time distribution #5: $\tau = 200$ s.
right of the highest class of relative frequency. In the following two diagrams one sees how the probability “flows” from long waiting times to short times and thereby shapes a maximal peak area for driving period $\tau = 50s$. The last two residence time distributions collect nearly all the probability within peak number “zero”, indicating a high probability for the particles to change wells multiple times during a single half-period.

4.2. Optimising the timestep

In the following, I look at the same situation as in the previous section, with coupling still switch off, and investigate mean and variance of the resulting quantifiers for different timestep widths. For each timestep, 10 simulations with different realisations of pseudo random numbers were performed, yielding the data for a small statistical analysis.

![Graph](image)

**Figure 4.11.** Quantifiers vs. $\Delta t$: 10 simulations of 1 000 periods for each data point.

The results for the Langevin simulation are shown in fig. 4.11. The number of simulated periods of the driving force was 1 000, yielding a total simulation time of 16 h 40 min. The $HLA$ averages stay close to $-13.6$ for over one order of magnitude in $\Delta t$, whereas the long timestep values ($\Delta t \geq 0.01s$) deviate distinctly. The behaviour of $RES$ is similar with two differences: the mean deviates already for $\Delta t \leq 0.003s$, and the variance is unequal for all timestep widths. For even longer times of order 0.1 s, there was actually an effective probability for the particle to
leave the trap completely. For too large time steps the dynamics became instable and the quartic potential played ping-pong with a particle until the simulation aborted.

![Figure 4.12: Quantifyers vs. Δt: 10 simulations of 5000 periods at each data point.](image)

The next figure, 4.12, demonstrates the same quantifiers from 5 times 10 Langevin simulations of 5000 periods (83 h 20 min). The shortest timestep was left out, since it was already found reliable, so the computing time could be saved. The $HLA$ indicates the same result with greater accuracy as before, whereas the accuracy of $RES$ is but slightly enhanced. $RES$ would suggest that all examined timesteps are good, but $HLA$ can distinguish an optimal point near $Δt = 0.003$ s.

![Figure 4.13: Timesteps for Kramers rates: relative values of $HLA(Δt)$](image)
4.2. Optimising the timestep

In the Kramers rate program, there is no stochasticity involved, so we do not see any error bars. But therefore, in fig. 4.13, the influence of the timestep width $\Delta t$ on the hysteresis loop area is clear to see: the $HLA$ values stay almost unchanged for one order of magnitude, up to $\Delta t = 0.01\text{s}$, and take then an upward turn denoting the increasing error produced by the finite integration interval of the Euler-algorithm. Therefore, $\Delta t = 0.01\text{s}$ is chosen for Kramers rates integration.
4. Testing the Programs
5. Results

5.1. Stochastic resonance and coupling strength

Both techniques, Langevin simulation as well as Kramers rates, provide evidence for an optimisation of the system response to external harmonic driving in dependence on the coupling strength. In the first simulation results to be presented here, hysteresis loop area and residence time distribution are investigated for driving periods of

\[ \tau_1 = 100 \, \text{s}, \quad \tau_2 = 200 \, \text{s}. \]  

The results from the Langevin simulations were obtained by simulating 2000 periods, equivalent to 55 h 33 min and 111 h 6 min, with a timestep of

\[ \Delta t = 5 \, \text{ms}. \]  

The phase difference is

\[ \varphi := \varphi_2 = \pi, \]  

so that the micro beads are driven to reside within the preferred diagonally opposing wells. Each data point refers to 10 simulations, each with a different realisation of the stochastic force. The variance of each data point is below the point size and therefore not represented within the following diagrams.

In fig. 5.1, we can see an almost perfect correspondence of both methods except for the maximal HLA-values. The Kramers rates could not be computed for distances lower than \( d \approx 3.35 \, \mu \text{m} \) because of the distortion of the potential landscape. It prohibits the application of Kramers rates irretrievably since the barrier height of the unfavourable wells (those with equal sign in the \((x_1, x_2)\)-coordinates) is diminished too much. But the tendency towards absolute zero in the high coupling limit is unquestionable. At the maximum itself there is the shortcoming of the Euler-algorithm already mentioned, which yields greater values than the exact solution of the underlying differential equation. But this deviation of approximately 5% can by no means distract us from the fact that there is a clear-cut maximum peak.

There are some obvious differences for distinct driving periods: the absolute height of the HLA peaks is about 5% larger for the longer driving period. The asymptotic values for large distances (i.e. weak coupling) end up in two straight lines:

\[
\lim_{d \to \infty} HLA (\tau = 100 \, \text{s}, d) = -10.4, \quad (5.4)
\]

\[
\lim_{d \to \infty} HLA (\tau = 200 \, \text{s}, d) = -6.1. \quad (5.5)
\]
5. Results

This corresponds to the behaviour of uncoupled systems for extremely low driving frequencies. For these driving frequencies, the position of the maximal $HLA$-value tends towards stronger coupling. Since stronger coupling evokes a higher potential ridge along the main diagonal (cf. fig. 2.5), which in case of two vanishing wells can be seen as the only remaining barrier (but with two passes!), this leads automatically to longer residence times and thereby to the observable frequency shift.

With only two exceptions, everything I said about $HLA$ holds for $RES$, too (cf. fig. 5.2). The first exception is the height of the maximum, which is for both residence time distributions the same. The maximum is reached at nearly identical values of the distance: for $\tau = 100\,\text{s}$, it is

$$d_{\text{max}}^{HLA} = (3.55 \pm 0.05) \, \mu\text{m},$$

$$d_{\text{max}}^{RES} = (3.60 \pm 0.05) \, \mu\text{m},$$

and for $\tau = 200\,\text{s}$, it is

$$d_{\text{max}}^{HLA} = (3.45 \pm 0.05) \, \mu\text{m},$$

$$d_{\text{max}}^{RES} = (3.45 \pm 0.05) \, \mu\text{m}.$$
5.1. Stochastic resonance and coupling strength

The reasons are quite obvious: RES only mirrors the relative proportion of transitions that occur within a given time interval. Whether these are many or few cannot be inferred from RES, whereas HLA measures, in a way, the absolute amount of transitions that occur at the right moment minus those that occur at false moments. A larger amount of transitions at the right moment makes the system trajectory \( x(t) \) spread vertically and thus increases the HLA-value.

The second exception lies within the greater variance of the RES-values. Of course, if the transition rates are extremely low, inter-well jumps of the particles are very rare so that only few events can be counted and measurements diverge stronger. But the errorbars are also visible for high RES-values, thereby indicating a minor exactitude as opposed to the hysteresis loop area, since both quantifiers result from identical simulations.

Nevertheless, the detailed residence time distribution, as depicted in fig. 5.3, leaves no doubt where the maximum of RES originates from. If we apply the “definition” for bona fide stochastic resonance, it is plain to see: the periodic structure with the characteristic peaks at odd integer multiples of \( \tau/2 \) does indicate
resonant behaviour, even if our driving frequency (5 mHz) seems so small. The diagram on the left hand side shows a wide spread distribution over many peaks, for the inter-well motion is still limited by strong repulsion. The diagram in the middle is representative for stochastic resonance. Almost 75% of the jump events occur within the 2σ-interval around \( \tau/2 = 100 \) s. The right hand side diagram shows the growing dominance of the asymptotic \( RES \)-behaviour towards weak coupling: for further extended driving periods, the whole jump probability cumulates under peak number zero.

5.2. Stochastic resonance and phase difference

Having established the interdependence of stochastic resonance and the coupling strength in the preceding section, I will now take another \textit{Ansatz} for an investigation of the phenomenon appearing at long driving times and at a characteristic coupling strength. Up to this point, I did not specially mention the reason why the phase difference in eqn. (5.3) was chosen as \( \varphi = \pi \), except for an \textit{ad hoc} explanation concerning the preference of maximal inter-particle distance. But there is certainly more to discover, as I will point out in this section.

Here, I will present the results of a thorough examination at one specific point of the \((\tau, d)\)-configuration space (which is in first approximation equivalent to the \((\Omega, \phi)\)-configuration space that might be more appealing to a theoretical fundamentalist). This is the

\begin{align*}
\tau &= 300 \text{ s}, \quad (5.10) \\
d &= 3.5 \mu\text{m} \quad (5.11)
\end{align*}

configuration, with all other parameter values identical to those listed in section 2.6. To obtain “presentable” hysteresis loops, an exorbitant simulated time of 20,000 periods, corresponding to 69 d 10 h 40 min, was set, also to ensure smooth graphs of the hysteresis loops. This time, the decisive parameter was the phase difference which naturally ranges from 0 to 2\( \pi \). A stepwidth of 5° was chosen, corresponding to \( \pi/36 \), thus generating 37 data points.

In two diagrams, fig. 5.4 concerning the hysteresis loop area, and fig. 5.5 concerning the residence time distribution, the summary of the results is visualised. The first diagram combines the results from Langevin simulation and Kramers rates, again showing the good accordance between both methods and the “overshoot” of the Kramers rates. The first thing that is still easily understood is the behaviour of the combined system \textit{HLA} which is \textit{HLA}_1 + \textit{HLA}_2, the sum of both single particle quantifiers. It simply changes between minima at \( \varphi = 0, 2\pi \) and a maximum at \( \varphi = \pi \) in a sinusoidal manner. Here the \textit{ad hoc} explanation mentioned above works fine: with zero phase difference, the drives attempt to force the particles to stay closely together, i.e. in neighbouring wells. But the strong coupling prevents this by destabilising this configuration. If both particles really
5.2. Stochastic resonance and phase difference

Figure 5.4.: Hysteresis loop areas at various phase differences.

Figure 5.5.: RES at various phase differences.
stay within corresponding wells, the first trying to escape of anyone of both particles will be successful. Consequently, each of the two particles will stay in the “bad” well for half of the time and thereby lose all the profit from the driving it gained in the other half when it stays in the “good” well. HLA therefore is close to zero.

The opposite is true for \( \varphi = \pi \): the drives are in congruence with the effort of the particles to maximise their distance. As the situation becomes unfavourable for both particles at the same instance of the driving period, their jumps are synchronised for mutual benefit. If either of the micro beads jumps first, the escape rate of the other will be significantly larger by a factor of

\[
\frac{k_1}{k_2} \approx \exp \left( \frac{x_1 A_1 + x_2 A_2}{k_B T} \cos (\Omega t) \right).
\]

The argument of the exponential is minus the potential difference between both particles over \( k_B T \), approximated by the driving forces \( f_1 \) and \( f_2 \). This mechanism vastly diminishes the probability of each particle to be trapped in the “bad” well. The situation must be explained with more delicacy if the single particle behaviour in between these poles of the phase difference is taken into consideration. Of course, the equivalence of the single particle dynamics, as I termed this notion in section 3.3, is mirrored by the symmetries in the single particle results of figures 5.4 and 5.5. Therefore everything stated for \( \varphi = 0 \ldots \pi \) holds also for \( 2\pi - \varphi = 0 \ldots \pi \) with exchanged particle numbers.

When we start at \( \varphi = 0 \) and increase the phase difference slightly, then particle 1 is permanently behind particle 2 with respect to the phase of both driving forces at any given moment. If eqn. (5.12) is applied to this more general situation, it reads:

\[
k_B T \log \frac{k_1}{k_2} \approx (x_1 A_1 \cos (\Omega t) - x_2 A_2 \cos (\Omega t + \varphi)) \approx \rho \cos (\Omega t + \theta)
\]

with new amplitude \( \rho \) and phase \( \theta \):

\[
\rho^2 = x_1^2 A_1^2 + x_2^2 A_2^2 - 2x_1 x_2 A_1 A_2 \cos \varphi, \quad (5.14)
\]

\[
cot \theta = \cot \varphi - \frac{x_1 A_1}{x_2 A_2} \csc \varphi. \quad (5.15)
\]

The sign of \( \cos (\Omega t + \theta) \) now determines, which particle in a nearest well competition will probably win. Furthermore, the interplay of \( \theta \) and the instantaneous drive phase of each particle results in a structural disadvantage for the particle that “hurries ahead”. This is the reason why \( RES_2 \) grows stronger than \( RES_1 \) in the beginning. Most of the time, both particles stay in opposing wells. Any particle will try to jump if it “feels” to sit in the “bad” well, so both will often meet in the “good” well where particle 2 has a structural disadvantage. Now it
becomes clear: with increasing phase difference, particle 2 more often loses the battle for the “good” well, it stays longer in the “bad” well and must often climb its way uphill because it was kicked out by particle 1. In return, particle 2 earns a higher $RES$-value.

![Figure 5.6.: Driving forces for phase difference $\varphi = \pi/2$.](image)

If $\varphi$ increases further, up to $\pi/2$, then the work balance of particle 2 is even, $HLA_2$ close to zero again, whereas particle 1 stays in the prime of its life: a maximum of $HLA_1$. The reason is that every other quadrant both particles are willing to change their respective wells because the driving forces are opponent, cf. fig. 5.6. This happens early for particle 1 and late for particle 2. Since the driving frequency is so very small, the jump will occur very soon after the particles entered this opportune jump phase. Thus it is particle 1 that still profits from the situation in terms of a hysteresis loop gain, whereas particle 2 stays behind. The situation becomes better for particle 2 when the phase difference approaches $\pi$. The timespan in which the forces $f_1$ and $f_2$ are opponent increase further and the disadvantage of particle 2 disappears gradually until equity is reached at $\varphi = \pi$.

In figures 5.7 and 5.8, the hysteresis loops of both particles are plotted. The left hand column of each figure represents particle 1, the right hand column particle 2. The results of the Langevin simulation are depicted with solid lines, whereas the Kramers rates results are drawn with dashed lines. Please, note that the phase difference is implicitly added to the phase $\Omega t$ in all diagrams of the right hand column. From top to bottom, four different stages are picked out:

1. $\varphi = 0$: Both particles describe the same narrow loop. Langevin and Kramers results are nearly indistinguishable. The loop area is close to zero. If not otherwise declared, the rotational direction on the time parametrised path is mathematically positive, i.e. anti-clockwise. Thus a negative $HLA$ is returned from integration.

2. $\varphi = 5/18 \pi = 50^\circ$: At this particular phase difference, particle 2 has a maximal positive hysteresis loop area, signified by the arrows in clockwise rotational direction. The influence on particle 1 is recognisable in the continued growth of $\langle x_1(t) \rangle$ that reaches beyond the instance of maximal force, almost up to the zero node of $f_1$. Although the maximum value of the averaged coordinate is far below $x_0$, $HLA$ is close to its maximum because of the wide spread at $\cos (\Omega t) = 0$. 

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Figure 5.7.: Hysteresis loops at various phase differences, 1.
5.2. Stochastic resonance and phase difference

Figure 5.8.: Hysteresis loops at various phase differences, 2.
3. \( \varphi = 7/12 \pi = 105^\circ \): The zero node of \( HLA_2 \) coincides with the maximum of \( HLA_1 \). The average coordinates of both particles reach maximal amplitude, but the difference is obvious. It is interesting to see that \( \langle x_2(t) \rangle \) behaves as if it was the averaged coordinate of an infinitely slow driven particle which in the mean resides in each well in agreement with the Boltzmann-distribution.

4. \( \varphi = \pi = 180^\circ \): Finally, equity is reached and both particles statistically follow the same hysteresis loop.

![Figure 5.9: Residence time distribution at various phase differences.](image)

The histograms of the residence time distribution, shown in fig. 5.9, refer to particle 1 (solid line) and particle 2 (dashed line) of the Langevin simulation. Please, note that the probabilities are scaled logarithmically. Every “straight line” downwards is a sign of exponential decay, e.g. in the upper left corner. The zero peak is always very prominent since each jump into the direction of the other micro bead has always a chance of direct reversal. The growth of the \( \tau/2 \)-peak is mainly fed by the right flank of the zero peak. This indicates a decrease of the respective decay rate, yielding a more gentle slope.
5.2. Stochastic resonance and phase difference

Figure 5.10.: Potential wells at critical distance; part 1.

The development of the $\tau/2$-peak is more distinctive for particle 2 than for particle 1. With respect to RES as a \textit{bona fide} quantifier of stochastic resonance, in the middle histogram of the lower row there is a clear signal, comparable with fig. 5.3. The fact that both quantifyers reach a maximum value at different phase shifts of the driving forces is a remarkable hint to be cautious. It is remarkable, too, that it is a cooperation where one partner gains a work surplus from its drive while the other partner has to work against its own drive. Naturally, this work is done by the surrounding water molecules, and the water eventually gains all the energy back via viscous dampening. Since the “working” particle also gains something in this process, namely a high phase synchronisation with its driving force, I would denote this cooperation a \textit{coupling induced} stochastic resonance.

There must be a cause for this unexpected property of the colloidal system. Since we can describe it so formidably by Kramers rates, and since the Kramers
5. Results

**Figure 5.11.** Potential wells at critical distance; part 2.

rates exclusively rely on the underlying potential landscape, there must be a good reason to be found in $V(x_1, x_2, t)$, eqn. (2.6). The figures 5.10 and 5.11 show the potential landscape in the $(x_1, x_2)$-coordinate system. All parameter values are chosen identically to the simulation in this section. The four contour-plots present the instance when both driving forces pass through a zero node. In other words, there is momentarily no external bias. The most important feature is obvious: with decreasing distance between the two pairs of optical tweezers, two of the four potential wells disappear. Furthermore, a substantial barrier rises along the $(x_1 = x_2)$-diagonal. The system is at a bifurcation point, transforming the four wells and four saddle points into effectively 2 wells and 2 saddle points.

On the one hand, this explains why the two micro beads undergo such a rivalry about the optimal jump time, even at a distance of 3.5 µm where there are still shallow wells present. If one particle jumps, a next jump will immediately occur,
5.3. Stochastic resonance with four coupled particles

either by the same particle backwards, or by the other particle because, for the two-particle system, the probability to stay in a well with barrier height $\Delta U \approx k_B T$ is very low. If a tilt into one direction is added, the chances get unequally distributed, with all the consequences described above.

On the other hand, the rising barrier on the diagonal through quadrants I and III explains the immensely long driving periods. This time is needed to "climb the wall". In a way this suggests that a proper renormalisation could bring up a connection between coupling strength and noise intensity, thereby leading to a description that is more in accord with classical, noise related concepts dealing with stochastic resonance. The decisive figure therein is the relation between barrier height and noise intensity. We just have seen this also applying to the relation between barrier height and coupling strength, apart from further fundamentally transforming effects like evoking bifurcations.

Because at the actual bifurcation point the wall separating the two remaining wells has risen by more than $12 k_B T$, it prevents any inter-well dynamics (at least on a human time scale). In our models the important quantity is the barrier height of the shallow potential wells. I am convinced that the Kramers rates that are computed for these wells are far from realistic since the whole approximation scheme Kramers adopted to this problem does no longer hold for barrier heights close to thermal energy. Fortunately, this does not play any role since the bulk sojourn probability is concentrated within the two deep wells representing cross-diagonally occupied laser traps. Their properties alone determin what happens dynamically, the shallow wells are merely transition states. Therefore I would denote the distance between double wells that leads to a remaining barrier height of $1 k_B T$ of the transition states a critical distance. For the given parameter values of this section, $d = 3.5 \mu m$ is a good first order estimate of the critical distance.

5.3. Stochastic resonance with four coupled particles

The examination of a four-particle system (cf. fig. 3.1) was initiated by the experiment in Stuttgart. As I already pointed out, there seemed to be "more harmony among four" than with only a pair of micro beads. Willingly I adopted new parameter values and applied them in my third program. The idea was to have 3D-plots of stochastic resonance quantifiers over frequency and coupling strength as a two-dimensional parameter space. For the experimentalist who cannot meticulously control one of those parameters, namely the coupling strength, the search for stochastic resonance could be guided by a map that tells him, in which region of the parameter space his experiment dwells right at the moment.

Most parameters stayed the same. The only new adopted values were the screening constant and the potential barrier that was reduced in height:

$$\kappa = 3.4 \mu m^{-1},$$
$$\Delta U = 2.6 k_B T.$$
Instead of the driving frequency I preferred the period time, thus ensuring equidistant scanning ranges which allow for grids and contour lines to be plotted. The time ranged from \( \tau = 5 \text{s} \) up to 200 \text{s} in steps of 5 \text{s}. The distance between opposing double wells varied from \( d = 4.1 \mu \text{m} \) up to 7.0 \( \mu \text{m} \) in 0.1 \( \mu \text{m} \) steps. Since the distance enters the exponent of the coupling potential, \( \Phi \) passed through four orders of magnitude.

![Figure 5.12.](image1)

**Figure 5.12.:** Quantifiers without coupling.

Two diagrams (fig. 5.12) show the dependence of each quantifier on the period \( \tau \) when the coupling is switched off. They represent the asymptotic behaviour for large distances (i.e. vanishing coupling) of all following diagrams. At \( \tau \approx 35 \text{s} \), both quantifiers are at their maximum and thus demonstrate the property of resonance for the uncoupled systems.

![Figure 5.13.](image2)

**Figure 5.13.:** Quantifiers at very large driving period, \( \tau = 2000 \text{s} \).

The next diagram (fig. 5.13) is enclosed herein for similar reasons as the pre-
5.3. Stochastic resonance with four coupled particles

Figure 5.14.: Configuration #1: HLA over τ and d.

ceeding one: to clarify asymptotic behaviour. In this case it is the large τ limit, and both quantifiers show plain zero lines for certain ranges. The non-zero parts I will discuss later in this section.

In the following, I will present two of the total four configurations that were explained in section 3.3: #1 and #2. As we will see, they differ remarkably in terms of their resonant behaviour at long driving periods at the critical distance. The respective diagrams of the other two configurations are enclosed in appendix A. The sign of HLA is usually negative since the work employed by the driving force is directed into the system. Please, note that for better reading of the grid-plots I have depicted HLA with reversed sign throughout this section.

I start with a discussion of the features to be read off from fig. 5.14:

- At low d-values (strong coupling), HLA is zero because the extremely strong repulsive force prohibits the system from leaving its ultra-deep potential well.
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- At high $d$-values (weak coupling), $HLA$ tends to the asymptotic values for uncoupled systems, as depicted in fig. 5.12.

- At short $\tau$-values, $HLA$ tends to zero. It is the high-frequency limit where the response of the system becomes more and more damped.

- At long $\tau$-values, $HLA$ stays at its maximal value. There is no “closing in” of the hysteresis loop for very long driving periods, as long as the system experiences a critical coupling. Elsewhere, $HLA$ converges to zero, as usual.

- The ridge parallel to the $d$-axis at $\tau \approx 30\,s$ stems from the one-particle stochastic resonance that is also present when the coupling is switched off.

- The second ridge runs almost parallel to the $\tau$-axis at $d \approx 4.6\,\mu m$. It indicates stochastic resonance at a critical distance.

The results for $RES$, shown in fig. 5.15, perfectly coincide with the $HLA$ findings. The ridge at long driving periods indicates the probability flow within the residence time distribution from short times (for zero coupling) to long times (at critical coupling) and to infinity (for overcritical coupling). “Short” is not to be understood literally but with respect to the long-time driving period regime. By this notion I mean driving periods beyond the stochastic resonance for an equivalent uncoupled system, in our case: $\tau \gg 30\,s$. For very long driving periods there is even a higher value of $RES$ than for normal periods (cf. fig. 5.13). This indicates an extremely rigid enforcement of synchronisation by the coupling.

These are the features of fig. 5.16, concerning configuration #2:

- With respect to low and high $d$-values, the behaviour of $HLA$ in configuration #2 mirrors that of configuration #1. Because the effect of the overcritical repulsive force stays the same, nothing else could have been expected.

- For low $\tau$-values, the same effect of the dampend system response is present.

- The high $\tau$-limit drops slowly down to zero.

- The maximal value of $HLA$ is reduce by approximately 40%.

- Most obviously the ridge at the critical distance is missing. Moreover, at the critical distance, the system response is stronger dampend for longer periods than for short periods where the influence of the resonance that governs the uncoupled system is still detectable. This remarkable property is easily seen in the contour plot, where the contour lines at small distances recede to larger distances for longer times.

In this configuration, pairs of particles are forced to meet their neighbours at short distance every half-period. The obstructive effect of the repulsion onto the inter-well dynamics permits no resonance in the sense of an $HLA$-maximum.
5.3. Stochastic resonance with four coupled particles

Figure 5.15.: Configuration #1: RES over $\tau$ and $d$.

When we look at the RES-quantifier in fig. 5.17, HLA seems to miss out on a resonance because, for this configuration, RES also shows a ridge for nearly the same critical coupling strength ($d \approx 4.7 \mu m$) as in the preceding configuration. Moreover, this ridge is present with nearly the same size at very long driving periods, then again shifted to shorter distances meaning stronger coupling. Admittedly, it is significantly diminished in contrast to configuration #1, and by far outranged by the asymptotic towards zero coupling. Alas, it is not HLA but RES that seems to mock us. In order to find out what really happens with RES, we take a look at the residence time distribution.

The six snapshots (fig. 5.18) of the residence time distribution give perfect insight into the artificial nature of the RES-peak in fig. 5.17 resulting from shifts of the average exponential decay rate:

1. For extremely strong coupling, $P_{res}(t)$ is zero everywhere except for $t \to \infty$. 

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If the coupling strength approaches its critical value, scarce transitions occur which instantaneously “jump back”, leading to an increasing delta-like peak at $t \approx 0$.

2. At the critical distance, there is still the delta-like zero-peak, but we recognise an exponential decay (gently declined linear slope at logarithmic scale) with increasing rate. This increase is evoked by the flow of the bulk probability from infinitely long residence times to short times, thereby producing an artificial $RES$-maximum!

3. Below the critical coupling, the exponential decay becomes predominant.

4. At $d \approx 5.3\,\mu m$, the slope reaches its maximum whereas the zero-peak has

Figure 5.16.: Configuration #2: $HLA$ over $\tau$ and $d$. 
5.3. Stochastic resonance with four coupled particles

totally vanished, in other words, it has been completely incorporated by the bulk probability.

5. Further decrease of the coupling reverts the tendency of the slope: the decay rates decrease again.

6. At last, for vanishing coupling, the residence time distribution tends towards its familiar appearance. In this case with a very long period of $\tau = 200\,\text{s}$, there is but a faint reminiscence of the $\tau/2$-peak.

What fig.5.17 really tells us is the stupendous increase of the decay rates for a subcritical distance of $d \approx 5.3\,\mu\text{m}$. This increase leads to the valley behind the phantom ridge in the $RES$ diagram! Furthermore, the predominant exponential
5. Results

![Figure 5.18: Residence time distribution for $\tau = 200\, s$ at various distances.](image)

The decay that is disclosed by the residence time distribution is the original inhibitor of $H\!L\!A$ in this region of the $(\tau, d)$-configuration space. And, as a last result, we can see why this phantom-resonance is still present at very, very long driving periods: because of the percolation of probability from longer to shorter residence times.
6. Conclusions and Outlook

In this thesis, it is shown that there is stochastic resonance in coupled systems of colloidal particles, and that for such systems stochastic resonance is strongly dependent on the coupling strength. A critical distance is identified that leads to an increased coupling induced stochastic resonance.

These findings are verified by two different methods, the simulation of a Langevin stochastic differential equation and the numerical integration of Kramers rates for transitions within a corresponding discretised model. This verification additionally reveals an astounding accordance of both methods that particularly for the case of extremely shallow potential wells could not be expected in advance.

Furthermore, it is shown that the properties of the resonance that is evoked by the coupling differs considerably from the stochastic resonance previously known for an equivalent uncoupled system. By this difference, a specific imperfection of the residence time distribution as a quantifier for stochastic resonance is identified. In all other cases where there is good reason to set confidence into this quantifier, the reliability of the hysteresis loop area is ascertained.

The dependence of stochastic resonance on the phase difference between the harmonically modulated driving forces is investigated, too. An interesting loop inversion and its relation to a cooperative synchronisation effect is detected. It is found out that one of two particles must work against the external driving force, thereby allowing the other particle to gain a surplus of energy out of the drive.

As an extension, the dependence of stochastic resonance on the coupling strength is shown also for a system containing four coupled colloidal particles. In return, I hope for an experimental validation by the collaborating group in Stuttgart.

In my opinion, the connection between the distorting influences of the coupling on the overall potential landscape on the one hand, and the conventionally applied investigation of noise dependant stochastic resonance on the other hand could further clarify the effects mentioned above. In particular the use of Kramers rates turns out to be of greater value than expected. The idea to apply a discrete two or four state drive, thereby allowing for analytical integration of constant rates, was already begun with and will hopefully yield a link between coupling strength, Kramers rates, and hysteresis loop area as a reliable quantifier of stochastic resonance. For better experimental verification, particles coupling via induced magnetic dipoles open up new perspectives for modelling coupling induced stochastic resonance by an excellently controllable coupling potential.
6. Conclusions and Outlook
A. Supplementary Diagrams

These are the contour- and grid-plots of the two further configurations of the four particle Langevin simulation. They were not enclosed in the main part because they resemble configurations #1 and #2 so much.

Figure A.1.: Configuration #3: HLA over $\tau$ and $d$. 
Figure A.2.: Configuration #3: RES over $\tau$ and $d$. 
Figure A.3.: Configuration #4: HLA over $\tau$ and $d$. 
Figure A.4.: Configuration #4: $RES$ over $\tau$ and $d$. 
B. Source Code

I am aware of the missing elegance and clarity of the following source code. However, it worked fine. The code was printed directly from the source files. As it was customised for each purpose, this printout is a mere snap-shot.

B.1. Program c.for

```fortran
program CoupledSystems
  ************************************************************************
  Simulation of one-dimensional Brownian motion of two micro-beads.
  Each bead is trapped in a quartic potential and periodically driven. Both beads are subject to a stochastic force realised by Gaussian white noise with zero mean and temperature dependent variance, and are coupled by a screened Coulomb potential. Discretised Langevin equations are numerically integrated as follows:
  \[
  dx_1 = dt (-x_1^3 + x_1 + \alpha_1 \cos(\omega t) + \xi_1)
  \]
  \[
  dx_2 = dt (-x_2^3 - x_2 - \alpha_2 \cos(\omega t + \phi_{\text{diff}}) + \xi_2)
  \]
  Results are represented in the form of hysteresis diagrams, displaying periodically averaged positions vs. driving force, and residence time distributions (RTD). Hysteresis loop area and area of the first peak of the RTD are analysed.
  ************************************************************************
  implicit none

  integer count(2), i, j, jflag(2), jumps(2,40), n, sets
  integer, allocatable :: period(:), steps(:)
  double precision ar(2), dx(2), ef(2), fp(2), omega, phase(2,500),
    + pi, rtd(2), sigma, thresh, uprime(2), width, x(2), xi(2),
    + z(2)
  double precision, allocatable :: amp(:,:), dist(:), dt(:), du(:),
    + et(sets), kappa(:), kbt(sets), pdiff(sets), phi(sets), tilt(:),
    + x0(:), y(:,:)
  external G05CBF, G05FDF

  integer, allocatable ::
  integer, parameter ::
  double precision, parameter ::
  integer, parameter ::

  sets = 1
  allocate( steps(sets), period(sets), amp(2,sets), dist(sets),
    + dt(sets), du(sets), eta(sets), kappa(sets), kbt(sets),
    + pdiff(sets), phi(sets), x0(sets) )
  pi = dacos(-1.d0)
  period = 50000
  steps = 50000 * period
  kbt = 4.033d0
  x0 = 0.8d0
  dist = 3.7d0
  dt = 5.d-2
  du = 2.9d0 * kbt
  eta = 5.95d-2
```
kappa = 2.5d0
phdiff = pi * 0.5d0
phi = 3.7d5
amp(1,1:sets) = 1.9d0 * kbt / x0(1:sets)
amp(2,1:sets) = 1.9d0 * kbt / x0(1:sets)
c ******** further initialisations *******************************
call G05CBF(222)
open (unit=1, file='c_plt.dat')
open (unit=2, file='c_hla.dat')
open (unit=3, file='c_rpa.dat')
c ******** beginning of data set loop *****************************
do n = 1, sets
allocate ( tilt(2,period(n)), y(2,period(n)) )
jflag = 0
jumps = 0
omega = 2 * pi / dble(period(n))
do i = 1, 500
phase(1,i) = dcos(i * pi / 2.5d2)
phase(2,i) = dcos(i * pi / 2.5d2 + phdiff(n))
enddo
sigma= sqrt(2 * eta(n) * kbt(n) * dt(n))
thresh = x0(n) * .75d0
x = 0.d0
y = 0.d0
c ******* beginning of main loop --> simulation steps ************
do i = 1, period(n)+steps(n)+1
j = mod(i-1,period(n)) + 1
if (i.le.period(n)) then
tilt(1,i) = -amp(1,n) * dcos((i-1) * omega)
tilt(2,i) = -amp(2,n) * dcos((i-1) * omega + phdiff(n))
else
y(1:2,j) = y(1:2,j) + x
end if
call G05FDF(0.d0, sigma, 2, xi)
call eforce(x,dist(n),kappa(n),phi(n), ef)
uprime = 4 * du(n) * x / x0(n)**2 * ((/ 1.d0, 1.d0/) + x / x0(n)**2) * (x / x0(n))**2
x = x + dx
demo
endo
c ******** end of main loop ****************************************
y = y * period(n) / dble(steps(n))
width = period(n) / 500
c ******** segmentation of averaged positions *********************
do i = 1, 500
z = 0.d0
do j = 1, width
z = z + y(1:2,(i-1) * width + j)
endo
z = z / width
write (1,100) phase(1:2,i), z, i
100 format (4f10.5,i10)
demo
c ******** calculation of hysteresis loop area ********************
call area(tilt,period(n),y,ar)
c ******** calculation of residence time distribution ************
count = 0
do i = 1, 40
count = count + jumps(1:2,i)
demo
do i = 1, 2
if (count(i).ne.0) then
rtd(i,1:40) = jumps(i,1:40) / float(count(i))
else
rtd(i,1:39) = 0
rtd(i,40) = 1
endif
demo
do i = 1, 40
write (3,300) (i-1) * period(n) * dt(n) / 20, rtd(1:2,i)
300 format (f10.1,2f10.5)
demo
c ******** first peak of residence time distribution **************
fp = rtd(1:2,6) + rtd(1:2,7) + rtd(1:2,8) + rtd(1:2,9) +
rtd(1:2,10) + rtd(1:2,11) + rtd(1:2,12) + rtd(1:2,13) +
rtd(1:2,14)
write (2,200) period(n), ar, fp
write (6,200) period(n), ar, fp
200 format (i10,4f15.5)
write (1,999)
write (3,999)
999 format (/)
deallocate (tilt, y)
enddo
c ******** end of loop of data sets ***********************
close (1)
close (2)
close (3)
stop
c ********** end of subroutine eforce(x,dist,kappa,phi,ef) **********

subroutine eforce(x,dist,kappa,phi,ef)
  implicit none
  double precision x(2), dist, ef(2), kappa, phi, r, xdiff

  xdiff = x(1) - x(2)
  r = sqrt(dist**2 + xdiff**2)
  ef = (/ 1.d0, -1.d0 /) * phi * (kappa * r + 1) * xdiff * 
      + exp(-kappa * r) / r**3
  return
end

subroutine jumpcount(x,thresh,jflag,i,count,period,jumps)
  implicit none
  integer k, jflag(2), i, j, count(2), period, jumps(2,40)
  double precision x(2), thresh

  do j = 1, 2
    if (jflag(j).eq.0) then
      if (x(j).le.-thresh) then
        jflag(j) = -1
        count(j) = i
      else if (x(j).ge.thresh) then
        jflag(j) = 1
        count(j) = i
      endif
    else
      if (-jflag(j)*x(j).ge.thresh) then
        k = (20 * (i - count(j))) / period + 1
        if (k.gt.40) k = 40
        jumps(j,k) = jumps(j,k) + 1
        jflag(j) = -jflag(j)
        count(j) = i
      endif
    endif
  enddo

  return
end

subroutine area(tilt,period,y,ar)
  implicit none
  integer i, period
  double precision ar(2), tilt(2,period), y(2,period)

  ar = .5d0 * (tilt(1:2,period) - tilt(1:2,1)) * 
      + (y(1:2,period) + y(1:2,1))
  do i = 1, period-1
    ar = ar + .5d0 * (tilt(1:2,i+1) - tilt(1:2,i)) * 
      + (y(1:2,i) + y(1:2,i+1))
  enddo
B. Source Code

return
data
end

B.2. Program r.for

program rates

c ******************************************************************
c corresponds to program CoupledSystems with equivalent structural
parameters; applies Kramers rates description to 2-dimensional
potentials consisting of four quadratically
xy plus an electrostatically repulsive ridge along
xy plus a t-periodic sinusoidal tilt proportional to x.
c ******************************************************************
implicit none
integer i, j, n, m, run, steps
type h(2,2), dist, du, dt, det(8), e(8)
func, g(2), k(2,2), eta, kappa, l(4), mini2(2),
+ pdiff, phi(1:2,0:200), phi0, pi, p(8,2), q, s1, s2, spt(2), t
+ kbt, x(2), x0(2), x(2)
double precision allocatable :: r(:,::), u(:,::), y(:,::)
logical far

pi = dacos(-1.d0)
steps = 7000
kbt = 4.033d0
x0 = (/ .8d0, .8d0 /)
amp = 1.9d0 * kbt / x0
dist = 3.7d0
du = 2.9 * kbt
dt = 0.025d0
eta = 16.75d0
kappa = 2.5d0
cdiff = 0.5d0 * pi
phi0 = 3.7d5
allocate ( r(4,4,0:steps), u(4,0:steps), y(2,8,0:steps) )

** Starting values for approximation of minima and saddle points are **
given with p(i,j); i=1,...,4 : minimum i; i=5,...,8 : saddle point
linking minima i-4 to i-3 mod 4; j=1,2 : x,y coordinate; u(i,n) :
initial probability distribution to find system in minimum i.

** Determination of minima and saddle points following the variable **
metric method in multidimensions in "Numerical Recipes in
FORTRAN 77", section 10.7 at
www.library.cornell.edu/nr/bookfpdf/f10-7.pdf

open (unit = 1, file = 'r6.dat')
open (unit = 2, file = 'r6ar.dat')
open (unit = 1, file = 'r6.dat')
B.2. Program r.for

do m = 0, steps - 1
t = m * 2.d0 * pi / dble(steps)
do i = 1, 8
  z = p(i,1:2)
  far = .true.
do while (far)
    call grad(z,x0,amp,dist,du,kappa,phi0,t,g)
    call hess(z,x0,amp,dist,du,kappa,phi0,t,h)
    far = (dot_product(g,g) .gt. 2.d-20)
    if (far) then
      call minv(h,b)
      z = z - matmul(b,g)
    endif
endo

p(i,1:2) = z
det(i) = h(1,1) * h(2,2) - h(1,2) * h(2,1)
if (det(i) .eq. 0.d0) det(i) = 1.d-9
y(1:2,i,m) = z
if (i .gt. 4) then
  q = (h(1,1) + h(2,2)) / 2.d0
  l(i-4) = abs(q - sqrt(q**2 - det(i)))
endif
endo

l= l / (2.d0 * pi * eta)

do i = 1, 4
  n = mod(i, 4) + 1
  do j = 1, 2
    min1(j) = p(i,j)
    min2(j) = p(n,j)
    spt(j) = p(i+4,j)
  enddo
  q = func(spt,x0,amp,dist,du,kappa,pdiff,phi0,t)
  s1 = func(min1,x0,amp,dist,du,kappa,pdiff,phi0,t)
  s2 = func(min2,x0,amp,dist,du,kappa,pdiff,phi0,t)
  e(i) = exp((s1 - q) / kbt)
  e(i+4) = exp((s2 - q) / kbt)
endo
endo

r(i,i,m) = - r(j,i,m) - r(n,i,m)
endo
endo

r = dt * r
y(1:2,1:4,steps) = y(1:2,1:4,0)
do i = 1, 4
  r(i,i,0:steps) = r(i,i,0:steps) + 1.d0
endo

far = .true.
do m = 0, steps-1
  w(1:4,m+1) = matmul( r(1:4,1:4,m), w(1:4,m) )
endo

far = (abs( maxval( w(1:4,steps) - w(1:4,0) ) ) .gt. 1.d-10)
w(1:4,0) = w(1:4,steps)
B. Source Code

c ***************
c twodimensional system's limit cycle of probability distribution is
reassigned to two onedimensional two-state-systems; using position
of minima at time step m, a t-periodic average of the particle
c position is recorded to output file <'r4.dat'>.
c ***************
do m = 0, 200
phi(1,m) = dcos(pi * m / 1.d2)
phi(2,m) = dcos(pi * m / 1.d2 + pdiff)
enddo
do m = 0, 200
n = m * steps / 200
x(1:2,m) = matmul( y(1:2,1:4,n), w(1:4,n) )
write (1,100) phi(1:2,m), x(1:2,m), w(1:4,n)
enddo
write (1,110)
call area(phi,200,x,ar,amp)
write (2,105) pdiff/pi, ar
print 105, pdiff/pi, ar
enddo
close(1)
close (2)
stop
100 format (8f9.5)
105 format (3f10.4)
110 format (/)
c ***************
function func(x,x0,amp,dist,du,kappa,pdiff,phi0,t)
c ***************
c implicit none
double precision x(2), x0(2), amp(2), dist, du, kappa, pdiff,
+ phi0, t, func, r, xdiff
xdiff = x(1) - x(2)
r = sqrt(xdiff**2 + dist**2)
func = (((x(1) / x0(1))**2 - 1)**2 + ((x(2) / x0(2))**2 - 1)**2)
+ * du + amp(1) * x(1) * dcos(t) + amp(2) * x(2) * dcos(t +
+ pdiff) + phi0 * exp(-kappa * r) / r
return
end

c ***************
subroutine grad(x,x0,amp,dist,du,kappa,pdiff,phi,t,g)
c ***************
implicit none
double precision x(2), x0(2), amp(2), dist, du, kappa, pdiff,
+ phi, t, g(2), c, r, xdiff
xdiff = x(1) - x(2)
r = sqrt(xdiff**2 + dist**2)
c = - phi * (kappa + r + 1) * dxdiff * exp(-kappa * r) / r**3
g(1) = 4.d0 * du/x0(1) + ((x(1)/x0(1))**2 - x(1)/x0(1)) +
+ amp(1) * dcos(t) + c
+ amp(2) * dcos(t + pdiff) - c
return
end

c ***************
subroutine hess(x,x0,dist,du,kappa,phi0,h)
c ***************
implicit none
double precision x(2), x0(2), dist, du, kappa, phi0, h(2,2), c,
+ r, s, xdiff2
xdiff2 = (x(1) - x(2) +)**2
r = sqrt(xdiff2 + dist**2)
s = kappa + r
c = phi0 * exp(-s) / r**3 * (xdiff2 + (s + (s+3.d0) + 3) -
+ r**2 + (s+1.d0))
h(1,1) = du/x0(1)**2 * (12.d0 * (x(1)/x0(1))**2 - 4.d0) + c
h(1,2) = - c
h(2,1) = - c
h(2,2) = du/x0(2)**2 * (12.d0 * (x(2)/x0(2))**2 - 4.d0) + c
B.3. Program q.for

**subroutine minv(h,hinv)**
```fortran
C minv : inverse of matrix h(i,j) is returned in hinv(i,j)
C ******************************************************************
implicit none
double precision h(2,2), hinv(2,2), det
C if (det .eq. 0.d0) det = 1.d0
hinv(1,1) = h(2,2)
hinv(1,2) = - h(2,1)
hinv(2,1) = - h(1,2)
hinv(2,2) = h(1,1)
hinv = hinv / det
return
end
```

**subroutine area(phi,max,y,ar,amp)**
```fortran
C Computation of loop-area within the periodic cycle of the mean
C position of the two system coordinates by trapezoids.
C ******************************************************************
implicit none
integer i, max
double precision ar(2), phi(1:2,0:max), y(2,0:max), amp
ar = 0.d0
C do i = 1, 200
ar = ar + .5d0 * ( phi(1:2,i) - phi(1:2,i-1) ) *
+ ( y(1:2,i) + y(1:2,i-1) )
C enddo
ar = ar * amp
return
end
```

**program Quadrat**
```fortran
C ******************************************************************
implicit none
integer count(4), i, j, jlock(4), jumps(4,40), k, n, sets
integer, allocatable :: period(:), steps(:)
double precision ar(4), dx(4), ef(4), omega, phase(4,250), pi,
+ rtd(4,40), sigma, thresh, width, x(4), xi(4), x0, z(4)
double precision, allocatable :: amp(:,:), dist(:), dt(:), du(:),
+ eta(:), kappa(:), kbt(:), pdiff(:,:), phi(:), tilt(:,:),
+ y(:,:)
C external G05CBF, G05FDF
C ******************************************************************
sets = 30
allocate ( steps(sets), period(sets), amp(2,sets), dist(sets),
+ dt(sets), du(sets), eta(sets), kappa(sets), kbt(sets),
+ pdiff(4,sets), phi(sets) )
pi = dacos(-1.d0)
do i=1, 30
  dist(i) = i * 0.1d0 + 4.d0
endo
C period = 5000
dt = 4.d-2
steps = 10000 * period
kbt = 4.033d0
x0 = 0.8d0
du = 2.6d0 * kbt
dt = 15.75d0
kappa = 3.4d0
pdiff(1,1:sets) = 0.d0
pdiff(2,1:sets) = pi
pdiff(3,1:sets) = 0.d0
pdiff(4,1:sets) = pi
phi = 3.7d5
amp(1,1:sets) = 1.9d0 * kbt / x0
amp(2,1:sets) = 1.9d0 * kbt / x0
```
B. Source Code

```fortran
amp(3,1:sets) = 1.9d0 * kbt / x0
amp(4,1:sets) = 1.9d0 * kbt / x0
call G05CBF(333)
open (unit=1, file='q1.dat')
open (unit=2, file='q1.are')
open (unit=3, file='q1.rtd')

do n = 1, sets
  allocate ( tilt(4,period(n)), y(4,period(n)) )
jlock = 0
jumps = 0
omega = 2 * pi / dble(period(n))
do i = 1, 4
  do j = 1, 250
    phase(i,j) = dcos(j * pi / 1.25d2 + pdiff(i,n))
  enddo
  sigma= sqrt(2 / eta(n) * kbt(n) * dt(n))
  thresh = x0 * .75d0
  x = 0.d0
  y = 0.d0
  c ******************************************************************
do i = 1, steps(n)+1
    j = mod(i-1,period(n)) + 1
    if (i.le.period(n)) then
      do k = 1, 4
        tilt(k,i) = amp(k,n) * dcos((i-1)*omega + pdiff(k,n))
      enddo
    else
      y(1:4,j) = y(1:4,j) + x
      call jumpcount(x,thresh,jlock,i,count,period(n),jumps)
    endif
    call G05FD(0.d0,sigma,4,xi)
    call eforce(x,dist(n),kappa(n),phi(n),ef)
    dx = 4.d0 * du(n) / x0**2 * (x - x**3 / x0**2)
    dx = dt(n) / eta(n) * (dx - tilt(1:4,j) + ef) + xi
    x = x + dx
  enddo
  c ******************************************************************
y = y * period(n) / dble(steps(n))
width = period(n) / 250
do i = 1, 250
  z = 0.d0
  do j = 1, width
    z = z + y(1:4,(i-1) * width + j)
  enddo
  z = z / width
  write (1,100) phase(1:4,i), z
100  format (8f9.4)
  enddo
  call area(tilt,period(n),y,ar)
count = 0
  do i = 1, 40
    count = count + jumps(1:4,i)
  enddo
  do i = 1, 4
    rtd(i,1:40) = jumps(i,1:40) / float(count(i))
  enddo
  do i = 1, 40
    write (3,300) (i-1) * period(n) * dt(n) / 20, rtd(1:4,i)
300  format (f8.1,4f9.4)
  enddo
  z = 0.d0
  do i = 6, 14
    z = z + rtd(1:4,i)
    write (2,200) period(n)*dt(n), dist(n), (ar(1)+ar(2)+ar(3)+ar(4) + )/4.d0, (z(1)+z(2)+z(3)+z(4))/4.d0
200  format (4f15.6)
  endif
  write (3,300) (i-1) * period(n) * dt(n) / 20, rtd(1:4,i)
  write (6,300) period(n)*dt(n), dist(n), (ar(1)+ar(2)+ar(3)+ar(4) + )/4.d0, (z(1)+z(2)+z(3)+z(4))/4.d0
  endif
  if (mod(n,40).eq.0) then
    write (2,*)
    write (6,*)
  endif
  write (1,999)
  write (3,999)
68
```
B.3. Program q.for

999 format (/)
deallocate (tilt, y)
enddo
close (1)
close (2)
close (3)
stop
end

format (/)
deallocate (tilt, y)
enddo
close (1)
close (2)
close (3)
stop
end

c of program

**** subroutine eforce(x,dist,kappa,phi,ef)****

implicit none
integer i, j
double precision x(4), dist, ef(4), kappa, phi, r(4,4), dr(4,4)
do i = 1, 4
j = mod(i,4) + 1
r(i,j) = sqrt((dist/2.d0 - x(i))**2 + (dist/2.d0 + x(j))**2)
x(j,i) = r(i,j)
dr(i,j) = x(i) - dist/2.d0
dr(j,i) = x(j) + dist/2.d0
enddo
do i = 1, 2
r(i, i+2) = sqrt(dist**2 + (x(i) + x(i+2))**2)
x(i+2,i) = r(i,i+2)
dr(i, i+2) = x(i) + x(i+2)
dr(i+2,i) = dr(i,i+2)
enddo
ef = 0.d0
do i = 1, 4
do j = 1, 4
if (i.ne.j) ef(i) = ef(i) + phi * (kappa * r(i,j) + 1) * + exp(-kappa * r(i,j)) / r(i,j)**3 * dr(i,j)
enddo
enddo
return
end

** subroutine jumpcount(x,thresh,jlock,i,count,period,jumps)****

implicit none
integer k, jlock(4), i, j, count(4), period, jumps(4,40)
double precision x(4), thresh
do j = 1, 4
if (x(j).le.-thresh.and.jlock(j).ne.-1) then
if (jlock(j).ne.0) then
k = (20 * (i - count(j))) / period + 1
if (k.gt.40) k = 40
jumps(j,k) = jumps(j,k) + 1
endif
jlock(j) = -1
endif
if (x(j).ge.thresh.and.jlock(j).ne.1) then
if (jlock(j).ne.0) then
k = (20 * (i - count(j))) / period + 1
if (k.gt.40) k = 40
jumps(j,k) = jumps(j,k) + 1
endif
jlock(j) = 1
endif
enddo
return
end

** subroutine area(tilt,period,y,ar)****

implicit none
integer i, period
double precision ar(4), tilt(4,period), y(4,period)
ar = .5d0 * (tilt(1:4,period) - tilt(1:4,1)) * + (y(1:4,period) + y(1:4,1))
do i = 1, period-1
ar = ar + .5d0 * (tilt(1:4,i+1) - tilt(1:4,1)) * + (y(1:4,i) + y(1:4,i+1))
enddo

B. Source Code

```fortran
  enddo
  return
  end

  c  of subroutine
```

Bibliography


Danksagung

Raffiniert ist der Herrgott, aber bosshaft ist er nicht.
Albert Einstein (1879–1955)


Weiteren, mittelbaren Anteil haben alle lieben Menschen aus der ehemaligen Christus-Kirchengemeinde Bielefeld-Senne, aus meinem Kirchenchor und vom C-Kurs, meiner Kirchenmusiker-Ausbildung, die mich, ob bewusst oder unbewusst, zusätzlich in meinem Voranschreiten begleitet und gestärkt haben. Das sollte mir erst gelingen, als ich ziemlich spät auf eigenen Beinen zu stehen gelernt hatte.


Hiermit versichere ich, dass ich außer den angegebenen Quellen keine weitere Literatur verwendet und diese Arbeit selbst erstellt habe.