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# Quantum limits in microscopy and spectroscopy 

by

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## Declarations

This thesis is submitted to the University of Warwick in support of my application for the degree of Doctor of Philosophy. It has been composed by myself and has not been submitted in any previous application for any degree. The work presented (including data generated and data analysis) was carried out by the author. Part of this thesis have been published by the author:

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## Abstract

In recent years quantum metrology and quantum sensing have enabled the advancement of quantum technologies and research in fundamental physics. Through statistical analysis, in the context of parameter estimation, the amount of information about a parameter of interest encoded in a quantum state can be quantified. In this thesis we focus on the application of quantum estimation theory to imaging and spectroscopy with quantum light. We study the fundamental bound of the mean square error for an unbiased estimator in terms of the quantum Fisher information (QFI) for two problems, one in microscopy and one in spectroscopy.

Firstly, we study the problem of localising multiple point sources below the diffraction limit. We show that localisation microscopy of multiple weak, incoherent point sources with possibly different intensities in one spatial dimension is equivalent to estimating the amplitudes of a classical mixture of coherent states of a simple harmonic oscillator. We obtain the QFI matrix elements analytically. In the regime of arbitrarily small separations we find it to be no more than rank two - implying that no more than two independent parameters can be estimated irrespective of the number of point sources. We use the eigenvalues of the classical and quantum Fisher information matrices to compare the performance of spatial-mode demultiplexing and direct imaging in localisation microscopy with respect to the quantum limits.

Secondly, we study the estimation of the electric dipole moment (EDM) of a two-level atom through its interaction with quantum pulses of light in free space. We derive analytical expressions for the states of one-photon wavepackets and entangled photon pairs (EPP). We numerically calculate the QFI for different states of light, including coherent and squeezed pulses, and compare their performance in estimating the EDM. We find that the one-photon wavepacket and the EPP have similar performances, while the performance of the entangled photon pair is not improved by increasing the entanglement of the EPP. Our results indicate that using Fock states to estimate the EDM of a two-level atom is preferable to using entangled light.

## Abbreviations

CFI Classical Fisher information
CFIM Classical Fisher information matrix

CRB Cramér-Rao bound
EDM Electric dipole moment

EPP Entangled photon pair
HG Hermite-Gauss

MSE Mean square error
NA Numerical aperture
PDC Parametric down-conversion

POVM Positive operator-valued measure
PSF Point spread function

QCRB Quantum Cramér-Rao bound
QFI Quantum Fisher information

QFIM Quantum Fisher information matrix

SLD Symmetric logarithmic derivative
SM Schmidt modes

SPADE Spatial-mode demultiplexing

## Chapter 1

## Introduction

Metrology is the field of science that engages in the study of measurement processes. The aim of metrology is to quantify the information available in a system about a given parameter, and to determine the optimal way to extract this information. A measurement process consists of preparing a probe, letting it interact with a target system and eventually measuring the probe. By repeating this process multiple times and analysing the measurement outcomes one can obtain an estimate of the parameter of interest. Metrology provides the tools to attain the most precise estimate of this parameter by using an optimal probe, by improving the detection scheme or, if possible, by optimising both.

The precision of the estimate is affected by statistical errors that lurk in any measurement process. The effects of statistical error on the precision of the estimate can be reduced by repeating the measurement multiple times and averaging the outcomes as a consequence of the central limit theorem. The above process provides the classical shot noise scaling of the estimation errors as $\epsilon=\mathcal{O}\left(N^{-1 / 2}\right)$, where $N$ is the number of repetitions [Kay93]. Ultimately, the fundamental limitations of the precision of the estimate is subject to the principles of quantum mechanics. Using quantum resources for probe states and detection devices the shot noise scaling, which is obtained by classically correlated probes, can be surpassed. Specifically, the Heisenberg scaling on the precision which is achieved for quantum correlated probes is $\epsilon=\mathcal{O}\left(N^{-1}\right)$ [GLM11]. The field of metrology which aims at reaching the fundamental bounds on estimation precision is called quantum metrology.

In recent years quantum metrology and quantum sensing have played a piv-
otal role in the advancement of quantum technologies, in the study of fundamental physics and research in other fields of science. Notably, it has allowed to rigorously determine the ultimate precision of fundamental constants, such as the speed of light [BSF17] or the gravitational constant $[\mathrm{Li}+18]$, parameters of fundamental physics effects, as for example the Unruh-Hawking Effect or spacetime parameters $[A h m+14$; Dow +17 ; Hog12; KBF19; How +18$]$, gravitational wave detection $[\mathrm{Abb}+21 ; \mathrm{BMD} 18 ; \mathrm{Cho}+17]$ and parameters whose precise estimation is vital for the testing of physical models [Bra +19 ; Ahm +18 ; Hog12]. New quantum measurement techniques and quantum probe states have made it possible to develop quantum sensors or optimal detecting schemes [DRC17; Pol +20 ; Law +19 ]. This has led to the advancement of other sciences, from biology [TB16] and medical science [Zie +19 ; Cou +21 ] to earth sciences [RML20].

In this thesis, we are interested in the application of the tools of quantum metrology to imaging and spectroscopy. Improving imaging schemes is particularly important in the measurement of delicate samples for which low illumination and large signal-to-noise ratio are necessary. Quantum imaging techniques may take advantage of quantum states of light and their correlations for producing greater visibility at low intensities and a greater signal-to-noise ratio, such as ghost imaging or imaging with undetected photons, or devise new measurements that achieve resolution beyond the diffraction limit [Mor+19; Alb+20].

Spectroscopy is an indispensable tool for the study of condensed matter physics, chemistry, and molecular biology, in which the structure and dynamics of samples depend on the light that interacts with them [Muk+20; TB16]. Spectroscopy can benefit from the quantum imaging techniques. For instance, a similar method to ghost imaging has been applied to spectroscopy where the spectrum of a sample in the infrared is obtained by detecting the entangled visible photons $[\mathrm{Kal}+16$; Mor +19$]$. Besides the detection schemes, the potential advantages of quantum light, i.e. few-photon or entangled light, are being investigated in spectroscopy in recent years. For example, the two-photon absorption rate of time-frequency entangled photons scales linearly with the beam intensity, in contrast to classical signals that scale quadratically. This could provide another way to perform nonlinear spectroscopy at lower photon fluxes, which is especially significant in photosensitive samples, where large intensity must be avoided [DSM16; Sch17a]. Quantum light also allows to shape and control dynamic processes in molecules in a way that is not possible with
classical light [DSM16; Muk+20]. However, it is still unclear under which techniques and conditions a 'quantum advantage' can be achieved or which quantum states are optimal for a given spectroscopic task [Muk+20; RW20].

## Summary of results

The first contribution of this thesis is in the field of localisation microscopy. We focus on the estimation of the position of $N$ incoherent point sources whose separation is smaller than the diffraction limit. The diffraction limit hinders the resolution performance of optical instruments and has been circumvented by modulating the fluorescence pattern of emitters within a diffraction-limited region [TPR14]. By studying the problem of estimating the separation of two light sources in terms of estimation theory, it has been shown that the diffraction limit is a consequence of the choice of measurement and not a true resolution limit [TNL16]. For realistic imaging scenarios, however, one must consider more than two sources. Order-of-magnitude bounds on the precision of estimating the normalised moments of extended sources smaller the Rayleigh limit have been obtained [ZJ19; Tsa19b].

We calculate an analytical lower bound on an unbiased estimator's covariance (mean square error) matrix for the simultaneous estimation of the locations of $N$ incoherent, weak point sources of unequal but known intensities in one spatial dimension, with the assumption that the point spread function is Gaussian. Our analysis shows that no more than two independent parameters can be estimated in localisation microscopy in the limit of arbitrarily small separations. Furthermore, we compare the performance of conventional direct imaging and the SPADE measurement [TNL16] in localisation microscopy with the quantum bounds we obtain.

The second contribution is in the field of spectroscopy with quantum light. The advantages of using entangled light in spectroscopy remain unclear and a way to quantify them is in the context of quantum metrology. Previous works investigate the role of entanglement in spectroscopy with photon pairs [Ste17] and have calculated the two-photon absorption cross section [SFS21] and both of them indicate that in two-photon interactions entanglement does provide an advantage. We focus on a two-level atom interacting with different states of light and particularly on the estimation of its electric dipole moment. Measuring the electric dipole moment (EDM) of an atom with high precision is important for investigating fundamental
physical models [Chu +19$]$ and for characterising the interaction between atoms and light which is of interest in quantum technologies.

We calculate the lower bound on an unbiased estimator's variance for the EDM of a two-level atom when it interacts with short - picosecond - pulses of light in a free space configuration. We find that an one-photon wavepacket and an entangled photon pair have similar performances, as in Ref. [Ste17]. However, we find that the performance of the entangled photon pair is not improved by increasing the entanglement of the photon pair. Hence, entanglement works as an impediment for the estimation of the electric dipole moment.

The structure of this thesis is the following: in Chapter 2 we introduce the basics of quantum mechanics. We discuss in detail the quantum states of light that we use in this thesis. In Chapter 3 we establish the basic concepts of estimation theory and their generalisation in the context of quantum mechanics. In Chapter 4 we present our results for the estimation of the position of of $N$ incoherent, weak point sources with separations below the diffraction limit. The results presented in this Chapter are based on the publication [BBD19]. Finally, in Chapter 5 we demonstrate our results for the estimation of the electric dipole moment of a twolevel atom with pulses of light. We summarise our results in the conclusions in Chapter 6.

## Chapter 2

## Quantum mechanics

In this chapter we introduce the basics of quantum mechanics. We present in detail the quantum states of light in a cavity and free space setup.

### 2.1 Basics of quantum mechanics

A quantum state can be described by a density operator $\hat{\rho} \in \mathcal{L}(\mathcal{H})$, which is the space of linear operators acting on the vectors in a Hilbert space $\mathcal{H}$. The density operator is a Hermitian, positive semi-definite operator of trace one. Given a basis of the Hilbert space $\mathcal{H}$, the density operator can be represented by a density matrix $\rho$. The quantum states can be classified into pure and mixed states. A pure state is described by a vector (ket) $|\psi\rangle$ in a Hilbert space $\mathcal{H}$ of unit norm $\langle\psi \mid \psi\rangle=1$, where $\langle\psi|$ (bra) is the conjugate transpose of $|\psi\rangle$. The density matrix of a pure state has the form $\rho=|\psi\rangle\langle\psi|$ and satisfies $\operatorname{Tr}\left(\rho^{2}\right)=1$ as a consequence of their unit norm. A mixed state can be expressed as an ensemble of pure states

$$
\begin{equation*}
\rho=\sum_{n} p_{n}\left|\psi_{n}\right\rangle\left\langle\psi_{n}\right| \tag{2.1}
\end{equation*}
$$

where $0<p_{n} \leq 1$ are probabilities and satisfy $\sum_{n} p_{n}=1$.
The measurements performed on a quantum state are represented by the measurement operators $\left\{\hat{\Pi}_{n}\right\}$ associated with each possible measurement outcome
$n$. The probability of each outcome given a state $\rho$ is

$$
\begin{equation*}
\operatorname{Prob}(n)=\operatorname{Tr}\left(\rho \hat{\Pi}_{n}\right) . \tag{2.2}
\end{equation*}
$$

The measurement operators must be Hermitian, positive semi-definite and must satisfy the completeness relation $\sum_{n} \hat{\Pi}_{n}=\hat{\mathbf{I}}$. The completeness relation ensures that the probabilities $\operatorname{Prob}(n)$ sum up to one. This partition of unity by the positive operators $\left\{\hat{\Pi}_{n}\right\}$ is called positive operator-valued measure (POVM). Given an orthonormal basis $\left\{\left|\psi_{i}\right\rangle\right\}$ of the Hilbert space, the simplest example of measurement operators is the set of projection operators $\left\{\hat{P}_{i}=\left|\psi_{i}\right\rangle\left\langle\psi_{i}\right|\right\}$ which satisfy $\hat{P}_{i}^{2}=\hat{P}_{i}$ and $\hat{P}_{i} \hat{P}_{j}=\delta_{i j}$.

### 2.1.1 Entanglement

The quantum states can be composed by one or more subsystems. If a mixed state $\rho$ or pure state $|\psi\rangle$ can be written as a product of the composite $N$ subsystems' states

$$
\begin{equation*}
\rho=\bigotimes_{i=1}^{N} \rho_{i} \quad \text { or } \quad|\psi\rangle=\bigotimes_{i=1}^{N}\left|\psi_{i}\right\rangle \tag{2.3}
\end{equation*}
$$

the state is called separable. A state that is not separable is entangled. The entangled subsystems demonstrate quantum correlations. Entanglement does not account for all non-classical correlations and separable states can demonstrate correlations that are not entirely classical [Mod+12]. In entangled systems the state of one constituent cannot be fully described without considering the other parts of the total, a phenomenon that does not have a classical counterpart.

A bipartite pure system in the Hilber space $\mathcal{H}_{\mathrm{AB}}$ can be decomposed in the so-called Schmidt decomposition as:

$$
\begin{equation*}
\left|\Psi_{\mathrm{AB}}\right\rangle=\sum_{i=1} \sqrt{p_{i}}\left|\phi_{i}\right\rangle_{A}\left|\psi_{i}\right\rangle_{B} \tag{2.4}
\end{equation*}
$$

where $\left\{\left|\phi_{i}\right\rangle\right\},\left\{\left|\psi_{i}\right\rangle\right\}$ are orthornomal basis in $\mathcal{H}_{A}$ and $\mathcal{H}_{B}$ respectively. By calculating the partial density matrix of one of the subsystems, let us say $A$

$$
\begin{equation*}
\rho_{A}=\operatorname{Tr}_{B}\left(\left(\left|\Psi_{A B}\right\rangle\left\langle\Psi_{A B}\right|\right)=\sum_{i=1} p_{i}\left|\phi_{i}\right\rangle\left\langle\left.\phi_{i}\right|_{A},\right.\right. \tag{2.5}
\end{equation*}
$$

we see that the scalars $p_{i}$ are the common eigenvalues of the two sub-systems. Thus, we can associate a Schmidt number, which is the number of non-zero $p_{i}$, to any bipartite pure state. In this way, we can determine whether a pure state is entangled: the state $\left|\Psi_{\mathrm{AB}}\right\rangle$ is entangled if its Schmidt number is greater than one, otherwise it is separable.

There exist different measures for quantum correlations [Mod+12]. In this thesis, we only encounter pure bipartite entangled states $\left|\Psi_{A B}\right\rangle$ for which the von Neumann entropy, or entropy of entanglement, is a good measure of the degree of entanglement between the two subsystems $\mathcal{H}_{A}$ and $\mathcal{H}_{B}$. We note that for mixed states $\rho_{A B}$ the von Neumann entropy fails to distinguish classical and quantum mechanical correlations. The entropy of entanglement is defined as:

$$
\begin{equation*}
S\left(\rho_{A}\right)=-\operatorname{Tr}\left(\rho_{A} \log \rho_{A}\right)=-\operatorname{Tr}\left(\rho_{B} \log \rho_{B}\right) \tag{2.6}
\end{equation*}
$$

and can be expressed in terms of the Schmidt decomposition coefficients as

$$
\begin{equation*}
S\left(\rho_{A}\right)=S\left(\rho_{B}\right)=\sum_{i=1} p_{i} \tag{2.7}
\end{equation*}
$$

In the case of a separable pure state $S\left(\rho_{A}\right)$ is zero, and for maximally entangled states it is $\log 2$.

### 2.1.2 Quantum dynamics

The time evolution of a closed system is fully determined by the system's Hamiltonian operator $\hat{\mathcal{H}}$. The evolution of the density matrix under the Hamiltonian $\hat{\mathcal{H}}$ is given by the von Neumann equation

$$
\begin{equation*}
\frac{\partial}{\partial t} \rho(t)=-\frac{i}{\hbar}[\hat{\mathcal{H}}, \rho(t)] \tag{2.8}
\end{equation*}
$$

Equivalently, the density matrix at some final time $t$ can be related to the density matrix at an initial time $t_{0}$ by the transformation $\rho(t)=\hat{U}\left(t, t_{0}\right) \rho\left(t_{0}\right) \hat{U}\left(t, t_{0}\right)^{\dagger}$. The unitary operator $\hat{U}\left(t, t_{0}\right)$ is called the time evolution operator and satisfies the equation

$$
\begin{equation*}
\frac{\partial}{\partial t} \hat{U}\left(t, t_{0}\right)=-\frac{i}{\hbar} \hat{\mathcal{H}} \hat{U}\left(t, t_{0}\right) \tag{2.9}
\end{equation*}
$$

subject to the initial condition $\hat{U}\left(t_{0}, t_{0}\right)=\hat{\mathbf{I}}$. For a time-independent Hamiltonian the evolution operator has the form

$$
\begin{equation*}
\hat{U}\left(t, t_{0}\right)=\mathrm{e}^{-\frac{i}{\hbar} \hat{\mathcal{H}}\left(t-t_{0}\right)} \tag{2.10}
\end{equation*}
$$

The above description of evolution in quantum mechanics is called the Schrödinger picture. In this description it is the quantum states that evolve in time and not the operators. Conversely, in the Heisenberg picture the quantum states do not e volve in time and the operators do. The two pictures are connected by the transformation of the operators and states under the evolution operator of Eq. (2.10). If $|\psi(t)\rangle$ is a ket in the Schrödinger picture then the state vector in the Heisenberg picture is $\left|\psi_{H}\right\rangle=\hat{U}\left(t, t_{0}\right)^{\dagger}|\psi(t)\rangle$. The operators are transformed according to $\hat{A}^{(\mathrm{H})}(t)=\hat{U}\left(t, t_{0}\right)^{\dagger} \hat{A}^{(S)} \hat{U}\left(t, t_{0}\right)$.

An intermediate picture between the Schrödinger and Heisenberg picture is the so-called interaction picture. In this picture both the operators and states evolve in time. It is a useful description in cases where the Hamiltonian of a system includes a perturbation term or an interaction term between its composite parts. Let us consider a bipartite system whose two parts interact and let the Schrödinger Hamiltonian be written as $\hat{\mathcal{H}}^{(S)}=\hat{\mathcal{H}}_{0}^{(S)}+\hat{\mathcal{H}}_{\text {int }}^{(S)}$, where $\hat{\mathcal{H}}_{0}^{(S)}$ is the free (uncoupled) Hamiltonian and $\hat{\mathcal{H}}_{\text {int }}^{(S)}$ is the interaction (coupling) Hamiltonian between the two parts. The interaction Hamiltonian and the density matrix are transformed according to the following rule

$$
\begin{align*}
& \hat{\mathcal{V}}(t)=\mathrm{e}^{i \hat{\mathcal{H}}_{0}^{(S)} t / \hbar} \hat{\mathcal{H}}_{\mathrm{int}}^{(S)} \mathrm{e}^{-i \hat{\mathcal{H}}_{0}^{(S)} t / \hbar},  \tag{2.11}\\
& \rho_{I}(t)=\mathrm{e}^{i \hat{\mathcal{H}}_{0}^{(S)} t / \hbar} \rho^{(S)}(t) \mathrm{e}^{-i \hat{\mathcal{H}}_{0}^{(S)} t / \hbar}, \tag{2.12}
\end{align*}
$$

where $\hat{U}_{0}(t)=\mathrm{e}^{i \hat{\mathcal{H}}_{0}^{(S)} t / \hbar}$ is the free evolution operator. The states evolve according to the equation

$$
\begin{equation*}
\frac{\partial}{\partial t} \rho_{I}(t)=-\frac{i}{\hbar}\left[\hat{\mathcal{V}}(t), \rho_{I}(t)\right], \tag{2.13}
\end{equation*}
$$

which is the von Neumann equation in the interaction picture.

### 2.2 Quantum states of light

In this section we discuss the quantum description of the electromagnetic field that is useful for quantum optics experiments. It is usually assumed that the field is contained in a 3-dimensional cavity. The Hamiltonian for the quantised radiation field is [MW95]

$$
\begin{equation*}
\hat{\mathcal{H}}(t)=\frac{1}{2} \sum_{\mathbf{k}, s}\left(\hat{p}_{\mathbf{k}, s}^{2}+\omega_{k}^{2} \hat{q}_{\mathbf{k}, s}^{2}\right) \tag{2.14}
\end{equation*}
$$

where the index $s=1,2$ indicates the two possible polarisation vectors of the field and the index $\mathbf{k}$ labels the infinite discrete set of the wavevectors $\mathbf{k}$ which define the modes of the light field. The quantity $\omega_{k}=c|\mathbf{k}|$ is the $k^{t h}$ mode's angular frequency, where $c$ is the speed of light, and $\hat{p}_{\mathbf{k}, s}, \hat{q}_{\mathbf{k}, s}$ are the momentum and position operators that obey the commutation relation

$$
\begin{align*}
{\left[\hat{q}_{\mathbf{k}, s}, \hat{p}_{\mathbf{k}^{\prime}, s^{\prime}}\right] } & =i \hbar \delta_{\mathbf{k}, \mathbf{k}^{\prime}}^{3} \delta_{s, s^{\prime}},  \tag{2.15}\\
{\left[\hat{q}_{\mathbf{k}, s}, \hat{q}_{\mathbf{k}^{\prime}, s^{\prime}}\right] } & =\left[\hat{p}_{\mathbf{k}, s}, \hat{p}_{\mathbf{k}^{\prime}, s^{\prime}}\right]=0 .
\end{align*}
$$

The Hamiltonian of Eq. (2.14) can be recast in terms of the dimensionless annihilation and creation operators

$$
\begin{align*}
& \hat{a}_{\mathbf{k}, s}=\frac{1}{\sqrt{2 \hbar \omega}}\left(\omega_{k} \hat{q}_{\mathbf{k}, s}+i \hat{p}_{\mathbf{k}, s}\right),  \tag{2.16}\\
& \hat{a}_{\mathbf{k}, s}^{\dagger}=\frac{1}{\sqrt{2 \hbar \omega}}\left(\omega_{k} \hat{q}_{\mathbf{k}, s}-i \hat{p}_{\mathbf{k}, s}\right), \tag{2.17}
\end{align*}
$$

which obey the bosonic commutation relations

$$
\begin{equation*}
\left[\hat{a}_{\mathbf{k}, s}, \hat{a}_{\mathbf{k}^{\prime}, s^{\prime}}^{\dagger}\right]=\delta_{\mathbf{k}, \mathbf{k}^{\prime}}^{3} \delta_{s, s^{\prime}}, \quad\left[\hat{a}_{\mathbf{k}, s}, \hat{a}_{\mathbf{k}^{\prime}, s^{\prime}}\right]=\left[\hat{a}_{\mathbf{k}, s}^{\dagger}, \hat{a}_{\mathbf{k}^{\prime}, s^{\prime}}^{\dagger}\right]=0 \tag{2.18}
\end{equation*}
$$

and give the form

$$
\begin{equation*}
\hat{\mathcal{H}}(t)=\sum_{\mathbf{k}, s} \hbar \omega_{k}\left(\hat{a}_{\mathbf{k}, s}^{\dagger} \hat{a}_{\mathbf{k}, s}+\frac{1}{2}\right) . \tag{2.19}
\end{equation*}
$$

The electric field operator is given by [MW95]

$$
\begin{equation*}
\hat{\mathbf{E}}(\mathbf{r}, t)=\sum_{\mathbf{k}, s}\left(\frac{\hbar \omega}{2 \epsilon_{0} V}\right)^{1 / 2}\left(\hat{a}_{\mathbf{k}, s} \varepsilon_{\mathbf{k}, s} \mathrm{e}^{i\left(\mathbf{k} \cdot \mathbf{r}-\omega_{k} t\right)}+\hat{a}_{\mathbf{k}, s}^{\dagger} \varepsilon_{\mathbf{k}, s}^{*} \mathrm{e}^{-i\left(\mathbf{k} \cdot \mathbf{r}-\omega_{k} t\right)}\right) \tag{2.20}
\end{equation*}
$$

where $\varepsilon_{\mathbf{k}}$ is the polarisation vector, $\epsilon_{0}$ is the electric permittivity of vacuum and $V$ is the volume of the cavity.

The eigenstates of the Hamiltonian operator (2.19) are the number or Fock states

$$
\begin{equation*}
\hat{\mathcal{H}}(t)\left|n_{\mathbf{k}, s}\right\rangle=E_{n, \mathbf{k}}\left|n_{\mathbf{k}, s}\right\rangle, \tag{2.21}
\end{equation*}
$$

where $E_{n, \mathbf{k}}=\hbar \omega_{k}(n+1 / 2), n=0,1, \ldots$ is the energy value of the eigenstate $\left|n_{\mathbf{k}, s}\right\rangle$. These states correspond to a specific energy and frequency value and are monochromatic states of light. The state $\left|n_{\mathbf{k}, s}\right\rangle$ is also an eigenstate of the number operator $\hat{N}_{\mathbf{k}, s}=\hat{a}_{\mathbf{k}, s}^{\dagger} \hat{a}_{\mathbf{k}, s}$ with eigenvalue $n$, i.e. the Fock states are states of light of specific photon number $n$ in the specific mode $\mathbf{k}$. The Fock states form a complete set of orthonormal vectors and can be used as a basis. The creation and annihilation operators act on the Fock states as

$$
\begin{equation*}
\hat{a}_{\mathbf{k}, s}\left|n_{\mathbf{k}, s}\right\rangle=\sqrt{n}\left|n_{\mathbf{k}, s}-1\right\rangle, \quad \hat{a}_{\mathbf{k}, s}^{\dagger}\left|n_{\mathbf{k}, s}\right\rangle=\sqrt{n+1}\left|n_{\mathbf{k}, s}+1\right\rangle, \tag{2.22}
\end{equation*}
$$

that is they destroy and create a photon in the mode $\mathbf{k}$. Finally, it is convenient to define the dimensionless quadrature operators

$$
\begin{equation*}
\hat{X}_{\mathbf{k}, s}=\frac{1}{2}\left(\hat{a}_{\mathbf{k}, s}^{\dagger}+\hat{a}_{\mathbf{k}, s}\right), \quad \hat{P}_{\mathbf{k}, s}=\frac{1}{2} i\left(\hat{a}_{\mathbf{k}, s}^{\dagger}-\hat{a}_{\mathbf{k}, s}\right), \tag{2.23}
\end{equation*}
$$

which are equivalent to the position and momentum operators.
The second state of light that interests us are the coherent states of light $\left|\left\{\alpha_{\mathbf{k}, s}\right\}\right\rangle$ which are the eigenstates of the annihilation operator

$$
\begin{equation*}
\hat{a}_{\mathbf{k}, s}\left|\left\{\alpha_{\mathbf{k}, s}\right\}\right\rangle=\alpha_{\mathbf{k}}\left|\left\{\alpha_{\mathbf{k}, s}\right\}\right\rangle, \tag{2.24}
\end{equation*}
$$

where $\alpha_{\mathbf{k}}$ is the amplitude of the $k^{\text {th }}$ mode of the above multi-mode coherent state. By defining the unitary displacement operator $\hat{\mathcal{D}}\left(\alpha_{\mathbf{k}}\right)$ as

$$
\begin{equation*}
\hat{\mathcal{D}}\left(\alpha_{\mathbf{k}}\right)=\mathrm{e}^{\alpha_{\mathbf{k}} \hat{a}_{\mathbf{k}, s}^{\dagger}-\alpha_{\mathbf{k}}^{*} \hat{a}_{\mathbf{k}, s}}=\mathrm{e}^{-\left|\alpha_{\mathbf{k}}\right|^{2} / 2} \mathrm{e}^{\alpha_{\mathbf{k}} \hat{a}_{\mathbf{k}, s,}^{\dagger}} \mathrm{e}^{-\alpha_{\mathbf{k}}^{*} \hat{a}_{\mathbf{k}, s}}, \tag{2.25}
\end{equation*}
$$

the coherent states can be redefined as $\left|\alpha_{\mathbf{k}}\right\rangle=\hat{\mathcal{D}}\left(\alpha_{\mathbf{k}}\right)|0\rangle$. In the following we focus on the single-mode case for convenience. The displacement operator acts on the
annihilation operators of each mode as

$$
\begin{equation*}
\hat{\mathcal{D}}(\alpha)^{\dagger} \hat{a} \hat{\mathcal{D}}(\alpha)=\hat{a}+\alpha, \quad \hat{\mathcal{D}}(\alpha) \hat{a} \hat{\mathcal{D}}(\alpha)^{\dagger}=\hat{a}-\alpha \tag{2.26}
\end{equation*}
$$

By taking the hermitian conjugate of the above expression we can obtain the identities for the creation operator $\hat{a}^{\dagger}$. The derivative of the displacement operator with respect to the amplitude $\alpha$ can be found by differentiating the Eq. (2.25)

$$
\begin{equation*}
\frac{\partial}{\partial \alpha} \hat{\mathcal{D}}(\alpha)=\hat{\mathcal{D}}(\alpha)\left(\hat{a}^{\dagger}-\frac{\alpha^{*}}{2}\right) . \tag{2.27}
\end{equation*}
$$

The coherent states can be expanded onto the Fock basis and obtain the expression

$$
\begin{equation*}
|\alpha\rangle=\mathrm{e}^{-|\alpha|^{2} / 2} \sum_{n=0}^{\infty} \frac{\alpha^{n}}{\sqrt{n!}}|n\rangle . \tag{2.28}
\end{equation*}
$$

Using the above relation and the properties of the Fock states, it can be proven that the set of all coherent states $|\alpha\rangle$ is a non-orthogonal $\left|\left\langle\alpha \mid \alpha^{\prime}\right\rangle\right|^{2}=\exp \left\{-\left|\alpha-\alpha^{\prime}\right|^{2}\right\}$ and overcomplete set $\int d \alpha^{2}|\alpha\rangle\langle\alpha|=\mathbb{I} / \pi$. Hence, any state can decomposed onto the set of coherent states.

The coherent states are minimum-uncertainty states so that $\Delta \hat{p} \Delta \hat{q}=\hbar / 2$, or equivalently using the quadrature operators, $\Delta \hat{X} \Delta \hat{P}=1 / 4$ with $\Delta \hat{X}=\Delta \hat{P}$. A state of light can have one of its quadrature uncertainty less than $1 / 4$. One example of such states is the so-called squeezed state. The single-mode squeezed vacuum state is defined by

$$
\begin{equation*}
|\zeta\rangle=\hat{S}(\zeta)|0\rangle \tag{2.29}
\end{equation*}
$$

where $\hat{S}(\zeta)$ is the unitary squeezing operator defined as

$$
\begin{equation*}
\hat{S}(\zeta)=\mathrm{e}^{\frac{1}{2}\left(\zeta^{*} \hat{a}^{2}-\zeta \hat{a}^{\dagger 2}\right)} \tag{2.30}
\end{equation*}
$$

and $\zeta$ is the squeeze parameter with amplitude and phase defined by $\zeta=r \mathrm{e}^{i \vartheta}$. The number-state expansion of the squeezed vacuum is

$$
\begin{equation*}
|\zeta\rangle=\frac{1}{\sqrt{\cosh r}} \sum_{n=0}^{\infty}(-1)^{n} \frac{\sqrt{(2 n)!}}{2^{n} n!}\left(\mathrm{e}^{i \vartheta} \tanh r\right)^{n}|2 n\rangle \tag{2.31}
\end{equation*}
$$

The variance of the quadratures of a squeezed state with real $\zeta$ are $\Delta^{2} \hat{X}=\mathrm{e}^{-2 r} / 4$ and $\Delta^{2} \hat{P}=\mathrm{e}^{+2 r} / 4$.

### 2.3 Pulses of quantum light

The description of the electric field of Sec. 2.2 assumes that the electric field is confined in a finite cavity space. A typical optical experiment, however, involves light beams propagating in free space for which the cavity quantisation of the electric field is not adequate. In order to describe the continuous-mode field operators we use the one-dimensional formalism of Ref. [Blo +90$]$. In this formalism the light is considered to propagate in a straight line and the transverse propagation is ignored, i.e. the field is well described by a single direction of the wavevector $\mathbf{k}$. The field is assumed to propagate along the the $z$-axis and to have a finite and fixed cross-section $A$ perpendicular to the $z$-axis. To quantise the field we consider that

$$
\begin{equation*}
\Delta k=\frac{2 \pi}{L} \Rightarrow \Delta \omega=\frac{2 \pi c}{L} \tag{2.32}
\end{equation*}
$$

which tends to zero as the length along the propagation axis $L$ tends to infinity. Hence, light is described by a continuum of modes and the sum that appears in the Hamiltonian (2.19) and electric field operator (2.20) is converted to an integral according to the rule

$$
\begin{equation*}
\sum_{k} \rightarrow \frac{1}{\Delta \omega} \int_{0}^{\infty} d \omega \tag{2.33}
\end{equation*}
$$

The continuous-mode creation and annihilation operators are obtained by the transformation

$$
\begin{equation*}
\hat{a}_{k} \rightarrow \sqrt{\Delta \omega} \hat{a}(\omega), \quad \text { and } \quad \hat{a}_{k}^{\dagger} \rightarrow \sqrt{\Delta \omega} \hat{a}^{\dagger}(\omega) \tag{2.34}
\end{equation*}
$$

and satisfy $\hat{a}(\omega)|0\rangle=0$ as in the discrete-mode quantisation. Their commutation relation is obtained by Eq. (2.34) along with the relation $\delta_{k k^{\prime}} \rightarrow \Delta \omega \delta\left(\omega-\omega^{\prime}\right)$ and is

$$
\begin{equation*}
\left[\hat{a}(\omega), a^{\dagger}\left(\omega^{\prime}\right)\right]=\delta\left(\omega-\omega^{\prime}\right) \tag{2.35}
\end{equation*}
$$

Following the above procedure we obtain the continuous-mode free electromagnetic field Hamiltonian

$$
\begin{equation*}
\hat{\mathcal{H}}=\int_{0}^{\infty} d \omega \hbar \omega \hat{a}^{\dagger}(\omega) \hat{a}(\omega) \tag{2.36}
\end{equation*}
$$

where we neglect the energy of the vacuum. The number operator is defined as

$$
\begin{equation*}
\hat{N}=\int_{0}^{\infty} d \omega \hat{a}^{\dagger}(\omega) \hat{a}(\omega) \tag{2.37}
\end{equation*}
$$

Likewise, we can obtain the expression for the electric field operator. The positivefrequency part of the electric field operator in the Heisenberg picture is

$$
\begin{equation*}
\hat{E}^{+}(z, t)=i \int_{0}^{\infty} d \omega \sqrt{\frac{\hbar \omega}{4 \pi \epsilon_{0} A c}} \hat{a}(\omega) \mathrm{e}^{-i \omega(t-z / c)} \tag{2.38}
\end{equation*}
$$

The transverse quantisation area $A$ is formally given by the value of the transverse electric field at the location $\left(x_{0}, y_{0}, z\right)$ that we are interested in each time.

We are interested in situations where the light source is narrow-band, i.e. the frequency bandwidth of the light beam is much smaller than its central frequency $\omega_{c}$. Under this narrow bandwidth approximation we can replace the $\sqrt{\omega}$ with its value the central frequency $\omega_{c}$ and extend the limits of integration from 0 to $-\infty$ without significant errors. The electric field operator then becomes

$$
\begin{equation*}
\hat{E}^{+}(z, t)=i \sqrt{\frac{\hbar \omega_{c}}{2 \epsilon_{0} c}} a\left(t-\frac{z}{c}\right), \tag{2.39}
\end{equation*}
$$

since the narrow bandwidth approximation allows us to define the Fourier transformed operators

$$
\begin{equation*}
\hat{a}(t)=\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} d \omega \hat{a}(\omega) \mathrm{e}^{-i \omega t} \tag{2.40}
\end{equation*}
$$

The operators of Eq. (2.40) obey the commutation relation

$$
\begin{equation*}
\left[\hat{a}(t), \hat{a}^{\dagger}\left(t^{\prime}\right)\right]=\delta\left(t-t^{\prime}\right) \tag{2.41}
\end{equation*}
$$

Let us consider a narrow-band function $\xi(\omega)$ in frequency, the spectral density function, and define the discrete photon-wavepacket creation operator

$$
\begin{equation*}
\hat{A}_{\xi}^{\dagger}=\int d \omega \xi(\omega) \hat{a}^{\dagger}(\omega)=\int d t \xi(t) \hat{a}^{\dagger}(t) \tag{2.42}
\end{equation*}
$$

where the time-dependent form is obtained by the use of Fourier transforms. For simplicity, we consider the spectral density function to be normalised, so that the operator $\hat{A}_{\xi}^{\dagger}$ creates a single photon in the wavepacket or temporal mode $\xi(t)$ defined
as

$$
\begin{equation*}
\left|1_{\xi}\right\rangle=\hat{A}_{\xi}^{\dagger}|0\rangle . \tag{2.43}
\end{equation*}
$$

The operator $\hat{A}_{\xi}$ satisfies the commutation relation $\left[\hat{A}_{\xi}, \hat{A}_{\xi}^{\dagger}\right]=1$ and can be considered a linear superposition of the operators $\hat{a}(\omega)$ with respect to the weight function $\xi(\omega)$. The continuous-mode photon number states are constructed in a similar way as the discrete-mode Fock states by successive application of the creation operator $\hat{A}_{\xi}^{\dagger}$ [Blo+90; RMS07]

$$
\begin{equation*}
\left|n_{\xi}\right\rangle=\frac{1}{\sqrt{n!}} \hat{A}_{\xi}^{\dagger}|0\rangle, \tag{2.44}
\end{equation*}
$$

which are eigenstates of the number operator (2.37). The action of the annihilation operator (2.40) onto the continuous Fock states is

$$
\begin{equation*}
\hat{a}(t)\left|n_{\xi}\right\rangle=\sqrt{n} \xi(t)\left|n_{\xi}-1_{\xi}\right\rangle, \tag{2.45}
\end{equation*}
$$

by taking into account that $\left[\hat{a}(\omega), \hat{A}_{\xi}^{\dagger n}\right]=n \xi(\omega) \hat{A}_{\xi}^{\dagger n-1}$ and taking its Fourier transform. The action of $\hat{a}^{\dagger}(t)$ can be defined in a similar manner.

The continuous-mode coherent states $\left|\alpha_{\xi}\right\rangle$ are generated by a continuousmode displacement operator [Blo +90 ]

$$
\begin{equation*}
\left|\alpha_{\xi}\right\rangle=\mathrm{e}^{\int d \omega\left[\xi(\omega) \hat{a}^{\dagger}(\omega)-\xi^{*}(\omega) \hat{a}(\omega)\right]}|0\rangle=\mathrm{e}^{-\alpha^{2} / 2} \mathrm{e}^{\int d \omega \xi(\omega) \hat{a}^{\dagger}(\omega)}|0\rangle, \tag{2.46}
\end{equation*}
$$

where $\alpha^{2}=\langle N\rangle=\int d \omega|\xi(\omega)|^{2}$ is the average photon number of the coherent state. As the discrete-mode coherent states, they satisfy $\hat{a}(\omega)\left|\alpha_{\xi}\right\rangle=\xi(\omega)\left|\alpha_{\xi}\right\rangle$.

It is useful to express the continuous-mode operator $\hat{a}(\omega)$ in terms of an arbitrary discrete set of basis functions. By considering a complete set of orthonormal functions $\left\{f_{k}(\omega)\right\}$, which satisfy the orthonormality and completeness conditions

$$
\begin{equation*}
\int d \omega f_{k}^{*}(\omega) f_{l}(\omega)=\delta_{k, l}, \quad \text { and } \quad \sum_{k} f_{k}^{*}(\omega) f_{k}\left(\omega^{\prime}\right)=\delta\left(\omega-\omega^{\prime}\right) \tag{2.47}
\end{equation*}
$$

we can define a new set of operators

$$
\begin{equation*}
\hat{A}_{k}^{\dagger}=\int d \omega f_{k}(\omega) \hat{a}^{\dagger}(\omega) \tag{2.48}
\end{equation*}
$$

which obey the boson commutation relation $\left[\hat{A}_{k}, \hat{A}_{l}^{\dagger}\right]=\delta_{k l}$. The inverse relation also
holds

$$
\begin{equation*}
\hat{a}^{\dagger}(\omega)=\sum_{k=0}^{\infty} f_{k}^{*}(\omega) \hat{A}_{k}^{\dagger} . \tag{2.49}
\end{equation*}
$$

An example of a set of orthogonal functions are the Hermite-Gauss functions that we will use for the description of the entangled photon pair.

We can also define the Fourier transform with a frequency shifting equal to the central frequency of the light beam as

$$
\begin{equation*}
\hat{b}(t)=\frac{1}{\sqrt{2 \pi}} \int_{-\infty}^{\infty} d \omega \hat{a}(\omega) \mathrm{e}^{-i\left(\omega-\omega_{c}\right) t} \tag{2.50}
\end{equation*}
$$

The operators $\hat{b}(t), \hat{b}^{\dagger}(t)$ obey the commutation relation of Eq. (2.41). In the same way, we can define the shifted Fourier transform of the spectral function as

$$
\begin{equation*}
\tilde{\xi}(t) \rightarrow \xi(t) \mathrm{e}^{i \omega_{c} t} \tag{2.51}
\end{equation*}
$$

The action of $\hat{b}^{\dagger}(t)$ onto the one-photon wavepacket is

$$
\begin{equation*}
\hat{b}(t)\left|n_{\xi}\right\rangle=\sqrt{n} \tilde{\xi}(t)\left|n_{\xi}-1_{\xi}\right\rangle \tag{2.52}
\end{equation*}
$$

Finally, we note that in this thesis we consider Gaussian shaped spectral density functions of the form

$$
\begin{equation*}
\xi(\omega)=\frac{1}{\sqrt{\sqrt{\pi} \sigma}} \mathrm{e}^{-\frac{\left(\omega-\omega_{c}\right)^{2}}{2 \sigma^{2}}} \rightarrow \xi(t)=\sqrt{\frac{\sigma}{\sqrt{\pi}}} \mathrm{e}^{-\sigma^{2} t^{2} / 2} \mathrm{e}^{-i \omega_{c} t} \tag{2.53}
\end{equation*}
$$

where $\omega_{c}$ is the central frequency of the pulse and $\sigma$ is the bandwidth which satisfy the relation $\omega_{c} \gg \sigma$ and $\xi(t)$ is the Fourier transform of $\xi(\omega)$.

### 2.3.1 Entangled light

The Fock states of Eq. (2.44) are a sub-case of the more general set of $n$-photon states whose spectral density function is not necessarily in a product form [RMS07]. In this section we will describe two special cases of multi-photon states with non-factorisable spectral density functions, the single-continuous-mode squeezed vacuum state and the entangled photon pair (EPP). Both of those states are produced by a parametric down-conversion (PDC) process, with the squeezed state being produced in the high-
gain regime of the PDC [Was+06] and the EPP in the low-gain regime [GW97]. The PDC process is a non-linear optical process in which a photon of frequency $\omega_{p}$ from an incident pump beam propagating in a non-linear crystal may split into two photons of lower energy. The PDC unitary is described by [Chr +13 ]

$$
\begin{equation*}
\hat{U}_{\mathrm{PDC}}=\exp \left\{-\frac{i}{\hbar} \int d \omega_{1} d \omega_{2} \Phi\left(\omega_{1}, \omega_{2}\right) \hat{a}_{1}^{\dagger}\left(\omega_{1}\right) \hat{a}_{2}^{\dagger}\left(\omega_{2}\right)+\text { h.c. }\right\} \tag{2.54}
\end{equation*}
$$

where $\Phi\left(\omega_{1}, \omega_{2}\right)$ is defined as

$$
\begin{equation*}
\Phi\left(\omega_{1}, \omega_{2}\right)=B f\left(\omega_{1}+\omega_{2}\right) \operatorname{sinc}\left(\frac{\Delta k\left(\omega_{1}, \omega_{2}\right) L}{2}\right) . \tag{2.55}
\end{equation*}
$$

Here $B$ is proportional to the pump amplitude that determines the strength of the PDC process, and hence the mean photon number, $f$ is the pump pulse envelope, $L$ is the length of the PDC crystal and the wavevector mismatch is equal to

$$
\begin{equation*}
\Delta k\left(\omega_{1}, \omega_{2}\right)=\left(k_{p}-k_{1}\right)\left(\omega_{1}-\bar{\omega}_{1}\right)+\left(k_{p}-k_{2}\right)\left(\omega_{2}-\bar{\omega}_{2}\right) \tag{2.56}
\end{equation*}
$$

with $\bar{\omega}_{i}$ being the central frequency of each photon.
The continuous-mode squeezed vacuum state $|\xi\rangle$ is the result of a degenerate PDC in which the photons that are emitted from the non-linear crystal are indistinguishable. Mathematically they can be constructed in a similar manner to the discrete-mode squeezed states by a generalisation of the squeezing operator as [Lou00, §6.9]

$$
\begin{equation*}
|\xi\rangle=\hat{\mathrm{S}}(\xi)|0\rangle=\mathrm{e}^{\frac{1}{2} \int d \omega \xi(\omega) \hat{a}^{\dagger}(\omega) a^{\dagger}\left(\omega_{p}-\omega\right)}|0\rangle . \tag{2.57}
\end{equation*}
$$

The spectral density function $\xi(\omega)$ must satisfy the following symmetry requirement $\xi\left(\omega_{p}-\omega\right)=\xi(\omega)$. The central frequency of the down-converted beam is $\omega_{c}=\omega_{p} / 2$. The spectral density function can be written in terms of an amplitude and a phase function as $\xi(\omega)=r(\omega) \mathrm{e}^{i \vartheta(\omega)}$. The average photon number for the above state is $\langle n\rangle=\int d \omega|\xi(\omega)|^{2}=\sinh ^{2} r\left(\omega_{c}\right)$.

The entangled photon pair, also called biphoton state, is obtained from an spontaneous PDC (SPDC) process for low pump intensities. In the weak PDC regime the $\hat{U}_{\text {PDC }}(2.54)$ can be approximated up to the first order term and be applied on the vacuum. The zeroth order term, which is the vacuum, is ignored and, hence, the

EPP | $\left.11_{\text {ent }}\right\rangle$ is described by [GUW01]

$$
\begin{equation*}
\left|11_{\mathrm{ent}}\right\rangle=\int d \omega_{1} d \omega_{2} \Phi\left(\omega_{1}, \omega_{2}\right) a_{1}^{\dagger}\left(\omega_{1}\right) a_{2}^{\dagger}\left(\omega_{2}\right)|00\rangle . \tag{2.58}
\end{equation*}
$$

We assume that the pump envelope is a Gaussian function and that the phase matching function can be approximated by a Gaussian function. Under these assumptions the joint spectral density function has the form [GUW01]

$$
\begin{equation*}
\Phi\left(\omega_{1}, \omega_{2}\right)=\alpha \frac{1}{\sqrt{\pi \sigma_{p}}} \mathrm{e}^{-\left(\omega_{1}+\omega_{2}-2 \omega_{p}\right)^{2} / \sigma_{p}^{2}} \mathrm{e}^{-\gamma\left(\Delta k\left(\omega_{1}, \omega_{2}\right) L\right)^{2}} \tag{2.59}
\end{equation*}
$$

where $\gamma=0.04822$ is the numerical value for which the Gaussian function best approximates the sinc function, $\sigma_{p}$ is the width of the pump beam and $\omega_{p}$ the central frequency of the pump. In Fig. 2.1 the joint spectral density functions of an EPP with different entropies of entanglement are plotted.

As any bipartite state, the EPP admits a Schmidt decomposition [LWE00]:

$$
\begin{equation*}
\Phi\left(\omega_{1}, \omega_{2}\right)=\sum_{n} \sqrt{\lambda_{n}} \psi_{n}\left(\omega_{1}\right) \phi_{n}\left(\omega_{2}\right) \tag{2.60}
\end{equation*}
$$

where the two sets of eigenfunctions must each obey the orthogonality conditions of Eq. (2.47). For the spectral density function of Eq. (2.59) the Schmidt decomposition can be derived analytically. The formulas can be found in Ref. [GUW01; UBW03] and a detailed derivation can be found in Ref. [Sch17b, App. A]. The Schmidt decomposition is given by

$$
\begin{equation*}
\Phi(x, y)=\sum_{n=0}^{\infty} \sqrt{\lambda_{n}} H_{n}\left(k_{1} x\right) H_{n}\left(k_{2} y\right) \tag{2.61}
\end{equation*}
$$

where $x=\omega_{1}-\bar{\omega}_{1}, y=\omega_{2}-\bar{\omega}_{2}$, for $i=1,2$,

$$
\begin{equation*}
H_{n}\left(k_{i} x\right)=\frac{\sqrt{k_{i}}}{\sqrt{2^{n} n!\sqrt{\pi}}} \mathrm{e}^{-\left(k_{i} x\right)^{2} / 2} \mathrm{~h}_{n}\left(k_{i} x\right) \tag{2.62}
\end{equation*}
$$

are the Hermite-Gauss functions, $\mathrm{h}_{n}(x)$ are the Hermite polynomials and the vari-


Figure 2.1: The joint spectral density functions of an entangled photon pair with different entropies of entanglement. The entropies of entanglement $S$ are (a) $S=$ 0.72 , (b) $S=0.85$, (c) $S=3.3$.
ables that appear are defined as follows:

$$
\begin{array}{ll}
\lambda_{n}=w^{2 n}\left(1-w^{2}\right), & w=\frac{-\sqrt{a c}+\sqrt{a c-b^{2}}}{b}, \\
r_{n}=\sqrt{\lambda_{n}}, & a=\frac{1}{2 \sigma_{p}}+\gamma T_{1}^{2}, \\
k_{1}=\sqrt{\frac{2 a\left(1-w^{2}\right)}{1+w^{2}}}, & b=\frac{1}{2 \sigma_{p}}+\gamma T_{1} T_{2},  \tag{2.63}\\
k_{2}=\sqrt{\frac{2 c\left(1-w^{2}\right)}{1+w^{2}}}, & c=\frac{1}{2 \sigma_{p}}+\gamma T_{2}^{2} .
\end{array}
$$

The difference $T=T_{2}-T_{1}$ defines the entanglement time, the quantities $T_{1}, T_{2}$ are defined as $T_{1}=\left(1 / v_{p}-1 / v_{1}\right) L, T_{2}=\left(1 / v_{p}-1 / v_{2}\right) L$ from Eq. (2.56) and $v_{i}$ is the speed of the $i^{\text {th }}$ beam in the PDC crystal. When $b>0$ the photons exhibit frequency anti-correlations, when $b<0$ positive frequency correlations and for $b=0$ the two photons are uncorrelated. The value of the parameter $w \in[0,1)$ indicates the degree of entanglement of the photon pair. High values of $w$ correspond to high values of entanglement, while for $w \rightarrow 0$ the photon pair is in a product state. We also note that $\sum_{n} \lambda_{n}=1$.

The electromagnetic field operator can be expressed in terms of the Schmidt modes. Its expression can be found by the use of the orthogonality relations of Eq. (2.47)[Sch17b, App A]:

$$
\begin{equation*}
\hat{E}_{i}(t)=\mathcal{A}(\bar{\omega}) \sum_{k=1}^{\infty} \hat{C}_{k}^{(i)} \tilde{H}_{k}^{(i)}(t), \quad i=1,2 \tag{2.64}
\end{equation*}
$$

where $\hat{C}_{k}^{(1)}=\hat{A}_{k}, \hat{C}_{k}^{(2)}=\hat{B}_{k}$ are the two effective annihilation operators for each mode and $\tilde{H}_{k}(t)$ is the Fourier transform of the Hermite-Gauss polynomial $\mathcal{F}\left\{H_{k}(\omega)\right\}$ and

$$
\begin{equation*}
\hat{C}_{k}^{(i)}=\int d \omega H_{k}^{*}\left(k_{i} \omega\right) \hat{a}_{i}(\omega), \tag{2.65}
\end{equation*}
$$

as per Eq. (2.48). We also define the function $u_{n}^{(i)}(t)=\mathcal{F}\left\{H_{n}\left(k_{i}\left(\omega-\bar{\omega}_{i}\right)\right)\right\} \mathrm{e}^{i \bar{\omega}_{i} t}$. Since the Hermite polynomials are eigenfunctions of the Fourier transform we have:

$$
\begin{equation*}
u_{n}^{(i)}(t)=\frac{i^{n}}{\sqrt{2^{n} \sqrt{\pi} n!k_{i}}} \mathrm{e}^{-t^{2} / 2 k_{i}^{2}} \mathrm{~h}_{n}\left(\frac{t}{k_{i}}\right) \tag{2.66}
\end{equation*}
$$

Using the above definitions the two photon entangled state of Eq. (2.58) can be written as [GUW01]

$$
\begin{equation*}
\left|11_{\text {ent }}\right\rangle=\sum_{k=0}^{\infty} r_{k} \hat{A}_{k}^{\dagger} \hat{B}_{k}^{\dagger}|00\rangle . \tag{2.67}
\end{equation*}
$$

## Chapter 3

## Metrology

The main goal of metrology is to determine the information available in a system about a given parameter and devise a measurement scheme in order to obtain an estimate of the parameter as accurate as possible. Using tools from statistical inference one can construct an estimator of the parameter based on observed data. The performance of an estimator is assessed in terms of a cost function, such as the mean square error. Once the appropriate cost function is chosen, one can calculate theoretical bounds on the precision of the estimator.

This chapter is organised as follows: In Sec. 3.1, we discuss about single parameter estimation and the classical and quantum Cramér-Rao bound (CRB) [Kay93]. The classical CRB is a lower bound on the variance of unbiased estimators, where it is assumed that the parameter to be determined has a fixed but unknown value within a known interval. With the use of quantum resources, however, the concepts of classical estimation theory must be generalised and applied in the context of quantum mechanics. In Sec. 3.2 we present the multi-parameter Cramér-Rao bound.

### 3.1 Simgle parameter estimation

Let $\{\mathbf{x}\}=X_{1}, X_{2}, \ldots, X_{n}$ be a set of independent identically distributed measurement outcomes which depend on an unknown parameter $\vartheta$. These outcomes are described by an underlying probability distribution function $p(\mathbf{x} \mid \vartheta)$ conditioned on the value of $\vartheta$. The goal of parameter estimation is to infer the value of the parameter $\vartheta$, i.e. obtain an estimator $\tilde{\vartheta}$, by post-processing the measurement outcomes. The
performance of the estimator can be quantified by choosing the mean square error (MSE)

$$
\begin{equation*}
\operatorname{MSE}(\tilde{\vartheta})=\mathrm{E}\left[(\tilde{\vartheta}-\vartheta)^{2}\right], \tag{3.1}
\end{equation*}
$$

as a cost function. In the above definition, E denotes the expectation value, which indicates the average mean square deviation of the estimator from the true value of the parameter. It is desirable for the estimator to have a minimal MSE. The MSE can be written as

$$
\begin{equation*}
\operatorname{MSE}(\tilde{\vartheta})=\operatorname{var}(\tilde{\vartheta})+\mathrm{b}^{2}(\vartheta) . \tag{3.2}
\end{equation*}
$$

Here, $\operatorname{var}(\tilde{\vartheta})$ is the variance

$$
\begin{equation*}
\operatorname{var}(\tilde{\vartheta})=\mathrm{E}\left[(\tilde{\vartheta}-\mathrm{E}[\tilde{\vartheta}])^{2}\right], \tag{3.3}
\end{equation*}
$$

which indicates the square deviation of the estimates from the expected value of the estimates, while $\mathrm{b}(\vartheta)$ is the bias of the estimator

$$
\begin{equation*}
\mathrm{b}(\vartheta)=\mathrm{E}(\tilde{\vartheta}-\vartheta), \tag{3.4}
\end{equation*}
$$

which is the expected value of the error. In this thesis we focus on unbiased estimators $\mathrm{E}[\tilde{\vartheta}]=\vartheta$, i.e. estimators which on average yield the true value of the unknown parameter for all possible values of the parameter.

### 3.1.1 Classical and quantum Cramér-Rao bound

For any given set of outcomes, a lower bound on the variance for an unbiased estimator can be calculated theoretically and it provides the so called Cramér-Rao bound (CRB) [CT06; Kay93]

$$
\begin{equation*}
\operatorname{var}(\tilde{\vartheta}) \geq \frac{1}{N \mathcal{C}(\vartheta)} \tag{3.5}
\end{equation*}
$$

Here, $N$ is the number of the independent repetitions of the measurements from which we extract the data and $\mathcal{C}(\vartheta)$ is the (classical) Fisher information (CFI) defined as

$$
\begin{equation*}
\mathcal{C}(\vartheta)=\int d x p(x \mid \vartheta)\left(\frac{\partial \ln p(x \mid \vartheta)}{\partial \vartheta}\right)^{2}=\int d x \frac{1}{p(x \mid \vartheta)}\left(\frac{\partial p(x \mid \vartheta)}{\partial \vartheta}\right)^{2}, \tag{3.6}
\end{equation*}
$$

where the regularity condition $\mathrm{E}[\partial \ln p(x \mid \vartheta) / \partial \vartheta]=0$ is assumed.
We are now interested in estimating the parameter $\theta$ on which a quantum
state depends. In this section we follow the derivation of Ref. [Par09; Hel09] and we assume that the quantum state depends only on one parameter $\vartheta$. In quantum mechanics the probability outcome of a measurement is given by Eq. (2.2), that is $p(x \mid \vartheta)=\operatorname{Tr}\left(\rho_{\vartheta} \hat{\Pi}_{x}\right)$, where $\left\{\Pi_{x}\right\}$ is a POVM. The CFI of Eq. (3.6) is written as

$$
\begin{equation*}
\mathcal{C}\left(\rho_{\vartheta}, \Pi_{x}\right)=\int d x \frac{\left(\operatorname{Tr}\left(\partial_{\vartheta} \rho_{\vartheta} \hat{\Pi}_{x}\right)\right)^{2}}{\operatorname{Tr}\left(\rho_{\vartheta} \hat{\Pi}_{x}\right)}=\int d x \frac{\operatorname{Re}\left(\operatorname{Tr}\left(\rho_{\vartheta} \hat{\Pi}_{x} \mathcal{L}_{\vartheta}\right)\right)^{2}}{\operatorname{Tr}\left(\rho_{\vartheta} \hat{\Pi}_{x}\right)}, \tag{3.7}
\end{equation*}
$$

where $\mathcal{L}$ is a hermitian operator, the symmetric logarithmic derivative (SLD) operator defined as

$$
\begin{equation*}
\partial_{\vartheta} \rho_{\vartheta}=\frac{\mathcal{L}_{\vartheta} \rho_{\vartheta}+\rho_{\vartheta} \mathcal{L}_{\vartheta}}{2}, \tag{3.8}
\end{equation*}
$$

that is the SLD operator is a solution of the Lyapunov equation. As it can be seen fron Eq. (3.7) the CFI $\mathcal{C}\left(\rho_{\vartheta}, \Pi_{x}\right)$ depends on the detection procedure, i.e the POVM $\left\{\hat{\Pi}_{x}\right\}$, as well as the quantum state itself. To identify the ultimate bound on the precision of the estimator, the CFI must be maximised over all the possible measurements. The maximisation is bounded as

$$
\begin{equation*}
\max _{\Pi_{x}} \mathcal{C}\left(\rho_{\vartheta}, \Pi_{x}\right) \leq \mathcal{Q}\left(\rho_{\vartheta}\right) \tag{3.9}
\end{equation*}
$$

by the quantum Fisher information (QFI) which is given by

$$
\begin{equation*}
\mathcal{Q}\left(\rho_{\vartheta}\right)=\operatorname{Tr}\left(\rho_{\vartheta} \mathcal{L}_{\vartheta}^{2}\right)=\operatorname{Tr}\left(\partial_{\vartheta} \rho_{\vartheta} \mathcal{L}_{\vartheta}\right) . \tag{3.10}
\end{equation*}
$$

Having defined the above quantities, we can now write the quantum Cramér-Rao bound (QCRB) as

$$
\begin{equation*}
\operatorname{var}(\tilde{\vartheta}) \geq \frac{1}{N \mathcal{C}\left(\rho_{\vartheta}, \Pi_{x}\right)} \geq \frac{1}{N \mathcal{Q}\left(\rho_{\vartheta}\right)} . \tag{3.11}
\end{equation*}
$$

The QCRB provides a fundamental bound that depends only on the quantum state and not on the detection system.

We note that for a single parameter estimation problem, the classical CRB is saturated in the limit $N \rightarrow \infty$ by the maximum likelihood estimator [Kay93]. Also, for the case of a single parameter, there exists an optimal POVM that saturates the inequality (3.9). This POVM $\hat{\Pi}_{\vartheta}$ is the set of projectors over the eigenstates of $\mathcal{L}_{\vartheta}$ [Par09; BC94].

### 3.2 Multiple parameter estimation

All the above definitions can be extended to a vector parameter, that is, to outcomes that depend on a set of parameters $\boldsymbol{\vartheta}=\left(\vartheta_{1} \vartheta_{2} \ldots \vartheta_{m}\right)$. In this case, the estimator is unbiased if $\mathrm{E}\left(\tilde{\vartheta}_{i}\right)=\vartheta_{i} \forall i$, efficient if $\operatorname{var}\left(\tilde{\vartheta}_{i}\right)$ is minimum among all the unbiased estimators for all $i$, and the CRB becomes a bound on the covariance matrix $\operatorname{Cov}(\tilde{\boldsymbol{\vartheta}})$ of the estimators [CT06; Kay93]

$$
\begin{equation*}
\operatorname{Cov}(\tilde{\boldsymbol{\vartheta}}) \geq \frac{1}{N} \mathcal{C}(\boldsymbol{\vartheta})^{-1} \tag{3.12}
\end{equation*}
$$

The elements of the covariance matrix are defined as $\operatorname{Cov}(\tilde{\boldsymbol{\vartheta}})_{i j}=\mathrm{E}\left[\left(\tilde{\vartheta}_{i}-\vartheta_{i}\right)\left(\tilde{\vartheta}_{j}-\vartheta_{j}\right)\right]$ and $\mathcal{C}(\boldsymbol{\vartheta})$ is the CFI matrix with elements

$$
\begin{equation*}
\mathcal{C}(\boldsymbol{\vartheta})_{i j}=\int d x \frac{1}{p(x \mid \vartheta)} \frac{\partial p(x \mid \vartheta)}{\partial \vartheta_{i}} \frac{\partial p(x \mid \vartheta)}{\partial \vartheta_{j}} \tag{3.13}
\end{equation*}
$$

Note that the inequality of Eq. (3.12) is a matrix inequality, which means that the difference $\operatorname{Cov}(\tilde{\boldsymbol{\vartheta}})-(N \mathcal{C}(\boldsymbol{\vartheta}))^{-1}$ is a positive semi-definite matrix. The QFI matrix $\mathcal{Q}\left(\rho_{\vartheta}\right)$ elements are defined as

$$
\begin{equation*}
\mathcal{Q}\left(\rho_{\boldsymbol{\vartheta}}\right)_{i j}=\frac{1}{2} \operatorname{Tr}\left(\rho_{\boldsymbol{\vartheta}}\left(\mathcal{L}_{i} \mathcal{L}_{j}+\mathcal{L}_{j} \mathcal{L}_{i}\right)\right) \tag{3.14}
\end{equation*}
$$

where $\mathcal{L}_{i}$ is the SLD corresponding to the parameter $\vartheta_{i}$ is given by Eq. (3.8). The QCRB can then be generalised for a vector parameter as

$$
\begin{equation*}
\operatorname{Cov}(\tilde{\boldsymbol{\vartheta}}) \geq \frac{1}{N} \mathcal{C}\left(\rho_{\boldsymbol{\vartheta}}, \hat{\Pi}_{x}\right)^{-1} \geq \frac{1}{N} \mathcal{Q}\left(\rho_{\boldsymbol{\vartheta}}\right)^{-1} \tag{3.15}
\end{equation*}
$$

One can obtain a scalar bound from Eq. 3.15 as follows:

$$
\begin{equation*}
\operatorname{Tr}(W \operatorname{Cov}(\tilde{\boldsymbol{\vartheta}})) \geq \frac{1}{N} \operatorname{Tr}\left(W \mathcal{C}\left(\rho_{\boldsymbol{\vartheta}}, \hat{\Pi}_{x}\right)^{-1}\right) \geq \operatorname{Tr}\left(W \frac{1}{N} \mathcal{Q}\left(\rho_{\vartheta}\right)^{-1}\right) \tag{3.16}
\end{equation*}
$$

where $W$ is some positive weight matrix which allows to prioritise the uncertainty of different parameters.

There are cases where it might be convenient to change the parameterisation of the system from a set of (old) parameters $\boldsymbol{\vartheta}$ to a new set of parameters $\boldsymbol{\vartheta}^{\prime}=$ $f(\boldsymbol{\vartheta})$. Given that the Jacobian matrix $B$ of the transformation with elements $B_{i j}=$
$\partial \vartheta_{i} / \partial \vartheta_{j}^{\prime}$ is non-singular, the CFI matrix is transformed according to [Kay93]

$$
\begin{equation*}
\mathcal{C}\left(\boldsymbol{\vartheta}^{\prime}\right)=B \mathcal{C}(\boldsymbol{\vartheta}) B^{\mathrm{T}} . \tag{3.17}
\end{equation*}
$$

The transformation of QFI matrix to a new set of parameters follows the same transformation rule.

Finally, we note that there are cases in which the FI matrix is singular. This implies that there are linear combinations of parameters that cannot be estimated from the data, i.e. the variance of the estimators is infinite. In such cases, the FI matrix can still provide a meaningful bound for the combination of parameters that correspond to the non-zero eigenvalues of the CFI matrix [SM01; Val08].

## Attainability of the QFI

As mentioned in Sec. 3.1.1, in the case of a single parameter estimation the set of projectors over the eigenstates of $\mathcal{L}_{\vartheta}$ saturates the QCRB. However, this strategy does not generalise to multiple parameters in general. In the case where all SLDs commute, the SLDs share a common eigenbasis and, thus, there exists a common measurement optimal for extracting information for all the parameters $\vartheta_{i}$. A necessary and sufficient condition for the saturability of QCRB is the satisfaction of the weak commutativity condition [RJD16]

$$
\begin{equation*}
\operatorname{Tr}\left(\rho_{\vartheta}\left[\mathcal{L}_{i}, \mathcal{L}_{j}\right]\right)=2 \operatorname{Tr}\left(\operatorname{Im}\left\{\rho_{\vartheta} \mathcal{L}_{i} \mathcal{L}_{j}\right\}\right)=0 \tag{3.18}
\end{equation*}
$$

where only the expectation value of the SLD operators needs to be equal to zero.
For pure states, the weak commutativity condition is equivalent to some SLDs commuting, which could make possible to find an optimal measurement as the common eigenbasis of these SLDs. For mixed states, the weak commutativity guaranties the saturability for the scalar bound of Eq. (3.16) through its equivalency to the Holevo bound [RJD16]. However, one might need to perform collective measurements in the the asymptotic limit of many independent copies of the state $\rho_{\vartheta}$ in order to saturate that bound [RJD16].

### 3.2.1 Calculating the SLDs

The general solution to the SLD equation requires the diagonalisation of the density matrix. We assume that the eigenvalues $\left\{\lambda_{n}\right\}$ and eigenvectors $\left\{\left|\psi_{n}\right\rangle\right\}$ of the density operator are known and, hence, write the density operator as $\rho_{\vartheta}=\sum_{n} \lambda_{n}\left|\psi_{n}\right\rangle\left\langle\psi_{n}\right|$. The the SLD operator can be calculated by the formula [Par09]

$$
\begin{equation*}
\mathcal{L}_{i}=2 \sum_{n, m} \frac{\left\langle\psi_{m}\right| \partial_{\vartheta_{i}} \rho_{\vartheta}\left|\psi_{n}\right\rangle}{\lambda_{n}+\lambda_{m}}\left|\psi_{m}\right\rangle\left\langle\psi_{n}\right|, \tag{3.19}
\end{equation*}
$$

where these sums include only the terms for which $\lambda_{n}+\lambda_{m} \neq 0$. We also note that the set of $\left\{\left|\psi_{n}\right\rangle\right\}$ must include the vectors that span the support of the density matrix and its derivative ${ }^{1}$.

There are cases where one might need to manipulate the SLD equation in order to avoid finding the eigendecomposition of the density matrix. One such way is the one described in Ref. [GT19], which is also the method we used to calculate the QFI matrix in the case of localisation microscopy. We pick a convenient, not necessarily orthogonal, basis $\mathcal{B}_{\rho}=\left\{\left|\phi_{n}\right\rangle\right\}$ that spans the support of the density matrix $\rho_{\vartheta}$. We calculate the set's non-zero derivatives $\mathcal{B}_{\rho}^{\prime}=\left\{\partial_{\vartheta_{i}}\left|\phi_{n}\right\rangle\right\}$. From the set $\mathcal{B}_{\rho}^{\prime}$ we keep the vectors $\left\{\partial_{\vartheta_{i}}|\phi\rangle\right\}$ such that the set $\mathcal{B}=\left\{\partial_{\vartheta_{i}}|\phi\rangle\right\} \cup \mathcal{B}_{\rho}$ is a set of linearly independent vectors. The density operator and the SLD operator can be expressed on terms of the vectors $\left\{\left|\phi_{i}\right\rangle\right\}$ as

$$
\begin{align*}
\rho_{\vartheta} & =\sum_{i, j} \varrho_{i j}\left|\phi_{i}\right\rangle\left\langle\phi_{j}\right|,  \tag{3.20}\\
\partial_{\vartheta_{\nu}} \rho_{\vartheta} & =\sum_{i, j} \mathrm{D}_{i j}^{(\nu)}\left|\phi_{i}\right\rangle\left\langle\phi_{j}\right|,  \tag{3.21}\\
\mathcal{L}_{\nu} & =\sum_{i, j} \Lambda_{i j}^{(\nu)}\left|\phi_{i}\right\rangle\left\langle\phi_{j}\right| . \tag{3.22}
\end{align*}
$$

We construct the so-called Grammian matrix $\Upsilon$ whose elements consist of the products

$$
\begin{equation*}
\Upsilon_{i j}=\left\langle\phi_{i} \mid \phi_{j}\right\rangle . \tag{3.23}
\end{equation*}
$$

[^0]The Lyapunov equation (3.15) then takes the form

$$
\begin{equation*}
2 \mathrm{D}^{(\nu)}=\Lambda^{(\nu)} \Upsilon \varrho+\varrho \Upsilon \Lambda^{(\nu)}, \tag{3.24}
\end{equation*}
$$

where $\varrho, \Lambda^{(\nu)}$ and $D^{(\nu)}$ are defined from Eq. (3.20)-(3.22). Eq. (3.24) may be solved using standard linear algebraic methods, such as solving a linear system of equations or by matrix vectorisation as in Ref. [Saf18]. In the case where we choose matrix vectorisation to solve Eq. (3.24), the elements of $\Lambda^{(\nu)}$ are found by the following expression

$$
\begin{equation*}
\operatorname{vec}\left[\Lambda^{(\nu)}\right]=2\left(\varrho \otimes \Upsilon^{-1}+\Upsilon^{-1} \otimes \varrho\right)^{-1}(\Upsilon \otimes \Upsilon) \operatorname{vec}\left[D^{(\nu)}\right] \tag{3.25}
\end{equation*}
$$

A simple example for which the QFI can be calculated analytically is for the case of a pure state $\rho_{\boldsymbol{\vartheta}}=\left|\psi_{\boldsymbol{\vartheta}}\right\rangle\left\langle\psi_{\boldsymbol{\vartheta}}\right|$. Since $\rho_{\boldsymbol{\vartheta}}^{2}=\rho_{\boldsymbol{\vartheta}}$ we have $\partial_{\vartheta_{i}} \rho_{\boldsymbol{\vartheta}}=\left(\partial_{\vartheta_{i}} \rho_{\boldsymbol{\vartheta}}\right) \rho_{\boldsymbol{\vartheta}}+$ $\rho_{\vartheta}\left(\partial_{\vartheta_{i}} \rho_{\vartheta}\right)$ from which we recognise $\mathcal{L}_{i}=2\left(\partial_{\vartheta_{i}} \rho_{\vartheta}\right)$. The derivative of the state is $\partial_{\vartheta_{i}} \rho_{\vartheta}=\left|\partial_{\vartheta_{i}} \psi_{\vartheta}\right\rangle\left\langle\psi_{\boldsymbol{\vartheta}}\right|+\left|\psi_{\boldsymbol{\vartheta}}\right\rangle\left\langle\partial_{\vartheta_{i}} \psi_{\boldsymbol{\vartheta}}\right|$ which leads to the following expression for the QFI matrix elements [Par09]

$$
\begin{equation*}
\mathcal{Q}_{i j}=4 \operatorname{Re}\left\{\left\langle\partial_{\vartheta_{i}} \psi_{\boldsymbol{\vartheta}} \mid \partial_{\vartheta_{j}} \psi_{\boldsymbol{\vartheta}}\right\rangle+\left\langle\psi_{\boldsymbol{\vartheta}} \mid \partial_{\vartheta_{i}} \psi_{\boldsymbol{\vartheta}}\right\rangle\left\langle\psi_{\boldsymbol{\vartheta}} \mid \partial_{\vartheta_{j}} \psi_{\boldsymbol{\vartheta}}\right\rangle\right\} \tag{3.26}
\end{equation*}
$$

An alternative formula for the pure state QFI is [FN95]

$$
\begin{equation*}
\mathcal{Q}_{i j}=\operatorname{Tr}\left(\left(\partial_{\vartheta_{i}} \rho_{\vartheta}\right)\left(\partial_{\vartheta_{j}} \rho_{\vartheta}\right)\right), \tag{3.27}
\end{equation*}
$$

where the density matrix is given by $\rho=\left|\psi_{\boldsymbol{\vartheta}}\right\rangle\left\langle\psi_{\boldsymbol{\vartheta}}\right|$.

### 3.2.2 Estimation of Hamiltonian parameters and QFI in the interaction picture

In this section, we examine whether the expression for QFI is the same in the Schrödinger and interaction picture. We follow the formalism of Sec. 2.1.2 and we assume that the Hamiltonian in the Schrodinger picture depends on two parameters, $\vartheta$ in the free evolution Hamiltonian $\hat{\mathcal{H}}_{0}(\vartheta)$ and $\varphi$ in the interaction Hamiltonian $H_{\text {int }}^{(S)}(\varphi)$. We assume that our state $\rho(\vartheta, \varphi)$ is pure for which the QFIM elements are given by Eq. (3.27) with $i, j=\vartheta, \varphi$. For convenience, we examine just the diagonal
elements. In the Schrodinger picture the diagonal QFI elements are given by

$$
\begin{equation*}
Q_{i i}=\operatorname{Tr}\left[\left(\partial_{i} \rho^{(S)}\right)\left(\partial_{i} \rho^{(S)}\right)\right] . \tag{3.28}
\end{equation*}
$$

The transformation of the density matrix to the interaction picture is given by Eq. (2.12) and, thus, the derivative of the state in the interaction picture is

$$
\begin{equation*}
\partial_{i} \rho_{S}=\partial_{i}\left(U_{o} \rho_{I} U_{o}^{\dagger}\right)=\partial_{i}\left(U_{o}\right) \rho_{I} U_{o}^{\dagger}+U_{o} \rho_{I} \partial_{i}\left(U_{o}^{\dagger}\right)+U_{o} \partial_{i} \rho_{I} U_{o}^{\dagger} . \tag{3.29}
\end{equation*}
$$

In the case where we are interested in the parameter $\varphi$ of the interaction Hamiltonian, the derivatives in the two pictures are connected by the transformation $\partial_{\varphi} \rho_{S}=$ $U_{o} \partial_{\varphi} \rho_{I} U_{o}^{\dagger}$ and the QFIs in the two pictures are equivalent. However, if we are interested in the parameter $\vartheta$ in the free evolution operator, then the derivatives in the two pictures are given by

$$
\begin{equation*}
\partial_{\vartheta} \rho_{S}=\partial_{\vartheta}\left(U_{o} \rho_{I} U_{o}^{\dagger}\right)=U_{o} \partial_{\vartheta} \rho_{I} U_{o}^{\dagger}-i t U_{o}\left[\mathcal{H}_{0}(\vartheta), \rho_{I}\right] U_{o}^{\dagger} \tag{3.30}
\end{equation*}
$$

Therefore, the two QFIs are different as $\operatorname{Tr}\left[\left(\partial_{i} \rho_{S}\right)\left(\partial_{i} \rho_{S}\right)\right] \neq \operatorname{Tr}\left[\left(\partial_{i} \rho_{I}\right)\left(\partial_{i} \rho_{I}\right)\right]$. In this case, the QFI must be calculated in the Schrödinger picture [SRP17]. In the derivation of the QFI there is the implicit assumption in Eq. (3.7) that

$$
\begin{equation*}
\partial_{\vartheta} p(x \mid \vartheta)=\partial_{\vartheta} \operatorname{Tr}\left[\rho_{\vartheta} \Pi_{x}\right]=\operatorname{Tr}\left[\partial_{\vartheta} \rho_{\vartheta} \Pi_{x}\right] . \tag{3.31}
\end{equation*}
$$

However, in the interaction picture the POVM operator is transformed according to Eq. (2.12) as $\Pi_{x}(\vartheta)=U_{o}(\vartheta) \Pi_{x} U_{o}(\vartheta)^{\dagger}$, that is the POVM is parameter-dependent. For such cases of non-regular measurements it is already known that a different approach to the one we have presented in this chapter must be adopted in order to obtain the correct lower bound to the variance of an estimator [SP20; SRP17]. In this thesis we are not interested in parameters that appear in the free Hamiltonian.

## Chapter 4

## Quantum limits to localisation microscopy

### 4.1 Introduction

Precisely locating multiple single emitters is a key challenge in fluorescence microscopy. The process of estimating these locations depends on the quality of the image obtained by the microscope. One of the major limitations to the image quality, known since Abbe and Rayleigh, lies in spatially resolving objects substantially smaller than half the wavelength of the light involved [BW99]. Known as the Rayleigh limit or diffraction limit, it is a consequence of the diffraction of light due to its wave nature.

Over the last couple of decades, ways to circumvent the Rayleigh limit in far-field fluorescence microscopy have been invented [TPR14]. Confocal methods such as STED, RESOLFT, and SSIM [HW94; HJC02; Gus05; Hof+05] use patterned illumination to spatially modulate the fluorescence pattern of emitters within a diffraction-limited region such that not all of them emit simultaneously, thereby achieving sub-Rayleigh resolution. Other far-field methods such as PALM, fPALM and STORM [RBZ06; Bet+06; HGM06] temporally modulate the fluorescence pattern of emitters with weak laser pulses stochastically such that only a low density of emitters are active within the Rayleigh limit at one time. Repeating the process many times, images with sub-Rayleigh resolution are reconstructed from the measured positions of individual emitters. These techniques, with resolution of tens of


Figure 4.1: Illustration of localisation microscopy with five point sources, imaged by a diffraction-limited system and the resultant intensity distribution on the image plane.
nanometers, have provided insights into biological processes at the cellular scale that were hitherto unattainable [HBZ10].

Though immensely powerful and impressive, none of these methods seek to extract all the information available in the emitted light field. As in conventional fluorescence microscopy these techniques use 'direct imaging' - intensity measurements on the image plane - to extract information from the incident light. That there is indeed more information in the light field to be extracted was shown by [TNL16]. Using methods from classical and quantum estimation theory, it was shown theoretically that two arbitrarily close incoherent point sources may be resolved, and that this may be achieved in practice using a spatial-mode demultiplexing (SPADE) measurement. In the few years since, theoretical studies have considered different source arrangements or parameters of interest [NT16a; LP16; KGA17; Chr +17 ; Řeh+17; Dut+19] in one as well as in two and three spatial dimensions [ANT17; YP18; Nap+19; BSW18]. Other theoretical studies have explored various detection systems that could achieve the ultimate precision in imaging or get close to it [NT16b; Yan +17 ; Reh +17 ; Řeh +18 ]. Several experiments have demonstrated some of the principles underlying these detection systems [TDL16; Paú +16 ; Yan +16 ; Don +18 ; Paú +18 ; Par +18 ; Zho +19 ; Bon +19 ]. Advances in this area have been recently reviewed by [Tsa19b].

Realistic imaging scenarios typically involve more than two point sources or even extended objects. It has been shown that an extended one-dimensional object much smaller than the Rayleigh limit described only in terms of its centroid and effective radius can be approximated by a two-level quantum system [Chr+17]. Theoretical optimality of certain measurement techniques in estimating this effective radius size has also been established in one and two spatial dimensions [Tsa17; Tsa18; Dut+19]. Order-of-magnitude bounds on the precision of estimating the normalised moments of extended sources smaller the Rayleigh limit have also been obtained [ZJ19; Tsa19a].

In this chapter, we provide an analytical lower bound on an unbiased estimator's covariance (mean square error) matrix for localisation microscopy - simultaneously estimating the locations of $N$ incoherent, weak point sources of unequal but known intensities in one spatial dimension. The bound is provided by the quantum Fisher information matrix. For a Gaussian point spread function (PSF), we first describe the light field on the image plane as a classical mixture of coherent states. We use this to derive the quantum Fisher information matrix analytically. In the limit of the point sources approaching a single point, we find its rank to be no more than two. As the inverse of the quantum Fisher information matrix lower bounds the covariance matrix, our result implies that no more than two independent parameters can be estimated in localisation microscopy in the limit of arbitrarily small separations. In this limit, we provide a mathematical explanation for our observation in terms of an approximation of the light field involving only the first two HermiteGauss modes. Finally, we compare performance of conventional direct imaging and the recently proposed SPADE [TNL16] in localisation microscopy with the quantum bounds we obtain. In the limit of the point sources approaching a single point, we find the classical Fisher information matrices for both these detection systems to be rank one. Furthermore, in the sub-Rayleigh limit, SPADE does not attain the quantum limit for localisation microscopy. For the subset of parameters where scalings may be optimal, we find SPADE to be short of the quantum limit in absolute precision.

This chapter is organised as follows: In Section 4.2 we provide a quantum mechanical description of localisation microscopy. In Section 5.4 we provide an analytic expression of the QFIM for localisation microscopy, our main technical result. We then draw conclusions about its rank and its implications for localisation
microscopy. We end in Section 4.4 with further insights and discussions about the sinc PSF and the potential of detection systems attaining the quantum limits of localisation microscopy.

### 4.2 Quantum description of localisation microscopy

We consider localisation microscopy - the problem of estimating the locations of $N$ incoherent point sources or emitters located in a one-dimensional spatial configuration as in Fig. 4.1. As we assume them to be weak, such that on average no photons arrive on the image place within a coherence time with probability $(1-\epsilon)$, where $\epsilon \ll 1$ and one photon arrives with probability $\epsilon$. We also assume the optical field on the image plane to be quasi-monochromatic and paraxial [TNL16]. The quantum state of this optical field is then

$$
\begin{equation*}
\rho_{\mathrm{opt}} \approx(1-\epsilon) \rho_{\mathrm{vac}}+\epsilon \rho, \tag{4.1}
\end{equation*}
$$

where we have neglected terms of second and higher orders in $\epsilon$ and $\rho_{\mathrm{vac}}=|\mathrm{vac}\rangle\langle\mathrm{vac}|$ is the vacuum state and $\rho$ is the one-photon state.

The one-photon density matrix on the object plane is an incoherent mixture of position eigenstates $\rho=\sum_{i=1}^{N} w_{i}\left|\chi_{i}\right\rangle\left\langle\chi_{i}\right|$, where $w_{i}$ are the relative intensities with $\sum_{i=1}^{N} w_{i}=1$. An imaging system maps $\hat{c}_{x}^{\dagger}$, the creation operator producing one photon in the position $x$ on the object plane, to the corresponding image plane operator $\hat{c}_{i}^{\dagger}$ [LP16]

$$
\begin{equation*}
\hat{c}_{i}^{\dagger}=\int d x \Psi_{\mathrm{PSF}}\left(x-\chi_{i}\right) \hat{c}_{x}^{\dagger} \tag{4.2}
\end{equation*}
$$

where $\chi_{i}$ is the position on the source on the object plane and $\psi_{\mathrm{PSF}}(x)$ is the PSF. On the image plane this becomes

$$
\begin{equation*}
\rho=\sum_{i=1}^{N} w_{i}\left|\psi_{i}\right\rangle\left\langle\psi_{i}\right| \tag{4.3}
\end{equation*}
$$

where

$$
\begin{equation*}
\left|\psi_{i}\right\rangle=\int d x \Psi_{\mathrm{PSF}}\left(x-\chi_{i}\right)|x\rangle \tag{4.4}
\end{equation*}
$$

as follows from Eq. (4.2).

An ideal imaging system with $\Psi_{\text {PSF }}(x)=\delta(x)$ is free of any Rayleigh limit as it transmits all spatial frequencies from the object to the image plane. In practice, a Gaussian PSF

$$
\begin{equation*}
\psi_{\mathrm{PSF}}(x)=\frac{1}{\left(2 \pi \sigma^{2}\right)^{1 / 4}} e^{-\frac{x^{2}}{4 \sigma^{2}}}, \tag{4.5}
\end{equation*}
$$

with $\sigma=\lambda /(2 \pi \mathrm{NA})$, where NA is the numerical aperture of the imaging system is a good approximation for quasimonochromatic paraxial light [ZZO06; TNL16] and also allows us to obtain analytical results. For such a PSF, the state of Eq. (4.3) has an intensity distribution of the form illustrated in Fig. 4.1. For a Gaussian PSF, the $\left|\psi_{i}\right\rangle$ can be expanded in the Hermite-Gauss (HG) basis as (See Appendix 4.5.1)

$$
\begin{equation*}
\left|\psi_{i}\right\rangle=\sum_{k=0}^{\infty} \frac{\alpha_{i}^{k}}{\sqrt{k!}} e^{-\alpha_{i}^{2} / 2}\left|\phi_{k}\right\rangle \equiv\left|\alpha_{i}\right\rangle, \tag{4.6}
\end{equation*}
$$

where $\left|\phi_{k}\right\rangle$ are the HG modes ${ }^{1}$ This has the same mathematical form as the coherent states, produced by the displacement operator $\mathcal{D}\left(\alpha_{i}\right)=e^{\alpha_{i} \hat{a}^{\dagger}-\alpha_{i}^{*} \hat{a}}$ [KL10] acting on the ground state of the harmonic oscillator with $\alpha_{i}=\chi_{i} / 2 \sigma \in \mathbb{R}$ the dimensionless positions of the sources. Thus the one-photon state on the image plane is

$$
\begin{equation*}
\rho \equiv \rho_{\boldsymbol{\alpha}}=\sum_{i=1}^{N} w_{i}\left|\alpha_{i}\right\rangle\left\langle\alpha_{i}\right|, \tag{4.7}
\end{equation*}
$$

a classical mixture of coherent states in the HG basis.
The above is a quantum optical rendition of localisation microscopy-a classical optics problem. It enables us to harness the mathematical formalism associated with coherent states and provides a basis that spans the space of the quantum state as well as its derivative. The latter is an essential ingredient of deriving the quantum Fisher information matrix analytically in Section 4.3.1. We also hope that this description will provide insights into the quantum limits to localisation microscopy in the presence of shot noise and assist in designing detection systems that attain these quantum limits.

[^1]
### 4.2.1 Previous works on two point sources



Figure 4.2: Illustration of localisation microscopy with five point sources, imaged by a diffraction-limited system and the resultant intensity distribution on the image plane

In this subsection, we discuss the Rayleigh limit and the previous work on the quantum limits on super-resolution of two point sources by Tsang et.al. [TNL16]. The system of two point sources with positions $\chi_{1}, \chi_{2}$ is depicted in Fig. 4.2 and is described by Eq. (4.1)-(4.5) with a total number of sources $N=2$. The density matrix for two equally bright sources is (Eq. (4.1)):

$$
\begin{equation*}
\rho=\frac{1}{2}\left(\left|\psi_{1}\right\rangle\left\langle\psi_{1}\right|+\left|\psi_{2}\right\rangle\left\langle\psi_{2}\right|\right) . \tag{4.8}
\end{equation*}
$$

The above system can also be described in terms of the centroid $x_{c}=\left(\chi_{1}+\chi_{2}\right) / 2$ and the separation $d=\chi_{1}-\chi_{2}$ between the two points.

The direct imaging measurement is a photon-counting measurement at a specific location $z$ on the image plane. Thus, the POVM of direct imaging is $\Pi_{z}=$ $|n\rangle\left\langle\left. n\right|_{z}, n=0,1,2, .\right.$. in each pixel $z$ of the image plane. In our case, we have assumed that the point sources emit one photon at most. We also assume that the data is
collected over $\mu$ coherence time intervals, leading to the average photon number detected at the image plane $M=\epsilon \mu$, where $\epsilon$ is the probability of a single photon detection of Eq. (4.1). With the above assumptions, the direct imaging measurement is given by the operator $\Pi_{z}=|1\rangle\left\langle\left. 1\right|_{z}\right.$, which results in the probability distribution

$$
\begin{equation*}
P\left(z \mid s_{12}\right)=\frac{1}{2}\left(\left|\left\langle z \mid \psi_{1}\right\rangle\right|^{2}+\left|\left\langle z \mid \psi_{2}\right\rangle\right|^{2}\right)=\frac{1}{2}\left(\left|\psi_{1}\right|^{2}+\left|\psi_{2}\right|^{2}\right) \tag{4.9}
\end{equation*}
$$

Using Eq. (3.6) we calculate the CFIM for the original locations $\chi_{1}, \chi_{2}$. The Rayleigh's limit regards the separations and, hence, we transform the CFIM according to Eq. (3.17) for the parameters $x_{c}$ and $d$. We find that the CFI for the separation $d$ for the direct measurement is [Bet+99]:

$$
\begin{equation*}
\mathcal{C}_{d} \approx \frac{d}{\sigma} \tag{4.10}
\end{equation*}
$$

where $\sigma$ characterises the PSF of the diffraction limited system as in Eq. (4.40). It is obvious from Eq. (4.10) that the CFI for the source's separations tends to 0 as their separation tends to 0 . This means that then mean-square error for the estimator of the separation diverges for direct imaging (see Fig. 4.3).

However, as proven by Tsang et.al. [TNL16], this divergent behaviour is not observed when we calculate the QFI. (For a full derivation of the SLD operators and the QFIM for the centroid and separation of the two point sources see [TNL16]). Tsang et al. proved that the QFI for the separation does not depend on its value, but it is a constant quantity depending only on the width of the Gaussian PSF $\sigma$, i.e. the properties of the imaging system:

$$
\begin{equation*}
\mathcal{Q}_{d} \approx \frac{M}{4 \sigma^{2}} \tag{4.11}
\end{equation*}
$$

where $M$ is the average photon number detected at the image plane. Therefore, the Rayleigh's 'curse' arises as a result of a specific measurement, namely direct imaging(see Fig. 4.3).


Figure 4.3: Mean-square error for separation

Additionally, Tsang et al. constructed the spatial-mode demultiplexing (SPADE) measurement that saturates the separation QFI. In SPADE we decompose the light on the image plane in terms of the Hermite-Gaussian modes $\left\{\phi_{k}\right\}$ and then we count photons in each mode, i.e. the SPADE POVM consists of the projectors $\Pi_{q}=\left|\phi_{q}\right\rangle\left\langle\phi_{q}\right|$.

### 4.3 Results

Localisation has long been treated as an estimation problem with the unknown locations of the sources $\chi \equiv\left\{\chi_{i}\right\}, i=1, \ldots, N$ being the parameters to be estimated [ORW04; CWO16]. In our formulation, the limits to the localisation of the point sources are the same as estimating the amplitudes $\boldsymbol{\alpha} \equiv\left\{\alpha_{i}\right\}, i=1, \ldots, N$ of the coherent states in Eq. (4.7). Let these estimates be $\tilde{\boldsymbol{\alpha}} \equiv\left\{\tilde{\alpha}_{i}\right\}$. We now present our main result - the analytical expression of the QFIM for localisation microscopy. Any scalar function of the covariances can be bounded by the inverse of QFIM with the lower bound following from the spectral decomposition of QFIM. To that end, calculating the eigenvalues of the QFIM and their scaling is of importance for the
multi-parameter estimation. We conclude that the QFIM is a rank two matrix as $\alpha_{i} \rightarrow 0$. Eq. (3.15) then implies that the eigenvalues of $\operatorname{Cov}[\boldsymbol{\alpha}]$ remains finite for no more than two independent parameters. Thus, no more than two independent parameters can be estimated from the entire set $\boldsymbol{\alpha}$ as $\boldsymbol{\alpha} \rightarrow 0$.

We lack a fully satisfactory physical explanation for this restriction on the number of estimable parameters, but provide an explanation involving only the first two Hermite-Gauss modes for $\alpha_{i} \ll 1$.

### 4.3.1 Analytical expression of QFIM

The state in Eq. (4.7) can be expressed in the basis of $\left\{\left|\alpha_{i}\right\rangle, \hat{a}^{\dagger}\left|\alpha_{i}\right\rangle\right\}$ as

$$
\rho_{\boldsymbol{\alpha}}=A\left(\begin{array}{cc}
D_{\boldsymbol{w}} & 0  \tag{4.12}\\
0 & 0
\end{array}\right) A^{\dagger} \equiv A \rho_{A} A^{\dagger}
$$

where

$$
A=\left(\begin{array}{lllllll}
\left|\alpha_{1}\right\rangle & \left|\alpha_{2}\right\rangle & \cdots & \left|\alpha_{N}\right\rangle & \hat{a}^{\dagger}\left|\alpha_{1}\right\rangle & \cdots & \hat{a}^{\dagger}\left|\alpha_{N}\right\rangle \tag{4.13}
\end{array}\right)
$$

and $D_{\boldsymbol{w}}=\operatorname{diag}\left(w_{1}, w_{2}, \cdots, w_{N}\right)$ denotes a diagonal matrix. Although the basis used in Eq. (4.12) is non-orthogonal this representation can still be used to evaluate the QFIM [GT19]. The coherent states $\left\{\left|\alpha_{i}\right\rangle\right\}$ are linearly independent and span the support of the state in Eq. (4.7). The support of the derivative is spanned by $\left\{\left|\alpha_{i}\right\rangle\right\}$ and $\left\{\hat{a}^{\dagger}\left|\alpha_{i}\right\rangle\right\}$, which are also linearly independent.

The Grammian matrix

$$
\begin{equation*}
\Upsilon=A^{\dagger} A, \tag{4.14}
\end{equation*}
$$

whose elements consist of the scalar products between the basis vectors $\left\langle\alpha_{j} \mid \alpha_{k}\right\rangle$, $\left\langle\alpha_{j}\right| \hat{a}^{\dagger}\left|\alpha_{k}\right\rangle,\left\langle\alpha_{j}\right| \hat{a}\left|\alpha_{k}\right\rangle$, and $\left\langle\alpha_{j}\right| \hat{a} \hat{a}^{\dagger}\left|\alpha_{k}\right\rangle$ is in block form,

$$
\Upsilon=\left(\begin{array}{ll}
\Upsilon_{\alpha \alpha} & \Upsilon_{\alpha d}  \tag{4.15}\\
\Upsilon_{d \alpha} & \Upsilon_{d d}
\end{array}\right)
$$

where

$$
\begin{align*}
\left(\Upsilon_{\alpha \alpha}\right)_{i j} & =\left\langle\alpha_{i} \mid \alpha_{j}\right\rangle=e^{-\left(\alpha_{i}-\alpha_{j}\right)^{2} / 2} \\
\left(\Upsilon_{\alpha d}\right)_{i j}=\Upsilon_{d \alpha}^{\dagger} & =\left\langle\alpha_{i}\right| \hat{a}^{\dagger}\left|\alpha_{j}\right\rangle=\alpha_{i} e^{-\left(\alpha_{i}-\alpha_{j}\right)^{2} / 2}=D_{\boldsymbol{\alpha}} \Upsilon_{\alpha \alpha}  \tag{4.16}\\
\left(\Upsilon_{d d}\right)_{i j} & =\left\langle\alpha_{i}\right| \hat{a} \hat{a}^{\dagger}\left|\alpha_{j}\right\rangle=\left(\alpha_{i} \alpha_{j}+1\right) e^{-\left(\alpha_{i}-\alpha_{j}\right)^{2} / 2} \\
& =D_{\boldsymbol{\alpha}} \Upsilon_{\alpha \alpha} D_{\boldsymbol{\alpha}}+\Upsilon_{\alpha \alpha}
\end{align*}
$$

and $D_{\boldsymbol{\alpha}}=\operatorname{diag}\left(\alpha_{1}, \alpha_{2}, \cdots, \alpha_{n}\right)$.
Since $\partial_{\alpha}|\alpha\rangle=\left(\hat{a}^{\dagger}-\alpha\right)|\alpha\rangle$ for real $\alpha$, the derivative of the quantum state is

$$
\partial_{j} \rho_{\boldsymbol{\alpha}}=A w_{j}\left(\begin{array}{cc}
-2 \alpha_{j} E_{j} & E_{j}  \tag{4.17}\\
E_{j} & 0
\end{array}\right) A^{T} \equiv A\left(\partial_{j} \rho\right)_{A} A^{\dagger}
$$

where $\partial_{j}$ denotes the derivative with respect to $\alpha_{j}$ and $\left(E_{j}\right)_{k l}=\delta_{j k} \delta_{j l}$. Similarly, the SLD $L_{A}^{i}$ can be written in the generic form

$$
L^{j}=A L_{A}^{j} A^{T}=A\left(\begin{array}{cc}
L_{\alpha \alpha}^{j} & L_{\alpha d}^{j}  \tag{4.18}\\
L_{d \alpha}^{j} & L_{d d}^{j}
\end{array}\right) A^{T}
$$

where $L_{\alpha \alpha}^{j}$ corresponds to the elements $\left\langle\alpha_{i}\right| L^{j}\left|\alpha_{j}\right\rangle, L_{\alpha d}^{j}$ to $\left\langle\alpha_{j}\right| L^{j} \hat{a}^{\dagger}\left|\alpha_{i}\right\rangle$ etc. The Lyapunov equation Eq. (3.8) can be now rewritten as

$$
\begin{equation*}
2 \partial_{j} \rho_{A}=\rho_{A} \Upsilon L_{A}^{j}+L_{A}^{j} \Upsilon \rho_{A} \tag{4.19}
\end{equation*}
$$

and the QFIM elements from Eq. (3.14) as

$$
\begin{equation*}
\left[\mathcal{Q}\left(\rho_{\boldsymbol{\alpha}}\right)\right]_{j k}=\operatorname{Tr} \partial_{j} \rho L^{k}=\operatorname{Tr} \partial_{j} \rho_{A} \Upsilon L_{A}^{k} \Upsilon \tag{4.20}
\end{equation*}
$$

Using the Tracy-Singh block kronecker product $\odot$ and the block column vectorisation "vecb" operator [KNT91] defined as

$$
\operatorname{vecb}\left(L_{A}^{j}\right)=\left[\begin{array}{c}
\left.\mid L_{\alpha \alpha}^{j}\right)  \tag{4.21}\\
\left.\mid L_{d \alpha}^{j}\right) \\
\left.\mid L_{\alpha d}^{j}\right) \\
\left.\mid L_{d d}^{j}\right)
\end{array}\right],
$$

where $\mid X)=\operatorname{vec}(X)$ is the column vectorisation of a matrix and $(X \mid$ its transpose.

Eq. (4.19) can be blockwise vectorised to

$$
\begin{equation*}
2 \operatorname{vecb}\left(\partial_{j} \rho_{A}\right)=\left(\mathbb{I} \odot\left(\rho_{A} \Upsilon\right)+\left(\rho_{A} \Upsilon\right) \odot \mathbb{I}\right) \operatorname{vecb}\left(L_{A}^{j}\right) \tag{4.22}
\end{equation*}
$$

with $\mathbb{I}$ being the identity matrix. Using the matrix identity [KNT91]

$$
\begin{equation*}
\operatorname{Tr} A^{T} B C D^{T}=\operatorname{vecb}\left(A^{T}\right)^{T}(D \odot B) \operatorname{vecb}(C) \tag{4.23}
\end{equation*}
$$

the QFIM elements from Eq. (4.20) can be re-expressed as

$$
\begin{align*}
{\left[\mathcal{Q}\left(\rho_{\boldsymbol{\alpha}}\right)\right]_{i j} } & =\operatorname{vecb}\left(\partial_{i} \rho_{A}\right)^{T}(\Upsilon \odot \Upsilon) \operatorname{vecb}\left(L_{A}^{j}\right) \\
& =w_{i}\left[\begin{array}{lll}
-2 \alpha_{i}\left(E_{i} \mid\right. & \left(E_{i} \mid\right. & \left(E_{i} \mid\right. \\
0
\end{array}\right]\left[\begin{array}{l}
\left.\mid \Gamma_{\alpha \alpha}^{j}\right) \\
\left.\mid \Gamma_{d \alpha}^{j}\right) \\
\left.\mid \Gamma_{\alpha d}^{j}\right) \\
\left.\mid \Gamma_{d d}^{j}\right)
\end{array}\right], \tag{4.24}
\end{align*}
$$

where we have defined

$$
(\Upsilon \odot \Upsilon) \operatorname{vecb}\left(L_{A}^{j}\right)=\operatorname{vecb}\left(\Gamma^{j}\right)=\left[\begin{array}{l}
\left.\mid \Gamma_{\alpha \alpha}^{j}\right)  \tag{4.25}\\
\left.\mid \Gamma_{d \alpha}^{j}\right) \\
\left.\mid \Gamma_{\alpha d}^{j}\right) \\
\left.\mid \Gamma_{d d}^{j}\right),
\end{array}\right]
$$

which is the outstanding quantity to be determined.
We now recast Eq.(4.22) and (4.24) as

$$
\begin{align*}
2 \operatorname{vecb}\left(\partial_{i} \rho_{A}\right) & =\left(\Upsilon^{-1} \odot \rho_{A}+\rho_{A} \odot \Upsilon^{-1}\right)(\Upsilon \odot \Upsilon) \operatorname{vecb}\left(L_{A}^{i}\right) \\
& =\left(\Upsilon^{-1} \odot \rho_{A}+\rho_{A} \odot \Upsilon^{-1}\right) \operatorname{vecb}\left(\Gamma^{i}\right) \tag{4.26}
\end{align*}
$$

Putting it all together, we obtain

$$
\left[\begin{array}{c}
\left.-4 w_{i} \alpha_{i} \mid E_{i}\right)  \tag{4.27}\\
\left.2 w_{i} \mid E_{i}\right) \\
\left.2 w_{i} \mid E_{i}\right) \\
0
\end{array}\right]=\left[\begin{array}{llll} 
& & & 0 \\
& \mathbb{A} & & 0 \\
& & & 0 \\
0 & 0 & 0 & 0
\end{array}\right]\left[\begin{array}{c}
\left.\mid \Gamma_{\alpha \alpha}^{j}\right) \\
\left.\mid \Gamma_{d \alpha}^{j}\right) \\
\left.\mid \Gamma_{\alpha d}^{j}\right) \\
\left.\mid \Gamma_{d d}^{j}\right)
\end{array}\right]
$$

where

$$
\mathbb{A}=\left[\begin{array}{ccc}
D_{\boldsymbol{w}} \otimes v_{\alpha \alpha}+v_{\alpha \alpha} \otimes D_{\boldsymbol{w}} & D_{\boldsymbol{w}} \otimes v_{\alpha d} & v_{\alpha d} \otimes D_{\boldsymbol{w}}  \tag{4.28}\\
D_{\boldsymbol{w}} \otimes v_{d \alpha} & D_{\boldsymbol{w}} \otimes v_{d d} & 0 \\
v_{d \alpha} \otimes D_{\boldsymbol{w}} & 0 & v_{d d} \otimes D_{\boldsymbol{w}}
\end{array}\right]
$$

and $\left\{v_{\alpha \alpha}, v_{\alpha d}, v_{d \alpha}, v_{d d}\right\}$ defines the inverse of $\Upsilon$ via

$$
\Upsilon^{-1}=\left[\begin{array}{ll}
v_{\alpha \alpha} & v_{\alpha d}  \tag{4.29}\\
v_{d \alpha} & v_{d d}
\end{array}\right]
$$

Note that the inverse $\Upsilon^{-1}$ always exists since $\Upsilon$ is the Grammian matrix of linearly independent vectors. The elements of $\Upsilon^{-1}$ can be found using the formula of blockwise inversion (See Appendix 4.5.2).

Noticing that $\left.\mid \Gamma_{d d}^{j}\right)$ does not contribute in Eq. (4.24), Eq. (4.27) can be reduced to

$$
\left[\begin{array}{c}
\left.-4 w_{i} \alpha_{i} \mid E_{i}\right)  \tag{4.30}\\
\left.2 w_{i} \mid E_{i}\right) \\
\left.2 w_{i} \mid E_{i}\right)
\end{array}\right]=\mathbb{A}\left[\begin{array}{c}
\left.\mid \Gamma_{\alpha \alpha}^{j}\right) \\
\left.\mid \Gamma_{d \alpha}^{j}\right) \\
\left.\mid \Gamma_{\alpha d}^{j}\right)
\end{array}\right]
$$

where $\mathbb{A}$ is a $3 N^{2} \times 3 N^{2}$ invertible matrix unless $\alpha_{i}=\alpha_{j}$ for some $i, j$, which is a singular case for which the rank of the density matrix reduces. Hence the unique solution to Eq. (4.30) is

$$
\left[\begin{array}{l}
\left.\mid \Gamma_{\alpha \alpha}^{j}\right)  \tag{4.31}\\
\left.\mid \Gamma_{d \alpha}^{j}\right) \\
\left.\mid \Gamma_{\alpha d}^{j}\right)
\end{array}\right]=\mathbb{A}^{-1}\left[\begin{array}{c}
\left.-4 w_{j} \alpha_{j} \mid E_{j}\right) \\
\left.2 w_{j} \mid E_{j}\right) \\
\left.2 w_{j} \mid E_{j}\right)
\end{array}\right]
$$

where the block matrices that compose the $\mathbb{A}^{-1}$ can be found by using the formulas for blockwise inversion (See Appendix 4.5.2).

Substituting Eq. (4.31) into Eq. (4.24) gives us the QFIM elements

$$
\begin{align*}
{\left[\mathcal{Q}\left(\rho_{\boldsymbol{\alpha}}\right)\right]_{i j}=} & 2 w_{i} w_{j}\left(E_{i} \mid\left[\mathbb{I} \otimes \Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1}+\Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1} \otimes \mathbb{I}-2 \alpha_{i} \mathbb{I} \otimes \mathbb{I}\right] S^{-1}\right. \\
& {\left.\left[\mathbb{I} \otimes \Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d}+\Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d} \otimes \mathbb{I}-2 \alpha_{j} \mathbb{I} \otimes \mathbb{I}\right] \mid E_{j}\right) }  \tag{4.32}\\
& +4 w_{i} \delta_{i j}\left[1+\alpha_{i}^{2}-\left(\Upsilon_{\alpha \alpha} D_{\alpha} \Upsilon_{\alpha \alpha}^{-1} D_{\alpha} \Upsilon_{\alpha \alpha}\right)_{i j}\right]
\end{align*}
$$

where $S^{-1}=\left(\Upsilon_{\alpha \alpha}^{-1} \otimes D_{\boldsymbol{w}}+D_{\boldsymbol{w}} \otimes \Upsilon_{\alpha \alpha}^{-1}\right)^{-1}$ is an $N^{2} \times N^{2}$ matrix and $\Upsilon_{\alpha \alpha}^{1}$ is the inverse of the submatrix of $\Upsilon$ which exists, as it is the Grammian matrix of linearly independent vectors $\left\{\left|\alpha_{i}\right\rangle\right\}$. Eq. (4.32) is an analytic expression for the QFIM elements for localisation microscopy and our main result.


Figure 4.4: Diagonal and off diagonal elements of the QFIM for the case of 3 sources with equal intensities. The sources are separating from each other at equal distances, $\left(\alpha_{1}, \alpha_{2}, \alpha_{3}\right)=(x, 2 x, 3 x)$. The element $\mathcal{Q}_{12}$ and $\mathcal{Q}_{23}$ elements are equal, as are the $\mathcal{Q}_{11}$ and $\mathcal{Q}_{33}$ elements.

Fig. 4.4 shows the elements of the QFIM for the localisation microscopy of three point sources. We choose them to be equidistant, that is, $\left(\alpha_{1}, \alpha_{2}, \alpha_{3}\right)=$ $(x, 2 x, 3 x)$ and $w_{1}=w_{2}=w_{3}=1 / 3$ for illustration purposes, however, this is not a necessary condition for our results. Note the non-zero off-diagonal elements evidencing correlations in the precision around and below the Rayleigh limit of $x \sim 1$.

While the diagonal elements are all non-vanishing, more crucially as $x \rightarrow 0$ the diagonal and off-diagonal elements combine to make the QFIM singular. This is revealed by a closer analysis of the QFIM matrix as in Fig. 4.5 which shows that only two of its eigenvalues remain non-zero as the sources approach each other. This is in spite of all the diagonal elements of the QFIM remaining non-zero even as $x \rightarrow 0$, as Fig. 4.4 shows.

This behaviour of only two non-zero eigenvalues also holds for other values of $N$. We have explicitly checked this for $N=4, \ldots, 10$ as well as when the sources


Figure 4.5: The eigenvalues of the QFIM matrix for 3 sources with equal intensities. The sources are separating from each other at equal distances, $\left(\alpha_{1}, \alpha_{2}, \alpha_{3}\right)=$ ( $x, 2 x, 3 x$ ).
are not equally spaced. In Fig. 4.9 in Appendix 4.5.2 we plot the eigenvalues of the QFIM for $N=4,5$ as further examples. In the case of different relative intensities the results are the same except of the limiting case of one extremely bright source $w_{j} \gg 1, w_{i \neq j} \ll 1$, where the rank of the QFIM is approximately one (Fig. 4.10 in Appendix 4.5.2).

Since the QFIM has rank two as $x \rightarrow 0$, its inverse is ill-defined except on a two-dimensional subspace. This implies that the $N \times N$ covariance matrix for localisation microscopy, as per Eq. (3.15), will also be unbounded except on a twodimensional subspace. Thus, no more than two independent parameters can be estimated in localisation microscopy as the point sources approach each other.

In other words, the rank-deficient nature of the QFIM shows that a form of the Rayleigh limit resurfaces for any $N>2$. This had been suggested by previous works based on order-of-magnitude bounds for the diagonal elements on the CFIM [ZJ19] or uppers bounds on the diagonal elements of the QFIM [Tsa19a]. Our analytical expression for the full QFIM - its diagonal and off-diagonal elements for any $N$-shows that this rank two behaviour is truly quantum mechanical in origin. Furthermore, knowing the full QFIM matrix allows us to uncover the nature in which $N-2$ of the eigenvalues approach zero. We return to the behaviour in which this
rank deficiency or Rayleigh limit emerges in Sec. 4.4.

### 4.3.2 Why rank two?

We now provide an explanation for the rank deficiency of the QFIM in the regime of small separations which can be seen as the re-emergence of the Rayleigh limit. To that end, we expresses the state in Eq. (4.7) in terms of the real-valued displacement operator $\mathcal{D}\left(\alpha_{i}\right)=e^{\alpha_{i} \hat{c}_{i}^{\dagger}-\alpha_{i} \hat{c}_{i}}$ as

$$
\begin{equation*}
\rho=\sum_{i} \sqrt{w_{i}} \mathcal{D}\left(\alpha_{i}\right)|0\rangle\langle 0| \sqrt{w_{i}} \mathcal{D}^{\dagger}\left(\alpha_{i}\right) . \tag{4.33}
\end{equation*}
$$

In the limit of very small separations $\left(\alpha_{i} \ll 1\right)$, the displacements are approximately

$$
\begin{equation*}
\mathcal{D}\left(\alpha_{i}\right)=\mathbb{I}+\alpha_{i}\left(\hat{a}^{\dagger}-\hat{a}\right)+\mathcal{O}\left(\alpha_{i}^{2}\right) \tag{4.34}
\end{equation*}
$$

where $\mathbb{I}$ is the identity operator and the displacement $\alpha_{i}$ is real. Up to the second order in $\alpha_{i}$, the normalised quantum state of the light field on the image place is then

$$
\rho_{\boldsymbol{\alpha}}^{(2)}=\left(\begin{array}{cc}
1-\mathcal{C}_{2} & \mathcal{C}_{1}  \tag{4.35}\\
\mathcal{C}_{1} & \mathcal{C}_{2}
\end{array}\right)
$$

where $\mathcal{C}_{i}$ are the first two moments

$$
\begin{equation*}
\mathcal{C}_{1}=\sum_{i=1}^{N} w_{i} \alpha_{i}, \quad \mathcal{C}_{2}=\sum_{i=1}^{N} w_{i} \alpha_{i}^{2} \tag{4.36}
\end{equation*}
$$

Eq. (4.35) describes the state of two-level quantum system-the two levels being the first two HG modes. A similar approximation which described the state relative to a PSF centred at a fixed reference point was used in Ref. [Chr +17 ] to estimate the centroid and the effective radius of a distribution of incoherent point sources. We now consider the more general problem of estimating the location of $N$ point sources.

The QFIM for $\boldsymbol{\alpha}$ (See Appendix 4.5.3) is

$$
\mathcal{Q}\left(\rho_{\boldsymbol{\alpha}}^{(2)}\right) \equiv \mathscr{Q}=\frac{1}{\mathcal{A}}\left(\begin{array}{ll}
\mathbf{I} & \boldsymbol{\alpha} \tag{4.37}
\end{array}\right) \mathcal{M}\binom{\mathbf{I}^{T}}{\boldsymbol{\alpha}^{T}}
$$

with

$$
\mathcal{M}=\left[\begin{array}{ll}
M_{11} & M_{12}  \tag{4.38}\\
M_{21} & M_{22}
\end{array}\right]
$$

where $M_{11}=\left(\mathcal{C}_{2}-1\right) \mathcal{C}_{2}, M_{12}=M_{21}=\mathcal{C}_{1}\left(1-2 \mathcal{C}_{2}\right), M_{22}=4 \mathcal{C}_{1}^{2}-1, \mathcal{A}=\left(\mathcal{C}_{2}-1\right) \mathcal{C}_{2}+$ $\mathcal{C}_{1}^{2}$, and $\boldsymbol{I}=(11 \ldots 1)^{T}$.

The QFIM $\mathscr{Q}$ is an $N \times N$ matrix, which is a product of three matrices of dimensions $N \times 2,2 \times 2$ and $2 \times N$. Since $\operatorname{rank}(A B) \leq \min \{\operatorname{rank}(A), \operatorname{rank}(B)\}$, and the matrix $\mathcal{M}$ has rank 2, the QFIM $\mathscr{Q}$ has rank no more than two. Although a two-level quantum system has the potential of estimating three real parameters, localisation microscopy in this limit can estimate only two as the two-level system possesses a real density matrix ${ }^{2}$ This is another way of arguing that as the point sources get closer, the light field on the image plane has enough information to estimate only two parameters. A physical reason for this observation would be highly desirable.

### 4.4 Discussion

Our analytical expression for the QFIM for localisation microscopy has enabled us to show that as point sources get closer, no more than two independent parameters can be estimated. A rank-deficient QFIM occurs when the quantum state does not contain enough information to permit the estimation of some of the parameters or combinations thereof. The parameters that can be estimated correspond to the non-zero eigenvalues of the QFIM.

[^2]

Figure 4.6: Fitting of the eigenvalues of the QFIM matrix for the case of 9 sources in the limit of small distribution size. The sources are positioned at $\alpha_{i}=i x$. The size of the distribution is denoted $l=8 x$. The scale on both axes is logarithmic. The sources are separating from each other at equal distances, as in the previous plots. The slope of each line corresponding to different eigenvalues appears in the box in the plot.

Without additional knowledge of the source distribution this restricts us to estimating functions of the first two moments $f\left(\mathcal{C}_{1}, \mathcal{C}_{2}\right)$ only deep in the sub-Rayleigh limit. As Eq. (4.37) shows, when all $\left\{\alpha_{j}\right\}$ are unknown as in localisation microscopy, there is vanishing information about any single $\alpha_{i}$ itself. This is in contrast to the scalar QFI $\left[\mathcal{Q}\left(\rho_{\boldsymbol{\alpha}}\right)\right]_{i i}$ for $\alpha_{i}$ which is non zero, but assumes that all the other $\left\{\alpha_{j}\right\}$ are known. The manner in which the eigenvalues of the QFIM tend to zero is of interest in the search for optimal detection systems for localisation microscopy. Numerical fitting in Fig. 4.6 shows the vanishing eigenvalues of the QFIM approach zero polynomially. The degree of the polynomial is given by $d=2\left\lfloor\frac{\mu-1}{2}\right\rfloor$, where $\mu$ is the order the eigenvalue when arranged in descending order and $\lfloor\cdot\rfloor$ is the floor function. These scalings are now extracted from the elements of the full QFIM of the localisation parameters $\boldsymbol{\alpha}$ - rather than from bounds on estimating the various moments independently as in previous works [ZJ19; Tsa19a].


Figure 4.7: The eigenvalues of the QFIM and CFIM for the SPADE with 20 HG modes and 9 equally bright sources. The sources are positioned at $\alpha_{i}=(i-5) x$ such that the peak of $\left|\phi_{0}\right\rangle$ is at the centroid of the distribution. The $x$ axis is the size $l$ of the distribution, with $l=8 x$. The QFI eigenvalues scale as in Fig. 4.6. By SPADE with 20 modes, we mean the POVM $\left\{\left|\phi_{0}\right\rangle\left\langle\phi_{0}\right|,\left|\phi_{1}\right\rangle\left\langle\phi_{1}\right|, \ldots,\left|\phi_{20}\right\rangle\left\langle\phi_{20}\right|, \mathbb{I}-\right.$ $\left.\sum_{i=0}^{20}\left|\phi_{i}\right\rangle\left\langle\phi_{i}\right|\right\}$.

Unlike the latter, we can now compare the absolute performance of detection systems for localisation microscopy relative to its quantum limit. Indeed, while Fig. 4.7 shows the $2 n$-th eigenvalue of the QFIM closely parallel to the $n$-th eigenvalue of the CFIM for SPADE [TNL16], there is a large gap in the absolute terms. This could be due to the sub-optimality of SPADE for estimating the $\lfloor N / 2\rfloor$ parameters it is sensitive to ${ }^{3}$. Similar scalings were observed with detection using superpositions of the conventional SPADE basis [Tsa17; Tsa18; ZJ19] that are sensitive to the other half of the moments. For reference over a range of separations, Fig. 4.12 in Appendix 4.5.5 shows the eigenvalues of the CFIM for SPADE as well as direct imaging. Note that for both, the CFIM tends towards a rank one matrix.

[^3]

Figure 4.8: The eigenvalues of the QFIM matrix in the case of 3 sources with equal intensities and a sinc PSF. The sources are separating from each other at equal distances, i.e. $\left(\alpha_{1}, \alpha_{2}, \alpha_{3}\right)=(x, 2 x, 3 x)$.

Since it is a multiparameter problem the issue of attainability must be discussed. It can be seen that the density matrix $\rho_{\boldsymbol{\alpha}}$ of Eq. (4.7) as well as its derivative are real-valued in the orthonormal $\left\{\left|\phi_{k}\right\rangle\right\}$ basis. Therefore, Eq. (3.8) is a system of equations with real coefficients and $L^{\mu}, L^{\nu}$ are also real and the weak commutativity condition (3.18) is satisfied. The quantum limit for localisation microscopy is therefore attainable, at least in principle, although collective measurements over multiple copies [Mat02; RJD16] of the light field on the image plane may be required.

Finally, although our analytical result is derived with a Gaussian PSF, we expect the rank deficiency of the QFIM to be present in a more general family of PSFs. To that end, Fig. 4.8 shows the numerically obtained eigenvalues of the QFIM for three equidistant point sources of equal intensities under a sinc PSF (See Appendix 4.5.4) defined as

$$
\begin{equation*}
\psi_{\mathrm{PSF}}(x)=\frac{1}{\sqrt{\sigma}} \operatorname{sinc}\left(\frac{\pi x}{\sigma}\right) \tag{4.39}
\end{equation*}
$$

This PSF is the exact form for diffraction through a sharp one-dimensional slit which
in its principal peak is well-approximated as Gaussian.
An approximation involving the first two spherical Bessel modes as in Sec. 4.3.2 can be performed for a sinc PSF as well, leading to similar insights. A proof of this rank deficiency for arbitrary PSFs and a physical explanation remains an open question.

### 4.5 Proofs

### 4.5.1 Expressing the density matrix in the HG basis

The density matrix is written in terms of the kets $\left|\psi_{i}\right\rangle$, which are expressed in the position space as in Eq. (4.3). We assume a normalised Gaussian point spread function (PSF) of the form

$$
\begin{equation*}
\psi_{\mathrm{PSF}}(x)=\frac{1}{\left(2 \pi \sigma^{2}\right)^{1 / 4}} e^{-\frac{x^{2}}{4 \sigma^{2}}}, \tag{4.40}
\end{equation*}
$$

and so

$$
\begin{equation*}
\left|\psi_{i}\right\rangle=\int d x \psi_{\mathrm{PSF}}\left(x-\chi_{i}\right)|x\rangle \tag{4.41}
\end{equation*}
$$

The kets $\left|\psi_{i}\right\rangle$ can be expressed in terms of the complete Hermite-Gauss modes as

$$
\begin{equation*}
\left|\psi_{i}\right\rangle=\sum_{q=0}^{\infty}\left\langle\phi_{q} \mid \psi_{i}\right\rangle\left|\phi_{q}\right\rangle, \tag{4.42}
\end{equation*}
$$

where $\left|\phi_{q}\right\rangle$ are the Hermite-Gauss modes, which can be expressed in the position space as [TNL16]

$$
\begin{equation*}
\left|\phi_{q}\right\rangle=\frac{1}{\left(2 \pi \sigma^{2}\right)^{1 / 4}} \frac{1}{\sqrt{2^{q} q!}} \int d x \mathrm{H}_{q}\left(\frac{x}{\sqrt{2} \sigma}\right) e^{-\frac{x^{2}}{4 \sigma^{2}}}|x\rangle, \tag{4.43}
\end{equation*}
$$

where $\mathrm{H}_{q}(x)$ are the Hermite polynomials. The coefficients of the expansion Eq. (4.42) are

$$
\begin{align*}
\left\langle\phi_{q} \mid \psi_{i}\right\rangle & \left.=\frac{1}{\sqrt{2 \pi \sigma^{2}}} \frac{1}{\sqrt{2^{q} q!}} \int d x d x^{\prime} \mathrm{H}_{q}\left(\frac{x}{\sqrt{2} \sigma}\right) e^{-\frac{x^{2}}{4 \sigma}} e^{-\frac{\left(x^{\prime}-\chi_{i}\right)^{2}}{4 \sigma^{2}}}\langle x| x^{\prime}|x| x^{\prime}\right\rangle \\
& =\frac{e^{-\frac{-\chi_{i}^{2}}{8 \sigma^{2}}}}{\sqrt{2 \pi \sigma^{2} 2^{q} q!}} \int d x \mathrm{H}_{q}\left(\frac{x}{\sqrt{2} \sigma}\right) e^{-\left(\frac{x}{\sqrt{2} \sigma}-\frac{\chi_{i}}{\sqrt{2} \sigma}\right)^{2}}  \tag{4.44}\\
& =\left(\frac{\chi_{i}}{2 \sigma}\right)^{q} \frac{e^{-\frac{1}{2}\left(\frac{\chi_{i}}{2 \sigma}\right)^{2}}}{\sqrt{q!}}
\end{align*}
$$

Setting $\frac{\chi_{i}}{2 \sigma}=\alpha_{i}$ we get

$$
\begin{equation*}
\left|\alpha_{i}\right\rangle \equiv\left|\psi_{i}\right\rangle=\sum_{q=0}^{\infty} \frac{\alpha_{i}^{q}}{\sqrt{q!}} e^{-\alpha_{i}^{2} / 2}\left|\phi_{q}\right\rangle \tag{4.45}
\end{equation*}
$$

which has the same mathematical form as the coherent states with $\left\{\left|\phi_{q}\right\rangle\right.$ \} forming the Fock basis [KL10].

The state in Eq. (4.3) can be also written in terms of the displacement operators $\mathcal{D}\left(\alpha_{i}\right)=e^{\alpha_{i}\left(a^{\dagger}-a\right)}$, with $\alpha_{i}=\frac{\chi_{i}}{2 \sigma} \in \mathbb{R}$

$$
\begin{equation*}
\rho_{\boldsymbol{\alpha}}=\sum_{i} \sqrt{w_{i}} \mathcal{D}\left(\alpha_{i}\right)|0\rangle\langle 0| \sqrt{w_{i}} \mathcal{D}^{\dagger}\left(\alpha_{i}\right) \tag{4.46}
\end{equation*}
$$

where $\mathcal{D}(\alpha)$ is the displacement operator.
The derivative of each coherent state with respect to its real amplitude $\alpha$ is given by

$$
\begin{align*}
\frac{\partial|\alpha\rangle}{\partial \alpha} & =\frac{\partial D(\alpha)}{\partial \alpha}|0\rangle=\left(\hat{a}^{\dagger}-\alpha\right)|\alpha\rangle  \tag{4.47}\\
\frac{\partial\langle\alpha|}{\partial \alpha} & =\frac{\partial D^{\dagger}(\alpha)}{\partial \alpha}\langle 0|=(\hat{a}-\alpha)|\alpha\rangle
\end{align*}
$$

which yields the formula Eq. (4.17).

### 4.5.2 Analytic results for $N$ sources

The Tracy-Singh product [TS72; KNT91] defined for matrices $A$ and $B$ subdivided into blocks $A_{i j}$ and $B_{k l}$ is $A \odot B$ where the $(i, j)$-th block of $A \odot B$ is $A_{i j} \odot B$ whose
( $k, l$ )-th block is in turn $A_{i j} \otimes B_{k l}$. That is if $A, B$ are block matrices with

$$
A=\left(\begin{array}{ll}
A_{11} & A_{12} \\
A_{21} & A_{22}
\end{array}\right), \text { and } B=\left(\begin{array}{ll}
B_{11} & B_{12} \\
B_{21} & B_{22}
\end{array}\right),
$$

then the Tracy-Singh product is

$$
A \odot B=\left(\begin{array}{c|c}
A_{11} \odot B & A_{12} \odot B \\
\hline A_{21} \odot B & A_{22} \odot B
\end{array}\right)=\left(\begin{array}{ll|ll}
A_{11} \otimes B_{11} & A_{11} \otimes B_{12} & A_{12} \otimes B_{11} & A_{12} \otimes B_{12} \\
A_{11} \otimes B_{21} & A_{11} \otimes B_{22} & A_{12} \otimes B_{21} & A_{12} \otimes B_{22} \\
\hline A_{21} \otimes B_{11} & A_{21} \otimes B_{12} & A_{22} \otimes B_{11} & A_{22} \otimes B_{12} \\
A_{21} \otimes B_{21} & A_{21} \otimes B_{22} & A_{22} \otimes B_{21} & A_{22} \otimes B_{22}
\end{array}\right)
$$

Using the above definition, the matrix of Eq. (4.26) is found to be

$$
\left(\Upsilon^{-1} \odot \rho_{A}+\rho_{A} \odot \Upsilon^{-1}\right)=\left[\begin{array}{cccc}
D_{\boldsymbol{w}} \otimes v_{\alpha \alpha}+v_{\alpha \alpha} \otimes D_{\boldsymbol{w}} & D_{\boldsymbol{w}} \otimes v_{\alpha d} & v_{\alpha d} \otimes D_{\boldsymbol{w}} & 0  \tag{4.48}\\
D_{\boldsymbol{w}} \otimes v_{d \alpha} & D_{\boldsymbol{w}} \otimes v_{d d} & 0 & 0 \\
v_{d \alpha} \otimes D_{\boldsymbol{w}} & 0 & v_{d d} \otimes D_{\boldsymbol{w}} & 0 \\
0 & 0 & 0 & 0
\end{array}\right] .
$$

where the elements of $\Upsilon^{-1}$ can be found using the formula of blockwise inversion:

$$
\Upsilon^{-1}=\left[\begin{array}{ll}
v_{\alpha \alpha} & v_{\alpha d}  \tag{4.49}\\
v_{d \alpha} & v_{d d}
\end{array}\right]
$$

with

$$
\begin{align*}
& v_{\alpha \alpha}=\Upsilon_{\alpha \alpha}^{-1}+\Upsilon_{\alpha \alpha}^{-1} D_{\alpha} \Upsilon_{\alpha \alpha}\left(\Upsilon_{d d}-\Upsilon_{\alpha \alpha} D_{\alpha} \Upsilon_{\alpha \alpha}^{-1} D_{\alpha} \Upsilon_{\alpha \alpha}\right)^{-1} \Upsilon_{\alpha \alpha} D_{\alpha} \Upsilon^{-1} \\
& v_{\alpha d}=-\Upsilon_{\alpha \alpha}^{-1} D_{\alpha} \Upsilon_{\alpha \alpha}\left(\Upsilon_{d d}-\Upsilon_{\alpha \alpha} D_{\alpha} \Upsilon_{\alpha \alpha}^{-1} D_{\alpha} \Upsilon_{\alpha \alpha}\right)^{-1} \\
& v_{d \alpha}=-\left(\Upsilon_{d d}-\Upsilon_{\alpha \alpha} D_{\alpha} \Upsilon_{\alpha \alpha}^{-1} D_{\alpha} \Upsilon_{\alpha \alpha}\right)^{-1} \Upsilon_{\alpha \alpha} D_{\alpha} \Upsilon_{\alpha \alpha}^{-1}  \tag{4.50}\\
& v_{d d}=\left(\Upsilon_{d d}-\Upsilon_{\alpha \alpha} D_{\alpha} \Upsilon_{\alpha \alpha}^{-1} D_{\alpha} \Upsilon_{\alpha \alpha}\right)^{-1}
\end{align*}
$$

The inverse of the block matrix $\Upsilon_{\alpha \alpha}$ exists, because it is the Grammian matrix of the linear independent vectors $\left|\alpha_{i}\right\rangle$.

For the QFIM elements we need to evaluate the inverse of the top left $3 N^{2} \times$ $3 N^{2}$ part of the matrix of Eq. (4.48) which we denote $\mathbb{A}$. In order to obtain the
inverse of $\mathbb{A}$, we need to further partition $\mathbb{A}$ as

$$
\mathbb{A}=\left[\begin{array}{ll}
\varepsilon & \vartheta  \tag{4.51}\\
\varphi & \varpi
\end{array}\right]
$$

with

$$
\begin{array}{ll}
\varepsilon=\left[\begin{array}{lll}
D_{\boldsymbol{w}} \otimes v_{\alpha \alpha}+v_{\alpha \alpha} \otimes D_{\boldsymbol{w}}
\end{array}\right] & \vartheta=\left[\begin{array}{ll}
D_{\boldsymbol{w}} \otimes v_{\alpha d} & v_{\alpha d} \otimes D_{\boldsymbol{w}}
\end{array}\right] \\
\varphi=\left[\begin{array}{c}
D_{\boldsymbol{w}} \otimes v_{d \alpha} \\
v_{d \alpha} \otimes D_{\boldsymbol{w}}
\end{array}\right] & \varpi=\left[\begin{array}{cc}
D_{\boldsymbol{w}} \otimes v_{d d} & 0 \\
0 & v_{d d} \otimes D_{\boldsymbol{w}}
\end{array}\right] \tag{4.52}
\end{array}
$$

The inverse of $\varpi$ is

$$
\varpi^{-1}=\left(\begin{array}{cc}
D_{\boldsymbol{w}}^{-1} \otimes v_{d d}^{-1} & 0  \tag{4.53}\\
0 & v_{d d}^{-1} \otimes D_{\boldsymbol{w}}^{-1}
\end{array}\right)
$$

The elements of $\mathbb{A}^{-1}$ will be given by the formulas

$$
\begin{align*}
& \left(\mathbb{A}^{-1}\right)_{11}=\left(\varepsilon-\vartheta \varpi^{-1} \varphi\right)^{-1}=S^{-1} \\
& \left(\mathbb{A}^{-1}\right)_{12}=-S^{-1} \vartheta \varpi^{-1} \\
& \left(\mathbb{A}^{-1}\right)_{21}=-\varpi^{-1} \varphi S^{-1}  \tag{4.54}\\
& \left(\mathbb{A}^{-1}\right)_{22}=\varpi^{-1}+\varpi^{-1} \varphi S^{-1} \vartheta \varpi^{-1}
\end{align*}
$$

After calculations and by substituting the $\Upsilon^{-1}$ elements from Eq. (4.50), we derive the explicit form of $\mathbb{A}^{-1}$ elements:

$$
\begin{align*}
& \left(\mathbb{A}^{-1}\right)_{11}=S^{-1}=\left(\Upsilon_{\alpha \alpha}^{-1} \otimes D_{\boldsymbol{w}}+D_{\boldsymbol{w}} \otimes \Upsilon_{\alpha \alpha}^{-1}\right)^{-1} \\
& \left(\mathbb{A}^{-1}\right)_{12}=S^{-1}\left(\mathbb{I} \otimes\left(\Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d}\right) \quad\left(\Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d}\right) \otimes \mathbb{I}\right) \\
& \left(\mathbb{A}^{-1}\right)_{21}=\binom{\mathbb{I} \otimes\left(\Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1}\right)}{\left(\Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1}\right) \otimes \mathbb{I}} S^{-1} \\
& \left(\mathbb{A}^{-1}\right)_{22}=\left(\begin{array}{cc}
D_{\boldsymbol{w}}^{-1} \otimes v_{d d}^{-1} & 0 \\
0 & v_{d d}^{-1} \otimes D_{\boldsymbol{w}}^{-1}
\end{array}\right)+\binom{\mathbb{I} \otimes\left(\Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1}\right)}{\left(\Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1}\right) \otimes \mathbb{I}} S^{-1}\left(\mathbb{I} \otimes\left(\Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d}\right) \quad\left(\Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d}\right) \otimes \mathbb{I}\right) \tag{4.55}
\end{align*}
$$

The QFIM elements are then obtained from Eq. (4.31) and (4.55)

$$
\begin{align*}
\mathcal{Q}_{i j} & =2 w_{i} w_{j}\left[\begin{array}{lll}
-2 \alpha_{i}\left(E_{i} \mid\right. & \left(E_{i} \mid\right. & \left(E_{i} \mid\right.
\end{array}\right] \mathbb{A}^{-1}\left[\begin{array}{c}
\left.-2 \alpha_{j} \mid E_{j}\right) \\
\left.\mid E_{j}\right) \\
\left.\mid E_{j}\right)
\end{array}\right] \\
& =2 w_{i} w_{j}\left(E_{i}\left|\left[-2 \alpha_{i} \mathbb{I} \otimes \mathbb{I} \mathbb{I} \otimes \mathbb{I} \mathbb{I} \otimes \mathbb{I}\right] \mathbb{A}^{-1}\left[\begin{array}{c}
-2 \alpha_{j} \mathbb{I} \otimes \mathbb{I} \\
\mathbb{I} \otimes \mathbb{I} \\
\mathbb{I} \otimes \mathbb{I}
\end{array}\right]\right| E_{j}\right) \\
& =2 w_{i} w_{j}\left(E_{i} \mid\left[\mathbb{I} \otimes \Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1}+\Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1} \otimes \mathbb{I}-2 \alpha_{i} \mathbb{I} \otimes \mathbb{I}\right] S^{-1}\right. \\
& \left.\times\left[\mathbb{I} \otimes \Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d}+\Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d} \otimes \mathbb{I}-2 \alpha_{j} \mathbb{I} \otimes \mathbb{I}\right] \mid E_{j}\right)+2 w_{i} w_{j}\left(E_{i}\left|E_{\boldsymbol{w}}^{-1} \otimes v_{d d}^{-1}+v_{d d}^{-1} \otimes E_{\boldsymbol{w}}^{-1}\right| E_{j}\right) \\
= & 2 w_{i} w_{j}\left(E_{i} \mid\left[\mathbb{I} \otimes \Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1}+\Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1} \otimes \mathbb{I}-2 \alpha_{i} \mathbb{I} \otimes \mathbb{I}\right] S^{-1}\right. \\
& \left.\times\left[\mathbb{I} \otimes \Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d}+\Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d} \otimes \mathbb{I}-2 \alpha_{j} \mathbb{I} \otimes \mathbb{I}\right] \mid E_{j}\right)+4 w_{i} \delta_{i j}\left(v_{d d}^{-1}\right)_{i j} \\
& =2 w_{i} w_{j}\left(E_{i} \mid\left[\mathbb{I} \otimes \Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1}+\Upsilon_{d \alpha} \Upsilon_{\alpha \alpha}^{-1} \otimes \mathbb{I}-2 \alpha_{i} \mathbb{I} \otimes \mathbb{I}\right] S^{-1}\right. \\
& \left.\times\left[\mathbb{I} \otimes \Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d}+\Upsilon_{\alpha \alpha}^{-1} \Upsilon_{\alpha d} \otimes \mathbb{I}-2 \alpha_{j} \mathbb{I} \otimes \mathbb{I}\right] \mid E_{j}\right) \\
& +4 w_{i} \delta_{i j}\left[1+\alpha_{i}^{2}-\left(\Upsilon_{\alpha \alpha} D_{\alpha} \Upsilon_{\alpha \alpha}^{-1} D_{\alpha} \Upsilon_{\alpha \alpha}\right)_{i j}\right] \tag{4.56}
\end{align*}
$$

Finally, to complement the discussion in the main text, we present some further examples of the QFIM eigenvalues for $N=4,5$ sources and in Fig. 4.10 we present the eigenvalues of the QFIM for 3 sources in the case of unequal weights (relative intensities) Fig.(4.10).


Figure 4.9: The eigenvalues of the QFIM for 4 (top) and 5 (bottom) sources with equal intensities. The sources are separating from each other by equal distances: $\left(\alpha_{1}, \alpha_{2}, \alpha_{3}, \alpha_{4}\right)=(x, 2 x, 3 x, 4 x)$ and $\left(\alpha_{1}, \alpha_{2}, \alpha_{3}, \alpha_{4}, \alpha_{5}\right)=(x, 2 x, 3 x, 4 x, 5 x)$.


Figure 4.10: The eigenvalues of the QFIM matrix in the case of 3 sources. The sources are separating from each other at equal distances, i.e. $\left(\alpha_{1}, \alpha_{2}, \alpha_{3}\right)=(x, 2 x, 3 x)$. It can be noticed that the limiting values of the two non zero eigenvalues are different as the weights become different. However, the rank 2 of the QFIM remains. In Fig. (b) the limiting case of one extremely bright source wand two very weak ones is displayed. The inset shows the two vanishing eigenvalues.

### 4.5.3 Analytic results for $x_{i} \ll \sigma$

The state in the sub-diffraction regime is given by Eq. (4.35). The derivative can be calculated immediately from this formula and it is

$$
\frac{\partial}{\partial \alpha_{i}} \rho=-2 \alpha_{i}|0\rangle\langle 0|+[|0\rangle\langle 1|+|1\rangle\langle 0|]+2 \alpha_{i}|1\rangle\langle 1|=\left[\begin{array}{cc}
-2 \alpha_{i} & 1  \tag{4.57}\\
1 & 2 \alpha_{i}
\end{array}\right]
$$

By solving the SLD equation $\partial_{a_{i}} \rho_{\alpha}=\left(\rho_{\alpha} L_{i}+L_{i} \rho_{\alpha}\right)$, we can determine the SLDs in the $\{|0\rangle,|1\rangle\}$ basis:

$$
L_{i}=\frac{2}{\left(\mathcal{C}_{2}-1\right) \mathcal{C}_{2}+\mathcal{C}_{1}^{2}}\left[\begin{array}{cc}
\mathcal{C}_{2} \mathcal{C}_{1}+\left(\mathcal{C}_{2}-\mathcal{C}_{1}^{2}\right) \alpha_{i} & \left(\mathcal{C}_{2}-1\right) \mathcal{C}_{2}+\left(\mathcal{C}_{1}-2 \mathcal{C}_{2} \mathcal{C}_{1}\right) \alpha_{i}  \tag{4.58}\\
\left(\mathcal{C}_{2}-1\right) \mathcal{C}_{2}+\left(\mathcal{C}_{1}-2 \mathcal{C}_{2} \mathcal{C}_{1}\right) \alpha_{i} & \mathcal{C}_{1}-\mathcal{C}_{1} \mathcal{C}_{2}+\left(2 \mathcal{C}_{1}^{2}+\mathcal{C}_{2}-1\right) \alpha_{i}
\end{array}\right]
$$

Knowing the SLDs, we can obtain the QFIM of Eq. (4.37).
As already mentioned in the main text, the rank of the QFIM only depends on the matrix

$$
\left[\begin{array}{ll}
M_{11} & M_{12}  \tag{4.59}\\
M_{21} & M_{22}
\end{array}\right]
$$

of Eq. (4.37), with the elements of this matrix given by Eq. (4.38). The eigenvalues $\mu_{1}, \mu_{2}$ of the matrix Eq. (4.59) are

$$
\begin{align*}
& \mu_{1}=\frac{1}{2}\left(\mathcal{C}_{2}^{2}-\sqrt{\left(\left(\mathcal{C}_{2}-1\right) \mathcal{C}_{2}+4 \mathcal{C}_{1}^{2}-1\right)^{2}+4\left(\left(\mathcal{C}_{2}-1\right) \mathcal{C}_{2}+\mathcal{C}_{1}^{2}\right)}-\mathcal{C}_{2}+4 \mathcal{C}_{1}^{2}-1\right), \\
& \mu_{2}=\frac{1}{2}\left(\mathcal{C}_{2}^{2}+\sqrt{\left(\left(\mathcal{C}_{2}-1\right) \mathcal{C}_{2}+4 \mathcal{C}_{1}^{2}-1\right)^{2}+4\left(\left(\mathcal{C}_{2}-1\right) \mathcal{C}_{2}+\mathcal{C}_{1}^{2}\right)}-\mathcal{C}_{2}+4 \mathcal{C}_{1}^{2}-1\right) \tag{4.60}
\end{align*}
$$

The condition for the eigenvalues to be zero is

$$
\begin{align*}
& \left(0=3 \mathcal{C}_{1}^{2}-1-\sqrt{\left(3 \mathcal{C}_{1}^{2}-1\right)^{2}} \wedge \mathcal{C}_{2}=\frac{1}{2}\left(1-\sqrt{1-4 \mathcal{C}_{1}^{2}}\right)\right) \vee \\
& \left(0=3 \mathcal{C}_{1}^{2}-1-\sqrt{\left(3 \mathcal{C}_{1}^{2}-1\right)^{2}} \wedge \mathcal{C}_{2}=\frac{1}{2}\left(1+\sqrt{1-4 \mathcal{C}_{1}^{2}}\right)\right) \tag{4.61}
\end{align*}
$$

The first part $0=3 \mathcal{C}_{1}^{2}-1-\sqrt{\left(3 \mathcal{C}_{1}^{2}-1\right)^{2}}$ is always true, as it reduces to the identity
$\left(3 \mathcal{C}_{1}^{2}-1\right)^{2}=\left(3 \mathcal{C}_{1}^{2}-1\right)^{2}$. For the second part we have

$$
\begin{equation*}
\mathcal{C}_{2}=\frac{1}{2}\left(1 \pm \sqrt{1-4 \mathcal{C}_{1}^{2}}\right) \Leftrightarrow\left(2 \mathcal{C}_{2}-1\right)^{2}=1-4 \mathcal{C}_{1}^{2} \Leftrightarrow \mathcal{C}_{2}^{2}-\mathcal{C}_{2}+\mathcal{C}_{1}^{2}=0 \tag{4.62}
\end{equation*}
$$

Substituting $\mathcal{C}_{2}$ and $\mathcal{C}_{1}$ we get

$$
\begin{align*}
& \left(\sum_{i=1}^{N} \alpha_{i}^{2}\right)^{2}-\sum_{i=1}^{N} \alpha_{i}^{2}+\left(\sum_{i=1}^{N} \alpha_{i}\right)^{2}=0 \Leftrightarrow \\
& \left(\sum_{i=1}^{N} \alpha_{i}^{2}\right)^{2}-\sum_{i=1}^{N} \alpha_{i}^{2}+\sum_{i=1}^{N} \alpha_{i}^{2}+2 \sum_{i, j=1, i \neq j}^{N} \alpha_{i} \alpha_{j}=0  \tag{4.63}\\
& \left(\sum_{i=1}^{N} \alpha_{i}^{2}\right)^{2}+2 \sum_{i, j=1, i \neq j}^{N} \alpha_{i} \alpha_{j}=0
\end{align*}
$$

Since $\alpha_{i}$ are strictly positive, except one that can be zero, this sum of positive terms cannot be equal to zero. Therefore, this statement is always false. Thus, the Eq. (4.61) becomes $(1 \wedge 0) \vee(1 \wedge 0)=0$, which means that the two eigenvalues can never be zero and the QFIM will be rank 2 .

### 4.5.4 Calculation of the QFI for the Sinc PSF

The expansion of the Sinc function on the HG modes is not ideal for numerical calculations. Instead we use the spherical Bessel function of the $1^{\text {st }}$ kind and express the states onto those modes in which we then truncate. If the PSF is a sinc function, the $\left|\psi_{i}\right\rangle$ are

$$
\begin{equation*}
\left|\psi_{i}\right\rangle=\frac{1}{\sqrt{\sigma}} \int_{-\infty}^{\infty} \operatorname{sinc}\left(\frac{\pi\left(x-X_{i}\right)}{\sigma}\right)|x\rangle \tag{4.64}
\end{equation*}
$$

We can use the identity [AS64]

$$
\begin{equation*}
\operatorname{sinc}\left(\frac{\pi\left(x-x^{\prime}\right)}{\sigma}\right)=\sum_{q=0}^{\infty}(2 q+1) J_{q}\left(\frac{\pi x}{\sigma}\right) J_{q}\left(\frac{\pi x^{\prime}}{\sigma}\right), \tag{4.65}
\end{equation*}
$$

where $J_{q}(x)$ is the spherical Bessel function of the $1^{\text {st }}$ kind. The spherical Bessel function are orthogonal in all $\mathbb{R}$

$$
\begin{equation*}
\int_{-\infty}^{\infty} d x J_{q}(x) J_{p}(x)=\frac{\pi}{2 q+1} \delta_{q p}, \tag{4.66}
\end{equation*}
$$

therefore we can define the orthonormal basis

$$
\begin{equation*}
\left|j_{q}\right\rangle=\sqrt{\frac{2 q+1}{\sigma}} \int_{-\infty}^{\infty} d x J_{q}\left(\frac{\pi x}{\sigma}\right)|x\rangle \tag{4.67}
\end{equation*}
$$

The set of the spherical Bessel functions is a basis in $\mathbb{R}$, but is not complete since it is not a resolution of identity as we can see from Eq. (4.65). Hence, we can expand the sinc function on the bessel function basis, using the identity Eq. (4.65):

$$
\begin{align*}
\left|\psi_{i}\right\rangle & =\frac{1}{\sqrt{\sigma}} \int_{-\infty}^{\infty} \sum_{q=0}^{\infty}(2 q+1) J_{q}\left(\frac{\pi x}{\sigma}\right) J_{q}\left(\frac{\pi X_{i}}{\sigma}\right)|x\rangle \\
& =\sum_{q=0}^{\infty} \sqrt{2 q+1} J_{q}\left(\frac{\pi X_{i}}{\sigma}\right)\left|j_{q}\right\rangle \tag{4.68}
\end{align*}
$$

Using the identity for the Bessel functions

$$
\begin{equation*}
\frac{\partial J_{q}(x)}{\partial x}=J_{q-1}(x)-\frac{q+1}{2} J_{q}(x) \tag{4.69}
\end{equation*}
$$

we can also have an expression for the derivative of $\left|\psi_{i}\right\rangle$

$$
\begin{equation*}
\frac{\partial\left|\psi_{i}\right\rangle}{\partial X_{i}}=\frac{\pi}{\sigma}\left(J_{q-1}\left(\frac{\pi X_{i}}{\sigma}\right)-\frac{q+1}{2} J_{q}\left(\frac{\pi X_{i}}{\sigma}\right)\right) \tag{4.70}
\end{equation*}
$$

We see that both the state $\rho$ and its derivatives are completely expressed within the basis $\left|j_{q}\right\rangle$. This means that we can use the definition of the SLD (Eq. 4.71) and express the SLD in the same basis.

$$
\begin{equation*}
2 \frac{\partial \rho}{\partial \alpha_{\mu}}=\rho L^{\mu}+L^{\mu} \rho \tag{4.71}
\end{equation*}
$$

In this way the fact that the specific basis is not complete does not affect our calculations.

For the numerical calculations we have to truncate our state in the appropriate amount of modes. From Figs. 4.8 and 4.11, we can see that our conclusions do not change with the use of a non-Gaussian PSF.


Figure 4.11: The eigenvalues of the QFIM matrix in the case of 3 sources with equal intensities for the sinc PSF. The sources are separating from each other at equal distances, i.e. $\left(\alpha_{1}, \alpha_{2}, \alpha_{3}\right)=(x, 2 x, 3 x)$.

### 4.5.5 Eigenvalues of the CFIM for SPADE and Direct Imaging

Finally, we present the eigenvalues of the CFIM for SPADE and direct imaging for a large range of separations.


Figure 4.12: The eigenvalues of the CFIMs in the case of 9 sources for SPADE (left) and direct imaging (right). The sources are positioned at $\alpha_{i}=(i-5) x$.

## Chapter 5

## Quantum limits to spectroscopy

### 5.1 Introduction

Measuring the electric dipole moment (EDM) of an atom with high precision is a task of great importance for investigating fundamental physical models. For example, the EDM can be used to test models that explain phenomena that are not encompassed by the Standard model, such as CP violations [CY19], or that indicate the existence of undiscovered particles [Saf+18; Chu+19]. Moreover, estimating the EDM is crucial for characterising the interaction between atoms and light. Indeed, the atom-light coupling is proportional to the EDM, which plays an important role in applications such as an atom-light interface in free space [Cir +97 ; Dua +01 ; Tey +08 ; Slo +10 ].

The experimental setups used to measure the EDM of an atom employ spectroscopic methods [Chu +19 , where the atom is probed by light. Recent technological developments have made it possible to use quantum light, i.e. few-photon states or entangled states of light, in spectroscopy [Muk+20]. This resulted in enhanced sensing to limits below the classical shot noise limit [Wal15], in obtaining different scaling of the spectroscopic signals [DSM16] and in new spectroscopic techniques [YK04; Ray +13 ]. However, the advantages of using entangled light in spectroscopy remain unclear. Previous works have explored the role of entanglement in one-photon processes and have shown that in this class of processes the results of the entangled photon pair can be reproduced with correlated separable states [Ste17]. Additionally, in the case of absorption spectroscopy the ultimate precision limit is
achieved by using single-photon states [Ade +09 ; Whi +17 ] and two-mode squeezed vacuum [Shi +20 ], with the one-photon states performing better on a per-photon basis [Bie +21].

In this chapter, we calculate the QFI for the EDM of a two-level atom when it interacts with different states of light in a free space configuration. Specifically, we consider the quantum pulses of light that are described in Sec. 2.3. We focus on the regime of ultra-short pulses, i.e. of pulses whose time duration is of the order of a picosecond or less. In this regime, we find that the standard deviation of the estimator is larger than the value of EDM. This means that estimating the EDM precisely requires repeating the experiment a large number of times, which is the regime in which the QFI is meaningful [Bra92]. The one-photon wavepacket and the entangled photon pair have similar performances, while the performance of the entangled photon pair is not improved by increasing the entanglement of the photon pair. Overall, our results indicate that using Fock states to estimate the EDM of a two-level atom is preferable to using entangled light. This complements the results of Ref. [Ste17], where it is shown that entanglement does not necessarily provide advantage in one-photon interactions.

### 5.2 Physical description of the problem

We consider a two level atom which interacts with a light pulse in free space. In the case of two-mode light, we consider that only one mode interacts with the atom (Fig. 5.1). This system is described by the Hamiltonian $\mathcal{H}=\mathcal{H}_{A}+\mathcal{H}_{F}+\mathcal{H}_{\text {int }}$. The free Hamiltonian of the atom $\mathcal{H}_{A}$ is given by

$$
\begin{equation*}
\mathcal{H}_{A}=\hbar \omega_{0}|e\rangle\langle e|, \tag{5.1}
\end{equation*}
$$

where we set the energy of the ground state of the atom equal to 0 and we are interested only in the energy difference $\hbar \omega_{0}$. The quantity $\omega_{0}$ is called the transition frequency of the atom. The free Hamiltonian of the light $\mathcal{H}_{F}$ is given by Eq. (2.36). Following the formalism of Sec. 2.1.2, the interaction Hamiltonian is written in the interaction picture as [SZ97]

$$
\begin{equation*}
\mathcal{V}(t)=\left(\mathbf{d}(t)+\mathbf{d}^{\dagger}(t)\right)\left(\mathrm{E}^{\dagger}(t)+\mathrm{E}(t)\right) . \tag{5.2}
\end{equation*}
$$



Figure 5.1: Schematic of the atom-light interaction: (a) a wavepacket interacts with the sample and gets measured (b) the sample interacts with only one beam of the EPP (as defined in Sec. 2.3) and both beams get measured.

The operator $\mathbf{d}(t)$ is the positive frequency part of the dipole operator

$$
\begin{equation*}
\mathbf{d}(t)=\boldsymbol{\mu}_{\boldsymbol{e} \boldsymbol{g}} \sigma_{-} \mathrm{e}^{-i \omega_{0} t} \tag{5.3}
\end{equation*}
$$

where $\boldsymbol{\mu}_{\boldsymbol{e g}}=-q_{e}\langle e| \mathbf{r}|g\rangle$ is the dipole moment operator, $q_{e}$ is the charge of the electron and $\sigma_{-}=|g\rangle\langle e|=\sigma_{+}^{\dagger}$. The electric field operator $\mathrm{E}(t)$ is given by Eq. (2.38), since we are in a free space configuration. For convenience we set the quantisation constant of the field $\mathcal{A}(\bar{\omega})=\sqrt{\hbar \bar{\omega} /\left(2 \epsilon_{0} c A\right)}$. We also assume that the central frequency of the field is equal to the transition frequency of the atom $\bar{\omega}=\omega_{0}$.

The term $\mathbf{d}^{\dagger}(t) \mathrm{E}^{\dagger}(t)$ describes a process where a photon is created while the atom transitions to the excited state, while the hermitian conjugate of this term describes the opposite process. In the rotating wave approximation these terms that correspond to higher-order processes are dropped. By substitution of the dipole moment and electric field operator by their definitions, the interaction Hamiltonian
takes the form

$$
\begin{equation*}
\mathcal{V}(t)=-i \hbar \mu \int \frac{d \omega}{\sqrt{2 \pi}}\left(\sigma_{+} a(\omega) \mathrm{e}^{-i\left(\omega-\omega_{0}\right) t}-\sigma_{-} a^{\dagger}(\omega) \mathrm{e}^{i\left(\omega-\omega_{0}\right) t}\right) \tag{5.4}
\end{equation*}
$$

where $\mu=\mu(\bar{\omega})=-\mu_{e g} \mathcal{A}(\bar{\omega}) / \hbar$ and has the units of $1 / \sqrt{\sec }$. Using the definition of Eq. (2.50) the above equation is re-written as

$$
\begin{equation*}
\mathcal{V}(t)=-i \hbar \mu\left[\sigma_{+} b(t)-\sigma_{-} b^{\dagger}(t)\right] \tag{5.5}
\end{equation*}
$$

By choosing a complete set of orthonormal functions, as described in Sec. 2.3 and using Eq. (2.47)-(2.49), the interaction Hamiltonian (5.4) can be recast as

$$
\begin{equation*}
\mathcal{V}(t)=-i \hbar \mu \sum_{n=0}^{\infty}\left(\tilde{f}_{n}(t) \sigma_{+} A_{n}-\tilde{f}_{n}^{*}(t) \sigma_{-} A_{n}^{\dagger}\right) \tag{5.6}
\end{equation*}
$$

where $\tilde{f}(t)$ is the (frequency shifted) Fourier transform of $f(\omega)$. The above expression allows us to use a discrete and orthonormal basis for our calculations.

We consider an atom with infinite lifetime, that is we ignore any coupling with modes other than the incoming pulse. The atom, however, emits into the pulse modes. This approximation is valid in the regime in which the pulse duration is much smaller than the atom's lifetime. A detailed discussion about the subtleties around the dynamics of atoms interacting with $N$-photon wavepackets can be found in Ref. [KCW21]. By disregarding the spontaneous emission of the atom, the evolution is unitary and is described by Eq. (2.13).

We are interested in estimating the dipole moment $\mu_{e g}$. This parameter appears in the interaction Hamiltonian and not in the free evolution Hamiltonian, which allows us to continue working in the interaction picture and use the usual formulas for the QFI calculations, as discussed in Sec. 3.2.2. In order to calculate the QFI for the dipole moment we need to calculate the state of light by solving Eq. (2.13). In the remainder of the chapter, we study the interaction between a two-level atom and the states of light described in Sec. 2.3. Specifically, we consider one-photon wavepackets, coherent, squeezed and entangled states of light.

### 5.3 Analytical calculation of the QFI

In this section we provide analytical expressions for some states of light. Following Ref. [Bar14; KG16], we derive the solution for the $n$-photon wavepacket which are defined in Sec. 2.3. Given the Hamiltonian in Eq. (5.5), the state of the atom-light system can be found by solving the Schrödinger equation for a closed system, namely

$$
\begin{equation*}
\partial_{t}|\psi(t)\rangle=-\frac{i}{\hbar} \mathcal{V}(t)|\psi(t)\rangle . \tag{5.7}
\end{equation*}
$$

The general expression for the total state of the atom-light system is of the form:

$$
\begin{equation*}
|\psi(t)\rangle=|g\rangle\left|\phi_{g}(t)\right\rangle+|e\rangle\left|\phi_{e}(t)\right\rangle, \tag{5.8}
\end{equation*}
$$

where $\left|\phi_{e}(t)\right\rangle$ and $\left|\phi_{g}(t)\right\rangle$ correspond to the state of light when the atom is in the excited and ground state respectively. The states $\left|\phi_{e}(t)\right\rangle$ and $\left|\phi_{g}(t)\right\rangle$ are not normalised, but the total state of $|\psi(t)\rangle$ is. Substituting the above equation into the Schrödinger equation, we obtain the following system of differential equations [Bar14; KG16]:

$$
\begin{align*}
\partial_{t}\left|\phi_{e}(t)\right\rangle & =-\mu b(t)\left|\phi_{g}(t)\right\rangle \\
\partial_{t}\left|\phi_{g}(t)\right\rangle & =\mu b^{\dagger}(t)\left|\phi_{e}(t)\right\rangle \tag{5.9}
\end{align*}
$$

By integrating the above equations, we obtain the solutions [Bar14; KG16]:

$$
\begin{align*}
\left|\phi_{g}(t)\right\rangle= & \left|\phi_{g}(0)\right\rangle+\mu \int_{-\infty}^{t} d t^{\prime} b^{\dagger}\left|\phi_{e}\left(t^{\prime}\right)\right\rangle \\
\left|\phi_{e}(t)\right\rangle= & -\mu \int_{-\infty}^{t} d t^{\prime} \mathrm{e}^{-\mu^{2}\left(t-t^{\prime}\right) / 2} b\left(t^{\prime}\right)\left|\phi_{g}(0)\right\rangle  \tag{5.10}\\
& -\mu^{2} \int_{-\infty}^{t} d t^{\prime} \int_{-\infty}^{t^{\prime}} d t^{\prime \prime} \mathrm{e}^{-\mu^{2}\left(t-t^{\prime}\right) / 2} b^{\dagger}\left(t^{\prime \prime}\right) b\left(t^{\prime}\right)\left|\phi_{e}\left(t^{\prime \prime}\right)\right\rangle,
\end{align*}
$$

where we have assumed that the pulse envelope is zero at $t \rightarrow-\infty$. By successive substitutions of the expression of $\left|\phi_{e}(t)\right\rangle$, we obtain the following expressions for the states of light [Bar14; KG16]:

$$
\begin{align*}
\left|\phi_{e}(t)\right\rangle=\sum_{k=1}^{n+1}(-1)^{k}(\mu)^{2 k-1} & \sqrt{n \ldots(n-k+1)} \int_{-\infty}^{t} d t_{1} \ldots \int_{-\infty}^{t_{2 k}} d t_{2 k+1} \xi\left(t_{1}\right) \ldots \xi\left(t_{2 k+1}\right) \\
& \times \mathrm{e}^{-\mu^{2}\left(t-t_{1}\right) / 2-\mu^{2}\left(t_{2}-t_{3}\right) / 2-\ldots b\left(t_{2}\right) \ldots b\left(t_{2 k}\right)\left|n_{\xi}-(k+1)_{\xi}\right\rangle} \tag{5.11}
\end{align*}
$$

and

$$
\begin{align*}
\left|\phi_{g}(t)\right\rangle=\left|\phi_{g}(0)\right\rangle+\sum_{k=1}^{n} & (-1)^{k}(\mu)^{2 k} \sqrt{n \ldots(n-k)} \int_{-\infty}^{t} d t_{1} \ldots \int_{-\infty}^{t_{2 k-1}} d t_{2 k} \xi\left(t_{2}\right) \ldots \xi\left(t_{2 k}\right) \\
& \times \mathrm{e}^{-\mu^{2}\left[\left(t_{1}-t_{2}\right)+\ldots+\left(t_{2 k-1}-t_{2 k}\right)\right] / 2} b^{\dagger}\left(t_{1}\right) \ldots b^{\dagger}\left(t_{2 k}\right)\left|n_{\xi}-k_{\xi}\right\rangle . \tag{5.12}
\end{align*}
$$

From the above solutions, it can be seen that the analytical solution for a general $n$ photon wavepacket or superpositions therof, such as the coherent or squeezed pulses, is difficult to calculate and handle. For those states we employ numerical methods to compute the state of the atom-light system. The solution of the state for the case of a one-photon wavepacket, however, has a simple expression. By noticing that when the atom is in the excited state the light is in the vacuum state, the general atom-light state is written as

$$
\begin{equation*}
|\phi(t)\rangle=c_{e}(t)|e\rangle|0\rangle+|g\rangle\left|\phi_{g}(t)\right\rangle . \tag{5.13}
\end{equation*}
$$

Assuming that the (frequency shifted) temporal function is $\tilde{\xi}(t)$, which is the shifted Fourier transform defined in Eq. (2.51), the coefficient $c_{e}(t)$ is given by

$$
\begin{equation*}
c_{e}(t)=-\mu \mathrm{e}^{-\mu^{2} t / 2} \int_{-\infty}^{t} d t^{\prime} \mathrm{e}^{\mu^{2} t^{\prime} / 2} \tilde{\xi}\left(t^{\prime}\right) \tag{5.14}
\end{equation*}
$$

and the ket $\left|\phi_{g}(t)\right\rangle$ is given by

$$
\begin{align*}
\left|\phi_{g}(t)\right\rangle & =\left|\phi_{g}(0)\right\rangle+\mu \int_{-\infty}^{t} d t^{\prime} c_{e}\left(t^{\prime}\right) b^{\dagger}\left(t^{\prime}\right)|0\rangle \\
& =\int_{-\infty}^{t} d t^{\prime}\left(\tilde{\xi}\left(t^{\prime}\right)-\mu^{2} \mathrm{e}^{-\mu^{2} t^{\prime} / 2} \int_{-\infty}^{t^{\prime}} d t^{\prime \prime} \mathrm{e}^{\mu^{2} t^{\prime \prime} / 2} \tilde{\xi}\left(t^{\prime \prime}\right)\right) b^{\dagger}\left(t^{\prime}\right)|0\rangle . \tag{5.15}
\end{align*}
$$

It can be seen from the above equation that the temporal shape of the light pulse changes as the light interacts with the atom. Therefore, the initial pulse mode $\left|1_{\xi}\right\rangle$ does not suffice to describe the state of light. In order to describe the above state in a discrete basis it is necessary to choose a complete set of orthonormal functions as described in Sec. 2.3 to fully characterise the state. Choosing the Hermite-Gauss polynomials $\left\{H_{n}(t)\right\}$, with $H_{0}(t)=\tilde{\xi}(t)$, as the basis and using Eq. (2.47)-(2.49), the ket $\left|\phi_{g}(t)\right\rangle$ can be recast as

$$
\begin{equation*}
\left|\phi_{g}(t)\right\rangle=\sum_{n=0}^{\infty} c_{g}(n, t)\left|1_{n}\right\rangle, \tag{5.16}
\end{equation*}
$$

where

$$
\begin{equation*}
c_{g}(n, t)=\delta_{n 0}+\mu \int_{-\infty}^{t} d t^{\prime} H_{n}\left(t^{\prime}\right) c_{e}\left(t^{\prime}\right) \tag{5.17}
\end{equation*}
$$

and $c_{e}(t)$ is given by Eq. (5.14). The same result can be obtained by using the interaction Hamiltonian of Eq. (5.6).

In the limit of infinite time $t \rightarrow \infty$, the atom is in the ground state as $c_{e}(\infty)=0$. We note that the decay to the ground state is due to the decay in the pulse mode. The state of the atom-light system is pure, as well as the state of the light $\left|\phi_{g}(\infty)\right\rangle$. The QFI for this asymptotic state is calculated by the formula for pure states (3.26) and gives

$$
\begin{equation*}
\mathcal{Q}_{\mu}(+\infty)=4 \int_{-\infty}^{\infty} d t\left|\partial_{\mu} c_{e}^{\prime}(t)\right|^{2}+4\left(\int_{-\infty}^{\infty} d t\left[\tilde{\xi}(t)+c_{e}^{\prime}(t)\right] \partial_{\mu} c_{e}^{\prime}(t)\right)^{2}, \tag{5.18}
\end{equation*}
$$

where $c_{e}^{\prime}(t)=\mu c_{e}(t)=-\mu^{2} \mathrm{e}^{-\mu^{2} t} \int_{-\infty}^{t} d t^{\prime} \mathrm{e}^{\mu^{2} t^{\prime}} \tilde{\xi}\left(t^{\prime}\right)$.

### 5.3.1 Entangled photon pair

An analytical solution can also be obtained for the case of an entangled photon pair, which was defined in Sec. 2.3. We assume that only one of the photons, the first mode of the field, interacts with the atom as depicted in Fig. 5.1(b). Taking into account the expansion of the electric field operator onto the Schmidt modes given
by Eq. (2.64), the interaction Hamiltonian (5.5) is written as

$$
\begin{equation*}
\mathcal{V}(t)=\hbar \mu \sum_{k=1}^{\infty}\left(u_{k}^{(1)}(t) \hat{A}_{k} \sigma_{+}+u_{k}^{(1) *}(t) \hat{A}_{-}^{\dagger} \sigma_{+}\right) \otimes \mathbb{I}_{2} \tag{5.19}
\end{equation*}
$$

where $\mathbb{I}_{2}$ is the identity of the space of the second mode. The time evolved state can be written as

$$
\begin{equation*}
|\psi(t)\rangle=|g\rangle\left|\phi_{g}(t)\right\rangle+|e\rangle\left|\phi_{e}(t)\right\rangle \tag{5.20}
\end{equation*}
$$

The kets corresponding to the state of light can be expanded onto the Schmidt mode basis and have the form

$$
\begin{align*}
\left|\phi_{g}(t)\right\rangle & =\sum_{m, n} c_{m n}(t)\left|1_{m} 1_{n}\right\rangle  \tag{5.21}\\
\left|\phi_{e}(t)\right\rangle & =\sum_{n} d_{n}(t)\left|01_{n}\right\rangle
\end{align*}
$$

Substituting Eq. (5.20) into the Schrödinger equation (5.7) we obtain

$$
\begin{align*}
& \partial_{t}\left|\phi_{g}(t)\right\rangle=\mu \sum_{k} u_{k}^{*} \hat{A}_{k}^{\dagger}\left|\phi_{e}(t)\right\rangle \\
& \partial_{t}\left|\phi_{e}(t)\right\rangle=-\mu \sum_{k} u_{k} \hat{A}_{k}\left|\phi_{g}(t)\right\rangle \tag{5.22}
\end{align*}
$$

which because of Eq. (5.21) becomes

$$
\begin{align*}
\dot{c}_{n m} & =\mu u_{n}^{*}(t) d_{m}(t) \\
\dot{d}_{m} & =-\mu \sum_{n} u_{n}(t) c_{n m}(t) \tag{5.23}
\end{align*}
$$

The solution for the $c_{n m}(t)$ coefficient is

$$
\begin{equation*}
c_{n m}(t)=c_{n m}(0)+\mu \int_{-\infty}^{t} d t^{\prime} u_{n}^{*}(t) d_{m}\left(t^{\prime}\right) \tag{5.24}
\end{equation*}
$$

with $c_{n m}(0)=r_{m} \delta_{n m}$. Substituting the above solution into the differential equation for $d_{n}$ we get

$$
\begin{equation*}
\dot{d}_{m}=-\mu \sum_{n} u_{n}(t) c_{n m}(0)-\mu^{2} \int_{-\infty}^{t} d t^{\prime} \sum_{n} u_{n}(t) u_{n}^{*}\left(t^{\prime}\right) d_{m}\left(t^{\prime}\right) \tag{5.25}
\end{equation*}
$$

Taking into consideration the fact that ${ }^{1}$

$$
\begin{equation*}
\sum_{n} u_{n}(t) u_{n}^{*}\left(t^{\prime}\right)=\sum_{n} \frac{1}{2^{n} \sqrt{\pi} n!k_{i}} \mathrm{e}^{-\left(t+t^{\prime}\right)^{2} / 2 k_{i}^{2}} \mathrm{~h}_{n}\left(\frac{t}{k_{i}}\right) \mathrm{h}_{n}\left(\frac{t^{\prime}}{k_{i}}\right)=\frac{1}{k_{i}} \delta\left(\frac{t-t^{\prime}}{k_{i}}\right)=\delta\left(t-t^{\prime}\right) \tag{5.26}
\end{equation*}
$$

and $c_{n m}(0)=r_{m} \delta_{n m}$, we obtain the differential equation

$$
\begin{equation*}
\dot{d}_{m}(t)=-\mu r_{m} u_{m}(t)-\mu^{2} d_{m}(t), \tag{5.27}
\end{equation*}
$$

which can be solved by the integrating factor method. Finally, since the atom is initially in the ground state, $d_{m}(0)=0 \forall m$ and we obtain the solutions

$$
\begin{align*}
d_{n}(t) & =-\mu r_{n} \mathrm{e}^{-\mu^{2} t / 2} \int_{-\infty}^{t} d t^{\prime} \mathrm{e}^{\mu^{2} t^{\prime} / 2} u_{n}\left(t^{\prime}\right) \\
c_{n m}(t) & =r_{m} \delta_{n m}-\mu^{2} r_{m} \int_{-\infty}^{t} d t^{\prime} \int_{-\infty}^{t^{\prime}} d t^{\prime \prime} \mathrm{e}^{-\mu^{2}\left(t^{\prime}-t^{\prime \prime}\right) / 2} u_{n}^{*}\left(t^{\prime}\right) u_{m}\left(t^{\prime \prime}\right) \tag{5.28}
\end{align*}
$$

i.e. the kets $\left|\phi_{g}(t)\right\rangle,\left|\phi_{e}(t)\right\rangle$ are explicitly written as
$\left|\phi_{g}(t)\right\rangle=\sum_{n} r_{n}\left|1_{n} 1_{n}\right\rangle-\mu^{2} \sum_{n m} r_{m}\left(\int_{-\infty}^{t} d t^{\prime} \int_{-\infty}^{t^{\prime}} d t^{\prime \prime} \mathrm{e}^{-\mu^{2}\left(t^{\prime}-t^{\prime \prime}\right) / 2} u_{n}^{*}\left(t^{\prime}\right) u_{m}\left(t^{\prime \prime}\right)\right)\left|1_{m} 1_{n}\right\rangle$
$\left|\phi_{e}(t)\right\rangle=|0\rangle \sum_{m} \mu r_{m} \mathrm{e}^{-\mu^{2} t / 2} \int_{-\infty}^{t} d t^{\prime} \mathrm{e}^{\mu^{2} t^{\prime} / 2} u_{n}\left(t^{\prime}\right)\left|1_{m}\right\rangle$

The state of the light after we trace out the atom part is $\rho_{F}=\left|\phi_{g}(t)\right\rangle\left\langle\phi_{g}(t)\right|+$ $\left|\phi_{e}(t)\right\rangle\left\langle\phi_{e}(t)\right|$.

An asymptotic expression for $t \rightarrow \infty$ can also be obtained in the case of the EPP. However, the Hermite-Gauss basis is not appropriate for calculating the asymptotic state. The numerical integrals that must be evaluated are highly oscillatory for $t \rightarrow \infty$, especially for the higher order HG polynomials. Additionally, the numerical solution of the system of differential equations that are derived from Eq. (5.7) have large numerical errors for large times. The above things makes the numerical evaluation of the asymptotic QFI unreliable.

[^4]
### 5.4 Numerical results

The state of light for the one-photon wavepacket and EPP, as well as their derivatives, are calculated from the expressions of the previous section 5.3 with numerical integration methods. However, the SLD and therefore the QFI is calculated numerically using Eq. (3.19). For the coherent and squeezed states, both the state and the QFI are calculated numerically, due to the difficulty to obtain an analytical expression of the state as discussed in Sec. 5.3.

In our numerical calculations we assume that the atom is initially in the ground state. We use the experimental data reported in Ref. [Ste19] for the $D_{2}$ transition of a sodium atom. Specifically, we set the dipole moment $\mu=2.988 \cdot 10^{-29} \mathrm{C} \cdot \mathrm{m}$ $=1.868 \cdot 10^{-8} \mathrm{e} \cdot \mathrm{cm}$ and the transition frequency $\omega_{0}=2 \pi \cdot 508.333 \mathrm{THz}$. The lifetime of the sodium atom is 16.2492 ns and is much larger than the pulse durations we consider, which are of the order of $10^{-2} \mathrm{ps}$, and, hence, the infinite lifetime approximation is valid. We calculate the quantisation constant $\mathcal{A}(\bar{\omega})$ of the field by considering the transverse quantisation area to be proportional to the atomic radiative cross section $\sigma \propto \lambda_{0}^{2} / 2 \pi$, with $\lambda_{0}$ the central wavelength of the light $\lambda_{0}=2 \pi c / \omega_{0}$.

We assume that the spectral density functions of the one-photon wavepacket, coherent and squeezed states are Gaussian of the form of Eq. (2.53) with central frequency equal to the transition frequency of the atom $\omega_{0}$. We consider pulses with a bandwidth of $\sigma_{p}=3 \mathrm{THz}$ (i.e. a pulse duration of $\tau_{p}=2 \ln 2 /(\pi \sigma)=0.15 \mathrm{ps}$ ). We consider the same bandwidth of $\sigma_{p}=3 \mathrm{THz}$ for the pump pulse of the entangled states.

### 5.4.1 QFI for one-photon wavepacket

We start by discussing the case where the atom interacts with an one photon wavepacket. We compute the asymptotic QFI of Eq. (5.18) for the values of $\mu$ and $\sigma_{P}$ mentioned before, and the QFI that is obtained by the numerical solution of the system of differential equations that are derived from Eq. (5.7). In calculating the QFI that is plotted in Fiq. 5.2 we assumed that only the first HG mode interacts with the atom.

By considering the interaction between the atom and the $0^{\text {th }} \mathrm{HG}$ mode we attain half of the total (asymptotic) QFI (Fig. 5.2). Adding more modes does not lead to significant variations in the QFI. That is, the $0^{t h}$ mode has the only measur-


Figure 5.2: One-photon wavepacket: QFI for the state of the light, the atom and the pure atom-light state. The three of them overlap. The light pulse envelope is plotted for reference, as well as the asymptotic value of the QFI.
able contribution to the state and the other modes have infinitesimal contributions. Nevertheless, by summing the infinitesimal contributions of infinite modes we attain the asymptotic QFI (see discussion in App. 5.6.1 and the figures within).

Finally, we note that the QFI of the atom and light states is identical and equal to the total state QFI for both cases. For a Fock state interacting with a twolevel atom initially in the ground state this result is known for the cavity case [GI12].

### 5.4.2 QFI for different states of light

We now discuss the performance of various states of light. We have already assumed a Gaussian spectral density function with a bandwidth of $\sigma_{p}=3 T H z$. We additionally assume that the coherent and squeezed state have an average photon number of $\langle n\rangle=1$. The squeezed state has an amount of squeezing of 6 dB in the transition frequency, as a result of the specific pulse bandwidth. We also assume that only the first HG mode interacts with the atom. For the entangled light we vary the entanglement time, which can be done by setting a different length of the PDC crystal, to obtain entangled light with different entropies of entanglement [GUW01]. Specifically, we consider entropies of entanglement equal to $S_{\text {EPP }}=0.71,0.85,1.24$


Figure 5.3: The standard deviation of the estimator of the dipole moment for the various states of light. The dipole moment is of the order of $10^{-8} e \cdot \mathrm{~cm}$ and the standard deviation is two orders of magnitude larger. It can be noted that the Fock and the entangle photon pair perform similarly. The bandwidth $\sigma_{a}$ is the bandwidth that maximises the probability (5.30) in Sec. 5.4.3.
and 1.85. In all of these cases, we consider entangled light that is anti-correlated in frequency.

The standard deviation for the various states of light is plotted in Fig. 5.3. As it can be seen, all the states of light have a large variance compared to the value of the parameter for a single repetition. Indeed, the dipole moment value is $\mu=$ $1.868 \cdot 10^{-8} e \cdot \mathrm{~cm}$ and the standard deviation is $6.91 \cdot 10^{-6} e \cdot \mathrm{~cm}$, i.e. $\sqrt{(\Delta \mu)^{2}} \approx 100 \mu$. Thus, according to Eq. (3.15) a large number of repetitions of the experiment are needed in order for the mean square error of the estimator to become comparable with the value of the parameter, for example $\mathcal{N} \approx 1000$ in our setting. This should not pose an obstacle, as the laser repetition rates vary between $0.1-100 \mathrm{MHz}$ [CMK14]
with recent works reporting lasers with repetition rates of the order of GHz [Mis+19]. The standard deviation of the coherent state is not plotted in Fig. 5.3 as it is six orders of magnitude larger than the deviations of the other states of light, since the QFI of the coherent state is very close to zero. To demonstrate how the QFI changes as the atom interacts with the incoming pulse, we plot the QFI for the same states of light along with the Gaussian pulse temporal envelope of the states of light in Fig. 5.4. We note that the standard deviation plotted in Fig. 5.3 is the inverse of the square root of the QFI.

The Fock and entangled states have similar standard deviations but, depending on the Fock state's pulse bandwidth, the Fock state can slightly outperform the entangled light. In particular, the relative difference of the standard deviation between the entangled states is around $8 \%$ and that between the Fock and entangled states is around $11 \%$. Moreover, the standard deviation for the case of the biphoton increases as the entropy of entanglement decreases, indicating that entangled light is a disadvantage for the estimation of EDM. This complements the results of Ref. [Ste17] in the sense that not only entanglement does not provide a fundamental advantage, but it is also a drawback for the estimation process.

Finally, we elaborate on the efficiencies of the Fock and entangled photon pair generations, since many repetitions of the experiment are required. The efficiency of a BBO down-converting crystal is between $3.6 \times 10^{4} \mathrm{cps}$ and $1.42 \times 10^{5} \mathrm{cps}$ [SI08], which is the same efficiency rate for the generation of ultrafast single photons [Mos+08; Ans+18] and between $1 \times 10^{4}$ counts-per-second (cps) and $7 \times 10^{6}$ cps for single quantum emitters sources [Loh +17 ]. Thus, there is no significant advantage of choosing one state of light over the other.

### 5.4.3 Light-matter entanglement

In a spectroscopic setting the information about the properties of the atom is transferred to the light through the entanglement between them when they interact. To explain our results we calculate the entropy of entanglement of the atom-light system (which we denote by $S_{\mathrm{AF}}$ ) for the various states of light. We find $S_{\mathrm{AF}}=3.7 \cdot 10^{-5}$ for the Fock state, $S_{\mathrm{AF}}=3.0 \cdot 10^{-5}$ for the biphoton of $S_{\mathrm{EPP}}=0.7$, and $S_{\mathrm{AF}}=1.1 \cdot 10^{-6}$ for the squeezed state. For the coherent state we find $S_{\mathrm{AF}}$ is very close to zero, which is the reason for the its very small QFI. The Fock state has the largest light-


Figure 5.4: The QFI of the dipole moment for the various states of light and the Gaussian pulse envelope. It can be noted that the Fock state and the entangle photon pair perform similarly. The bandwidth $\sigma_{a}$ is the bandwidth that maximises the probability (5.30) in Sec. 5.4.3.
matter entanglement compared to the other states, which means that Fock states couple more efficiently with the two-level atom. Additionally to the efficient coupling, the Fock and the entangled states of light attain the maximum QFI of the system (Fig. 5.2).

Having observed that the entropy of entanglement between the atom and the light is what characterises the enhancement in the EDM estimation, we now provide an intuitive explanation for the disadvantage of the entangled photon pair in the EDM estimation. To do so, we determine the entangled photon pair with the minimum standard deviation by maximising the quantity $S_{A F}$ for the case of an infinite lifetime atom. The state of the atom for the entangled photon pair is diagonal (see Sec. 5.6.2). Since the atom is initially in the ground state, a higher entropy of entanglement is equivalent to a higher probability of excitation. We note that this is also true for the Fock state, since the state of the atom is also diagonal. The probability of excitation $P_{e}$ can be calculated for short times by approximating
the integrals of Eq. (2.67):

$$
\begin{align*}
P_{e}= & \sum_{m}\left|d_{m}(t)\right|^{2}=\frac{\mu^{2}}{\sqrt{2 a \pi}} \mathrm{e}^{-2 \mu^{2} t}\left(\frac{t^{2}}{2}+\mu^{2} t^{3}+\right.  \tag{5.30}\\
& \left.\frac{t^{4}}{12}\left(3 \gamma+7 \mu^{4}-8 \beta\right)+\frac{t^{5}}{12}\left(4 \gamma+3 \mu^{2}-10 \beta\right)\right)+\mathcal{O}\left(t^{6}\right)
\end{align*}
$$

where $\beta=\frac{w^{4}+1}{4 a\left(1-w^{2}\right)^{2}}, \gamma=\frac{w^{2}\left(1+w^{2}\right)}{a\left(1-w^{2}\right)^{2}}$ and $a, w$ are the Schmidt decomposition parameters. The probability of excitation has a maximum for $w=0$, i.e. for a Fock state with bandwidth $\sigma_{p}=\sigma_{a}=1 / \sqrt{2 a}$. Indeed, as it is shown in Fig. 5.5 , the one-photon wavepacket with $\sigma_{a}$ has the highest excitation probability and the lowest standard deviation (Fig. 5.3). As it can be seen, the probability of excitation decreases with the degree of entanglement for the light.


Figure 5.5: Probability of the atom to be in the excited state after it has interacted with the various states of light. The bandwidth $\sigma_{a}$ is the bandwidth that maximises the probability (5.30) in Sec. 5.4.3.

Finally, as it can be seen in Fig. 5.5, the high excitation probability for the case of the coherent light-which is almost identical to the excitation probability for the case of the Fock state - does not correspond to high entanglement between the atom and the coherent light. The reason is that the atom is in a pure state. The same observation was made for the coherent light-atom interaction in a cavity [PK91].


Figure 5.6: Schematic for "binary" mode-resolved photon counting measurement, where only the first component of the Schmidt decomposition is measured and the higher orders are not discriminated.

Hence, the excitation probability is not necessarily indicative of efficient atom-light coupling in the presence of coherences in the state of the atom.

### 5.5 Discussion

In the following, we focus on the Fock and EPP states, since they are the ones with higher QFI. We are interested in calculating the CFI for these states. We specifically consider the mode-resolved photon counting [HK13; EBS11; RW20], which counts the photons in each mode, in our case in each HG mode. The POVM for this measurement consists of the set $\left\{\Pi_{i}=\left|1_{i}\right\rangle\left\langle 1_{i}\right|, i=-1,0,1, \ldots\right\}$ for the single-mode case and $\left\{\Pi_{i, j}=\left|1_{i} 1_{j}\right\rangle\left\langle 1_{i} 1_{j}\right|, i, j=-1,0,1, \ldots\right\}$ for the two-mode (EPP) case, where we denote as $\left|1_{-1}\right\rangle$ the vacuum state for convenience and $i \in \mathbb{N}$ labels the $i^{\text {th }}$ Hermite-Gauss mode.

The photon resolved measurement saturates the QFI in the case of the onephoton wave packet. As discussed in the previous section, for the short pulses we are considering the atom interacts mainly with the initial pulse mode, the $0^{t h} \mathrm{HG}$ mode in our case. Under this assumption and by ignoring the higher modes, the state of the light is diagonal in the $\left\{|0\rangle,\left|1_{0}\right\rangle\right\}$ basis. Therefore, a POVM consisting of the projectors onto the basis $\left\{|0\rangle,\left|1_{0}\right\rangle\right\}$, i.e. the photon-counting measurement,
saturates the QFI.
The two-mode mode-resolved photon counting measurement also saturates the QFI for the entangled photon pair. We also consider a "binary" mode-resolved photon counting measurement ${ }^{2}$, where only the $n=0$ component of the Schmidt decomposition is measured and the higher orders are not discriminated (Fig. 5.6). The POVM of this measurement is $\left\{\Pi_{0}=\left|01_{1}\right\rangle\left\langle 01_{1}\right|, \Pi_{1}=\left|1_{1} 1_{1}\right\rangle\left\langle 1_{1} 1_{1}\right|, \Pi_{1}=\mathbb{I}\right.$ -$\left.\Pi_{0}-\Pi_{1}\right\}$. This measurement achieves approximately $98 \%$ of the QFI of the entangled states. This indicates that most of the information about the EDM is in the first mode of the Schmidt decomposition.


Figure 5.7: Value of the QFI for the asymptotic state of an one-photon wavepacket (5.18). It can be seen that the QFI is larger for longer pulses and there exists an optimal pulse length for which the QFI maximises.

Finally, in Fig. 5.7 the asymptotic value of the QFI (5.18) for a one-photon wavepacket with Gaussian spectral function for different pulse durations is plotted. It can be seen that the QFI is larger for ns pulses and there exists an optimal pulse length for which the QFI maximises. For example, the standard deviation for a 2 ns pulse is $\sqrt{(\Delta \mu)^{2}}=1.26 \cdot 10^{-8} \mathrm{e} \cdot \mathrm{cm}$ which is comparable to the value of the dipole moment $\mu=1.86 \cdot 10^{-8} \mathrm{e} \cdot \mathrm{cm}$. Therefore, longer pulses perform better in estimating

[^5]the EDM. This could be due to the fact that the pulses are narrower around the transition frequency of the atom, making the excitation probability and the atomlight entanglement larger, and to the fact that there is more time for the atom to interact with the light. However, for a fair comparison one must take into account the many repetitions of the experiment, which for shorter pulses might need less time to perform. Furthermore, as the pulses get longer the infinite lifetime assumption for the atom must be waived and the spontaneous emission into the environment must be considered.

The work presented in this chapter leaves room for further investigations. Firstly, a natural question is how to estimate the other parameter that characterises the two-level atom, the transition frequency. Secondly, an atom with finite lifetime could be considered and, hence, the interaction with the environment should be included. Finally, the two-level atom is limited to one-photon transitions for which we know that entanglement does not offer true advantage [Ste17]. One can consider more complex systems, such as coupled dimer, for which genuine two-photon interactions can occur and entanglement could lead to results that cannot be reproduced by non-entangled states. Indeed, for the estimation of the two-photon absorption cross section squeezed states of light outperform the classical-coherent states, as it has been shown theoretically in Ref. [SFS21]. Therefore, it is important to determine the advantage of entangled light in estimating parameters in those more complex systems.

Finally, we note the results presented here have been independently verified using the formalism presented in Ref. [KM20]. The contents of this chapter will be part of a publication that is currently in preparation.

### 5.6 Proofs

### 5.6.1 Expansion of Coherent and Squeezed states onto the Fock basis

The set of these Fock states $\left\{\left|n_{\xi}\right\rangle\right\}$ is a complete basis (see Sec. 2.3). As such, we can expand an arbitrary density matrix of the light field onto this basis as

$$
\begin{equation*}
\rho=\sum_{n, m=0}^{\infty} c_{n, m}\left|n_{\xi}\right\rangle\left\langle m_{\xi}\right| . \tag{5.31}
\end{equation*}
$$

We can rewrite the coherent and squeezed states defined in Sec. 2.3 as a superposition of Fock states.

The coherent states of Eq. (2.46) are defined in terms of $\xi(\omega)$ which is an arbitrary complex function not normalised. Instead its norm gives the average photon number of the coherent state, $\alpha^{2}=\int d \omega|\xi(\omega)|^{2}$. By re-defining the spectral amplitude as $\xi(\omega)=\alpha \bar{\xi}(\omega)$ with $\int d \omega|\bar{\xi}(\omega)|^{2}=1$ we obtain the following expression

$$
\begin{equation*}
|a\rangle=\mathrm{e}^{-\alpha^{2} / 2} \mathrm{e}^{\alpha \int d \omega \bar{\xi}(\omega) a^{\dagger}(\omega)}|0\rangle=\mathrm{e}^{-\alpha^{2} / 2} \sum_{n=0}^{\infty} \frac{\alpha^{n}}{\sqrt{n!}}\left|n_{\bar{\xi}}\right\rangle, \tag{5.32}
\end{equation*}
$$

which is a similar expression to the discrete-mode coherent state.


Figure 5.8: Squeezing Spectrum: The uncertainties $\Delta X(\omega)^{2}$ and $\Delta P(\omega)^{2}$ around the central frequency of the pulse.

A similar procedure can be followed for the continuous-mode squeezed vacuum of Eq. (2.57). Their expansion onto the Fock basis is

$$
\begin{equation*}
|\xi\rangle=\left(1-\left(\frac{\mathcal{Y}_{2}}{\mathcal{Y}_{1}}\right)^{2}\right)^{1 / 4} \sum_{n=0}^{\infty}(-1)^{n} \frac{\sqrt{2 n!}}{2^{n} 2!}\left(\frac{\mathcal{Y}_{2}}{\mathcal{Y}_{1}}\right)^{n}\left|2 n_{\xi}\right\rangle \tag{5.33}
\end{equation*}
$$

where

$$
\begin{align*}
& \mathcal{Y}_{1}=\int d \omega \xi(\omega) \cosh (r(\omega)), \text { and }  \tag{5.34}\\
& \mathcal{Y}_{2}=\int d \omega \xi^{*}(\omega) \sinh (r(\omega))
\end{align*}
$$

The q-quadrature uncertainty is given by

$$
\begin{align*}
(\Delta X(\omega))^{2} & =\left\langle\hat{Q}(\omega)^{2}\right\rangle=\frac{1}{2}\langle\xi|\left(a(\omega)+a^{\dagger}(\omega)\right)^{2}|\xi\rangle= \\
& =\sinh ^{2}(r(\omega))+\cosh ^{2}(r(\omega))-\cosh (r(\omega)) \sinh \left(r\left(\omega_{p}-\omega\right)\right) \cos \left(\theta\left(\omega_{p}-\omega\right)\right) \tag{5.35}
\end{align*}
$$

and the uncertainty for the p-quadrature is given by a similar expression. The spectrum of a squeezed state is plotted in Fig. 5.8. The squeezed state is assumed to have a Gaussian amplitude function with central frequency $\omega_{0}$.

### 5.6.2 EPP excitation probability

In order to calculate the excitation probability, firstly we need to derive the density matrix of the atom.

$$
\begin{align*}
\rho_{A}(t)= & \operatorname{Tr}_{F}\left[\rho_{A F}(t)\right]=\langle 00| \rho_{A F}(t)|00\rangle+\sum_{n}\left\langle 01_{n}\right| \rho_{A F}(t)\left|01_{n}\right\rangle+ \\
& +\sum_{n}\left\langle 1_{n} 0\right| \rho_{A F}(t)\left|1_{n} 0\right\rangle+\sum_{n m}\left\langle 1_{n} 1_{m}\right| \rho_{A F}(t)\left|1_{n} 1_{m}\right\rangle=  \tag{5.36}\\
= & |e\rangle\langle e| \sum_{n}\left|d_{n}(t)\right|^{2}+|g\rangle\langle g| \sum_{n m}\left|c_{n m}(t)\right|^{2}
\end{align*}
$$

where $\rho_{A F}(t)=|\psi(t)\rangle\langle\psi(t)|$. We can calculate the summations by using the identity of Eq. (5.26) and Mehler's formula [MF53, p.781]:

$$
\begin{equation*}
\sum_{n=0}^{\infty} \frac{(\epsilon / 2)^{n}}{n!} \mathrm{h}_{n}(x) \mathrm{h}_{n}(y)=\frac{1}{\sqrt{1-\epsilon^{2}}} \mathrm{e}^{-\frac{\epsilon^{2}\left(x^{2}+y^{2}\right)-2 \epsilon x y}{1-\epsilon^{2}}} \tag{5.37}
\end{equation*}
$$

The summations that appear in Eq. (5.36) correspond to

$$
\begin{equation*}
\sum_{m}\left|d_{m}(t)\right|^{2} \rightarrow \sum_{m} r_{m}^{2} u_{m}^{*}\left(t^{\prime}\right) u_{m}\left(t^{\prime \prime}\right)=\frac{1}{\sqrt{2 a \pi}} \mathrm{e}^{-\beta t^{\prime 2}+\gamma t^{\prime} t^{\prime \prime}-\beta t^{\prime \prime 2}} \tag{5.38}
\end{equation*}
$$

$$
\begin{equation*}
\sum_{n m}\left|c_{n m}(t)\right|^{2} \rightarrow \sum_{n m} r_{m}^{2} u_{n}\left(t^{\prime}\right) u_{n}^{*}\left(t_{1}\right) u_{m}^{*}\left(t^{\prime \prime}\right) u_{m}\left(t_{2}\right)=\frac{1}{\sqrt{2 a \pi}} \mathrm{e}^{-\beta t_{2}^{2}+\gamma t_{2} t^{\prime \prime}-\beta t^{\prime \prime 2}} \delta\left(t^{\prime}-t_{1}\right) \tag{5.39}
\end{equation*}
$$

where

$$
\begin{align*}
& \beta=\frac{w^{4}+1}{4 a\left(1-w^{2}\right)^{2}}, \text { and } \\
& \gamma=\frac{w^{2}\left(1+w^{2}\right)}{a\left(1-w^{2}\right)^{2}} \tag{5.40}
\end{align*}
$$

The quantities $w, a$ and $k_{1}$ are defined in Eq. (2.63). Using the above expressions, the atom coefficients have the explicit form of

$$
\begin{align*}
\sum_{m}\left|d_{m}(t)\right|^{2} & =\frac{\mu^{2}}{\sqrt{2 a \pi}} \mathrm{e}^{-2 \mu^{2} t} \int_{0}^{t} d t^{\prime} \int_{0}^{t} d t^{\prime \prime} \mathrm{e}^{\mu^{2}\left(t^{\prime}+t^{\prime \prime}\right)} \mathrm{e}^{-\beta t^{\prime 2}+\gamma t^{\prime} t^{\prime \prime}-\beta t^{\prime \prime 2}} \\
\sum_{n m}\left|c_{n m}(t)\right|^{2} & =1-\frac{2 \mu^{2}}{\sqrt{2 a \pi}} \int_{0}^{t} d t^{\prime} \int_{0}^{t} d t^{\prime \prime} \mathrm{e}^{\mu^{2}\left(t^{\prime}-t^{\prime \prime}\right)} \mathrm{e}^{-\beta t^{\prime 2}+\gamma t^{\prime} t^{\prime \prime}-\beta t^{\prime \prime 2}}+  \tag{5.41}\\
& +\frac{4 \mu^{4}}{\sqrt{2 a \pi}} \int_{0}^{t} d t^{\prime} \int_{0}^{t^{\prime}} d t_{1} \int_{0}^{t^{\prime}} d t_{2} \mathrm{e}^{-\mu^{2} t^{\prime}} \mathrm{e}^{\mu^{2}\left(t^{\prime}+t^{\prime \prime}\right)} \mathrm{e}^{-\beta t_{1}^{2}+\gamma t_{1} t_{2}-\beta t_{2}^{2}}
\end{align*}
$$

The above integrals cannot be calculated analytically, since they involve Error function integrals. For this reason, we approximately calculate them for short times by Taylor expanding them using the Leibniz integral rule. The coefficients are approximately

$$
\begin{align*}
& P_{e}= \sum_{m}\left|d_{m}(t)\right|^{2}=  \tag{5.42}\\
& \times\left[\frac{\mu^{2} \mathrm{e}^{-2 \mu^{2} t}}{\sqrt{2 a \pi}}\right. \\
& \sum_{n m}\left|c_{n m}(t)\right|^{2}=1-\frac{2 \mu^{2} t^{3}}{\sqrt{2 a \pi}}\left[\frac{t^{4}}{12}\left(3 \gamma+7 \mu^{4}-8 \beta\right)+\frac{t^{5}}{12}\left(4 \gamma+3 \mu^{2}-10 \beta\right)\right]+\mathcal{O}(6)  \tag{5.43}\\
&\left.+\frac{\mu^{4}}{\sqrt{2 a \pi}}\left[3 \gamma+\mu^{4}-8 \beta\right)+\frac{t^{5}}{6} \mu^{2} \beta\right]+ \\
&\left.+\frac{t^{3}}{4} \mu^{2}+\frac{t^{5}}{60}\left(3 \gamma+7 \mu^{4}-8 \beta\right)\right]+\mathcal{O}(6)
\end{align*}
$$

Since $\beta$ and $\gamma$ depend on $w$, the maximum $P_{e}(w)$ with regards to the value of $w$ can
be found. The values of $w$ that give maximum probability of excitation are:

$$
\begin{equation*}
w_{1}=0, \quad w_{2}=\frac{\sqrt{20-11 t \mu^{2}}}{\sqrt{5} \sqrt{20+t \mu^{2}}}, \quad w_{3}=-\frac{\sqrt{20-11 t \mu^{2}}}{\sqrt{5} \sqrt{20+t \mu^{2}}} \tag{5.44}
\end{equation*}
$$

The values $w_{2}, w_{3}$ are rejected since they depend on the interaction time, whereas $w$ depends only on the properties of the PDC. For $w=0$ the photon pair is in a product state of two Fock states. Therefore, the maximum excitation probability is obtained for a Fock state.


Figure 5.9: One-photon wavepacket QFI for different number of HG modes truncation. The QFI is the same for all different truncations.

### 5.6.3 Necessary orthogonal modes

Here, we provide numerical evidence that, for the case of ultra-short one-photon pulses, the $0^{\text {th }}$ mode has the only measurable contribution to the state and that other modes have infinitesimal contributions, but their infinite sum results in the asymptotic QFI.

(a) The values of the coefficients $c_{g}(n, t)$ of the state $\left|\phi_{g}(t)\right\rangle$ for each mode $n$ for time equal to $10 \tau_{p}$, i.e. 10 pulse durations. For comparison we mention that $\left|c_{g}\left(0,10 \tau_{p}\right)\right| \simeq 0.97$ and $\left|c_{e}\left(10 \tau_{p}\right)\right| \simeq 0.02$. It is clear that the greatest contribution is from the $0^{t h}$ mode.

(b) The values of the coefficients $\operatorname{Re}\left\{c_{g}(n, t) \partial_{\mu} c_{g}(n, t)\right\}$ of the state $\left|\phi_{g}(t)\right\rangle$ for each mode $n$ for time equal to $10 \tau_{p}$, i.e. 10 pulse durations. For comparison we mention that $\left|c_{g}\left(0,10 \tau_{p}\right)\right| \simeq 0.97$ and $\left|c_{e}\left(10 \tau_{p}\right)\right| \simeq 0.02$. It is clear that the greatest contribution is from the $0^{t h}$ mode.

Figure 5.10: The values of $c_{g}(n, t)$ and the derivatives $\operatorname{Re}\left\{c_{g}(n, t) \partial_{\mu} c_{g}(n, t)\right\}$ for each mode for time equal to $10 \tau_{p}$, i.e. 10 pulse durations, much after the light has interacted with the atom.

In Fig. 5.10a we plot the values of $c_{g}(n, t)$ for each mode for time equal to $10 \tau_{p}$, i.e. 10 pulse durations, much after the light has interacted with the atom. It can be seen that with the exception of the $0^{t h}$ mode with a value $c_{g}\left(0,10 \tau_{p}\right)=0.9$ the rest of the values are much smaller than 1 . The same is true for the elements of the derivative of the state $\left|\partial_{\mu} \phi(t)\right\rangle$, as it can be seen from Fig. 5.10b. In Fig. 5.9 the value of the QFI for different truncations of the state is plotted. We can see that adding extra modes does not change the value of the QFI by a notable amount. Therefore, in order to obtain the value of the asymptotic QFI we need to take into consideration the all the infinite modes of light.

### 5.6.4 Necessary number of modes for the EPP

We start this section with two remarks. Firstly, we want to point out that even the initially vacant Schmidt modes might become occupied after the interaction with the light pulse. This is because after a photon from a specific Schmidt mode has been absorbed, it can be re-emitted into an initially vacant one. This can be seen more clearly if we consider the example of the second order term of the expansion of the evolution operator $\mathrm{U}(t, 0)=\mathrm{e}^{-i / \hbar \int_{0}^{t} \mathcal{V}\left(t^{\prime}\right) d t^{\prime}}$ acting on the initial state $\left|11_{\text {ent }}\right\rangle|g\rangle=|g\rangle \sum_{m=1}^{c_{f}} r_{m}\left|1_{m} 1_{m}\right\rangle$ :

$$
\begin{aligned}
\mathrm{U}^{(2)}(t, 0)|g\rangle \sum_{m=1}^{c_{f}} r_{m}\left|1_{m} 1_{m}\right\rangle= & -\mu^{2}|g\rangle \sum_{k, l=1}^{\infty} \sum_{m=1}^{c_{f}} r_{m} c_{k l}(t) \hat{A}_{k}^{\dagger} \hat{A}_{l}\left|1_{m} 1_{m}\right\rangle \\
& =-\mu^{2}|g\rangle \sum_{k=1}^{\infty} \sum_{m=1}^{c_{f}} r_{m} c_{k m}(t) \hat{A}_{k}^{\dagger}\left|01_{m}\right\rangle
\end{aligned}
$$

where the $k$ index is running over all the infinite modes.
Secondly, we would like to note that the $|0\rangle$ of the first light mode that appears in Eq. (5.21) is the vacuum for all modes. This can be seen if we apply the first order term of the evolution operator:

$$
\begin{aligned}
& \mathrm{U}^{(1)}(t, 0)|g\rangle \sum_{m=1}^{c_{f}} r_{m}\left|1_{m} 1_{m}\right\rangle=-i \mu|e\rangle \sum_{k=1}^{\infty} \sum_{m=1}^{c_{f}} r_{m} c_{k}(t) \hat{A}_{k}\left|1_{m} 1_{m}\right\rangle \\
& \quad=-i \mu|e\rangle \sum_{k=1}^{\infty}\left(r_{1} c_{k}(t) \hat{A}_{k}\left|1_{1} 00 \ldots 0\right\rangle\left|1_{1}\right\rangle+\ldots+r_{f} c_{k}(t) \hat{A}_{k}\left|000 \ldots 1_{f}\right\rangle\left|1_{f}\right\rangle\right)
\end{aligned}
$$

The terms where $k \neq m$ are 0 and the terms for which $k=m$ the photon in this mode annihilates giving the vacuum for all modes.

(a) The initial truncation of the bi-photon is for Schmidt modes $\mathrm{SM}=7$. We use different number of initially empty Schmidt modes to see if the QFI converges. We can see that the QFI is identical for all different number of empty Schmidt modes.

(b) The QFI for different truncation of the bi-photon. We do not consider any additional vacant modes. We can see that the QFI is identical for all different number of empty Schmidt modes.

Figure 5.11: QFI values for the same bi-photon state with entropy of entanglement $S=0.7$ and: (a) different number of vacant modes, (b) different truncation of the state.

### 5.6.5 QFI for the individual states of light

Here, we present the QFI for each case separately. In each figure, the QFI of the light, the atom and the total atom-light system is plotted.


Figure 5.12: Coherent state: QFI for the state of the light, the atom and the pure atom-light state. The state of light has a QFI of six orders of magnitude less than the total QFI.


Figure 5.13: Squeezed state: QFI for the state of the light, the atom and the pure atom-light state. The three of them overlap.


Figure 5.14: Entangled photon pair: QFI for the state of the light, the atom and the pure atom-light state. The three of them overlap.

### 5.6.6 Measurements and probability distributions

The measurements can only be performed on the state of light. For the probability derivations for the cases of Fock, coherent and squeezed light we consider the following general expression, As follows from Eq:

$$
\begin{equation*}
\rho_{F}(t)=\operatorname{Tr}_{\mathrm{A}}\left[\rho_{A F}(t)\right]=\left|c_{e}(t)\right|^{2}|0\rangle\langle 0|+\sum_{m, n=0}\left|1_{n}\right\rangle\left\langle 1_{m}\right|\left(c_{g}(n, t) c_{g}^{*}(m, t)\right), \tag{5.45}
\end{equation*}
$$

where $c_{g}(n, t)$ are defined from Eq. (5.17). Similarly for the EPP, the light density matrix is given by

$$
\begin{equation*}
\rho_{E P P}(t)=\sum_{m, n=1}^{\infty} d_{n}(t) d_{m}^{*}(t)\left|01_{m}\right\rangle\left\langle 01_{n}\right|+\sum_{k, l, m, n=1}^{\infty} c_{k, m}(t) c_{l, n}^{*}(t)\left|1_{k} 1_{m}\right\rangle\left\langle 1_{l} 1_{n}\right| \tag{5.46}
\end{equation*}
$$

The POVM of the mode-resolving measurement is defined in Sec. 5.5 as [RW20] $\left\{\Pi_{i}=\left|1_{i}\right\rangle\left\langle 1_{i}\right|, i=-1,0,1, \ldots\right\}$ for the single-mode case and $\left\{\Pi_{i, j}=\left|1_{i} 1_{j}\right\rangle\left\langle 1_{i} 1_{j}\right|, i, j=\right.$ $0,1, \ldots\}$ for the two-mode case, where for $i=-1$ the detector in the $1^{\text {st }}$ mode does not detect any photons. Since the POVMs are the projectors onto the basis elements
the states (5.46) and (5.45) are projected, the probability distribution is given by

$$
\begin{align*}
P_{n} & =\left|c_{g}(n, t)\right|^{2},  \tag{5.47}\\
P_{-1} & =\left|c_{e}(t)\right|^{2},
\end{align*}
$$

for the single-mode case and

$$
\begin{align*}
P_{n \neq-1, m} & =c_{n, m}(t) c_{n, m}^{*}(t),  \tag{5.48}\\
P_{-1, m} & =d_{m}(t) d_{m}^{*}(t),
\end{align*}
$$

for the two-mode case. The CFI is given by substitution into Eq. (3.17).

## Chapter 6

## Conclusions

This thesis has explored the quantum limits of microscopy and spectroscopy. Although we have limited ourselves to the study of the simplest configurations of these vast fields that have highly sophisticated methods, we hope that this thesis sheds light on the fundamental limits of these two ubiquitous tools in science and technology.

In chapter 4 we examined the estimation problem of $N$ positions under the diffraction limit. We have obtained several insights into the quantum limits of localisation microscopy via an analytical expression for the QFIM. In particular, the behaviour of the eigenvalues of the QFIM deep in the sub-Rayleigh limit revealed that only two parameters are eventually estimable. It also enabled us to compare the performance of known detection systems relative to the quantum limit in absolute terms, a question left open in the literature [Tsa19b].

Our work is included in the general effort to determine the resolution limits in imaging. The non-orthogonal basis approach for expressing the quantum states [GT19] and our formalism with block-matrices have enabled the study of the two-point sources problem in three dimensions with an arbitrary PSF [Fid+21]. Theoretical works have also contributed to the understanding of the role of realistic, and hence noisy, measurement devices in two-point sources' separation estimation. Analysis of SPADE measurements with noisy detectors and detectors that introduce cross-talks between the decomposed modes have been made and have found that the super-resolution feature for arbitrarily small separation is lost in such cases. Instead, a minimum resolvable spatial separation is introduced which depends on the
the signal-to-noise ratio or the number of detected photons respectively [Len +20 ; GFT20]. Additionally, adaptive strategies for simultaneous centroid and separation estimation has been proposed [Gra+20], as well as schemes for measuring the separation of two sources of unequal and unknown intensities, in which case the Rayleigh curse returns [Bon+19]. Most recent experimental demonstrations include SPADE measurements using multiple spatial modes for measuring the separation of two point sources [Bou +20$]$, as well as the the effect of partial coherence on the sub-diffraction limit localisation of two sources [Wad+21]. In the case of two point sources an optimal strategy to simultaneously estimate centroid and separation has still not been determined [Alb +20 ]. Also based on our results, it can be seen that the SPADE measurement does not achieve the quantum bounds. The gap identified by us should motivate the search for detection systems, ideally on a single copy of the light field on the image plane, seeking to reduce or eliminate it.

In chapter 5 we examined the estimation problem of the electric dipole moment (EDM) of a two-level atom interacting with light pulses in free space. We considered the case of ultra-short - picosecond - pulses that are usually employed in spectroscopy experiments. We derived analytical expressions for the QFI in the asymptotic limit for the Fock and entangled states of light. We numerically calculated the QFI for finite times for different states of light and compared their performance in estimating the EDM. We found that entanglement does not offer an advantage compared to non-entangled light, as expected from Ref. [Ste17], but entanglement acts as an impediment in the estimation process.

Even though it is shown that entanglement offers true advantage only in genuine two-photon processes [Ste17], the conditions under which such an advantage can be achieved is not clear yet. Several experiments have presented evidence that entangled two-photon absorption in molecules does provide advantages, however, other experiments, as well as theory, have disputed those conclusions, as reviewed in Ref. [RW20]. Studies which make use of quantum estimation theory can provide the means for a clear understanding. The methods described in this thesis fall into the above category and can be extended to two or multiphoton processes. Another such example is the recent publication of Ref. [SFS21] in which the estimation of the two-photon absorption cross section has been calculated. Since there have not been many works that examine spectroscopy with quantum light in the context of quantum metrology, it is a fertile ground for future research. It could be of great importance
both in determining the dynamical processes in light-matter interactions, as well as the properties of quantum materials [Szo +20 ; KM17].

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[^0]:    ${ }^{1}$ The support of a matrix is defined as the space that is orthogonal to the kernel of the matrix.

[^1]:    ${ }^{1}$ Unlike the conventional quantum optical coherent states which reside in the phase space of the electromagnetic field, our coherent states reside in physical space on the image plane. This mathematical form was also identified by [Dut+19] but only used for numerical calculations.

[^2]:    ${ }^{2}$ As the localisation parameters $\boldsymbol{\alpha}$ are real, $\operatorname{Tr}\left(\rho_{\alpha}^{(2)} \sigma_{y}\right)=2 \operatorname{Im}\left(\mathcal{C}_{1}\right)=0$, where $\sigma_{y}$ is the Pauli $Y$ matrix.

[^3]:    ${ }^{3}$ Conventional SPADE is not sensitive to all the parameters needed to describe the sources' distribution, only its even moments [Tsa19a; ZJ19; Tsa19b].

[^4]:    ${ }^{1}$ The summation identity can be found at: functions.wolfram.com/05.01.23.0014.01

[^5]:    ${ }^{2}$ The mode-resolved photon counting measurement that is considered here is mathematically identical to SPADE measurement with the additional element $\{|0\rangle\langle 0|\}$.

