2 Introduction to various tools

This chapter is devoted to a brief description of various tools used to study the dynamics of quantum systems. Specifically, the dynamics of open and closed systems is spelled out, followed by the description of dynamical maps. Further, a detailed account of various quantum correlation, both temporal as well as spatial is presented.

2.1 Dynamics of closed systems

The state of a closed physical system is described by a state vector $|\psi\rangle$ which is an element of some Hilbert space \mathcal{H} . The scalar product of two states $|\psi\rangle$ and $|\phi\rangle$ in \mathcal{H} is defined as $\langle \phi | \psi \rangle$. Accordingly, one defines the norm of $|\psi\rangle$ as $||\psi|| = \sqrt{\langle \psi | \psi \rangle}$ [210]. Let q be the set of various parameters apart from time t on which the state vector depends, one shows this dependence as $|\psi(q,t)\rangle$. The time evolution is given by the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\left|\psi(q,t)\right\rangle = H\left|\psi(q,t)\right\rangle.$$
(2.1)

Here, H is the generator of time evolution known as Hamiltonian of the system, and is generally a Hermitian operator. Though one can arrive at Schrödinger equation by different arguments, its ultimate validity relies on the fact it agrees with the experiments [211–213]. The solution of Eq. (2.1) can be represented in terms of unitary operator $U(t, t_0)$ which takes the state $|\psi(q, t_0)\rangle$ at time t_0 to $|\psi(q, t)\rangle$ at some later time t, such that, $|\psi(q, t)\rangle = U(t, t_0) |\psi(q, t_0)\rangle$. This leads to a time evolution equation of the unitary operator $i\partial U(t, t_0)/\partial t = H(t)U(t, t_0)$, subjected to the initial condition $U(t_0, t_0) = \mathbf{I}$. If Hamiltonian is time independent, the system is said to be *closed* and *isolated*; such a system is described by the unitary $U(t - t_0) = \exp[-iH(t - t_0)]$. If the system is under the influence of an external driving, the dynamics may still be described by a time dependent Hamiltonian H(t), the system is still called *closed* but not *isolated*. In this case, the time evolution is given in terms of the time ordered operator $U(t, t_0) = T \exp[-i \int_{t_0}^t ds H(s)]$, where T means that the product of time dependent operators are ordered such that the operators at earlier times are at the left of the operators at later times.

The above description holds true for *pure* states. However, if the system is represented by an ensemble of pure states $\{p_i, |\psi_i(q, t)\rangle\}$, p_i being the weight associated with pure state $|\psi_i\rangle$, one has to resort to the description of density matrix description. At time t_0 , one defines the density matrix as $\rho(q, t_0) = \sum_i p_i |\psi_i(q, t_0)\rangle \langle \psi_i(q, t_0)|$, which, under unitary evolution, evolves at some later time t

$$\rho(q,t) = \sum_{i} p_i U(t,t_0) |\psi_i(q,t_0)\rangle \langle \psi_i(q,t_0) | U^{\dagger}(t,t_0).$$
(2.2)

This can be written more compactly as $\rho(q,t) = U(t,t_0)\rho(q,t_0)U^{\dagger}(t,t_0)$, which upon differentiation leads to the *von Neumann* or *Liouville-von Neumann* equation

$$\frac{\rho(q,t)}{dt} = -i[H(t),\rho(q,t)].$$
(2.3)

This equation is often written as $d\rho(q,t)/dt = \mathcal{L}(t)\rho(q,t)$, with the understanding that $\mathcal{L}(t)\sigma$ stands for $-i[H(t),\sigma]$. The operator \mathcal{L} is called as *Liouville super-operator*, since it acts on operators. The complete solution of the Liouville equation becomes $\rho(q,t) = \overleftarrow{T} \exp[-i \int_{t_0}^t ds \mathcal{L}(s)]\rho(q,t_0)$. In what follows, we will drop the explicit q-dependence and call the state $\rho(t)$.

2.2 Dynamics of open systems

An open system is a quantum system S interacting with another quantum system E called the environment with the underlying Hilbert spaces \mathcal{H}_S and \mathcal{H}_E , respectively [57, 214]. The combined system, which belongs to the space $\mathcal{H}_S \otimes \mathcal{H}_E$, is assumed to be *closed* obeying Hamiltonian dynamics. The interaction of system with its ambient environment leads to the development of correlations between them. As a consequence the state of the system alone can no longer be described by unitary Hamiltonian dynamics. One then extracts the *reduced dynamics* of the system from the unitary Hamiltonian dynamics of the combined system S + E with the total Hamiltonian

$$H(t) = H_S + H_E + H_{SE}.$$
 (2.4)

Here, H_S and H_E are the system and environment Hamiltonians, respectively, and H_{SE} describes the interaction between them.

At time t_0 , let the combined state be $\rho(t_0) \in \mathcal{H}_S \otimes \mathcal{H}_E$, at some later time t, we have $\rho(t) = U(t, t_0)\rho(t_0)U^{\dagger}(t, t_0)$ and the *reduced state*

$$\rho_S(t) = \operatorname{Tr}_E \left[\rho_{SE}(t) \right], \tag{2.5}$$

such that the definition of the partial trace ensures $\operatorname{Tr}_{SE}\left[(\mathcal{M}_S \otimes \mathbb{I})\rho_{SE}\right] = \operatorname{Tr}_S\left[\mathcal{M}_S \operatorname{Tr}_E[\rho]\right]$ for all observables \mathcal{M}_S . The equation of motion for the reduced state becomes

$$\frac{d}{dt}\rho_S(t) = -i\operatorname{Tr}_E\left[H(t),\rho(t)\right].$$
(2.6)

It is important to note that the unitary dynamics is reversible due to the symmetry, i.e., $U^{-1}(t) = U(-t)$. However, this reversibility no longer holds in an open system scenario as the dynamics contains a random element; a consequence of the system environment interaction.

2.3 Quantum measurement theory

The measurements in quantum mechanics play a dual role by telling us (i) how the state of a system changes after a measurement is performed on it, and (ii) how to prepare a system in a given state. Consider an observable A associated with the system under consideration. The quantum measurement on a quantum statistical ensemble Σ described by state ρ is then given by a set $\{A_m\}$ of the *measurement operators*, with the corresponding outcomes $\{\lambda_m\}$, satisfying the completeness condition $\sum_m A_m^{\dagger} A_m = \mathbf{1}$. The probability of obtaining λ_m is given by $p(\lambda_m) =$ $\text{Tr}[A_m A_m^{\dagger} \rho]$. The post measurement state

$$\rho' = \frac{A_m \rho A_m^{\dagger}}{\text{Tr}[A_m \rho A_m^{\dagger}]},\tag{2.7}$$

describes the sub-ensemble Σ' of systems for which observable A has been measured. It should be noted that Eq. (2.7) does not represent the most general form of measurement since it restricts the post-measurement state to be pure when the input state is pure. The most general mathematical abstraction of quantum measurement is known as *quantum instrument* proposed by Davies [215], such that $\rho_x = \frac{\mathcal{E}_x[\rho]}{\text{Tr}\{\mathcal{E}_x[\rho]\}}$, where $\{\mathcal{E}_x : x \in X\}$ is the set of trace non-increasing completely positive operators, and X is a countable set pertaining to the outcomes of the measurement.

For the special case of *projective measurements*, the measurement operators are given by *projectors* Π_m . The action on the state $\rho = |\psi\rangle\langle\psi|$ then results in $\rho' = [p(\lambda_m)]^{-1}\Pi_m(|\psi\rangle\langle\psi|)\Pi_m = |\phi\rangle\langle\phi|$, where $p(\lambda_m) = \text{Tr}[\Pi_m\rho\Pi_m]$ is the probability of obtaining the outcome λ_m . Thus the measured ensemble is represented by the normalized state vector $|\phi\rangle$. The post measurement state is not a pure state if the pre-measurement state ρ is mixed. However, if the eigenvalue λ_m is nondegenerate and the projectors are $\Pi_m = |\psi_m\rangle\langle\psi_m|$, then the measurement does lead to a pure state $|\phi\rangle = |\psi_m\rangle$, even if the pre-measurement state is mixed.

The generalized measurement theory is often described in terms of operations and effects and is based on the following concepts [57]:

- 1. The outcome λ_m of a measurement is a random number with probability distribution $p(\lambda_m) = \text{Tr}[E_m \rho]$, such that E_m is a positive operator called as *effect*, satisfying the normalization condition $\sum_m E_m^{\dagger} E_m = \mathbf{1}$, such that the total probability $\sum_m p(\lambda_m) = \mathbf{1}$.
- 2. An experimental situation where a measurement on an ensemble leads to splitting into various ensembles conditioned on a specific measurement outcome is called a *selective* measurement. The state corresponding to outcome λ_m is given by ρ'_m = E_m(ρ)/p(λ_m), where E_m is a positive superoperator also called as *operation*, obeying the condition Tr[E_m(ρ)] = Tr[E_mρ].
- 3. In an experimental scenario where the post measurement ensembles again mix, the measurement is called as *non-selective*. In this case, the post measurement state becomes $\rho' = \sum_m p(\lambda_m) \rho'_m = \sum_m \mathcal{E}_m(\rho)$.

2.4 Dynamical maps

Consider the composite state of the system and environment $\rho(0) = \rho_S(0) \otimes \rho_E$, such that ρ_E is fixed, that is, we assume that the system can be prepared in a given state independent of the environment. The evolution of the system state from time t = 0 to some later time t can be described as

$$\rho_S(t) = \mathcal{E}_{(t,0)}[\rho(0)] = \operatorname{Tr}_E \left[U(t)\rho_S(0) \otimes \rho_E U^{\dagger}(t) \right].$$
(2.8)

The map $\mathcal{E}_{(t,0)}$ is called as a dynamical map for a fixed time t such that it satisfies the following properties:

- 1. Complete positivity (CP): It means that not only \mathcal{E} is positive, but the combined operation $\mathcal{E} \otimes \mathbf{1}_n$, where $\mathbf{1}_n$ is the identity operator, is also positive for all dimensions n.
- 2. Trace preserving (TP): $\operatorname{Tr}[\mathcal{E}_{(t,0)}[\rho]] = \operatorname{Tr}[\rho]$, for all $\rho \in \mathcal{H}$.

The positivity and trace preserving is important for a map to connect an input density matrix to an output density matrix. The complete positivity is also a physically motivated axiom, since a combined operation $\mathcal{E} \otimes \mathbf{1}_n$ may be viewed as a local operation on first of the two widely separated

systems without influencing the second. A famous result by Kraus [216] is that any CP map can be written in the form

$$\rho_S(t) = K_\mu(t_1, t_0) \rho_S(t_0) K_\mu^{\dagger}(t_1, t_0).$$
(2.9)

The condition $\sum_{\mu} K_{\mu}^{\dagger} K_{\mu} = \mathbb{I}$ then implies that $\operatorname{Tr}[\rho_{S}(t)] = 1$ for any input state $\rho_{S}(t_{0})$.

2.4.1 Quantum channel

A quantum channel in the Schrödinger picture is a completely positive and trace preserving map $\Phi : \mathcal{T}(\mathcal{H}_A) \to \mathcal{T}(\mathcal{H}_B)$, where $\mathcal{T}(\mathcal{H}_A)$ and $\mathcal{T}(\mathcal{H}_B)$) denote the set of operators defined in the underlying Hilbert space \mathcal{H}_A and \mathcal{H}_B , respectively. A corresponding description in the Heisenberg picture would invoke the *dual* channel [109].

The operator sum representation of a channel is given as

$$\Phi[\boldsymbol{\rho}] = \sum_{\mu} \mathbf{M}_{\mu} \boldsymbol{\rho} \mathbf{M}_{\mu}^{\dagger}, \qquad (2.10)$$

such that the operators \mathbf{M}_{μ} , called as *Kraus operators*, obey the completeness condition, $\sum \mathbf{M}_{\mu}^{\dagger} \mathbf{M}_{\mu} =$

I. Here, *I* is the identity operator. Note that ρ , in Eq. (2.10), need not be a pure state. A linear map given in Eq. (2.10), is called a *quantum channel* or *superoperator* (as it maps operators to operators) or *completely positive trace preserving* (CPTP) map. A quantum channel is characterized by the following properties:

1. They are *linearity* transformations, that is, for states ρ_1 and ρ_2 , we have

$$\Phi[\alpha \rho_1 + \beta \rho_2] = \alpha \Phi(\rho_1) + \beta \Phi(\rho_2),$$

where α and β are complex numbers.

2. They represent Hermicity preserving maps, that is,

$$oldsymbol{
ho}=oldsymbol{
ho}^{\dagger}\implies \Phi[oldsymbol{
ho}]=\Phi[oldsymbol{
ho}]^{\dagger}$$

3. They preserve positivity, that is,

$$\boldsymbol{\rho} \ge 0 \implies \Phi[\boldsymbol{\rho}] \ge 0.$$

4. They are trace preserving, that is,

$$Tr(\Phi[\boldsymbol{\rho}]) = Tr(\boldsymbol{\rho}).$$

5. They are completely positive

$$I_{k \times k} \otimes \Phi[\rho] \ge 0, \quad \text{for all } k \tag{2.11}$$

2.5 Markovian and Non-Markovian processes

A precise definition of Markov process involves considering a random variable X with sample space $\{x_1, x_2, \ldots, x_n\}$. One can ask about the probability that any given value from

this sample space is attained at a given time, denoted by $p_1(x_j, t_j)$, where subscript reminds us that this is a one time probability. Similarly, one can ask the questions about joint probability $p_2(x_2, t_2; x_1, t_1)$, $p_3(x_3, t_3; x_2, t_2; x_1, t_1)$, and so on. Thus a stochastic process is described by an infinite hierarchy of probability. The conditional and joint probabilities are related as

$$p_n(x_j, t_j; x_{n-1}, t_{n-1}, \dots, x_1, t_1) = p_n(x_j, t_j | x_{n-1}, t_{n-1}, \dots, x_1, t_1) \\ \times p_{n-1}(x_{n-1}, t_{n-1}, \dots, x_1, t_1).$$
(2.12)

It is a formidable task to specify all the joint probabilities. If the probability of a random variable taking value x_n at time t_n is conditioned only to the values x_{n-1} at time t_{n-1} , that is, $p_n(x_j, t_j | x_{n-1}, t_{n-1}; \ldots; x_0, t_0) = p_2(x_n, t_n | x_{n-1}, t_{n-1})$, for all time *n*, then the process is called as *Markov process*. Therefore, under this assumption,

$$p_n(x_j, t_j; x_{n-1}, t_{n-1}, \dots, x_1, t_1) = \prod_{s=1}^{n-1} p_2(j_{s+1}, t_{s+1} | j_s, t_s) p_1(x_1, t_1).$$
(2.13)

A further simplification can be made in case the processes do not change statistically with time. Such processes are called *stationary* processes and obey the condition $p_2(x_k, t|x_j, t') = p_2(x_k, (t-t')|x_j, 0)$ (the right hand side is often written simply as $p_2(x_k, (t-t')|x_j)$). In other words, shifting the origin of time does not change the probabilities. This also implies that for one time probability, the time dependence disappears, i.e., $p_1(x_j, t) = p_1(x_j)$ must be time independent. For the stationary Markov processes

$$p_n(x_j, t_j; x_{n-1}, t_{n-1}, \dots, x_1, t_1) = \prod_{s=1}^{n-1} p_2(j_{s+1}, t_{s+1} - t_s | j_s) p_1(x_1).$$
(2.14)

For the stationary Markov processes, one can write the "chain equation" for the probability of going from any state x_j to a state x_k in time t as the probability of going from x_j to x_l in time t' and then from x_l to x_k in the remaining time t - t' (0 < t' < t) by summing over all the intermediate paths

$$p(x_k, t|x_j) = \sum_{l=1}^{N} p(x_k, t - t') p(x_l, t'|x_j).$$
(2.15)

This is known as Chapman–Kolmogorov equation. It is often desirable to avoid the nonlinearity of this equation by considering the probability for the scenario involving small time difference, i.e., when $t - t' = \delta t$. In this case, one expects $p(x_k, \delta t | x_j) = w(x_k | x_j) \delta t$, where $w(x_k | x_j)$ is the transition probability per unit time or transition rate. Note that this is true even for the non stationary processes where one would have $p(x_k, t + \delta t | x_j)$ and should be proportional to δt when δt is small. However, the transition rate in this case will depend on time $w(x_k | x_j; t)$. One obtains the following "Master equation"

$$\frac{dp(x_k, t|x_j)}{dt} = \sum_{l=1, x_l \neq x_k}^{N} [w(x_k|x_l)p(x_l, t|x_j) - w(x_l|x_k)p(x_k, t|x_j)].$$
(2.16)

2.5.1 Theory of Markovian open quantum systems

The theory of open quantum systems constitutes the study of quantum systems in presence of ambient environment. The environment is indeed fundamentally quantum mechanical in nature, however, under certain circumstances it is useful to treat it classical, stochastically varying driving terms in the Hamiltonian of the system. Either way, one inevitably deals with a master equation, the equation of motion for state of the system $\rho_S(t) = \text{Tr}[\rho_{SE}(t)]$, which upon using Nakajima-Zwanzig projection technique, is given by

$$\frac{d}{dt}\rho_S(t) = -i[H_S, \rho_S(t)] + \int_{t_0}^t \mathcal{K}_{t,t'}[\rho_S(t')]dt'.$$
(2.17)

Here, H_S denotes the system Hamiltonian in the absence of system environment interaction. Also, $\mathcal{K}_{t,t'}$, called as *memory kernel*, is a linear map which describes the effects of environment on the system. The notion of Markovian dynamics as traditionally advocated in quantum statistical mechanics involves two assumptions [57, 58, 217]:

1. Born-Markov approximation: This involves neglecting the memory effects by approximating the memory kernel as

$$\mathcal{K}_{t,t'}[\rho_S(t')] = \mathcal{K}\delta(t-t')[\rho_S(t')]$$
(2.18)

2. Rotating wave approximation: Here, the fast rotating terms in the memory kernel are ignored.

When the timescale of system environment interaction is compared to timescales of environment, the system couples to all the frequencies of environment (white noise scenario). This happens when the system environment coupling is weak and leads to the Markovian dynamics.

A simple quintessential example of a Markov process of an open system dynamics is the one for which all memory effects are neglected and the dynamics is *stationary* in time. Under these conditions, the family of maps has quantum dynamical semigroup (QDS) structure [218]

$$\Phi_{(t_2+t_1,0)} = \Phi_{(t_2,0)}\Phi_{(t_1,0)}, \qquad t_1, t_2 \ge 0.$$
(2.19)

Such a quantum dynamical semigroup can be associated with a generator \mathcal{L} , defined as $\Phi_{t,0} = \exp(-\mathcal{L}t)$ [59] which governs the dynamics of the reduced state, i.e., $d\rho_S(t)/dt = \mathcal{L}\rho_S(t)$. The most general form of \mathcal{L} is given by

$$\mathcal{L}\rho_S = -i[H,\rho_S] + \sum_i \gamma_i \left(A_i \rho_S A_i^{\dagger} - \frac{1}{2} \{ A_i^{\dagger} A_i, \rho_S \} \right).$$
(2.20)

Here, H is the Hamiltonian of the system, A_i 's are the system jump operators and $\gamma_i \ge 0$ the corresponding decay rates. The equation of the form $d\rho_S(t)/dt = \mathcal{L}\rho_S(t)$ with generator given by Eq. (2.20) are often known as Markovian master equation.

The master equations for subsystem dynamics are usually obtained either phenomenologically or derived with microscopic approaches under various approximations [57]. It is possible that the assumptions made in the derivation may lead to unphysical equation of motion. However, a powerful theorem proved in [219] says that irrespective of how one derives the master equation, if the generator is of the form of Eq. (2.20), it is guaranteed to lead to physically consistent and feasible solutions, i.e., it generated a family of CP maps. No such elegant mathematical framework is available for a general non-Markovian process.

2.5.2 Theory of non-Markovian open quantum systems

The Lindblad master equation represents the universally accepted prototype of Markovian dynamics, the memoryless dynamics implied by the semi-group property which make the future

evolution of the system independent of the past states. However, there is divided opinion on what is *not* Markovian and, to date, various points of view have been advocated, and as a result each non-Markovian map is non-Markovian in its own way. This subsection is devoted to a quantitative description of non-Markovianity by various existing measures.

The semi-group property defined in Eq. (2.19) is a characteristic feature of all master equations in the Lindblad form. Two independent studies reported by Lindblad in [219] and V. Gorini, A. Kossakowski, E.C.G. Sudarshan in [220] proved one-to-one correspondence between CPTP dynamical maps with semi-group property and master equations in the Lindblad form. In [76], it was proposed that any deviation from semi-group property should be considered the principal characteristic of non-Markovian dynamical maps, and constructed a quantitative measure of non-Markovianity as the minimum amount of isotropic noise needs to be added to the dynamics of open system to make it Markovian. Recently, the concept of *self-similarity* was defined and non-Markovian behaviour was quantified as the departure from *self-similarity* [221].

The time independence of the Lindbladian leads to the master equation of the form $d\Phi/dt = \mathcal{L}\Phi$, with the solution $\Phi_{(t_j,t_i)} = \exp[\mathcal{L}(t_j - t_i)]$ which represents a family of completely positive trace preserving (CPTP) maps for all $t_j > t_i$. The stationarity is implied by the dependence of the evolution on the time difference $t_j - t_i$ such that the maps form a one parameter semigroup, Eq. (2.19). When the system environment time scales are smaller than the environment time scales, the system couples to the environment at certain frequencies [94]. As a result, the generator \mathcal{L} , Eq. (2.20), acquires additional time dependences in the form of $H_S(t)$, $\gamma_i(t)$, and $A_i(t)$ [222]. A non trivial time dependence of $\gamma_i(t)$ on time breaks the QDS structure, Eq. (2.19), of the maps; however, if $\gamma_i \geq 0$, the map is CP divisible and represents the *time-dependent Markov processes*. The resulting dynamics is given in terms of a two parameter family of CPTP maps with time inhomogenous composition law $\Phi_{t,t_0} = \Phi_{t,t'}\Phi_{t',t_0}$, for $t > t' > t_0$, with the map Φ_{t,t_0} obeying the master equation $d\Phi_{t,t_0}/dt = \mathcal{L}\Phi_{t,t_0}$. It turns out that the full map Φ_{t,t_0} depends on the time difference $t - t_0$ but violates the semigroup property, Eq. (2.19), because the intermediate map $\Phi_{t',t_0} \neq \Phi(t' - t_0)$.

2.6 Quantum correlations

In this section, we describe various quantum correlations, both temporal as well as spatial, which have been analyzed in various systems, as discussed in this thesis.

2.6.1 Temporal quantum correlations

Temporal correlations exist between the outcomes of an observable measured on a single system at different times. In this thesis, we have analyzed a family of temporal quantum correlations which go by the name of Leggett-Garg inequalities (LGIs). Here, we provide a brief description of various avatars of LGIs.

Standard Leggett-Garg inequality (SLGI): Leggett Garg inequalities are based on the concept of macrorealism (MR) and non-invasive measurability (NIM). MR means that the system which has available to it two or more macroscopically distinct states, pertaining to an observable $\hat{M}(t)$, always exists in one of these states irrespective of any measurement performed on it. NIM states that, in principle, we can perform the measurement without disturbing the future dynamics of the system [23]. To derive a simple form of the SLGI, let the observable $\hat{M}(t)$ be dichotomic, i.e., it takes values $m_i = \pm 1$ at time t_i (dichotomicity is not necessary here, but is invoked for simplification. In fact, the results obtained hold true provided \hat{M} is bounded, i.e., $|\hat{M}| < 1$ [23, 223, 224]). The measurement of this observable is performed on a single system at different times $t_1 < t_2 < t_3$. After a series of measurements, one can estimate the value of the two time correlation function

$$C(t_i, t_j) = \frac{1}{N} \sum_{s=1}^{N} m_i^{(s)} m_j^{(s)}.$$
(2.21)

Here, $m_i^{(n)}$ (or $m_j^{(n)}$) is the outcome of *n*-th measurement of $\hat{M}(t_i)$ (or $\hat{M}(t_j)$). The MR assures that the system exists in a well defined state even when no measurement is performed on it. As a consequence, one can say the joint probability of outcomes m_1 , m_2 and m_3 is determined *a priori* at some initial time say t_0 , say $P(m_1, m_2, m_3)$. Further, the two time probability can be obtained as a marginal $P_{ij}(m_i, m_j) = \sum_{k,k \neq i,j} P_{ij}(m_1, m_2, m_3)$, where the indices i, j remind us that only two observables are measured. In general, $P_{12}(m_1, m_2, m_3)$, $P_{23}(m_1, m_2, m_3)$ and $P_{13}(m_1, m_2, m_3)$ are different since the measurements at different times may affect the dynamics differently. However, invoking NIM one assumes that $P_{ij}(m_1, m_2, m_3) = P(m_1, m_2, m_3)$ for all i, j. Therefore, we have $P(m_1, m_2) = \sum_{m_3=\pm 1} P(m_1, m_2, m_3)$, $P(m_2, m_3) = \sum_{m_1=\pm 1} P(m_1, m_2, m_3)$ and $P(m_1, m_3) = \sum_{m_2=\pm 1} P(m_1, m_2, m_3)$. The correlation functions are written in terms of joint probabilities $P(m_i, m_j)$

$$C(t_i, t_j) = P(m_i = +1, m_j = +1) - P(m_i = +1, m_j = -1) - P(m_i = -1, m_j = +1) + P(m_i = -1, m_j = -1).$$
(2.22)

We define the Leggett Garg parameter K_3 as

$$K_3 = C(t_1, t_2) + C(t_2, t_3) - C(t_1, t_3).$$
(2.23)

The subscript in K_3 reminds us that we are considering three measurements, made at t_1 , t_2 and t_3 . We have

$$K_3 = 1 - 4 \left[P(m_1 = 1, m_2 = -1, m_3 = 1) + P(m_1 = -1, m_2 = 1, m_3 = -1) \right].$$
(2.24)

The sum of the probabilities inside the bracket can range from 0 to 1, therefore we have the simplest SLGI given by $-3 \le K_3 \le 1$. The maximum quantum value of K_3 for a two level system in 3/2 [17] and has been found to hold for any system, *irrespective* of the number of levels, as long as the measurements are given by just two projectors Π^{\pm} [225], a fact revealed in several studies [22, 226–228]. One can talk about n measurement scenario leading to more SLGIs: $-n \le K_n \le n-2$ for $n \ge 3$ (and odd), and $-(n-2) \le K_n \le n-2$, for $n \ge 4$ (and even) [23]. It was shown in [229] that in the limit $N \to \infty$, the SLGI can be violated up to its maximum algebraic sum.

Stationarity based Leggett-Garg inequality: It is often convenient to deviate from the original formulation of LGI and study instead a variant form of it, known as Leggett-Garg type inequalities (LGIs) introduced in [38, 230–232] and experimentally verified in [31, 233]. These inequalities were derived to avoid the requirement of noninvasive measurements at intermediate times. This feature makes them more suitable for the experimental verification as compared to LGIs. The assumption of NIM is replaced by a weaker condition known as stationarity. This asserts that the conditional probability $p(\phi, t_j | \psi, t_i)$ that the system is found in state ϕ at time t_j given that it was in state ψ at time t_i is a function of the time difference $(t_j - t_i)$. Invoking stationarity leads to the following form of LGIs

$$K_{\pm} = \pm 2C(t_1, t) - C(t_1, 2t) \le 1.$$
(2.25)

Here, $t = t_3 - t_2 = t_2 - t_1$, is the time between two successive measurements. Though the assumption of stationarity helps to put the inequalities into easily testable forms, it reduces the class

of macrorealist theories which are put to the test [38]. The stationarity condition holds provided the system can be prepared in a well-defined state and the system evolves under unitary or Markovian dynamics.

Wigner and CHSH forms of Leggett-Garg inequality: From the assumptions of joint probability and non-invasive measurability, we obtain the pairwise statistics of measurement of \hat{M}_2 and \hat{M}_3 having outcome m_2 and m_3 as $P(m_2, m_3) = \sum_{m_1=\pm} P(m_1, m_2, m_3)$ and similarly for others. We can write the expression, $P(-m_1, m_2) + P(m_1, m_3) - P(m_2, m_3) = P(-m_1, m_2, -m_3) + P(m_1, -m_2, m_3)$. By invoking the non-negativity of the probability, Wigner form of LGIs can be derived as

$$P(m_2, m_3) - P(-m_1, m_2) - P(m_1, m_3) \le 0.$$
(2.26)

One can obtain eight variants Wigner form of LGIs from (6.65). Similarly, sixteen more inequalities can be derived from

$$P(m_1, m_3) - P(m_1, -m_2) - P(m_2, m_3) \le 0,$$
(2.27)

$$P(m_1, m_2) - P(m_2, -m_3) - P(m_1, m_3) \le 0.$$
(2.28)

Thus one has twenty four variants of Wigner form of LGI characterized by different measurement settings. This richness turns out to be very useful especially in systems where experimental constraints put limitation on arbitrary preparation and detection process, viz., in subatomic systems like neutrinos and mesons. It has been recently shown that Wigner form of LGIs are stronger than the standard LGIs [100, 234].

The single marginal statistics of the measurement of the observable, for example, probability of getting outcome, when M_2 measurement is performed can be obtained as $P(m_2) = \sum_{m_1,m_3=\pm} P(m_1,m_2,m_3)$ and similarly for $P(m_1)$ and $P(m_3)$. By combining single and pairwise statistics, we can get the expression, $P(m_1,m_3) + P(m_2) - P(m_1,m_2) - P(m_2,m_3) = P(m_1,-m_2,m_3) + P(-m_1,m_2,-m_3)$, which gives

$$P(m_1, m_2) + P(m_2, m_3) - P(m_1, m_3) - P(m_2) \le 0.$$
(2.29)

Inequality (2.29) can lead to eight variants of Clauser-Horne form of LGIs [234]. Similarly, sixteen more inequalities can be derived in this manner. In compact notation, we can write,

$$P(m_1, m_3) + P(m_1, m_2) - P(m_2, m_3) - P(m_1) \le 0,$$
(2.30)

$$P(m_1, m_3) + P(m_2, m_3) - P(m_1, m_2) - P(m_3) \le 0.$$
(2.31)

Note that in the Wigner form of LGIs only pair-wise probabilities are involved but in Clauser-Horne form of LGIs single probabilities are also involved along with pair-wise ones. Wigner and Clauser-Horne forms of LGIs can be shown to be equivalent to standard LGIs in macrorealist model, but inequivalent in quantum theory [234].

Entropic Leggett-Garg inequality: We now provide a brief review of some rudiments of information theory used in the development of the entropic Leggett-Garg inequality. We begin

by considering the observable A which can take discrete values denoted by a_i at time t_i , that is, $A(t_i) = a_i$. We define the joint probability of the measurement of A at times t_i and t_j giving results a_i and a_j , respectively, as $P(a_i, a_j)$. According to Bayes's theorem the joint probability is related to the conditional probability as,

$$P(a_i, a_j) = P(a_j | a_i) P(a_i) = P(a_i | a_j) P(a_j).$$
(2.32)

Here, $P(a_j|a_i)$ is the conditional probability of obtaining the outcome a_j at time t_j , given that a_i was obtained at time t_i .

A classical theory can assign well defined values to all observables of the system with no reference to the measurement process. This assumption lies at the heart of Bell and Leggett-Garg inequalities, leading to bounds which may not be respected by the nonclassical systems. In other words, this assumption demands a joint probability distribution, $P(a_i, a_j)$, yielding information about the marginals of individual observations at time t_i . The assumption of non-invasive measurability implies that the measurement made on a system at any time does not disturb its future dynamics and hence any measurement made at a later time t_j where $t_j > t_i$. The mathematical statement would be that the joint probabilities be expressed as a convex combination of the product of probabilities $P(a_i|\lambda)$, averaged over a hidden variable probability distribution $\rho(\lambda)$ [36, 235, 236]:

$$P(a_1, a_2, \dots, a_n) = \sum_{\lambda} \rho(\lambda) P(a_1|\lambda) P(a_2|\lambda) \cdots P(a_n|\lambda), \qquad (2.33)$$

such that the following properties are satisfied

$$0 \le \rho(\lambda) \le 1$$
, $\sum_{\lambda} \rho(\lambda) = 1$; $0 \le P(a_i|\lambda) \le 1$, $\sum_{\lambda} P(a_i|\lambda) = 1$. (2.34)

A close resemblance of Eq. (2.33) may be noted to the corresponding assumption made in the derivation Bell inequality [1] with the difference that in later case one talks about the joint probabilities of outcomes in spatially separated systems. One can use the conditional probability given by Eq. (2.32) to define the conditional entropy as

$$H[A(t_j)|A(t_i)] = -\sum_{a_i, a_j} P(a_j|a_i) \log_2 P(a_j|a_i).$$
(2.35)

Now using chain rule and the fact that conditioning reduces entropy [237], one obtains [238]

$$H[A(t_{N-1}), ..., A(t_0)] \le H[A(t_{N-1})|A(t_{N-2})] + ... + H[A(t_1)|A(t_0)] + H[A(t_0)].$$
(2.36)

This temporal entropic inequality was used in [238] to study the role of quantum coherence in Grover's algorithm. Using the relation $H[A(t_i), A(t_{i+j})] = H[A(t_{i+j})|A(t_i)] + H[A(t_i)]$, one can derive the temporal analogues of the spatial entropic Bell inequalities

$$\sum_{k=1}^{N-1} H[A(t_k)|A(t_{k-1})] - H[A(t_{N-1})|A(t_0)] \ge 0.$$
(2.37)

These are the entropic Leggett-Garg inequalities [239]. Here, N denotes the number of measurements, inclusive of the preparation; the case of N = 3 was experimentally tested in [35].

2.6.2 Spatial quantum correlations

Here, we discuss some of the important quantum correlations existing between spatially separated systems. The simplest scenario for realizing correlations is by considering a bipartite system consisting of subsystems A and B, with respective Hilbert spaces \mathcal{H}_A and \mathcal{H}_B . The tensor product of these spaces, i.e., $\mathcal{H}_A \otimes \mathcal{H}_B$ harbors the states of the total system.

Entanglement: If the two subsystems are completely independent, then the composite state (of the total system) is a tensor product of the states of subsystems, i.e., if $|\psi_A\rangle \in \mathcal{H}_A$ and $|\psi_B\rangle \in \mathcal{H}_B$, then $|\psi_{AB}\rangle \in \mathcal{H}_A \otimes \mathcal{H}_B$, such that $|\psi_{AB}\rangle = |\psi_A\rangle \otimes |\psi_B\rangle$. In this case, no correlations of any kind (neither classical nor quantum) exist between the subsystems. However, it may not be possible to write the composite state as a product of two states of the respective subsystems. In this case $|\psi_{AB}\rangle$ describes an *entangled* state of the two subsystems. In a pure bipartite scenario, entanglement manifests in different yet equivalent ways. For example, all pure entangled states are non-local and hence can violate Bell's inequality [240]. Further, every pure entangled state is definitely disturbed by an action of any possible local measurement [5, 241].

Steering: Steering is the manipulation of the state of one subsystem by performing local operations on the other. It captures the original notion of inseparability which was appreciated by Schrödinger [136], and was recently formalized from the quantum information theoretic point of view [242]. Steering is an example of an asymmetric correlation which means that A can steer B but not the other way around. A recent review on steering [243] summarizes the important developments in this direction.

Nonlocality: Nonlocality provides an example of the most radical departure from a classical description of the world. It says that the probabilities of outcomes of the measurements performed on subsystems cannot be generated from classical correlations. It is a symmetric correlation like entanglement, i.e., invariant under the swap of the subsystems A and B [244].

2.7 Nonclassical properties of light

We now briefly revisit some important notions relevant for understanding the nonclassical nature of light. An important property in this context is the *coherence*, i.e., the ability to show *interference*, and is encountered both in classical as well as in quantum description of light. Recall that the interference in the Mach Zehnder or Michealson interferometric setups, where interference means that there is a well defined correlation between the field at two different times, is an example of *temporal coherence*. However, from the perspective of quantum mechanics, the coherence is described as interference of two *processes* associated with photons. The two processes are associated with wave amplitudes ψ_1 and ψ_2 . If the processes are *indistinguishable*, the total amplitude for the detection is the sum $\psi = \psi_1 + \psi_2$. Consequently the probability $|\psi|^2$ contains the cross term $Re[\psi_1\psi_2^*]$. The degree to which we see the interference is proportional to the degree to which the two processes are indistinguishable. It is worth mentioning that the interaction of such systems with the environment can increase the distinguishablity of these processes and thereby destroy the ability of the system to show interference, a process known as *decoherence*.

To better appreciate the nonclassical features of light, we discuss in a nutshell, the quantization of electromagnetic field. Recall that the classical field can be decomposed in terms of plane waves in three dimensions $\vec{u}_{\vec{k},\mu}(\vec{r}) = \hat{\epsilon}_{\vec{k},\mu} e^{i\vec{k}\cdot\vec{r}}/\sqrt{V}$ with complex amplitudes $\alpha_{\vec{k},\mu}(t) =$

 $e^{-i\nu_k t} \alpha_{\vec{k}}(0)$ of normal modes as:

$$\vec{E}(\vec{r},t) = \sum_{\vec{k}} \mathcal{E}_{\vec{k}} \, \alpha_{\vec{k},\mu}(t) \, \vec{u}_{\vec{k},\mu}(\vec{r}) + c.c., \tag{2.38}$$

Here, *c.c.* denotes the complex conjugate, $\mathcal{E}_{\vec{k}} = \sqrt{\hbar \nu_k / 2\epsilon_0}$ is a normalization constant, and $\hat{\epsilon}_{\vec{k},\mu}$ is a unit polarization vector where μ labels two orthogonal polarization directions in plane perpendicular to \vec{k} . The sum is taken over an infinite discrete set of values of wave vector $\vec{k} = (k_x, k_y, k_z)$, such that the periodic boundary conditions require $k_i = 2\pi n_i/L$, (i = x, y, z). Each triplet (n_x, n_y, n_z) defines a mode of the electromagnetic field.

The quantized field is achieved by identifying $\alpha_{\vec{k}}$ and $\alpha_{\vec{k}}^*$ with the harmonic oscillator operators $a_{\vec{k}}$ and $a_{\vec{k}}^{\dagger}$, respectively. The quantized electric field takes the form [144]

$$\hat{E}(\vec{r},t) = \sum_{\vec{k},\mu} \hat{\epsilon}_{\vec{k}} \mathcal{E}_{\vec{k}} \ \hat{a}_{\vec{k}} \ e^{-i\nu_k t + i\vec{k}.\vec{r}} + h.c.$$
(2.39)

The operator $\hat{a}_{\vec{k}} (\hat{a}_{\vec{k}}^{\dagger})$ annihilates (creates) a photon - an excitation in the plane wave mode $\hat{\epsilon}_{\vec{k}} e^{-i\nu_k t + i\vec{k}.\vec{r}}$. The commutation relation $[\hat{a}_{\vec{k}}, \hat{a}_{\vec{k}'}^{\dagger}] = \delta_{\vec{k},\vec{k}'} \delta_{\mu,\mu'}$ assures that different modes commute with each other. A useful picture to look at fields is in terms of Fock space $\mathcal{H}_{Fock} = \bigotimes_{n=0}^{\infty} h_{n_k}$ spanned by

$$\{|n_1, n_2, \dots, n_k, \dots\rangle : n_k = 0, 1, 2, \dots \infty\},\$$

such that n_k number of particles are in the mode labeled by (composite) index $k = (\vec{k}, \mu) = 0, 1, 2, \ldots$ This "discrete variable" description is intimately related to the particle picture, i.e., associates particle degrees of freedom to the field. Each constituent space h_{n_k} , which is "simple harmonic oscillator" Hilbert space, is spanned by $\{|n_k\rangle : n_k = 0, 1, 2 \ldots \infty\}$ with

$$|n_k\rangle = \frac{a_{n_k}^{\dagger}}{\sqrt{n_k!}} |0\rangle \,. \tag{2.40}$$

It follows from the above equation that a single photon is not necessarily in one mode, e.g., a single photon at the input of a beam-splitter leads to the output which is a superposition of two plane waves $e^{-i\vec{k}_a.\vec{r}}$ and $e^{-i\vec{k}_b.\vec{r}}$, $|\psi_{out}\rangle = (t\hat{a}^{\dagger}_{\vec{k}_a} + r\hat{a}^{\dagger}_{\vec{k}_b})|0\rangle$, where t(r) is the transmission (reflection) coefficient. Let us now revisit various witnesses of nonclassicality of electromagnetic field in terms of the field creation and annihilation operators discussed above.

2.7.1 Criteria for nonclassicality of light

Nonclassicality is a multifaceted entity. From the perspective of quantum optics there are different witnesses of nonclassicality of the radiation field. For example, the Mandel parameter $Q_M < 0$, gives a sufficient condition for the field to be nonclassical [143]; single and multimode squeezing conditions reveal the nonclassical character of a state arising due to the field fluctuation [133]; Hillery-Zubairy criteria provide sufficient conditions in the form of a family of inequalities for detecting entanglement [245]. These criteria can be casted in terms of the bosonic creation and annihilation operators as discussed below.

• The Mandel Q_M parameter: Defined as the normalized variance of the boson distribution, this measure characterizes the nonclassicality of a radiation field in the context of the photon number distribution. Quantitatively,

$$Q_M = \frac{\langle (a^{\dagger}a)^2 \rangle - \langle a^{\dagger}a \rangle^2 - \langle a^{\dagger}a \rangle}{\langle a^{\dagger}a \rangle}.$$
(2.41)

Since the minimum value of $\langle (a^{\dagger}a)^2 \rangle - \langle a^{\dagger}a \rangle^2$ is zero, the Mandel parameter has a lower bound of -1, and it provides the criterion for observing different photon statistics as follows:

$$Q_M \begin{cases} < 0 & \text{sub} - \text{Poissonian field,} \\ = 0 & \text{coherent (Poissonian) field,} \\ > 0 & \text{super} - \text{Poissonian field.} \end{cases}$$
(2.42)

• *Antibunching*: A closely related phenomena is photon antibunching, given usually in terms of the two-time light intensity correlation function [246],

 $g^{(2)}(\tau) = \langle n_1(t)n_2(t+\tau) \rangle / \langle n_1(t) \rangle \langle n_2(t+\tau) \rangle$, where $n_i(t)$ is the number of counts registered on *i*th detector at time *t*. A quantum state is referred to as an antibunched if $g^{(2)}(0) < g^{(2)}(\tau)$. Interestingly, it was shown in the past to be closely related to the Mandel parameter [247]. The correlation $g^{(2)}(0)$ characterizes the antibunched, the coherent and the bunched fields as:

$$g^{(2)}(0) \begin{cases} < 1 & \text{antibunched,} \\ = 1 & \text{coherent,} \\ > 1 & \text{bunched.} \end{cases}$$
(2.43)

Therefore, for a single field with annihilation operator a, the criterion for antibunching can also be written as [248]

$$\mathcal{A}_a = \langle a^{\dagger 2} a^2 \rangle - \langle a^{\dagger} a \rangle^2 < 0, \tag{2.44}$$

i.e., the negative values of Mandel parameter also establish antibunching. Further, the intermodal antibunching is witnessed by using the following criterion [157]

$$\mathcal{A}_{ab} = \langle a^{\dagger}b^{\dagger}ba \rangle - \langle a^{\dagger}a \rangle \langle b^{\dagger}b \rangle < 0.$$
(2.45)

• Squeezing: This measure delineates the nonclassicality of a field in the context of the fluctuations in the quadratures X_a and Y_a of the field (with annihilation operator a), defined as

$$X_a = \frac{a+a^{\dagger}}{2} \qquad Y_a = \frac{a-a^{\dagger}}{2i}.$$
 (2.46)

The criteria for the nonclassical signature in the field is given, in terms of the variances in the quadratures, as follows [133]

$$\langle X_a^2 \rangle - \langle X_a \rangle^2 = (\Delta X_a)^2 < \frac{1}{4}$$
(2.47)

or

$$\langle Y_a^2 \rangle - \langle Y_a \rangle^2 = (\Delta Y_a)^2 < \frac{1}{4}.$$
(2.48)

We can also define the intermodal quadrature operators $X_{ab} = (a + a^{\dagger} + b + b^{\dagger})/2\sqrt{2}$ and $Y_{ab} = (a - a^{\dagger} + b - b^{\dagger})/2i\sqrt{2}$, such that the intermodal squeezing criterion is given by

or

$$(\Delta X_{ab})^2 < \frac{1}{4} \tag{2.49a}$$

$$(\Delta Y_{ab})^2 < \frac{1}{4}.$$
 (2.49b)

• Duan el al.'s criterion of entanglement: For two systems A and B, the non-separability means the impossibility of factorizing the density matrix of the combined system ρ as $\rho = \sum_k \lambda_k \rho_A^k \rho_B^k$, with $\sum_k \lambda_k = 1$. In [249], a criterion for inseparability was developed by Duan et al., which provides a sufficient condition for the entanglement of any two party continuous variable states [250]. For two radiation fields with annihilation operators a and b, this criterion translates to

$$\mathcal{D}_{ab} = 4(\Delta X_{ab})^2 + 4(\Delta Y_{ab})^2 - 2 < 0, \qquad (2.50)$$

where $(\Delta X_{ab})^2$ and $(\Delta Y_{ab})^2$ are defined in Eq. (2.49). The presence of squeezing does not ensure the existence of entanglement as at a given time squeezing can happen only in one quadrature. Thus, this criterion captures the asymmetry in the fluctuations in X and Y and this is why it's studied independently. In what follows, we refer to this criterion of entanglement as Duan's criterion.

• *Hillery-Zubairy(HZ) criteria of entanglement*: In [245], it was shown that for two field modes *a* and *b*, two inseparability criteria are

$$\mathcal{E}_{ab} = \langle a^{\dagger}ab^{\dagger}b \rangle - |\langle ab^{\dagger} \rangle|^2 < 0, \qquad (2.51)$$

and

$$\tilde{\mathcal{E}}_{ab} = \langle a^{\dagger}a \rangle \langle b^{\dagger}b \rangle - |\langle ab \rangle|^2 < 0.$$
(2.52)

• Steering: The notion of steering, as an apparent action at a distance, was introduced by Schrödinger while discussing the EPR paradox [136], and shares logical differences both with non-separability and Bell non-locality. While as non-separability and Bell non-locality are symmetric between two parties, say Alice and Bob, steering is inherently asymmetric, addressing whether Alice can change the state of Bob's system by applying local measurements. An operational definition of steering was first provided in [242], wherein they proved that steerable states are a strict subset of the entangled states and a strict superset of the states that can exhibit Bell non-locality. In the context of field modes a and b, the EPR - steering entanglement is confirmed if it satisfies [251]

$$0 < 1 + \frac{\langle a^{\dagger}ab^{\dagger}b \rangle - |\langle ab^{\dagger} \rangle|^2}{\langle a^{\dagger}a(bb^{\dagger} - b^{\dagger}b) \rangle} < \frac{1}{2}.$$
(2.53)

This result can be proved by the methods given in [252]. The above steering condition (2.53) can be expressed in terms of the HZ criterion Eq. (2.51), the condition reads:

$$\mathcal{S}_{AB} = \mathcal{E}_{ab} + \frac{\langle a^{\dagger}a \rangle}{2} < 0.$$
(2.54)

The concept of steering being inherently asymmetric [253], it will be interesting to compare S_{AB} and $S_{BA} = \mathcal{E}_{ab} + \frac{\langle b^{\dagger}b \rangle}{2}$.

• *Multimode entanglement*: In [254], a class of inequalities was derived for detecting the entanglement in multimode systems. In the case of a tripartite state, viz., the one corresponding to the three modes a, b, and c, the sufficient conditions for not being bi-separable of the form ab|c (in which a compound mode ab is entangled with mode c), are given as follows:

$$E_{ab|c} = \langle a^{\dagger}ab^{\dagger}bc^{\dagger}c \rangle - |\langle abc^{\dagger} \rangle|^2, \qquad (2.55)$$

$$E'_{ab|c} = \langle a^{\dagger}ab^{\dagger}bc^{\dagger}c \rangle - |\langle abc \rangle|^2.$$
(2.56)

A three-mode quantum state is fully entangled by the satisfaction of either or both of the following sets of inequalities:

$$E_{ab|c} < 0, \quad E_{bc|a} < 0, \quad E_{ac|b} < 0, \tag{2.57}$$

$$E'_{ab|c} < 0, \quad E'_{bc|a} < 0, \quad E'_{ac|b} < 0.$$
 (2.58)

It is worth mentioning here that the analysis of the above mentioned witnesses of nonclassicality may involve higher order products of the operators. These higher order correlations can be decorrelated by the prescription given in [171]. For example $\langle \hat{a}\hat{b}\hat{c}\rangle \approx \langle \hat{a}\hat{b}\rangle \langle \hat{c}\rangle + \langle \hat{a}\rangle \langle \hat{b}\hat{c}\rangle + \langle \hat{a}\hat{c}\rangle \langle \hat{b}\rangle - 2\langle \hat{a}\rangle \langle \hat{b}\rangle \langle \hat{c}\rangle$, which basically makes use of the Bogoliubov theory of linearized quantum corrections to mean field effects.

We now proceed to investigate in detail various facets of nonclassicality in different systems and will often refer to the tools developed in this chapter.