

## TWO NOVEL PURE-STATE COHERENCE MEASURES IN QUANTIFYING COHERENCE

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Received May 16, 2024

Revised October 7, 2024

Quantification is one of the primary goals of quantum coherence resource theory. Here, we put forward two coherence measures, analytically prove their validity in the pure-state regime following the axiomatic definition of a legitimate pure-state coherence measure (PSCM), and provide their general normalized forms. We further discuss their extensions in the domain of mixed states with the assistance of the convex roof of coherence, and top coherence (quantification in terms of pure state coherence) and define their classes (coherence *monotone* or *measure*); in this regard, we thoroughly investigate the top coherence class. For the quantitative demonstration, we pick up different laser pulse-two-qubit interaction scenarios and compare the evolutions of these two coherence measures along with  $l_1$ -norm of coherence and relative entropy of coherence; this study also signifies the importance of coherence in probing an atomic or molecular system.

*Keywords:* Quantifying Coherence, Pure State Coherence Measure (PSCM), mixed-state coherence, Convexity of Top Coherence, Laser-Qubit Interaction

### 1 Introduction

One essential aspect of quantum theory is coherence, which is represented by the superposition of the quantum levels in a fixed reference frame. In quantum optics, the quest for coherence began as a significant quantum resource that was being quantitatively characterized in terms of phase space distributions and multi-point correlation functions [1], [2], [3], techniques that are rooted in classical electromagnetic theory. However, coherence is a general quantum resource; it emphasizes most of the quantum features, such as quantum correlation [4], [5], entanglement [6], [7], [8], and symmetry [9]. The resource theory of coherence [10], [11] has recently emerged as a means of effectively utilizing coherence as a resource in general (beneficial to any quantum field). It provides a robust mathematical framework for quantifying coherence [12], [13] and accounts for quantum advantages in numerous fields, such as quantum thermodynamics [14], [15], [16], [17], [18], quantum metrology [19], [20], [21], quantum cryptography [22], [23], quantum biology, [24], [25], etc. It also creates a framework for in-depth examination of the impact of coherence in fundamental physics [26].

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One of the main objectives of any coherence resource theory is to quantify state coherence. In this sense, it is crucial to look for novel coherence measures since they add new computability as well as broaden the operational area of a coherence resource theory [10], [12], [27], [28], [29], [30], [31], [32], [33]. A coherence quantifier should satisfy some or all the requirements proposed by Baumgratz et. al., in 2014 [12], depending on free operations and free states. In the context of *standard coherence resource theory* (where the *incoherent operation* (IO) class and its associated set of *incoherent states* are the free operations and free states, respectively) [12], [10], these criteria are the following: *non-negativity*, *monotonicity* and *strict monotonicity* under IO, *convexity* under incoherent mixing, and the presence of *maximally coherent states* (MCS). The  $l_1$ -norm of coherence ( $C_{l_1}$ ) [12], the relative entropy of coherence ( $C_{r,e}$ ) [12], the geometric coherence ( $C_g$ ) [6] are a few coherence quantifiers that satisfy these criteria, thus considered bona fide coherence measures.

However, it is not straightforward to achieve a novel coherence quantifier that is equally applicable to both pure- and mixed-state regimes since the coherence information hidden in the participating pure states is not directly retrievable from the mixed density matrix. In this regard, two techniques (for quantifying mixed states) are found helpful: (a) the traditional convex-roof of coherence ( $C_{conv.roof}$ ) [34], and (b) coherence measure in terms of pure-state coherence, or, in short, the top coherence ( $C_P$ ) [35]. Both of these quantifier classes are solely dependent on the coherence measures defined for the entire pure-state regime, called the *pure-state coherence measures* (PSCMs) [36].

Unearthing a new PSCM is vital for the following reasons: (a) a PSCM provides a proper measure for any pure state; (b) it is simple to compute since the diagonal elements of a density matrix are the only participants; and (c) it contributes towards different coherence measures (monotones) based on the expansion techniques (such as  $C_{conv.roof}$  or  $C_P$ ) that effectively introduce new computability as well as broaden the operational aspect, greatly improving resource theory [37], [38], [39].

Realizing the importance of a PSCM, Shuanping Du et. al. [36] recommended four constraints (based on the quantifying criteria provided by Baumgratz et. al. [12] that a PSCM must satisfy. These constraints, in fact, act as guidelines for finding a new PSCM. Nonetheless, the number of valid PSCMs proposed so far is not adequate. In this work, we put forth two PSCMs that fulfill all four criteria. We also provide their general normalized expressions so that they can be applied to any finite-dimensional system.

We further discuss the expansion of these PSCMs in the mixed-state regime through the assistance of  $C_{conv.roof}$  and  $C_P$ , and reveal the nature of the updated versions (to clear up the confusion with the top coherence being a *coherence measure* or *monotone* class [35], we present a thorough analytical investigation in the **Appendix-C**). Nevertheless,  $C_{conv.roof}$  and  $C_P$  are the indirect methods. PSCMs can also be applied directly to a mixed state (using the simple expression given by ref. [40]) if the participating pure states and their mixing probabilities are known; we cover that part too.

Probing an atomic or molecular system through a laser pulse ( $<10^{-12}$ m) is a very well-known and useful technique for unraveling its rovibrational structure, dynamics and for coherent control of the quantum state populations of the atomic or molecular system [41], [42], [43], [44]. In these studies, the state populations, or the basis-state probabilities (of the quantum system), are the important parameters. However, since the cumulative effect of all the associated state populations is reflected in the overall coherence, the study of coherence dynamics could be more beneficial on these occasions. In view of this, we consider a model two-qubit system (quantum), perturbed by a laser pulse, and quantitatively demonstrate the overall coherence evolutions for different interaction scenarios, such as qubit-qubit *coupling* or *decoupling* situations, along with laser-qubit *resonance* or *detuned* conditions.

The sole purpose of this quantitative investigation is twofold: firstly, to compare the evolutions of the PSCMs (we propose) to the most prevalent coherence measures,  $C_{l_1}$  and  $C_{r.e.}$ , and secondly, to provide a brief account of the coherence responses for different interacting situations.

We organize the remainder of the paper as follows: In Sec. 2, we briefly review the basics of the coherence resource theory framework, particularly emphasizing the coherence criteria, followed by a discussion of the methodology of  $C_P$  and the four necessary conditions for any bona fide PSCM. In Sec. 3, we set forth two functions of coherence vector ( $\mu$ ) and prove their validity as two bona fide PSCMs; thereafter, provide the general normalized expressions of the two PSCMs. In Sec. 4: appendix-a, we consider a laser pulse-two-qubit interacting system and quantitatively demonstrate the coherence evolutions of the qubit system based on these PSCMs, along with two other coherence measures,  $C_{l_1}$  and  $C_{r.e.}$ . We then discuss the extensions of the two PSCMs in the mixed-state regime (Sec. 5). Finally, we draw the conclusion in Sec. 6.

## 2 Theoretical Background

### 2.1 Resource Theory of coherence Framework

As with the resource theory of quantum entanglement or any other quantum resource [11], [37], the framework of the standard coherence resource theory is well-defined by free operations and free states. In the reference basis,  $\{|i\rangle\}$ , a free or incoherent state is denoted by  $\delta$ , where  $\delta = \sum_i \delta_i |i\rangle\langle i|$  with the basis-state probabilities  $\delta_i$ . Thus, the incoherent class  $\mathfrak{I} : \delta \in \mathfrak{I}$  contains the states abstaining from any non-zero off-diagonal elements in their density-matrix forms. The free (or incoherent) operation ( $\Lambda$ ) is a completely positive and trace-preserving (CPTP) map that cannot increase coherence. Therefore, it maps an incoherent state to any other incoherent state:  $\Lambda\delta\Lambda^\dagger \in \mathfrak{I}$ .  $\Lambda$  is defined in the Kraus representation as  $\Lambda(\cdot) = \sum_n K_n(\cdot)K_n^\dagger$  with  $K_n(\cdot)K_n^\dagger$  and  $\sum_n K_n K_n^\dagger = I$ , where  $I$  denotes the unity operator and “ $\cdot$ ” stands for an arbitrary quantum state. The coherence framework sets the following criteria that a bona fide coherence quantifier ( $C$ ) should fulfill [12], [35], [36].

C1: *Non-negativity*. For a quantum state  $\rho$ ,  $C(\rho) = 0 \forall \rho \in \mathfrak{I}$ , otherwise  $C(\rho) > 0$ .

C2: *Monotonicity*.  $C(\rho) \geq C(\Lambda\rho\Lambda^\dagger)$ . This criterion basically reflects the definition of free or incoherent operation,  $\Lambda$ .

C3: *Strong monotonicity*. State coherence should not increase under sub-selection measurements on average, i.e.,  $C(\rho) \geq \sum_n p_n C(K_n\rho K_n^\dagger/p_n) = \sum_n p_n C(K_n\rho_n K_n^\dagger)$ , with  $p_n = \text{Tr}[K_n\rho K_n^\dagger]$ , the probability that the incoherent Kraus operation  $K_n$  acting on  $\rho$  (in other words,  $p_n$  is expectation value of the state  $\rho_n = K_n\rho K_n^\dagger/\text{Tr}[K_n\rho K_n^\dagger]$ , achieved after the action of  $K_n$ ).

C4: *Convexity*. for any mixed state  $\rho (= \sum p_i \rho_i)$ , the coherence of  $\rho$  is not greater than the average coherence of the participating pure states  $\rho_i$ , i.e.,  $C(\rho) \leq \sum p_i C(\rho_i)$ .

C5: *Maximal coherence*.  $C(\rho) < C(|\Phi_d\rangle\langle\Phi_d|)$ , for any  $\rho$  other than  $|\Phi_d\rangle\langle\Phi_d|$ , where  $|\Phi_d\rangle (= \frac{1}{\sqrt{d}} \sum_{n=1}^d e^{i\theta_n} |n\rangle)$  with real  $\theta_n$  [45] is the set of *maximally coherent states* (MCS) of dimension  $d$ .

Note: When  $C(\rho)$  satisfies all five criteria C1–C5, it is classified as a *coherence measure*; conversely, if it satisfies all the given criteria except C4, it is called a *coherence monotone*, in the same way as an entanglement monotone. Due to its’ *convex* (C4) nature, the handling of a *coherence measure* is mathematically convenient [35]. However, it is seen in some instances that *coherence monotones* have an operational advantage over *coherence measures* and thus play an important role as coherence quantifiers [46].

## 2.2 Concept of $C_P$

In this work, we redefine  $C_P$  and investigate its *convexity*. Here, the basic idea of it is discussed below:

In quantum state transformation, an objective state  $\rho$  can be achieved from another state  $\varphi$  through incoherent operations (IO), but only if the coherence of  $\varphi$  is not less than  $\rho$  [12], i.e.,  $\varphi \xrightarrow{IO} \rho$  iff  $C(\varphi) \geq C(\rho)$  for  $C$  being any valid coherence monotone or measure. The above statement implies that a general state  $\rho$  (pure, or mixed) can be realized from different pure states through distinct incoherent channels. In that sense, *for a particular  $\rho$ , the set of pure states is non-empty*. This non-empty set for  $\rho$  is denoted by  $R(\rho)$ . In ref. [35], it is shown that the minimal coherence achieved from the set of pure states  $|\varphi\rangle \in R(\rho)$  can be a valid coherence quantifier of  $\rho$ .

**Theorem 1**[35]. *If  $\rho$  is the given state (pure or mixed) that can be converted from a set of pure states  $|\varphi\rangle$  in  $R(\rho)$  through IO, then  $C_P(\rho)$  is a coherence monotone with*

$$C_P(\rho) = \inf_{|\varphi\rangle \in R(\rho)} \check{C}(|\varphi\rangle). \quad (1)$$

Here,  $\check{C}(\cdot)$  is any *pure-state coherence measure (PSCM)*.

## 2.3 Coherence Vector and Criteria for PSCM

**Coherence vector:** Let  $|\psi\rangle \in \mathcal{H}^d$  be an arbitrary pure state in a  $d$ -dimensional Hilbert space ( $\mathcal{H}^d$ ). Then, its *probability vector* [35]  $\boldsymbol{\mu}(|\psi\rangle) \equiv (\langle 1|\psi\rangle\langle\psi|1\rangle, \langle 2|\psi\rangle\langle\psi|2\rangle, \dots, \langle d|\psi\rangle\langle\psi|d\rangle)^T$ .  $\boldsymbol{\mu}(|\psi\rangle)$  contains all the diagonal elements of the density matrix  $|\psi\rangle\langle\psi|$ , in the reference basis  $\{|i\rangle\}_{i=1}^d$  (coherence is basis-dependent). Since coherence does not depend on the phase terms, the total coherence of a pure state can be measured only considering the absolute values of the probability amplitude elements of that state (or the diagonal elements of its density operator). Therefore, a PSCM ( $\check{C}$ ) is a function of *probability vector  $\boldsymbol{\mu}$* , i.e.,  $\check{C}(|\psi\rangle) \equiv \check{C}(\boldsymbol{\mu}(|\psi\rangle))$ . For this reason,  $\boldsymbol{\mu}$  is known as *coherence vector* in the study of coherence.

**PSCM criteria:** A bona fide PSCM must satisfy the following four conditions [36], [35], [47]:

*Condition 1.* If  $\boldsymbol{\mu}$  is any permutation of  $(1, 0, \dots, 0)^T$ ,  $\check{C}(\boldsymbol{\mu}) = 0$ .

*Condition 2.* Under any permutation operation  $P_\pi$ ,  $\check{C}(\boldsymbol{\mu})$  is invariant, i.e.,  $\check{C}(P_\pi(\boldsymbol{\mu})) = \check{C}(\boldsymbol{\mu})$ ; here,  $P_\pi$  is the permutation matrix corresponding to  $\pi$ , which is a permutation of  $\{1, 2, \dots, d\}$ .

*Condition 3.*  $\check{C}(\boldsymbol{\mu})$  should be a concave function of  $\boldsymbol{\mu}$ :  $\check{C}(\lambda\boldsymbol{\mu}(\rho_1) + (1-\lambda)\boldsymbol{\mu}(\rho_2)) \geq \lambda\check{C}(\boldsymbol{\mu}(\rho_1)) + (1-\lambda)\check{C}(\boldsymbol{\mu}(\rho_2))$  for  $\forall \lambda \in [0, 1]$ .

*Condition 4.*  $\check{C}(\boldsymbol{\mu})$  reaches the maximal ( $\check{C}(\boldsymbol{\mu}) = 1$ , in the case of normalized PSCM) if all the elements of  $\boldsymbol{\mu}$  is  $\frac{1}{d}$ , i.e.,  $\boldsymbol{\mu} = \left(\frac{1}{d}, \frac{1}{d}, \dots, \frac{1}{d}\right)^T$ .

For any incoherent pure state,  $\boldsymbol{\mu}$  always takes the form of any of the permutations of  $(1, 0, \dots, 0)^T$ . Therefore, *condition-1* basically signifies the coherence measure criterion, C1 (see Sec.2.1). *Condition-2* arises from the symmetric nature of  $\check{C}(\boldsymbol{\mu})$ . Again, concavity fulfills *strict monotonicity*, C3 (see *Theorem 1* of Ref. [36]); hence,  $\check{C}(\boldsymbol{\mu})$  must be a concave function (i.e., *condition-3*). Lastly, *condition-4* directly resembles the criterion C5, indicating the presence of *maximally coherent states (MCS)*. Keeping in mind all these requirements we put forth two new PSCMs in the following section.

## 3 Two New PSCM: Propositions and Proofs

### 3.1 Diagonal Difference of PSCM ( $\check{C}_{DD}$ )

**Proposition 1:** *Let  $\rho$  represents any pure-state density matrix in  $\mathcal{H}^d$ , and let its coherence vector be*

defined as  $\boldsymbol{\mu}(\rho) = (\rho_{11}, \rho_{22}, \dots, \rho_{dd})^T$ , where the diagonal entries of  $\rho$  are  $\{\rho_{ii}\}_{i=1}^d$ . Then, the following function  $\check{C}_{DD}(\rho)$  based on the modulus of the difference between any two distinct elements of  $\boldsymbol{\mu}(\rho)$  in all possible combinations, serves as a coherence measure of  $\rho$ :

$$\check{C}_{DD}(\rho) = (d-1) - \sum_{i=1}^{d-1} \sum_{j=i+1}^d |\rho_{ii} - \rho_{jj}|. \quad (2)$$

**Proof.** In the following, we establish  $\check{C}_{DD}(\rho)$  as a valid PSCM through the fulfillment of all four necessary criteria, from *condition-1* to *condition-4*.

*Proof of condition-1.* When the pure state  $\rho$  is incoherent,  $\boldsymbol{\mu}(\rho)$  is confined to any of the permutation of  $(1, 0, \dots, 0)^T$ . In the RHS of Eq.2,  $\sum_{i=1}^{d-1} \sum_{j=i+1}^d |\rho_{ii} - \rho_{jj}|$ , is the sum of  $C_2^d$  (i.e.,  $\frac{d(d-1)}{2}$ ) absolute terms,  $|\rho_{ii} - \rho_{jj}|$ . For  $\boldsymbol{\mu}(\rho) = (1, 0, \dots, 0)^T$ , we can simply see that  $|\rho_{ii} - \rho_{jj}| = 0 \forall i \neq 1$ , whereas  $|\rho_{ii} - \rho_{jj}| = 1$  for  $i = 1$ ; therefore, it gives  $\sum_{i=1}^{d-1} \sum_{j=i+1}^d |\rho_{ii} - \rho_{jj}| = (d-1)$ . Similarly, when  $\boldsymbol{\mu}(\rho) = (0, 1, \dots, 0)^T$ , only  $|\rho_{11} - \rho_{22}|$  and  $(d-2)$  terms involving  $i = 2$  are non-zero (each of them giving one), which again makes the sum  $(d-1)$ . Continuing this way for all possible  $\boldsymbol{\mu}(\rho)$ , one can conclude that for any pure state  $\rho \in \mathfrak{J}$ ,  $\sum_{i=1}^{d-1} \sum_{j=i+1}^d |\rho_{ii} - \rho_{jj}| = (d-1)$ . Apply of this result to Eq.2 shows that  $\check{C}_{DD}(\rho)$  meets *condition-1*.

*Proof of condition-2.* Let  $\boldsymbol{\mu}(\rho_0) = (\rho_{11}, \rho_{22}, \dots, \rho_{dd})^T$ , and after a permutation operation (where the positions of  $\rho_{ii}$  and  $\rho_{11}$  are interchanged) we have  $\boldsymbol{\mu}(\rho_1) = P_\pi(\boldsymbol{\mu}(\rho_0)) = (\rho_{ii}, \rho_{22}, \dots, \rho_{11}, \dots, \rho_{dd})^T$ . As  $|\rho_{ii} - \rho_{jj}| = |\rho_{jj} - \rho_{ii}| \forall (i, j)$ , it is obvious from Eq. (2) that  $\check{C}_{DD}(\boldsymbol{\mu}(\rho_0)) = \check{C}_{DD}(\boldsymbol{\mu}(\rho_1))$ . Therefore,  $\check{C}_{DD}$  is permutation invariant or a symmetric function of  $\boldsymbol{\mu}$ .

*Proof of condition-3.* A function  $f: \mathbb{R}^n \rightarrow \mathbb{R}$  is *concave* if its domain  $dom(f)$  is a *convex set*, and for  $\forall x, y \in dom(f)$  and  $\forall \lambda \in [0, 1]$ , the following inequality holds [48]:

$$f(\lambda x + (1-\lambda)y) \geq \lambda f(x) + (1-\lambda)f(y). \quad (3)$$

Conversely,  $f(\cdot)$  is *convex* when the inequality is reversed.

In this case,  $\boldsymbol{\mu}(\rho) \in \mathbb{R}^d$  always belongs to a *convex set*, as the elements of  $\boldsymbol{\mu}(\rho)$  i.e.,  $\{\rho_{ii}\}$  are constrained by the following two conditions:  $\forall \rho_{ii} \geq 0$  and  $\sum_{i=1}^d \rho_{ii} = 1$ . It can be easily verified that  $\mathcal{M} = \sum_{i=1}^{d-1} \sum_{j=i+1}^d |\rho_{ii} - \rho_{jj}|$  is a *convex* function of  $\boldsymbol{\mu}(\rho)$ , and thus  $\check{C}_{DD}$  is *concave* (Eq. (2)). A detailed proof in this regard is given in **Appendix-A**. Two illustrations are the following: Let  $\boldsymbol{\mu}(\rho_1) = (0.2, 0.3, 0.5)^T$ ,  $\boldsymbol{\mu}(\rho_2) = (0.4, 0.3, 0.3)^T$  and  $\lambda = 0.5$ ; then  $\mathcal{M}(\lambda\boldsymbol{\mu}(\rho_1) + (1-\lambda)\boldsymbol{\mu}(\rho_2)) = 0.2$ , whereas  $\lambda\mathcal{M}(\boldsymbol{\mu}(\rho_1)) + (1-\lambda)\mathcal{M}(\boldsymbol{\mu}(\rho_2)) = 0.4$ , thus obeying *convexity*. In another example, let  $\lambda = 0.25$ , keeping the *coherence vectors* the same. Here again, the result follows *convexity*:  $\mathcal{M}(\lambda\boldsymbol{\mu}(\rho_1) + (1-\lambda)\boldsymbol{\mu}(\rho_2)) = 0.1$  and  $\lambda\mathcal{M}(\boldsymbol{\mu}(\rho_1)) + (1-\lambda)\mathcal{M}(\boldsymbol{\mu}(\rho_2)) = 0.3$ . Therefore,  $\check{C}_{DD}$  fulfils concavity in the two examples since  $\mathcal{M}$  follows *convexity* for both the occasions. Fig. 1(a) (below), numerically verifies the concavity of  $\check{C}_{DD}$  for all values of  $\lambda$  for the same participant states,  $\rho_1$  and  $\rho_2$ .

*Proof of condition-4.* All the diagonal elements  $\{\rho_{ii}\}$  of any MCS (which is obviously a pure state) are of the same value  $\frac{1}{d}$ , i.e.,  $\boldsymbol{\mu}(MCS) = (\frac{1}{d}, \frac{1}{d}, \dots, \frac{1}{d})^T$ . Thus, for any MCS,  $\mathcal{M} = \sum_{i=1}^{d-1} \sum_{j=i+1}^d |\rho_{ii} - \rho_{jj}| = 0$ . Applying this result in Eq. (2), it gives that  $\check{C}_{DD}(MCS) = (d-1)$ , which is the maximal (as  $\mathcal{M} \geq 0$ ). So, *condition-4* is satisfied, and with this, the proof of *proposition-1* is completed.

### 3.2 Diagonal Multiplication of PSCM ( $\check{C}_{DM}$ )

**Proposition 2:** Let  $\rho$  represents any pure-state density matrix in  $\mathcal{H}^d$ , and let its coherence vector be defined as  $\boldsymbol{\mu}(\rho) = (\rho_{11}, \rho_{22}, \dots, \rho_{dd})^T$ , where the diagonal entries of  $\rho$  are  $\{\rho_{ii}\}_{i=1}^d$ . Then, the fol-

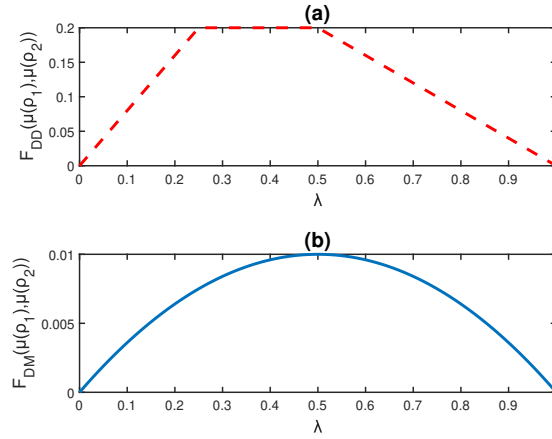


Fig. 1. The concavity of  $\check{C}_{DD}$  (Fig. 1(a)) and  $\check{C}_{DM}$  (Fig. 1(b)) is shown numerically in Fig. 1. For this, two qutrit states,  $\rho_1$  and  $\rho_2$ , are taken into account, with corresponding coherence vectors of  $\mu(\rho_1) = (0.2, 0.3, 0.5)^T$  and  $\mu(\rho_2) = (0.4, 0.3, 0.3)^T$ , respectively. The weighting factor  $\lambda \in [0, 1]$  is the x-variable and  $F_{DD(DM)} = \check{C}_{DD(DM)}(\lambda\mu(\rho_1) + (1-\lambda)\mu(\rho_2)) - \lambda\check{C}_{DD(DM)}(\mu(\rho_1)) - (1-\lambda)\check{C}_{DD(DM)}(\mu(\rho_2))$  are the y-variables. In this scenario, both  $\check{C}_{DD}$  and  $\check{C}_{DM}$  adhere to the concavity inequality (Eq. (3)), since  $F_{DD(DM)} \geq 0$  for each  $\lambda$ .

lowing function  $\check{C}_{DM}(\rho)$  containing the products between two distinct elements of  $\mu(\rho)$  in all possible combinations, serves as a coherence measure of  $\rho$ .

$$\check{C}_{DD}(\rho) = \sum_{i=1}^{d-1} \sum_{j=i+1}^d \rho_{ii}\rho_{jj}. \quad (4)$$

**Proof.** To be a PSCM,  $\check{C}_{DM}$  must meet the same requirements as  $\check{C}_{DD}$ , which are conditions 1 through 4.

*Proof of condition-1.* If  $\rho \in \mathfrak{J}$ ,  $\mu(\rho) \in \{P_\pi((1, 0, \dots, 0)^T)\}$ . Since  $\mu(\rho)$  has only one non-zero element, all the  $C_2^d$  number of  $\rho_{ii}\rho_{jj}$  terms in the RHS of Eq. (4) yield zero. Hence,  $\check{C}_{DM}(\rho) = 0$ .

*Proof of condition-2.* Since  $\rho_{ii} \geq 0 \forall \rho_{ii}$  ( $i \in \{1, 2, \dots, d\}$ ) and the RHS of Eq. (4) gives the sum of all possible  $\rho_{ii}\rho_{jj}$ , it is easy to conclude that  $\check{C}_{DM}(\mu(\rho))$  is invariant under any permutation operation, i.e.,  $\check{C}_{DM}(P_\pi(\mu(\rho))) = \check{C}_{DM}(\mu(\rho)) \forall \pi \in \{1, 2, \dots, d\}$ .

*Proof of condition-3.* A detailed, general proof regarding the concavity of  $\check{C}_{DM}$  is given in **Appendix-B**. Here, two examples are given for verification: Let,  $\mu(\rho_1) = (0.2, 0.3, 0.5)^T$ ,  $\mu(\rho_2) = (0.4, 0.3, 0.3)^T$  and  $\lambda = 0.5$ ; then  $\check{C}_{DM}(\lambda\mu(\rho_1) + (1-\lambda)\mu(\rho_2)) = 0.33$ , whereas,  $\lambda\check{C}_{DM}(\mu(\rho_1)) + (1-\lambda)\check{C}_{DM}(\mu(\rho_2)) = 0.32$ , thus obeying concavity. In another example, let  $\lambda = 0.25$  keeping the coherence vectors unchanged. Here again, the result follows concavity by fulfilling Eq. (3):  $\check{C}_{DM}(\lambda\mu(\rho_1) + (1-\lambda)\mu(\rho_2)) = 0.3325$  and  $\lambda\check{C}_{DM}(\mu(\rho_1)) + (1-\lambda)\check{C}_{DM}(\mu(\rho_2)) = 0.3250$ . The concavity of  $\check{C}_{DM}$  for all values of  $\lambda \in [0, 1]$  for the same participant states,  $\rho_1$  and  $\rho_2$ , is verified statistically in Fig.1(b).

*Proof of condition-4.* A  $d$ -dimensional pure state  $\rho$  is MCS only if all its diagonal elements are of the same value  $\frac{1}{d}$ ; thus,  $\mu(MCS) = (\frac{1}{d}, \frac{1}{d}, \dots, \frac{1}{d})^T$ . To prove that  $\check{C}_{DM}(MCS)$  is the maximal, it is first necessary to prove the following lemma:

**Lemma 1.** If a finite set of positive-valued variables has a constant sum, then the sum of the

products of two distinct variables (from that set) in all possible combinations is maximal if all the variables have equal values.

*Proof of Lemma-1.* First, consider a two-variable system  $x_1, x_2 \in \mathbb{R}^+$  bound by the condition:  $x_1 + x_2 = S$  where  $S$  is any finite constant. Thus,  $x_1x_2 = x_1(S - x_1)$ . Now if we take the first order derivative of  $x_1x_2$  w.r.t  $x_1$  and equate it to zero we simply find the maxima of  $x_1x_2$  at  $x_1 = x_2 = \frac{S}{2}$  (this is the maximal point as the second order derivative of  $x_1x_2$  is negative). Hence lemma-1 is satisfied for the two-variable case.

Now, take a three-variable system  $x_1, x_2, x_3 \in \mathbb{R}^+$  where  $x_1 + x_2 + x_3 = S$ . To find the maximal point of ' $x_1x_2 + x_1x_3 + x_2x_3$ ,' let's assume  $x_1 = k$  (positive constant less than  $S$ ). Then  $x_1x_2 + x_1x_3 + x_2x_3 = k(S - k) + x_2x_3$  effectively becomes a two-variable problem (discussed above) with its maxima at:  $x_2 = x_3$ . Conversely, if we assume  $x_3 = m$  (positive constant less than  $S$ ), Then  $x_1x_2 + x_1x_3 + x_2x_3 = x_2x_3 + m(S - m)$  again becomes a two-variable problem with its maxima at:  $x_1 = x_2$ . ' $x_1x_2 + x_1x_3 + x_2x_3$ ' is maximal if and only if both the situations occur simultaneously, that means when  $k = m$ ; it implies that  $x_1x_2 + x_1x_3 + x_2x_3$  is maximal when  $x_1 = x_2 = x_3 = \frac{S}{3}$ .

Similarly, for a four-variable case assuming the first or last variable constant, it becomes a three-variable case. Thereby applying the same logic used in the three-variable situation it is easy to prove that the maximality is achieved when all the four variables are equal to  $\frac{S}{4}$ .

Therefore, applying the mathematical induction it is proved that for a general  $d$ -variable system  $\sum_{i=1}^{d-1} \sum_{j=i+1}^d x_i x_j$  is maximal if and only if  $x_1 = x_2 = \dots = x_d = \frac{S}{d}$ . Hence, the proof of lemma-1 is completed.

As we know that  $\check{C}_{DM}(\rho) = \sum_{i=1}^{d-1} \sum_{j=i+1}^d \rho_{ii}\rho_{jj}$  with  $\sum_{n=1}^d \rho_{nn} = 1$  and  $\forall \rho_{nn} \geq 0$ ,  $\check{C}_{DM}$  is maximal when  $\forall \rho_{nn} = \frac{1}{d}$ ; thus, the proof of condition-4 is completed.

Alternatively, condition-4 can be directly proved with the help of the  $l_1$ -norm of coherence ( $C_{l_1}$ ).  $C_{l_1}$  is defined by the sum of the modulus of all the off-diagonal elements of  $\rho$ , i.e.,  $C_{l_1}(\rho) = \sum_{i,j=1;i \neq j}^d |\rho_{ij}|$  [12]. Now, for any pure state,  $|\rho_{ij}| = \sqrt{\rho_{ii}\rho_{jj}}$ ; therefore,  $C_{l_1}(\rho)$  can be rewritten as below:

$$C_{l_1}(\rho) = 2 \sum_{i=1}^{d-1} \sum_{j=i+1}^d \sqrt{\rho_{ii}\rho_{jj}}. \tag{5}$$

This expression of  $C_{l_1}$  is very similar to  $\check{C}_{DM}$  (Eq. (4)) except of the presence of twice multiplicity and the power-root of two (associated with each term in the summation) for  $C_{l_1}$ . As  $C_{l_1}$  is a coherence measure, it obeys the criterion C5, i.e.,  $C_{l_1}(\rho)$  is maximal only if  $\rho \in MCS$ ; in other words,  $C_{l_1}$  obeys condition-4. Therefore, the close resemblance between the expressions of  $C_{l_1}(\rho)$  and  $\check{C}_{DM}(\rho)$  makes it obvious that  $\check{C}_{DM}(\rho)$  is maximal only if  $\rho \in MCS$ , i.e., when  $\mu(\rho) = \left(\frac{1}{d}, \frac{1}{d}, \dots, \frac{1}{d}\right)^T$ . With this, the proof of  $\check{C}_{DM}(\rho)$  being a valid PSCM is completed.

### 3.3 Normalization of ( $\check{C}_{DD}$ ) and ( $\check{C}_{DM}$ )

Normalization of a coherence measure is necessary when it comes to dealing with multi-dimensional systems or to compare among different coherence measures. In this regard, a general normalized (to one) expression of a coherence measure applicable to any dimension is desirable. The normalizing process relies on determining the MCS values for a few different dimensional systems and, from these results, achieving a general MCS value (by applying mathematical induction), which is a function of system dimension ( $d$ ). However, the general normalizing factors are straightforward to attain for  $\check{C}_{DD}$  and  $\check{C}_{DM}$ .

For  $\check{C}_{DD}$ , it is apparent from Eq.(2) that  $\check{C}_{DD}(|\Phi_d\rangle) = (d - 1)$ , where  $|\Phi_d\rangle \in MCS$ ; therefore, the normalizing factor for  $\check{C}_{DD}$  is ' $\frac{1}{d-1}$ ,' and with this the normalized expression for  $\check{C}_{DD}$  is the following:

$$\check{C}_{DD}(\rho) = 1 - \frac{1}{d-1} \sum_{i=1}^{d-1} \sum_{j=i+1}^d |\rho_{ii} - \rho_{jj}|. \tag{6}$$

In the case of  $\check{C}_{DM}$ , the normalizing factor cannot be directly achieved from its expression (like  $\check{C}_{DD}$ ), however it's easy to calculate. From Eq. (4) we can achieve the  $\check{C}_{DM}(|\Phi_d\rangle)$  values, which are  $\frac{1}{2^2}$ ,  $\frac{3}{3^2}$ ,  $\frac{6}{4^2}$ , and  $\frac{10}{5^2}$  for  $d = 2, 3, 4$  and  $5$ , respectively. Therefore, by applying mathematical induction, it can be found that  $\check{C}_{DM}(|\Phi_d\rangle) = C_2^d \frac{1}{d^2} = \frac{d(d-1)}{2(d^2)}$ . With this, the normalized expression for  $\check{C}_{DM}$  is the following:

$$\check{C}_{DM}(\rho) = 1 - \frac{1}{d-1} \sum_{i=1}^{d-1} \sum_{j=i+1}^d \rho_{ii}\rho_{jj}. \tag{7}$$

From now on, when we refer to  $\check{C}_{DD(or DM)}$ , we mean the normalized version (Eq.(6) (or Eq.(7)) that is presented above, rather than its earlier form (Eq. (2) (or Eq.(4))).

#### 4 Evolutions of $\check{C}_{DD}$ and $\check{C}_{DM}$ (Qubit-Laser Pulse Interaction Model)

In the previous section, we presented two new PSCMs ( $\check{C}_{DD}$  and  $\check{C}_{DM}$ , respectively) together with their proofs in Sec. 3.1 and Sec. 3.2 and their normalized expressions in Sec. 3.3. Here, we numerically demonstrate the responses of these two PSCMs, along with two additional bona fide coherence measures,  $C_{l_1}$  and  $C_{r.e}$ , for the state change of a fictitious two-qubit system when perturbed by a laser pulse, in the pure-state regime. The purpose of this quantitative study is twofold: (a) to validate  $\check{C}_{DD}$  and  $\check{C}_{DM}$  as two novel coherence measures through the comparison with  $C_{l_1}$  and  $C_{r.e}$ , and (b) a brief demonstration of coherence evolutions for different kinds of laser-qubit and qubit-qubit interaction scenarios. We employ normalized versions of these four measures in order to preserve scalability.

##### 4.1 Interaction Model

We have seen before that state coherence is a function of the basis-state probabilities  $\{\rho_{ii}\}$  (which are sometimes called the state populations, in the light-matter interaction scenario). A simple and well-studied setup that allows population dynamics is a laser pulse-two-level (or multilevel) interaction system [41], [42], [44], a semi-classical approach where the laser pulse (considered classical, without loss of generality [42]–[44]) makes the *Hamiltonian* of the qubit-system (quantum) time (or pulse area) dependent. Based on this model, we think of an isolated two-qubit system (no decay loss) and simulate the evolution of the qubit system when perturbed by a laser pulse. To ease calculation, we use the *FM-transformed* form (a detailed discussion of it is given in [41], [49]) of the *perturbed Hamiltonian*:

$$H_i = \begin{pmatrix} \Delta_i & \Omega_i/2 \\ \Omega_i^*/2 & 0 \end{pmatrix} \tag{8}$$

where  $i$  takes the value 1 (or 2) for *qubit-1* (or *qubit-2*);  $H_i$ ,  $\Delta_i$ , and  $\Omega_i$  are the *Hamiltonian*, *detuning*, and *Rabi frequency* [41], [50] of the  $i^{th}$  qubit, respectively. Here, we study two cases: (a) *qubit-qubit non-interacting*:



$$H = I \otimes H_1 + H_2 \otimes I , \quad (9)$$

(where  $I$  is the  $(2 \times 2)$  identity operator), and

(b) *qubit-qubit interacting*:

$$H = H_2 \otimes H_1 , \quad (10)$$

keeping an interaction strength of “one.” For each one of these cases, we examine both *resonance* ( $\Delta_i = 0$ ) and *detuned* ( $\Delta_i \neq 0$ ) interactions. ‘ $H$ ’ is the total *Hamiltonian* of the *two-qubit system*. The governing equation used here is the famous *Von Neumann-Liouville equation* [42], [51]]:

$$\dot{\rho} = i\hbar^{-1}[\rho, H] . \quad (11)$$

To compare coherence evolutions, we use normalized forms of  $C_{r,e}$  and  $C_{l_1}$  [52].

#### 4.2 Numerical results

Figs. 2, 3, 4, and 5 depict the evolution of  $\check{C}_{DD}$  and  $\check{C}_{DM}$  in comparison with  $C_{l_1}$  and  $C_{r,e}$  in the pure state regime.

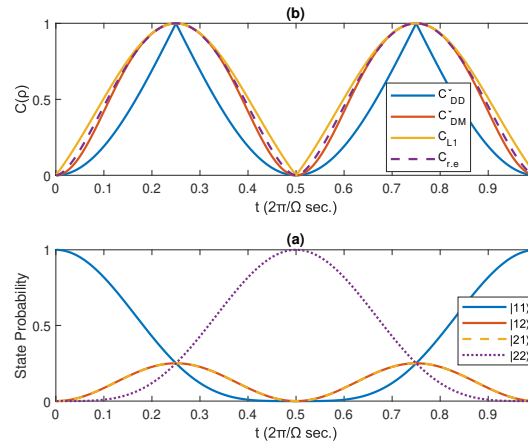


Fig. 2. Comparison of the evolutions of the coherence measures ( $\check{C}_{DD}$ ,  $\check{C}_{DM}$ ,  $C_{r,e}$ , and  $C_{l_1}$ ) vs. time for an isolated two-qubit system perturbed by a laser pulse (Figs.2(b)). It shows the laser pulse-qubit interaction at resonance ( $\Delta_i = 0$ ;  $\Delta_i$ : detuning of the  $i$ -th qubit for  $i = \{1, 2\}$ ) with qubit-qubit unentangled to each other. The evolutions of the basis-state populations indicated by  $|00\rangle$ ,  $|01\rangle$ ,  $|10\rangle$  and  $|11\rangle$  are also captured in Fig.2(a).

As both  $C_{l_1}$  and  $C_{r,e}$  are bona fide coherence measures (that fulfill all five necessary and sufficient coherence criteria, C1–C5), the similar trends of  $\check{C}_{DD}$  and  $\check{C}_{DM}$  for each of the qubit-qubit *non-interacting* (see figs. 2, 3) and *interacting* (see figs. 4, 5) cases substantiate that these two are valid coherence measures; furthermore, the different pathways for all four measures provide the proofs of distinctness of  $\check{C}_{DD}$  or  $\check{C}_{DM}$  from the other three. The *resonant and non-interacting* plot (fig. 2) shows that while all the population curves meet at the same point (i.e., the formation of MCS), all four coherence measures reach “1”; likewise, wherever only the state  $|2-2\rangle$  achieves the population “one” and all remaining state populations stay at “0” (implies  $\mathfrak{J}$ ), all the measures become “0”, and in between

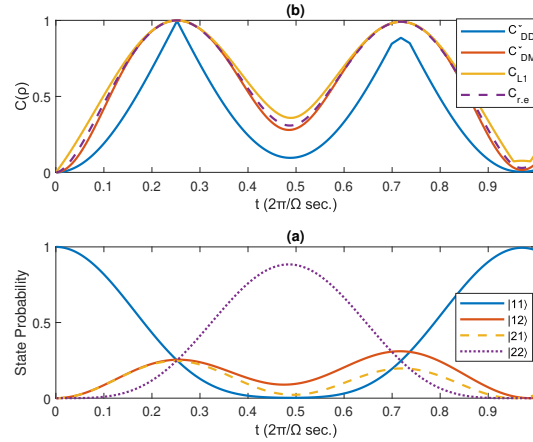


Fig. 3. Comparison of the evolutions of the coherence measures ( $\check{C}_{DD}$ ,  $\check{C}_{DM}$ ,  $C_{r,e}$ , and  $C_{l_1}$ ) vs. time for an isolated two-qubit system perturbed by a laser pulse (Figs.3(b)) It shows the detuned laser pulse-qubit interaction ( $\Delta_1 = 1\Omega$  and  $\Delta_2 = 2\Omega$ ; detuning of the  $i$ -th qubit for  $i = \{1, 2\}$ ) with qubit-qubit unentangled to each other. The evolutions of the basis-state populations indicated by  $|00\rangle$ ,  $|01\rangle$ ,  $|10\rangle$  and  $|11\rangle$  are also captured in Fig.3(a).

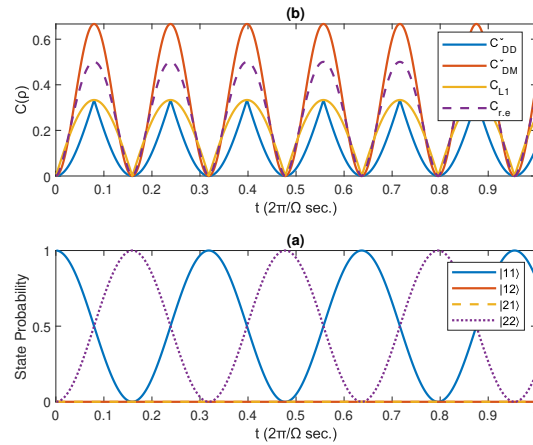


Fig. 4. Comparison of the evolutions of the coherence measures ( $\check{C}_{DD}$ ,  $\check{C}_{DM}$ ,  $C_{r,e}$ , and  $C_{l_1}$ ) vs. time for an isolated two-qubit system perturbed by a laser pulse (Figs.4(b)) It shows the laser pulse-qubit interaction at resonance ( $\Delta_i = 0$ ;  $\Delta_i$ : detuning of the  $i$ -th qubit for  $i = \{1, 2\}$ ) with qubit-qubit unentangled to each other. The evolutions of the basis-state populations indicated by  $|00\rangle$ ,  $|01\rangle$ ,  $|10\rangle$  and  $|11\rangle$  are also captured in Fig.4(a).

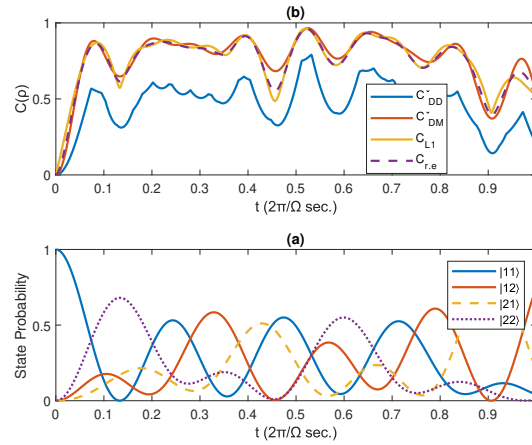


Fig. 5. Comparison of the evolutions of the coherence measures ( $\check{C}_{DD}$ ,  $\check{C}_{DM}$ ,  $C_{r,e}$ , and  $C_{L1}$ ) vs. time for an isolated two-qubit system perturbed by a laser pulse (Figs.5(b)) It shows the detuned laser pulse-qubit interaction ( $\Delta_1 = 1\Omega$  and  $\Delta_2 = 2\Omega$ ; detuning of the  $i$ -th qubit for  $i = \{1, 2\}$ ) with qubit-qubit unentangled to each other. The evolutions of the basis-state populations indicated by  $|00\rangle$ ,  $|01\rangle$ ,  $|10\rangle$  and  $|11\rangle$  are also captured in Fig.5(a).

these two extremes, they follow the condition  $0 < C(\rho) < 1$ . This means that the normalized  $\check{C}_{DD}$  and  $\check{C}_{DM}$  are well bounded within “1” and “0”.

The qubits are unentangled (following Eq.(9-a)) from each other for both Figs.2 and 3. Figs.3, 5 exhibit the *detuned* pulse-qubit interactions, whereas Figs. 4, 5 show the situations where qubits are *entangled* with each other (when the pulse is on).

The coherence measures are also responsive to these changed interactions, such as, for the qubit-qubit *entangled* with pulse-qubit *resonance* case (fig. 4), the oscillation frequencies of all four measures are much higher than the *unentangled* cases. On the other hand, the periodicity of the coherence measures is lost when the *detuned* pulse-qubit interaction is added to the *entangled* situation (fig.5). Therefore, different evolution patterns of a coherence measure can address various interaction situations. It indicates that the study of the *coherence pathway* can act as an instrument in tracing the interaction type between two quantum systems. However, it needs a thorough investigation including various types of interactions and for different dimensional systems, which is out of scope here.

### 5 $\check{C}_{DD}$ and $\check{C}_{DM}$ in the mixed-state regime

We have seen that both  $\check{C}_{DD}$  and  $\check{C}_{DM}$  are defined for any pure state. Therefore, in the more general context of the mixed-state domain, we can safely apply these to the quantifier classes, like  $C_P$  or  $C_{conv. roof}$ , to achieve new *coherence monotones* or *measures* (depending on the natures of  $C_P$  and  $C_{conv. roof}$ ). In the following, we define two such new *coherence monotones* (normalized) based on  $C_P$ : diagonal difference of coherence ( $C_{DD}^P$ ) and diagonal multiplication of coherence ( $C_{DM}^P$ ).

- *Diagonal difference of coherence under  $C_P$  ( $C_{DD}^P$ ):* Definition 1. If  $\rho$  is a given state (pure or mixed) with its associated pure state set  $R(\rho)$ , then  $C_{DD}^P(\rho)$  is a coherence monotone with

$$C_{DD}^P(\rho) = \inf_{|\psi\rangle \in R(\rho)} \check{C}_{DD}(|\psi\rangle) = 1 - \frac{1}{d-1} f(DD) . \quad (12)$$

where  $f(DD) = \sup_{|\psi\rangle \in R(\rho)} \sum_{i=1}^{d-1} \sum_{j=i+1}^d |\varphi_{ii} - \varphi_{jj}|$  with  $\varphi_{ii(jj)} = |\psi\rangle\langle\psi|_{ii(jj)}$ ; Here, the minimal of all the  $\check{C}_{DD}(|\psi\rangle \in R(\rho))$  is considered the measure of  $\rho$ .

- Diagonal multiplication of coherence under  $C_P$  ( $C_{DM}^P$ ): Definition 2. If  $\rho$  is a given state (pure or mixed) with its associated pure states set  $R(\rho)$ , then  $C_{DM}^P(\rho)$  is a coherence monotone with

$$C_{DM}^P(\rho) = \inf_{|\psi\rangle \in R(\rho)} \check{C}_{DM}(|\psi\rangle) = \frac{2d}{d-1} \inf_{|\psi\rangle \in R(\rho)} \sum_{i=1}^{d-1} \sum_{j=i+1}^d \varphi_{ii}\varphi_{jj} . \tag{13}$$

where  $\varphi_{ii(jj)} = |\psi\rangle\langle\psi|_{ii(jj)}$ .

Note that for any pure-state  $\rho = |\psi\rangle\langle\psi|$ , both Eq. (10) and Eq.11) reduce to the following forms, respectively:  $C_{DD}^P(\rho) = \check{C}_{DD}(|\psi\rangle)$  and  $C_{DM}^P(\rho) = \check{C}_{DM}(|\psi\rangle)$ .

In a similar manner,  $\check{C}_{DD}$  and  $\check{C}_{DM}$  provide two new coherence measures when applied in the coherence measure class  $C_{conv. roof}$ . The definitions of these two measures,  $C_{DD}^{conv. roof}$  and  $C_{DM}^{conv. roof}$ , are presented below in a compact form:

$$C_j^{conv. roof}(\rho) = \inf_{\{p_i, |\psi_i\rangle\}} \sum_i p_i \check{C}_j(|\psi_i\rangle) . \tag{14}$$

with  $j = 1$  or  $2$  stands for “DD” or “DM,” respectively. Here, the infimum is taken over all the pure state decompositions of  $\rho$  ( $\rho = \sum_i p_i (|\psi_i\rangle\langle\psi_i|)$ ).

Note that we do not consider  $C_{DD}^P$  or  $C_{DM}^P$  as a coherence measure but rather classify them as coherence monotones. This is due to the fact that  $C_P$  does not fulfil convexity (C4) in general. A detailed discussion on  $C_P$  and the analytical proof for its nonconvexity is given in **Appendix-C**.

These two techniques discussed above (i.e.,  $C_P$  and  $C_{conv. roof}$ ) are two different coherence quantifiers classes that just make use of  $C_{DD}$  or  $C_{DM}$  being two PSCMs; not only that, both these techniques suffer from optimization issues. However,  $C_{DD}$  or  $C_{DM}$  can be applied to a mixed state (with the help of a simple technique) if the associated pure states and their respective mixing probabilities of the mixed state are previously known.

Recently, ref. 40 shows that if the constituting pure states  $|\psi_i\rangle$  and their respective mixing probabilities  $p_i$  of a mixed state  $\rho$  are known beforehand, then a PSCM can be employed to measure  $\rho$  by using the following formula:

$$C_{PSCM}(\rho) = A \sum_{i=1}^n p_i \check{C}_{PSCM}(|\psi_i\rangle) . \tag{15}$$

where  $A = \sqrt{\sum_{j=1}^n p_j^2}$ . Eq.(13) is applicable to any finite-dimensional system if  $C_{PSCM}$  is properly normalized (to unity). As both  $\check{C}_{DD}$  and  $\check{C}_{DM}$  possess general normalized forms (Eqs. 6, 7), we can simply apply these to Eq. (13). In that case,

$$\begin{aligned} C_{DD}(\rho) &= A \sum_{i=1}^n p_i \check{C}_{DD}(|\psi_i\rangle) \\ C_{DM}(\rho) &= A \sum_{i=1}^n p_i \check{C}_{DM}(|\psi_i\rangle). \end{aligned} \tag{16}$$

In (Fig. 6) we demonstrate the evolutions of  $C_{DD}$  and  $C_{DM}$  along with  $C_{r,e}$ , and  $C_{l_1}$  for the same two-qubit-laser pulse interacting scenario (discussed in Sec.4.1) where  $\rho$  is initially prepared as a mixture of  $|\psi_1\rangle = |11\rangle$  and  $|\psi_2\rangle = |10\rangle$  with their respective mixing probabilities of 0.35 and 0.65. To enable generality, the qubits of  $|\psi_1\rangle$  and  $|\psi_2\rangle$  are kept in unentangled and entangled mode, respectively, and the pulse-qubit interaction is detuned for the first qubit (from right) whereas it is in resonant for the other qubit.

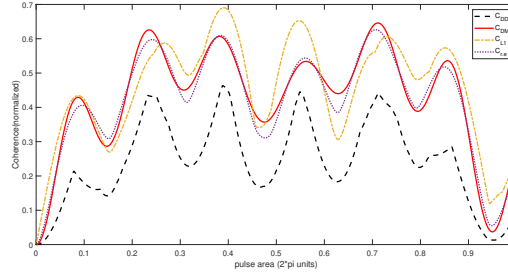


Fig. 6. A comparison of the evolution of  $C_{DD}$  and  $C_{DM}$  with  $C_{l_1}$  and  $C_{r,e}$  for a two-qubit system perturbed by a laser pulse. Here, the two-qubit system is considered a mixture of two pure states  $|\psi_1\rangle$  and  $|\psi_2\rangle$ , with their mixing probabilities  $p_1 = 0.35$  and  $p_2 = 0.65$ , respectively. Both  $|\psi_1\rangle$  and  $|\psi_2\rangle$  are varying with the pulse area (or time), and the participating qubits are in *unentangled* and *entangled* mode (between each other) for  $|\psi_1\rangle$  and  $|\psi_2\rangle$ , respectively. Initially, the mixed state is prepared as:  $|\psi\rangle = p_1 |11\rangle + p_2 |10\rangle$ . Pulse-qubit interaction is detuned ( $\Delta_1 = 1\Omega$ ) for the first qubit (from right), whereas it is resonant ( $\Delta_2 = 0$ ) for the second qubit.

## 6 Conclusions

In conclusion, we propose two novel coherence measures ( $\check{C}_{DD}$  and  $\check{C}_{DM}$ ) that are defined for the entire domain of pure states (both of them meet all four PSCM-criteria listed in Sec.2.3). Since  $\check{C}_{DD}$  and  $\check{C}_{DM}$  are simple to calculate and have available general normalized formulations, they can be used in any finite-dimensional system.

$\check{C}_{DD}$  and  $\check{C}_{DM}$  can also be extended to the mixed-state regime through any mixed-state extending approach, such as  $C_P$  or  $C_{conv. roof}$ .  $C_{conv. roof}$  creates two coherence measures,  $C_{DD}^{conv. roof}$  and  $C_{DM}^{conv. roof}$ , respectively, from  $\check{C}_{DD}$  and  $\check{C}_{DM}$ , whereas  $C_P$  yields the coherence monotones,  $C_{DD}^P$  and  $C_{DM}^P$  (respectively). The study of convexity (Appendix-C) provides a detailed explanation of why  $C_{DD}^P$  and  $C_{DM}^P$  are two monotones, as well as a redefinition of the top coherence ( $C_P$ ). Apart from the assistance of  $C_P$  or  $C_{conv. roof}$ , another direct extension approach [40] is also discussed in brief.

Finally, to see how the total coherence of a qubit system evolves whenever interacted by a laser pulse, along with comparing  $C_{DD}$  and  $C_{DM}$  with  $C_{l_1}$  and  $C_{r,e}$ , we consider an isolated two-qubit system interacted by the laser pulse and quantitatively demonstrate different interaction scenarios both in pure- and mixed-state regimes.

## Acknowledgements

We thank S. Goswami for language correction and editing. DG acknowledges funding support from MEITY, SERB, and STC ISRO of the Govt. of India.

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### Appendix A

#### Proof of concavity of $\check{C}_{DD}$ :

From Eq.(3), a function  $f : \mathbb{R}^n \rightarrow \mathbb{R}$  is *concave* if its domain  $dom(f)$  is a *convex set*, and for  $\forall x, y \in dom(f)$  and  $\forall \lambda \in [0, 1]$ , the following inequality holds:  $f(\lambda x + (1 - \lambda)y) \geq \lambda f(x) + (1 - \lambda)f(y)$ . Conversely,  $f(\cdot)$  is *convex* when the inequality is reversed. In this case,  $\mu(\rho) \in \mathbb{R}^d$  always belongs to a *convex set*, as the elements of  $\mu(\rho)$  i.e.,  $\{\rho_{ii}\}$  are constrained by the following two conditions:  $\forall \rho_{ii} \geq 0$  and  $\sum_{i=1}^d \rho_{ii} = 1$ . Now, the expression of  $\check{C}_{DD}$  (Eq.(2)) can be rewritten as

$$\check{C}_{DD} = (d - 1) - \mathcal{M}(\mu(\rho)). \tag{A.1}$$

where  $\mathcal{M}(\mu(\rho)) = \sum_{i=1}^{d-1} \sum_{j=i+1}^d |\rho_{ii} - \rho_{jj}|$ .  $\mathcal{M}(\mu(\rho))$  must be a convex function in order to satisfy concavity for  $\check{C}_{DD}$ . To prove the convexity of  $\mathcal{M}(\mu(\rho))$ , first consider two pure two-level states  $\rho^A$  and  $\rho^B$  with their respective coherence vectors:  $\mu^A = [\rho_{11}^A, \rho_{22}^A]^T$ , and  $\mu^B = [\rho_{11}^B, \rho_{22}^B]^T$ . Then,

$$\mathcal{M}(\mu^A) = |\rho_{11}^A - \rho_{22}^A|, \quad \mathcal{M}(\mu^B) = |\rho_{11}^B - \rho_{22}^B|. \tag{A.2}$$

Now,

$$\begin{aligned} \mathcal{M}(\lambda\mu^A + (1 - \lambda)\mu^B) &= \mathcal{M}\left(\begin{matrix} \lambda\rho_{11}^A + (1 - \lambda)\rho_{11}^B \\ \lambda\rho_{22}^A + (1 - \lambda)\rho_{22}^B \end{matrix}\right) \\ &= \left| \lambda(\rho_{11}^A - \rho_{22}^A) + (1 - \lambda)(\rho_{11}^B - \rho_{22}^B) \right| \\ &\leq \lambda|\rho_{11}^A - \rho_{22}^A| + (1 - \lambda)|\rho_{11}^B - \rho_{22}^B| \\ &= \lambda\mathcal{M}(\mu^A) + (1 - \lambda)\mathcal{M}(\mu^B) \end{aligned} \tag{A.3}$$

The inequality in Eq. (A-3) is true since  $\lambda \in [0, 1]$ .

Again, let  $\rho^A$  and  $\rho^B$  be two pure states in three-dimensional Hilbert space with their coherence vectors  $\mu^A = [\rho_{11}^A, \rho_{22}^A, \rho_{33}^A]^T$  and  $\mu^B = [\rho_{11}^B, \rho_{22}^B, \rho_{33}^B]^T$ , respectively. Thus,

$$\begin{aligned} \mathcal{M}(\mu^A) &= |\rho_{11}^A - \rho_{22}^A| + |\rho_{11}^A - \rho_{33}^A| + |\rho_{22}^A - \rho_{33}^A| \\ \mathcal{M}(\mu^B) &= |\rho_{11}^B - \rho_{22}^B| + |\rho_{11}^B - \rho_{33}^B| + |\rho_{22}^B - \rho_{33}^B| \end{aligned} \tag{A.4}$$

On the other hand,



$$\begin{aligned}
 \mathcal{M}(\lambda\boldsymbol{\mu}^A + (1-\lambda)\boldsymbol{\mu}^B) &= M \begin{pmatrix} \lambda\rho_{11}^A + (1-\lambda)\rho_{11}^B \\ \lambda\rho_{22}^A + (1-\lambda)\rho_{22}^B \\ \lambda\rho_{33}^A + (1-\lambda)\rho_{33}^B \end{pmatrix} \\
 &= \left| \lambda(\rho_{11}^A - \rho_{22}^A) + (1-\lambda)(\rho_{11}^B - \rho_{22}^B) \right| \\
 &\quad + \left| \lambda(\rho_{11}^A - \rho_{33}^A) + (1-\lambda)(\rho_{11}^B - \rho_{33}^B) \right| \\
 &\quad + \left| \lambda(\rho_{22}^A - \rho_{33}^A) + (1-\lambda)(\rho_{22}^B - \rho_{33}^B) \right| \\
 &\leq \lambda \left| \rho_{11}^A - \rho_{22}^A \right| + (1-\lambda) \left| \rho_{11}^B - \rho_{22}^B \right| \\
 &\quad + \lambda \left| \rho_{11}^A - \rho_{33}^A \right| + (1-\lambda) \left| \rho_{11}^B - \rho_{33}^B \right| \\
 &\quad + \lambda \left| \rho_{22}^A - \rho_{33}^A \right| + (1-\lambda) \left| \rho_{22}^B - \rho_{33}^B \right| \\
 &= \lambda \left\{ \left| \rho_{11}^A - \rho_{22}^A \right| + \left| \rho_{11}^A - \rho_{33}^A \right| + \left| \rho_{22}^A - \rho_{33}^A \right| \right\} \\
 &\quad + (1-\lambda) \left\{ \left| \rho_{11}^B - \rho_{22}^B \right| + \left| \rho_{11}^B - \rho_{33}^B \right| + \left| \rho_{22}^B - \rho_{33}^B \right| \right\} \\
 &= \lambda \mathcal{M}(\boldsymbol{\mu}^A) + (1-\lambda) \mathcal{M}(\boldsymbol{\mu}^B)
 \end{aligned} \tag{A.5}$$

By applying Eq. (A-4). The inequalities (Eqs.A-3) and (A-5)) show that  $\mathcal{M}(\boldsymbol{\mu}(\rho))$  is *convex* when two- and three-dimensional cases are considered. Following a similar procedure, it can be easily verified that  $\mathcal{M}(\boldsymbol{\mu}(\rho))$  is a *convex* function of any  $d$ -dimensional-system. Hence,  $\check{C}_{DM}$  (Eq.(A-1)) is a *concave* function in general.

## Appendix B

### Proof of concavity for $\check{C}_{DM}$ :

Consider  $\rho^A$  and  $\rho^B$  are two pure states in  $\mathcal{H}^d$ . Then from Eq. (3)  $\check{C}_{DM}$  is concave if

$$\check{C}_{DM}(\lambda\rho^A + (1-\lambda)\rho^B) - \lambda\check{C}_{DM}(\rho^A) - (1-\lambda)\check{C}_{DM}(\rho^B) \geq 0. \tag{B.1}$$

$d = 2$ : Let  $\rho^A, \rho^B \in \mathcal{H}^2$  with  $\boldsymbol{\mu}^A = [\rho_{11}^A, \rho_{22}^A]^T$ , and  $\boldsymbol{\mu}^B = [\rho_{11}^B, \rho_{22}^B]^T$ . From Eq. (4)

$$\check{C}_{DM}(\rho^A) = \rho_{11}^A \cdot \rho_{22}^A; \quad \check{C}_{DM}(\rho^B) = \rho_{11}^B \cdot \rho_{22}^B. \tag{B.2}$$

On the other hand, through simple algebra, it can be shown that

$$\begin{aligned}
 \check{C}_{DM}(\lambda\rho^A + (1-\lambda)\rho^B) &= \check{C}_{DM} \begin{bmatrix} \lambda\rho_{11}^A + (1-\lambda)\rho_{11}^B \\ \lambda\rho_{22}^A + (1-\lambda)\rho_{22}^B \end{bmatrix} \\
 &= \lambda^2\check{C}_{DM}(\rho^A) + (1-\lambda)^2\check{C}_{DM}(\rho^B) + \lambda(1-\lambda)\delta_2.
 \end{aligned} \tag{B.3}$$

where  $\delta_2 = \rho_{11}^A \cdot \rho_{22}^B + \rho_{22}^A \cdot \rho_{11}^B$ . Applying Eq. (B-3) in Eq.(B-1), the condition for concavity can be re-written as the following:

$$\begin{aligned}
 \lambda(1-\lambda) \left[ \delta_2 - \check{C}_{DM}(\rho^A) - \check{C}_{DM}(\rho^B) \right] &\geq 0 \\
 \implies \delta_2 - \check{C}_{DM}(\rho^A) - \check{C}_{DM}(\rho^B) &\geq 0.
 \end{aligned} \tag{B.4}$$

since  $\lambda(1-\lambda) \geq 0$ . Now,

$$\begin{aligned}\delta_2 - \check{C}_{DM}(\rho^A) - \check{C}_{DM}(\rho^B) &= \rho_{11}^A \cdot \rho_{22}^B + \rho_{22}^A \cdot \rho_{11}^B - \rho_{11}^A \cdot \rho_{22}^A - \rho_{11}^B \cdot \rho_{22}^B \\ &= (\rho_{11}^A - \rho_{11}^B)(\rho_{22}^B - \rho_{22}^A) \geq 0.\end{aligned}\quad (\text{B.5})$$

since  $\rho_{11}^A + \rho_{22}^A = \rho_{11}^B + \rho_{22}^B = 1$ . Thus,  $\check{C}_{DM}$  fulfills the condition of *concavity* (Eq.B-4) for the case of two-level system.

**$d = 3$ :** Let,  $\rho^A, \rho^B \in \mathcal{H}^3$ , with  $\mu^A = [\rho_{11}^A, \rho_{22}^A, \rho_{33}^A]^T$ , and  $\mu^B = [\rho_{11}^B, \rho_{22}^B, \rho_{33}^B]^T$ . Then,

$$\begin{aligned}\check{C}_{DM}(\rho^A) &= \rho_{11}^A \cdot \rho_{22}^A + \rho_{11}^A \cdot \rho_{33}^A + \rho_{22}^A \cdot \rho_{33}^A \\ \check{C}_{DM}(\rho^B) &= \rho_{11}^B \cdot \rho_{22}^B + \rho_{11}^B \cdot \rho_{33}^B + \rho_{22}^B \cdot \rho_{33}^B.\end{aligned}\quad (\text{B.6})$$

It can be simply shown that the condition for *concavity* for  $\mathcal{H}^3$  is similar to  $\mathcal{H}^2$  (Eq.(B-4)) which is the following:

$$\delta_3 - \check{C}_{DM}(\rho^A) - \check{C}_{DM}(\rho^B) \geq 0, \quad (\text{B.7})$$

with  $\delta_3 = \rho_{11}^A \cdot \rho_{22}^B + \rho_{22}^A \cdot \rho_{11}^B + \rho_{11}^A \cdot \rho_{33}^B + \rho_{33}^A \cdot \rho_{11}^B + \rho_{22}^A \cdot \rho_{33}^B + \rho_{33}^A \cdot \rho_{22}^B$ .

Now, applying Eq. (B-6) in the LHS of Eq.(B-7) we get:

$$\begin{aligned}\delta_3 - \check{C}_{DM}(\rho^A) - \check{C}_{DM}(\rho^B) &= \rho_{11}^A \cdot \rho_{22}^B + \rho_{22}^A \cdot \rho_{11}^B + \rho_{11}^A \cdot \rho_{33}^B + \rho_{33}^A \cdot \rho_{11}^B + \rho_{22}^A \cdot \rho_{33}^B + \rho_{33}^A \cdot \rho_{22}^B \\ &\quad - (\rho_{11}^A \cdot \rho_{22}^A + \rho_{11}^A \cdot \rho_{33}^A + \rho_{22}^A \cdot \rho_{33}^A + \rho_{11}^B \cdot \rho_{22}^B + \rho_{11}^B \cdot \rho_{33}^B + \rho_{22}^B \cdot \rho_{33}^B) \\ &= (\rho_{11}^A - \rho_{11}^B)^2 - (\rho_{22}^A - \rho_{22}^B)(\rho_{33}^A - \rho_{33}^B)\end{aligned}\quad (\text{B.8})$$

Since  $\rho_{11}^A + \rho_{22}^A + \rho_{33}^A = \rho_{11}^B + \rho_{22}^B + \rho_{33}^B = 1$ , then

$$(\rho_{11}^A - \rho_{11}^B) + (\rho_{22}^A - \rho_{22}^B) + (\rho_{33}^A - \rho_{33}^B) = 0. \quad (\text{B.9})$$

Sending the remaining part except  $(\rho_{11}^A - \rho_{11}^B)$  Eq. (B-9) to the right and then squaring both sides of Eq.(B-9) it can be simply shown that

$$(\rho_{11}^A - \rho_{11}^B)^2 \geq (\rho_{22}^A - \rho_{22}^B)(\rho_{33}^A - \rho_{33}^B) \quad (\text{B.10})$$

Applying the above inequality (Eq. (B-10)) in B-8) it gives:  $\delta_3 - \check{C}_{DM}(\rho^A) - \check{C}_{DM}(\rho^B) \geq 0$ , which is the required condition of *concavity* for 3-dimensional case.

By applying mathematical induction, the concavity condition (of  $\check{C}_{DM}$ ) for any general  $d$ -dimensional system can be written as:

$$\delta_d - \check{C}_{DM}(\rho^A) - \check{C}_{DM}(\rho^B) \geq 0 \quad (\text{B.11})$$

where  $\delta_d = \sum_{i=1}^{d-1} \sum_{j=i+1}^d (\rho_{ii}^A \rho_{jj}^B + \rho_{jj}^A \rho_{ii}^B)$ , and the LHS of Eqn. (B-11) can be written as

$$\delta_d - \check{C}_{DM}(\rho^A) - \check{C}_{DM}(\rho^B) = (\rho_{11}^A - \rho_{11}^B)^2 - \sum_{i=2}^{d-1} \sum_{j=i+1}^d (\rho_{ii}^A - \rho_{ii}^B)(\rho_{jj}^A - \rho_{jj}^B) \quad (\text{B.12})$$

Following the same logic applied in  $d = 3$ , it is easy to show that

$$(\rho_{11}^A - \rho_{11}^B)^2 \geq \sum_{i=2}^{d-1} \sum_{j=i+1}^d (\rho_{ii}^A - \rho_{ii}^B)(\rho_{jj}^A - \rho_{jj}^B) \tag{B.13}$$

Thus,  $\delta_d - \check{C}_{DM}(\rho^A) - \check{C}_{DM}(\rho^B) \geq 0$ ; Hence proved.

### Appendix C

#### The study of convexity (for $C_{DD}^P$ and $C_{DM}^P$ ):

According to (C4),  $C_{DM}$  is convex under mixing iff  $\delta_d \geq 0$  with  $d = 2, 3$ , where  $\rho^A$  and  $\rho^B$  are the participating pure states with their mixing probabilities  $\lambda$  and  $1 - \lambda$ , respectively. It is not difficult to see that, by definition,  $C_{DM}$  fulfills convexity (C4) as  $\delta_d \geq 0$ , irrespective of any valid  $\rho^A$  and  $\rho^B$  and for any  $\lambda \in [0, 1]$ . Thus,  $C_{DM}$  naturally falls under the coherence measure category, obeying all five criteria (C1) (C5).

However, in the case of  $d = 3$ , it has been shown (see Theorem 3 of [35]) that  $C_{DM}$  is convex if, for any ensemble  $\{\rho^A, \rho^B, \lambda\}$ , there always exists a pure state  $\rho^C$  such that  $\delta_3 \geq 0$ . Putting it another way,  $C_{DM}$  is convex if  $\delta_3 \geq 0$  (Theorem 4 of ref. [35]). Here, through a few logical steps, we are going to explain why this condition cannot be met for a bona fide coherence measure; in other words, why  $C_{DM}$  is not convex in general. But before that, we redefine  $\delta_3$ , so that the remaining task becomes easier.

The theorem 2 of ref. [35] shows that the set of pure states  $\{\rho^A, \rho^B\}$  (see Theorem 1) corresponding to  $\delta_3 = 0$  can effectively be narrowed down to the subset  $\{\rho^A, \rho^B\}^{opt}$  that also contains the optimal pure state  $\rho^C$ , i.e.,  $\rho^C \in \{\rho^A, \rho^B\}^{opt}$ . In the following, we show that a subset  $\{\rho^A, \rho^B\}^{opt}$  is also available that contains  $\rho^D$ . Before coming to that, we briefly recap the theory of majorization as an important pre-requisite.

**Definition C.1** Majorization: A vector  $\mathbf{p} \in \mathbb{R}^n$  majorizes [48, 53] another vector  $\mathbf{q} \in \mathbb{R}^n$  or  $\mathbf{p} \succcurlyeq \mathbf{q}$  if

$$\sum_{i=1}^l p_i^\downarrow \geq \sum_{i=1}^l q_i^\downarrow, \quad \forall l \in \{1, 2, \dots, n\}$$

where  $p_{[i]}^\downarrow$  and  $q_{[i]}^\downarrow$  are the  $i$ -th elements of  $\mathbf{p}$  and  $\mathbf{q}$ , respectively, when arranged in the descending order, i.e.,  $p_{[1]}^\downarrow \geq p_{[2]}^\downarrow \geq \dots \geq p_{[n]}^\downarrow$  and  $q_{[1]}^\downarrow \geq q_{[2]}^\downarrow \geq \dots \geq q_{[n]}^\downarrow$ .

In quantum resource theory (QRT), majorization is used as an important albeit simple mathematical tool that determines the transformation from one quantum state  $\varphi \in \mathcal{H}^d$  to another state  $\rho \in \mathcal{H}^d$  via free or incoherent operations (IO); if  $\varphi \xrightarrow{IO} \rho$ , then it must be that  $\mu(\rho) \succcurlyeq \mu(\varphi)$  [11, 28, 47]. Again, according to the postulates of the resource theory of coherence, if  $\varphi \xrightarrow{IO} \rho$ , then it is obvious that  $C(\varphi) \geq C(\rho)$  [12].

Therefore, we can translate this very concept of free operation into majorization through the following statement:

$$\begin{aligned} \varphi \xrightarrow{IO} \rho &\iff \mu(\rho) \succcurlyeq \mu(\varphi) \\ &\implies \sum_{i=1}^l \mu^\downarrow(\rho) \geq \sum_{i=1}^l \mu^\downarrow(\varphi) \quad \forall l \in \{1, \dots, d\}. \end{aligned} \tag{C.1}$$

With this piece of information in hand, we are now ready to discuss the further redefinition of  $C_P$ .

**Theorem 2.** *The coherence monotone  $C_P(\rho)$  of a density matrix  $\rho$  with its pure state decompositions  $\{p_i|\psi_i\rangle\}$  can be redefined as*

$$C_P(\rho) = \underset{|\varphi\rangle \in M(\rho)}{\text{all}} \check{C}(|\varphi\rangle) \tag{C.2}$$

where  $\check{C}(\cdot)$  is any bona fide PSCM, and  $M(\rho)$  is the set of pure states  $|\varphi\rangle$  that fulfil the following relation:

$$\mu^\perp(|\varphi\rangle) = \sup_{\{p_i, |\psi_i\rangle\}} \sum_i p_i \mu^\perp(|\psi_i\rangle), \tag{C.3}$$

where the supremum is considered based on majorization.

**Proof of Theorem 2** Let there exist 3 possible pure-state decompositions of  $\rho$ :  $\sum_i p_i(|\psi_i\rangle)$ ,  $\sum_j p_j(|\psi_j\rangle)$ ,  $\sum_k p_k(|\psi_k\rangle)$ ; again, let  $\sum_i p_i \mu^\perp(|\psi_i\rangle) \leq \sum_j p_j \mu^\perp(|\psi_j\rangle) \leq \sum_k p_k \mu^\perp(|\psi_k\rangle)$  (without loss of generality). Now, theorem 2 of [35], says that the pure states  $|\vartheta\rangle$  corresponding to all the different decompositions of  $\rho$  collectively form the pure-state set  $Q(\rho)$  that contains the optimal state  $|\psi_o\rangle$ :  $|\vartheta\rangle \in Q(\rho) \subset R(\rho)$ , and for any member  $|\vartheta_s\rangle$  of  $Q(\rho)$  there exists a decomposition  $\{p_s, |\psi_s\rangle\}$  of  $\rho$  such that  $\mu^\perp(|\vartheta_s\rangle) = \sum_i p_s \mu^\perp(|\psi_s\rangle)$ . It implies that each of the different decompositions of  $\rho$  corresponds to a pure-state set. In this case, let  $\{|\varphi_m\rangle\}_{m \in \{i,j,k\}}$  represent three such sets for the three decompositions, where for any element  $|\varphi_D\rangle$  in the set  $\{|\varphi_m\rangle\}_{m \in \{i,j,k\}}$ , the following relation holds:

$$\mu^\perp(|\varphi_l^m\rangle) = \sum_m p_m \mu^\perp(|\psi_m\rangle) \quad \forall \quad |\varphi_l^m\rangle \in \{|\varphi^m\rangle\}. \tag{C.4}$$

Thus,  $\{|\varphi^m\rangle\} \subseteq Q(\rho)$  for all  $m$ ; and  $\{|\varphi^i\rangle\} \cup \{|\varphi^j\rangle\} \cup \{|\varphi^k\rangle\} = Q(\rho)$ . Again, from the connection between the majorization relation and incoherent transformation (see Eq. C-2), it is clear that  $\{|\varphi^i\rangle\} \xrightarrow{IO} \{|\varphi^j\rangle\} \xrightarrow{IO} \{|\varphi^k\rangle\} \xrightarrow{IO} \sum_k p_k(|\psi_k\rangle)$ . As  $C_P$  always goes for the minimal coherent state, the subset  $\{|\varphi^k\rangle\} (= M(\rho))$  of  $Q(\rho)$  only holds the optimum state  $|\varphi_o\rangle$ ; thus,  $|\varphi_o\rangle \in M(\rho) \subseteq Q(\rho) \subset R(\rho)$ .

Next, to get the optimum state  $|\psi_o\rangle$  from  $M(\rho)$ , it can be emphasized that the coherence vectors in descending order ( $\mu^\perp$ ) for all the elements  $|\varphi_l^m\rangle$  in the  $m$ -th subset  $\{|\varphi^m\rangle\}$  are identical (Eq. C-5). It means that all the pure states in  $M(\rho)$  are of the same coherence; the coherence vectors of any two states in  $M(\rho)$  only differ by a permutation operation  $P_\pi$ . It means that all the states in  $M(\rho)$  can produce the optimum result; therefore, Eq. C-3 is true. The same logic can be applied to any number of possible pure-state decompositions of  $\rho$ , hence proved.

**Theorem 3.** *In general,  $C_P$  does not obey convexity (C4). Hence, it cannot be treated as a coherence-measure class.*

**Proof of Theorem 3** It is seen from Theorem 2 that  $C_P(\rho) = C(|\varphi\rangle)$  where  $|\varphi\rangle$  is any pure state that belongs to  $M(\rho)$ : ( $|\varphi\rangle \in M(\rho)$ ).  $|\varphi\rangle$  is related to the pure-state decomposition of  $\rho$  through the coherence vector s.t  $\mu^\perp(\varphi) = \sup_{\{p_i, |\psi_i\rangle\}} \sum_i p_i \mu^\perp(|\psi_i\rangle)$ . According to the majorization principle (see Eqs. C-1 and C-2), the coherence vector for the decomposition that majorizes all other decompositions of  $\rho$ , gives the least coherence. Thus,  $|\varphi\rangle$  points to the optimal decomposition of  $\rho$  for  $C_P$ ; lets say  $\{p_k, |\psi_k\rangle\}$ ; then,  $C_P(\sum_k p_k(|\psi_k\rangle)) = C_P(\rho) = \check{C}(|\varphi\rangle)$ . As a result,

$$\mu^\perp(|\varphi\rangle) = \sum_k p_k \mu^\perp(|\psi_k\rangle). \tag{C.5}$$

Thus,

$$\check{C}(\mu^\perp(|\varphi\rangle)) = \check{C}\left(\sum_k p_k \mu^\perp(|\psi_k\rangle)\right) \geq \sum_k p_k \check{C}(\mu^\perp(|\psi_k\rangle)) \tag{C.6}$$

The inequality in Eq. C-6 comes from the *concavity (condition-3)* of any valid PSCM,  $\check{C}$ . As  $\check{C}$  must be invariant under any permutation (*condition-2*),  $\check{C}(\mu^\perp(|\theta\rangle)) = \check{C}(\mu|\theta)$  for any pure state  $|\theta\rangle$ , then from Eqs. C-3 and C-6

$$\check{C}(\mu^\perp(|\varphi\rangle)) = \check{C}(|\varphi\rangle) = C_P\left(\sum_k p_k(|\psi_k\rangle)\right) \geq \sum_k p_k \check{C}(|\psi_k\rangle) \tag{C.7}$$

Eq. C-7 implies that  $C_P$  does not satisfy convexity (C-4) in general. As  $C_P$  fulfills all the remaining criteria C1-C3 and C5 [35], it is a bona fide *coherence monotone class*. However, it cannot be treated as a *coherence-measure class*.

It is now apparent from Theorem 3, that  $C_{DD}$  and  $C_{DM}$  are two new *coherence monotones* (not *coherence measures*), as they do not satisfy (C-4). However, there is no problem in considering  $C_{DD}^{conv.roof}$  and  $C_{DM}^{conv.roof}$  as two new *coherence measures*.