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# On Macroscopic Quantum Coherence with Synchronized Atoms and Molecules: Superradiance

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**Abstract:** The collective behavior of quantum particles is one of the most intriguing phenomena in quantum optics. In particular, superradiance refers to spontaneous collective emissions from a group of quantum particles behaving collectively as a whole due to the buildup of macroscopic quantum coherence. An important question is whether macroscopic quantum coherence is constructed by means of a quantum synchronization (i.e., a quantum analog of classical synchronization) or not. The purpose of this article is to draw attention to this question from the author's perspective. A few selected studies relevant to synchronized atoms and molecules are discussed. The author concludes that collective behaviors of quantum particles may be formulated as quantum synchronizations, but extensive studies are still needed to confirm this hypothesis.

**Keywords:** macroscopic quantum coherence; collective spontaneous emissions; Dicke superradiance; superfluorescence; Kuramoto model; classical synchronization; quantum synchronization



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## 1. Introduction

One of the fundamental concepts necessary to understand the principles of quantum technologies is the concept of macroscopic quantum coherence (MQC) [1–3]. Macroscopic quantum coherence shows prevalent quantum coherent behavior at the macroscopic scale, rather than at the individual atomic scale. For example, macroscopic quantum coherence exists in superfluidity, superradiance, and superconductivity. MQC is an extremely fragile state susceptible to rapid environmental disturbances [1–3]. Surprisingly, these disturbances also help to isolate the quantum system from decoherence in the sense that they heat water and, counterintuitively, turn it to ice. Current quantum technologies have been realized at ultralow temperatures [3], at which the environmental disturbances, including dephasing (i.e., decoherence), are rather negligible. In practice, however, dephasing is highly intrinsic and rapidly overwhelms a delicate MQC. Sustaining MQC under ultrafast dephasing at high temperatures is an issue of the utmost importance that has incited recent scientific breakthroughs. MQC in quantum materials was observed first at a high temperature [4] and later at room temperature [5]. To interpret these groundbreaking results, a proposal of the quantum analog of vibration isolation (QAVI) mechanism was postulated [6]. The QAVI mechanism is universal in nature and is proposed to explain both high-temperature superconductivity and room-temperature superfluorescence in solid-state quantum materials. The QAVI mechanism may "potentially lead to a transformation of quantum technologies similar to that of electronics technology in the 20th century" [6]. However, it has yet to be conclusively confirmed. In particular, the important knowledge of whether MQC may be isolated in atomic and molecular gases via this mechanism is still missing. Before considering a QAVI problem, the primary study needs to test the hypothesis that the atomic (also molecular) MQC is initiated by means of a quantum synchronization (QS, a quantum analog of classical synchronization). This hypothesis is formulated by the following reasoning. Synchronization (synch) is a universal phenomenon [7], and it has been proposed in very specific cases of quantum particles [8–10].

From the author's viewpoint, upon successful testing of this hypothesis for QS, the QAVI can be established. Thus, the purpose of this perspective is to draw attention to the question of whether atomic and molecular MQC should be explained in terms of the QS phenomenon or not.

The collective behavior in systems of quantum particles such as atoms, molecules, excitons, and polarons is one of the most intriguing phenomena in quantum optics [2,11,12]. In particular, superradiance (SR), or spontaneous collective emission, is produced when a group of atoms behaves collectively as a giant dipole due to the buildup of MQC among atoms [13–19]. In the case of SR, MQC is studied by measuring the temporal and spatial characteristics of collective emissions from quantum particles. Creating and sustaining MQC in atomic systems in the realistic environment is particularly important for future practical quantum technologies. MQC among atoms numbering in the trillions was observed [20,21]. MQC may have the potential to be used in developing an ultranarrow (in mHz) linewidth laser, which, in turn, may be used to test the fundamental constants in physics [22–24] and facilitate the entangled light sources for quantum information and computational sciences [1,3,25,26]. The SR process is sketched in Figure 1A. SR is initiated spontaneously from distinguishable (non-synched) atoms in the inverted system via quantum fluctuations [27–32]. At a certain time, maximum MQC is formed, and all atoms become indistinguishable (synched) to emit an SR pulse. Thus, the atoms fall to their ground state and no MQC feature is left. However, the knowledge of a transition near threshold from non-synched to synched atoms is still lacking. This perspective recognizes this knowledge gap. Throughout the text, by the term SR, the author refers to the collective phenomenon; in general, this includes Dicke SR [13,33,34], SR in an extended medium [35,36], and superfluorescence (SF) [27,28,37–39] for a two-level atomic system and, if not specified, also cascade SR [40], cascade SF [21,41,42], and yoked SF [29–31,39,43–45] from a cascade three-level atomic system. It is also not the author's intention to review the topic of SR, as the recent reviews are reported in [15,16] (see references therein).

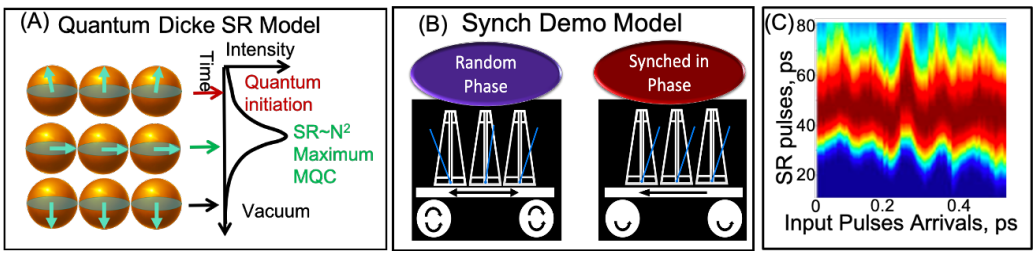


**Figure 1.** (**A**) Collective spontaneous emissions. A pictorial explanation of superradiance. (**B**) Synch demo. A sketch of a system of oscillating three metronomes and platform. (**C**) Data for a temporal coherent control of atomic macroscopic quantum coherence.

## 2. Superradiance and Synch Phenomenon

The universality of the fascinating classical synch phenomenon, such as the collective dynamics of fish schools and bird flocks, synched flashing of fireflies, synched neurons, synched galaxies and more [7], will become a breakthrough for the macroscopic quantum world of synched atoms. A classic demo of synch is a system consisting of the N number of oscillating metronomes on a common platform that is capable of oscillating. A sketch of a system with N = 3 metronomes on a platform (a thin wooden board) isolated from the ground by two soda cans is shown in Figure 1B. The metronomes start swinging in random phase; however, eventually they catch up and start oscillating in phase. Thus, the metronome-platform system becomes stable. The most relevant properties of this system to SR are as follows [46,47]. (i) The synch is a classical phase transition phenomenon, wherein phase I consists of distinguishable metronomes, and phase II consists of them becoming indistinguishable; hence, they represent a giant oscillating "metronome." (ii) The synch is initiated from instability and it takes time. (iii) The synch needs a feedback. In this demo, it is the platform that couples each metronome. The platform is able to oscillate but

at a rather slower pace during which the fast oscillations (that some metronomes could have) are filtered out. In this sense, the platform naturally becomes a vibration isolator or shock absorber. When the platform is restricted to vibrate (placed on the floor), metronomes start oscillating randomly on their own. Being a universal phenomenon [7], the synch may be extended to the quantum world at atomic level. The equations representing the synch are given by the known Kuramoto model [47].

For the sake of better understanding the question raised by the author in this perspective, the following oversimplified classical-to-quantum correspondences may be assumed. For example, atoms are metronomes. The platform is assumed to be a radiation field (a cavity mode) emitted by excited atoms in the case of SR initiation due to quantum fluctuations. The platform feedback means there are atom–light interactions. This feedback consideration is questionable and entirely missing in any type of SR description and must be properly addressed in the future. Classical phase transition corresponds to QPT. Classical instability corresponds to the quantum fluctuations originating from atom–vacuum interactions, and, similarly, classical VI to QAVI. The SR classical equation, in terms of single collective Bloch vector dynamics [13] for indistinguishable atoms [16], is equivalent to the equation in the Kuramoto model [47] for all synched oscillators. Recently, classical and quantum Kuramoto, and classical and quantum Dicke SR Hamiltonian models have been emerging [48–56]. For example, Akkermans et al. concluded in [48] that in "photon cooperative emission results from the synchronization of the atomic dipoles induced by long range atomic correlations, this connexion becomes even more interesting and relevant". Indeed, what is missing is how the classical (and quantum) oscillators synch (become indistinguishable) from unsynched (distinguishable) oscillators. Therefore, an all-inclusive quantum analogy to synch based on the abovementioned three main properties is needed to qualify to be QS phenomenon.

Furthermore, the question of whether SR is a QPT phenomenon or not remains open [16,57–60]. An inclusion of the rotating wave approximation (RWA) in the Dicke Hamiltonian (not reduced to the indistinguishable atoms) is, therefore, important. For example, with RWA, the no-go theorem [60] rules out the QPT. The research to test whether the buildup of atomic MQC is a QPT phenomenon analogous to classical synch requires development of the new theoretical SR model analogous to the synch model. The Kuramoto model [47] will serve as a simple tool by which to understand the essentials of synch [7]. In general, synch is an open problem for a system of a large N number of oscillators because of the $2^N$ (exponential) complexity that no classical computer can solve. However, an approximation of considering only nearest-neighbor coupled oscillators, which has a polynomial complexity, can reveal the main characteristics of synch. On the other hand, fully quantum mechanical treatment for SR [12] is also an open problem due to the complexity of non-commuting atomic operators. To overcome the insolvable exponential complexity, the Dicke Hamiltonian is either reduced to the indistinguishable atoms [13,14,61] or uses "mean-field" approximation [12,15]. Analogously, the effective SR Hamiltonian, in which only atomic operators are involved, can be solved by using the nearest-neighbor approximation. This formalism using the atomic operators [48,61] has proven to be a powerful tool. Dicke's classical equation [16] already bears a hint that it is equivalent to Kuramoto's in-phase equation [47]. To date, no known analogy has been conducted for the Dicke and Kuramoto models. The author suggests that the Kuramoto model involves stochastic noise terms for both Brownian and Ornstein–Uhlenbeck processes. under the nearest-neighbor approximations.

## 3. Macroscopic Quantum Coherence with Synched Atoms

Meeting the specific criteria to generate SR (and thus, build up MQC), which requires a high number of atoms to become indistinguishable and tolerant to their environment, is manifold for condensed matter [16] compared to atomic gas. In atomic gas, SR reflects to moderate homogenous spectral linewidth broadening, a small atomic dephasing rate [62] and a relatively highly efficient inversion of the atomic density [12]. Therefore, MQC in

atomic gases will be discussed in this section. Exploration of atomic MQC study has been the main tool of the SR phenomenon [21,27,28,37,38,63]. In particular, atomic MQC study in the time domain has been emergent as a research topic [20,21,28,30,31,36,41,43–45,64–74]. For example, the temporal coherent control of SR was demonstrated for the first time in [30]. In Figure 1C, the streak camera data for temporal profiles of SR pulses versus time arrivals of the input fs laser pulses is shown. When the input pulses overlap, the buildup of MQC occurs abruptly, whereas when they do not overlap, the buildup of MQC occurs much later. These data contain the following three SR quantities: (i) the SR time delay, (ii) the SR peak intensity, and (iii) the SR pulse width. These quantities, thus, can be controlled by the input double pulses with variable delay.

To date, the following have recently been accomplished: (i) observations of the shortest-ever SR emissions from Rb [44], Na [31], and Cs [45] atomic vapors; (ii) observations of quantum fluctuations for Rb [29,30]; (iii) observations of quantum fluctuations for Na [31], for the first time; (iv) introduction and demonstration of an SR temporal coherent control for the first time [30]; (v) observations of SR quantum beatings in Rb [30,36,71,74] and Cs [45]; (vi) introduction of a new cascade SR model [40]; and (vii) observations of a cascade SF [21].

An atomic MQC study in an ultrafast time domain requires the fs laser system, including optical parametric amplifiers, a high-temporal resolution streak camera, and atomic vapor cells (see Figure 2A). The typical SR temporal and spatial profiles are depicted in Figures 2B and Figure 2C, respectively. The rings in Figure 2C are called conical emissions, due to spatially extended, excited atoms. The SR time delay is ideally determined as relative to the arrival time of the excitation pulse. Nevertheless, measuring the "absolute" average SR time delay is still a challenging task, and to address this challenge, a pair of SR pulses are used [21,29,31,40]. In Figure 2D, a non-interfering pair of SR pulses was observed to suitably obtain not only average SR time delay [31], but also quantum fluctuations in a time delay that is relative to each other as in Figure 2E [29]. Distributions of time delay differences are shown in Figure 2F) [29].

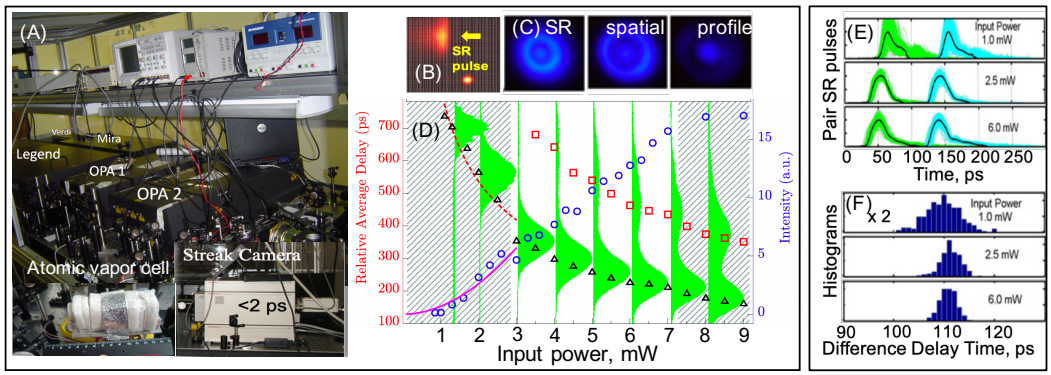


**Figure 2.** (**A**) A typical experimental setup for superradiance. A femtosecond laser system is used to excite a heated-up atomic vapor. The emitted superradiance pulses are detected by a picosecond streak camera. (**B**) A typical temporal profile of the recorded picosecond superradiance pulse relative to the reference pulse. (**C**) Spatial beam profiles of the superradiance light on a paper screen far away from the sample. For higher excitation power, the conical emissions are formed because of multiple transverse modes involved in the overall process. For lower power, superradiance emission is directional originating from a cigar-shaped sample. (**D**) Superradiance time delays and peak intensities as functions of input power. As power increases, delay time decreases, whereas peak intensity increases nonlinearly. In this particular case, pairs of superradiance pulses, originating from two macroscopically extended groups of atoms, are recorded. (**E**) To measure fluctuations in delay times, which have quantum mechanical origin, pairs of pulses are needed to obtain relative delays for each laser shot. (**F**) Distributions of time delay differences.

## 4. Macroscopic Quantum Coherence with Synched Molecules

Collective emission processes in a molecular system are much more complex than those for an atomic system. From a physicist's point of view, the collective phenomenon is less studied for the molecular system. This is a very vast topic when it comes to the molecular level; therefore, with this perspective, the author focuses mainly on the pump-probe type of experimental setup. A pump-probe study of molecular dynamics is the key method in femtochemistry [75]. Typical pump-probe experiments are not quite suitable to reveal a collective phenomenon in the molecular system. Therefore, it is the author's intention to interpret the well-known coherent Stokes and anti-Stokes Raman scattering (CSRS and CARS) processes [76,77] in terms of the synch phenomenon. A typical three-color CARS process involves four waves: two excitation pulses (pump pulses) prepare a number of molecules vibrating in phase synchronization, and then the third pulse (probe) scatters off from phased oscillators (molecules) to generate the fourth, which is either red-shifted (CSRS) or blue-shifted (CARS) relative to the probe pulse center wavelength [78–88]. In this case, it brings up the natural question of how long it takes to synch molecular vibrations after pumping arises. This question has recently been addressed. Namely, the existence of molecular MQC buildup was theoretically predicted [82,83] and experimentally observed for benzene and pyridine molecular systems [86] for the first time. The molecular MQC data are shown in Figure 3A,B. Analytical (Figure 3A) and experimental (Figure 3B) data and good agreement (Figure 3C) are shown as functions of probe width and delay [86].

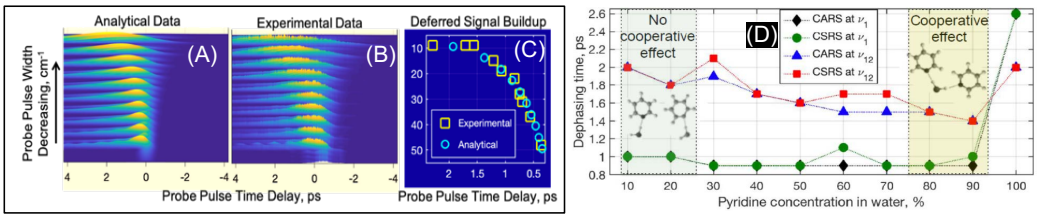


**Figure 3.** (**A**–**C**) Data for the new effect: A deferred macroscopic quantum coherence buildup in a molecular system. (**A**) Analytical results based on the Faddeeva function for a pair of vibrational ring modes of pyridine molecules. (**B**) Experimental results as probe pulse width varies but pump pulses remain unchanged. (**C**) A comparison between analytical and experimental data for the triangular ring vibrational mode of pyridine molecules. (**D**) A cooperative effect observed in pyridine–water complexes. At higher concentration of pyridine molecules, more water molecules tend to bond with pairs of pyridine molecules, and effectively escalate dephasing rate of the mixture.

To reveal molecular MQC in this particular experimental setup, a new two-dimensional correlation analysis (2DCA) has been introduced [84,85,87,88]. This new approach to reveal molecular MQC has been further generalized to pyridine–water complexes [85,87]. An interesting collective behavior was observed while studying pyridine complexes (see Figure 3D). At a higher pyridine concentration in the pyridine–water mixture, the dephasing of synched molecules is escalated. The reason is interpreted in terms of a collective phenomenon. At this concentration ratio, a specific complex is dominated wherein a single additional water molecule tends to make a hydrogen bonding to a pair of pyridine molecules. The shaded area in Figure 3D shows decreased dephasing for both vibrational ring modes [87]. As the number of water molecules increases, this perfect pairing is eventually disrupted. In this case, MQC is twofold: (i) macroscopic coherence for pyridine molecules, and (ii) macroscopic coherence of paired pyridine molecules. One of the advantages of this method is that both CARS and CSRS are measured simultaneously. In this sense, the CARS/CSRS noise correlation spectroscopy was also introduced [84].

## 5. Conclusions

Superradiance is known as spontaneous collective emission from a group of atoms behaving collectively as a giant dipole due to the buildup of atomic macroscopic quantum coherence. This article focuses on the question of whether macroscopic quantum coherence is initiated by means of a quantum analog of classical synchronization or not. A few selected examples relevant to synchronized atoms and molecules has been discussed. In current models for collective behaviors of quantum particles, a detailed understanding of how quantum oscillators become synched (or become indistinguishable) after being unsynched (distinguishable) is missing. This perspective recognizes this knowledge gap. In conclusion, collective behaviors in an ensemble of atoms and molecules may be formulated as quantum synchronizations; however, all-inclusive, rigorous theoretical and experimental research studies are still needed to confirm this hypothesis.

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## Abbreviations

The following abbreviations are used in this manuscript:

| | |
|---|---|
| MDPI | Multidisciplinary Digital Publishing Institute |
| DOAJ | Directory of open access journals |
| QAVI | Quantum Analog of Vibration Isolation |
| SR | Superradiance |
| MQC | Macroscopic Quantum Coherence |
| Synch | Synchronization |
| QS | Quantum Synchronization |

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