Testing the quantum superposition principle in the frequency domain

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New technological developments allow to explore the quantum properties of very complex systems, bringing the question of whether also macroscopic systems share such features, within experimental reach. The interest in this question is increased by the fact that, on the theory side, many suggest that the quantum superposition principle is not exact, departures from it being the larger, the more macroscopic the system. Here we propose a novel way to test the possible violation of the superposition principle, by analyzing its effect on the spectral properties of a generic two-level system. We will show that spectral lines shapes are modified, if the superposition principle is violated, and we quantify the magnitude of the violation. We show how this effect can be distinguished from that of standard environmental noises. We argue that accurate enough spectroscopic experiments are within reach, with current technology.

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Introduction. – Quantum theory is an extremely successful theory of microscopic phenomena. Yet, still an open question is if and how it applies also to macroscopic systems. Indeed it would be absolutely fascinating to see quantum mechanical behavior for a macroscopic object. But until today macroscopic systems are always found to behave according to classical mechanics and the open question is exactly if they can show quantum behaviour or if the quantum superposition principle is violated at some level. Here we show how such violations would modify spectral lineshapes of atoms and molecules, and we propose a new class of spectroscopic experiments to test such modifications. These violations have a universal character, which cannot be controlled by experimental parameters. We will show how this specific signature can be used to distinguish them from decoherence.

In general, when performing a quantum experiment, two different limitations can be identified. We distinguish the technological and the fundamental limitations. Technological limitations are induced through coupling of the quantum system with the environment. Decoherence theory explains how environmental noise can rapidly destroy quantum coherence before it can be even observed 1. For macroscopic systems we can safely expect a plethora of decohering interactions and it will be a technological challenge to isolate large quantum systems from such interactions. On the other hand fundamental limitations are due to intrinsic non-linearity in the quantum dynamics that are induced e.g., by the gravity as in the Diósi-Penrose model 2, or by a generic stochastic field as in the Continuous Spontaneous Localization model 3–5, 51. In all such models, Schrödinger dynamics is modified by adding non-linear terms, in such a way that quantum coherence is preserved at the microscopic level, while it is destroyed when approaching the macroscopic scale 6–9.

Recently, there has been rapid experimental progress in revealing quantum features such as particle-wave duality for large objects with tiny de Broglie wavelength of only a few hundred femtometer. Such objects were successfully decoupled from environmental noises, thus overcoming the technological limit and thereby extending the realm of quantum theory to new regimes 10–18. This progress not only confirms the predictions of decoherence theory, but also provides the possibility to search for intrinsic non-linearity.

Current experiments are mainly focused on the preparation of macroscopic systems in a spatial quantum superposition state. Non-linearity would then manifest as loss of visibility in observed interference pattern 5. Promising schemes to observe such non-linearity are provided by optomechanical techniques 15, 16 and matter-wave interferometry 17, 18. They have not yet found any evidence for intrinsic non-linearity, however allow to introduce upper bounds on their strength 17. The definite test of the existence of fundamental limitations to the quantum superposition principle will require quantum interference with single particles of mass 106 – 109 amu 16, 17, 18.

Here, we provide an alternative test of fundamental limitations by measuring light emitted by radiative transitions of excited states of matter. We compute modifications to the standard quantum optical formulation of spontaneous photoemission, assuming quantum linearity is violated. We derive a analytical formula for both shift and broadening of spectral lines, which is applicable to a broad class of systems. We explicitly compute these effects for specific models and then quantify them for relevant physical systems. We show that observation of line broadening induced by intrinsic non-linearity is at the reach of current technology.

The frequency shift and broadening. – Very similar to vacuum fluctuations, intrinsic non-linearity produces two
FIG. 1: Collapse models predicts that the broadening \( \beta_c \) depends only on the geometry of involved states, and scales quadratically with the mass of the system (see Eq. 3). On the other hand, decoherence broadening in general scales differently with the mass, and moreover depends on the details of the interaction with the environment. In Fig. (a) we show the different scaling behavior between CSL broadening and decoherence broadening (in the recoil-free regime \( M \gg m \)) while the environment does not change and the system increases in mass. Assuming the density of the system to remain constant so that \( r \propto M^{1/3} \) (\( r \) is the radius of the systems and \( M \) its mass), then in the Hard sphere limit one has for collisional broadening \[ \beta_c = 4d^2p\sqrt{\pi/mk_BT} \propto M^{2/3} \] (where \( d \sim r \) is the closet distance among the colliding particles, \( p \) the pressure of the bath, \( T \) its temperature and \( m \) the mass of bath particles). In the inset, the dependence of the experimental Hard sphere radius on mass is shown, for \( H_2, \) He, NH, Ne, \( N_2, \) O, Ar, CO, Kr, and Xe \[ 28] \). The behaviour is qualitatively similar to the oversimplified \( M^{1/3} \) dependence we estimated, therefore the mass dependence of \( \beta_c \) is different from the \( M^2 \) behavior of the CSL broadening. In Fig. (b) we assume the opposite situation. The system does not change and the bath particle increases in mass (still in the recoil-free regime \( M \gg m \)). In such a case, CSL broadening remains constant, while decoherence broadening scales as \( m^{-1/2} \).

types of radiative corrections in the emitted light spectrum: frequency shift and broadening. Both phenomena appear naturally in QED analysis of the spectrum of spontaneously emitted light. We formulate the intrinsic non-linearity in terms of a stochastic potential added to the Schrödinger equation, since observable effects of non-linearity, at the statistical level, can be reproduced to the Schrödinger equation, since observable effects of non-linearity in terms of a stochastic potential added. This is a lengthy calculation, fully reported in \[ 19 \]; here, we present only the final result for the stochastic averaging approximation, given by \[ H_0 = h \sum_{s,k} \omega_k \hat{\alpha}_s^\dagger \hat{\alpha}_s \hat{\alpha}_s + (\hbar\omega_0/2)\hat{\sigma}_z + \hbar\omega_0 \sum_{s,k} \{ g_{s,k} \hat{\sigma}_y \hat{\alpha}_s \hat{\alpha}_s \hat{\alpha}_s - H.C. \}, \] with \( g_{s,k} = (2\hbar\hbar\omega_L)^{-1/2} d_{12} \cdot \hat{e}_{s,k} \). Here the system’s Hamiltonian is \( (\hbar\omega_0/2)\hat{\sigma}_z \), the position operator is proportional to \( \hat{\sigma}_x = \hat{\sigma}_+ + \hat{\sigma}_- \), and the transition dipole matrix element between two levels is not zero \( (d_{12} = (\varepsilon_1, d_{12} \neq 0) \). All other terms have the usual meanings. According to the Wiener-Khinchin theorem \[ 27 \], the light spectrum is given by the Fourier transform of the normalized dipole-dipole autocorrelation function. Measurable quantities are obtained after taking the stochastic averages. Therefore, all we need is to solve the corresponding differential equations for dipole-dipole autocorrelation functions. This is a lengthy calculation, fully reported in \[ 19 \]; here, we present only the final result for the stochastic averaged equations that we need to compute the dipole-dipole autocorrelation. They read:

\[
\frac{d}{dt} \mathbb{E}(\hat{\sigma}_g(t)\hat{\sigma}_g(t+\tau)) = \Omega_{\text{QED}} \mathbb{E}(\hat{\sigma}_x(t)\hat{\sigma}_x(t+\tau))
\]

\[
-\beta_{\text{QED}} + 2\lambda_x + 2\lambda_z \mathbb{E}(\hat{\sigma}_x(t+\tau)\hat{\sigma}_x(t))
\]

\[
\frac{d}{dt} \mathbb{E}(\hat{\sigma}_x(t)\hat{\sigma}_x(t+\tau)) = -\Omega_{\text{QED}} \mathbb{E}(\hat{\sigma}_y(t+\tau)\hat{\sigma}_z(t))
\]

\[
-\beta_{\text{QED}} + 2\lambda_x \mathbb{E}(\hat{\sigma}_x(t+\tau)\hat{\sigma}_x(t))
\]

where \( \beta_{\text{QED}} = \omega_0^2 |d_{12}|^2 / 6\pi\varepsilon_0\hbar c^2 \) is the standard spontaneous
photo-emission rate and \( \Omega_{\text{QED}} = \omega_0 - \gamma_{\text{QED}} \) with 
\[
\gamma_{\text{QED}} = \frac{\omega_0^2 |d_{12}|^2}{8\epsilon_0e^2c^3} \left( \mathcal{P} \int_0^\infty \frac{d\omega}{\omega - \omega_0} - \mathcal{P} \int_0^\infty \frac{d\omega}{\omega + \omega_0} \right)
\]
The Lamb shift (where \( \mathcal{P} \) is Cauchy principal part). Accordingly, for the dipole-dipole autocorrelation function we find:
\[
E(\langle \hat{\sigma}_+(t + \tau) \hat{\sigma}_-(t) \rangle) = e^{-i(2\lambda_0 + 2\lambda_3)\tau} \cos \Omega \tau + i \frac{\lambda_0 \Omega_{\text{QED}}}{\Omega} \sin \Omega \tau \langle \hat{\sigma}_+(t + \tau) \hat{\sigma}_-(t) \rangle
\]
with \( \Omega = \sqrt{\Omega_{\text{QED}}^2 - \lambda_2^2} \) for \( \Omega_{\text{QED}} > \lambda_2 \). For \( \Omega_{\text{QED}} \gg \lambda_2 \), which is the case for most cases of experimental interest, we can Taylor-expand \( \Omega_{\text{QED}}/\Omega \) to the first leading term in \( \lambda_2/\Omega_{\text{QED}} \), and we get:
\[
E(\langle \hat{\sigma}_+(t + \tau) \hat{\sigma}_-(t) \rangle) \approx e^{-i(2\lambda_0 + 2\lambda_3)\tau} e^{i\Omega_{\text{QED}}/\Omega} E(\langle \hat{\sigma}_+(t) \hat{\sigma}_-(t) \rangle).
\]
Using Wiener-Khinchin relation, for the spectral density of emitted radiation we obtain:
\[
S(\omega) = \frac{1}{\pi} \left( \frac{\beta}{\beta^2 + (\omega - \Omega)^2} \right),
\]
which is a Lorentzian distribution with the central frequency \( \Omega \approx \Omega_{\text{QED}} - \gamma_N \) where \( \gamma_N = \lambda_2^2/2\omega_0 \) is the shift induced by intrinsic non-linearity; and the full width at half-maximum of \( 2\beta = 2\lambda_{\text{QED}} + 2\beta_N \) where \( \beta_N = \lambda_2 + 2\lambda_3 \) is the broadening induced by intrinsic non-linearity. For \( \beta_N = 0 \), the standard quantum optical spectrum is recovered. Accordingly, the radiative corrections due to the modified dynamics manifest both as frequency shift and broadening. We shall see that broadening is much bigger than the shift.

Quantifying the shift and the broadening. – We now obtain the shift and the broadening from two most studied collapse models in the literature: the mass proportional Continuous Spontaneous Localization (CSL) model \[3, 6, 11\] and the Diósi-Penrose (DP) gravitation model \[2\]. For both models, we have \[62\]:
\[
\hat{L}(x) = \int dy e^{-|x-y|^2/2\gamma^2} \sum_{j=1}^N m_j \hat{a}_j^+(y) \hat{a}_j(y)
\]
where the sum is over the type of particles (electrons and nucleons, in the nonrelativistic limit), \( r_C \approx 10^{-7} \) m the correlation length, and \( \hat{a}_j(y) \) is the annihilation operator of a particle with mass \( m_j \) at the position \( y \).

Regarding Eq. \[15\], for the CSL model we have \( \lambda = \gamma/m_0^2 \) (with \( m_0 = 1 \) amu, \( \gamma \approx 10^{-22} \text{cm}^2/\text{s} \) \[3\]) and \( \xi_t(x) \) is a white noise field with correlation \( E(\xi_t(x)\xi_s(y)) = \delta(t_1 - t_2)\delta(x-y) \). For the DP model, \( \lambda = 1 \) and \( E(\xi_t(x)\xi_s(y)) = G \delta(t-s)/h|x-y| \) with \( G \) the gravitational constant.

Taking into the account Eq. \[2\] and the correlation properties of the noises, one can show that:
\[
E(\langle \hat{V}_1 | \hat{V}_1 | \hat{V}_2 | \hat{V}_2 \rangle) - E(\langle \hat{V}_2 | \hat{V}_1 | \hat{V}_2 | \hat{V}_1 \rangle) \equiv 2\lambda_2 \gamma^2 \delta(t_1 - t_2) \quad \text{and} \quad E(\langle \hat{V}_1 | \hat{V}_1 | \hat{V}_2 | \hat{V}_2 \rangle) = \lambda_3 \gamma^2 \delta(t_1 - t_2).
\]
From these relations, together with Eq. \[6\], one can obtain the analytical expression for the broadening \( \beta_N \) and the shift \( \gamma_N \). We compute the shift and broadening for the CSL model and DP model when the region in which the amplitude of the eigenstates is different from zero, is smaller than \( r_C \approx 10^{-7} \) m, which is valid for most cases of interest such as molecules.

After a lengthy computation, reported in \[19\], we get:
\[
\beta_N = \frac{\Lambda}{2r_C m_0^2} \left( D_{12}^2 + \frac{M}{2} (I_2 - I_1) \right),
\]
\[
\gamma_N = \frac{\Lambda^2}{8\xi_0} \left( \frac{D_{12}^2}{r_C m_0^2} \right)^4,
\]
with \( I_\alpha = \langle \varepsilon_\alpha(\sum_{j=1}^N m_j q_j^2) | \varepsilon_\alpha \rangle \) the average momentum of inertia (\( \alpha = 1, 2, \) and \( N \) the total number of particles), \( D_{12} = \langle \varepsilon_2(\sum_{j=1}^N m_j q_j) | \varepsilon_1 \rangle, M = \sum_{j=1}^N m_j \) the total mass, and \( \Lambda \) is the coupling constant with the collapse or gravitational field. For the CSL model we get \( \Lambda_{\text{CSL}} \approx 1.12 \times 10^{-9} \text{s}^{-1} \) and for the DP model \( \Lambda_{\text{DP}} \approx 7.39 \times 10^{-25} \text{Hz} \) \[19\]. From now on, we consider only CSL as the effect of DP model is by far smaller.

Universality of broadening. – A crucial feature of Eqs. \[8\] and \[9\] is that both broadening and shift induced by collapse models are universal, in the sense that they depend only on the mass of the system (at the practical level, only on the mass of those particles, whose position changes significantly during the transition) and on the geometry of the levels, and nothing else. This has to be compared with decoherence broadening and shift, which depends also on the details of the surrounding environment: mass of the bath particle, cross section, pressure, temperature. Moreover, in case of collapse models, they roughly scale quadratically with the mass of the system, while the mass dependence with decoherence is different. This behavior represents a specific signature, which can be used to discriminate collapse broadening from decoherence broadening (see Fig.1).

Experimental implication. – In general, spectroscopy can be done in any domain of the electro-magnetic spectrum and any physical system with discrete energy levels is potentially good for tests. This means in principle any degree of freedom of particles: electronic, vibration or rotation, as well as collective rearrangements of atoms in bigger structures such as conformation changes of molecules or any other many-body system such as atoms in optical lattices as well as any internal degree of freedom of a condensed matter system such as spin can be used as a probe. Spectroscopy techniques with very different degree of resolution exist in such different frequency domains. Surely, to resolve the broadening effect one needs not only to resolve the spectral line centres as typically done in precision frequency spectroscopy, one also needs to resolve the width of the spectral line with the same resolution.

Resolution of the spectroscopic detection system. To detect small deviations as those shown in Table 1, one needs ultra-high resolution spectroscopic techniques. In gen-
TABLE I: Collapse broadening and shift as predicted by the CSL model. Three relevant situations have been considered: the transition from the 2P to 1S state in a Hydrogen-like atom; the transition from the first excited state to the ground state for a harmonic potential, and for a double-well potential (see supplementary material for a description of these systems). The latter case is particularly relevant to describe chiral molecules. The constant Λ ≃ 2πℏ/mω0 is the separation of the minima of double-well potential. For the double-well potential, we assume the range of the molecular vibration: ω0 ≃ 10^{12}−10^{14} Hz. We have considered only the predictions of the CSL model, since those of gravity induced models (Diosi-Penrose) are much smaller. Note, that all numbers in this table are exemplary to illustrate the magnitude of the spectral effects, and not necessarily realised in experiments yet.

| System                     | β_N (Hz)   | γ_N (Hz)  |
|---------------------------|------------|-----------|
| Hydrogen-like Atoms        | 10^{-20}−10^{-18} | ~10^{-53} |
| Harmonic oscillator       | 5.3×10^{-13} | 6.2×10^{-36} |
| μ = 1 amu and ω₀ = 10^{10} Hz |            |           |
| μ = 10^{7} amu and ω₀ = 1.7 × 10^{8} Hz | 3.1×10^{-4} | 1.3×10^{-16} |
| Double-well               | 4.2×10^{-23} | 10^{-57} − 10^{-55} |
| μ = m_e × 5.5 × 10^{-4} amu and q₀ = 1 Å |            |           |
| μ = 1 amu and q₀ = 1 Å    | 1.4×10^{-16} | 10^{-44} − 10^{-42} |
| μ = 10^{7} amu and q₀ = 1 Å | 0.014 | 10^{-16} − 10^{-18} |

eral to observe small spectral effects the quality factor (Q = ω/β) of the spectroscopic system has to be as high as possible. State of the art for frequency stability of a laser (1/Q) is of the order 10^{-16} in the visible spectral range, which can be transferred to other frequency domains in principle by frequency combs. However, most recently the frequency stability for an atomic frequency measurement in the visible/near-infrared spectral range [25], which can be transferred to other frequency domains, would need to be detected with a spectral resolution of about 30 µHz to show a nonlinear effect, which is only three orders of magnitude away from today's resolution [26]. Further relevant are solid state systems, like semiconductor microcavities, quantum dots, or nanodiamonds with vacancy centers and spin structure as well as opto-mechanical systems. Conclusion. – The mass proportional CSL model predicts a measurable spectral line broadening effect. Ultrastable lasers as used in state-of-the-art experiments are sufficient to observe this effect, if the appropriate system can be identified, according to the significant parameters: spectral lifetime and mass. Competing line-broadening decoherence effects can be controlled by maintaining experimental conditions. In general and in contrast to decoherence effects, CSL broadening scales quadratically with mass and does not depend on experimental controllable parameters. This opens the door for systematic investigations on the hunt for intrinsic non-linearities and a
Supplementary information

Here we provide an extensive analysis on how modifying Schrödinger equation with non-linearities, changes the spectral density of emitted radiation from a general two-level system. We will show that the non-linearity manifests as a broadening and shift in the spectral density of emitted radiation. We first discuss how the non-linearity can be introduced into the dynamics. Then, following standard QED formulations, we obtain the broadening and the shift induced by non-linearity. We finally compute analytically and quantify these radiative corrections from two most studied collapse models.

S1: Dynamical equations.

It was first proven by Gisin \cite{34} that, in order to avoid superluminal signalling, nonlinear terms can be added to the Schrödinger equation only combined with stochastic terms, in such a way that the equivalence relation among statistical ensembles of states is preserved by the dynamics \cite{35–37}. In more mathematical terms, this means that the modified dynamics for the wave function must generate a closed linear dynamics for the density matrix. Given these premises, it was recently proven \cite{38} that such a dynamics must be of the Lindblad type. It is important to notice that in the proof, complete positivity—usually requested in order to derive Lindblad’s theorem—is not a necessary hypothesis, but comes about from the existence of a (Markovian) dynamics for the wave function. Therefore, we start from a dynamics for the density matrix of the form:

\[
\frac{d\hat{\rho}_t}{dt} = -\frac{i}{\hbar} [\hat{H}_0, \hat{\rho}_t] + \lambda \sum_{k=1}^{n} \left( \hat{L}_k \hat{\rho}_t \hat{L}_k^\dagger - \frac{1}{2} \hat{L}_k \hat{L}_k^\dagger \hat{\rho}_t - \frac{1}{2} \hat{\rho}_t \hat{L}_k^\dagger \hat{L}_k \right)
\] (10)

where the Lindblad operators \( \hat{L}_k \) can describe decoherence effect or, as it is the case here, intrinsic non-linearities in the dynamics for the wave function. Collapse models induce a dynamics of this type, but here we want to stay more general. The most convenient unraveling of Eq. (10) for solving the equations of motions, is given in terms of a stochastic potential added to the Schrödinger equation \cite{39–43}:

\[
i\hbar \frac{d}{dt} \psi_t = \left[ \hat{H}_0 + \hat{V}_t \right] \psi_t, \quad \hat{V}_t = -\hbar \sqrt{\lambda} \sum_{k=1}^{n} \hat{L}_k \xi^{(k)}_t,
\] (11)

where \( \xi^{(k)}_t \) are \( n \) independent white noises. Here, we have assumed that the Lindblad operators \( \hat{L}_k \) are self-adjoint, which is the case for most proposals for nonlinear and stochastic modifications of the Schrödinger equation.

Since with violations of the superposition principle we mean superpositions in space, the Lindblad operators are taken as functions of space variables, therefore we have:

\[
\hat{V}_t = -\hbar \sqrt{\lambda} \int d^3x \, \hat{L}(x) \xi_t(x),
\] (12)

where \( \xi_t(x) \) is a noise-field, white both in space and time. Note that, in this form, the dynamical equation is still linear. As discussed several times in the literature \cite{39–43}, the effects of nonlinear terms introduced in the Schrödinger equation, at the statistical level, can be mimicked also by linear random potentials. For individual realizations of the noise, the affects are very different (those of a linear dynamics vs those of a nonlinear one), while at the statistical level they coincide, if the potential is suitably chosen.
We consider the situation in which the system’s dynamics effectively involves only two levels, whose transition dipole matrix element is not zero \( \langle d_{12} \rangle = \langle \varepsilon_1 | \mathbf{d} | \varepsilon_2 \rangle \neq 0 \), with the dipole operator defined as \( \mathbf{d} = \sum_i \varepsilon_i \mathbf{d}_i \). This is means that the higher energy level eventually decays to the lower one, by emitting a photon. Therefore, the standard quantum Hamiltonian characterizing the interaction between a two-level system and a quantized radiation field can be, in the dipole approximation, written in the form [18] [17]:

\[
\hat{H}_0 = \hbar \sum_{s,k} \omega_s \hat{a}_s^\dagger \hat{a}_s + (\omega_0 / 2) \hat{\sigma}_z - i \omega_0 \sum_{s,k} \{ g_{s,k} (\hat{\sigma}_+ - \hat{\sigma}_- \hat{a}_s^\dagger \hat{a}_s - \text{H.C.}) \},
\]

with \( g_{s,k} = (2 \varepsilon_0 \hbar \omega L^3)^{-1/2} d_{12} \cdot \mathbf{e}_s \cdot \mathbf{e}_k \) the coupling constant of radiation-matter, \( \hat{\sigma}_+ = |\varepsilon_2 \rangle \langle \varepsilon_1 | \), \( \hat{\sigma}_- = |\varepsilon_1 \rangle \langle \varepsilon_2 | \), \( \hat{\sigma}_z = \hat{\sigma}_+ \hat{\sigma}_- - \hat{\sigma}_- \hat{\sigma}_+ \), \( \omega_0 = (\varepsilon_2 - \varepsilon_1) / \hbar \) and \( \{ |\varepsilon_1 \rangle, |\varepsilon_2 \rangle \} \) the two levels of matter. All other terms have the usual meanings.

The two-level representation of \( \hat{H}_0 \) is obtained by calculating \( \langle \varepsilon_\alpha | \hat{V}_i | \varepsilon_\beta \rangle \) with \( \alpha, \beta = 1, 2 \). In general, one has:

\[
\hat{V}_i = -\hbar \left( \sqrt{\lambda_x} w_t^{(z)} \hat{\sigma}_z + \sqrt{\lambda_y} w_t^{(x)} \hat{\sigma}_x + \sqrt{\lambda_z} w_t^{(y)} \hat{\sigma}_y \right)
\]

where \( \lambda_i (i = x, y, z) \) are collapse rates and \( w_t^{(i)} \) are three white noises. Eigenenergies are real functions in most cases, thus one finds: \( \lambda_y = 0 \). Therefore \( \hat{V}_i \) simplifies to:

\[
\hat{V}_i = -\hbar \left( \sqrt{\lambda_x} w_t^{(z)} \hat{\sigma}_z + \sqrt{\lambda_x} w_t^{(x)} \hat{\sigma}_x \right).
\]

S3: Solution of the equations of motion: shift and broadening.

The radiative corrections of the non-linearities appear in a very natural way from the formulation of the spectral density of emitted light [14] [17] which is given by the stochastic expectation (averaging) of the Fourier transform of the normalized dipole-dipole autocorrelation function. The dipole-dipole autocorrelation function is given by \( \rho_\omega (\omega) \) thus one finds:

\[
\hat{\sigma}_z(t) = \int d\omega \omega \rho_\omega (\omega) \hat{a}_s^\dagger \hat{a}_s.
\]

With the Hamiltonian \( \hat{H} = \hat{H}_0 + \hat{V}_i \), the Heisenberg equations of motion for the system operators take the forms:

\[
\frac{d}{dt} \hat{\sigma}_z(t) = -2 \beta_{QED} \left( \hat{\sigma}_z(t) + \hat{I} \right) - 2 \omega_0 \left( \hat{\sigma}_z(t) \mathbf{d}_{12} \cdot \hat{A}^{(+)}_{\text{free}} (0, t) + \mathbf{d}_{12} \cdot \hat{A}^{(-)}_{\text{free}} (0, t) \hat{\sigma}_x(t) \right) - 2 \sqrt{\lambda_x} w_t^{(z)} \hat{\sigma}_y(t),
\]

\[
\frac{d}{dt} \hat{\sigma}_y(t) = - \left( \Omega_{QED} - 2 \sqrt{\lambda_x} w_t^{(z)} \right) \hat{\sigma}_x(t) - \beta_{QED} \hat{\sigma}_y(t) + 2 \sqrt{\lambda_x} w_t^{(x)} \hat{\sigma}_z(t),
\]

\[
\frac{d}{dt} \hat{\sigma}_x(t) = - \left( \Omega_{QED} - 2 \sqrt{\lambda_x} w_t^{(z)} \right) \hat{\sigma}_y(t) - \beta_{QED} \hat{\sigma}_x(t) + 2 \omega_0 \left( \hat{\sigma}_z(t) \mathbf{d}_{12} \cdot \hat{A}^{(+)}_{\text{free}} (0, t) + \mathbf{d}_{12} \cdot \hat{A}^{(-)}_{\text{free}} (0, t) \hat{\sigma}_z(t) \right),
\]

where last terms on the right hand side of Eqs. [16] and [17] are the new contributions to the dynamics due to the noise terms describing nonlinear effects. Other terms are obtained using standard derivations given by quantum statistical theories of spontaneous emission [18] [17]. In the above equations, \( \Omega_{QED} = \omega_0 - \gamma_{QED} \) where

\[
\gamma_{QED} = \frac{\omega_0^3 |\mathbf{d}_{12}|^2}{3 \varepsilon_0 \hbar \pi^2 \varepsilon^3} \left( \mathcal{P} \int_0^\infty \frac{d\omega \omega}{\omega - \omega_0} - \mathcal{P} \int_0^\infty \frac{d\omega \omega}{\omega + \omega_0} \right),
\]

is the Lamb shift, with \( \mathcal{P} \) is the Cauchy principal part, which can be normalized in the standard fashion [18]; the parameter

\[
\beta_{QED} = \frac{\omega_0^3 |\mathbf{d}_{12}|^2}{6 \pi \varepsilon_0 \hbar c^3}
\]

is the standard spontaneous emission rate; and:

\[
\mathbf{d}_{12} \cdot \hat{A}^{(+)}_{\text{free}} (0, t) = \sum_{s,k} g_{s,k} e^{-i \omega_t} \hat{a}_s^\dagger (0).
\]
We now average Eqs. (16) and (17) over the initial state |ψ⟩|0⟩, according to which matter is in a generic state |ψ⟩ and the radiation field in the vacuum state. Therefore we get:

\[
\frac{d}{dt} \langle \hat{a}_z (t) \rangle = -2\beta_{\text{QED}} \langle \hat{a}_z (t) \rangle - 2\sqrt{\lambda_x} w_t^{(x)} \langle \hat{a}_y (t) \rangle
\]

(22)

\[
\frac{d}{dt} \langle \hat{a}_y (t) \rangle = \left( \Omega_{\text{QED}} - 2\sqrt{\lambda_x} w_t^{(z)} \right) \langle \hat{a}_x (t) \rangle - \beta_{\text{QED}} \langle \hat{a}_y (t) \rangle + 2\sqrt{\lambda_x} w_t^{(x)} \langle \hat{a}_z (t) \rangle
\]

(23)

\[
\frac{d}{dt} \langle \hat{a}_x (t) \rangle = - \left( \Omega_{\text{QED}} - 2\sqrt{\lambda_x} w_t^{(z)} \right) \langle \hat{a}_y (t) \rangle - \beta_{\text{QED}} \langle \hat{a}_x (t) \rangle.
\]

(24)

The above stochastic differential equations should be understood in Stratonovich sense. Since we want to compute stochastic averages, it is more convenient to switch to the Itô formalism. To this end, one can use Eqs. (10.2.5) to (10.2.7) of Ref. [49]. Then, once expressed in the Itô form, by using theorem (8.5.5) of Ref. [49], one can prove that the stochastic expectations satisfy the following equations:

\[
\frac{d}{dt} \mathbb{E}(\langle \hat{a}_x (t) \rangle) = -2(\beta_{\text{QED}} + \lambda_x) \mathbb{E}(\langle \hat{a}_x (t) \rangle) - 2\beta_{\text{QED}}
\]

(25)

\[
\frac{d}{dt} \mathbb{E}(\langle \hat{a}_y (t) \rangle) = \Omega_{\text{QED}} \mathbb{E}(\langle \hat{a}_x (t) \rangle) - (\beta_{\text{QED}} + 2\lambda_x + 2\lambda_z) \mathbb{E}(\langle \hat{a}_y (t) \rangle)
\]

(26)

\[
\frac{d}{dt} \mathbb{E}(\langle \hat{a}_x (t) \rangle) = -\Omega_{\text{QED}} \mathbb{E}(\langle \hat{a}_y (t) \rangle) - (\beta_{\text{QED}} + 2\lambda_z) \mathbb{E}(\langle \hat{a}_x (t) \rangle)
\]

(27)

Using the solutions of above equations, one can find:

\[
\mathbb{E}(\langle \hat{a}_x (t) \rangle) = \left( \frac{\beta_{\text{QED}}}{\beta_{\text{QED}} + \lambda_x + \langle \hat{a}_z (0) \rangle} \right) e^{-2(\beta_{\text{QED}} + \lambda_x) t} - \frac{\beta_{\text{QED}}}{\beta_{\text{QED}} + \lambda_x},
\]

(28)

\[
\mathbb{E}(\langle \hat{a}_y (t) \hat{a}_x (t) \rangle) = \frac{1}{2} \left( \frac{\beta_{\text{QED}}}{\beta_{\text{QED}} + \lambda_x + \langle \hat{a}_z (0) \rangle} \right) e^{-2(\beta_{\text{QED}} + \lambda_x) t} + \frac{\lambda_x}{\beta_{\text{QED}} + \lambda_x},
\]

(29)

where \( h\omega_0 \mathbb{E}(\langle \hat{a}_z (t) \rangle) \) gives the rate of energy emission by matter; and \( \mathbb{E}(\langle \hat{a}_x (t) \hat{a}_x (t) \rangle) \) represents the change in the population of the excited state |\varepsilon_2\rangle. When the initial state is |\varepsilon_1\rangle, we have: \( \langle \hat{a}_z (0) \rangle = -1 \), and for |\varepsilon_2\rangle, we have: \( \langle \hat{a}_z (0) \rangle = 1 \). On the other hand, for both initial states |\varepsilon_{1,2}\rangle we get:

\[
\mathbb{E}(\langle \hat{a}_x (t) \hat{a}_x (t) \rangle) = \mathbb{E}(\langle \hat{a}_y (t) \hat{a}_y (t) \rangle) = 0 \quad \text{for} \quad |\varepsilon_{1,2}\rangle |0\rangle.
\]

(30)

Using Eq. (29), one can compute for example the mean light intensity of emitted radiation in the far-field limit [41-47] as follows:

\[
\langle \hat{I}(r, t) \rangle = \left( \frac{\omega_0^2 |\mathbf{d}_{12}|}{8\pi\varepsilon_0 c^2 r} \right)^2 \left( 1 - \frac{1}{2} \sin^2 \theta \right) \left[ \left( \frac{\beta_{\text{QED}}}{\beta_{\text{QED}} + \lambda_x + \langle \hat{a}_z (0) \rangle} \right) e^{-2(\beta_{\text{QED}} + \lambda_x) (t - \frac{r}{c})} + \frac{\lambda_x}{\beta_{\text{QED}} + \lambda_x} \right], \quad t > \frac{r}{c},
\]

(31)

with \( \theta \) the polar angle of the r-vector, and the complex dipole moment \( \mathbf{d}_{12} \) lies in the \( xy \)-plane, where \( r \) is the vector connecting the center-of-mass of system to the detector. \( \langle \hat{I}(r, t) \rangle \) is a very interesting quantity for experimental research; however, here we are concerned with the spectral density of emitted light, which we now compute.

The exponential nature of energy decay, as given by Eqs. (28) and (31), suggests that the spectral distribution of the emitted radiation is Lorentzian. The explicit mathematical form of spectral density can be obtained by computing the dipole-dipole autocorrelation function, \( \mathbb{E}(\langle \hat{a}_x (t + \tau) \hat{a}_x (t) \rangle) \), and then using the Wiener-Khinchin theorem [44]. The time derivative of this autocorrelation function can be obtained by making the change \( t \rightarrow t + \tau \) in Eqs. (16), (17) and (18), and then writing down the derivatives with respect to \( \tau \). After multiplying the result from the right with \( \hat{a}_x (t) \) and then taking the quantum average over the initial state |ψ⟩|0⟩, we find:

\[
\frac{d}{d\tau} \langle \hat{a}_z (t + \tau) \hat{a}_x (t) \rangle = -2\beta_{\text{QED}} \langle \hat{a}_z (t + \tau) \hat{a}_x (t) \rangle - \langle \hat{a}_x (t) \rangle - 2\sqrt{\lambda_x} w_{\tau}^{(x)} \langle \hat{a}_y (t + \tau) \hat{a}_z (t) \rangle,
\]

(32)

\[
\frac{d}{d\tau} \langle \hat{a}_y (t + \tau) \hat{a}_x (t) \rangle = \left( \Omega_{\text{QED}} - 2\sqrt{\lambda_x} w_{\tau}^{(z)} \right) \langle \hat{a}_x (t + \tau) \hat{a}_x (t) \rangle - \beta_{\text{QED}} \langle \hat{a}_y (t + \tau) \hat{a}_x (t) \rangle + 2\sqrt{\lambda_x} w_{\tau}^{(x)} \langle \hat{a}_z (t + \tau) \hat{a}_x (t) \rangle + 2\sqrt{\lambda_x} w_{\tau}^{(z)} \langle \hat{a}_x (t + \tau) \hat{a}_y (t + \tau) \hat{a}_z (t + \tau) \rangle,
\]

(33)

\[
\frac{d}{d\tau} \langle \hat{a}_x (t + \tau) \hat{a}_x (t) \rangle = -\beta_{\text{QED}} \langle \hat{a}_x (t + \tau) \hat{a}_x (t) \rangle - \langle \Omega_{\text{QED}} - 2\sqrt{\lambda_x} w_{\tau}^{(z)} \rangle \langle \hat{a}_y (t + \tau) \hat{a}_x (t + \tau) \rangle - \beta_{\text{QED}} \langle \hat{a}_x (t + \tau) \hat{a}_x (t + \tau) \rangle + 2\sqrt{\lambda_x} w_{\tau}^{(x)} \langle \hat{a}_z (t + \tau) \hat{a}_x (t + \tau) \rangle.
\]

(34)
Using the aforementioned theorems to switch between Stratonovich and Itô forms and also to obtain the stochastic expectations, and also taking into the account the result given by Eq. (30), we find:

\[
\frac{d}{dt} \mathbb{E}(\langle \hat{\sigma}_z(t + \tau)\hat{\sigma}_z(t) \rangle) = -2(\beta_{\text{QED}} + \lambda_z) \mathbb{E}(\langle \hat{\sigma}_z(t + \tau)\hat{\sigma}_z(t) \rangle)
\]

\[
\frac{d}{dt} \mathbb{E}(\langle \hat{\sigma}_y(t + \tau)\hat{\sigma}_y(t) \rangle) = \Omega_{\text{QED}} \mathbb{E}(\langle \hat{\sigma}_y(t + \tau)\hat{\sigma}_y(t) \rangle) - (\beta_{\text{QED}} + 2\lambda_z + 2\lambda_z) \mathbb{E}(\langle \hat{\sigma}_y(t + \tau)\hat{\sigma}_y(t) \rangle)
\]

\[
\frac{d}{dt} \mathbb{E}(\langle \hat{\sigma}_x(t + \tau)\hat{\sigma}_x(t) \rangle) = -(\beta_{\text{QED}} + 2\lambda_z) \mathbb{E}(\langle \hat{\sigma}_x(t + \tau)\hat{\sigma}_x(t) \rangle) - \Omega_{\text{QED}} \mathbb{E}(\langle \hat{\sigma}_x(t + \tau)\hat{\sigma}_x(t) \rangle)
\]

Accordingly, for the dipole-dipole autocorrelation function we get:

\[
\mathbb{E}(\langle \hat{\sigma}^\dagger_+ + \tau \hat{\sigma}_- - \tau \rangle) = e^{-(\beta_{\text{QED}} + \lambda_z + 2\lambda_z)\tau} \left( \cosh(i\Omega\tau) + \frac{\Omega_{\text{QED}} \sinh(i\Omega\tau)}{\Omega} \right) \mathbb{E}(\langle \hat{\sigma}^\dagger_+ \hat{\sigma}_- - \rangle)
\]

\[
\mathbb{E}(\langle \hat{\sigma}^\dagger_+ + \tau \hat{\sigma}_- - \tau \rangle) = e^{-(\beta_{\text{QED}} + \lambda_z + 2\lambda_z)\tau} \left( \cos(\Omega\tau) + i \frac{\Omega_{\text{QED}}}{\Omega} \sin(\Omega\tau) \right) \mathbb{E}(\langle \hat{\sigma}^\dagger_+ \hat{\sigma}_- - \rangle)
\]

with \( \Omega = \sqrt{\Omega_{\text{QED}}^2 - \lambda_z^2} \) for \( \Omega_{\text{QED}} > \lambda_z \). For \( \Omega_{\text{QED}} \gg \lambda_z \), which is the case for most cases of experimental interest, we can expand \( \Omega_{\text{QED}}/\Omega \) to the first leading term in \( \lambda_z/\Omega_{\text{QED}} \), and we get:

\[
\mathbb{E}(\langle \hat{\sigma}^\dagger_+ + \tau \hat{\sigma}_- - \tau \rangle) \approx e^{-(\beta_{\text{QED}} + \lambda_z + 2\lambda_z)\tau} e^{i\Omega\tau} \mathbb{E}(\langle \hat{\sigma}^\dagger_+ \hat{\sigma}_- - \rangle)
\]

Using Wiener-Khinchin relation, for the spectral density of emitted radiation we obtain:

\[
S(\omega) = \frac{1}{\pi} \left( \frac{\beta_{\text{QED}} + \lambda_z + 2\lambda_z}{(\beta_{\text{QED}} + \lambda_z + 2\lambda_z)^2 + (\omega - \Omega)^2} \right) = \frac{1}{\pi} \left( \frac{\beta}{\beta^2 + (\omega - \Omega)^2} \right),
\]

with the full width at half-maximum of \( \beta \) where

\[
\beta = \beta_{\text{QED}} + \beta_N, \quad \text{with} \quad \beta_N = \lambda_z + 2\lambda_z
\]

and the central frequency of

\[
\Omega = \sqrt{\Omega_{\text{QED}}^2 - \lambda_z^2} \approx \Omega_{\text{QED}} - \frac{\lambda_z^2}{2\Omega_{\text{QED}}} \approx \Omega_{\text{QED}} - \frac{\lambda_z^2}{2\omega_0} = \Omega_{\text{QED}} - \Omega_N, \quad \text{with} \quad \Omega_N = \frac{\lambda_z^2}{2\omega_0}.
\]

Accordingly, the radiative corrections given by violations of the quantum superposition principle produce two observable effects: a frequency-shift and a line broadening, whose magnitude is controlled by the rates \( \lambda_{x,z} \). Their numerical value depends on the specific model used to describe nonlinear (collapse) effects.

S4: Calculation of rates \( \lambda_{x,z} \)

We now derive the rates \( \lambda_{x,z} \) as predicted by the two most-studied collapse models in the literature: the mass proportional Continuous Spontaneous Localization (CSL) model [52], and the Diósi-Penrose gravitational (DP) model [53, 54].

Rates for the CSL model. The stochastic potential \( \hat{V}_i \) associated to the CSL model is [50–52]:

\[
\hat{V}_i = -\hbar \sqrt{\gamma/m_0} \int dx \xi_i(x) \hat{L}(x), \quad \text{with} \quad \hat{L}(x) = \int dy g(x - y) \sum_j m_j \sum_s \hat{d}^\dagger_s(s,y) \hat{d}_s(s,y),
\]

where \( m_0 = 1 \text{ amu}, \gamma \approx 10^{-22} \text{cm}^3\text{s}^{-1} \) [55], \( \xi_i(x) \) is a white noise with correlation \( \mathbb{E}(\xi_i(x)\xi_r(y)) = \delta(t - \tau)\delta(x - y) \), and \( \hat{d}^\dagger_s(s,y) \) is the annihilation operator of the particle type-\( s \) with mass \( m_j \) and the spin \( s \) at position \( y \); and \( g(r) = \exp(-r^2/2r_c^2)/((\sqrt{2\pi}r_c)^3) \) with \( r_c \approx 10^{-5} \text{cm} \) the correlation length.

In the two-level representation, the matrix elements of \( \hat{V}_i \) are given by:

\[
V^\alpha_\beta = \langle \varepsilon_{\alpha} | \hat{V}_i | \varepsilon_{\beta} \rangle = -\hbar \sqrt{\gamma/m_0} \int dQ \psi^\alpha(Q) \psi^\beta(Q) \int dx \xi_i(x) \sum_{j=1}^N m_j g(x - q_j),
\]
with \( \alpha, \beta = 1, 2 \), and \( \psi_{\alpha}(Q) = (Q|\xi_{\alpha}) \) where we use improper states: \( |Q\rangle \equiv |q_1; q_2; \cdots; q_N\rangle \) (with \( q_j \) the position of \( j \)-th particle) for which we have: \( \hat{L}(x)|Q\rangle = \left( \sum_{j=1}^{N} m_j \, g(x - q_j) \right) |Q\rangle \). We also assume that the wave functions \( \psi_{\alpha} \) are real. Since the right side of Eq. (46) contains a Gaussian white noise, \( \lambda_{x,z} \) can be calculated as follow:

\[
\mathbb{E} \left( V_{t_1}^{\alpha\beta} V_{t_2}^{\alpha'\beta'} \right) = \frac{\delta(t_1 - t_2)}{8\pi^3/2r_C^3} \int dQ \, dQ' \, \psi_\alpha(Q) \psi_\beta(Q) \psi_{\alpha'}(Q') \psi_{\beta'}(Q') \sum_{j,l=1}^{N} \frac{m_j \, m_l}{m_0^2} \exp \left[ -\frac{(q_j - q'_l)^2}{4r_C^2} \right],
\]

We consider the situation where the effective size of the region in which \( \psi_{1,2} \) is different from zero is smaller than \( r_C \approx 10^{-7} m \), which is the case for atomic and molecules systems. This is the small scale limit of the CSL model. Accordingly, by expanding the exponential term in Eq. (46) to first order in \( (q_j - q'_l)^2/4r_C^2 \), and then by performing the integrations, we get:

\[
\lambda_{11}^{11} \approx \frac{\Lambda_{\text{CSL}} \, M^2}{m_0^2} \left( 1 - \frac{1}{M} \int dQ \, |\psi_1(Q)|^2 \sum_j m_j \left( \frac{q_j}{2r_C} \right)^2 \right),
\]

\[
\lambda_{22}^{22} \approx \frac{\Lambda_{\text{CSL}} \, M^2}{m_0^2} \left( 1 - \frac{1}{M} \int dQ \, |\psi_2(Q)|^2 \sum_j m_j \left( \frac{q_j}{2r_C} \right)^2 \right),
\]

\[
\lambda_{12}^{12} = \lambda_{21}^{21} \approx \frac{\Lambda_{\text{CSL}}}{2r_C^2 \, m_0^2} \left( \int dQ \, \psi_1(Q) \psi_2(Q) \sum_j m_j q_j \right)^2,
\]

with \( \Lambda_{\text{CSL}} = \gamma/(8\pi^3/2r_C^3) = 1.12 \times 10^{-9} s^{-1} \) and \( M = \sum_j m_j \). In the derivation of above equations, we used the parity considerations and the orthogonality of \( \psi_1 \) and \( \psi_2 \). Accordingly, we have:

\[
\lambda_z = \frac{\lambda_{11}^{11} - \lambda_{22}^{22}}{2} = \frac{\Lambda_{\text{CSL}} \, M}{8r_C^2 \, m_0^2} \int dQ \, \left( |\psi_2(Q)|^2 - |\psi_1(Q)|^2 \right) \sum_j m_j \, q_j^2,
\]

\[
\lambda_x = \lambda_{12}^{12} = \frac{\Lambda_{\text{CSL}}}{2r_C^2 \, m_0^2} \left( \int dQ \, \psi_1(Q) \psi_2(Q) \sum_j m_j q_j \right)^2.
\]

S5: Rates for the Diósi-Penrose (DP) model

The stochastic potential in the DP model is given by:

\[
\hat{V}_t = -\hbar \int dx \, \xi_t(x) \hat{L}(x),
\]

where \( \hat{L}(x) \) is the same like the CSL model, and \( \xi_t(x) \) is a white noise with correlation \( \mathbb{E}(\xi_t(x)\xi_s(y)) = G \, \delta(t - s)/h |x - y| \) with \( G \) the gravitational constant. This form of \( \hat{L}(x) \) is different from Diósi’s original proposal [53]. To avoid the divergence due to the Newton self-energy, Diósi initially proposed a Lindblad operator with a length cutoff equal to the nuclear size. Then, it was shown [50] that with this cutoff, predictions of the DP model are in contradiction with known observations. To avoid this problem, Ghirardi, Grassi and Rimini [50] proposed this new form of Lindblad operator whose cutoff is \( r_C \approx 10^{-7} m \). This adjustment of the model was eventually acknowledged by Diósi [57].

For the matrix elements of \( \hat{V}_t \) in two-level representation, one gets:

\[
V_t^{\alpha\beta} = \langle \varepsilon_\alpha | \hat{V}_t | \varepsilon_\beta \rangle = \hbar \int dQ \, \psi_\alpha(Q) \psi_\beta(Q) \int dx \, \xi_t(x) \sum_{j=1}^{N} m_j \, g(x - q_j),
\]
Following the same approach that we used for the CSL model, we find:

\[
E \left(V_{t_1}^{\alpha\beta} V_{t_2}^{\alpha'\beta'} \right) = \delta(t_1 - t_2) G \hbar \int dQ dQ' \psi_\alpha(Q) \psi_\beta(Q) \psi_\alpha'(Q') \psi_\beta'(Q') \times \\
\sum_{j,l} m_j m_l \int \frac{dx dx'}{|x - x'|} g(x - q_j) g(x' - q'_l) \\
= \delta(t_1 - t_2) G \hbar \frac{4\pi}{4\pi} \int dQ dQ' \psi_\alpha(Q) \psi_\beta(Q) \psi_\alpha'(Q') \psi_\beta'(Q') \sum_{j,l} m_j m_l \int dx g(x) \Phi_{jl}(x) \\
= \delta(t_1 - t_2) \hbar^2 \lambda^{\alpha\beta}_{\alpha'\beta'}
\]

with

\[
\Phi_{jl}(x) = \frac{\text{erf} \left( \frac{|x - (q_j - q'_l)|}{\sqrt{2r_C}} \right)}{|x - (q_j - q'_l)|},
\]

where \(\text{erf}\) is the error function. Here \(\Phi_{jl}(x)\) is slowly varying with respect to \(g(x)\) (for more detail, see Section (4.2) of Ref. [58]). Therefore \(g(x)\) acts like a Dirac-delta, practically selecting the value of \(\Phi_{jl}(x)\) in the origin \(x = 0\). Accordingly, we can write:

\[
E \left(V_{t_1}^{\alpha\beta} V_{t_2}^{\alpha'\beta'} \right) \simeq \delta(t_1 - t_2) G \hbar \frac{4\pi}{4\pi} \int dQ dQ' \psi_\alpha(Q) \psi_\beta(Q) \psi_\alpha'(Q') \psi_\beta'(Q') \sum_{j,l} m_j m_l \text{erf} \left( \frac{|q_j - q'_l|}{\sqrt{2r_C}} \right) \\
|q_j - q'_l| \ll r_C.
\]

Like before, we are interested in the cases where the spatial width of eigenenergies \(\psi_{1,2}\) are smaller than \(r_C\), meaning \(|q_j - q'_l| \ll r_C\). This implies that \(\Phi_{jl}(0)\) can be Taylor expanded, and to the leading order, one finds:

\[
\text{erf} \left( \frac{|q_j - q'_l|}{\sqrt{2r_C}} \right) \simeq \frac{2}{r_C \sqrt{2\pi}} \left( 1 - \frac{|q_j - q'_l|^2}{6r^2_C} \right), \text{ where } |q_j - q'_l| \ll r_C.
\]

Then, one can follow the same line of reasoning that we followed from Eq. (46) to Eqs. (50) and (51) to solve the rest of integrations. Accordingly, the rates \(\lambda_{x,z}\) of the DP model become:

\[
\lambda_z = \frac{\Lambda_{dp}}{8r^2_C m_0^3} \int dQ \left( |\psi_2(Q)|^2 - |\psi_1(Q)|^2 \right) \sum_j m_j q_j^2,
\]

\[
\lambda_x = \lambda^{12}_{12} = \frac{\Lambda_{dp}}{2r^2_C m_0^3} \left( \int dQ \psi_1(Q) \psi_2(Q) \sum_j m_j q_j \right)^2.
\]

with \(\Lambda_{dp} = \frac{G m_0^3}{3\sqrt{2}\pi^{3/2} \hbar r_C} \approx 7.39 \times 10^{-25} \text{ s}^{-1}\). These rates have the same form as those of the CSL model, but with a different coupling constant, which is \(10^{-16}\) times smaller than the CSL coupling constant, and therefore negligible. Accordingly, in the subsequent analysis we will report just the CSL values.

**S6: Application to relevant physical systems.**

We now provide quantitative estimation of these rates for some interesting physical systems.

**Hydrogen-like atoms.** For an atom that contains only one electron, we have: \(D_{12} = (m_e/e) d_{12}\) with \(m_e\) the mass and \(e\) the charge of electron, and \(d_{12}\) the off-diagonal element of the dipole moment with typical values of a few Debye. In addition, \(\sqrt{I_{12}/m_e}\) has typical values of a few Bohr radius. Accordingly, we get:

\[
\lambda_z \approx 10^{-20} - 10^{-18} \text{ s}^{-1}; \quad \lambda_x \approx 10^{-23} - 10^{-21} \text{ s}^{-1}.
\]

For example, for the transition \(2P \rightarrow 1S\) of the hydrogen atom, where the emitted light is a K-level X-ray radiation [59], we find: \(\lambda_z \approx 5.2 \times 10^{-19} \text{ s}^{-1}\) and \(\lambda_x \approx 1.4 \times 10^{-22} \text{ s}^{-1}\).
Harmonic oscillator. We consider the two lowest states of a harmonic oscillator with mass $\mu$ and frequency $\omega_0$. Introducing these eigenstates into Eqs. (50) and (51) and performing the integration, we find:

$$\lambda_x = 4\lambda_z = \frac{\Lambda}{2} \left( \frac{\mu x_0}{m_0 v_C} \right)^2,$$

(62)

with $x_0 = \sqrt{\hbar/\mu \omega_0}$ the zero-point fluctuation amplitude.

Double-well potential. We consider a system of mass $\mu$ moving in a symmetric double-well potential at low temperatures, where the meaningful eigenstates are the two lowest ones: $|\varepsilon_1\rangle = \frac{1}{\sqrt{2}} (|R\rangle + |L\rangle)$ and $|\varepsilon_2\rangle = \frac{1}{\sqrt{2}} (|R\rangle - |L\rangle)$. The tunnelling frequency is $\omega_0 = (\varepsilon_2 - \varepsilon_1)/\hbar$. We denote the separation of minima by $q_0$. The states $|L\rangle$ and $|R\rangle$ are localized states at left and right minima, respectively. They can be transformed to each other by the displacement operator where the displacement distance is $q_0$, and $\langle L|R\rangle \approx 0$. Accordingly, using Eqs. (50) and (51) and performing the integration, we find:

$$\lambda_x \approx 0; \quad \lambda_z \approx \frac{\Lambda}{8} \left( \frac{\mu q_0}{m_0 v_C} \right)^2.$$

(63)

For example, some internal motions of non-rigid molecules and complexes (e.g., the inversion motion in Ammonia [60] or hindered torsional rotation in X-Y-Y-X molecules [61]) can be effectively described by a double-well potential. For these systems, we have $q_0 \sim 1 - 10 \AA$ and $\mu \sim 1 - 100$amu [60, 61]. Accordingly, the order of magnitude of the strongest collapse rate for the internal motions of molecular systems is $\lambda_x \sim 10^{-9}$s$^{-1}$.
[38] Bassi, A., Duerr, D., Hinrichs, G. Uniqueness of the equation for state-vector collapse. arXiv:1303.4284 (2013).

[39] Gisin, N. Quantum measurements and stochastic processes. Phys. Rev. Lett. 52, 1657 (1984).

[40] Diósi, L. Continuous quantum measurement and Ito formalism. Phys. Lett. A 129, 419 (1988).

[41] Adler, S. L., Horwitz, L. P. Structure and properties of Hughston’s stochastic extension of the Schrödinger equation. J. Math. Phys. 41, 2485 (2000).

[42] Adler, S. L. Statistical Dynamics of Global Unitary Invariant Matrix Models as Pre-Quantum Mechanics. hep-th/0206120 (2002), Section 5F.

[43] Adler, S. L. Weisskopf-Wigner decay theory for the energy-driven stochastic Schrödinger equation. Phys. Rev. D 67, 025007 (2003), Added note.

[44] Heitler, W. Quantum theory of radiation. 3rd Edition, International Series of Monographs on Physics, Oxford: Clarendon, (1954), Sections 16, 17, 18 and 34.

[45] Agarwal, G. S. Quantum Statistical Theories of Spontaneous Emission and their Relation to other Approaches. Springer Tracts Mod. Physics 70 (Springer, Berlin 1974).

[46] Garrison, J. C. and Chiao, R. Y. Quantum Optics. Oxford Univ. Press (2008), Chapters 4 and 11.

[47] Mandel, L. and Wolf, E. Optical Coherence and Quantum Optics. Cambridge Univ. Press (1995), Chapter 15.

[48] Bethe, H. A. The electromagnetic shift of energy levels. Phys. Rev. 72, 339 (1947).

[49] Arnold, L. Stochastic Differential Equations: Theory and Applications. Dover Publications (2013).

[50] Ghirardi, G. C., Rimini, A., and Weber, T. Unified dynamics for microscopic and macroscopic systems. Phys. Rev. D 34, 470-491 (1986).

[51] Ghirardi, G.C., Pearle, P., and Rimini, A. Markov processes in Hilbert space and continuous spontaneous localization of systems of identical particles. Phys. Rev. A 42, 78 (1990).

[52] Ghirardi, G.C., Grassi, R. and Benatti, F. Describing the macroscopic world: closing the circle within the dynamical reduction program. Found. Phys. 25, 5-38 (1995).

[53] Diósi, L. A universal master equation for the gravitational violation of the quantum mechanics. Phys. Lett. A 120, 377-381 (1987).

[54] Penrose, P. On gravity’s role in quantum state reduction. Gen. Rel. Grav. 28, 581 (1996).

[55] Adler, S. L. Lower and upper bounds on CSL parameters from latent image formation and IGM heating. J. Phys. A 40, 2935-2958 (2007).

[56] Ghirardi, G.C., Grassi, R. Rimini, A. Continuous spontaneous-reduction model involving gravity. Phys. Rev. A 42, 1057 (1990).

[57] Diósi, L. Intrinsic time-uncertainties and decoherence: comparison of 4 models. Braz. J. Phys. 35, 260 (2005).

[58] Jackson, J. D. Classical Electrodynamics. (John Wiley & Sons, New York, 1962).

[59] IUPAC, Analytical Compendium - X-Ray Spectroscopy. International Union of Pure and Applied Chemistry (2003), Chapter 10.3.4.8.

[60] Townes, C. H. and Schawlow, A. L. Microwave Spectroscopy. McGraw-Hill, New York, (1955), Chapter 12.

[61] Herzberg, G. Molecular Spectra and Molecular Structure. Electronic Spectra and Electronic Structure of Polyatomic Molecules. Krieger: Malabar, FL (1991) Vol. II, Sec.II.5(d).

[62] This form of $\hat{L}(x)$ is different from Diósi’s original proposal [2]. To avoid the divergence due to the Newton self-energy, Diósi initially proposed a Lindblad operator with a length cutoff equal to the nuclear size ($10^{-15}$ – $10^{-14}$ m). Then, it was shown that with this small cutoff, DP model contradicts some known observations [56]. To avoid this problem, Ghirardi, Grassi and Rimini [56] proposed this new form of Lindblad operator whose cutoff is $r_C \simeq 10^{-7}$ m. This adjustment of the model was eventually acknowledged by Diósi [57].