Probing Quantum Dynamical Couple Correlations with Time-Domain Interferometry

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Time domain interferometry is a promising method to characterize spatial and temporal correlations at x-ray energies, via the so-called intermediate scattering function and the related dynamical couple correlations. However, so far, it has only been analyzed for classical target systems. Here, we provide a quantum analysis, and suggest a scheme which allows to access quantum dynamical correlations. We further show how TDI can be used to exclude classical models for the target dynamics, and illustrate our results with the archetype system of a single particle in a double well potential.

Introduction — Spatial and temporal correlations among particles are key to the exploration of complex many-body phenomena. Scattering experiments provide access to the scattering function \( S(p, \omega) \) that is proportional to the cross section for scattering with energy transfer \( \hbar \omega \) and momentum transfer \( \hbar \mathbf{p} \). It characterizes the evolution of correlations on time scales \( \sim 1/\omega \) and length scales \( \sim 1/|\mathbf{p}| \). In practice, knowledge of the correlations over a broad range of time and momentum transfer scales is desirable, and various scattering techniques such as x-ray \([2, 3]\) and neutron \([4]\) scattering can be used to access complementary energy and momentum transfer scales. Similarly, depending on the properties of the scatterer, it can be favorable to characterize correlations directly in the time domain, via the intermediate scattering function (ISF)

\[
S(p, t_1, t_2) = \int_V G(r, t_1, t_2) e^{i \mathbf{p} \cdot \mathbf{r}} d^3r, \tag{1}
\]

with the dynamical couple-correlation function (DCF)

\[
G(r, t_1, t_2) = \int_V \text{Tr} \left[ \hat{\rho}(r', t_1) \hat{\rho}(r' + \mathbf{r}, t_2) \right] d^3r'. \tag{2}
\]

Here, the system described by the density matrix \( \rho \) covers the volume \( V \), and \( \hat{\rho}(r, t) \) is the particle-density operator. The DCF quantifies the spatial and temporal correlations between particles at \( (r, t_1) \) and \( (r' + \mathbf{r}, t_2) \).

A particular technique to access the ISF is the so-called time-domain interferometry (TDI) \([5, 11]\) (see Fig. 1 for the extended scheme used here). It has recently been suggested as a promising candidate for x-ray free electron laser experiments \([2]\), since it allows to measure ISF over much longer times than competing techniques, and since it is essentially background-free even for intense x-ray pulses. TDI uses filter foils containing long-lived Mössbauer isotopes, which are placed in front of and behind the actual target. The incident x-ray frequency is chosen in resonance with the Mössbauer nuclear transition. The first foil (“split unit”) then splits the incoming pulse into two parts. The first part comprises photons which did not interact with the nuclei. The second part is delayed, due to the interaction with the long-lived nuclear transition. As a consequence, the two parts probe the target at different times \( t_1, t_2 \). After the interaction, the second Mössbauer foil (“overlap unit”) again splits each of the two parts. This “overlap operation” creates contributions to the detected signal, which were either delayed in the split unit or in the overlap unit, but not in both, and thus reach the detector at the same time. For these, it is not possible to distinguish if the interaction with the target took place at time \( t_1 \) or \( t_2 \), and the interference of these two pathways leads to temporal modulations of the detection signal, which in turn provide access to the ISF. Depending on the chosen Mössbauer species, different momentum and energy transfer ranges can be accessed \([11]\). Recently, also a modified scheme using Mössbauer foils with two resonances has been suggested \([12]\).

So far, TDI has been analyzed and demonstrated experimentally \([5, 11, 14]\) for targets which can be described by classical mechanics \([15]\). With the growth of interest toward strongly correlated materials \([16, 17]\) and the possibility of simulating their features in lab \([18, 19]\), the development of experimental techniques for measuring quantum dynamical correlations is attracting more and more interest. This raises the question, whether time domain techniques can also be used to explore correlations in targets which require a quantum mechanical treatment.

Here, we provide a quantum mechanical analysis of TDI, and suggest a scheme which allows one to measure the intermediate scattering function both, for quantum and classical targets. DCF and ISF have different properties for classical and quantum targets, and we show how TDI can be used to exclude classical models for the targets. Finally, we illustrate our main results for a minimal archetype system composed of a single particle hopping between two sites.

Properties of DCF and ISF — We start with symmetry properties of DCF and ISF, which will enable us to distinguish quantum mechanical targets from classical ones. As already noted by van Hove himself in \([1, 20]\), for quantum systems (subscript \( qu \), the DCF is in general a complex-valued function due to the non-commutativity of particle-density operators at different times. It directly
FIG. 1. (Color online) Schematic setup. The incoming wave packet propagating along $k$ (red) is separated into two identical copies with a mutual delay by a “split unit”. The advanced (violet) component is quasi-elastically scattered by the target at time $t_1$, while the delayed one (blue) scatters at $t_2$. Subsequently, the light scattered in direction $k + p$ passes an “overlap unit”, which acts identical to the split unit. The central component of the outgoing signal contains two indistinguishable copies with a mutual delay by a “split unit”, which acts identical to the split unit. The central component of the outgoing signal contains two indistinguishable photons, which were either delayed in the split unit or in the overlap unit, but not in both. It is therefore not possible to distinguish if the interaction with the target took place at time $t_1$ or $t_2$. In the following, we will concentrate on this part. Note that the corresponding quantum analysis of the original setup with Mössbauer foils is given in the Supplemental Material [21].

We proceed by calculating the probability amplitude that a photon from the central pulse is registered by a detector placed at position $R$ at time $t$, by summing up the detection amplitudes for the two indistinguishable scattering pathways. These evaluate to ($j \in \{1, 2\}$, see Supplemental Material [21] and the references therein [22–24] for details)

$$\frac{\epsilon^{i\omega_D(R/c-t)}}{R} e^{i\phi_j} f(t) \int_V \! d^3 r e^{-ip \cdot r} \hat{\rho}(r, t_j) |\psi\rangle,$$

As expected, the amplitudes are spherical wave packets with carrier frequency and envelope $f(t)$ identical to those of the incoming photon. The amplitudes depend on the target’s density operator at the scattering times and on the initial state of the target $|\psi\rangle$. Here, $p$ is the exchanged momentum between the photon and the target. The signal recorded by the detector will be proportional to the probability of detecting the photon, which in turn is

$$P(p, t) \propto f(t)^2 \left( \sum_{j=1,2} S_{qu}(p, t_j, t_j) ight. \\
+ 2 \cos[\phi] S^R_{qu}(p, t_1, t_2) - \sin[\phi] S^I_{qu}(p, t_1, t_2) \bigg),$$

where $\phi = \phi_2 - \phi_1$ is the phase difference between the two scattering pathways. Here and in the following, a superscript $R$ [$I$] denotes the real [imaginary] part, such
that $S_{qu} = S_{qu}^R + i S_{qu}^f$. Note that Eq. (7) applies to targets initially in a pure quantum state. Otherwise, it has to be averaged over all possible initial states.

As our first main result, we find that from Eq. (7) that control over the relative phase $\phi$ and the delay $\Delta t$ enables one to individually access the real and the imaginary parts of the ISF as function of momentum transfer $p$ and time $t$, as desired.

Next, in order to extract information about the quantum or classical nature of the target, we consider the sum $I^+$ and the difference $I^-$ of the intensities at two opposite exchanged momenta $\pm p$. From Eq. (7), we find

$$I^+_{qu}(\phi, t) \propto f(t)^2 \left( \sum_{j=1,2} [S_{qu}(p, t_j, t_j) \pm S_{qu}(-p, t_j, t_j)] \right.$$

$$+ 2 \left\{ \cos[\phi] \left[ S_{qu}^R(p, t_1, t_2) \pm S_{qu}^R(-p, t_1, t_2) \right] +

- \sin[\phi] \left[ S_{qu}^f(p, t_1, t_2) \pm S_{qu}^f(-p, t_1, t_2) \right] \right\} \right),$$

(8)

If a classical model for the target is assumed, such that the ISF satisfies the symmetry Eq. (5), then Eq. (8) simplifies to

$$I^+_{cl}(\phi, t) \propto f(t)^2 \left( \sum_{j=1,2} S(p, t_j, t_j) +

+ 2 \cos[\phi] S^R(p, t_1, t_2) \right),$$

(9)

$$I^-_{cl}(\phi, t) \propto -2 f(t)^2 \sin[\phi] S^f(p, t_1, t_2).$$

(10)

We find that recording $I_{\pm}$ for different values of $\phi$ enables one to distinguish quantum or classical symmetries of the target. If, for example, $I_{-}$ does not vanish at $\phi = n\pi$, then the classical relation Eq. (10) is ruled out. It follows that the ISF of the target has no inversion symmetry, such that the DCF is a complex valued function and a quantum model for the target is needed. In the opposite case, DCF is real valued. Then, it may still be possible to violate Eq. (7) to exclude a classical model. However, it is important to note that a real DCF alone does not imply a classical target. Rather, also quantum targets may exhibit real valued DCF for particular parameter choices. This fact is explicitly shown for a concrete system in the next section.

**Archetype model** — In the final part, we illustrate our results with the concrete toy model of a single particle in a double well potential, which is of relevance since it serves as elementary building block for a number of related phenomena. For example, cold atoms trapped in atomic lattices serve as quantum simulators for complex solid state phenomena, structured potentials appear on surfaces of materials, and certain complex materials may intrinsically offer various quantum states placed in a potential landscape. Few-particle systems in single- or double-well potentials have also been directly studied [25].

Furthermore, the model is simple enough to allow for an exact calculation of DCF and ISF, yet it already suffices to illustrate the main results of this work.

We denote the two wells by $L$ and $R$, and the particle dynamics is governed by the Hamiltonian

$$H = -\hbar \frac{\Omega}{2} (|L\rangle\langle R| + |R\rangle\langle L|).$$

(11)

A generic state of the particle at time $t$ in the $|L\rangle, |R\rangle$ representation is given by the density matrix

$$\rho(t) = \begin{pmatrix} P_L(t) & \Gamma(t) \\ \Gamma(t)^* & P_R(t) \end{pmatrix},$$

where $P_L(t), P_R(t)$ are the probabilities of finding the particle at time $t$ at position $j$ which satisfy the condition $P_L(t) + P_R(t) = 1$, while $\Gamma(t)$ is the coherence coefficient. The DCF calculated for the state (12) is

$$G_{qu}(d, t_1, t_2) = S^2 P_L(t_1) + \frac{i}{2} S' \Gamma(t_1)^*,$$

(13)

$$G_{qu}(-d, t_1, t_2) = S^2 P_R(t_1) + \frac{i}{2} S' \Gamma(t_1),$$

(14)

$$G_{qu}(0, t_1, t_2) = C - i S' \Gamma^R(t_1),$$

(15)

where $S = \sin[\Omega \Delta t/2], C = \cos[\Omega \Delta t/2], S' = \sin[\Omega \Delta t], \Delta t = t_2 - t_1$ and $\Gamma^R(t_1)$ indicates the real part of $\Gamma(t_1)$. Expressions (13)- (15) are complex valued, if $\Gamma^R(t_1)$ is non-zero. Thus, the presence of imaginary parts is strictly linked to (partially) coherent superpositions of position eigenstates, such that the complex nature of the DCF clearly relates to quantum mechanical properties of the target. Conversely, if $\Gamma(t_1)$ is purely imaginary, then the DCF is real, even though the system is in a quantum mechanical superposition of position eigenstates. This shows that a real DCF does not imply a classical target, while an imaginary contribution to the DCF indicates a quantum target. The ISF corresponding to (13-15) is

$$S_{qu}(p, t_1, t_2) = S^2 \cos[p \cdot d] + C^2 +$$

$$+ i \left\{ [P_L(t_1) - P_R(t_1)] S^2 \sin[p \cdot d] +$$

$$+ \frac{S'}{2} \left[ \Gamma^I(t_1) \sin[p \cdot d] + \Gamma^R(t_1) \left( \cos[p \cdot d] - 2 \right) \right] \right\}$$

(16)

which evidently satisfies the identity (5) only if $\Gamma^R(t_1) = 0$, consistently with the condition found to have a real DCF.

It turns out that $\Gamma^R$ is a constant of motion under the action of Hamiltonian Eq. (11). This allows us to relate the results better to an actual experimental implementation, in which it may only be possible to control the
delay $\Delta t$, but not $t_1$ itself. Averaging over $t_1$, we find

$$
\bar{G}_{\text{step}}(\pm d, \Delta t) = \frac{1}{2} S + \frac{i}{2} S' \Gamma^R,
$$

$$
\bar{G}_{\text{step}}(0, \Delta t) = C - i S' \Gamma^R,
$$

$$
\bar{S}_{\text{step}}(p, \Delta t) = S^2 \cos|p|d + C^2 + \frac{i}{2}(\cos|p|d - 2)S' \Gamma^R.
$$

As before, the complex nature of the DCF is linked to $\Gamma^R$. From Eq. (10), we further find $\bar{I}^- = 0$, such that a classical model cannot be excluded. But $\bar{I}^+$ has a contribution proportional to $\Gamma^R \sin \phi$, which is at odds with Eq. (9) if $\Gamma^R \neq 0$, such that then a classical model can be excluded.

Summary and discussion — DCF and ISF have different properties for quantum and classical systems. The non-commutativity of particle-density operators at different times in general leads to imaginary contributions to the DCF for quantum systems, and DCF and ISF have different symmetry properties under complex conjugation for classical and quantum systems. Using the quantum mechanical analysis presented here, we have shown that time-domain techniques can be used to measure the complex-valued ISF. Moreover, the comparison of the ISF at two opposite values of the exchanged momentum $p$ in the form Eq. (8) provides access to the symmetry properties of the system’s ISF, and gives a handle to exclude classical models for the target.

Throughout the analysis, we used a simplified model for the split and overlap units, but our results carry over to the case of Mössbauer filter foils, as shown in the supplemental materials. For Mössbauer foils, it has recently been demonstrated that it is possible to control the relative phase $\phi$ with sub-Ångstrom precision on a nanosecond time scale using piezo elements [24], as required in the TDI scheme suggested here.

We illustrated our results for the archetype double well system, and found that it is sufficient to control the delay time $\Delta t$, while the absolute times $t_1$ and $t_2$ can be averaged over. However, we did not assume a stationary state of the target system, and it would be interesting to study extended setups in which the incident x-ray pulses are synchronized to the preparation of the target in a specific quantum state, e.g., using laser fields.

Our TDI scheme is not restricted to the x-ray domain, but could also be used to explore correlations on other time and length scales. This requires the availability of suitable split- and overlap units, and a system whose internal dynamics has no resonance in the spectrum of the probing photon pulse, so that only quasi-elastic scattering of the photon is relevant.

Finally, we note that the appearance of imaginary parts in such quantities poses practical and interpretative problems [27, 29], which could be explored experimentally using TDI techniques.

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Supplemental Material to “Probing Quantum Dynamical Couple Correlations with Time-Domain Interferometry”

Derivation of the single photon detection amplitudes

In order to derive the explicit form for the detection amplitude, we need to solve the problem of the scattering of the photon by the target system after it passed through the split stage. Next, we have to evaluate how the initial state of the system composed of target and photon evolves for long times under the influence of the matter-radiation interaction. At the initial time after the split unit, the photon is in a state given by the superposition of two spatially separated incoming wave packets, \(| \gamma_1 \rangle + | \gamma_2 \rangle\), whereas the target system is in the state \(| \psi \rangle\), so that the initial state of the global system is the product of the two

\[ | \Psi \rangle = | \gamma_1 \rangle | \psi \rangle + | \gamma_2 \rangle | \psi \rangle . \]  (20)

Let us fix a reference frame with the \(x\)-axis parallel to the initial direction of propagation of the incoming photon, and such that the edge of the target facing the split unit is parallel to the \(x = 0\) plane. The two wave packets have the form

\[ | \gamma_j \rangle = \int dk c(\omega_k - \omega_0) e^{-ikx_j} | 1_k \rangle \]  (21)

where \(| 1_k \rangle\) are single photon states with momentum parallel to \(x\), \(c(\omega_k - \omega_0)\) is a function centered at \(\omega_0\) with bandwidth \(\Delta \omega \ll \omega_0\), and \(x_j\) is the distance the wave packet must travel to reach the target.

The scattering state

The full dynamics of the global system is generated by \(\hat{H}_0\), which is the sum of the free Hamiltonians for the radiation and the target, plus an interaction term \(\hat{H}_I\), which, assuming that the radiation is non-resonant with the target’s internal structure, can be written as

\[ \hat{H}_I = \lambda \int d^3k' d^3k'' \frac{\hat{a}_k^\dagger \hat{a}^\dagger_{k''}}{\sqrt{\omega_k \omega_{k''}}} \int_V d^3r \rho(r)e^{i(k'' - k') \cdot r} . \]  (22)

Here, \(\lambda\) is a coupling constant depending on the charge and the mass of the scatterers in the target, \(\hat{a}_k\) and its conjugate are the photon destruction and creation operators, \(\hat{\rho}(r)\) is the density of scatterers operator at point \(r\) and \(V\) the volume of the target.

Denoting the time evolution operator associated to \(\hat{H}_0\) as \(\hat{U}_0(t)\), the full time evolution operator can be evaluated perturbatively to give

\[ \hat{U}(t) \approx \hat{U}_0 - \frac{i}{\hbar} \hat{U}_0(t) \int_0^t dt' \hat{U}_0^\dagger(t') \hat{H}_I \hat{U}_0(t') dt' . \]  (23)

The evolved state obtained by applying to \(\hat{U}_0\) as \(\hat{U}_0(t)\), the full time evolution operator can be evaluated perturbatively to give

\[ | \delta \Psi_j \rangle = -\frac{i}{\hbar} \hat{U}_0(t) \int_0^t dt' \hat{U}_0^\dagger(t') H_I \hat{U}_0(t') \, dt' | \gamma_j \rangle | \psi \rangle \]  (24)

Substituting (21) and (22) into this expression leads to

\[ | \delta \Psi_j \rangle = -\frac{i}{\hbar} \hat{U}_0(t) \int_0^t dt' \int_V d^3r \int d^3k' \int d^3k'' \frac{e^{-i(k' \cdot r - \omega_{k'} t')}}{\sqrt{\omega_{k'}}} \frac{e^{i(k'' \cdot r - \omega_{k''} t')}}{\sqrt{\omega_{k''}}} e^{-ikx_j} c(\omega_k - \omega_0) \hat{\rho}(r, t') | \psi \rangle | 1_k \rangle . \]  (25)

Here, the time-integration interval is considered large compared to the other timescales involved in the problem. Then, some of the integrals appearing can be approximated by the Fourier transforms of their respective integrands.
Simplified case

In what follows, we neglect all global constant pre-factors to simplify the notation. The probability of detecting a scattered photon at position \( \mathbf{R} \) at time \( t \) is \([24]\)

\[
\langle \delta \Psi_1 | + \langle \delta \Psi_2 | \hat{E}^{(+)}(\mathbf{R}) \hat{E}^{(-)}(\mathbf{R}) (| \delta \Psi_1 \rangle + | \delta \Psi_2 \rangle) = \\
= (\langle \delta \Psi_1 | + \langle \delta \Psi_2 | \hat{E}^{(+)}(\mathbf{R}) \hat{U}_0(t) \hat{U}_0^\dagger(t) \hat{E}^{(-)}(\mathbf{R}) (| \delta \Psi_1 \rangle + | \delta \Psi_2 \rangle) .
\]

(26)

Here, \( \hat{E}^{(\pm)}(\mathbf{R}) \) is the positive/negative frequency part of the electric field operator, corresponding to the destruction or creation of a photon at position \( \mathbf{R} \). Since \( | \delta \Psi_j \rangle \) are single photon states, the application of \( \hat{E}^{(-)}(\mathbf{R}) \) induces transition to the electromagnetic vacuum state \( |0\rangle \) \([24]\). Thus, we can insert an identity relation in the middle of the scalar product to give

\[
\langle \delta \Psi_1 | + \langle \delta \Psi_2 | \hat{E}^{(+)}(\mathbf{R}) \hat{U}_0(t)|0\rangle \langle 0| \hat{U}_0^\dagger(t) \hat{E}^{(-)}(\mathbf{R}) (| \delta \Psi_1 \rangle + | \delta \Psi_2 \rangle) .
\]

(27)

It follows that the detection amplitude is given by \( \langle 0| \hat{U}_0^\dagger(t) \hat{E}^{(-)}(\mathbf{R}) (| \delta \Psi_1 \rangle + | \delta \Psi_2 \rangle) \) and in this case, it is given by the sum of two terms originating from the two different scattering channels. With the explicit form of the electric field operator,

\[
\hat{E}^{(-)}(\mathbf{R}) = \frac{i}{(2\pi)^{3/2}} \int d^3q \sqrt{\omega_q} a_q e^{i\mathbf{q}\cdot\mathbf{R}} ,
\]

(28)

and using the explicit form of \( | \delta \Psi_j \rangle \), the detection amplitude for the \( j \)-th channel becomes

\[
\langle 0| \hat{U}_0^\dagger(t) \hat{E}^{(-)}(\mathbf{R}) | \delta \Psi_j \rangle \propto \int_0^t dt' \int V d^3r \hat{\rho}(\mathbf{r}, t') | \psi \rangle \int d^3q \sqrt{\omega_q} (\mathbf{R} - \mathbf{r})/\omega_q (t-t') \int dke^{-i[\omega_k t' - k(x+j) + x]} e^{-(\omega_k - \omega_0)}/\sqrt{\omega_k} .
\]

(29)

Because of the properties of \( c \), we can approximate

\[
\int dke^{-i[\omega_k t' - k(x+j)]} e^{-(\omega_k - \omega_0)}/\sqrt{\omega_k} \approx \frac{1}{\sqrt{\omega_0}} e^{-i\omega_0 (t' - x+j)/c} f \left( t' - x+j/c \right) ,
\]

(30)

where \( f \) is the envelope function of the photon wave packet which has a temporal extend \( \sim 1/\Delta \omega \). The finite duration of the wave packet reduces the time-integration interval to the time the pulse needs to cross the target, which is of order \( 1/\Delta \omega + L/c \), with \( L \) the longitudinal size of the target. Assuming that the dynamics of the scatterers in the target has slower timescale than the crossing time, \( \hat{\rho} \) can be considered constant at the instant of arrival of the photon wave packet \( t_j \equiv x+j/c \) and brought out of the time integral. The detection amplitude then becomes

\[
\int V d^3r \hat{\rho}(\mathbf{r}, t_j) | \psi \rangle \int d^3q \sqrt{\omega_q} (\mathbf{R} - \mathbf{r})/\omega_q t \int_0^t dt' f \left( t' - t_j - x/c \right) e^{i\omega_q t' - i\omega_0 (t' - t_j - x/c)}
\]

\[
\times \int V d^3r \hat{\rho}(\mathbf{r}, t_j) | \psi \rangle \int d\omega q d\theta q e^{i\omega_q t} \sin \theta e^{i\omega_q (\mathbf{R} - \mathbf{r})/c} e^{-i\omega_q \sin \theta} e^{-i\omega_q \sin \theta} e^{i\omega_q (t - t_j - x/c)} e^{i\omega_q - \omega_0} .
\]

(31)

Upon integration over \( \theta \) taking into account that the distance of the detection point is much larger than the size of the target, the scattering amplitude becomes proportional to

\[
\frac{e^{i\omega_0 (R/c - t)} - e^{-i\omega_0 (R/c + t)}}{R} e^{i\omega_0 t_j} \int V d^3r e^{-i(k_0 - k_0) \cdot r} f \left( t - t_j - | \mathbf{R} - \mathbf{r} | + x/c \right) \hat{\rho}(\mathbf{r}, t_j) | \psi \rangle ,
\]

(32)

where \( k_0 \equiv \omega_0 / (cR) \) and \( k_0 \equiv x_0 / (cx) \). The last line is a superposition of outgoing and ingoing spherical waves centered at the target, of which the ingoing does not correspond to the boundary conditions of interest here and therefore is dropped. Supposing moreover that the envelope does not vary significantly over the size of the target, i.e., \( L \ll c/\Delta \omega \), and neglecting the propagation time to the detector, one finds the final form of the detection amplitude

\[
\frac{e^{i\omega_0 (R/c - t)}}{R} e^{i\omega_0 t_j} f (t - t_j) \int V d^3r e^{-i(k_0 - k_0) \cdot r} \hat{\rho}(\mathbf{r}, t_j) | \psi \rangle .
\]

(33)

After the interaction with the target, the overlap unit creates a contribution in which the two scattering pathways temporally overlap, by delaying the advanced one.
Realization with Mössbauer foils

In the original proposal [5], the split and overlap units consist of two identical Mössbauer foils. When a pulse impinges on a Mössbauer foil, the component of the pulse resonant with the Mössbauer transition at frequency $\omega_0$ is scattered on a time scale $T$. One of the two foils used in the original arrangement is slightly detuned from the other by moving it at a constant velocity, this causing a Doppler shift $\Omega \gg T^{-1}$ of its transition energy. In the following, we assume that the split unit is subject to the Doppler shift. Thus, the initial state of the photon is a superposition of a state of the kind \([21]\), that is a copy of the temporally short incoming wave packet, and a temporally long wave packet with an approximately Lorentzian spectral shape $\mathcal{L}(\omega_k - \omega_0 - \Omega)$. The Lorentzian spectral shape corresponds to an approximately exponential decay in the time domain, which starts immediately after the excitation, such that $x_1 = x_2 \equiv x_0$. The first component of the initial photon state gives a contribution to the detection amplitude similar to \([33]\). However, the second foil acts on it by scattering its component at frequency $\omega_0$, such that the envelope $f$ becomes an exponentially decaying function

$$f(\xi) \rightarrow h(\xi) \equiv \Theta(\xi)e^{-\frac{\xi}{\tau}}$$

with $\Theta$ the Heaviside step function.

The scattering of the Lorentzian part of the initial photon state needs a different treatment because the characteristic time $T$ of its exponentially decaying envelope is comparable with timescale of the internal dynamics of the target. This fact does not allow us to assume that the scatterer’s density operator is constant during the crossing time of the wave packet. In order to take into account the time dependency of $\hat{\rho}$ in formula \([29]\) we decompose it into the energy eigenvectors of the target,

$$\hat{\rho}(r, t') = \sum_{m,n} e^{i\omega_{mn}t'} \langle m | \hat{\rho}(r) | n \rangle | n \rangle$$

where $\omega_{mn}$ are the characteristic frequencies of the target’s internal dynamics. Since we assume that they are non-resonant with the radiation, they are not within the support of the Lorentzian wave packet. As a consequence, the detection amplitude due to the second component of the initial photon state is

$$\sum_{m,n} \int_V d^3r \langle m | \hat{\rho}(r) | n \rangle \langle n | \psi \rangle | m \rangle \int d^3q e^{i\{q(r-R)+\omega_k t\}} \int_0^t dt' e^{i(\omega_0+\omega_{mn}) t'} \int dk e^{-i|\omega_k t' - k(x+x_0)|} \frac{\mathcal{L}(\omega_k - \omega_0 - \Omega)}{\sqrt{\omega_k}}$$

The explicit calculation of the integrals over time and momenta transform the last expression into

$$\frac{1}{R} \int_V d^3r \hat{\rho}(r, t - \frac{|R+r|}{c}) | \psi \rangle h \left( t - \frac{|R-r| + x + x_0}{c} \right) e^{i(\omega_0 + \Omega)(t - \frac{|R-r| + x + x_0}{c})}$$

Assuming that $T \gg L/c$, defining the wave vectors $\mathbf{k}_0 \equiv (\omega_0 + \Omega)\mathbf{R}/(cR) \simeq \omega_0 \mathbf{R}/(cR)$ and $\mathbf{k}_0 \equiv (\omega_0 + \Omega)\mathbf{x}/(cx) \simeq \omega_0 \mathbf{x}/(cx)$ the detection amplitude in the second channel is

$$\frac{e^{i\omega_0(R/c-t)}}{R} e^{i(\omega_0(t - \frac{x_0}{c}) h)} \left( t - \frac{R + x_0}{c} \right) \int_V d^3r \hat{\rho}(r, t - \frac{R}{c}) | \psi \rangle e^{-i(\mathbf{k}_0 - \mathbf{k}) \cdot r}$$

It can be assumed that the second foil has no effect on the scattered photon because, due to the Doppler shift, the spectrum of the latter is far from the resonance of the former. In addition the factor $\Omega x_0/c$ usually is very small giving no relevant phase contributions. The total amplitude for detection after the overlap unit is then

$$\frac{e^{i\omega_0(R/c-t)}}{R} e^{i\omega_0 \frac{x_0}{c} h} \left( t - \frac{R + x_0}{c} \right) \left( \int_V d^3r \hat{\rho}(r, t_0) | \psi \rangle e^{-i(\mathbf{k}_0 - \mathbf{k}) \cdot r} + e^{i\Omega t} \int_V d^3r \hat{\rho}(r, t - \frac{R}{c}) | \psi \rangle e^{-i(\mathbf{k}_0 - \mathbf{k}) \cdot r} \right)$$

which is the quantum correspondent of the result found in \([5]\). Notice that the phase difference in this case is given by the Doppler-shift factor $\Omega t$.

**Calculation of DCF for one particle in a Double Well potential**

When the particles in the target system can only occupy discrete positions, the integral defining the DCF at the separation $r$ \([Eq. (2) in the main text]\) reduces to a discrete sum over all the possible pairs of points with mutual
distance \( r \),

\[
G_{\text{qu}}(r, t_1, t_2) = \sum_{r_n} \text{Tr}[\mu(\hat{\rho}_n(t_1)\hat{\rho}_{n+r}(t_2))] = \sum_{r_n} \text{Tr}[\mu(t_1)\hat{\rho}_n\hat{\rho}_{n+r}(t_2 - t_1)].
\] (40)

The ISF then is given by a discrete Fourier transform

\[
S_{\text{qu}}(p, t_1, t_2) = \sum_r G_{\text{qu}}(r, t_1, t_2)e^{ip\cdot r}.
\] (41)

In our case, the particle can only occupy the two minima of the double-well potential labeled by \( L, R \) whose distance is \( d \). As a consequence, the DCF has only three values in correspondence of the three distances \( r = 0, \pm d \), given by

\[
G_{\text{qu}}(d, t_1, t_2) = \text{Tr}[\mu(t_1)\hat{\rho}_L\hat{U}^\dagger(t_2 - t_1)\hat{\rho}_R\hat{U}(t_2 - t_1)] ,
\] (42)

\[
G_{\text{qu}}(-d, t_1, t_2) = \text{Tr}[\mu(t_1)\hat{\rho}_R\hat{U}^\dagger(t_2 - t_1)\hat{\rho}_L\hat{U}(t_2 - t_1)] ,
\] (43)

\[
G_{\text{qu}}(0, t_1, t_2) = \text{Tr}[\mu(t_1)\hat{\rho}_L\hat{U}^\dagger(t_2 - t_1)\hat{\rho}_L\hat{U}(t_2 - t_1)] + \text{Tr}[\mu(t_1)\hat{\rho}_R\hat{U}^\dagger(t_2 - t_1)\hat{\rho}_R\hat{U}(t_2 - t_1)].
\] (44)

As only one particle is considered, the explicit form of the density operators at the two position \( L, R \) are simply

\[
\hat{\rho}_L = |L\rangle\langle L| ,
\] (45)

\[
\hat{\rho}_R = |R\rangle\langle R| .
\] (46)

The Hamiltonian for the single particle is

\[
\hat{H} = -\hbar \frac{\Omega}{2} (|L\rangle\langle R| + |R\rangle\langle L|) ,
\] (47)

and the time evolution operator can be calculated exactly to give

\[
\hat{U}(t) = \cos \left( \frac{\Omega}{2} t \right) + i(|L\rangle\langle R| + |R\rangle\langle L|) \sin \left( \frac{\Omega}{2} t \right).
\] (48)

The expression of the evolved density matrix is kept implicit in the calculation as we wanted to point out the role of coherences at the time at which the first variable is considered, but its explicit time dependency follows in a straightforward way from (48).