Quantum Interference Enforced by Time-Energy Complementarity

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The interplay of the concepts of complementarity and interference in the time-energy domain are studied. In particular, we theoretically investigate the fluorescence light from a $J = 1/2$ to $J = 1/2$ transition that is driven by a monochromatic laser field. We find that the spectrum of resonance fluorescence exhibits a signature of vacuum-mediated interference effects, whereas the total intensity is not affected by interference. We demonstrate that this result is a consequence of the principle of complementarity, applied to time and energy. Since the considered level scheme can be found, e.g., in $^{198}Hg^{+}$ ions, our model system turns out to be an ideal candidate to provide evidence for as yet experimentally unconfirmed vacuum-induced atomic coherences.

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Complementarity is a key concept of quantum mechanics which has no classical analogue. According to Niels Bohr [1], complementarity arises from the inseparability of detector and object. This leads to mutually exclusive observables that cannot be measured using a single experimental setup. A famous example is the wave-particle duality in a two-slit experiment. One can decide to observe either the interference pattern exposing wavelike features, or particle properties by measuring the path taken. The interference pattern is observed under conditions where it is principally impossible to know through which of the two slits each object has moved. On augmenting the experiment by any means which, in principle, allow us to measure the path taken, the interference pattern vanishes. In famous thought experiments [1,2] this wave-particle duality is attributed to the position-momentum uncertainty relation. This has led to an inspiring discussion [3,4] about the interrelation between complementarity and this uncertainty relation. A recent experiment demonstrates that these two are not equivalent [5].

Up to now, the focus has been on spatially separated pathways resulting in an interference pattern in position space. Here, we demonstrate that quantum optical experiments can reveal complementarity of time and energy. In this class of setups, different temporal paths lead to interference in the energy domain. An attempt to extend interference and complementarity to the time-energy domain raises several questions. First, it is not obvious what the equivalence is of (spatial) pathway interference in the time-energy domain. How can paths differing "in time" be realized? Two approaches are given in a recent double-slit experiment in the time-energy domain where the slits are related to different time windows of attosecond duration [6], and in the intensity correlations of different spectral components in a two-level atom [7]. Second, what makes these paths indistinguishable in principle, as required for interference? Finally, what is the role of the time-energy uncertainty relation, which is special in that time is a parameter rather than an operator in quantum mechanics?

In the following, we discuss these questions on the basis of the resonance fluorescence of a single laser-driven atom with a $J = 1/2$ to $J = 1/2$ transition as found, e.g., in $^{198}Hg^{+}$ ions. We show that complementarity enforces a signature of interference in the spectrum of resonance fluorescence, whereas the total fluorescence intensity exhibits no interference. The results can be described quantitatively via the time-energy uncertainty relation. Further, we demonstrate that the interference in the fluorescence spectrum results from vacuum-induced coherences, which have been intensively studied theoretically, but are experimentally unconfirmed. Thus we provide a realistic experimental setup to verify the presence of these effects.

The schematic setup of the discussed experiment is shown in Fig. 1(a). An atomic system is located at the point of origin and irradiated by a monochromatic laser beam polarized along the z axis and propagating in x direction. We assume that the incident driving field of frequency ω_L is nearly resonant with a $J = 1/2$ to $J = 1/2$ transition of the atom; the corresponding level structure is shown in Fig. 1(b). The external light field couples

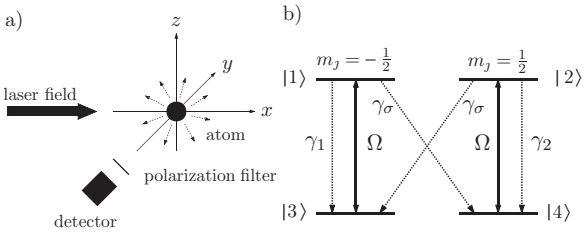

FIG. 1. (a) Considered experimental setup showing the atom interacting with the laser field. Either the total intensity or the fluorescence spectrum emitted on the π transitions is observed. (b) Level scheme of the $J = 1/2$ to $J = 1/2$ transition. The coupling between the laser field and the π transitions is characterized by the Rabi frequency Ω. γ_1, γ_2, and γ_σ are spontaneous decay rates indicated by dotted lines.

only to the transitions $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |4\rangle$ that will be designated as the π transitions, and the transitions $|1\rangle \rightarrow |4\rangle$ and $|2\rangle \rightarrow |3\rangle$ will be denoted as the σ transitions. Throughout this Letter we are only concerned with the fluorescence light emitted on the π transitions that can be selected with a suitable polarization filter in front of the detector. The time evolution of the driven four-level atom is modeled by a master equation, $\dot{\varrho} = -i[H, \varrho]/\hbar + \mathcal{L}_\gamma \varrho$. In the rotating wave approximation and in a frame rotating with the laser frequency ω_L, the Hamiltonian is given by

$$H = -\hbar\Delta(A_{11} + A_{22}) + \hbar[(A_{13} - A_{24})\Omega + \text{H.c.}], \quad (1)$$

where $\Delta = \omega_L - \omega_0$ is the detuning of the driving field from resonance, the $A_{ij} = |i\rangle\langle j|$ denote the atomic transition operators, and the Rabi frequency is labeled by Ω. The damping term $\mathcal{L}_\gamma \varrho$ takes the form [8]

$$\mathcal{L}_\gamma \varrho = -\frac{1}{2}\sum_{i,j=1}^{2} \gamma_{ij}\{[S_i^+, S_j^- \varrho] + [\varrho S_i^+, S_j^-]\} - \frac{\gamma_\sigma}{2}\sum_{i=3}^{4}\{[S_i^+, S_i^- \varrho] + [\varrho S_i^+, S_i^-]\}, \quad (2)$$

where the transition operators $S_i^\pm$ are defined as $S_1^+ = A_{13}$, $S_2^+ = A_{24}$, $S_3^+ = A_{23}$, $S_4^+ = A_{14}$, and $S_i^- = (S_i^+)^\dagger$. γ_σ is the decay constant on each of the σ transitions, the parameters γ_{ij} are defined as $\gamma_{ij} = (\boldsymbol{d}_i \cdot \boldsymbol{d}_j^*)/(|\boldsymbol{d}_i||\boldsymbol{d}_j|)\sqrt{\gamma_i \gamma_j}$, and γ_1 and γ_2 are the decay constants of the π transitions [see Fig. 1(b)]. Although γ_1 and γ_2 are equal in our setup, we will continue to label them differently to facilitate the physical interpretation later on. Both matrix elements $\boldsymbol{d}_1 = \langle 1|\hat{\boldsymbol{d}}|3\rangle$ and $\boldsymbol{d}_2 = \langle 2|\hat{\boldsymbol{d}}|4\rangle$ of the electric-dipole moment operator $\hat{\boldsymbol{d}}$ correspond to π transitions ($\Delta m_j = 0$) and are thus proportional to each other. More precisely, it follows from the Wigner-Eckart theorem [9] that $\boldsymbol{d}_1$ and $\boldsymbol{d}_2$ are antiparallel, and hence $\gamma_{12} = \gamma_{21} = -\sqrt{\gamma_1 \gamma_2}$.

Here we discuss two distinct photodetection signals, namely, the total intensity emitted by the atom and the spectrum of resonance fluorescence. Extensive theoretical studies [10] show that the so-called V system, an atomic level scheme consisting of two near-degenerate excited levels and one ground state, exhibits a rich variety of interference effects, including the modification of spontaneous emission, provided that the two dipole moments are nonorthogonal. However, so far there is no convincing experimental evidence for these effects in atomic systems due to the lack of suitable candidates. Although the driven four-level system in Fig. 1(b) is a realistic level scheme with antiparallel dipole moments, it is not self-evident that it also displays interference effects in spontaneous emission. Quantum interference does only occur if various indistinguishable transition amplitudes connect a common initial state to a common final state. In contrast to the V system, the two π transitions end up in different ground states that are orthogonal to each other, and hence our four-level system is not expected to display any interference effects at all. Surprisingly, we find that the spectrum of resonance fluorescence does show a signature of interference, whereas the total intensity exhibits no indication of interference.

First, we illustrate that the total intensity, being determined by the normally ordered first-order correlation function of the electric field, is not affected by interference. In the far-field zone, the source-field contributions to the positive and negative frequency parts $\hat{\boldsymbol{E}}^{(\pm)}(\boldsymbol{r}, t)$ of the electric field operator can be expressed in terms of the atomic transition operators [11]. The total intensity emitted on the π transitions in the direction of the detector is $I_{\text{st}}^\pi = \phi_\pi \sum_{i,j=1}^{2} \gamma_{ij}\langle S_i^+ S_j^-\rangle_{\text{st}}$, where ϕ_π is a geometrical factor that we set equal to one in the following and $\langle \ldots \rangle_{\text{st}}$ denotes the steady-state mean value [8]. The terms proportional to $\gamma_{12} = \gamma_{21}$ describe the cross damping between the two π transitions that arises as a consequence of quantum interference. However, these interference terms do not contribute to the total intensity since the ground states are orthogonal, $\langle S_1^+ S_2^-\rangle_{\text{st}} = \langle |1\rangle\langle 3|4\rangle\langle 2|\rangle_{\text{st}} = 0$.

We now turn to the spectrum of resonance fluorescence that is determined by the Fourier transform of the two-time correlation function $\langle [\boldsymbol{e}_z \cdot \hat{\boldsymbol{E}}^{(-)}(\boldsymbol{r}, t + \tau)][\boldsymbol{e}_z \cdot \hat{\boldsymbol{E}}^{(+)}(\boldsymbol{r}, t)]\rangle_{\text{st}}$. If a filter with setting frequency ω and bandwidth λ is placed in front of a broadband detector, the fluorescence spectrum is found to be [8,12]

$$S^\pi(\tilde{\omega}, \lambda) = \frac{1}{\pi}\sum_{i,j=1}^{2} \gamma_{ij}\,\text{Re}\int_0^\infty e^{-i\tilde{\omega}\tau} e^{-\lambda\tau} \times \langle \tilde{S}_i^+(t + \tau)\tilde{S}_j^-(t)\rangle_{\text{st}} d\tau, \quad (3)$$

where $\tilde{\omega} = \omega - \omega_L$ is the difference between the observed frequency and the laser frequency and $\tilde{S}_i^\pm = \exp(\mp i\omega_L t) S_i^\pm$. The two-time correlation functions in Eq. (3) can be evaluated via the quantum regression theorem [8]. In order to prove that the fluorescence spectrum from the π transitions does show a signature of interference, we point out that the terms proportional to γ_{12} are now determined by the *two-time averages* $\langle \tilde{S}_1^+(t + \tau)\tilde{S}_2^-(t)\rangle_{\text{st}}$ rather than by the one-time averages. Indeed, we find that the correlation function

$$G_{12}(\tau) = -\sqrt{\gamma_1 \gamma_2}\langle \tilde{S}_1^+(t + \tau)\tilde{S}_2^-(t)\rangle_{\text{st}} \quad (4)$$

is different from zero for $\tau > 0$; a plot of G_{12} is shown in Fig. 2(a). To illustrate this result we decompose the transition operators $\tilde{S}_i^\pm$ in mean values and fluctuations according to $\tilde{S}_i^\pm = \langle \tilde{S}_i^\pm\rangle_{\text{st}} \hat{1} + \delta\tilde{S}_i^\pm$. The mean values $\langle \tilde{S}_1^+\rangle_{\text{st}} = \varrho_{31}$ and $\langle \tilde{S}_2^+\rangle_{\text{st}} = \varrho_{42}$ are equal to the matrix elements of the steady-state density operator that represent the coherences between the states $|1\rangle$, $|3\rangle$ and $|2\rangle$, $|4\rangle$, respectively. These matrix elements are both different from zero since the driving field couples to both π transitions. Since the two-time average of the fluctuations $\langle \delta\tilde{S}_1^+(t + \tau)\delta\tilde{S}_2^-(t)\rangle_{\text{st}}$ decays exponentially with a time constant on the order of

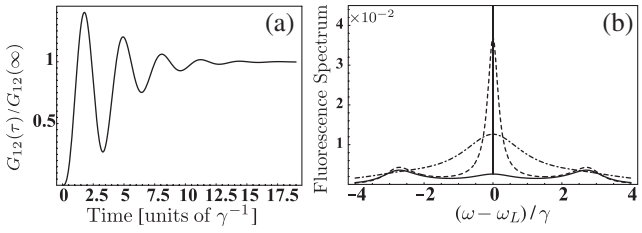

FIG. 2. (a) Plot of the normalized correlation function $G_{12}(\tau)/G_{12}(\infty)$. The parameters are $\Omega = 10^7\ \mathrm{s}^{-1}$, $\Delta = 4 \times 10^6\ \mathrm{s}^{-1}$, and $\gamma = 10^7\ \mathrm{s}^{-1}$. (b) The solid line shows the fluorescence spectrum for perfect detector resolution $\lambda = 0$. The dashed line is the spectrum without the interference terms proportional γ_{12}, γ_{21} in Eq. (3). The Rayleigh peak (the vertical line at $\omega = \omega_L$) is present both with and without interference terms. Note that its weight is larger if the interference terms are taken into account. However, the sums of the integrated coherent and incoherent spectra with and without the interference terms are identical, making the total intensity independent of the interference terms. The parameters are $\Omega = 10^7\ \mathrm{s}^{-1}$, $\Delta = 2 \times 10^7\ \mathrm{s}^{-1}$, and $\gamma = \gamma_1 + \gamma_\sigma = \gamma_2 + \gamma_\sigma = 10^7\ \mathrm{s}^{-1}$. If observed with a finite frequency resolution $\lambda = \gamma$, the spectra with and without interference terms are virtually identical and represented by the dot-dashed line.

γ^{-1}, the long-time limit of G_{12} is equal to $G_{12}(\infty) = -\sqrt{\gamma_1\gamma_2}\langle\tilde{S}_1^+\rangle_{\mathrm{st}}\langle\tilde{S}_2^-\rangle_{\mathrm{st}}$.

Next we investigate how the interference terms alter the fluorescence spectrum and consider the case of an ideal detector with perfect frequency resolution, i.e., $\lambda = 0$. The solid line in Fig. 2(b) shows $S^\pi(\tilde{\omega}, \lambda = 0)$ as it should be observable in an experiment. By contrast, the dashed line has been generated by omitting the interference terms in Eq. (3). It is distinguished by a narrow peak centered at the laser frequency that occurs in addition to the elastic Rayleigh peak. It follows that an experimental observation of the fluorescence spectrum confirming the solid line would give evidence for vacuum-mediated interference effects as described by terms proportional to γ_{12}. So far, interference effects of this kind have not been observed in atomic systems.

In the following we will show that the preceding results are a consequence of the principle of complementarity. If the observer decides to measure the total intensity, the detector must not distinguish between different photon energies. Consequently, complementarity does not impose any restrictions on the time resolution of such a measurement, and hence it is in principle possible to detect the photons in a time resolved way. Thus the experimental conditions allow, at least in principle, to determine the atomic ground state immediately after the detection of a π photon. This implies that the π photons cannot interfere, since one could decide on which of the two π transitions the photon was emitted and hence reveal the quantum path taken by the system.

A totally different situation arises if the detector records the spectrum of resonance fluorescence. An ideal detector is only sensitive to a single frequency, which is swept across the relevant frequency range to measure the whole spectrum. The crucial difference that distinguishes the measurement of the total intensity from the recording of the fluorescence spectrum is that the observer decides in the latter case to measure the photon energies precisely. Since time and energy are complementary observables, no information on the time sequence of the emission can be obtained simultaneously. A more quantitative analysis can be obtained by employing the time-energy uncertainty. If one determines the photon frequencies (energies) with a precision of $\Delta\omega$, the time-energy uncertainty relation enforces that the time of observation has to be at least on the order of $1/\Delta\omega$. This implies that the photon emission times are indeterminate within a time interval of $\Delta t = 1/\Delta\omega$, since the observer can only decide whether a photon has been detected or not after the observation time has elapsed. For the moment we envisage an ideal measurement of the fluorescence spectrum that is characterized by perfect frequency resolution. It follows that the photon emission times are indeterminate. In contrast to the measurement of the total intensity, it is now impossible to decide on which transition the π photon has been emitted. A similar argument explains why interference was observed in a recent attosecond time-energy double-slit experiment [6], where the spectral resolution of the detector makes the ''time slits'' indistinguishable. Next, we identify the interfering pathways. In general, the atom will emit many photons during the time of observation. Since it is impossible to determine the photon emission times, the time order in which these photons have been emitted is unknown. Thus the transition amplitudes that correspond

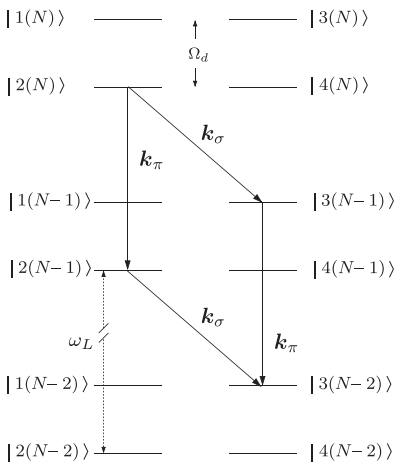

FIG. 3. The dressed states of the system. N is the number of laser photons. The splitting $\Omega_d = \sqrt{4|\Omega|^2 + \Delta^2}$ is not to scale. Two possible cascades are shown that involve the emission of a π photon and a σ photon with wave vector $\boldsymbol{k}_\pi$ and $\boldsymbol{k}_\sigma$, respectively. Depending on the time order of their emission, the π photon is either emitted on transition $|2(N)\rangle \rightarrow |2(N-1)\rangle$ or $|3(N-1)\rangle \rightarrow |3(N-2)\rangle$, corresponding to the bare state transitions $|1\rangle \rightarrow |3\rangle$ and $|2\rangle \rightarrow |4\rangle$, respectively. Since the final and initial states of the two cascades are identical, the corresponding transition amplitudes may interfere.

to the various time orderings of the photons will interfere. To illustrate this fact we consider only a cascade of two photons, one emitted on a π transition and the other on a σ transition.

This situation is shown in Fig. 3. The cascade of spontaneously emitted photons is most suitably described in the dressed state basis, $|1(N)\rangle = \sin\Theta|1, N\rangle + \cos\Theta|3, N+1\rangle$, $|2(N)\rangle = \cos\Theta|1, N\rangle - \sin\Theta|3, N+1\rangle$, $|3(N)\rangle = \sin\Theta|2, N\rangle - \cos\Theta|4, N+1\rangle$, $|4(N)\rangle = \cos\Theta|2, N\rangle + \sin\Theta|4, N+1\rangle$, where $\tan 2\Theta = 2\Omega/\Delta$ $(0 < \Theta < \pi/2, \Omega > 0)$. Consider that the atom is initially in the dressed state $|2(N)\rangle$, where N stands for the number of laser photons. By the emission of a σ photon, the atom may decay to the state $|3(N-1)\rangle$ within the manifold with $N-1$ laser photons. The subsequent emission of a π photon on the $|2\rangle \rightarrow |4\rangle$ transition will take the atom to $|3(N-2)\rangle$. If the time order of the two spontaneously emitted photons is reversed, the atom decays first to $|2(N-1)\rangle$ by the emission of a π photon on the $|1\rangle \rightarrow |3\rangle$ transition, and then to the final state $|3(N-2)\rangle$ under the emission of a σ photon. Since the two cascades in Fig. 3 have the same initial and final states, and since it is in principle impossible to determine the quantum path taken by the system, the two transition amplitudes corresponding to different time orders of photon emissions interfere. In one of the transition amplitudes the π photon stems from the $|1\rangle \rightarrow |3\rangle$ transition, and in the other from the $|2\rangle \rightarrow |4\rangle$ transition. Exactly this mechanism gives rise to the interference effects in the fluorescence spectrum that are mediated by the cross-damping terms in Eq. (3). The preceding discussion also implies that the experimental setup—potentially after the photon emissions—decides if interference takes place, a feature that is also known from quantum eraser schemes [4,13].

Finally, we consider a detector with a finite frequency resolution $\lambda = \Delta\omega$ that allows us to study the continuous transition from perfect frequency resolution to perfect time resolution. With increasing time resolution, the observer can in principle obtain more information about the quantum path taken by the atom. Since this rules out the interference mechanism as described above, we deduce that the signature of interference in the fluorescence spectrum diminishes with an increasing filter bandwidth λ. This is completely analogous to a two-slit experiment, where the visibility of the interference pattern is reduced at the cost of which-path information and vice versa [14]. The mean number of photons emitted per unit time is at most on the order of the total decay rate γ of each upper state. It follows that if the time resolution of the detector is improved beyond γ^{-1}, the radiative cascade of photons can be observed in a time resolved way, and hence the signature of interference in the fluorescence spectrum should vanish. Figure 2(b) shows the fluorescence spectrum as recorded by a detector with frequency resolution $\lambda = \gamma$ that could allow for a time resolution on the order of γ^{-1}. The fluorescence spectra with and without the interference terms are now virtually the same, implying that the signature of interference vanished. Thus the time-energy uncertainty analysis quantitatively describes our system, but no conclusion about the hierarchy between the uncertainty relation and complementarity can be drawn from our results.

In summary, we have shown that there is quantum interference in the spectrum of resonance fluorescence from the π transitions under conditions of no interference in the total intensity, enforced by the principle of complementarity. For the system considered here, it claims that it is impossible to observe the temporal and the energy aspect of the radiative cascade of the atom at the same time. If the fluorescence spectrum is observed, the photon emission times are indeterminate. This allows for interference between different time orders of photon emissions that explains the interference in the fluorescence spectrum. Finally, the system is an interesting candidate for an experimental verification of vacuum-induced interference effects.

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