In this article, we describe how to develop a mode converter that transforms a plane electromagnetic wave into an inward moving dipole wave. The latter one is intended to bring a single atom or ion from its ground state to its excited state by absorption of a single photon wave packet with near-100% efficiency.

I. MOTIVATION

The interaction of a single quantum of the electromagnetic field and a single absorber is one of the fundamental processes in optics and in physics in general. Yet it is difficult to study the dynamics of an individual absorption/emission process. In addition there is an apparent asymmetry between emission and absorption. While an isolated excited atom will definitely lead to the emission of a photon, the reverse process is usually not very efficient: a single photon impinging on an atom will only occasionally be absorbed, this corresponds to weak coupling. Placing the atom in a cavity [1, 2] constitutes one possibility of achieving strong coupling, and there exist numerous experimental demonstrations (see, e.g., Refs. [3, 4, 5, 6]). When attempting to achieve similar strong coupling in free space without a cavity – leaving the free space density of modes of the electromagnetic field unmodified – one could take guidance from the emission process [7].

In free space, the process of spontaneous emission of a single photon is characterized by an exponential decay of the upper state population of the emitter [9], which is due to the interaction of the emitter with an infinite number of plane wave electromagnetic modes. These modes can be considered as a heat bath [10], and hence the process of spontaneous emission in free space is often regarded as an irreversible process. This irreversibility is, however, to be understood in a pure thermodynamical sense, i.e., not as an in principle violation of time reversal symmetry. The latter is expected only in connection with CP-violation [11]. The reversibility of the process can be inferred from the fact that the Schrödinger equation is invariant under time reversal for a closed system with a Hamiltonian with CP-violation [11]. The reversibility of the process can be inferred from the fact that the Schrödinger equation is invariant under time reversal for a closed system with a Hamiltonian without any explicit time dependence. Since time reversal refers to an inversion of the evolution in all degrees of freedom, we thus conjecture that a single photon light field can be absorbed completely provided it was generated by such an inversion [8].

In the case of an atomic dipole transition, one should obtain this effect – absorption with a probability of one – if one is able to artificially create the time reversed version of the dipole wave that is emitted by the atom, i.e., a dipole wave moving towards the atom with its properties matched to the respective atomic dipole transition. Any deviation from the perfectly reverted wave reduces the achievable probability for absorption as evidenced by calculations in Ref. [12], where the temporal shape was not matched. We can rephrase the underlying question as follows: Is there a single-photon π-pulse that brings a system in free space from its ground state to its excited state?

In Sec. II we describe how such a wave may be created by use of a mode converter that basically consists of a parabolic mirror and optical elements which tailor the spatial and temporal distribution of the light field incident onto the parabolic mirror. The mathematical details of the procedure of obtaining the ideal spatial field distribution have been discussed recently elsewhere [13]. Therefore, the essential steps of this procedure are only reviewed briefly here. The mode converter proposed here may prove useful in quantum computation schemes, where a reliable transfer of information from photons to atoms or ions is desirable, in biophysical microscopy applications, as well as in investigations of fundamental light-matter interaction itself. In Sec. III we discuss the influence of the energy level structure of the absorber, highlighting the importance of a clean two-level system for obtaining an absorption probability close to unity and proposing possible atomic species which closely resemble a two-level system. Finally, in Sec. IV the coupling scheme proposed here is compared to already established coupling schemes.

II. DESCRIPTION OF THE MODE CONVERTER

It has already been pointed out in Refs. [8, 14, 15], that the probability to bring an atom from its ground state to its excited state upon illumination strongly depends on the overlap of the illuminating light wave with the dipole wave that corresponds to the atomic transition. In the mode converter described here (see Fig. 1), this overlap will be achieved by use of a deep parabolic mirror [16], as has been described recently [13]. Due to the deepness of the mirror, the illumination of the absorber is achieved from a solid angle of almost 4π.

The emitter, e.g., an ion, is located at the focus of the parabolic mirror. To obtain the optimum illuminating intensity distribution, one starts by studying the reverse process. In case of a linear dipole oscillating parallel to the optical axis, the emitted intensity pattern is proportional to \( \sin^2(\theta) \) [17], with \( \theta \) being the angle between the optical axis and the Poynting vector. Each angle \( \theta \) corresponds to a certain radial distance to the optical axis after reflection off the mirror. With this procedure one obtains the transverse intensity distribution of the light field collimated parallel to the optical axis with...
as a function of the radial mirror coordinate (see Ref. [13] for a complete derivation). The resulting transverse distribution shown in Fig. 2 is precisely the input intensity distribution required for excitation. This desired intensity distribution, which has considerable overlap with a Laguerre-Gaussian mode of zeroth radial order and first azimuthal order, can be obtained using a combination of two diffractive optical elements [13].

In order to obtain the correct dipole wave in the mirror focus, all partial waves have to arrive there with the same phase. This is simply achieved by a flat phase distribution of the incident wave in the case of a perfect parabolic mirror. In reality, the parabolic mirror exhibits deviations from the parabolic form leading to wave front aberrations. Furthermore, the phase shift which an incident wave experiences upon reflection is in general a function of the angle of incidence with respect to the surface normal at a certain position on the mirror. Both effects can be compensated for by placing an aberration compensating phase plate in front of the mirror. This ensures that after reflection off the mirror the incident wave arrives at the focus with the same phase from all directions. Finally, an additional diffractive optical lens which transmits most of the light in zeroth order and deflects some light directly to the focus in the first diffraction order may compensate for the otherwise incomplete angular distribution of the created dipole pattern. This has to be performed in such a way that the light directly incident onto the atom and the light reflected by the mirror interfere constructively. However, even without such an element a parabolic mirror of a depth of approximately six times the focal length already covers 94% of the light power required for the total radiation pattern of a linear dipole oscillating along the mirror axis [13].

The polarization of the dipole pattern of the atomic transition under investigation is created by tailoring the polarization of the incident field. For the case of a linear dipole oscillating along the optical axis of the mirror, the incident field has to be radially polarized. This is shown schematically in Fig. 1. In the case of a lens of high numerical aperture, radial polarization was proven to produce a small focus with the focussed electric field parallel to the optical axis in the focal spot [8, 18]. Radial polarization can be generated e.g. by sending a linearly polarized beam through a sectioned half wave plate [19, 20], where the optical axis of the half wave plate is rotated from segment to segment, or by use of liquid crystal cells [20].

The inversion of an outgoing atomic dipole wave must be realized for all relevant frequencies, i.e., over the entire atomic spectrum. Thus, a single photon π-pulse that is meant to mimic such an inverted wave has to have the spectral properties of the atomic transition. This is realized by a pulse of exponential shape, where the time constant of the exponential function is the lifetime of the atomic transition. The carrier frequency of the pulse is the optical frequency of the transition under consideration. Since spontaneous emission manifests itself by an exponentially decreasing probability to detect an emitted photon [21], the π-pulse that corresponds to the time inverted wave must have an exponentially increasing shape.

III. INFLUENCE OF ENERGY LEVEL STRUCTURE

The success of the coupling scheme employing a 4π mode converter depends also on the energy level structure of the emitter which is to be excited. Let us assume there is more than one decay channel from the upper level, say two as it is the case for an atom with Λ-like level structure. When exciting the system from one lower state, the system may decay into the second lower state before completing the π-rotation of the Bloch vector describing the transition from the first lower state to the excited state. Moreover, if there is more than one decay
channel from the excited state, the state of the emitter after the decay process is a superposition of the ground states of all decay channels. The corresponding emitted photon state is entangled with this atomic superposition state. One possibility would be to wait for a spontaneously emitted photon and to time reverse this single photon pulse and thus the whole process by, e.g., phase conjugation. Of course the time reversal of the atomic subsystem would pose a serious difficulty. Moreover, such a process would induce excess noise [23]. Therefore, we restrict the discussion to the ideal case of a simple two-level system.

To obtain a true two-level system several criteria must be fulfilled. First, there must be no hyperfine splitting, since otherwise the ground state will be a multiplet and more than one decay channel exist from the upper state. This criterion calls for an atom with an even number of protons and an isotope with an even number of neutrons. Furthermore, the ground state must have a total angular momentum of \( J = 0 \) for the same reasons (this sets \( J = 1 \) for the upper level). Finally, the only dipole allowed transition from the excited state must be the one to the ground state. One possible candidate for a true two-level transition in neutral atoms could be the \( ^1S_0 \rightarrow ^3P_1 \) transition at 657 nm in neutral \(^{40}\)Ca. If an ion is used it has to be doubly ionized. Otherwise the requirement for a nucleus with an even number of protons and neutrons and \( J = 0 \) total electron angular momentum of the ground state cannot be fulfilled. A possible candidate for such a transition is the \( ^1S_0 \rightarrow ^3P_1 \) transition at 252 nm in \(^{174}\)Yb\(^{2+}\).

We note that similar strong coupling will also be achievable for a \( \Lambda \)-like level structure when inducing the Raman coupling between the two lower states with a bimodal spectrum of the exciting laser fields as demonstrated in a cavity (see, e.g., Ref. [7]). This coupling scheme [23] can also be transferred to the free space mode converter situation.

One could also think of conducting the experiment with an atom having more than one ground state. This would require circularly polarized light to optically pump the system into a cycling transition. But again the circular polarization would lead to unwanted complications, such as an angular emission-absorption pattern requiring light intensity on the optical axis.

![FIG. 3: Sketch of the geometry inside a resonator. The gray solid lines depict the resonator mirrors. \( L \) is the resonator length. \( \omega_0 \) is the beam waist of a Gaussian beam. \( \theta \) is the divergence angle of the beam.](image)

IV. COMPARISON OF THE MODE CONVERTER WITH OTHER COUPLING SCHEMES

A quantitative comparison between the photon-atom coupling in free space and inside a resonator requires a properly defined figure of merit. In cavity quantum electrodynamics the established figure of merit is \( F_g = g^2/(\kappa \gamma) \) (see, e.g., Refs. [24, 25]). \( F_g \) > 1 defines the strong coupling regime, where \( g \), \( \kappa \) and \( \gamma \) are the coupling between the atom and the cavity mode, the cavity photon decay rate and the free space spontaneous emission rate. Miller et al. refer to \( 1/F_g \) as the 'number of strongly coupled atoms necessary to affect appreciably the intracavity field' [24]. This figure of merit is not immediately applicable to the free space case, because \( \kappa \) is not well defined in free space. However, as can be deduced from the discussion in Ref. [26], \( F_g \) is largely determined by the geometry of the cavity. Adapting this train of thought, we compare in what follows the photon-atom coupling achievable in free space to that demonstrated in cavity quantum electrodynamics, which is related to the figure of merit \( F_g \).

In free space maximum coupling is achieved if the incoming photon wave packet occupies the full solid angle which is \( \Omega = \pi R^2/\lambda \), including a weighting factor for the angular radiation pattern of the linear or circular dipole transition. Provided that the incident field has full overlap with the atomic dipole radiation pattern [8, 15], the coupling in free space is maximum if the covered solid angle is equal to \( \Omega \). A reduced solid angle of \( \Delta \Omega \) will result in an absorption probability \( P_{abs,cav} = \Delta \Omega/\Omega = 3\Delta \Omega/\Omega_{max} \leq 1 \). Thus, the maximum absorption probability achievable with the proposed mode converter is unity.

We will now connect this approach to the cavity case. The geometry of an atom residing inside a resonator is depicted in Fig. 3 We assume that the atom is located at the position of the beam waist and that it is illuminated by a single photon pulse of Gaussian beam shape with beam waist \( \omega_0 \) and wavelength \( \lambda \). The divergence angle of the beam is \( \theta \), which is taken to be small with \( 1 \gg \theta \approx \lambda/(\pi \omega_0) \). Then, the solid angle from which the photon is incident onto the atom is \( \Omega_{cav} = \pi \theta^2 \) for a single passage by the atom, where we have implicitly supposed that the quantization axis of the atom is oriented such that the angular radiation pattern has its maximum in the direction of the cavity mirrors.

One thus arrives at a single passage absorption probability of \( \Delta \Omega_{cav}/\Omega = 3\lambda^2/(8\pi^2\omega_0^2) \), which is a number much smaller than one. However, inside a cavity with a reflectivity \( R \) of each mirror the photon passes the atom \( N = 1/(1-R) \) times on average, which results in a figure of merit

\[
F_{\Omega} = N \frac{\Delta \Omega_{cav}}{\Omega} = \frac{3\lambda^2}{8\pi^2\omega_0^2} \frac{1}{1-R} .
\]

For high quality factor resonators or high \( R \), respectively, \( F_{\Omega} \) becomes much larger than one. This reflects the situation commonly encountered in the strong coupling regime of cavity quantum electrodynamics, where a photon is absorbed and re-emitted repeatedly for many times before the photon escapes the cavity. One can thus define the probability that the photon is absorbed at least once inside a cavity as

\[
P_{abs,cav} = \begin{cases} F_{\Omega}, & \text{if } F_{\Omega} \leq 1 \\ 1, & \text{if } F_{\Omega} > 1 \end{cases}.
\]
This demonstrates that by employing the mode converter efficiency absorption should be possible in free space to the same extent as in a cavity.

We note that it is straightforward to show that the two figures of merit $F_e$ and $F_{\Omega}$ are identical. Inserting the definitions of $g$ [25], $\gamma$ [21], using $\kappa \simeq (1-R)c_0/L$ and approximating the cavity mode volume with $\pi w_0^2L$ proves the identity.

In the remainder of this section, we argue that the mode converter does not modify the free space density of modes. The presence of the surface of the parabolic mirror might be expected to result in disturbing effects similar to the ones observed in the case of emitters that are positioned close to surfaces within a near field distance (see, e.g., Ref. [27] and citations therein). Since the distance of the atom/ion to the mirror surface in case of a realistic parabolic mirror is much larger than a wavelength, near field effects do not play a role [27, 28]. In other words, the focusing mirror does not modify the density of modes at the focus. This would be different if the atom was located on the optical axis at a distance of the vertex radius of curvature away from the mirror vertex — a situation similar to the scenario examined experimentally in Ref. [29] and theoretically in Ref. [30]. There, a finite fraction of the light emitted by an ion was focussed onto the ion by use of a lens and a mirror, leading to a change of the spontaneous emission rate despite a large distance between ion and surface. However, if the emitter is located at the mirror focus, the amount of light which is back reflected from the mirror to the emitter covers a vanishingly small part of the full solid angle and thus should not have a considerable effect. In the special case of a linear dipole oscillating along the mirror axis, emission does not occur in this direction at all. A modification of the spontaneous emission rate as observed for emitters inside resonators [3, 31] will therefore not occur. Thus, the mode converter allows for investigations of light-atom interaction in the ‘natural’ free space environment of the atom.

V. OUTLOOK

There are several reports in the literature on experiments involving optical excitation of single quantum systems in free space, including far field excitation [32, 33] as well as near field excitation [34]. The most successful free space coupling reported so far was characterized by 12% extinction of the excitation light by a single quantum dot [33]. However, in none of these experiments the quantum systems were excited with single photon pulses tailored to the specific optical transition in all aspects. We believe that this will be possible with the mode converter described here. The use of this mode converter is expected to result in a coupling efficiency or absorption probability, respectively, of almost 100%. The achieved absorption will sensitively depend on the overlap of the generated single photon wave packet with a time reversed dipole wave.

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