## Lecture 23: Effective interaction Hamiltonians

- Effective interaction Hamiltonians
- Effective squeezing operator, parametric down-conversion

Effective interaction Hamiltonians: We will go back to the question of effective interaction Hamiltonians and find out more about atom-light interactions beyond the linear approximation that led us to the optical Bloch equations. We have already seen in connection with the Jaynes-Cummings model in cavity QED that in certain limits, in this case the dispersive limit where the detuning is assumed to be large compared to the $n$-photon Rabi frequency, the time evolution operator can take rather simple forms. In the dispersive limit, it reads [Eq. (21.6)]

$$
\hat{U}(t)=\exp \left(-\frac{i}{\hbar} \hat{H}_{0} t\right)\left[\exp \left(-i \frac{g^{2}}{\delta}(\hat{n}+1) t\right)|e\rangle\langle e|+\exp \left(i \frac{g^{2}}{\delta} \hat{n} t\right)|g\rangle\langle g|\right] .
$$

The interaction part is, for small times $t$, given by

$$
\begin{align*}
\hat{U}_{\mathrm{int} t}(t) & \approx\left(1-i \frac{g^{2}}{\delta}(\hat{n}+1) t+\ldots\right) \hat{\sigma}_{e e}+\left(1+i \frac{g^{2}}{\delta} \hat{n} t+\ldots\right) \hat{\sigma}_{g g}  \tag{23.1}\\
& =1-i \frac{g^{2}}{\delta} \hat{n}\left(\hat{\sigma}_{e e}-\hat{\sigma}_{g g}\right) t-i \frac{g^{2}}{\delta} \hat{\sigma}_{e e} t+\ldots \tag{23.2}
\end{align*}
$$

The last term can be included in the free Hamiltonian and amounts to a shift of the excited state (Stark effect) whereas the second term stems from an effective interaction Hamiltonian

$$
\begin{equation*}
\hat{H}_{\mathrm{eff}}=\frac{\hbar g^{2}}{\delta} \hat{a}^{\dagger} \hat{a} \hat{\sigma}_{z} \tag{23.3}
\end{equation*}
$$

This Hamiltonian is trilinear in all operators and as such does not appear in the original Jaynes-Cummings Hamiltonian.

We will now return to the electric-dipole interaction Hamiltonian for multi-level atoms [Eq. (15.5)]

$$
\begin{equation*}
\hat{H}_{\mathrm{int}}=-i \sum_{i, j} \sum_{\lambda} \omega_{i j} \hat{\sigma}_{i j} \mathbf{d}_{i j} \cdot \mathbf{A}_{\lambda}\left(\mathbf{r}_{A}\right) \hat{a}_{\lambda}+\text { h.c. }=\hbar \sum_{i, j} \sum_{\lambda} g_{i j, \lambda} \hat{\sigma}_{i j} \hat{a}_{\lambda}+\text { h.c. } \tag{23.4}
\end{equation*}
$$

where we defined the coupling strengths $g_{i j, \lambda}=\omega_{i j} \mathbf{d}_{i j} \cdot \mathbf{A}_{\lambda}\left(\mathbf{r}_{A}\right) /(i \hbar)$. Heisenberg's equations of motion for the atomic and photonic operators are then

$$
\begin{align*}
& \dot{\hat{a}}_{\lambda}=-i \omega_{\lambda} \hat{a}_{\lambda}-i \sum_{i, j} g_{j i, \lambda}^{*} \hat{\sigma}_{i j},  \tag{23.5}\\
& \dot{\hat{\sigma}}_{i j}=i \omega_{i j} \hat{\sigma}_{i j}-i \sum_{k} \sum_{\lambda}\left[\left(g_{j k, \lambda} \hat{\sigma}_{i k}-g_{k i, \lambda} \hat{\sigma}_{k j}\right) \hat{a}_{\lambda}+\left(g_{k j, \lambda}^{*} \hat{\sigma}_{i k}-g_{i k, \lambda}^{*} \hat{\sigma}_{k j}\right) \hat{a}_{\lambda}^{\dagger}\right] \tag{23.6}
\end{align*}
$$

(note that $\left[\hat{\sigma}_{i j}, \hat{\sigma}_{i^{\prime} j^{\prime}}\right]=\delta_{j i^{\prime}} \hat{\sigma}_{i j^{\prime}}-\delta_{i j^{\prime}}{\hat{a^{\prime}} \mathbf{j}}$ ). The formal solution to Eq. (23.6) is then

$$
\begin{align*}
& \hat{\sigma}_{i j}(t)=e^{i \omega_{i j} t} \hat{\sigma}_{i j}-i \sum_{k} \sum_{\lambda} \int_{0}^{t} d t^{\prime} e^{i \omega_{i j}\left(t-t^{\prime}\right)}\left\{\left[g_{j k, \lambda} \hat{\sigma}_{i k}\left(t^{\prime}\right)-g_{k i, \lambda} \hat{\sigma}_{k j}\left(t^{\prime}\right)\right] \hat{a}_{\lambda}\left(t^{\prime}\right)\right. \\
&+ {\left.\left[g_{k j, \lambda}^{*} \hat{\sigma}_{i k}\left(t^{\prime}\right)-g_{i k, \lambda}^{*} \hat{\sigma}_{k j}\left(t^{\prime}\right)\right] \hat{a}_{\lambda}^{\dagger}\left(t^{\prime}\right)\right\} . } \tag{23.7}
\end{align*}
$$

Up until now, we have merely repeated some of the derivations that led to the optical Bloch equations, only for multilevel atoms. Note that we have not made any rotating-wave approximation yet.

Suppose now that the incoming light contains waves of only three frequencies that add up as $\omega_{1}=\omega_{2}+\omega_{3}$, none of which is resonant with any atomic transition (see Figure 31). Iterating Eq. (23.7) once more and inserting the result into Eq. (23.5), we obtain, amongst


FIG. 31: A multilevel atom interacts non-resonantly with light of only three distinct frequencies.
many other, the following terms:

$$
\begin{align*}
\dot{\hat{a}}_{\lambda}= & -i \omega_{\lambda} \hat{a}_{\lambda}+i \sum_{i, j, k, l} \sum_{\mu, \nu} g_{j i, \lambda}^{*} \int_{0}^{t} d t^{\prime} e^{i \omega_{i j}\left(t-t^{\prime}\right)}\{ \\
& g_{j k, \mu} \int_{0}^{t^{\prime}} d t^{\prime \prime} e^{i \omega_{i k}\left(t^{\prime}-t^{\prime \prime}\right)}\left[g_{k l, \nu} \hat{\sigma}_{l l}\left(t^{\prime \prime}\right)-g_{l i, \nu} \hat{\sigma}_{l k}\left(t^{\prime \prime}\right)\right] \hat{a}_{\nu}\left(t^{\prime \prime}\right) \hat{a}_{\mu}\left(t^{\prime}\right) \\
& \left.-g_{k i, \mu} \int_{0}^{t^{\prime}} d t^{\prime \prime} e^{i \omega_{k j}\left(t^{\prime}-t^{\prime \prime}\right)}\left[g_{j l, \nu} \hat{\sigma}_{k l}\left(t^{\prime \prime}\right)-g_{l k, \nu} \hat{\sigma}_{l j}\left(t^{\prime \prime}\right)\right] \hat{a}_{\nu}\left(t^{\prime \prime}\right) \hat{a}_{\mu}\left(t^{\prime}\right)\right\} \tag{23.8}
\end{align*}
$$

The dropped terms contain either atomic operators at the initial time or photonic creation operators which we neglect for the following reason. Suppose we let $\lambda=1$ and we perform the rotating-wave approximation (RWA) in which we keep all terms on the rhs of Eq. (23.8) that are resonant with the lhs, i.e. that evolve approximately as $e^{-i \omega_{1} t}$. Since $\omega_{1}=\omega_{2}+\omega_{3}$,
the time evolution can only come from the photonic operators $\hat{a}_{\nu}\left(t^{\prime \prime}\right) \hat{a}_{\mu}\left(t^{\prime}\right)(\mu, \nu=2,3)$, and all the time evolutions of the atomic operators have to cancel out. The only atomic operators that are allowed are the diagonal projectors $\hat{\sigma}_{i i}$. Hence, the equation of motion for the photonic operator can be brought into the form

$$
\begin{equation*}
\dot{\hat{a}}_{1}(t)=-i \omega_{1} \hat{a}_{1}(t)-i \sum_{i} G_{i, 23} \hat{\sigma}_{i i}(t) \hat{a}_{2}(t) \hat{a}_{3}(t), \tag{23.9}
\end{equation*}
$$

where the new effective coupling constants $G_{i, 23}$ are rather complicated functions (sums and products) of the original couplings $g_{i j, \lambda}$.
Effective squeezing operator, parametric down-conversion: The structure of the effective equation of motion (23.9) is such that it could have been derived from the effective Hamiltonian

$$
\begin{equation*}
\hat{H}_{\mathrm{eff}}=\hbar \hat{a}_{1}^{\dagger} \hat{a}_{2} \hat{a}_{3} \sum_{i} G_{i, 23} \hat{\sigma}_{i i}+\text { h.c. } \tag{23.10}
\end{equation*}
$$

From the same Hamiltonian (23.10) we find that $\dot{\hat{\sigma}}_{i i}=0$, so that the atomic variables can be completely eliminated from the equations of motion. If the atom was initially in its ground state, it will remain there for all times, and the effective Hamiltonian takes the even simpler form

$$
\begin{equation*}
\hat{H}_{\mathrm{eff}}=\hbar \kappa \hat{a}_{1}^{\dagger} \hat{a}_{2} \hat{a}_{3}+\text { h.c. } . \tag{23.11}
\end{equation*}
$$

This Hamiltonian describes the process of sum-frequency generation in which two light modes of frequencies $\omega_{2}$ and $\omega_{3}$ combine to a light mode at the sum frequency $\omega_{1}=\omega_{2}+\omega_{3}$. The atomic degrees of freedom do not appear explicitly in (23.11), only in the effective coupling constant $\kappa$.

If we furthermore assume that the light mode with frequency $\omega_{1}$ has been prepared in a strong coherent state $|\alpha\rangle$ so that its operator character can be neglected, and if we assume that $\omega_{2}=\omega_{3}$, we arrive at the parametric approximation with an effective Hamiltonian

$$
\begin{equation*}
\hat{H}_{\mathrm{eff}}=\hbar \kappa \alpha_{1}^{*} \hat{a}^{2}+\text { h.c. . } \tag{23.12}
\end{equation*}
$$

The unitary time evolution generated by this Hamiltonian is

$$
\begin{equation*}
\hat{U}(t)=e^{-\frac{i}{\hbar} \hat{H}_{\text {eff }} t}=e^{\left(-i \kappa \alpha_{1}^{*} \hat{a}^{2}-i \kappa^{*} \alpha_{1} \hat{a}^{\dagger}\right) t} \tag{23.13}
\end{equation*}
$$

which, letting $\kappa \mapsto i \kappa$, is equivalent to the squeezing operator $\hat{S}(\xi)$ [Eq. (5.17)] with $\xi=$ $2 \kappa \alpha_{1} t$. We can therefore generate squeezed light by interacting with atomic ensembles in the strongly dispersive regime in which the atoms remain in their ground states for all times.

In nonlinear optics, the derivation of effective Hamiltonians is performed in a similar way. The main difference is that one does not start with individual atoms but usually with a semi-classical theory in which a nonlinear dielectric material (the atomic ensemble in our case) is described by phenomenological susceptibilities $\chi^{(2)}$ which take the place of the (microscopic) coupling constant $\kappa$. Typical examples of crystals with a non-vanishing $\chi^{(2)}$ nonlinear coefficient are Barium titanate $\mathrm{BaTiO}_{3}$, Lithium niobate $\mathrm{LiNbO}_{3}$, and KDP $\mathrm{KH}_{2} \mathrm{PO}_{4}$. The process described by the Hamiltonian (23.12) is known as parametric downconversion which, in the low-intensity regime, leads to the production of correlated singlephoton pairs with entangled polarizations.


FIG. 32: Parametric down-conversion from a BBO crystal. In the points of intersection of the two cones the quantum state of the photons is exactly a maximally entangled state in the polarization basis.

## Lecture 24: Optical harmonic generation (frequency-doubling)

- Hamiltonian description of optical harmonic generation
- Squeezing in optical harmonic generation

Hamiltonian description of optical harmonic generation: We have seen in the previous lecture that off-resonant atom-light interactions can lead to an effective Hamiltonian of the form [see Eq. (23.11)]

$$
\begin{equation*}
\hat{H}_{\mathrm{eff}}=\hbar \kappa \hat{a}_{1}^{\dagger} \hat{a}_{2} \hat{a}_{3}+\text { h.c. } \tag{24.1}
\end{equation*}
$$

where the coupling constant $\kappa$ depends on the details of the atom-light coupling (dipole moments, transition frequencies etc.). The process of optical harmonic generation is one of the best known examples in nonlinear optics. Here a monochromatic light beam of frequency $\omega_{1}$ incident on a $\chi^{(2)}$ nonlinear medium generates light at twice the frequency, $\omega_{2}=2 \omega_{1}$ (see Fig. 33). Hence, we can describe the evolution of the electromagnetic field at these two


FIG. 33: Schematic description of second-harmonic generation. The relevant light frequencies are far away from any atomic transition so that the effective description as a nonlinear process holds.
frequencies by the Hamiltonian

$$
\begin{equation*}
\hat{H}=\hbar \omega_{1}\left(\hat{n}_{1}+\frac{1}{2}\right)+\hbar \omega_{2}\left(\hat{n}_{2}+\frac{1}{2}\right)+\hbar \kappa\left(\hat{a}_{1}^{\dagger 2} \hat{a}_{2}+\hat{a}_{2}^{\dagger} \hat{a}_{1}^{2}\right) . \tag{24.2}
\end{equation*}
$$

From the Hamiltonian (24.2) we find immediately that the combination $\hat{n}_{1}+2 \hat{n}_{2}$ is a constant of motion, i.e. $\left[\hat{n}_{1}+2 \hat{n}_{2}, \hat{H}\right]=0$. This means that for every emitted harmonic photon of frequency $\omega_{2}$ two fundamental photons of frequency $\omega_{1}$ have to be absorbed. Therefore, the process of harmonic generation conserves energy, but not photon numbers.

We will now try to solve the time evolution of the photonic amplitude operators for short interaction times. To do so, we split up the fast time evolution and introduce slowly varying
amplitude operators as usual: $\hat{a}_{1}=e^{-i \omega_{1} t} \hat{\tilde{a}}_{1}$ and $\hat{a}_{2}=e^{-i \omega_{2} t} \hat{\tilde{a}}_{2}$. Heisenberg's equations of motion for the slowly varying amplitude operators are (we omit the $\sim$ sign again)

$$
\begin{align*}
& \dot{\hat{a}}_{1}=\frac{1}{i \hbar}\left[\hat{a}_{1}, \hat{H}\right]+\frac{\partial \hat{a}_{1}}{\partial t}=-2 i \kappa \hat{a}_{1}^{\dagger} \hat{a}_{2},  \tag{24.3}\\
& \dot{\hat{a}}_{2}=\frac{1}{i \hbar}\left[\hat{a}_{2}, \hat{H}\right]+\frac{\partial \hat{a}_{2}}{\partial t}=-i \kappa \hat{a}_{1}^{2} . \tag{24.4}
\end{align*}
$$

The second time derivatives are computed similarly as

$$
\begin{align*}
& \ddot{\hat{a}}_{1}=-2 i g\left(\dot{\hat{a}}_{1}^{\dagger} \hat{a}_{2}+\hat{a}_{1}^{\dagger} \dot{\hat{a}}_{2}\right)=4 \kappa^{2}\left(\hat{n}_{2}-\frac{\hat{n}_{1}}{2}\right) \hat{a}_{1},  \tag{24.5}\\
& \ddot{\hat{a}}_{2}=-i g\left(\dot{\hat{a}}_{1} \hat{a}_{1}+\hat{a}_{1} \dot{\hat{a}}_{1}\right)=-4 \kappa^{2}\left(\hat{n}_{1}+\frac{1}{2}\right) \hat{a}_{2} . \tag{24.6}
\end{align*}
$$

The equations of motion are once again coupled differential equations that have no analytic solution. However, we can seek a solution for very short interaction times by Taylor expanding around the initial time $t=0$. The interaction time is more or less the propagation time through the nonlinear medium, such an approximation can be justified. The Taylor expansion leads to

$$
\begin{align*}
\hat{a}_{1}(t) & \approx \hat{a}_{1}(0)+t \dot{\hat{a}}_{1}(0)+\frac{t^{2}}{2} \ddot{\hat{a}}_{1}(0)+\ldots \\
& =\hat{a}_{1}(0)-2 i(\kappa t) \hat{a}_{1}^{\dagger}(0) \hat{a}_{2}(0)+2(\kappa t)^{2}\left(\hat{n}_{2}(0)-\frac{\hat{n}_{1}(0)}{2}\right) \hat{a}_{1}(0)+\ldots,  \tag{24.7}\\
\hat{a}_{2}(t) & \approx \hat{a}_{2}(0)+t \dot{\hat{a}}_{2}(0)+\frac{t^{2}}{2} \ddot{\hat{a}}_{2}(0)+\ldots \\
& =\hat{a}_{2}(0)-i(\kappa t) \hat{a}_{1}^{2}(0)-2(\kappa t)^{2}\left(\hat{n}_{1}(0)+\frac{1}{2}\right) \hat{a}_{2}(0)+\ldots \tag{24.8}
\end{align*}
$$

where we explcitly used the first and second derivatives. The expressions are valid as long as $\left\langle\hat{n}_{1}(0)\right\rangle(\kappa t)^{2} \ll 1$. Since we know the time evolution of the (slowly varying) photonic amplitude operators in the Heisenberg picture, we can construct all other composite operators like photon numbers from them and compute their expectation values.

Let us assume that the pump field at frequency $\omega_{1}$ is prepared in a coherent state with amplitude $\alpha$, and the harmonic mode is initially empty. Hence, the initial state of both modes at time $t=0$ is $|\psi(0)\rangle=\left|\alpha_{1}, 0_{2}\right\rangle$. For the expectation values of the photon number operators we obtain

$$
\begin{align*}
& \left\langle\hat{n}_{1}(t)\right\rangle=\langle\psi(0)| \hat{a}_{1}^{\dagger}(t) \hat{a}_{1}(t)|\psi(0)\rangle \approx|\alpha|^{2}-2(\kappa t)^{2}|\alpha|^{4}+\ldots,  \tag{24.9}\\
& \left\langle\hat{n}_{2}(t)\right\rangle=\langle\psi(0)| \hat{a}_{2}^{\dagger}(t) \hat{a}_{2}(t)|\psi(0)\rangle \approx(\kappa t)^{2}|\alpha|^{4}+\ldots, \tag{24.10}
\end{align*}
$$

showing that the intensity of the generated second harmonic field grows quadratically in time $\left(\left\langle\hat{n}_{2}(t)\right\rangle \propto t^{2}\right)$ as well as with the square of the pump intensity $\left\langle\hat{n}_{2}(t)\right\rangle \propto\left(|\alpha|^{2}\right)^{2}$.

Similarly, one can compute the photon-number variances $\left\langle\left(\Delta \hat{n}_{1}\right)^{2}\right\rangle$ and $\left\langle\left(\Delta \hat{n}_{2}\right)^{2}\right\rangle$. Before we state the results, let us briefly go back to the definition of the coherent states. Recall that their photon-number statistics is Poissonian, with the variance equal to their mean, $\langle\alpha|(\Delta \hat{n})^{2}|\alpha\rangle=\langle\alpha| \hat{n}^{2}|\alpha\rangle-\langle\alpha| \hat{n}|\alpha\rangle^{2}=\langle\alpha| \hat{n}|\alpha\rangle=|\alpha|^{2}$. Any state with a narrower, subPoissonian, photon-number distribution is nonclassical. Without going into the details of the calculations (you can check them yourself), the second moments of the photon numbers are

$$
\begin{align*}
& \left\langle\hat{n}_{1}^{2}(t)\right\rangle \approx\left(|\alpha|^{2}+|\alpha|^{2}\right)\left(1-4(\kappa t)^{2}|\alpha|^{2}\right)+\ldots,  \tag{24.11}\\
& \left\langle\hat{n}_{2}^{2}(t)\right\rangle \approx(\kappa t)^{2}|\alpha|^{4}\left(1+(\kappa t)^{2}|\alpha|^{2}\right)+\ldots, \tag{24.12}
\end{align*}
$$

from which it follows that

$$
\begin{align*}
& \left\langle\left(\Delta \hat{n}_{1}(t)\right)^{2}\right\rangle=\left\langle\hat{n}_{1}^{2}(t)\right\rangle-\left\langle\hat{n}_{1}(t)\right\rangle^{2}=\left\langle\hat{n}_{1}(t)\right\rangle-2(\kappa t)^{2}|\alpha|^{4}+\ldots,  \tag{24.13}\\
& \left\langle\left(\Delta \hat{n}_{2}(t)\right)^{2}\right\rangle=\left\langle\hat{n}_{2}^{2}(t)\right\rangle-\left\langle\hat{n}_{2}(t)\right\rangle^{2}=\left\langle\hat{n}_{2}(t)\right\rangle+\ldots \tag{24.14}
\end{align*}
$$

Therefore, the photon-number statistics of the harmonic field is close to Poissonian, and the quantum state close to a coherent state. On the other hand, the photon-number statistics of the pump field is narrower than the original Poissonian distribution, i.e. sub-Poissonian. Squeezing in optical harmonic generation: Generically, most nonclassical features are equivalent to or caused by other nonclassical features. In the situation considered here, the pump field becomes also squeezed by the nonlinear interaction. Recall that our criterion for squeezing that the fluctuations of the electric field has to drop below its vacuum value. This is equivalent to saying that the quadrature variance has to drop below 1 . The quadrature operator was defined as $\hat{x}(\varphi)=\hat{a} e^{i \varphi}+\hat{a}^{\dagger} e^{-i \varphi}$ (see Lecture 5). Using the solution (24.7), one finds (again without proof) that

$$
\begin{align*}
\left\langle\hat{x}_{1}(\varphi)\right\rangle= & \left(\alpha e^{i \varphi}+\alpha^{*} e^{-i \varphi}\right)\left(1-(\kappa t)^{2}|\alpha|^{2}\right)+\ldots,  \tag{24.15}\\
\left\langle\hat{x}_{1}^{2}(\varphi)\right\rangle= & \left(\alpha^{2} e^{2 i \varphi}+\alpha^{* 2} e^{-2 i \varphi}\right)\left(1-2(\kappa t)^{2}|\alpha|^{2}\right)+2|\alpha|^{2}\left(1-2(\kappa t)^{2}|\alpha|^{2}\right)+1 \\
& -(\kappa t)^{2}\left(\alpha^{2} e^{2 i \varphi}+\alpha^{* 2} e^{-2 i \varphi}\right)+\ldots \tag{24.16}
\end{align*}
$$

so that the quadrature variance becomes

$$
\begin{equation*}
\left\langle\left(\Delta \hat{x}_{1}(\varphi)\right)^{2}\right\rangle=1-2(\kappa t)^{2}|\alpha|^{2} \cos (\varphi+\arg \alpha)+\mathcal{O}(\kappa t)^{3} . \tag{24.17}
\end{equation*}
$$

This means that, depending on the phase of the coherent amplitude of the pump, one can always find a direction $\varphi$ in phase space in which the pump field is squeezed.

Since both sub-Poissonian statistics and squeezing are purely quantum mechanical phenomena, we know that optical harmonic generation cannot be described using purely classical nonlinear optics. Most of the interesting phenomena would be missed by a classical description.

## Lecture 25: Parametric down-conversion

- Solution of the equations of motion
- Photon statistics

Hamiltonian and equations of motion: Parametric down-conversion is a nonlinear process in which a pump photon of frequency $\omega_{0}$ converts into two photons with frequencies $\omega_{1}$ and $\omega_{2}$, commonly called signal and idler photons. Due to energy conservation, the relation $\omega_{0}=\omega_{1}+\omega_{2}$ must hold. Similarly, as the photons carry momentum, the so-called phase-matching condition $\mathbf{k}_{0}=\mathbf{k}_{1}+\mathbf{k}_{2}$ must be satisfied, i.e. the photon momentum is conserved.

The Hamiltonian of this process is, using the notation of the effective interaction Hamiltonian (23.11),

$$
\begin{equation*}
\hat{H}=\sum_{i=0}^{2} \hbar \omega_{i}\left(\hat{n}_{i}+\frac{1}{2}\right)+\hbar \kappa\left(\hat{a}_{0}^{\dagger} \hat{a}_{1} \hat{a}_{2}+\hat{a}_{1}^{\dagger} \hat{a}_{2}^{\dagger} \hat{a}_{0}\right) . \tag{25.1}
\end{equation*}
$$

The combination $2 \hat{n}_{0}+\hat{n}_{1}+\hat{n}_{2}$ is again a constant of the motion, i.e. $\left[2 \hat{n}_{0}+\hat{n}_{1}+\hat{n}_{2}, \hat{H}\right]=0$. In order to find the time evolution of the photonic amplitude operators, we could make in principle the same Taylor expansion for short interaction times as we did for optical harmonic generation. However, if it justified to assume that the pump field is sufficiently intense such that it can be treated as a classical field with complex amplitude $\alpha_{0} e^{-i \omega_{0} t}$, the Hamiltonian simplifies to

$$
\begin{equation*}
\hat{H}=\sum_{i=1}^{2} \hbar \omega_{i}\left(\hat{n}_{i}+\frac{1}{2}\right)+\hbar \kappa\left(\hat{a}_{1} \hat{a}_{2} \alpha_{0}^{*} e^{i \omega_{0} t}+\hat{a}_{1}^{\dagger} \hat{a}_{2}^{\dagger} \alpha_{0} e^{-i \omega_{0} t}\right) . \tag{25.2}
\end{equation*}
$$

The new constant of the motion is $\hat{n}_{1}-\hat{n}_{2}$ meaning that signal and idler photons are always created together.

As before, we introduce slowly-varying amplitude operators as $\hat{a}_{i}(t)=e^{-i \omega_{i} t} \hat{\tilde{a}}_{i}(t)$ where we omit the $\sim$ sign straightaway and write Heisenberg's equations of motion for the slowlyvarying quantities as

$$
\begin{align*}
& \dot{\hat{a}}_{1}=-i \kappa \alpha_{0} \hat{a}_{2}^{\dagger} e^{i\left(\omega_{1}+\omega_{2}-\omega_{0}\right) t}  \tag{25.3}\\
& \dot{\hat{a}}_{2}=-i \kappa \alpha_{0} \hat{a}_{1}^{\dagger} e^{i\left(\omega_{1}+\omega_{2}-\omega_{0}\right) t} . \tag{25.4}
\end{align*}
$$

Using energy conservation, the equations of motion simplify further to

$$
\begin{equation*}
\dot{\hat{a}}_{1}=-i \kappa \alpha_{0} \hat{a}_{2}^{\dagger}, \quad \dot{\hat{a}}_{2}=-i \kappa \alpha_{0} \hat{a}_{1}^{\dagger} . \tag{25.5}
\end{equation*}
$$

Differentiating once more leads to the uncoupled equations

$$
\begin{equation*}
\ddot{\hat{a}}_{1}=\kappa^{2}\left|\alpha_{0}\right|^{2} \hat{a}_{1}, \quad \ddot{\hat{a}}_{2}=\kappa^{2}\left|\alpha_{0}\right|^{2} \hat{a}_{2}, \tag{25.6}
\end{equation*}
$$

with the solutions

$$
\begin{align*}
& \hat{a}_{1}(t)=\hat{a}_{1}(0) \cosh \left(\kappa\left|\alpha_{0}\right| t\right)-i e^{i \arg \alpha_{0}} \hat{a}_{2}^{\dagger}(0) \sinh \left(\kappa\left|\alpha_{0}\right| t\right),  \tag{25.7}\\
& \hat{a}_{2}(t)=\hat{a}_{2}(0) \cosh \left(\kappa\left|\alpha_{0}\right| t\right)-i e^{i \arg \alpha_{0}} \hat{a}_{1}^{\dagger}(0) \sinh \left(\kappa\left|\alpha_{0}\right| t\right) . \tag{25.8}
\end{align*}
$$

Photon statistics: Instead of trigonometric function, the solutions contain hyperbolic functions so that we can expect a strong increase in the production of photons with time. Indeed, if we assume that both signal and idler modes where initially empty, $|\psi(0)\rangle=\left|0_{1}, 0_{2}\right\rangle$, the photon-number expectation values are

$$
\begin{equation*}
\left\langle\hat{n}_{1}(t)\right\rangle=\left\langle\hat{n}_{2}(t)\right\rangle=\sinh ^{2}\left(\kappa\left|\alpha_{0}\right| t\right) . \tag{25.9}
\end{equation*}
$$

For short interaction times, $\kappa\left|\alpha_{0}\right| t \ll 1$, the average number of downconverted photons grows quadratically in time, because $\sinh x \stackrel{x \ll 1}{\approx} x$. That is, initially the process is driven by spontaneous emission of signal and idler photons. For longer times, however, the average number of photons grows exponentially $\left(\sinh x \stackrel{x \gg 1}{\approx} e^{x} / 2\right)$ and the process is dominated by stimulated emission of photons. At this point one needs to make sure that the approximation of constant pump amplitude is still valid. The above solution only holds when the pump field is not substantially depleted by the down-conversion process.

From the solutions (25.7) and (25.8) it is also easy to find the photon-number variance with the result that

$$
\begin{equation*}
\left\langle\left(\Delta \hat{n}_{1}(t)\right)^{2}\right\rangle=\left\langle\left(\Delta \hat{n}_{2}(t)\right)^{2}\right\rangle=\sinh ^{4}\left(\kappa\left|\alpha_{0}\right| t\right)+\sinh ^{2}\left(\kappa\left|\alpha_{0}\right| t\right) . \tag{25.10}
\end{equation*}
$$

Going back to the Hamiltonian (25.2), we find that it generates a time evolution operator $\hat{U}=e^{-i \hat{H} t / \hbar}$ that looks similar to the squeeze operator $\hat{S}(\xi)$ introduced in Lecture 5, with $\xi=-2 i \kappa t \alpha_{0}^{*} e^{i \omega_{0} t}$. We will now verify that the downconverted photons are indeed squeezed. Note, however, that the resulting quantum state consists of two separate modes of the electromagnetic field, hence our squeezing criterion has to be generalized. The individual modes will in general not be squeezed, only the quantum-mechanical correlations between the modes will cause two-mode squeezing. From the correspondence between quantum states
and Wigner functions it is enough to check that the Wigner function shows features in phase space that are narrower than those of a vacuum state.

In connection with the Mach-Zehnder interferometer we have already used the covariance matrix $\mathbf{V}$ whose entries are the (symmetrically ordered) second-order quadrature moments $\left\langle\left(\hat{\lambda}_{i} \hat{\lambda}_{j}+\hat{\lambda}_{j} \hat{\lambda}_{i}\right) / 2\right\rangle$ where $\hat{\boldsymbol{\lambda}}=\left(\hat{x}_{1}, \hat{p}_{1}, \hat{x}_{2}, \hat{p}_{2}\right)^{T}$. In the present situation, after a suitable local rotation, the covariance matrix can be brought into the canonical form

$$
\mathbf{V}=\left(\begin{array}{cccc}
c & 0 & s & 0  \tag{25.11}\\
0 & c & 0 & -s \\
s & 0 & c & 0 \\
0 & -s & 0 & c
\end{array}\right)
$$

with $c=\cosh \left(2 \kappa\left|\alpha_{0}\right| t\right)$ and $s=\sinh \left(2 \kappa\left|\alpha_{0}\right| t\right)$. Its (doubly degenerate) eigenvalues are $e^{ \pm 2 \kappa\left|\alpha_{0}\right| t}$, meaning that there are two directions in the four-dimensional phase space of our two light modes in which the Wigner function is exponentially narrower than its vacuum counterpart. Hence, the down-converted photons are indeed squeezed.
Generation of entangled photon pairs: For very short interaction times, $\kappa\left|\alpha_{0}\right| t \ll 1$, we can a simple expression for the quantum state of the down-converted photons. Taylor expanding the unitary evolution operator to first order in the interaction time leaves us with

$$
\begin{equation*}
\hat{U}(t)=e^{-i \hat{H} t / \hbar} \approx 1-\frac{i t}{\hbar} \hat{H}+\mathcal{O}\left(t^{2}\right) \tag{25.12}
\end{equation*}
$$

which, acting on the initial vacuum state in the interaction picture (i.e. neglecting the free time evolution and concentrating on the interaction) gives

$$
\begin{equation*}
\hat{U}(t)\left|0_{1}, 0_{2}\right\rangle \approx\left|0_{1}, 0_{2}\right\rangle-i \kappa \alpha_{0} t\left|1_{1}, 1_{2}\right\rangle+\mathcal{O}\left(t^{2}\right) . \tag{25.13}
\end{equation*}
$$

That is, apart from a very high probability of doing nothing at all, parametric downconversion generates perfectly correlated (entangled) photon pairs that can be used for quantum information processing or metrology.

Generation of heralded single photons: The output of the parametric down-conversion consists mostly of vacuum and the probability of generating an entangled photon pair is small, $p=\left|\kappa \alpha_{0} t\right|^{2} \ll 1$. The probability of producing two photons in each mode is almost completely negligible. Moreover, signal and idler photons are completely correlated in frequency, wavevector, polarization and are produced in the same spatiotemporal mode. This
high degree of correlation makes them suitable for generating single photons in well-defined quantum states. We have previously seen that producing a photon via atomic decay is a probabilistic process with no success guarantee. In particular, spontaneous decay does not provide a trigger signal indicating that the photon has been released.

In parametric down-conversion such a trigger signal exists: the idler photon. The idea behind a 'heralded' single photon source is to make use of the quantum correlations in the output of parametric down-conversion. One places a photon detector in the path of the idler photon and waits until it has registered a photon. At the same moment one can be sure that the signal mode contains exactly one photon in the spatiotemporal mode selected by the detector. Such heralding schemes are widely used in quantum information processing


FIG. 34: Experimental scheme for the generation of six-photon entangled states for quantum information processing. After C.-Y. Lu, X.-Q. Zhou, O. Gühne, W.-B. Gao, J. Zhang, Z.-S. Yuan, A. Goebel, T. Yang, and J.-W. Pan, Nature Physics 3, 91 (2007).
with photons (see Fig. 34).

