On the feasibility of a nuclear exciton laser

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Nuclear excitons known from Mössbauer spectroscopy describe coherent excitations of a large number of nuclei – analogous to Dicke states (or Dicke super-radiance) in quantum optics. In this paper, we study the possibility of constructing a laser based on these coherent excitations. In contrast to the free electron laser (in its usual design), such a device would be based on stimulated emission and thus might offer certain advantages, e.g., regarding energy-momentum accuracy. Unfortunately, inserting realistic parameters, the window of operability is probably not open (yet) to present-day technology – but our design should be feasible in the UV regime, for example.

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I. INTRODUCTION

The invention of the laser lead to a giant leap in the field of classical and quantum optics. This light source offers unprecedented possibilities regarding features such as coherence, intensity, and brilliance etc. Unfortunately, however, it is not easy to transfer this successful concept beyond the optical or near-optical regime, cf. [1, 2]. Free-electron lasers, for example, work at much higher energies – but their principle of operation (in their usual design) is more similar to classical emission instead of stimulated emission. As a result, their properties (e.g., regarding coherence) are not quite comparable to optical lasers.

There is another phenomenon in this energy range $\mathcal{O}(\text{keV})$ in which coherence plays a crucial role – nuclear excitons known from Mössbauer spectroscopy [3, 4]. These coherent excitations of a large number of nuclei [5–7] are analogous to Dicke states [8] (also known as Dicke super-radiance [9–11]) in quantum optics. The coherence results in constructive interference of the emission amplitudes from many nuclei [12] and is facilitated by the fact that the photon recoil is absorbed by the whole lattice [3, 4] instead of the individual nuclei (which would destroy the coherence). For example, the coherent nature of the propagation of nuclear excitons through resonant media, showing quantum beats, was observed in [13, 14]. Other cooperative effects of coherently excited nuclei have been studied, such as the collective Lamb shift [15], coherent control of nuclear x-ray pumping [16], and electromagnetically induced transparency [17].

In the following, we study the possibility of constructing a laser-type device employing these nuclear excitons, which is based on stimulated emission [18]. Such a device could combine the advantages of the free-electron laser with the coherence and brilliance of nuclear excitons.

II. HAMILTONIAN

First, we describe a single nucleus as a two-level system with transition frequency $\omega$ interacting resonantly with a single-mode field. In rotating-wave and dipole approximation, the Hamiltonian can then be cast into the standard form ($\hbar = c = \varepsilon_0 = 1$)

$$
\hat{H}_{\text{single}} = (g\hat{a}\sigma^+_\ell + \text{H.c.}) + \frac{\omega}{2}(\hat{\sigma}_z^\ell + 1) + \omega\hat{a}^\dagger\hat{a}.
$$

As usual, the ladder operators $\sigma^\dagger_\ell = (\sigma^+_\ell \pm i\sigma^x_\ell)/2$ and the Pauli matrix $\sigma^\ell_z$ describe the two-level system. The first term governs the interaction (with coupling constant $g$) with the electromagnetic field and thus contains photonic annihilation and creation operators $\hat{a}/\hat{a}^\dagger$ and phase factors $e^{i\kappa \cdot \mathbf{r}_\ell}$ depending on the location of the nucleus, $\mathbf{r}_\ell$, and the wavenumber $\kappa = \mathbf{k}$ of the photon mode with $|\kappa| = \omega$. The second and third term account for the energy stored in the two-level nucleus and in the single-mode field, respectively. When dealing with many $S \gg 1$ two-level nuclei instead of one, we can sum up the individual-nucleus Hamiltonians and arrive at

$$
\hat{H} = (g\hat{a}\hat{\Sigma}^+ + \text{H.c.}) + \omega\left(\hat{\Sigma}^z + \frac{S}{2}\right) + \omega\hat{a}^\dagger\hat{a},
$$

where quasinpin-$S$-operators have been introduced

$$
\hat{\Sigma}^\pm = \sum_{\ell = 1}^S \sigma^\pm_\ell \exp\{\pm i\kappa \cdot \mathbf{r}_\ell\}, \quad \hat{\Sigma}^z = \frac{1}{2} \sum_{\ell = 1}^S \sigma^z_\ell.
$$

In the interaction picture, the perturbation Hamiltonian, originating from the first term in Eq. (2), reads

$$
\hat{V} = g\hat{a}\hat{\Sigma}^+ + \text{H.c.}.
$$

The quasinpin-$S$-operators $\hat{\Sigma}^\pm = \hat{\Sigma}^x \pm i\hat{\Sigma}^y$ and $\hat{\Sigma}^z$ generate an $SU(2)$ algebra [13]. Thus, the transition matrix elements for collective transitions not only depend on the

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number of nuclei involved, but also on the number of excitations \( s \)
\[
\hat{\Sigma}^+ |s\rangle = \sqrt{(S-s)(s+1)} |s+1\rangle,
\hat{\Sigma}^- |s\rangle = \sqrt{(S-s+1)s} |s-1\rangle,
\] (5)
where \( |s\rangle \propto (\hat{\Sigma}^+)^s |0\rangle \) denotes a coherent state with \( s \) excitons, often referred to as Dicke states \([8]\).

### III. COHERENT EMISSION

In contrast to the spontaneous decay of a single excited nucleus, where the resulting photon can be emitted in all directions, exciton states as in Eq. (4) predominantly emit photons in forward direction. Only in this case, all the phases \( e^{i\kappa \tau t} \) add up coherently (we assume random locations \( r_i \)), see Fig. 1. We will now investigate spontaneous and stimulated emission from an ensemble of \( S \) coherently excited nuclei in more detail.

![FIG. 1: Sketch of the coherent properties of nuclear excitons. An incident photon with wave-vector \( \kappa \) is absorbed (a) by an ensemble of \( S \gg 1 \) nuclei (two-level systems) and thus generates a Dicke state \( |s=1\rangle \). Then the decay amplitudes of all these nuclei add up coherently in forward direction such that the absorption is followed by collective spontaneous emission into the same direction (b).](image)

#### A. Spontaneous emission

We start with the case of collective spontaneous emission (a.k.a. Dicke super-radiance \([9,11]\)) from a coherent state \( |s\rangle \). First of all, as the \( S \) nuclei are not enclosed by a resonator or a cavity in our set-up, we have to consider all \( k \)-modes. Thus, the Hamiltonian (4) changes into
\[
\hat{V}_{sp}(\tau) = \int d^3k \ g_k \hat{\alpha}_k e^{-i(\omega_k - \omega)\tau} \hat{\Sigma}^+ (k) + \text{H.c.},
\] (6)
where \( \hat{\alpha}_k \) is the photonic annihilation operator for the mode \( k \) with frequency \( \omega_k \) and \( g_k \) the associated coupling strength. Note that we neglect polarization effects, i.e., we assume that the polarization vectors are directed along the same axis as the dipole moments of the absorbing nuclei. Furthermore, \( \hat{\Sigma}^+ (k) \) denotes the quasispin-\( S \)-operators with the wavenumber \( k \) instead of \( \kappa \). However, when \( k \) is not close to \( \kappa \), the phase factors of \( \hat{\Sigma}^\pm (k) \) and \( |s\rangle \) do not match, and the transition is not coherent, i.e., not enhanced by a factor \( S \) according to Eq. (5), and can thus be neglected. Note that this is the reason why collectively emitted photons are directed along (almost) the same axis as previously absorbed photons \([9,11]\), see also Fig. 1. For simplicity, the quasispin-\( S \)-operators are therefore approximated by introducing a cut-off function \( g(\kappa - k) \) that is only non-zero for small deviations \( \kappa - k \) of the supported direction \( \kappa \), i.e., \( \hat{\Sigma}^\pm (k) \approx g(\kappa - k) \hat{\Sigma}^\pm \). For large \( S \), we may approximate the quasispin-\( S \)-operators classically, i.e., \( \hat{\Sigma}^\pm \approx \sqrt{(S-1)S} |s\rangle \), and thus the effect of the Hamiltonian Eq. (6) acting on the vacuum state can be expressed by a coherent state
\[
\hat{U}_{sp}(t) |0\rangle \approx \exp \left( \int d^3k \ a_k a_k \hat{\Sigma}^+ - \text{H.c.} \right) |0\rangle,
\] (7)
with the amplitudes
\[
i a_k = g_k g(\kappa - k) \sqrt{(S-1)S} \int_0^t d\tau e^{i(\omega_k - \omega_\tau)\omega_\tau}.
\] (8)

The number of emitted photons per mode is given by \( |a_k|^2 \) and the total photon number grows linearly with \( t \)
\[
N_\gamma = \int d^3k |a_k|^2 \approx 2\pi^2 (S-1) s |g_k|^2 t \frac{1}{L^2},
\] (9)
where \( L^2 \) denotes the transversal cross-section area of the ensemble, which determines the transverse area in \( k \)-space where \( g(\kappa - k) \) is non-zero. In addition to this spatial resonance condition, the temporal resonance was incorporated via approximating the squared time-integral by \( t^2 \) for \( |\omega_k - \omega| < 1/t \) and zero otherwise.

Strictly speaking, this relation is only valid for a fixed number of excitations \( s \), i.e., the time-dependence of \( s(t) \) due to the emission of photons (energy conservation) is neglected. Assuming that this time-dependence \( s(t) \) is slow compared to \( \omega \) (i.e., that the coupling strength is small enough), we may take it into account approximately via defining the instantaneous spontaneous emission rate
\[
\Gamma_{sp}(t) = \frac{dN_\gamma}{dt} = \gamma [S - s(t) + 1] s(t),
\] (10)
with the abbreviation \( \gamma = 2\pi^2 |g_k|^2 / L^2 \). The change of \( s(t) \) in the time interval \( dt \) is then governed by \( \Gamma_{sp}(t) \)
\[
\frac{ds(t)}{dt} = -\Gamma_{sp}(t) = -\gamma [S - s(t) + 1] s(t).
\] (11)

For the initial condition \( s(0) = S/2 \) (see below), the solution for \( S \gg 1 \) is given by
\[
s(t) = \frac{S}{1 + e^{\gamma St}}.
\] (12)

This yields the intensity due to spontaneous emission
\[
I_{sp}(t) = -\frac{dE}{dt} L^2 = -\frac{ds(t)}{dt} \frac{\omega}{L^2} = \frac{1}{4} \gamma S^2 \text{sech}^2 \left( \frac{S t}{2} \right) \frac{\omega}{L^2}.
\] (13)
where \( L_s^2 \) is the cross-section area of the emitted beam, see also \[20\]. The time-dependence in the sech-function can be used to define an effective time constant via

\[
\tau_{\text{sp}} = \frac{4}{\gamma S}, \tag{14}
\]

after which \( I_{\text{sp}}(t) \) has dropped to 7% of its initial value. Let us now briefly compare this time-scale \( \tau_{\text{sp}} \) for the coherent spontaneous emission process with the time-scale in the incoherent case. For incoherent emission from \( s \) excited nuclei, we can regard each nucleus independently. According to standard Weisskopf-Wigner theory \[21\], the life-time of an excited nucleus is given by

\[
\tau_{\text{single}} = \frac{1}{\Gamma_{\text{single}}} = \frac{1}{8\pi^2} \frac{1}{|\gamma_k|^2 \omega^2} . \tag{15}
\]

Comparing the two time-scales Eq. (14) and (15)

\[
\frac{\tau_{\text{sp}}}{\tau_{\text{single}}} = 64\pi^2 L_s^2 \frac{1}{\lambda^2} \frac{1}{S} , \tag{16}
\]

we find that for large \( S \), the coherent spontaneous emission process is much faster than the incoherent process (see also \[22\]). Taking for example \( S = 10^{10} \) \( ^{57}\)Fe-nuclei with resonance at \( \Delta E_o = 14.4 \text{ keV} \), lifetime \( \tau_{\text{single}} = 141 \text{ ns} \) and \( L_s = 0.1 \mu \text{m} \), the quotient evaluates to \( \tau_{\text{sp}}/\tau_{\text{single}} \approx 0.09 \), i.e., the coherent emission runs over ten times faster than the incoherent emission. Note that there are also competing processes, such as decay via electron conversion – but they are incoherent and thus can be suppressed for large \( S \), i.e., small \( \tau_{\text{sp}} \).

### B. Stimulated emission

In order to study stimulated emission from a coherently excited \( S \)-nuclei ensemble, we regard the incoming field \( A_in(t) = \sqrt{I_in(t)/\omega} \) classically. That is, we use the Hamiltonian Eq. (11), but replace \( \hat{g} \) by \( \hat{g} A_in(t) \). For simplicity, we assume the transition matrix element \( \hat{g} \) of the nucleus to be real

\[
\hat{V}_{st} = \hat{g} A_in(t) \left( \hat{\Sigma}^+ + \hat{\Sigma}^- \right) = 2\hat{g} A_in(t) \hat{\Sigma}^z . \tag{17}
\]

Applying Heisenberg picture and employing the properties of the \( SU(2) \)-algebra yields

\[
\hat{U}_{st}(t) \hat{\Sigma}^z \hat{U}_{st}(t) = \cos \left( 2\hat{g} \int_0^t d\tau \; A_in(\tau) \right) \hat{\Sigma}^z + \sin \left( 2\hat{g} \int_0^t d\tau \; A_in(\tau) \right) \hat{\Sigma}^y . \tag{18}
\]

As envisaged for laser application (see below), we choose \( \langle 0 | \hat{\Sigma}^+ | 0 \rangle = + S/2 \) \( | S \rangle \) and thus the energy stored in the \( S \) nuclei at time \( t \) is

\[
E(t) = \frac{S \omega}{2} \left[ \cos \left( 2\hat{g} \int_0^t d\tau \; A_in(\tau) \right) + 1 \right] . \tag{19}
\]

This yields the emitted intensity \( I_{st}(t) \) stimulated by the incoming intensity \( I_{in}(t) \)

\[
I_{st}(t) = \frac{\hat{g} S}{L_s^2} \sin \left( 2\hat{g} \int_0^t d\tau \; \sqrt{I_{in}(\tau)} \right) \sqrt{I_{in}(t)} , \tag{20}
\]

where we have assumed that both beams have the same cross-section area \( L_s^2 \).

We define the time-scale of the stimulated emission as the time \( \tau_{st} \), after which all the energy initially stored in the \( S \) nuclei has been emitted, i.e.,

\[
\int_0^{\tau_{st}} d\tau \; \sqrt{I_{in}(\tau)} = \frac{\pi \omega}{2\hat{g}} . \tag{21}
\]

Now let us imagine that we have two separate ensembles (e.g., foils) of coherently excited nuclei, such that the first foil spontaneously emits the intensity \( I_{in}(t) \) as in Eq. (18) which causes stimulated emission according to Eq. (20) in the second foil. In this case, we can insert \( I_{in}(t) = I_{sp}(t) \), and Eq. (21) can be solved for \( \tau_{st} \)

\[
\tau_{st} = \frac{4}{\gamma S} \text{ArTanh} \left[ \tan \left( \frac{1}{8} \sqrt{\frac{\pi}{2}} \right) \right] \approx 0.16 \times \tau_{sp} . \tag{22}
\]

Since both foils contain the same nuclei (with the same coupling strengths), the time-scale for the stimulated emission of the second foil, \( \tau_{st} \), is completely determined by the time-scale of the spontaneous emission process of the first foil, \( \tau_{sp} \).

### IV. Pumping

After having discussed coherent spontaneous emission as well as coherent stimulated emission, let us investigate the pumping process for a single foil of \( S \) nuclei, which are initially in the state \( s = 0 \), i.e., \( \langle 0 | \hat{\Sigma}^+ | 0 \rangle = - S/2 \).

Note that it is very easy to over- or under-estimate the efficiency of the pumping process by using too simplified pictures. On the one hand, one might expect that the number of excitons in the foil grows linearly with the number of photons incident and thus linearly with the interaction time \( t \). However, this is only true for pumping with incoherent light (for further details, see the appendix), but not for coherent pumping, which is the case considered here. On the other hand, since the transition matrix elements in Eq. (5) scale with \( \sqrt{s} \) and thus the effective line-width increases with \( s \), one might expect a behavior like \( s \propto s \), which would imply an exponential growth \( s(t) \propto e^{\tau t} \), at least for small \( s \ll S \). This picture is also wrong, since – in view of the unitarity of the time-evolution – not just the absorption rate but also the emission rate increase with \( s \). Thus, the correct answer is that \( s(t) \) grows quadratically \( s(t) \propto t^2 \) for small \( s \), i.e., somewhere in between linear and exponential.

To show this, let us consider pumping with one coherent pump-pulse \( A_{\text{pump}}(t) \) for the whole interaction time. We can employ the Hamiltonian Eq. (17) again with the
sole difference that the incoming field $A_{\text{in}}(t)$ is now given by the pump-field $A_{\text{pump}}(t)$. Thus, Eq. (18) again holds, and the exciton number is given by

$$s(t) = \frac{S}{2} \left[ 1 - \cos \left( 2\tilde{g} \int_0^t d\tau A_{\text{pump}}(\tau) \right) \right]$$

$$= S\tilde{g}^2 \left( \int_0^t d\tau A_{\text{pump}}(\tau) \right)^2 + O(\tilde{g}^4 t^4), \quad (23)$$

i.e., the exciton number grows quadratically for small $t$ (for an alternative approach, see the appendix). We moreover find that a full cycle (i.e., a sign flip of $\Sigma^z \rightarrow -\Sigma^z$) occurs after the pump time $\tau_{\text{pump}}$ where

$$\int_0^{\tau_{\text{pump}}} d\tau A_{\text{pump}}(\tau) = \frac{\pi}{2\tilde{g}}. \quad (24)$$

The simplest example would be a constant pump pulse $A_{\text{pump}} = A_0$ with $\tau_{\text{pump}} = \pi/(2\tilde{g}A_0)$. In order to see if such a pump-field is feasible in general, we calculate a rough estimate for the required intensity of the pump-field. For simplicity, we assume that the intensity is constant over the pulse, i.e., $A_{\text{pump}}(t) = \sqrt{\text{pump}}/\omega$. Then we find $I_{\text{pump}} = \pi\gamma^2/(4\tilde{g}^2\tau_{\text{pump}}^2)$. Now $\tilde{g}$ can be expressed in terms of the (single-nucleus) decay rate $\Gamma_{\text{single}}$ and the frequency $\omega$ of the considered nuclear excitation (see the appendix).

Moreover, if we replace the coherent pulse-length $\tau_{\text{pump}} = \Omega\lambda = 2\pi\Omega/\omega$ by the number $\Omega$ of (coherent) wave-cycles, we find

$$I_{\text{pump}} = \frac{1}{32\pi\Omega^2} \frac{\omega^5}{\Gamma_{\text{single}}}. \quad (25)$$

Thus, nuclear resonances with low energies $\omega$ but high decay-rates $\Gamma_{\text{single}}$ require low pump intensities. Concrete examples will be discussed at the end of this article.

V. LASER

Now we have gathered all the tools required to understand the set-up of the proposed nuclear exciton laser. The envisaged set-up consists of a series of $N \gg 1$ foils $n = 1, 2, \ldots, N$, each foil containing $S_n$ nuclei (two-level systems) with a nuclear resonance at frequency $\omega$. At the beginning, we assume that all $n = 1, 2, \ldots, N$ foils are in the ground state, corresponding to a quasi-spin $\Sigma_n^z = -S_n/2$, see Fig. 2(a).

To prepare the emission of a laser pulse, the foils need to be pumped to suitable coherent states. Let us distinguish between the first foil and all later foils $n = 2, \ldots, N$. While the latter all should be pumped to the maximum of $\Sigma_n^z = S_n/2$, i.e., $s_n = S_n$, the first foil should only be pumped such that half of the nuclei are in the excited state, i.e., $\Sigma_1^z = 0$ and $s_1 = S_1/2$. For simplicity, we envisage the whole pumping process to be achieved by only one coherent pump-pulse, which goes through all the foils one after another and is only weakly changed by absorption. The pump-pulse should satisfy Eq. (24) in order to rotate the quasispin $\Sigma_1^z$ of each foil from $\Sigma_n^z = -S_n/2$ to $\Sigma_n^z = +S_n/2$. Additional measures need to be taken to ensure that the first foil is only pumped to $s_1 = S_1/2$. One option could be to have a different kind of nuclei in the first foil, which have the same resonance frequency as those in the other foils, while the coupling strengths differ by a factor of two (approximately). Another option could be to switch the first foil (mechanically or magnetically) during the pumping process.

When the set-up is prepared as shown in Fig. 2(b), the emission process automatically starts, as the first foil immediately begins with the spontaneous emission discussed above, Eq. (13). The idea is that, due to the “half-filled" coherent state $\Sigma_1^z = 0$, the emission process of the first foil happens much faster than the spontaneous emission of the subsequent foils. Taking, e.g., the second foil ($s_2 = S_2$), the time-scale for the emission of a single photon would be $1/\Gamma_{\text{sp}} = 1/(\gamma S_2)$. For the first foil, the time-scale for the whole emission process (of nearly all photons, not only one) is given by $\tau_{\text{sp}} = 4/(\gamma S_1)$. So by choosing $S_1 \gg S_2$, e.g., by making the first foil ten times thicker than the subsequent foils, it is assured that the second foil is still in the state $\Sigma_n^z = S_n/2$, when the intensity emitted from the first foil is incident.

Stimulated emission then occurs at the second foil according to Eq. (20) and the second foil has emitted all its energy after $\tau_{\text{st}} \approx 0.16 \times \tau_{\text{sp}}$, i.e., before the stimulating pulse coming from the first foil declines. After the second foil, the overall intensity thus adds up to $I_{\text{total}}^{(2)}(t) = I_{\text{sp}}(t) + I_{\text{st}}^{(2)}(t)$. This overall intensity then causes stimulated emission at the third foil, resulting in an even bigger intensity $I_{\text{total}}^{(3)}(t) = I_{\text{total}}^{(2)}(t) + I_{\text{st}}^{(3)}(t)$, etc. In this way, the intensity of the light pulse grows stepwise with each passed foil.

FIG. 2: Sketch of the operation sequence of the proposed nuclear exciton laser. Initially, all foils (here $N = 3$) are in the ground state $\Sigma_n^z = -S_n/2$ (a). The pump-pulse then rotates the quasispin of the first foil to $\Sigma_1^z = 0$ and the quasispin of all subsequent foils to $\Sigma_n^z = +S_n/2$ (b). Then, the “half-filled” first foil spontaneously emits a pulse $I_{\text{sp}}(t)$, which stimulates emission at foils 2 and 3, leading to an enhanced overall intensity $I_{\text{total}}^{(3)}(t)$ (c).
Numerical analysis has been done for the case of \( N = 50 \) foils. Iteratively, \( I_{\text{total}}^{(n)}(t) \) was calculated from \( I_{\text{total}}^{(n-1)}(t) \), where \( I_{\text{total}}^{(n)}(t) = I_{\text{total}}^{(n-1)}(t) + I_{\text{st}}^{(n)}(t) \), starting with \( I_{\text{total}}^{(1)}(t) = I_{sp}(t) \). It was assumed that the first foil consists of \( S_1 = 10^{10} \) \(^{57}\)Fe-nuclei (with \( \Delta E_{\gamma} = 14.4 \) keV and \( \tau = 141 \) ns) while all other foils are ten times thinner, i.e., \( S_n = 10^9 \). Transversal dimensions of the foils and the laser beam are chosen as \( L_\perp^2 = (0.1 \, \mu \text{m})^2 \).

Note that the useful part of the laser pulse \( I_{\text{total}}^{(n)}(t) \) is determined by the time after which the last foil has emitted all its excitations, \( \tau_{\text{st}}^{(n)} \), because afterwards re-absorption takes place. This time \( \tau_{\text{st}}^{(n)} \) becomes shorter with rising \( n \), as the intensity which causes the stimulated emission grows with \( n \). As a result, the average intensity of the useful part of the laser pulse,

\[
\bar{I}_{\text{total}}^{(n)} = \frac{1}{\tau_{\text{st}}^{(n)}} \int_0^{\tau_{\text{st}}^{(n)}} d\tau I_{\text{total}}^{(n)}(\tau),
\]

increases with a power law. In the concrete example given above, \( I_{\text{total}}^{(n)} \) grows roughly \( \propto n^{3/2} \), see Fig. 3.

FIG. 3: Average intensity \( \bar{I}_{\text{total}}^{(n)} \) over number of foil \( n \) from numerical analysis. In this example, \( I_{\text{total}}^{(n)} \) roughly increases as a power law \( \propto n^{3/2} \).

VI. CONCLUSIONS

In summary, we described a proposal for a laser in the \( \mathcal{O}(\text{keV}) \) regime which is based on stimulated emission and works with nuclear excitons. The pumping could be achieved with a free-electron laser, for example. Note that the pump pulse \( A_{\text{pump}} \) and the generated laser pulse \( A_{\text{laser}} \) both correspond to a 180°-rotation of the last foil according to Eq. (18) and thus are related via

\[
\int_0^{\tau_{\text{pump}}} d\tau A_{\text{pump}}(\tau) = \int_0^{\tau_{\text{st}}^{(N)}} d\tau A_{\text{laser}}(\tau) = \frac{\pi}{2\tilde{g}}.
\]

However, the intensity of the pump pulse \( \propto |A_{pump}^2| \) is much larger than that of the laser pulse \( \propto |A_{laser}^2| \). On the other hand, the duration \( \tau_{\text{st}}^{(N)} \) of the laser pulse is much larger and thus its frequency accuracy is much higher (see also [28] for a different approach). This could be important for spectroscopy etc.

Let us discuss some example data for the required intensity of the pump-pulse. First, we consider \(^{57}\)Fe-nuclei with a resonance at \( \Delta E_{\gamma} = 14.4 \) keV with a mean lifetime of \( \tau = 141 \) ns. If we assume that the pump pulse consists of \( \mathcal{R} = 10^6 \) coherent wave-trains, we would need a pump intensity of \( I_{\text{pump}} \approx 8.3 \cdot 10^{20} \text{ W/cm}^2 \) according to Eq. (25). (Comparable or even higher intensities have already been considered in e.g. [29] [31].) This is probably beyond the capabilities of present free-electron lasers, see, e.g., [32]. However, future light sources such as seeded free-electron lasers should achieve improved coherence times and higher intensities (especially after focusing with X-ray lenses).

On the other hand, when considering other nuclear resonances beyond the well-known \(^{57}\)Fe-example, we find that the requirements are somewhat easier to fulfill. For example, considering the \(^{201}\)Hg resonance at \( \Delta E_{\gamma} = 1.6 \) keV with \( \tau = 81 \) ns and again assuming \( \mathcal{R} = 10^6 \) coherent wave-trains, we would “only” need a pump intensity of \( I_{\text{pump}} \approx 8.0 \cdot 10^{15} \text{ W/cm}^2 \) according to Eq. (25).

Unfortunately, this intensity is probably still too large: After inserting typical values for the absorption cross section of 1.6 keV-photons in metals (or other solid materials), we find that the pump beam deposits enough energy in the foil to evaporate it. Even though the thermalization dynamics following the illumination with such a \( 8.0 \cdot 10^{15} \text{ W/cm}^2 \)-beam of 1.6 keV-photons is not well studied yet, one would expect that the foil starts to disintegrate after a few pico-seconds [33] and hence does not survive long enough for our purposes.

In summary, the major difficulty of our set-up is that it requires extremely large pump intensities. As we may infer from Eq. (23), the pump intensity scales with the fifth power of the transition energy \( \omega \). Thus, our scheme should be much easier to realize at lower energies. As one possible example, let us envisage a UV-laser. In this case, the nuclear transitions could be replaced by suitable electronic transitions in atoms or molecules. The pumping could be achieved either directly via a free-electron laser in the low-energy regime or indirectly via a two-photon transition generated by two optical lasers, for example.

Let us discuss the latter case using a three-level system as depicted in Fig. 4. Assuming two pump-lasers with optical frequencies \( \omega_1 \) and \( \omega_2 \), the laser could operate in the ultra-violet regime \( \omega_{\text{uv}} \). In this case, the expression \( \tilde{g}A_{\text{pump}} \) for one-photon pumping in Eq. (24) should be replaced by \( \tilde{g}_{34}A_{\text{pump}}^2/\Delta \). Note that the coupling constant \( \tilde{g}_{34} \) of the “dipole-forbidden” 2s-3d transition is typically much smaller than \( \tilde{g}_{33} \). Assuming typical values, such as a dipole coupling length of three Bohr radii, we would need pump-laser intensities of about \( I_{\text{pump}} = \mathcal{O}(10^{10} \text{ W/cm}^2) \) over a length
of $\mathcal{W} = 10^4$ coherent wave-trains with a detuning of $\Delta = \mathcal{O}(10^{13} \text{ Hz})$ in order to prevent unwanted excitations of the middle 3d level. The condition for dominant coherent emission, $\tau_{\text{pump}}/\tau_{\text{single}} \ll 1$, can be fulfilled for $S_1/L_2^2 = \mathcal{O}(10^5 \mu \text{m}^{-2})$, which is quite reasonable. In this scenario, the duration of the laser pulse $\tau_{\text{pump}}$ is comparable to the length of the pump pulse $\tau_{\text{pump}} \approx \tau_{\text{st}} = \mathcal{O}(10^5 \text{ W/cm}^2)$, depending on the number of foils.

Again, the main idea would be that the coherent emission is strongly enhanced for $S \gg 1$ in comparison to competing non-coherent decay channels. To this end, the two pump lasers must be parallel to ensure the spatial phase matching.

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**Appendix**

**A. Coherent versus incoherent pumping**

Let us review the pumping process by applying the Holstein-Primakoff [34] transformation

$$\hat{\Sigma}^+ = \hat{b}^\dagger \sqrt{S - \hat{b}^\dagger \hat{b}} = (\hat{\Sigma}^{-})^\dagger, \quad \hat{\Sigma}^z = \hat{b}^\dagger \hat{b} - S/2,$$

(28)

to the Hamiltonian Eq. [34] and considering the limit $S \gg s$, i.e., the beginning of the pumping process

$$\hat{V}_{st} \approx \tilde{g} A_{\text{pump}}(t) \left( \sqrt{S} \hat{b}^\dagger + \text{H.c.} \right).$$

(29)

We first analyze the case of coherent pumping, that is pumping with a coherent pulse $A_{\text{pump}}(t)$, i.e., the same time-evolution operator for the whole pumping process

$$\hat{U}_{st}(t) = \exp \left( \beta(t) \hat{b}^\dagger - \text{H.c.} \right).$$

(30)

A time-dependent coherent state of excitons is created

$$\beta(t) = -i \tilde{g} \sqrt{S} \int_0^t d\tau A_{\text{pump}}(\tau),$$

(31)

whose exciton number grows quadratically with time $t$

$$n(t) = |\beta(t)|^2 = \mathcal{O}(\tilde{g}^2 S A_{\text{pump}}^2 t^2).$$

(32)

An incoherent pump-pulse, in contrast, can be approximated as a succession of many uncorrelated coherent pulses $A_{\text{pump}}^{(i)}(t)$ incident on the target. The time-evolution operator then is a product of many coherent displacement operators

$$\hat{U}_{\text{eff}} \approx \prod_i \hat{U}_{st}^{(i)} = \exp \left( \sum_i \beta_i \hat{b}^\dagger - \text{H.c.} \right).$$

(33)

For uncorrelated pulses, the $\beta_i$ have random phases, such that the sum corresponds to a random walk

$$\beta_{\text{eff}}(j) = \sum_{i=1}^j \beta_i \propto \sqrt{j} \propto \sqrt{t},$$

(34)

such that the exciton number $n = |\beta_{\text{eff}}|^2$ grows merely linearly with time in this case.

**B. Expressing $\tilde{g}$ in terms of $\Gamma_{\text{single}}$ and $\omega$**

The coupling constant $|g_k|$ can be expressed via the decay rate $\Gamma_{\text{single}}$ and frequency $\omega$

$$\Gamma_{\text{single}} = 2\pi \int d^3k |g_k|^2 \delta(\omega_k - \omega) \approx 8\pi^2 |g_k|^2 \omega^2.$$  

(35)

The dimensionless coupling constant $\tilde{g}$ can be obtained via $\tilde{g} = |g_k|\sqrt{2(2\pi)^3} \omega$ since our Hamiltonian contains the classical field $A_{\text{pump}}(t)$. As a result we arrive at

$$\tilde{g} = \sqrt{\frac{2\pi \Gamma_{\text{single}}}{\omega}}.$$  

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