Optimized geometries for future generation optical lattice clocks

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Abstract – Atoms deeply trapped in magic wavelength optical lattices provide a Doppler- and collision-free dense ensemble of quantum emitters ideal for high-precision spectroscopy and they are the basis of some of the best optical atomic clocks to date. However, despite their minute optical dipole moments the inherent long-range dipole-dipole interactions in such lattices still generate line shifts, dephasing and modified decay. We show that in a perfectly filled lattice line shifts and decay are resonantly enhanced depending on the lattice constant and geometry. Potentially, this yields clock shifts of many atomic linewidths and reduces the measurement by optimizing the lattice geometry. Such collective effects can be tailored to yield zero effective shifts and prolong dipole lifetimes beyond the single-atom decay. In particular, we identify dense 2D hexagonal or square lattices as the most promising configurations for an accuracy and precision well below the independent ensemble limit. This geometry should also be an ideal basis for related applications such as superradiant lasers, precision magnetometry or long-lived quantum memories.

Since the turn of the century, the technology of manipulating and controlling ultracold atoms with lasers has seen breathtaking advances [1–3]. Following the seminal demonstration of a quantum phase transition in an optical lattice [4], nowadays the so-called Mott insulator state with exactly one atom per site can now be prepared routinely [5,6]. Experiments with photo-associated ultracold molecules have reached a comparable control [7–10]. Coherent interactions between the atoms at neighboring sites can be tailored [11] and by using large and sufficiently dense ensembles one overcomes the weak single-atom field coupling in free space [12].

For some of the world’s best optical clocks [13–15], atoms with a long-lived transition are prepared in an optical lattice using a differential light shift free (magic) trapping wavelength [16,17]. In principle, this provides for a Doppler-free and, for 3D confinement, also a collision-free cold and dense ensemble with negligible inhomogeneous broadening, eliminating major clock accuracy limitations. However, when excited optically, emitters are still not completely independent as they will inevitably interact via a long-range resonant dipole-dipole coupling [18].

At sufficient densities, i.e., small lattice constants, this dipole-dipole interaction strength surpasses the excited-state lifetime and collective excitations analogous to excitons appear [19]. For distances much smaller than the wavelength as for polar molecules in optical lattices they dominate the dynamics [20] and allow for studying generic phenomena of solid-state physics [1]. For clock transitions the extremely tiny dipole moment keeps these interactions small. However, the excitons effective transition frequencies and their spontaneous decay rate are still dominated by the dipole-dipole interaction [21] substantially deviating from the bare atom case. Although not fully reached in the current setups, this constitutes a fundamental limit for the accuracy and precision of the corresponding clock setups.

In an idealized Ramsey sequence for a clock setup, the first laser pulse creates a product state of all atoms prepared in a 50% superposition of ground and excited state with equal phase and all dipoles aligned in parallel. This state features the maximally possible dipole moment and typically exhibits super-radiance. Even a tiny single-particle spontaneous emission rate thus can be that strongly enhanced, so that collective decay becomes the dominant factor limiting measurement time and precision [22]. In the current setups based on 1D lattices with low filling, this perturbation is often negligible compared to other noise like collisions, transverse motion, black-body shifts or reference cavity fluctuations. However, in lattices with 3D confinement and unit filling, these dipole-dipole interaction shifts are much larger than the atomic
linewidth and represent a significant inherent perturbation. Note that their absolute magnitude scales with the atomic dipole moment and, thus, strongly depends on the chosen transition lifetime, which can change by almost 6 orders of magnitude from a calcium to a strontium clock. This, however, is very different from the single-excitation case, which has been investigated in recent experiments in Boulder [23].

In this work we quantitatively study such collective effects on the Ramsey spectroscopy in perfect optical lattice configurations. As a key quantity to capturing the collective modifications of the system dynamics, we use the decay and phase shift of the collective dipole generated by the first Ramsey pulse, which determines the contrast and shift of the central Ramsey fringes. Note, that due to the pairwise nature of dipole-dipole interactions, a simple rephasing pulse cannot correct these errors. We ignore interaction-induced perturbations during the Ramsey pulses, which introduce extra noise but could be reduced by very fast pulses or improved sequences [24]. In practice we set out to numerically solve the well-established master equation for the atomic density matrix \[ \rho \dot{=} [\rho, H] + \mathcal{L}[\rho]. \] (1)

As previously shown for small atom numbers \( N < 12 \) a numerical solution of the full master equation yields non-negligible shifts already [21,28]. Unfortunately, as the Hilbert space grows exponentially with the atom number, the full equation cannot be solved for ensembles of a realistic size. Since for precision measurements we need to evaluate collective effects precisely, reliable and in a converging manner alternative numerical methods are required. For larger ensembles at low densities a cluster approach has produced first estimates of the scaling of the dephasing with the system’s size and density [13]. In the opposite limit of a very high density, important self-synchronization effects through dipole coupling were studied recently using strongly simplifying assumptions for the coupling [29]. Synchronization via spin-spin coupling can also occur via collisions at high density [30] or within an optical resonator [31].

In this letter we present an extensive numerical analysis of the collective dynamics for fully populated lattices of different geometries and sizes containing a large or even an infinite number of particles. Our primary goal is to estimate the magnitude of the dipole phase shift and collective decay as a function of lattice and excitation geometry. Most interestingly, besides a resonant enhancement of shifts, decay and dephasing at certain lattice spacings, we find cases in which collective interactions even lead to improvements of the maximally achievable measurement precision beyond the independent particle level by virtue of subradiant states. Our considerations are based on an idealized setup ignoring lattice shifts, thermal effects or the hopping of atoms.

Numerically we apply an enhanced mean-field approach related to cumulant expansion methods. It was developed in a recent paper, where we have also checked its validity extensively [32]. Using this method we can scale up the ensemble towards experimentally relevant atom numbers of up to \( N \approx 10^5 \) particles. If the particle distribution exhibits symmetries, numbers up to even \( 10^{10} \) are possible, well approximating infinite systems in 1D and 2D. The accuracy of the approach, however, breaks down at very close distances, as it cannot correctly capture high-order correlations. Similar deliberations for classical dipoles have recently been put forward [33].

**Model.** – We consider an ensemble of \( N \) identical effective two-level atoms with transition frequency \( \omega_0 \) and inverse lifetime \( \gamma \) at positions \( r_i (i = 1, \ldots, N) \) interacting via optical dipole-dipole coupling described by the Hamiltonian [18,26]

\[
H = \sum_{i,j \neq j} \frac{G(r_{ij})}{|r_i - r_j|^3} \sigma^+_i \sigma^-_j.
\] (2)

Here, \( \sigma^\pm_i \) denotes the raising (lowering) operator of the \( i \)-th atom and \( \Omega_{ij} = \frac{G(r_{ij})}{|r_i - r_j|^3} \) represents the energy exchange with \( k_0 = \frac{\omega_0}{c} = 2\pi/\lambda_0 \) and \( r_{ij} = |r_i - r_j| \) being the distance between atoms \( i \) and \( j \). Collective spontaneous emission is accounted for by a Liouvillian of the form [18,25]

\[
\mathcal{L}[\rho] = \frac{1}{2} \sum_{i,j} \Gamma_{ij}(r_{ij})(2\sigma^+_i \sigma^-_j - \sigma^+_i \sigma^-_i - \sigma^+_j \sigma^-_j - \rho),
\] (3)

where the off-diagonal rates \( \Gamma_{ij} = \frac{2}{\omega_0}G(k_0r_{ij}) \) introduce super- and subradiant decay [26] and \( \Gamma_{ii} = \gamma \). Explicitly we have

\[
\begin{align*}
F(\xi) &= \alpha \sin \xi \xi + \beta \left( \cos \xi \xi^2 - \sin \xi \xi^2 \right), \quad (4a) \\
G(\xi) &= -\alpha \cos \xi \xi + \beta \left( \sin \xi \xi^2 + \cos \xi \xi^2 \right) \quad (4b)
\end{align*}
\]

with \( \alpha = 1 - \cos^2 \theta \) and \( \beta = 1 - 3 \cos^2 \theta \), where \( \theta \) represents the angle between the line connecting atoms \( i \) and \( j \) and the common atomic dipole orientation.

**Mean-field approximation.** – To study large particle numbers we derive the equations of motion for the expectation values of the Pauli operators for the \( k \)-th atom as detailed in the supplementary material in [34]. Assuming a separable density operator and factorizing the two-particle correlations via \( \langle \sigma^\mu_i \sigma^\nu_j \rangle \approx \langle \sigma^\mu_i \rangle \langle \sigma^\nu_j \rangle \) for \( \mu, \nu \in \{x, y, z\} \) they transform to a closed set. As shown previously [32], these equations still capture the major part of the interaction up to a moderate interaction strength. To obtain even more accurate results one can add second-order pair correlation corrections. As we have shown in some earlier work, these corrections significantly increase the precision of the results at increased computational effort [32] but do not induce qualitative changes.
Fig. 1: (Color online) Scheme of a 2D optical lattice filled with clock atoms interacting via dipole-dipole energy exchange $\Omega_{ij}$ and a collectively modified spontaneous emission $\Gamma_{ij}$ at two different lattice constants shown in blue and yellow. In a mean-field treatment with translation invariance the sum over all interaction terms yields two effective couplings $\Omega^{\text{eff}}$ and $\Gamma^{\text{eff}}$ only, which govern the approximate system dynamics.

**Symmetric configurations.** – For symmetric geometries with each atom initially in the same state and subject to the same effective interactions, the equations of motion for all particles become identical and read

\[
\langle \sigma^x \rangle = \Omega^{\text{eff}} \langle \sigma^y \rangle \langle \sigma^z \rangle - \frac{1}{2} \left( \gamma - \Gamma^{\text{eff}} \langle \sigma^z \rangle \right) \langle \sigma^x \rangle, \tag{5a}
\]

\[
\langle \sigma^y \rangle = -\Omega^{\text{eff}} \langle \sigma^y \rangle \langle \sigma^z \rangle - \frac{1}{2} \left( \gamma - \Gamma^{\text{eff}} \langle \sigma^z \rangle \right) \langle \sigma^y \rangle, \tag{5b}
\]

\[
\langle \sigma^z \rangle = -\gamma \left(1 + \langle \sigma^z \rangle \right) - \frac{1}{2} \Gamma^{\text{eff}} \left( \langle \sigma^x \rangle^2 + \langle \sigma^y \rangle^2 \right). \tag{5c}
\]

Hence, instead of solving a huge set of coupled nonlinear equations, we need to determine the effective couplings, i.e.

\[
\Omega^{\text{eff}} = \sum_{j=2}^{N} \Omega_{1j}, \quad \Gamma^{\text{eff}} = \sum_{j=2}^{N} \Gamma_{1j}, \tag{6}
\]

Of course, such a rigorous symmetry condition is fulfilled for very few atomic distributions only. In these cases, however, the essence of the interactions within the entire lattice is captured solely by two real numbers, the effective coupling $\Omega^{\text{eff}}$ and the collective decay rate $\Gamma^{\text{eff}}$. In a clock setup one seeks to minimize the energy shifts $\Omega^{\text{eff}}$ and find configurations with a maximally negative $\Gamma^{\text{eff}}$, minimizing decay and allowing for an as long as possible interrogation time (see fig. 1).

**Finite systems.** – Firstly, for finite symmetric configurations the effective quantities can be calculated easily. The most obvious symmetric structures are regular polygons. This might not be the most practical example but nicely displays the underlying physics [35]. In fig. 2 we compare the parameters for a square, a ten-sided and a 100000-sided polygon. The square shows a behavior quite similar to the underlying functions $F(\xi)$ and $G(\xi)$, while the two larger polygons exhibit strong size-dependent variations, particularly at integral values of $d/\lambda_0$ emerging from the accumulation of many $1/\xi$ contributions. Note that even with a relatively large atom spacing, cooperative collective effects are sizable and vary strongly with distance.

**Infinite systems.** – In practice, extended regular systems, i.e. large periodic lattices, are experimentally more relevant. Figure 3 depicts the effective couplings for an infinite chain, a square lattice and a hexagonal lattice. For comparison, we have overlaid the results for smaller atom numbers to demonstrate finite-size effects, where even unphysical values of $\Gamma^{\text{eff}} < -1$ can appear. We observe stronger variations and again divergences at integral values of $d/\lambda_0$. These manifest themselves in a much more pronounced way at huge atom numbers and, therefore, underpin the importance of properly treating long-range interactions.

Note that for the two-dimensional square lattice and the hexagonal lattice, $\Gamma^{\text{eff}}$ exhibits a broad minimum for the effective decay close to $\Gamma^{\text{eff}} = -1$ for $d < \lambda_0$, where the atomic decay is strongly inhibited. This favors such two-dimensional setups for lattice clocks as subradiant decay will dominate the system dynamics allowing for much longer Ramsey delay times and thus offering a higher overall precision [22]. Similarly we can identify lattice constants with a zero effective shift and, therefore, increased clock accuracy. Extending these calculations to three-dimensional lattices, we find that the necessary atom numbers to obtain smooth converging behavior are beyond our current numerical capabilities. For particle numbers of about $10^{12}$ the resulting effective quantities still fluctuate strongly, predicting potential problems for such 3D clock setups. A demonstration of this effect can be found in the supplementary material in [34].

**Tailoring atomic excitations.** – So far we have assumed a phase-symmetric excitation of all atoms by the first Ramsey pulse. In a practical excitation scheme this corresponds to illumination at right angle. In general, however, the effective couplings $\Omega^{\text{eff}}$ and $\Gamma^{\text{eff}}$ will change, when we allow for a local phase shift imprinted on the atoms. In a $\pi/2$ Ramsey sequence [36] the excitation phase appears on the excited state directly, i.e.,

\[
|\Psi\rangle = \bigotimes_{j=1}^{N} \frac{1}{\sqrt{2}} \left( |g\rangle + e^{i\Delta \phi (j-1)} |e\rangle \right), \tag{7}
\]
In our treatment we can exploit the system’s symmetry and absorb this phase into the effective couplings \([34]\). For \(\Delta \phi = 0\) we recover the above results. The closer the phase shift gets to \(\Delta \phi = \pi\), however, the more half-integral values of \(d/\lambda_0\) yield minimal shifts and the maximally negative \(\Gamma_{\text{eff}}\) as seen in fig. 4. Since the emitted light has interfered constructively at integral and destructively at half-integral distances for \(\Delta \phi = 0\), it will do exactly the opposite at \(\Delta \phi = \pi\). Furthermore, addressing atoms transversally \((\Delta \phi = 0)\) seems more favorable at typical magic wavelength trapping distances, e.g. \(d/\lambda_0 \approx 0.58\) for \(^{87}\text{Sr}\) \([16, 21, 37]\). Again, for \(d \ll \lambda_0\) the mean-field approach breaks down and one should rather turn to the Dicke model \([38]\), reducing \(N\) two-level emitters to one effective spin-\(N/2\) system \([29]\). Let us finally discuss the consequences for typical cases. Figure 5 shows the time evolution of the average spin for an infinite chain initialized in a symmetric Ramsey state with either no phase shift or with \(\Delta \phi = 0\). The parameters used are \(d = 0.792\lambda_0\) with \(\Delta \phi = 0\) (red triangles), where \(\Omega_{\text{eff}} = 0\) and \(\Gamma_{\text{eff}}\) is nearly optimal, as well as \(d = 0.49\lambda_0\) (green squares) and \(d = 0.51\lambda_0\) (blue circles), both with \(\Delta \phi = \pi\) which are close to a \(\Gamma_{\text{eff}}\) discontinuity. The solid lines correspond to a solution of a second-order cumulant expansion model with 200 particles and demonstrate a very good agreement with the infinite mean-field description.

Fig. 3: (Color online) Distance dependence of the effective quantities \(\Omega_{\text{eff}}\) and \(\Gamma_{\text{eff}}\) for an infinite equidistant chain, a square lattice and a hexagonal lattice (dashed black line) compared to their not yet converged finite counterparts of 10, 4 \(\times\) 10, and 10\(^5\) particles, respectively (solid red line). Again, we find divergences at integral \(d/\lambda_0\) owing to the \(1/\xi\) terms in \(F(\xi)\) and \(G(\xi)\). In the 2D configurations \(\Gamma_{\text{eff}}\) plateaus at \(-1\) for \(d < \lambda_0\), suggesting that this parameter range will be the most favorable for clock setups as decay is strongly suppressed. Finite sample sizes in our numerics can lead to strong oscillations of the effective quantities at small distances and even to unphysical values of \(\Gamma_{\text{eff}} < -1\).

Fig. 4: (Color online) Effective interactions \(\Omega_{\text{eff}}\) and \(\Gamma_{\text{eff}}\) for an infinite chain with spacing \(a\) where the spins are initially prepared with phase shift \(\Delta \phi\) between neighboring spins. The dashed lines indicate parameters with \(\Omega_{\text{eff}} = 0\) optimal for an optical clock.

Fig. 5: (Color online) Three different examples for the time evolution of the spin expectation values for a chain with spacing \(d\) where initially all spins are prepared in a coherent superposition of ground and excited state with a phase shift of \(\Delta \phi\). The parameters used are \(d = 0.792\lambda_0\) with \(\Delta \phi = 0\) (red triangles), where \(\Omega_{\text{eff}} = 0\) and \(\Gamma_{\text{eff}}\) is nearly optimal, as well as \(d = 0.49\lambda_0\) (green squares) and \(d = 0.51\lambda_0\) (blue circles), both with \(\Delta \phi = \pi\) which are close to a \(\Gamma_{\text{eff}}\) discontinuity. The solid lines correspond to a solution of a second-order cumulant expansion model with 200 particles and demonstrate a very good agreement with the infinite mean-field description.
shift or a phase shift of $\Delta \phi = \pi$ between neighboring atoms. The lattice constants have been chosen to be approximately $\lambda_0/2$ as would be typical [5]. We refrain from choosing exactly $\lambda_0/2$ to avoid the $1/\xi$ divergence. We observe that the dipoles’ lifetimes vary strongly, comparing the subradiant behavior (red) where the collective dipole lives much longer than the natural lifetime of the atom to the superradiant (green) regime where the excitation vanishes very quickly. Additionally, to highlight the validity of the mean-field approach, we add the results of a second-order expansion simulation. Corresponding results for a full Ramsey sequence are shown in the supplementary material in [34].

Conclusions. — In densely filled optical lattices dipole–dipole interaction and collective decay significantly change the evolution of an induced collective dipole strongly affecting the Ramsey spectroscopy. Due to the long-range nature of the coupling, sizable shifts appear even for long-lived clock states despite their minute dipole moment, which limits the accuracy and precision of the Ramsey spectroscopy. Shifts and dephasing in large systems strongly depend on the dimensionality and geometry of the lattice, exhibiting resonant enhancements at particular lattice constants. While at the current operating densities for strontium [13–15] these shifts are smaller than other technical imperfections, they constitute inherent fundamental perturbations even in perfectly filled lattice clocks.

In this work, we have identified optimal operation geometries, which combine a negligible effective shift with a strong suppression of decay. In particular, for a 1D lattice with a tailored excitation angle and for a 2D hexagonal lattice, favorable operation parameters for future generation clock setups were found. These results appear to be robust against small position fluctuations or a few lattice defects. In this sense it seems possible to implement a high-density dark-exciton based atomic clock geometry, where the fundamental limit to line shifts is many orders of magnitude below a single Hz and one gets almost unlimited exciton lifetimes. In three dimensions the interactions are particularly sensitive to a change in lattice constant and boundary effects, which dominate even for billions of particles rendering such setups very challenging [34].

We have considered perfectly filled and designed optical lattices, while in any experimental setup some imperfections in the form of defects or position fluctuations will be present. This can be of fundamental quantum nature [39,40] or simply stem from technical imperfections. Interestingly, at least in 1D geometries we found that this even leads to strong subradiant behavior and, thus, could be a useful resource [41,42]. This effect has to be confirmed for higher dimensions, though.

While for most considerations we have focused on the case of clock transitions, the same physics is present in a more prominent and experimentally easier observable form for broader transitions. Optimizing geometries will also be relevant for devices such as superradiant lasers [43,44] or lattice-based optical memories.

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