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To cite this version:
Mauro Antezza, Yvan Castin. Spectrum of Light in a Quantum Fluctuating Periodic Structure. Physical Review Letters, American Physical Society, 2009, 103, pp.123903. hal-00365761v2

HAL Id: hal-00365761
https://hal.archives-ouvertes.fr/hal-00365761v2
Submitted on 16 Sep 2009

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Spectrum of Light in a Quantum Fluctuating Periodic Structure

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(Dated: September 16, 2009)

We address the general problem of the excitation spectrum for light coupled to scatterers having quantum fluctuating positions around the sites of a periodic lattice. In addition to providing an imaginary part to the spectrum, we show that these quantum fluctuations affect the real part of the spectrum, in a way that we determine analytically. Our predictions may be observed with ultracold atoms in an optical lattice, on a \( J = 0 \to J' = 1 \) narrow atomic transition. As a side result, we resolve a controversy for the occurrence of a spectral gap in a fcc lattice.

PACS numbers: 42.50.Ct, 67.85.-d, 71.36.+c

The investigation of light propagation in a periodic structure is a fundamental problem in condensed matter physics \([3, 4]\), ranging from the physics of photonic crystals \([5, 6]\) to X ray \([7, 8, 9, 10, 11, 12]\) or even \( \gamma \) ray \([3, 4]\) scattering by a crystal. It recently gained a renewed interest \([5, 6, 15, 20]\) thanks to the possibility of producing artificial periodic structures of quantum dots \([3, 4]\) or of atoms \([3, 4]\), which have a much larger spatial period than natural crystals, allowing a measurement with a laser of the excitation spectrum over the whole Brillouin zone.

In reality, strictly periodic structures do not exist: the quantum (if not thermal) fluctuations of the positions of the scatterers in the structure are unavoidable. From general grounds, we know that these fluctuations affect the spectrum of light in two ways. First, they introduce a dissipative component: the elementary excitation spectrum acquires an imaginary part, a well known effect of phonon coupling in condensed matter physics. Second, they introduce a reactive component, modifying the real part of the excitation spectrum. Whereas the fluctuations of the scatterers positions have indeed been taken into account in simplified models \([1, 2]\), the only explicit prediction for the corresponding modification to the excitation spectrum was given in \([3, 4]\).

The problem however is not closed yet: in the limit of vanishing position fluctuations, Eq. (4.9) of \([3, 4]\) for the real part of the spectrum does not reduce to the prediction given in \([3, 4]\) for fixed scatterers, a prediction that also does not coincide with the one of \([4]\). This problem is not only formal, it may soon be addressed in current experiments with narrow line cold atoms trapped in optical lattices \([3, 4]\), where measurements of the spectrum with a good precision may be performed. Here we provide a conclusive analytical answer to this problem.

Model: Although our method to come is general, we assume for concreteness that the scatterers are coupled to the electromagnetic field on an electronic transition between the ground state \( g \) of spin 0 and an excited state \( e \) of spin 1. The atoms are trapped at the nodes of an optical lattice, with nowhere more than one atom per site \([3, 4]\). In the deep-lattice limit, tunneling is negligible and the \( i \)th atom is assumed to be harmonically trapped around lattice site \( \mathbf{R}_i \), with the potential \( U_i(\mathbf{r}_i) = m\omega_\text{ho}^2/2(\mathbf{r}_i - \mathbf{R}_i)^2/2 \). Here \( m \) is the atomic mass, \( \mathbf{r}_i \) is the position operator of atom \( i \), and \( \omega_\text{ho} \) is the atomic oscillation frequency. In this regime, the fluctuations of the atomic positions are purely on-site and uncorrelated, contrarily to the case of phonons in a crystal \([3, 4]\). The Hamiltonian of our system \([3, 4]\) may be split in the non-interacting term \( H_0 \) and the atom-field dipolar coupling \( V \), \( H = H_0 + V \), with

\[
H_0 = \sum_{\alpha} \hbar \omega_\alpha \left| \alpha \right\rangle \left\langle \alpha \right| + \frac{\hbar c}{2} \int_D \sum_{\epsilon \neq k} \hbar \epsilon \hat{a}_{\epsilon k}^\dagger \hat{a}_{\epsilon k}
\]

Here \( N \) is the number of atoms, \( \omega_\text{ho} \) is the bare atomic resonance frequency, the sum \( \sum_\alpha \) over the three directions of space \( x, y, z \) accounts for the three-fold degeneracy of \( \epsilon \), \( D \) is the three-dimensional Fourier space truncated by a cut-off \( k < k_M \), and the annihilation and creation operators obey usual bosonic commutation relations such as \( \left[ \hat{a}_{\epsilon k}, \hat{a}_{\epsilon' k'}^\dagger \right] = \delta_{\epsilon, \epsilon'} \delta(\mathbf{k} - \mathbf{k}') \), where \( \epsilon \) and \( k \) are the photon polarization and wavevector. The coupling \( V \) is \([4]\)

\[
V = -\sum_{i=1}^N \hat{D}_{i,\alpha} \cdot \mathbf{E}_\perp(\mathbf{r}_i)
\]

where \( \hat{D}_{i,\alpha} = d\left| i : e_\alpha \right\rangle \left\langle i : g \right| + \text{h.c.} \) is the component along direction \( \alpha \) of the dipole operator \( \hat{D}_i \) of the \( i \)th atom, proportional to the atomic dipole moment \( \mathbf{d} \), \( \mathbf{E}_\perp(\mathbf{r}) = \int_D d^3k \sum_{\epsilon \neq k} \mathbf{E}_{\epsilon k} \hat{a}_{\epsilon k} e^{ik\cdot\mathbf{r}} + \text{h.c.} \) is the transverse electric field operator, and \( \mathbf{E}_{\epsilon k} = i(2\pi)^{-3/2}[\hbar \epsilon/(2\epsilon_0)]^{1/2} \).

Method: We treat the coupling \( V \) to second order of perturbation theory, to calculate atom-field elementary excitations mainly of atomic nature. In practice for a lattice spacing \( \sim 1/k_0 \), where \( k_0 = \omega_\text{ho}/c \) is the resonant wavevector, this requires \( \Gamma \ll \omega_\text{ho} \), where \( \Gamma = \Delta^2 k_0^3/(3\pi\epsilon_0 h^3) \) is the free space atomic spontaneous emission rate. The energies of the system up to second order in \( V \) are eigenvalues of the effective Hamiltonian

\[
H_{\text{eff}} = PHP + PVQ + \frac{Q}{E(0)Q - QH_0Q} QVP,
\]
where $E^{(0)}$ is an eigenenergy of $H_0$, $P$ projects orthogonally onto the corresponding eigenspace of $H_0$, and $Q = I - P$. We now obtain the excitation energies of the system as the difference between excited states energies and the ground state energy.

For the perturbative calculation of the ground state energy $E_g$, we take for $P$ the projector over the state with all the atoms in the electronic and motional ground states, and the field in vacuum, so that $E_g^{(0)} = N \frac{1}{2} \omega \omega_{ho}$. We obtain $E_g = N \epsilon_g$, with

$$
\epsilon_g = \frac{3}{2} \omega_{ho} - \frac{d^2}{2 \omega_{ho}} \int_D d^3k \frac{\hbar c k}{2 \omega_{ho} + \hbar c k}.
$$

(4)

For the excited state energies, we take for $P$ the projector $P_e$ over all the states $|i : \epsilon_i\rangle$, where $|i : \epsilon_i\rangle$ represents the atom $i$ in the electronic state $\epsilon_i$, the $N - 1$ other atoms in the ground electronic state, all the atoms in the motional ground state, and the field in vacuum. Then $P$ projects over a subspace of dimension $3N$. Since this excited subspace is coupled by $V$ to the continuous part of the spectrum of $H_0$, one has to replace $E^{(0)}$ by $E^{(0)}_e + i0^+$ in (4), with $E^{(0)}_e = E^{(0)} + \omega_{ho}$, which gives rise to a complex excited state energy $E_e$, eigenvalue of

$$H_{eff} = [(N - 1) \epsilon_e + \epsilon_e] P_e + \sum_{i \neq j, \alpha \beta} g_{\alpha \beta}(R_i - R_j) |i : \epsilon_i\rangle \langle j : \epsilon_j|.
$$

(5)

Here $\epsilon_e$ is the complex energy of a single atom $\epsilon_e = \hbar \omega_{ho} + \frac{d^2}{2 \omega_{ho}} \int_D d^3k \frac{\hbar c k}{2 \omega_{ho} + \hbar c k}.$

(6)

To obtain the excitation energies of the system we subtract the ground state energy $E_g$ from (5). In this subtraction, the dangerous terms proportional to the number of atoms $N$ disappear and the interatomic distance independent term gives the excitation energy for a single atom, that we split in a real part and an imaginary part:

$$\epsilon_e - \epsilon_g \equiv \hbar \omega_A - i \frac{\hbar \Gamma}{2}.
$$

(7)

As expected, $\Gamma$ is the free space spontaneous emission rate, and the effective atomic resonance frequency $\omega_A$ deviates from $\omega_0$ by Lamb shift type terms.

Most interesting are the position dependent terms in (5), which contain the effective coupling amplitude $\tilde{g}_{\alpha \beta}(r)$ for the transfer of the atomic excitation between different sites separated by $r$. This coupling amplitude appears as the inverse Fourier transform of $g_{\alpha \beta}(k)$ of the function

$$\tilde{g}_{\alpha \beta}(k) = 3 \pi \hbar \Gamma \frac{k^2 \delta_{\alpha \beta} - k_a k_b}{k^2 a^2_{\alpha \beta}} e^{-k^2 a^2_{\alpha \beta}}.$$

(8)

The effect of the quantum fluctuations of the atomic positions on the interstate coupling here enters through the size of the harmonic oscillator ground state $a_{ho} = [\hbar/(2m\omega_{ho})]^{1/2}$.

The integral defining $\tilde{g}_{\alpha \beta}(r)$ is cut at large $k$ by the Gaussian factor of momentum width $1/a_{ho} \ll \kappa M$. Hence we can evaluate the coupling amplitude $g_{\alpha \beta}(r_i - r_j)$ by extending the integral over $k$ to the whole space. In this limit the coupling amplitude is the average, over the harmonic oscillator ground state probability distributions of $r_i$ and $r_j$, of the function $g_{\alpha \beta}(r_i - r_j)$, where

$$g_{\alpha \beta}(r) = \frac{3 \hbar \Gamma}{4 k_0^2} \left[ k_0^2 \delta_{\alpha \beta} + \partial_{r_a} \partial_{r_b} \right] \frac{e^{i k_0 r}}{r}$$

(9)

is proportional to the component along $\alpha$ of the classical electric field radiated by a point-like dipole of frequency $\omega_0$ oriented along direction $\beta$. A crucial consequence of the average is that, whereas $g_{\alpha \beta}(r)$ has the electrostatic $1/r^3$ divergence at the origin, the function $\tilde{g}_{\alpha \beta}(r)$ is regular even in $r = 0$, with a value

$$\tilde{g}_{\alpha \beta}(0) = \frac{3 \hbar \Gamma}{2 k_0^2} \left[ \frac{\text{Erfi}(k_0 a_{ho}) - i}{e^{k_0 a_{ho}^2}} - \frac{1 + 2(k_0 a_{ho}^2)}{2 \pi^{1/2} (k_0 a_{ho})^2} \right].$$

(10)

where $\text{Erfi}(x) = 2^{x^{-1/2}} \int_0^x dy \exp^2(y^2)$ is the imaginary error function.

For large $r$, the average over the atomic motion does not suppress the long range nature of the radiated dipolar field $\propto \exp(i k_0 r)/r$. From (10) one has indeed $\tilde{g}_{\alpha \beta}(r) \simeq g_{\alpha \beta}(r) \exp(-k^2 \omega_{ho}^2)$ remarkably as soon as $r \gg a_{ho}$.

Periodic case: We now take the limit $N \to +\infty$ with one atom per lattice site, realizing for the light field a periodic crystal, where an arbitrary Bravais lattice. To diagonalize $H_{eff}$ we rely on Bloch theorem: The eigenvectors $|\psi_\mathbf{q}\rangle$ depend on the lattice site position $R_i$ as

$$|i : \epsilon_i|\langle \psi_\mathbf{q}| = \tilde{d}_\mathbf{q} e^{i \mathbf{q} \cdot \mathbf{R}}.$$

(11)

where the Bloch vector $\mathbf{q}$ is chosen in the first Brillouin zone of the lattice. I.e. the "dipole" carried by atom $i$ differs from the one $\mathbf{d}$ carried by the atom in $\mathbf{R} = 0$ by a global phase factor. Thus the infinite dimension eigenvalue problem on the excitation spectrum $\epsilon_\mathbf{q}$, 

$$[H_{eff} - E_g P_e]|\psi_\mathbf{q}\rangle = \epsilon_\mathbf{q}|\psi_\mathbf{q}\rangle,$$

(12)

reduces to the diagonalization of the $3 \times 3$ matrix $M$, $M\mathbf{d} = \epsilon_\mathbf{q} \mathbf{d}$, with matrix elements

$$M_{\alpha \beta} = \left( \hbar \omega_A - i \frac{\hbar \Gamma}{2} \right) \delta_{\alpha \beta} + \sum_{\mathbf{R} \in \mathbf{L}} g_{\alpha \beta}(\mathbf{R}) e^{i \mathbf{q} \cdot \mathbf{R}},$$

(13)

where the sum runs over the lattice $\mathbf{L}$ excluding the origin. By adding and subtracting $g_{\alpha \beta}(0)$ and using Poisson’s summation formula we convert this sum into a sum over the reciprocal lattice $\mathbf{R}L$:

$$M_{\alpha \beta} = \left( \hbar \omega_A - i \frac{\hbar \Gamma}{2} \right) \delta_{\alpha \beta} - \tilde{g}_{\alpha \beta}(0) + \frac{1}{|\mathbf{L}|} \sum_{\mathbf{K} \in \mathbf{R}L} \tilde{g}_{\alpha \beta}(\mathbf{K} - \mathbf{q}).$$

(14)
where $V_L$ is the primitive unit cell volume of the lattice.

**Imaginary part:** Since each term of the sum over $K$ in (14) is real, the imaginary part of the excitation spectrum can be calculated explicitly:

$$\Im \varepsilon_q = -\frac{\hbar \Gamma}{2} \left(1 - e^{-k_0^2 a_{ho}^2}\right).$$

As already mentioned, this non-zero value is a direct consequence of the fluctuating scatterers positions. This is why an expression analogous, but not equal to (15) was derived in [5] in the different context of $\gamma$ ray nuclear scattering in a crystal.

The non-zero imaginary part of $\varepsilon_q$ is indeed due to the decay of the system out of the subspace where all the atoms are in their motional ground state. When an excited atom $\epsilon_a$ in its motional ground state $|0\rangle_{ho}$ spontaneously emits a photon of polarization $\epsilon$ and momentum $k_0n$, $|n| = 1$, its probability density to fall in $g$ with an excited motional state is $\langle 3\Gamma/2 \rangle |\epsilon_a|^2 |1 - |_{ho}(\exp(-i(k_0n \cdot r)|_{ho})|^2$, which after sum over $\epsilon, l, n$ and average over the direction $n$, exactly gives the decay rate $-2\Im \varepsilon_q/\hbar$. This process conserves the quasi-momentum $q$. If the emitted photon carries away the quasi-momentum $q_{ph}$, the resulting $g$ atom in the motional excited state is coherently delocalized over the whole lattice, with a probability amplitude $\propto e^{iq_{ph} \cdot R}$ of being in site $R$, and a quasi-momentum $q_{at} = q - q_{ph}$.

Since $q_{ph}$ belongs to a continuum, this spontaneous emission process opens up a continuum of final states, hence the possibility to have for a fixed $q$ a continuous spectrum for $H$ and a complex $\varepsilon_q$. Experimentally, to obtain long lived elementary excitations, one may operate in the so-called Lamb-Dicke regime, $k_0a_{ho} \ll 1$, where the loss rate $-2\Im \varepsilon_q/\hbar \approx \Gamma(k_0a_{ho})^2$ is much smaller than $\Gamma$.

**Real part:** In (14) we replace $\bar{g}_{\alpha \beta}(0)$ and $\bar{g}_{\alpha \beta}$ by their explicit expressions (13) and (8). Then $\Re \varepsilon_q - \hbar \omega_A$ is an eigenvalue of the $3 \times 3$ real symmetric matrix

$$\delta M_{\alpha \beta} = \frac{\hbar \Gamma}{2} \delta_{\alpha \beta} \left[1 + \frac{2(k_0a_{ho})^2}{2\pi^2/k_0^2a_{ho}^4} - \text{Erfi}(k_0a_{ho})^2 e^{-k_0^2a_{ho}^2}\right] + \frac{3\pi \hbar \Gamma}{k_0^4 V_L} \sum_{K \in RL} \delta_{\alpha \beta} K^2 - K'_{\alpha} K'_{\beta} \delta_{K - K'} e^{-K^2a_{ho}^2}$$

where $K' \equiv K - q$ [9]. Eq.(14) is useful for a numerical calculation of the spectrum, see Fig.1. Remarkably, one can even derive analytically the dependence of the spectrum with $a_{ho}$ from (13): one multiplies $\bar{g}_{\alpha \beta}(R)$ by $e^{K^2a_{ho}^2}$, and one takes the derivative with respect to $a_{ho}^2$. In the Fourier representation (8), this pulls out a factor $k_0^2 - K^2$ which exactly cancels the denominator. The resulting Fourier integrand is now essentially Gaussian and can be directly calculated:

$$u_{\alpha \beta}(r) = \partial_{a_{ho}} \left[ e^{K_{\alpha}^2a_{ho}^2} \bar{g}_{\alpha \beta}(r) \right] = \frac{3\hbar \Gamma e^{K_{\alpha}^2a_{ho}^2}}{8\pi^2/k_0^2a_{ho}^4} \times \left(-\delta_{\alpha \beta} \Delta_{r} + \partial_{r_{\alpha}} \partial_{r_{\beta}} \right) e^{-r^2/(4a_{ho}^2)}.$$  

**FIG. 1:** Spectrum of light in a quantum fluctuating periodic atomic structure. (a) For a simple cubic lattice, with a lattice constant $a$ ($V_L = a^3$), comparison of the analytical prediction (solid lines) with the numerical solution of (16) (symbols), for two values of $k_0a_{ho}$ and of the Bloch vector ($\times$: point $R$ and $\circ$: point $\Gamma$ of the first Brillouin zone). (b) and (c): Real part of the spectrum as a function of the Bloch vector along the standard irreducible path in the first Brillouin zone, for (b) a simple cubic lattice, and for (c) a face-centered cubic lattice with a lattice constant $2a$ ($V_L = 2a^3$): here $k_0a = 2$ and $k_0a_{ho} = 1/\sqrt{3}a$, which leads to $a_{ho}/a \simeq 0.09$. In both cases the band structure is gapless.

From (19) one then obtains a sum excluding the origin:

$$\partial_{a_{ho}} \left[ e^{K_{\alpha}^2a_{ho}^2} \delta M_{\alpha \beta} \right] = \sum_{R \in L} u_{\alpha \beta}(R) e^{iq_{R}.}$$

It is apparent that each term of the sum is exponentially small since, in the deep lattice limit, the harmonic length $a_{ho}$ is much smaller than the lattice spacing. Keeping these terms is actually beyond accuracy of our Hamiltonian (see [8]). One can thus set the right-hand side of (19) to zero, which leads to $\delta M_{\alpha \beta} = e^{-k_0^2a_{ho}^2} \delta M_{0\beta}$, where $\delta M_{0\beta}$ is independent of $a_{ho}$ and is simply the limit of $\delta M_{\alpha \beta}$ when $a_{ho} \to 0$. This shows that the real part of the excitation spectrum is a Gaussian function of $a_{ho}$:

$$\Re \varepsilon_q - \hbar \omega_A = e^{-k_0^2a_{ho}^2} (\varepsilon_0 - \hbar \omega_A).$$

where $\varepsilon_0$ is the limit of the excitation energy for $a_{ho} \to 0$. Eq.(19) is the main result of this work, it gives in a very
explicit way the influence of the fluctuations of the atomic positions on the real part of the light spectrum. It is in excellent agreement with the numerics, see Fig.3.

**Band structures:** We illustrate (14) by numerically calculating the band structure in the two lattice geometries of [3, 4]. In Fig.3 we show Re $\varepsilon_q - \hbar \omega_A$ as a function of the Bloch vector along the standard irreducible path in the first Brillouin zone, for the simple cubic (Fig.3a) and for the face-centered cubic (fcc) (Fig.3b) lattices. The figure reveals the lack of an omnidirectional gap, which confirms the prediction of [7, 9] against the one of [8]. In Fig.1 we show Re $\varepsilon_q$ and cannot open or close an energy gap. The situation may be different for anisotropic microtraps. Experimental issues: The perturbative regime $\Gamma \ll \omega_{\text{tao}}$ considered here was not usual in atomic lattice experiments. It is now available in experiments using very narrow transitions for atomic clock purposes: e.g. $^{88}$Sr was recently trapped in a deep 3D lattice, with $\omega_{\text{tao}} \sim 0.05 \omega_0 \sim 20$nm [4], and it has a narrow line $5s^2 \, ^1S_0 \rightarrow 5s5p \, ^3P_1$, realizing the needed $J = 0 \rightarrow J' = 1$ transition, with $\Gamma \sim 0.05 \omega_0$. To produce and spectroscopically probe elementary excitations with $q \neq 0$, one cannot use the direct $g \rightarrow e$ coupling with resonant light, but one can use an indirect Raman coupling [22].

We also assumed that $g$ and the three sublevels of $e$ experience the same trapping potential. This is the case in Wigner ion crystals [22], where $e$ and $g$ experience the same Coulomb shift. For neutral atoms, the lattice potential is a lightshift, that may deviate from one of the two assumptions of [4], (i) $e$ experiences a scalar lightshift, and (ii) the lightshifts of $e$ and $g$ are equal. For a lattice obtained by incoherent superposition of laser standing waves along $x$, $y$, and $z$ linearly polarized along $y$, $z$, and $x$ respectively, violation of (i) breaks the harmonic oscillator isotropy: the sublevel $\varepsilon_x$ (resp. $\varepsilon_y$, $\varepsilon_z$) has an oscillator length $\eta_x \omega_{\text{tao}}$ along $z$ (resp. $x$, $y$) different from the one $\eta_y \omega_{\text{tao}}$ along the other two directions. This does not break the three-fold degeneracy of the motional ground state in $e$ but it reduces the overlap between the motional ground state of $e$, and that of $g$. Hence after spontaneous emission, even if one neglects the atom recoil ($k_0 \omega_{\text{tao}} \rightarrow 0$), the atom in $g$ can populate an excited motional state, giving a non-zero decay rate to the elementary excitations. Violation of condition (ii) leads to an oscillator length in $g$ equal to $\eta_y \omega_{\text{tao}}$, $\eta_y \neq 1$, which also increases the decay rate. For $k_0 \omega_{\text{tao}} \rightarrow 0$, combining both violations gives

$$\text{Im } \varepsilon_q = \frac{\hbar \Gamma}{2} \left( 1 - \frac{8}{\eta_x \eta_y + (\eta_x \eta_y)^{-1}][\eta_y + \eta_y^{-1}]^2 \right).$$

E.g. if one has achieved $\eta_y \simeq 1$ at the expense of having an optical lattice depth in $g$ twice as small/large as in $e$, one still finds a small decay rate $\simeq 0.04 \Gamma$.

**Conclusion:** Quantum fluctuations of the positions of the scatterers in a periodic structure very generally give rise to an imaginary part in the spectrum of light and affects its real part. For scatterers tightly trapped in a periodic potential, we derived an expression for this spectrum. We showed that, amazingly, its dependence on the amplitude of the fluctuations of the positions is a Gaussian, not only for the imaginary part [22] but also for the real part. This effect on the real part can be large and may be observed in recent atomic lattice clock experiments. An intriguing perspective is the extension of this work to the disordered case and to localized states of light.

We acknowledge discussions with I. Carusotto, D. Willkowski, E. Arimondo, B. Duk, G. La Rocca, A. Sinatra, and support from IFRAF and ANR Gascor.

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