Trapping of cold atoms by the quadrupole force

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Abstract

Cold atoms are traditionally trapped by the dipole force in periodically spaced potential wells induced by the standing laser field. We derive here a theory beyond the conventional dipole approximation which provides field/atom coupling potential terms that so far have not been taken into consideration in theoretical or experimental studies. We show that for some atoms for specific laser parameters despite the absence of dipole transition laser trapping is still possible due to the quadrupole force. Illustrative numerical calculations for Ca and Na trapped by the quadrupole force are given.
The possibility of trapping cold neutral atoms in periodic potentials formed by standing laser waves, so called optical lattices, has opened a new subject of research. For reviews see for example Refs. [1, 2, 3, 4, 5] and references therein. Laser cooling and the formation of optical lattices were explained by the ability to influence the translational motion of neutral atoms by the laser induced dipole force [4]. Until now, the formation of optical lattices has been studied theoretically and experimentally exclusively due to the dipole interactions. An important question arises, however, on what happens when the dipole transition matrix element vanishes due to a symmetry property of the studied atomic system. This is the case, for example, when the laser frequency is detuned from an excited state which is associated with a \(d\)-type symmetry orbital whereas in the ground state the valence shell electron occupies an \(s\)-type symmetry orbital. Does this imply that in such a case atoms can not be trapped because one can not form an optical lattice?

The purpose of the present study is to demonstrate that it is possible to trap atoms even when the dipole transition between the two relevant electronic levels coupled by the laser is prohibited by symmetry. We derive here the Hamiltonian of optical lattices without imposing the dipole approximation, and study the possibility of generating optical lattices due to the quadrupole contributions to the trapping potential which until now have been neglected. Two illustrative examples are presented explicitly. Namely, we show that by using realistic laser parameters the quadrupole force induced optical lattice can be formed and trap \(Ca\) and \(Na\) atoms.

Let us consider an atom interacting with a linearly polarized laser light. The light propagation direction is chosen to be \(x\), and the field is assumed to oscillate along the \(z\)-direction. For the sake of simplicity and without loss of generality we describe the field free atomic Hamiltonian by an effective one electron model Hamiltonian, \(H_{\text{el}}^{FF}\). The formulation given here can be extended in a straightforward manner to the general case of a many electron atom (or ion) interacting with an arbitrary (not necessarily linearly polarized) light pulse. Such a generalization will be elaborated elsewhere [6]. By switching from the laboratory frame coordinates to the relative \(\vec{q} = (x, y, z)\) and center-of-mass (c.m.) \(\vec{R} = (X, Y, Z)\) coordinates, one obtains the full Hamiltonian of an effective one electron atom in a linearly polarized laser field,

\[
H(t) = -\frac{\hbar^2}{2M} \nabla^2_{\vec{R}} + H_{\text{el}}^{FF}(\vec{q}) + V_{\text{coup}}(\vec{q}, \vec{R}, t)
\]  

(1)
with an interaction term \( \mathbf{V}_{\text{coup}}(\mathbf{q}, \mathbf{R}, t) = e(m_e c)^{-1} A_{m_e} \hat{p}_z + e^2(2m_e c^2)^{-1} A_{m_e}^{(2)} + e(cM)^{-1} A_M \hat{P}_Z + e^2(2(M-m_e)c^2)^{-1} A_M^{(2)} \). Here, the \( A \)-factors are defined in terms of the time, relative coordinate, and c.m. coordinate dependent vector potential \( A(x, X, t) \), as follows:

\[
A_{m_e} = \left[ A(x\xi_1 + X, t) + (\xi_2/\xi_1)A(-x\xi_2 + X, t) \right]; \quad A_M = \left[ A(x\xi_1 + X, t) - A(-x\xi_2 + X, t) \right]; \quad A_{m_e}^{(2)} = A^2(x\xi_1 + X, t) \text{ and } A_M^{(2)} = A^2(-x\xi_2 + X, t).
\]

The symbols \( m_e \) and \( M \) stand, respectively, for the mass of the electron and the atom, and parameters \( \xi_1 = (M - m_e)/M, \xi_2 = m_e/M \).

When the field intensity \( I \) is weak (\( I \) is proportional to the squared field amplitude) the linear interaction terms \( A_{m_e}(x, X, t)\hat{p}_z \) and \( A_M(x, X, t)\hat{P}_Z \) become dominant over the quadratic interaction terms \( A_{m_e}^{(2)} \) and \( A_M^{(2)} \) so that the latter two contributions can be neglected. Moreover, since \( M \gg m_e \) it is clear that \( \xi_1 \approx 1 \) and thus the dominant term in \( \mathbf{V}_{\text{coup}}(\mathbf{q}, \mathbf{R}, t) \) is the first one. In summary, we conclude that the coupling between the c.m. and relative coordinates can be considered as

\[
\mathbf{V}_{\text{coup}}(\mathbf{q}, \mathbf{R}, t) = e(m_e c)^{-1} A_{m_e}(x, X, t)\hat{p}_z .
\] (2)

In standard treatments of the atom-field Hamiltonian the dependence of the vector potential on the relative coordinate has been neglected. That is, \( A(x + X, t) \sim A(X, t) \). Here, we do not use this so called dipole approximation. Instead, the spatial dependence of the vector potential is taken into account, via the Taylor expansion around \( x = 0 \). An important question arises on what should be taken as the small parameter in such a Taylor series expansion. In the case when the laser intensity is low, the associated time dependent electronic wavefunctions are only slightly different from their field-free counterparts. Therefore, they possess exponentially small values for \( q > a_0 \), where \( a_0 \) stands for the size of the atom. For weak fields, as in our case, \( a_0 \) is essentially equal to the radius of the field free atom, \( r_0 \). Of course, in the presence of very strong fields \( a_0 \) varies with the intensity as the electron oscillates with the field. Roughly speaking we may think that the relevant radius of the atom is then \( r_0 + a_0 \) where \( a_0 = (eA_0)^2/(c^2 m_e \omega_L) \) and \( A_0 \) is the field amplitude. In order to find a qualitative criterion for convergence of our Taylor series expansion of \( A(x + X, t) \), the above estimated atomic radius \( a_0 \) should be compared with the wavelength \( \lambda \) of the laser. Clearly, \( \lambda \) should get sufficiently large values such that \( \lambda \gg a_0 \). The small parameter of the considered Taylor series expansion is thus found to be \( k_L a_0 \), where \( k_L = 2\pi/\lambda \).

Following the above discussion, when the wavelengths which constitute the dominant part
of the laser pulse are sufficiently large, $\lambda \gg a_0$, the two leading order terms in the Taylor series expansion of the vector potential are given by

$$A(x + X, t) \sim A(X, t) + x \partial A(X, t)/\partial X.$$  

(3)

The first term in Eq. (3) is the dipole term, whereas the second one is the quadrupole term which is taken here into consideration for the first time in contrast to other theoretical studies published up to now. Substituting Eq. (3) into formula (2) one finds that

$$V_{\text{coup}}(\vec{q}, \vec{R}, t) = V_{\text{dip}}(z, X, t) + V_{\text{qd}}(z, x, X, t)$$

(4)

where the dipole and quadrupole potential terms that couple the internal (electronic) and c.m. degrees of freedom are respectively given by, $V_{\text{dip}}(z, X, t) = (e/c)m_\epsilon^{-1}A(X, t)\hat{p}_z$ and $V_{\text{qd}}(z, x, X, t) = (e/c)m_\epsilon^{-1}[\partial A(X, t)/\partial X]x\hat{p}_z$.

Since the field oscillates periodically in time the Hamiltonian is time periodic with the period $T = 2\pi/\omega_L$ where $\omega_L$ is the laser frequency. In such a case the solutions of the time dependent Schrödinger equation are the Floquet solutions, similarly as the Bloch states are the solutions of the Schrödinger equation for spatially periodic potentials. Within the framework of the Floquet theory, the time averaged optical potential, $\bar{V}_{\text{opt}}(X)$, is associated with the quasi-energy eigenvalue of the Floquet operator, $\mathcal{H}_F = -i\hbar\partial/\partial t + \mathbf{H}_{\text{el}}^{\text{FF}}(\vec{q}) + V_{\text{coup}}(\vec{q}, \vec{R}, t)$, (for Floquet formalism see for example Ref.9),

$$\mathcal{H}_F \Phi_{\text{opt}}^{\text{QE}}(\vec{q}, X, t) = \bar{V}_{\text{opt}}(X)\Phi_{\text{opt}}^{\text{QE}}(\vec{q}, X, t).$$

(5)

The eigenvalue solution of Eq. (5) provides the definition of the optical lattice potential. Strictly speaking, optical potentials for the atoms exist only in the adiabatic approximation where one can separate the atomic center of mass motion from the electronic motion (similarly to the Born-Oppenheimer or adiabatic approximation which separates the nuclear and electronic motions in molecules). Within the adiabatic approximation, our claim that the optical potential equals to an eigen-energy of the Floquet operator is exact. Of course, there are infinitely many eigen-energies and hence optical potentials (similarly to molecules where in each electronic state there is a potential for the vibrations). In equation (5) we have chosen that eigen-energy which correlates to the field-free atomic ground state. In the weak field limit we will show below that in the dipole case the same expression as that used in the literature results, whereas in the quadrupole case a novel expression for the quadrupole
force induced optical lattice potential is obtained. Note however that for sufficiently strong fields, the literature expression (which is found to be a second order perturbation theory result) fails whereas the eigen-energy of the Floquet operator still provides an exact optical potential.

Let us derive now an analytical expression for the optical lattice potential which includes the leading order term beyond the dipole approximation. From Eq. (3) it is clear that the only information we need in order to go beyond the dipole approximation is \( A(X, t) \) and its first order derivative with respect to the c.m. coordinate \( X \). For atoms in a standing laser beam \( A(X, t) = A_0 \cos(k_L X) \cos(\omega_L t) \), where \( \omega_L \) is the corresponding laser frequency, and \( k_L = \omega_L/c \). Suppose that the atomic ground state \( |g\rangle \) (with energy \( \mathcal{E}_g^0 \)) is significantly coupled by the laser light only to a single excited electronic state \( |e\rangle \) (with energy \( \mathcal{E}_e^0 = \mathcal{E}_g^0 + \hbar \omega_{\text{atom}} \)). By following the "traditional" approach based upon the adiabatic elimination scheme [7], we obtain the time dependent optical lattice potential (including the new term beyond the dipole approximation),

\[
V_{\text{opt}}(X, t) = |\langle e| V_{\text{dip}}(z, X, t) + V_{\text{qd}}(z, x, X, t)|g\rangle|^2/(\hbar \Delta_L),
\]

where, \( \Delta_L = \omega_{\text{atom}} - \omega_L \). The characteristic timescale of the center of mass dynamics is by several orders of magnitude smaller than the time period of an optical cycle, \( T = \lambda/c = 2\pi/\omega_L \). Therefore, the time averaged optical lattice potential reads as

\[
\bar{V}_{\text{opt}}(X) = \frac{1}{T} \int_0^T V_{\text{opt}}(X, t) \, dt.
\]

In case that the laser detuning with respect to the atomic frequency is such that it couples an \( s \)-type atomic state \( |g\rangle \) with a \( p \)-type atomic state \( |e\rangle \), the quadrupole contribution to \( \bar{V}_{\text{opt}}(X) \) vanishes, and the optical lattice potential is reduced to the expression

\[
\bar{V}_{\text{dip}}^{\text{opt}}(X) = \hbar |\Omega_{\text{dip}}|^2 (4\Delta_L)^{-1} \cos^2(k_L X) \tag{6}
\]

where the Rabi frequency is given by \( \Omega_{\text{dip}} = (e A_0)/(\hbar c m_e) \langle e|\hat{p}_z|g\rangle \). This is, of course, the well known conventional expression for the optical lattice potential formed by the dipole force [7].

Let us consider now the case when the laser detuning is off resonance but not from a \( p \)-type atomic state but from a \( d \)-type atomic state. Consequently, the force exerted on the atom is of quadrupole nature since the dipole contribution vanishes. Note that for Calcium, for example, the lowest excitation is from an \( s \) to a \( d \) orbital. Introducing the "generalized" Rabi frequency \( \Omega_{\text{qd}} = (e A_0)/(\hbar c m_e) k_L \langle e|x\hat{p}_z|g\rangle \) we obtain the formula

\[
\bar{V}_{\text{qd}}^{\text{opt}}(X) = \hbar |\Omega_{\text{qd}}|^2 (4\Delta_L)^{-1} \sin^2(k_L X) \tag{7}
\]
which exhibits remarkable similarity to the standard expression \[ (6) \] for the dipole optical lattice potential. Before proceeding further, let us recall once again that both relations \[ (6) \] and \[ (7) \] can be derived directly by applying the second order perturbation theory to the Floquet eigenproblem \[ (5) \].

Our theoretical analysis concludes with a very brief discussion of the as yet neglected incoherent effects (friction force, spontaneous emission) which are caused by the quantized nature of the electromagnetic field. It is known \[ [10] \] that the mentioned dissipation processes can be safely ignored when \( \Delta_L \gg \Gamma \), where \( \Gamma \) is the natural linewidth of the excited electronic state. For the dipole \( |e\rangle \rightarrow |g\rangle \) transitions,

\[
\Gamma_{\text{dip}} = 4e^2\omega_{\text{atom}}(3m_e^2\hbar^3)^{-1}|\langle e|\hat{p}_z|g\rangle|^2, \tag{8}
\]

see, e.g., Ref. \[ [11] \]. The derivation leading to Eq. \[ (8) \] can be extended in a straightforward manner to cover also the situation when the transition \( |e\rangle \rightarrow |g\rangle \) occurs solely via the quadrupole coupling. The final result is then given by formula

\[
\Gamma_{\text{qd}} = 4e^2\omega_{\text{atom}}^3(5m_e^2\hbar^5)^{-1}|\langle e|x\hat{p}_z|g\rangle|^2. \tag{9}
\]

Let us apply now our above theoretical results and study the feasibility of trapping \( Na \) and \( Ca \) atoms by the quadrupole force in the periodically spaced potential wells induced by the standing laser waves. Our model Hamiltonian consists of the ground S state, \( |E_S\rangle \), and the lowest excited P and D states, \( |E_P\rangle \) and \( |E_D\rangle \). The corresponding energy levels for \( Ca \) and \( Na \) were taken from the NIST atomic database. The field free electronic Hamiltonian is a diagonal \( 3 \times 3 \) matrix, \( E_e \), where the diagonal terms are the mentioned atomic energies \( E_S, E_P, E_D \). Following Eq. \[ (5) \], the interaction potential induced by the linearly polarized standing laser field is described by the symmetric \( 3 \times 3 \) matrix \( V_{\text{coup}}(X,t) \), where the diagonal matrix elements vanish. The off diagonal matrix elements are connected either with the dipole or the quadrupole transitions, and take an explicit form:

\[
(V_{\text{coup}})_{1,2} = eA_0/(m_e c)\langle E_S|\hat{p}_z|E_P\rangle \cos(k_L X) \cos(\omega_L t),
\]

\[
(V_{\text{coup}})_{2,3} = eA_0/(m_e c)\langle E_P|\hat{p}_z|E_D\rangle \cos(k_L X) \cos(\omega_L t)
\]

and \( (V_{\text{coup}})_{1,3} = -eA_0/(m_e c)k_L\langle E_S|x\hat{p}_z|E_D\rangle \sin(k_L X) \cos(\omega_L t) \). The corresponding dipole and quadrupole transition matrix elements for \( Na \) and \( Ca \) have been calculated using equations \[ (8) \] and \[ (9) \] from available theoretical/experimental data for the associated transition rates. More specifically, the dipole transition linewidths \( \Gamma_{\text{dip}}^{Na} \) and \( \Gamma_{\text{dip}}^{Ca} \) are extracted from the NIST
database, while the quadrupole terms $\Gamma^{Na}_{qd}$ and $\Gamma^{Ca}_{qd}$ are taken from the literature [12, 13]. Referring to previous discussion of Eq. (5), we recall that there is a single quasi-energy eigenvalue of the Floquet Hamiltonian $H_F = E_e + V_{coup}(X, t) - i\hbar(\partial/\partial t)$ which is associated with the field free ground state and which is interpreted physically as the appropriate numerically exact optical lattice potential.

In the first step of our calculations we computed the variation of the depth of the optical lattice potential well $V_{\text{max}} = \text{Max}[V_{\text{opt}}(X)]$ for Na atoms as a function of the laser intensity, $I$ [W/cm$^2$] $\propto (k_L A_0/2)^2$ for the laser parameters as were used recently in the experiment of Phillips group [14]. That is, Na atoms are trapped in a one dimensional optical lattice induced by a laser with $\lambda = 589$ nm, and the detuning from the first excited P state of Na is equal to $\Delta_L = 14$ GHz. Our calculations show that for $I < 100$ W/cm$^2$ the analytical results derived from the second order perturbation theory, Eq. (6), are in a remarkable agreement with the numerically exact values. Therefore, indeed within this region of field intensities the optical lattice is formed exclusively by the dipole force.

The situation is entirely different when the laser wavelength is taken to be $\lambda = 343$ nm and the detuning from the first excited D state is $\Delta_L = 1$ GHz. From Fig. 1 one can see that the quadrupole force induced optical lattice potential predicted by Eq. (7) is now dominant and in complete agreement with the numerically exact solutions of Eq. (5) for the entire range of displayed field intensities. The dipole force contribution to the formation of Na optical lattice as calculated from Eq. (6) is, for the considered laser parameters, by one order of magnitude smaller than the contribution of the quadrupole force. To achieve for the quadrupole induced optical lattice the same depth as being produced for the dipole induced optical lattice potential in the NIST experiment [14] one must increase the field intensity by 4 orders of magnitude. This is not a practical intensity in nowadays technology. However, by increasing the intensity by less than 2 orders of magnitude, the depth of the quadrupole induced optical lattice potential is smaller just by about 2 orders of magnitude than in the NIST experiments. Such a potential can trap Na atoms provided that they are cooled down to less than 0.1 $\mu$K. The required temperature is sufficiently low to keep the atoms trapped in the quadrupole induced optical lattice potential well since $k_B T < V_{\text{max}}$.

Let us now turn to the case of Ca atoms where the first excited state is a D state. The results of our calculations which are presented in Fig. 2 show that when the laser light is detuned slightly from the S $\rightarrow$ D transition ($\lambda = 457$ nm, $\Delta_L = 70$ kHz) it can trap the
Ca atoms solely by quadrupole interactions. The Ca atoms must, however, be pre-cooled. It is known that already today ultracold calcium can be prepared at 10 $\mu K$ [15]. At this temperature the Ca atoms can be trapped by the quadrupole lattice potential using a laser intensity of 10 W/cm$^2$. Other values of interest: In order to form a quadrupole induced optical lattice potential for field intensities which are smaller than 100 W/cm$^2$, the Ca atoms should be cooled to a temperature which is smaller than about 30 $\mu K$. For the intensity of $I = 1$ W/cm$^2$ the Ca atoms will be trapped by the quadrupole force when their temperature is smaller than 3 $\mu K$. In passing we note that the used value of detuning $\Delta_L = 70$ kHz is relatively small compared to that of Fig. 1, but still remains more than 200 times larger than the natural linewidth $\Gamma \approx 300$ Hz of the upper D state of Ca [13]. Therefore, the spontaneous emission effects can be safely neglected here. This statement has been confirmed also by an additional numerical calculation [6].

Above illustrative examples of course do not restrict the range of possible applications just to alkali metal or alkali earth atoms. Similar kind of quadrupole trapping can be exploited e.g. in the case of rare gases, which have been laser cooled and Bose condensed as well (see for example Ref. [16]).

In conclusion, we would like to re-stress that our formulation based upon the Floquet eigenstates is exact for all field intensities. This method provides a playground for a rational design of optical lattice potentials induced by the dipole or the quadrupole forces or by any combination of them. Design of quadrupole optical lattices might prove to be important in every practical situation where a specific lattice constant is required and where no dipole transition possesses a suitable wavelength. Such applications may include e.g. interference experiments with wavepackets of cold atoms, or an enhancement of the high harmonic generation efficiency via constructive interference/phase matching. Besides trapping of cold atoms in optical lattices, the theoretical framework described in the present paper can be used also for other purposes, e.g. for studying an effect of quadrupole interaction on the performance of atomic clocks (cf. Ref. [17]).

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FIG. 1: The depth $V_{\text{max}}$ of the optical lattice potential well for Na atoms plotted as a function of the laser intensity $I$. The laser parameters, $\lambda = 343$ nm and $\Delta L = 1$ GHz, correspond to the transition from the S ground state to the lowest D state of Na. The solid line represents the numerically exact solutions of Eq. (5) which coalesces to all significant digits with the second order perturbation theory contribution of the quadrupole forces, see Eq. (7). The dashed line shows the contribution of dipole forces calculated from Eq. (6).
FIG. 2: The depth $V_{\text{max}}$ of the optical lattice potential well for Ca atoms plotted as a function of the laser intensity $I$. The laser parameters, $\lambda = 457$ nm and $\Delta_L = 70$ kHz, correspond to the transition from the S ground state to the first excited D state of Ca. The solid line stands here for the numerically exact optical lattice potential as obtained from the numerical solution of Eq. (5). The dashed line shows the contribution of quadrupole forces accounted within the second order perturbation theory, see Eq. (7).