Laser-Induced Entangled Giant Quasi-Molecules in Optical Lattices

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We analyze atom-atom interactions in optical lattices due to a laser-induced long-range interatomic force which prevails over the usual London-van der-Waals forces. This force, which can be generated by an intense laser field at a wavelength longer than that of the lattice-generating laser, is shown to bind pairs of cold atoms trapped at different lattice sites, and cause their translational quantum correlations (spatial entanglement).

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The best-known laser-induced force acting upon or between atoms is the ”dipole force”, resulting from the action of the gradient of the laser field, which has been extensively used to confine cold atoms in optical traps [1]. This force has also been exploited in optical lattices, created by intersecting laser beams, which can trap cold atoms in periodic potential wells with typical depth of neV and inter-well separations on the order of the laser wavelength [2]. Recently, the mechanical effect of another laser-induced force has come to the fore, that of dipole-dipole interactions generated by off-resonant laser beams with small gradient (nearly plane waves) [3]. In the near-zone region of multiple, mutually-detuned and appropriately oriented laser beams, such dipole-dipole interactions yield a gravity-like interatomic 1/r attraction [4]. In contrast to the previously considered near-resonant dipole-dipole interaction [5] via real photon exchange between a ground-state atom and its excited partner, wherein their cooperative decay plays a role, the far-off resonant dipole-dipole interaction arises from virtual (nearly decay-free) photon exchange between two ground-state atoms, which acquire small induced dipole-moments [3].

Here we put forward a new effect of the off-resonant, laser-induced dipole-dipole interaction: its ability to bind together pairs of ground-state atoms localized at adjacent sites of an optical lattice. The resulting novel “giant” (submicron-size) quasimolecules will remain bound even if the lattice potential is switched off. An important feature of such quasimolecules are their translational quantum correlations or entanglement, i.e., nonfactorizability of the two-atom spatial wavefunction. The predicted effects should be contrasted with entanglement between internal states of closely separated atoms residing at the same site of a densely occupied optical lattice, via the essentially different near-resonant dipole-dipole interaction [6].

The retarded dipole-dipole interaction potential between atoms A and B, induced by an off-resonant circularly-polarized, plane-wave laser is [3]:

$$V_{AB} = -\frac{2\pi \omega_{A}^2 I}{c} F_{\theta}(kr); \quad F_{\theta}(kr) = \cos(kr \cos \theta)$$

$$\times \left\{ \cos(kr) + kr \sin(kr) \right\} \left(1 - 3 \cos^2 \theta \right) + \frac{1 + \cos^2 \theta \cos(kr)}{kr}. \quad (1)$$

Here $\theta$ is the angle between the interatomic axis and the wave-vector $k = \omega/c$ of the laser, $I$ is the laser intensity and $\alpha = \frac{2\omega_{A}^2 d^2}{\hbar (\omega_{A}^2 - \omega^2)}$ is the atomic dynamic polarizability, with $d$ and $\omega_{A}$ being the atomic dipole moment and transition frequency, respectively, assuming that both atoms $(A, B)$ have the same polarizability. In a 3D optical lattice, the $V_{AB}$ potential in Eq. (1) can be attractive for a particular relative position of two tightly-bound atoms whilst repulsive for another. Yet the lattice allows us to find a direction of propagation of the additional laser beam such that $V_{AB}(kr, \theta)$ is attractive for two atoms located at adjacent lattice sites, $k_{LR} \approx \pi$, along certain lattice axes, thus making the quasibinding effect observable, as illustrated below.

For pair binding at adjacent lattice sites to have a distinct spectral signature, the binding potential depth $|V_{AB}(r \approx \pi/k_{L})|$ should substantially exceed two parameters: (a) the linewidth for the off-resonant absorption of the additional laser, $\hbar \Gamma_{\text{S}}$, where $\Gamma_{\text{S}}$ is the resonant transition linewidth and the saturation parameter $S = \frac{2\Omega^2}{\Gamma^2 + \Gamma_{\text{S}}^2}$, with $\Omega$ being the laser Rabi frequency and $\delta = \omega - \omega_{A}$ its detuning, should satisfy $S \ll 1$, thus keeping the interacting atoms unexcited, as required by the validity of Eq. (1) [6]; (b) the heating energy $\hbar \Gamma_{\text{heat}}$ by the additional laser given by

$$\Gamma_{\text{heat}} \sim E_{\text{R}} F_{\text{Ray}}^{\text{(inelas)}} / |V_{AB}|, \quad \text{where} \quad E_{\text{R}} = \frac{h^2 k^2}{2m}$$

is the recoil energy of an atom with mass $m$, and the inelastic Rayleigh-scattering rate $F_{\text{Ray}}^{\text{(inelas)}}$ satisfies $\hbar F_{\text{Ray}}^{\text{(inelas)}} / |V_{AB}| \sim f_{\text{LD}}(\vec{k} \cdot \hat{r}) / |F_{\theta}(kr)|$, $f_{\text{LD}}(\vec{k} \cdot \hat{r})$ being the Lamb-Dicke factor of the system, which reduces to $(kr)^2 \ll 1$ in the near zone, and $F_{\theta}(kr)$ is given by (1). Conditions (a) and (b)
can be satisfied in the near-zone regime of $V_{AB}$, at $r \sim \pi/k_L \ll \pi/k$, such that $F_0(kr) \gg 1$ and $f_{LD}(\vec{r} \cdot \vec{r}) \ll 1$. This requires the additional laser to have a much longer wavelength than the lattice period, $k \ll k_L$.

We proceed with the general theoretical framework that allows one to treat atom-atom scattering in an optical lattice for any interatomic potential $V_{AB}(r)$, which is assumed to be a function of the interatomic separation $r = |\mathbf{r}_A - \mathbf{r}_B|$ only. The Hamiltonian of the two interacting atoms is $H = H_0 + V_{AB}$, where

$$H_0 = -\frac{\hbar^2}{2m_A} \nabla_A^2 - \frac{\hbar^2}{2m_B} \nabla_B^2 + U_A(r_A) + U_B(r_B), \quad (2)$$

$U_{A(B)}$ being the periodic potential of the lattice acting on atom $A(B)$.

The formation of a bound or quasi-bound pair is revealed by the analysis of the transition matrix

$$T = V_{AB}(1 - GV_{AB})^{-1} \quad (3)$$

$G$ being the unperturbed Green function (associated with $H_0$). Let $D$ be the determinant of the matrix $(1 - GV_{AB})$. When $\text{Re}(D) = 0$, a bound state (resonance with zero width) may be formed if its energy $E_b$ exceeds the total band energy of the two unperturbed atoms. A resonance may occur if $\text{Re}(D) = 0$ corresponds to an energy $E_r$ within the unperturbed total band energy. Such a resonance implies the existence of two-atom quasi-bound states in the lattice, but only if its width (its reciprocal lifetime) is given by the positive value $\Gamma_r$.

$$\Gamma_r = 2 \frac{\text{Im}(D)}{dE \text{Re}(D)}. \quad (4)$$

Information about the two-atom states can be inferred from Bragg scattering spectra, which depend on the atomic density of states in the lattice. The change in the two-atom density of states due to the interatomic interaction potential $V_{AB}$ is given by

$$\delta \rho = -\frac{1}{\pi N} \text{Im}[\frac{d}{dE} \text{Tr}(G)], \quad (5)$$

$N$ being the normalization constant. In the vicinity of a genuine resonance located at $E = E_r$ (i.e., if the conditions leading to Eq. (4) are satisfied), Eq. (5) reduces to the Lorentzian shape $\delta \rho$

$$\delta \rho = \frac{\Gamma_r / 2\pi}{(E - E_r)^2 + \Gamma_r^2 / 4}. \quad (6)$$

This expression measures the enhancement of the two-atom density of states near $E_r$, relative to that of a perfect optical lattice (without atom-atom interaction),

$$\rho^{(0)}(E) = -\frac{1}{\pi N} \text{Tr}(G). \quad (7)$$

Experimentally, however, $\Gamma_r$ will be swamped by the much larger line broadening due to the $\Gamma_{\text{heat}}$ discussed above.

We now consider that the atoms are located near the minima of the periodic potential wells of the optical lattice, for which the tight-binding approximation is valid. As the basis states, we choose the Wannier functions $\chi_n(r - \vec{R}_\mu)$ which have the property: $\int d\vec{r} \chi_n(\vec{r} - \vec{R}_\mu) H_0 \chi_n(\vec{r} - \vec{R}_\nu) = \epsilon_n(\vec{R}_\mu - \vec{R}_\nu)$, $\epsilon_n$ being the $n$th band energy eigenvalue and $\vec{R}_\mu$ the site location. The symmetrized or antisymmetrized product of the Wannier functions of the two atoms is denoted by

$$|nm; \vec{\mu}\vec{\nu}\rangle = N \chi_n(r_A - \vec{R}_\mu) \chi_m(r_B - \vec{R}_\nu) \pm \chi_n(r_B - \vec{R}_\mu) \chi_m(r_A - \vec{R}_\nu)], \quad (8)$$

where $N$ is a normalization constant, and + (-) refers to bosons (fermions). Here we assume that $\vec{\mu} \neq \vec{\nu}$, i.e., no lattice-site is occupied by more than one atom. In the plots and estimates given below only bosons are considered.

In a simple cubic (SC) lattice, $\vec{R}_\mu = (i\mu_x + j\mu_y + k\mu_z)a$, where $a$ is the lattice spacing, $\mu_i$ ($i = 1, 2, 3$) are integers and $\hat{i}, \hat{j}$ and $\hat{k}$ are the unit vectors along the X, Y and Z axes, respectively, we make use of the well-known dispersion relation for the tightly-bound energy bands $\epsilon_n(k)$

$$\epsilon_n(k) = \lambda_n(0) + 2\lambda_n(1) [\cos k_x a + \cos k_y a + \cos k_z a]. \quad (9)$$
Here $\lambda_n(0) = \int d^3r A_n^*(\vec{r} - \vec{R}_\mu) \hat{H}_0 A_n(\vec{r} - \vec{R}_\mu)$, $\lambda_n(1) = \int d^3r A_n^*(\vec{r} - \vec{R}_\mu) \hat{H}_0 A_n(\vec{r} - \vec{R}_\mu)$, and $\vec{R}_\mu = \vec{R}_\mu + \hat{u}_\mu$, $\hat{u}_\mu$ being any one of the unit vectors $\hat{i}$, $\hat{j}$ and $\hat{k}$. In the harmonic approximation of the lattice potential, $\chi_n(r)$ is the $n$th level 3D harmonic oscillator eigenfunction. Under these conditions, the unperturbed Green function can be expressed in the chosen basis as:

$$G_{nm}(\vec{R}, \vec{R}') = \langle mm'; \vec{R}' | E^+ - H_0 | nm; \vec{R} \rangle$$

$$= -iC \int_0^\infty dt \exp(itE') \left[ I_{\mu_1+}(t) I_{\mu_2+}(t) J_{\mu_3+}(t) + I_{\mu_1-}(t) I_{\mu_2-}(t) J_{\mu_3-}(t) \right], \quad E' < 3$$

$$= C \int_0^\infty dt \exp(-itE') \left[ I_{\mu_1+}(t) I_{\mu_2+}(t) J_{\mu_3+}(t) + I_{\mu_1-}(t) I_{\mu_2-}(t) J_{\mu_3-}(t) \right], \quad E' \geq 3$$

(10)

Here $\mu_{i \pm} = [\mu_i \pm \mu'_i]$, $J_n$ and $I_n$ are the $n$th order Bessel functions, $E' = \{E - \lambda_n(0) - \epsilon_m(0)\}/\lambda_n m(1)$ are the normalized energies and $C = 2N^2/(2\pi\lambda_n m(1))$, with $\lambda_n m(1) = 2\{\lambda_n(1) + \epsilon_m(1)\}$.

Let us take the circularly-polarized additional laser field to have direction cosines $(1/\sqrt{3}, 1/\sqrt{3}, 1/\sqrt{3})$ in a SC optical lattice formed by far-off resonance blue-detuned lasers. Then, for two atoms lying along either of the X, Y or Z axes, the interatomic potential in Eq. (1) becomes

$$V_{AB} = \frac{-8\alpha^2 I^2}{3c} \frac{\cos(kr/\sqrt{3}) \cos(kr)}{r}.$$

(11)

The crucial point is that for two atoms residing at the nearest lattice sites, this potential is attractive for $r \approx \pi/kL \ll \pi/k$. By contrast, if the two atoms lie along the direction $(0, \sqrt{2}/\sqrt{2}, 1)$, the corresponding potential

$$V_{AB} = \frac{2\alpha^2 I}{c} \cos(\sqrt{2kr}/\sqrt{3}) \left[ -\frac{5}{3} \cos(kr) + \frac{k \sin(kr)}{r^2} + \frac{\cos(kr)}{r^3} \right]$$

(12)

is repulsive for atoms at adjacent sites in this limit. For near-zone separations $r \approx \pi/kL \ll \pi/k$, Eq. (11) reduces to the “gravity”-like $1/r$ attractive potential. The competition of such a potential with the van der Waals (VdW) short-range repulsion (s-wave scattering) is analyzed in [17]. However, these VdW effects are irrelevant for atoms tightly bound at lattice sites separated by $100$ nm or more. We therefore can impose a cut-off separation $r_c(> r_0)$, where $r_0$ is of the order of $10^3$ Ångstroms, for calculating the low-energy collisional dynamics of the two atoms.

We shall illustrate the effect for two bosonic Li atoms that are in the lowest band ($n = m = 0$) of a SC lattice in the absence of the additional laser ($V_{AB} = 0$). The Lorentzian change in the density of states [Eq. (6)] is associated with resonance states formed (for an appropriate $V_{AB}$) within the energy range of the two unperturbed atoms $-3 < E' < 3$, that is, as long as the two atoms remain confined within the lowest band. In contrast, a genuine bound state (a resonance with zero width $\Gamma_r = 0$ in Eq. (4)) is formed at an energy exceeding that of the two unperturbed atoms in the lowest band ($E' > 3$), that is, when at least one atom populates a higher band in the presence of $V_{AB}$.

From Fig. 1 we see that for the laser propagation direction as in Eq. (11), the interatomic potential is attractive, only at $\cos \theta = \sqrt{1/3}$, for $r \approx \pi/kL \ll \pi/k$. Hence, only these atoms would contribute to the signature of a resonance (Fig. 2 - inset): the drastic increase of the two-atom density of states, i.e., the appearance of distinct peaks around $E_r$ (yet bearing in mind that $\Gamma_{\text{heat}}$ is their dominant experimental linewidth). The $E_r$ and $E_b$ in Fig.2 are shown to scale roughly linearly with laser intensity (at a fixed detuning $\delta$). For the $^6S_{1/2} \rightarrow ^6P_{3/2}$ ($852.1$ nm) transition in Cs, the recoil energy $E_R \lesssim 10$ kHz, whence conditions (a), (b) are best satisfied by a near-UV lattice, $kL \gg k$, $F_0(kr) \gg 1$, $f_{\text{LD}}(kR) \ll 1$ and an additional laser with $S \sim 10^{-4}, I \lesssim 0.1$ W/cm$^2$.

The solution of the Schrödinger equation, $H\Psi = E\Psi$ for the tightly-binding limit and in the presence of $V_{AB}$ can be written as

$$\Psi = \sum_{nm} \sum_{\mu\nu} C_{nm} |nm; \mu\nu\rangle.$$

(13)

This is a superposition of the two-atom Wannier functions (8) with coefficients $C_{nm}$ obtainable from the Lippmann-Schwinger matrix equation [16]:

$$\sum_{\mu'\nu'} |nm; \mu\nu\rangle \langle H_0 - E |nm; \mu'\nu'\rangle C_{nm}(R_{\mu}, R_{\nu}) + \sum_{\mu'\nu'} V_{nm;\mu\nu;\mu'\nu'} C_{nm}(R_{\mu'}, R_{\nu'}) = 0,$$

(14)
where $V_{nm;\mu\nu,\mu'\nu'} = \langle nm;\mu\nu | V_{AB} | nm;\mu'\nu' \rangle$. Equation (14) can be solved by using the unperturbed Green’s function Eq. (10). The solution is given by

$$C_{nm}(R_{\mu}, R_{\nu}) = C_{nm}^{(0)}(R_{\mu}, R_{\nu}) + \sum_{\mu'\nu', \mu''\nu''} G_{nm}(\mu\nu, \mu'\nu')(V_{nm;\mu\nu} V_{nm;\mu'\nu'} C_{nm}(R_{\mu''}, R_{\nu''})),$$

(15)

where $C_{nm}^{(0)}$ pertains to the uncoupled superposition of two-atom Wannier functions satisfying the equation

$$\sum_{\mu'\nu'} \langle nm;\mu\nu | (H_0 - E) | nm;\mu'\nu' \rangle C_{nm}^{(0)}(R_{\mu'}, R_{\nu'}) = 0.$$

(16)

The formation of a quasibound or resonance state gives rise to quantum translational correlations (spatial entanglement) of the two atoms. This entanglement is seen to correspond to strong deviations of the $C_{nm}$ in Eq. (15) from the uncoupled coefficients $C_{nm}^{(0)}$ at $E$ near a resonance. Its experimental manifestation would be the non-separability and redistribution of the two-atom density, as compared to the non-entangled (unperturbed) density of atoms at adjacent sites (Fig. 3).

We have concentrated here on the essential effects of laser-coupled atom pairs at adjacent sites of a moderately populated lattice, but at higher densities we can obtain multi-atom correlated arrays. A mean-field analysis reveals the possibility of a bosonic “supersolid”—a self-organized periodic array of atoms, under the influence of dipole-dipole interactions induced by a circularly-polarized plane-wave laser (without an optical lattice) [6].

To conclude, we have demonstrated the possibility of observing a novel effect: the quasi-binding or binding of cold atoms, initially residing at adjacent sites of an optical lattice, by a plane-wave off-resonant laser. Their controllable spatial entanglement may be used for matter-wave teleportation [10] and other quantum information applications. Experimentally, such giant quasimolecules can be revealed as satellite lines (with energies depending on the binding potential depth) shifted relative to the Bragg scattering spectral lines of a probe laser, analogously to excitonic lines in crystals.

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[1] C. Cohen-Tannoudji, Rev.Mod.Phys. 70, 707 (1998); S. Chu, Rev.Mod.Phys. 70, 685 (1998); W.D. Philips, Rev.Mod.Phys. 70, 721 (1998).
[2] G. Grynberg et al., Phys.Rev.Lett. 70, 2249 (1993); A. Hemmerich and T.W. Hansch, Phys.Rev.Lett. 70, 410 (1993); P.D. Lett et al., Annu.Rev.Phys.Chem. 46, 423 (1995); J. Weiner, Adv.At.Mol.Opt.Phys. 35, 45 (1995); A. Fioretti et al., Phys.Rev.Lett. 80, 4402 (1998).
[3] T. Thirunamachandran, Molecular Physics 40, 393 (1980); M.M. Burns et al., Science 249, 749 (1990); P.W. Milonni and A. Smith, Phys.Rev. A 53, 3484 (1996).
[4] D. O’Dell, S. Giovanazzi, G. Kurizki and V.M. Akulin, Phys.Rev.Lett. 84, 5687 (2000); S. Giovanazzi, D. O’Dell and G. Kurizki Phys. Rev. A 63, 031603(R) (2001).
[5] G. K. Brennen et al., Phys.Rev.Lett. 82, 1060 (1999); C. Menotti and H. Ritsch, Phys. Rev. A 60, R2653 (1999); A. M. Guzman and P. Meystre, Phys. Rev. A 57, 1139 (1996).
[6] S. Giovanazzi, D. O’Dell and G. Kurizki (preprint). Compare with D. Pines and P. Nozieres, The theory of Quantum Liquids (Benjamin, 1966).
[7] J. Callaway, Quantum Theory of the Solid State, (Academic,1974); J. Callaway, J.Math.Phys. 5, 783 (1964).
[8] G.F. Koster and J.C. Slater, Phys.Rev. 96, 1208 (1954).
[9] T. Müller-Seydlitz et al., Phys.Rev.Lett. 78, 1638 (1997).
[10] T. Opatrny and G. Kurizki, Phys. Rev. Lett. 86, 3180 (2001).
FIG. 1. The laser-induced off-resonant dipole-dipole potential for two Li atoms in their ground states (2S$_{1/2}$) as a function of $kr$, for three interatomic axes at different angles $\theta$ with the direction of propagation of the circularly polarized laser: $\cos \theta = 0$ (solid line), $\cos \theta = \sqrt{2/3}$ (dashed line), $\cos \theta = \sqrt{1/3}$ (dotted line). The laser frequency is blue-detuned by 300 $\Gamma$ from the atomic transition $2S_{1/2} \rightarrow 2P_{3/2}$ (wavelength 670.77 nm and linewidth $\Gamma/2\pi = 5.9$ MHz), $I = 5$ W/cm$^2$, $S = 10^{-4}$. Inset (top) shows the geometry of the system. The other inset shows the x-component of the periodic lattice potential $V(x) = V_0 \sin^2 k_L x$ generated by lasers blue-detuned by $10^4 \Gamma$ from the same transition ($k \simeq k_L$) with $S = 7.6 \times 10^{-5}$.

FIG. 2. Main figure: the dimensionless resonance energy $E'_r = \frac{E_r - 2\lambda_0(0)}{\lambda_0(1)}$ (marked by “+”) and the bound state energy $E'_b = \frac{E_b - 2\lambda_0(0)}{\lambda_0(1)}$ (marked by “*”) as a function of laser intensity $I$ for the parameters of Fig. 1. Here $\lambda_0(0) = 3.408$ MHz, $\lambda_0(1) = 0.98$ Hz. Inset: the drastic increase of the two-atom density of states $\delta \rho$ by 4 orders of magnitude (solid line) compared to the unperturbed scaled density of states $\rho = 10^4 \times \rho^{(0)}$ (dashed line) near the resonance $E'_r = 2$, for low laser intensity $I = 0.48$ mW/cm$^2$, $\delta = 300 \Gamma$. The resonance linewidth $\Gamma_r = 6.29$ Hz.
FIG. 3. Inset: Scattered intensity $|C(\vec{u})|^2$ as a function of laser intensity $I$ ($\delta = 300 \Gamma$). The incident atoms ($I = 0$) are assumed to be in sites (000, 111). Here solid line refers to $|C(000,111)|^2$, dashed lines to $|C(000,100)|^2$, “+” to $|C(000,110)|^2$ and dotted line to $|C(000,200)|^2$. Main figure: Two-atom wavefunction squared (times the unit cell volume $a^3$) $|\Psi(r)|^2 = \int d^3 R |\Psi(\vec{R}, \vec{r})|^2$, where $\vec{R}$ is their center-of-mass coordinate, as a function of the separation $r$ (in units of $a$) for $I = 0.48$ mW/cm$^2$ (dashed), $I = 5$ W/cm$^2$ (dotted) and $I = 0$ (solid). All adjacent sites become partially occupied by the nonseparable (entangled) atoms.