Optical binding with cold atoms

C. E. Máximo, R. Bachelard, and R. Kaiser

1 Instituto de Física de São Carlos, Universidade de São Paulo, 13560-970 São Carlos, SP, Brazil
2 Université Côte d’Azur, CNRS, INPHYNI, 06560 Valbonne, France

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Optical binding is a form of light-mediated forces between elements of matter which emerge in response to the collective scattering of light. Such phenomenon has been studied mainly in the context of equilibrium stability of dielectric spheres arrays which move amid dissipative media. In this letter, we demonstrate that optically bounded states of a pair of cold atoms can exist, in the absence of non-radiative damping. We study the scaling laws for the unstable-stable phase transition at negative detuning and the unstable-metastable one for positive detuning. In addition, we show that angular momentum can lead to dynamical stabilisation with infinite range scaling.

The interaction of light with atoms, from the microscopic to the macroscopic scale, is one of the most fundamental mechanisms in nature. After the advent of the laser, new techniques were developed to manipulate precisely objects of very different sizes with light, ranging from individual atoms [1] to macroscopic objects in optical tweezers [2]. It is convenient to distinguish two kinds of optical forces which are of fundamental importance: the radiation pressure force, which pushes the particles in the direction of the light propagation, and the dipole force, which tends to trap them into intensity extrema, as for example in optical lattices. Beyond single-particle physics, multiple scattering of light plays an important role in modifying these forces. For instance, the radiation pressure force is at the origin of an increase of the size of magneto-optical traps [3] whereas dipole forces can lead to optomechanical self-structuring in a cold atomic gas [4] or to optomechanical strain [5].

For two or more scatterers, mutual exchange of light results in cooperative optical forces, which may induce optical mutual trapping, and eventually correlations in the relative positions of the particles at distances of the order of the optical wavelength. This effect, called optical binding, has been first demonstrated by Golovchenko and coworkers [6, 7], using two dielectric microsized-spheres interacting with light fields within dissipative fluids. Since then a number of experiments with different geometries and with increasing number of scatterers have been reported, all using a suspension of scatterers in a fluid providing thus a viscous damping of the motion of the scatterers [8–15].

In this letter, we demonstrate theoretically the existence of optically bounded motion for a pair of cold atoms in the absence of non-radiative friction. We consider two atoms confined in two dimensions, e.g. by counterpropagating lasers. After introducing the model used we first derive the equilibrium positions of two atoms. We then study the scaling laws for bound states in the case of pairs of atoms without angular momentum, confronting our finding to known results of optical binding [6, 7]. We then turn to the more general situation allowing for angular momentum in the initial conditions of the atomic pairs and discuss the increased range of such a dynamically stabilized pair of atoms.

We consider a system composed of two two-level atoms of mass $m$ which interact with the radiation field. Using the dipole approximation, the atom-laser interaction is described by $H_{\text{AL}} = -\sum_{j=1}^{2} D_j \cdot E(r_j)$, where $D_j$ is the dipole operator and $E(r_j)$ the electric field calculated at the center-of-mass $r_j$ of each atom. We will describe the center of mass of each atom by its classical trajectory and for simplicity we consider here the scalar linear optics regime [16]. The equations of motion for the amplitudes of the dipoles and their positions are then given by:

\[
\dot{\beta}_j = \left( i \Delta - \frac{\Gamma}{2} \right) \beta_j - i \Omega - \frac{\Gamma}{2} G (|r_j - r_l|) \beta_l, \quad (1)
\]

\[
\ddot{r}_j = -\frac{\hbar \Gamma}{m} \text{Im} \left[ \nabla_{r_j} G (|r_j - r_l|) \beta^*_l \beta_l \right], \quad l \neq j. \quad (2)
\]

Here we assume that the atoms are confined in the $z = 0$ plane, which can e.g. be obtained by using counterpropagating waves. The strength of the atom-laser cou-
pling is given for a homogeneous laser by the Rabi frequency $\Omega$ and laser frequency $\omega_L = ck$ tuned close to the two-level transition frequency $\omega_{at}$. $\Gamma$ is the decay rate of the excited state of the atom and $\Delta = \omega_L - \omega_{at}$ the detuning between the laser frequency and the atomic transition frequency. For a single atom, a frictionless 2D motion emerges. Note that while the dipole amplitude $\beta_j$ responds linearly to the light field, for two atoms their coupling with the centers-of-mass $r_j$ is responsible for the nonlinearity emerging from Eqs.\((1-2)\). The light-mediated long-range interaction between the atoms is given, in the scalar light approximation, by the Green function

$$G(|r_j - r_l|) = \frac{e^{ik|r_j - r_l|}}{ik|r_j - r_l|}, \quad (3)$$

which is obtained from the Markovian integration over the vacuum modes of the electromagnetic field \(16\). This coupled dipole model, has been shown to provide an accurate description of many phenomena based on cooperative light scattering, such as the observation of a cooperative radiation pressure force \(17\) \(18\), superradiance \(19\) and subradiance \(20\) in dilute atomic clouds, linewidth broadening and cooperative frequency shifts \(21\) \(22\).

Central force problems, as the one described by Eqs. \((12)\), are best studied in the relative coordinate system is a conserved quantity, with no role \(23\). The angular momentum $L$ motion and the difference in dipole amplitudes thus play coupled only to the average dipole. The center-of-mass detuning, and their relative position $r_j$ get tuned close to $r_{at}$, the equilibrium points are given by the zeros of the function

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Central force problems, as the one described by Eqs. \((12)\), are best studied in the relative coordinate frame, so we define the average (and differential) positions and dipoles of the atoms. One can shown that after a transient of order $1/\Gamma$, the two atomic dipoles synchronize, independently of their distance, pump strength or detuning, and their relative position $r = r_1 - r_2$ gets coupled only to the average dipole. The center-of-mass motion and the difference in dipole amplitudes thus play no role \(23\). The angular momentum $L = mrv_\perp/2$ of the system is a conserved quantity, with $v_\perp$ the magnitude of the velocity perpendicular to the inter-particle separation $r = r\hat{r}$. This is a fundamental difference with other optical binding systems where the angular momentum is quickly driven to zero by friction. Note that the stochastic heating due to the random atomic recoil which comes from spontaneous emission recoils is neglected here.

To study bound states we first identify the equilibrium points of the system, which are given by the zeros of the equation \(23\)

$$\frac{\ell^2}{k^2} = \frac{\Omega^2}{\Gamma^2} \frac{\sin kr \frac{kr}{k^2r} + \cos kr \frac{k^2r}{k^2r^2}}{(1 + 2\sin kr \frac{kr}{k^2r} + (2\delta + \cos kr)^2) \omega}. \quad (4)$$

Note that Eq.\((4)\) depends on the pump strength $\Omega/\Gamma$, laser detuning $\delta \equiv \Delta/\Gamma$ and the dimensionless angular momentum $\ell \equiv L/2\sqrt{\hbar \Gamma m} = krv_\perp/\sqrt{32 \upsilon_{\text{Dopp}}}$. where $v_{\text{Dopp}} = \hbar k/2m$ corresponds to the Doppler temperature of the two-level laser cooling. This equation includes both stable and unstable equilibrium points. For the particular case $\ell = 0$, the equilibrium points are given by the simplified condition $\tan kr = -1/kr$ which does not depend on the light matter coupling $\Omega$ or $\Delta$, but only on the mutual distance between the atoms. The details of the light-matter interaction will however come into play when the stability of these equilibrium points is considered.

As pointed out in Ref.\((24)\), linear stability may not be sufficient to provide a phase diagram with bound states. We thus choose to study its stability by integrating numerically the dynamics, starting with a pair of atoms with initial velocities, and moving around the equilibrium point. Simulations realized with particles with an initial temperature $T = 1\mu K$, as throughout this work.
one-dimensional dynamics.

Fig. 2(a) presents the stability diagram of the $r \approx \lambda$ equilibrium point, for a pair of atoms with an initial temperature of $T = 1 \text{mK}$ (throughout this work, the conversion between temperature and velocity is realized using the $^{87}\text{Rb}$ atom mass $m = 1.419 \times 10^{-25} \text{kg}$). For zero angular momentum, the initial temperature is associated to the radial degree of freedom $T = mv_\parallel^2/2$. In the lower (black) part of the diagram (low pump strength), free-particle states are always observed after a short transient $\tau \approx 0.1 \text{ms}$, which means that the optical forces are unable to bind the atoms (see trajectory for $\delta = -2.5$ in Fig. 2(b)). For negative $\delta$, a free-particle to bounded motion phase transition occurs as the pumping strength $\Omega/T$ is increased. In the stable phase, the atoms mutual distance $r$ converges oscillating to $r \approx \lambda$, as displayed for $\delta = -1$ in Fig. 2(b). We verified that these bound states, characterized by an infinite escape time $\tau$, occur around the equilibrium points $r = n\lambda$, with $n \in \mathbb{N}$. The unstable equilibrium points are found around $(n + 1/2)\lambda$ and contains only free-particle states. We stress that the damping observed in the bound state region cannot be associated to a viscous medium as in [6]. In our case, the cooling of the relative motion for negative detuning can be understood as Doppler cooling in a multiply scattering regime [25–27].

For positive detuning ($\delta > 0$), we also observe a free-particle to bound states phase transition, symmetric to the negative detuning case (see Fig. 2(a)). However, we find that these bound states appear to be only metastable, with the pair of atoms separating on large time scales (see Fig. 2(b)). The binding time is indeed observed to grow exponentially with the detuning $\delta$. We have verified that the oscillations of an atom in the optical potential of another atom kept at a fixed position also results in an increase (decrease) of kinetic energy for positive (negative) detuning. These results of mutual heating or cooling is thus reminiscent of the asymmetry reported in multiple-scattering based atom cooling schemes [25–27], but differs from previous works on optical binding of dielectric spheres, which did not study the sign of the particles refractive index [14].

We have performed a systematic study of the free to bound states transition and identified the following scaling law for the critical temperature $T_c$ of this transition:

$$\frac{T_c}{T_{Dopp}} = \frac{\Omega^2}{\Gamma^2 + 4\Delta^2} \frac{16}{k_r},$$

with $T_{Dopp} = \hbar \Gamma^2/2k_r$ the Doppler temperature. This criterion can be obtained from the balance between the kinetic energy $E_{kin} = mk_Bv_\parallel^2/2$ and the dipole potential induced by the interference between the incident light beam and the scattered light field $V(r) = 4\hbar \Omega^2(\Gamma^2 + 4\Delta^2)^{-1} \cos(kr)/(kr)$. We recall that here we are considering zero angular motion dynamics, and the temperature is thus associated to the parallel (radial) velocity of the two atoms ($k_BT = m\langle v_\parallel^2 \rangle$). Eq. (5) is valid for atoms at large distances ($r \gg \lambda$) since for short distances, corrections due to their coupling should be included. We note that this scaling law corresponds to the law derived for dielectrics particles, where the interaction is given by $W = -\frac{1}{2}\alpha^2 E^2 k_r^2 \cos(kr)/r$ [6], where the $\alpha^2$ scaling of the polarisability is the indication of double scattering. The corresponding scaling law for two-level atoms yields a dipole potential ($\propto \Omega^2$) but with double scattering and a corresponding square dependence of the atomic polarisability, which at large detuning scales as $\alpha^2 \propto 1/\Delta^2$.

The difference to previous work lies in the fact that we do not have an external friction or viscous force, which would damp the atomic motion independently from the laser detuning. The metastable phase region is thus a novel feature for cold atoms compared to dielectrics embedded in a fluid.

A fine analysis of the transition between bound states to metastable states in Fig. 2(a) shows a small shift $\delta_0$ compared to the single atom resonance condition. The origin of this shift can partially be understood by the cooperative energy shift of $\cos(kr)/2kr$ for the two-atom state at the origin of the dynamics for these synchronized dipoles. We also identified an additional velocity dependence beyond this effect, with a total shift scaling as $\delta_0 \approx -\frac{\cos(kr)}{2kr} - \frac{kv_\parallel(r)}{\Gamma}$. An additional novel feature emerging from the frictionless nature of the cold atom system is the conservation of the angular momentum during the dynamics. This leads to a striking difference to optical binding with dielectric particles as we can obtain rotating bound states. Examples of such states are shown in Fig. 3: the pair of atoms can reach a rotating bound states with fixed interparticle distance on resonance (see Figs. 3(a) and (b)), but it may also support stable oscillations along the two-atom axis far from resonance (see Figs. 3(c) and (d)), analogue to a molecule vibrational mode. As in the 1D case, the atoms may remain coupled for long times, before eventually separating (see Figs. 3(e) and (f)). As can be seen in Eq. (4), for $\ell \neq 0$ the stationary points are circular orbits in the plane $z = 0$ instead of fixed points. We note that high values of angular momentum $\ell$ strongly modifies the equilibrium point landscape, suppressing the low-$r$ equilibrium points [23].

Rotating bound states are characterized by both radial $v_\parallel$ and tangential $v_\perp$ velocities, the latter being associated to the conserved angular momentum. For simplicity, we focus on initial states of atoms with purely tangential and opposite velocities: $T = mv_\perp^2/2$, neglecting thus any initial radial velocity, although it may appear dynamically. A phase diagram for the $r \approx \lambda$ and $\ell = 0.09$ (corresponding to an initial temperature $T = 1 \text{µK}$) states is presented in Fig. 4, where the escape time $\tau$ has been computed following the same procedure as before. Let us first remark that the purely-bounded to metastable tran-
FIG. 3. Rotating states for the pair of atoms for (a–b) Fixed inter-particle distance, (c–d) Vibrational mode and (e–f) Metastable state. As the orbits of both atoms coincides in (a), only a single orbit appears displayed. Simulations realized with $\ell = 0$, $\Omega/\Gamma = 0.25$ and $T = 1\mu K$, where the $^{87}\text{Rb}$ atom mass was adopted.

sition is not delimited by a sharp transition anymore, in contrast to the $\ell = 0$ case. For $\ell \neq 0$, this energy shift varies nonlinearly according to the field pump strength $\Omega/\Gamma$, allowing dynamically stable bound states for $\delta > \delta_0$, including the resonant line. The stable-metastable phase transition for $\ell \neq 0$ covers a larger part of the phase diagram, so the introduction of an angular momentum in the system allows to reach bound states for ranges of parameters where they do not exist at $\ell = 0$.

A more systematic investigation of the stability of states with different angular momentum and inter-particle distance, leads to the following scaling law for the $\ell \neq 0$:

$$\frac{T_c}{T_{\text{Dopp}}} = \frac{8}{\pi} \frac{\Omega^2}{\Gamma^2 + 4\Delta^2},$$

(6)

where the temperature is now related to the tangential velocity as $Tk_B = m\langle v^2 \rangle$. In excellent agreement with simulations, Eq. (6) differs substantially from the $\ell = 0$ case in that the distance between the atoms has disappeared. This peculiar effect originates in the fact that while the kinetic energy is initially associated to the rotan-}

tional degree of freedom, the conservation of the angular momentum implies that over a displacement of $\delta r = \lambda/2$ necessary to escape the radial potential well, only a portion of the kinetic energy $\delta E = m\langle v^2 \rangle L^2 \delta r/2r$ is converted to the radial degree of freedom. As an extra consequence of this constraint, the diagram of the $\ell \neq 0$ case presents lifetimes for the metastable states which are much longer than for the $\ell = 0$. Thus, the presence of a conserved angular momentum strongly promotes the stability of the system, and is particular promising for the optical stabilization of macroscopic clouds. This dynamical stabilization of optical binding in frictionless media is a important novel feature since it opens the possibility of enhanced long range and collective effects in cold atoms and beyond.

In conclusion, our study of optical binding in cold atoms has allowed to recover the prediction of optical binding of dielectric particles for negative detuning, where the viscosity of the embedding medium is replaced by a diffusive Doppler cooling analogue. For positive detunning, we find a metastable region, as Doppler heating eventually leads to an escape of the atoms from the mutually induced dipole potential. We also identified a novel dynamical stabilization with rotating bound states. We have shown that pairs of cold atoms can exhibit optically bound states in vacuum. The absence of non-radiative damping in the motion allows for a new class of dynamically bound states – a phenomenon not present in other optical binding setups. While this demonstration of optical binding for a pair of particles paves the way for the study of this phenomenon on larger atomic systems, the
generalization of these peculiar stability properties will be an important issue to understand the all-optical stability of large clouds.

One interesting generalization is the study of optical forces in astrophysical situations. Whereas radiation pressure forces are well studied and participate for instance in the determination of the size of a star, dipole forces are often neglected [28]. The possibility of trapping a large assembly of particles in space would allow to consider novel approaches in astrophysical imaging and could shed additional light on the motion of atoms such as the abundant hydrogen around high intensity regions of galaxies, where even small corrections to the pure gravitational attraction might be important [30].

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which yields the following dynamical equations:

\[
\dot{b} = -\frac{\Gamma}{2} \left[ 1 - \frac{\sin kr}{kr} - i \left( 2\delta - \frac{\cos kr}{k^2 r} \right) \right] b, \quad (S-5)
\]

\[
\dot{B} = -\frac{\Gamma}{2} \left[ 1 + \frac{\sin kr}{kr} - i \left( 2\delta + \frac{\cos kr}{k^2 r} \right) \right] B - i\Omega, \quad (S-6)
\]

\[
\dot{r} = \frac{2L^2}{m r^3} - \frac{\Gamma \hbar k}{2m} \left( \frac{\sin kr}{kr} + \frac{\cos kr}{k^2 r^2} \right) \left( 4|B|^2 - |b|^2 \right)^{\frac{1}{2}}, \quad (S-7)
\]

\[
\dot{L} = \frac{d}{dt} \left( \frac{mr^2}{2} \phi^2 \right) = 0, \quad (S-8)
\]

where the center-of-mass dynamics of the two-atom system has naturally decoupled from all equations.

The decay of the relative dipole \( b \), given by Eq. (S-5), is given by \( \Gamma \), independently of the pump strength or of the inter-atomic distance. Once \( b \) becomes negligible, the two atoms are synchronized, and their dipole is given by the average dipole \( B \). This synchronized mode has an energy \(-\cos kr/2kr\), relative to the atomic transition, and its decay rate is \( 1 + \sin(kr)/kr \). Nevertheless, the equilibrium points which emerge from the atom dynamics, at \( r \approx n\lambda \ (n \in \mathbb{N}) \), makes that this rate is approximately \( \Gamma \), so no superradiant or subradiant behaviour is expected.

### Derivation of the equilibrium condition Eq.(4)

As there is no source term Eq. (S-5), the relative dipoles coordinate \( b \) vanishes on a time scale of the order of \( 1/\Gamma \), which is very fast compared to the atomic motion. Therefore, the atoms can be considered always synchronized and the inter-atomic dynamics \( r \) couples only to the average dipole \( B \). It is thus possible to write

\[
B = \beta_1 = \beta_2 \equiv \beta e^{i\chi}, \quad (S-9)
\]

with \( \beta \geq 0 \) and \( \chi \in \mathbb{R} \). Then, the set of dynamical equations reduces to:

\[
\dot{\beta} = -\frac{\Gamma}{2} \left( 1 + \frac{\sin kr}{kr} \right) \beta - \Omega \sin \chi, \quad (S-10)
\]

\[
\dot{\chi} = \Delta + \frac{\Gamma \cos kr}{2 kr} - \frac{\Omega \cos \chi}{\beta}, \quad (S-11)
\]

\[
\ddot{r} = \frac{2L^2}{m r^3} - \frac{2\Gamma \hbar k}{m} \left( \frac{\sin kr}{kr} + \frac{\cos kr}{k^2 r^2} \right) \beta^2. \quad (S-12)
\]

The equilibrium points of these equations are given by setting \( \dot{\beta} = \dot{\chi} = 0 \):

\[
\beta_0 = 2\frac{\Omega}{\Gamma} \left[ \left( 1 + \frac{\sin kr}{kr} \right)^2 + \left( 2\delta + \frac{\cos kr}{k^2 r} \right)^2 \right]^{-\frac{1}{2}}, \quad (S-13)
\]

\[
\chi_0 = \arctan \left( -\frac{1 + \frac{\sin kr}{kr}}{2\delta + \frac{\cos kr}{k^2 r}} \right). \quad (S-14)
\]

which, combined with \( \ddot{r} = 0 \), gives the equilibrium condition for the interatomic separations, Eq.(4) in the main text. If one redefines Eq.(4) as

\[
F \equiv \frac{\ell^2}{k^3 r^3} - \frac{\Omega^2}{\Gamma^2} \left( \frac{\sin kr}{kr} + \frac{\cos kr}{k^2 r^2} \right)^2 = 0, \quad (S-15)
\]

the equilibrium points are obtained from the zeros of \( F \) (see Fig. 5). Fig. 5(a) illustrates the dependence of \( F \) on the angular momentum and on the detuning, showing that zeros of \( F \) tend to disappear as we get farther from resonance or at larger angular momentum.

**FIG. 5.** Two-atoms equilibrium distances pattern for different \( \ell \) (a) and different detunings (b).

**r \approx 2\lambda \ equilibrium point**

In Fig. 6 the phase diagram of the \( r \approx 2\lambda \) equilibrium point is shown: The pair of atoms presents a higher threshold in pump strength to become stable, in order to compensate for their weaker interaction at a larger distance.
FIG. 6. Stability diagram for the equilibrium point around $r \approx 2\lambda$. 