Controlled ripple texturing of suspended graphene membrane due to coupling with ultracold atoms

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We explore the possibility to create hybrid quantum systems that combine ultracold atoms with graphene membranes. We investigate a setup in which a cold atom cloud is placed close to a free-standing sheet of graphene at distances of a few hundred nanometers. The atoms then couple strongly to the graphene membrane via Casimir-Polder forces. Temporal changes in the atomic state of the atomic cloud changes the Casimir-Polder interaction, thereby leading to the creation of a backaction force in the graphene sheet. This setup provides a controllable way to engineer ripples in a graphene sheet with cold atoms.

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With the advances in trapping and coherently manipulating clouds of ultracold atoms near microstructured solid-state surfaces [1], the possibility of constructing hybrid atom/solid-state quantum systems have attracted considerable attention (see, e.g., Refs. [2–5]). Such a hybrid system would consist of ultracold atoms that can be manipulated by laser light, and a solid-state system that could for instance be controlled by electrical currents. The influence of the solid-state substrate on the atomic dynamics is well established; dispersion potentials [6] and their consequences such as quantum reflection [7] and line shifts [8] are sufficiently well understood. However, a backaction of the atom cloud on the solid-state system is rather challenging. If atoms are regarded as mechanical oscillators [4], the impedance mismatch due to the large mass difference between a single atom and a mechanical oscillator limits the atom–surface coupling. The routes that have been taken so far to alleviate this discrepancy are either to select a subsystem within the solid-state device [4], to decrease the effective size of the macroscopic system [9,11], or to enhance the coupling of ions to a membrane via resonant modes of an optical cavity [2,5]. It has been proposed that laser light could be used to couple also the motion of ultracold trapped atoms to the vibrational modes of a mechanical oscillator. In recent experiments, using magnetic [11] or surface–force coupling [12], atoms are used to study vibrations of micromechanical oscillators. In Ref. [13], the backaction of the atoms onto the oscillator vibrations as well as the effect of the membrane vibrations onto the atoms were observed.

Our goal is to find a coupling mechanism between ultracold atoms and a solid-state system that is strong enough to provide a mutual interaction between them. In this article we show that the Casimir–Polder force [6], a dispersion interaction that is due to quantum fluctuations of the electromagnetic field, can provide precisely that. The Casimir-Polder force on an atom in thermal equilibrium with its environment is typically attractive; however, in out-of-equilibrium situations such as for atoms prepared in energy eigenstates the sign of that force can be reversed [14] and can reach extremely large values for highly excited (Rydberg) atoms [15]. Any cycling transition between the ground state and an excited state thus translates into an oscillating dispersion force. The coupling could be increased by minimizing the impedance mismatch using oscillators with low mass such as carbon nanotubes or graphene membranes. Freely suspended graphene crystals can exist without a substrate, suspended graphene flakes or scaffolds have been observed for single layers and bilayers [16]. Suspended graphene membranes can be created with diameters that are comparable to the diameter of a Bose–Einstein condensate. Free-standing graphene membranes have a key advantage over bulk systems studied in previous works as the membrane can be cleaned from adsorbates by passing a current through it [17].

In the resulting hybrid quantum system, driving the atomic cloud to excited states could be used to engineer ripples on a graphene membrane. Ripples are an intrinsic feature of graphene sheets which influence their electronic properties; perturbations to nearest neighbour hopping might cause the same effects as inducing an effective magnetic fields and changing local potentials [18–20]. The ability to control ripple structures could allow a device design based on local strain and selective bandgap engineering [21]. The possibility of constructing an all-graphene circuit, one of the big goals in graphene science, could be achieved by applying the patterning of different devices and leads by means of appropriate cuts in the sheet. In Ref. [19] it has been proposed to deposit graphene onto substrates with regions that can be con-
trollably strained on demand, or by exploring substrates with thermal expansion heterogeneity; the generation of strain in the graphene lattice is then capable of changing the in-plane hopping amplitude in an anisotropic way. Controlled ripple texturing using both spontaneously and thermally generated strains was first reported in Ref. [22], where the possibility was shown to control ripple orientation, wavelength and amplitude by controlling boundary conditions and making use of graphene’s negative thermal expansion coefficient.

In the following, we evaluate the Casimir-Polder force between a single graphene sheet and a rubidium atom in various energy eigenstates and determine the minimal number of atoms needed to excite a ripple. For the calculation of the interaction potential we assume the sheet to be infinitely extended, thereby neglecting possible effects that may arise from the finite extent of the flake. For planar structures, the Casimir-Polder potential of an atom in an energy eigenstate $|n\rangle$ at a distance $z_A$ away from the macroscopic body with permittivity $\varepsilon(\omega)$ can be written as [6]

$$U_{CP}(z_A) = \frac{\hbar \mu_0}{8\pi^2} \int_0^\infty d\xi \xi^2 \alpha_n(i\xi) \int_0^\infty dk_z e^{-2k_z\gamma_0 z_A} \gamma_0 z_A$$

$$\times \left[ R_{\text{TE}} + R_{\text{TM}} \left( 1 - \frac{2k_z^2 \gamma_0 z_A^2}{\xi^2} \right) \right]$$

$$+ \frac{\mu_0}{4\pi} \sum_{k \neq n} \omega_n^2 d_{nk} \otimes d_{kn} \int_0^\infty d\kappa_0 e^{-\kappa_0 z_A} \times \text{Re} \left[ R_{\text{TE}} + R_{\text{TM}} \left( 1 + \frac{2\kappa_0^2 \omega_n^2}{\omega^2} \right) \right], \quad (1)$$

where $\gamma_{iz} = \sqrt{1 + \varepsilon(\omega)\xi^2/(c^2 k_A^2)}$, $\kappa_0 z_A = \sqrt{k_B^2 + \omega^2/c^2}$ and $\alpha_n(\omega)$ is the atomic polarizability defined by

$$\alpha_n(\omega) = \lim_{\varepsilon \rightarrow 0} \frac{2}{\hbar} \sum_{k \neq n} \omega_k d_{nk} \otimes d_{kn} \omega_k^2 - \omega^2 - i\omega \varepsilon \quad (2)$$

The first term in Eq. [1] describes the nonresonant part of the Casimir-Polder potential, recognizable by the integration along the imaginary frequency axis, $\omega = i\xi$, whereas the second term is related to resonant photon exchange between the atom and the graphene sheet. Equation [1] is strictly valid only at zero temperature. However, we can assume that a potential experiment with ultracold atoms could be performed at sufficiently low temperatures for thermal excitations to only play a subordinate role; in addition, the distance of those atoms from the graphene sheet will be much smaller than the thermal wavelength $\lambda_T = \hbar c/(k_B T)$. In situations in which either assumption fails to hold, a replacement of the frequency integral by a Matsubara sum,

$$\frac{\hbar}{\pi} \int_0^\infty d\xi f(\xi) \rightarrow 2k_B T \sum_{n=0}^\infty \left( 1 - \frac{1}{2} \delta_{mn} \right) f(i\xi_n), \quad (3)$$

with Matsubara frequencies $\xi_n = n2\pi k_B T/h$ [14], has to be employed. The material properties of graphene enter via their reflection coefficients $R_{\text{TE}}$ and $R_{\text{TM}}$. Due to graphene’s unique electronic structure, a full calculation of its electromagnetic reflection coefficients is in fact possible from first principles. Following Ref. [23], in order to derive the reflection coefficient of a graphene sheet, the dynamics of quasiparticles are described within the $(2+1)$-dimensional Dirac model. The quasiparticles in graphene obey the linear dispersion law $\omega = v_F k$, where $v_F \approx c/300$ is the Fermi velocity valid for energies below 2 eV [24]. More elaborate models for the conductivity are not needed here because at frequencies above the dominant atomic transitions the polarizability $\alpha_n(i\xi)$ does no longer contribute to the integral in Eq. [1].

Thermal corrections become important only for $k_B T \gtrsim \Delta$, where $\Delta$ is the gap parameter of quasiparticle excitations [25]. At finite temperature, the potential is well approximated by inserting the temperature-dependent reflection coefficients in the lowest term in the Matsubara sum ($j = 0$) while keeping the zero-temperature coefficients for all higher Matsubara terms. However, it has been shown in Ref. [26], that at room temperature the static value of the polarizability increases only by 10 percent. The TM reflection coefficient increases by one percent due to finite temperature and the TE coefficient vanishes altogether at zero frequency. For this reason and with the similar approach in Ref. [27] in mind, we have used the zero-temperature limit of the polarizability as a very good approximation.

The interaction of the quasiparticles with external electromagnetic fields can be described within this Dirac model. From the boundary conditions of the electric and magnetic fields one finds the reflection and transmission coefficients given the specific values of mass gap $m$ and chemical potential $\mu$. For simplicity we will set $m = \mu = 0$ (perfect Dirac cone) for which the difference between this approximation for suspended graphene samples ($m, \mu \sim 0.01 eV$) is less than 1% [23]. One then arrives at the reflection coefficients of a free standing graphene sheet in vacuum as

$$R_{\text{TM}} = \frac{4\pi\alpha}{\pi k_B^2 + k^2} \frac{\sqrt{k_B^2 + k^2}}{\sqrt{k_B^2 + k^2} + 8 \sqrt{k_B^2 + \hat{v}^2 k^2}}, \quad (4)$$

$$R_{\text{TE}} = -\frac{4\pi\alpha}{\pi k_B^2 + \hat{v}^2 k^2} \frac{\sqrt{k_B^2 + \hat{v}^2 k^2}}{\sqrt{k_B^2 + \hat{v}^2 k^2} + 8 \sqrt{k_B^2 + k^2}}, \quad (5)$$

where we defined $k_B^2 = \xi^2/c^2$ and $\hat{v} = v_F/c = (300)^{-1}$; $\alpha = 1/137$ is the fine structure constant.

For a ground-state rubidium atom the force $F_{\text{CP}}(z_A) = -\nabla_A U_{\text{CP}}(z_A)$ at $z_A = 200$ nm is rather small — $F_{\text{CP}}(5 S_{1/2}) = -1.05 \times 10^{-22}$ N. The atom–surface coupling can be vastly enhanced by promoting atoms to highly excited Rydberg states, i.e. states with very high principal quantum number $n$. 
TABLE I: Casimir-Polder force between rubidium atoms in different atomic states and graphene sheet and the number of atoms needed to excite a ripple of 1 nm amplitude.

| Atomic State | $T = 0K$ | $T = 300K$ |
|--------------|--------|--------|
|               | $F_{CP}(N)$ | $N_{atoms}$ | $F_{CP}(N)$ | $N_{atoms}$ |
| $26S_{1/2}$  | $2.29 \times 10^{-16}$ | 70 | $-1.89 \times 10^{-15}$ | 9 |
| $29S_{1/2}$  | $3.72 \times 10^{-16}$ | 43 | $-4.08 \times 10^{-15}$ | 4 |
| $32S_{1/2}$  | $5.72 \times 10^{-16}$ | 28 | $-8.15 \times 10^{-15}$ | 2 |
| $34S_{1/2}$  | $7.47 \times 10^{-16}$ | 22 | $-1.25 \times 10^{-14}$ | 2 |

Z = 200 nm

| Atomic State | $T = 0K$ | $T = 300K$ |
|--------------|--------|--------|
|               | $z_{A}$ = $z_{min}(n)$ | $z_{A}$ = $z_{min}(n)$ |
| $26S_{1/2}$  | $8.88 \times 10^{-15}$ | 2 | $-7.36 \times 10^{-14}$ | 1 |
| $29S_{1/2}$  | $6.04 \times 10^{-15}$ | 3 | $-6.65 \times 10^{-14}$ | 1 |
| $32S_{1/2}$  | $4.25 \times 10^{-15}$ | 4 | $-6.05 \times 10^{-14}$ | 1 |
| $34S_{1/2}$  | $3.41 \times 10^{-15}$ | 5 | $-5.69 \times 10^{-14}$ | 1 |

The primary motivation for the study of Rydberg atoms is to take advantage of the unique opportunities afforded by their exaggerated properties which make them extremely sensitive to small-scale perturbations of their environment and to dispersion forces. A second important aspect of atoms in Rydberg states is their regularity [28]. For example, the free-space radiative lifetime of high-lying Rydberg states increases as $n^3$. The influence of macroscopic bodies modifies those atomic relaxation rates [9]. Earlier results [15] showed a strong enhancement in the transition rates of Rydberg atoms near a surface. Intimately connected (via a Hilbert transform or Kramers–Kronig relation) to the lifetime is the dispersive energy shift that a Rydberg atom experiences in the vicinity of a macroscopic body (Casimir-Polder shift) [14] or in close proximity of another atom (van der Waals shift). An important consequence of the latter is the Rydberg blockade effect that prevents multiple Rydberg excitations within a volume of radius $a_{block}$, the blockade radius [29].

In Table I we show numerical values of the Casimir-Polder force acting on rubidium atoms. One observes that the force is attractive for ground-state atoms, but repulsive for highly excited atoms. This is due to the increased contributions of the resonant Casimir-Polder force associated with real-photon transitions as opposed to the nonresonant force components due to virtual-photon exchanges. A handy feature of Rydberg atoms is the tunability of their interaction strength by choosing a particular Rydberg state [28][30]. The excitation into Rydberg states with principal quantum numbers ranging from $n = 20$ up to the ionization threshold are typically accomplished by a two-photon excitation scheme (for an experimental review see [30]). Positioning the atom cloud at a fixed distance away from the surface, one can then excite atoms to the desired Rydberg state. The backaction of the atoms, mediated by the Casimir-Polder force, onto the membrane will be a periodic bending force $F_b$. Thus, when driving an atom from its ground state to a Rydberg state and back, one cycles between an attractive (when the atom is in the ground state) and a repulsive interaction (when the atom is in the Rydberg state) between atom and graphene sheet. In addition it is well known that a free-floating graphene sheet would always crumple at room temperature, hence the need to support the graphene sheet by a substrate. At very low temperatures, the graphene membrane experiences a combination of the following forces: (a) the substrate–pinning force $F_{pin}$ that prevents the graphene membrane from sliding and (b) the bending force $F_b$ due to the Casimir-Polder potential, see Fig. 1. Measurements on layered graphene sheets of thickness between 2 and 8 nm have provided spring constants that scale as expected with the dimensions of the suspended section, and range from 1 to 5 N/m [31]. Other experiments studied the fundamental resonant frequencies from electromechanical resonators made from graphene sheets [32]. For mechanical resonators under tension $T$ the fundamental resonance mode $f_0$ is given by

$$f_0 = \left(\frac{A}{\rho L^2} - 2A^20.57\frac{T}{\rho L^2 w t}\right)^{1/2}$$

where $E$ is Young’s modulus, $\rho$ is the mass density; $t, w, L$ are the thickness, width and length of the suspended graphene sheet and $A$ is a clamping coefficient ($A$ is equal to 1.03 for doubly clamped beams and 0.162 for cantilevers). The effective spring constant of the fundamental resonance mode is given by $\kappa_{eff} = m_{eff}\omega^2$, where $m_{eff} = 0.735Lwt\rho$ [32]. In the limit of vanishing tension, the fundamental resonance mode is $f_0 = A\sqrt{E/\rho t/L^2}$. However, we have to assume a finite value for the tension, for which we choose $T = 0.1$ nN. Tension between graphene and trenches is a random process depending on the production technique and the interaction with the substrate and for that reason very difficult to control [32]. Using the known values for bulk graphite $\rho = 2200$ kg/m$^3$ and $E = 1.0$ TPa, for a graphene can-
tile with \( t = 0.3 \text{ nm}, L = 3 \mu\text{m} \) and \( w = 2 \mu\text{m} \) the force needed to create a curvature on graphene with 1 nm amplitude is approximately 16 fN. In order to create a force necessary to produce a ripple of a determined amplitude — AFM imaging measures amplitudes in graphene sheets from 0.7 to 30 nm — one has to excite a particular number \( N \) of atoms from the cloud.

Upon inspection of Table II one observes that, for a cloud of cold \(^{87}\text{Rb}\) atoms at a fixed distance of 200 nm from the graphene membrane, one would need to excite one or more atoms in order to create a ripple with 1 nm amplitude. With the blockade radius \( a_m \) of several micrometers between neighbouring Rydberg atoms only a limited number of atoms can be resonantly excited to Rydberg states at the same time. If larger numbers of Rydberg excitations are needed, the laser line width has to be chosen large enough to bridge the van der Waals shift between two neighbouring Rydberg atoms. This is easily achieved by pulsed-laser excitation with typical line widths of several 10–100 MHz.

The interplay between atom–surface distance and principal quantum number \( n \) is of crucial importance in this process. For fixed atom–surface distance, the interaction increases with \( n \) so that fewer atoms are needed to induce a desired ripple amplitude. However, due to its increasing size, there is a limit to how close a Rydberg atom can possibly be brought to a surface, or to what Rydberg state an atom at a given distance can be excited. This limiting distance can be estimated simply from the classical atomic radius as \( z_{\text{min}}(n) \geq \sqrt{5} n^2 a_B \) where \( a_B \) is the Bohr radius and the numerical factor \( \sqrt{5} \) has been chosen to ensure that its wavefunction does not overlap with the surface. We see that the number of atoms decreases when placed at their minimal distance and, at finite temperature, this number may decrease to only one atom needed to create a 1 nm ripple.

An estimate of the number of atoms in a given Rydberg \( n \) state needed to generate a ripple with amplitude 1 nm can thus be obtained as follows. From Ref. [15] we know that the Casimir–Polder force in the non-retarded limit scales as \( F_{\text{CP}} \propto n^4/z^4 \). We then equate the necessary number of atoms to generate a force of, say 16 fN, by using the scaling law on a particular reference state, say 32S from Table I at zero temperature. Together with the constraint on distance, \( z_{\text{min}}(n) \geq n^2 (0.118 \text{ nm}) \), this yields a lower bound on the number of required atoms as \( N_{\text{min}}(n) \geq 3.6 \times 10^{-6} n^4 \). This result seems counterintuitive at first in the sense that excitation to higher Rydberg states does not seem to increase the force and lower the number of required atoms. This is due to the competition of increasing force at fixed distance and larger minimal separation with increasing \( n \). Numerical values for an estimate of the number of atoms needed to be held at their respective minimal distances are provided in Table II.

Realization of the proposed setup requires placing and controlling an atom very close to a surface. Achieving such control is challenging because atom-surface forces are comparable with typical trapping forces for cold atoms in this regime. Atomic ensembles have been stably trapped using magnetic traps formed by patterned electrodes at distances of 500 nm from a surface [12, 33] and down to 215 nm by using optical dipole traps based on evanescent waves [34, 35]. In Ref. [36], a tightly focused optical tweezer is used to achieve a minimum trap distance of about 100 nm for realistic laser intensities.

In conclusion, we have shown that it is possible to construct hybrid quantum systems consisting of cold atoms and solid devices in which a very small number of atoms exert influence on a much larger object. Here we investigated the creation of ripples in a graphene membrane due to laser-controlled atom–surface interactions. Because atoms in different quantum states, in particular highly excited Rydberg states show vastly different interaction strengths, the modification of the Casimir–Polder potential creates an effective force on the graphene sheet. This ability to control and manipulate ripples opens up a number of novel research possibilities such as the investigation of the effects of ripples on graphene’s electrical and optical properties.

The key idea in quantum emulators setups with cold gases (bosons, fermions or mixtures) is to control and simulate other systems of interest, based on the universality of quantum mechanics. Atom-light interaction can be used to generate artificial gauge potentials acting on neutral atoms [37]. In the same way, by tailoring ripples in graphene via Casimir–Polder forces is introducing the same effects onto graphene as those induced by an effective magnetic fields, similarly creating an artificial gauge potential. This technique also provides a route towards coherent manipulation of atom-graphene systems. For example, an atom in a coherent superposition of ground and (highly) excited states leaves the sheet in a similar superposition of curvatures, thus providing an effective backaction between cold atoms and a solid-state system that leaves the hybrid system potentially in an entangled state. We expect such quantum effects only to be achievable for amplitudes smaller than 1 nm which have been shown to exist [38]. The advantage of smaller ripples is also the lower number of atoms for their excitation. Another major advantage of using such a hybrid system is the fact that we could do a true non-destructive quan-

| Atomic State \( |n\rangle \) | \( z_{\text{min}}(n) \) | \( N_{\text{min}}(n) \) |
|----------------|-----------------|------------------|
| \( |23 \text{S} \rangle/2 \) | 62 nm | 1 |
| \( |30 \text{S} \rangle/2 \) | 106 nm | 3 |
| \( |36 \text{S} \rangle/2 \) | 153 nm | 6 |
| \( |43 \text{S} \rangle/2 \) | 218 nm | 12 |

TABLE II: Minimal number of atoms required to generate a ripple with 1 nm amplitude. The atoms are assumed to be held at their respective minimal distances at zero temperature.
tum measurement of the atomic state by testing only the graphene sheet.

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