Mesoscopic Molecular Ions in Bose-Einstein Condensates

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We study the possible formation of large (mesoscopic) molecular ions in an ultracold degenerate bosonic gas doped with charged particles (ions). We show that the polarization potentials produced by the ionic impurities are capable of capturing hundreds of atoms into loosely bound states. We describe the spontaneous formation of these hollow molecular ions via phonon emission and suggest an optical technique for coherent stimulated transitions of condensate atoms into a specific bound state. These results open up new possibilities for manipulating tightly confined ensembles.

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The experimental realization of Bose-Einstein condensation (BEC) in atomic samples [1] have stimulated a new wave of theoretical and experimental studies of degenerate systems in the dilute and weakly interacting regime. A broad range of techniques from atomic physics and optics have allowed the accurate manipulation of ultracold samples. In particular, various properties of degenerate gases have been explored, such as their excitation modes [2], superfluidity [3,4], and controlled vortex creation [5]. Excitations in the phonon regime and the dynamic structure factor were probed using Bragg spectroscopy [6]. Recent studies are probing ultracold atomic systems in which electric charges may play an important role [7]. These include ultracold plasmas [8], ultracold Rydberg gases [9], as well as ionization experiments in BEC [10].

In this Letter, we explore the behavior of a dilute atomic condensate doped with ionic impurities. We show that the polarization interaction leads to the capture of large numbers of atoms into weakly bound states, resulting in the rapid formation of mesoscopically large molecular ions. We study the spontaneous dynamics of the molecular ion formation and show that the degenerate nature of the condensate, as well as the properties of collective excitations (phonons), play an important role. We further describe a coherent optical technique to prepare molecular ions in specific states. Besides the fundamental interest of studying the formation of such large many-body objects, the effects described here may open up new ways to manipulate cold atoms. In particular, the charged and tightly trapped atomic cloud represents a microtrap that could be manipulated and ''transported'' by external fields. Controlled mechanisms to manipulate tightly confined, strongly interacting atoms may also allow for new approaches for quantum information processing and for studies of other phenomena such as quantum phase transitions. We note an analogy to early studies involving electron bubbles and ion ''snowballs'' in superfluid helium [11]. However, a dilute weakly interacting atomic BEC favors the formation of large mesoscopic ions in highly excited and metastable states (see below).

We consider the situation in which few ions with ultralow kinetic energy are introduced into an atomic BEC. This can be realized, e.g., by rapidly ionizing atoms from the BEC using lasers in a process where the ejected electrons carry essentially all the kinetic energy. This would leave the BEC doped with few ions, and the BEC-ion system in a nonequilibrium state. Alternatively, one could introduce charged impurities via controlled processes involving a combination of ion and atom traps, or using surface traps on semiconductor surfaces. As a relaxation process, large numbers of BEC atoms can be captured into loosely bound states of the polarization potentials, rapidly forming shells of atoms around ions. Such a process occurs spontaneously through collisions of condensate atoms in which atoms are stimulated down into the molecular ion bound state, and the excess energy is carried away by the condensate collective excitations.

We are interested in the limit $T \to 0$, and for simplicity, we consider a homogeneous BEC with the neutral gas being the parent atom of the doping ion [12]. The ion will polarize a nearby atom (separated by a distance r) and the two will interact via a polarization potential behaving asymptotically as $-C_4/2r^4$ [13], where C_4 is the dipole polarizability of the neutral atom.

Under the conditions mentioned above, two types of scattering processes involving the ion are possible: elastic and superelastic collisions. The energy of the colliding partners is not changed in the first type, while kinetic energy is released in the second type. Elastic scattering of an atom and an ion is often described as the contributions from two separate processes [7,14]: resonant charge transfer (the charge changes center), and pure elastic scattering (the charge remains with the same center). According to Wigner's threshold laws [15], the rate of all elastic processes vanishes as $T \to 0$. The second process, superelastic scattering, corresponds to an inelastic process where one BEC atom is captured by the ion and where kinetic energy is released via phonon emission. Here, BEC atoms are accelerated by the ionic field [see Fig. 1(a)], and after the collision, one of the neutral atoms is captured by

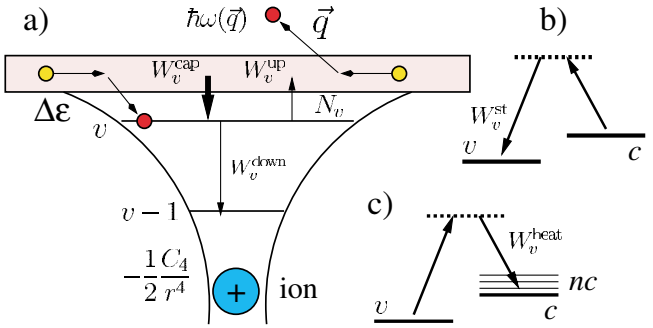

FIG. 1 (color online). Diagrams of atom capture by an ion. In (a), the spontaneous capture in level υ is followed phonon emission (with corresponding rates). In (b), the stimulated rate from the condensate c to the bound level υ, also may produce heating, i.e., population of noncondensate atoms nc, in (c).

the polarization potential, and the kinetic energy released is shared by the remaining free condensate atoms and the newly formed molecular ion [16]. Inelastic collisions with excitations of BEC collective modes usually cannot be decribed by simple binary collisions, unless in the limit where the binding energy of the uppermost bound level of the molecular ion is much larger than the BEC chemical potential. The rate for superelastic processes tends to a constant at zero temperature [15], and this scattering process will be dominant at $T \to 0$. Contrary to the slowdown of impurities in condensates, where phonon radiation is not possible below some critical velocity (because of momentum-energy conservation [4]), the capture of atoms by the long range potential created by the ion corresponds to free-bound transitions, and does not suffer from this restriction: phonon emission takes place at any velocity.

We describe the evolution of the number of atoms N_υ in the bound level υ of the polarization potential by a kinetic equation [see Fig. 1(a)]

$$\frac{dN_\upsilon}{dt} = W_\upsilon^{\rm cap}(N_\upsilon + 1) - (W_\upsilon^{\rm down} + W_\upsilon^{\rm up})N_\upsilon, \qquad (1)$$

where $W_\upsilon^{\rm cap}$ is the capture rate from the condensate, and $W_\upsilon^{\rm down}$ and $W_\upsilon^{\rm up}$ are the loss rates to more deeply bound states and back to the condensate, respectively. The factor $N_\upsilon + 1$ comes from the bosonic nature of the atoms: the capture will be bose enhanced (or stimulated), as opposed to the depletion of the level (proportional to N_υ).

Let us first calculate $W_\upsilon^{\rm cap}$. As mentioned above, the capture rate is equal to the phonon emission rate. For Bogoliubov quasiparticles (or phonons), the emission rate of phonons with momentum $\vec{q}$ and energy $\hbar\omega_{\vec{q}}$ can be estimated using the Fermi golden rule [4,17]

$$w_{\rm emis}(\vec{q}) = \frac{2\pi}{\hbar}\frac{\mu_c^2}{nV}S(\vec{q})I(\vec{q})(n_{\vec{q}} + 1)\delta(\Delta\varepsilon - \hbar\omega_{\vec{q}}), \qquad (2)$$

where $S(\vec{q}) = \hbar q^2/2m\omega_{\vec{q}}$ is the static structure factor (for free-free transitions [4,6,18]), V is the volume occupied by the condensate, $\mu_c = 4\pi\hbar^2 na/m$ its chemical potential (n, m, and a are the density, mass, and scattering length of the condensate atoms, respectively), and $n_{\vec{q}}$ the phonon occupation number. Here, $I(\vec{q})$ accounts for transitions from a continuum to a bound state. If we represent the initial condensate state containing N atoms ($N = nV$ is large) by the single particle wave function $\Psi_0(\vec{r})$ and the final bound state by the wave function $\Psi_\upsilon(\vec{r})$, with energy difference $\Delta\varepsilon = \varepsilon_0 - \varepsilon_\upsilon = \hbar\omega_{\vec{q}}$, then

$$I(\vec{q}) = N\left|\int d^3r\Psi_\upsilon^*(\vec{r})e^{-i\vec{q}\cdot\vec{r}}\Psi_0(\vec{r})\right|^2. \qquad (3)$$

At zero temperature in an infinite homogeneous system, we set $\varepsilon_0 = 0$ ($\vec{k} = 0$), and we have [19]

$$\Psi_0(\vec{r}) = \frac{1}{\sqrt{V}} \quad \text{and} \quad \Psi_\upsilon(\vec{r}) = \frac{1}{\sqrt{2\pi a_\upsilon}}\frac{e^{-r/a_\upsilon}}{r}, \qquad (4)$$

where the binding energy of the final bound state is related to the extent of the s-wave function $\Psi_\upsilon(\vec{r})$ via $\varepsilon_\upsilon = -\hbar^2/2\mu a_\upsilon^2$, with μ being the system reduced mass. Note that we have $\Delta\varepsilon = \hbar^2/2\mu a_\upsilon^2$. In an isotropic Bose gas, $I(\vec{q})$ depends only on the magnitude of $\vec{q}$, and with Eq. (4), $I(\vec{q}) = 8\pi a_\upsilon^3 n/(1 + q^2a_\upsilon^2)^2$.

The capture rate is obtained by integrating over all possible phonon states, i.e., $\int d^3q\, w_{\rm emis}(\vec{q})\, V/(2\pi)^3$, and

$$W_\upsilon^{\rm cap} = 4\frac{\mu_c}{\hbar}\sqrt{\frac{m}{\mu}}\frac{a}{a_\upsilon}\frac{(\sqrt{1+\xi^2} - \xi)^{3/2}}{\sqrt{1+\xi^2}}(n_{q_0} + 1)I(q_0)\,, \qquad (5)$$

where $\hbar\omega_{q_0} = \Delta\varepsilon$, and $\xi = \mu_c/\Delta\varepsilon = 8\pi n a_\upsilon^2 a\mu/m$. For Bogoliubov phonons, $\hbar\omega_q = \hbar qs\sqrt{1 + (\hbar q/2ms)^2}$, and $q_0^2a_\upsilon^2\ \mu/m = \sqrt{1+\xi^2} - \xi$, where the sound velocity s is related to the chemical potential by $\mu_c = ms^2$. Then

$$W_\upsilon^{\rm cap} = 4\frac{\mu_c}{\hbar}\left(\frac{m}{\mu}\right)^{3/2}\frac{\xi(\sqrt{1+\xi^2} - \xi)^{3/2}}{\sqrt{1+\xi^2}} \times\left[1 + \frac{m}{\mu}(\sqrt{1+\xi^2} - \xi)\right]^{-2}(n_{q_0} + 1). \qquad (6)$$

For infinitly massive ions, we have $m/\mu = 1$ [16]. The two limiting values of ξ correspond to different physical conditions: $\xi \to \infty$ implies $q_0 \to 0$, i.e., a phononlike regime, and $\xi \to 0$ implies dilute conditions with a binary collision regime. In Fig. 2, we illustrate $W_\upsilon^{\rm cap}$ as a function of ξ, and show its behavior for the asymptotic regimes: $W_\upsilon^{\rm cap} \propto \hbar n^2 a_\upsilon^2 a^2/m$ for small ξ, and $W_\upsilon^{\rm cap} \propto \hbar/ma_\upsilon^3\sqrt{na}$ for large ξ. The binary regime rate ($\xi \ll 1$) is proportional to n^2 as expected: the capture process is then reduced to three-body recombination. Similar processes have been studied for three identical atoms [20]. The $n^{-1/2}$ dependence for $\xi \gg 1$ clearly indicates the dominant contribution of phonon assisted transitions [17,21]. The increase in sound velocity $s \propto \sqrt{n}$ with the condensate density n reduces the rate of phonon emission (and $W_\upsilon^{\rm cap}$) [22].

As an example, we consider a sodium ion (Na^+) in a condensate of sodium atoms at $T = 0$, with $n \sim 10^{14}$ cm^{-3}

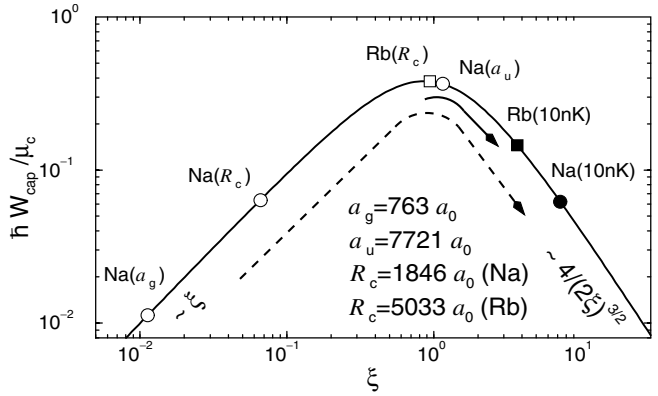

FIG. 2. $W_v^{\rm cap}$ as a function of ξ for various values of a_v corresponding to the states $2\Sigma_g^+$ and $2\Sigma_u^+$ of Na_2^+ [7], and $a_v \sim R_c$ for both Na and Rb. The two limiting behaviors and the equilibrium points for Na and Rb at 10 nK are shown.

and $a = 52\ a_0$ [23] (a_0: bohr radius). For the uppermost bound state of the polarization potential, a_v is well approximated by the atom-ion scattering length $a_i \sim 2000\ a_0$ [7,24], and assuming $m/\mu \approx 1$ [16], we obtain $\xi \sim 0.066$ (the system is closer to the binary regime than the phonon regime). Neglecting n_{q_0}, we find $\mathrm{W}_v^{\rm cap} \sim 600\ \mathrm{s}^{-1}$ (see Fig. 2), i.e., roughly 600 atoms will be captured by the ion, and another 600 will be emitted as phonons (excited out of the condensate) per second. In view of typical condensate lifetimes, this represents a sizable transfer mechanism that should be observable.

By comparison, the capture rate into more deeply bound states will be much smaller. Using the approach of LeRoy and Bernstein [25], we find for the second uppermost level of the pure polarization potential [26]

$$a_{v-1}^2 = \frac{a_v^2}{1 + (2\mu a_v^2/\hbar^2)^{1/4} K_4}, \tag{7}$$

where $K_4 = 4\sqrt{2\pi\hbar^2/\mu}\ \Gamma(5/4)/[\Gamma(3/4)C_4^{1/4}]$. Because $a_{v-1} < a_v \sim a_i$, the capture rate will be accompanied by atomlike excitations with $W_v^{\rm cap} \propto a_v^2$, and

$$\frac{W_v^{\rm cap}}{W_{v-1}^{\rm cap}} \approx \frac{a_v^2}{a_{v-1}^2} \simeq 1 + \left(\frac{2\mu a_v^2}{\hbar^2}\right)^{1/4} K_4. \tag{8}$$

For Na, $C_4 = 162.7$ a.u. [7], and assuming $m/\mu \approx 1$, we find that $W_v^{\rm cap}$ is about 10 times larger for the uppermost level than $W_{v-1}^{\rm cap}$ for the second uppermost level. The ratio is even larger for deeper levels: spontaneous capture occurs mostly in the uppermost level.

Let us now examine $W_v^{\rm up}$ and $W_v^{\rm down}$. Since only the capture in the uppermost level is relevant, we consider only this level. At very low temperatures, $k_B T \ll \Delta\varepsilon$, and there are no thermal phonons with sufficient energy to promote bound atoms to the free condensate state, and nonequilibrium phonons emitted from the capture process are moving far away from the location of the ion and can be neglected. Hence, because of energy conservation, transitions from bound levels to the continuum (into the condensate) are not allowed, and $W_v^{\rm up} \sim 0$. The collision of atoms within the upper bound level (or condensate atoms with the trapped ones) may result in the decay of the upper level to a lower level. The rate of this process ($W_v^{\rm down}$) is inversely proportional to the binding energy of the deeper level: $W_v^{\rm cap}/W_v^{\rm down} \sim a_v^2/a_{v-1}^2$. Hence, $W_v^{\rm down}$ is of the same order as $W_{v-1}^{\rm cap}$, and is at least one order of magnitude smaller than $W_v^{\rm cap}$.

As the number of trapped atoms N_v within the uppermost bound level v increases, the atom-atom repulsive mean field energy [27] will grow, and will eventually "push" the energy level up. Using a simple treatment based of the Gross-Pitaevskii equation, one can show that the binding energy of the uppermost level v, for $N_v \gg 1$, is approximately given by $\varepsilon_v(N_v) \simeq (ma_v/6\mu a N_v)^{2/3}\varepsilon_v$. Although the number of atoms trapped in the level v would apparently grow to extremely large numbers (since the decay rates out of it are much smaller), other effects, such as charge hopping [7] or thermal fluctuations, will limit that maximum number. The most important one is related to thermal fluctuations. If their energy ($\sim k_B T$) is equal to $|\varepsilon_v(N_v)|$, thermal equilibrium is reached, and as many atoms get kicked out of the level as there are being captured. In other words, $W_v^{\rm up}$ will be equal to $W_v^{\rm cap}$. At that point, $N_v^{\rm max}$ can be found from $\varepsilon_v(N_v^{\rm max}) \sim k_B T$, and ξ will have reached its equilibrium value $\xi_{\rm eq} \sim \mu_c/k_B T$. For a BEC of Na atoms with $a_v \sim R_c \sim 2000 a_0$ and $T \sim 100$ nK, we get $N_v^{\rm max} \sim 600$. In Fig. 2, we illustrate the trajectory of the system as a function of ξ for various initial values of a_v, for both sodium and rubidium condensates. In all cases, as ξ grows from its initial to its final equilibrium value, the capture rate $W_v^{\rm cap}$ passes through a maximum before reaching its final value: then the rates "in" and "out" of the uppermost level are equal, and the system has reached thermal equilibrium.

Lasers can also be used to control coherent stimulated transitions of BEC atoms into a specific bound state v of the molecular ion. Two off-resonant laser fields with a frequency difference matching the transition energy into a specific bound state v cause a stimulated two-photon transition from the BEC into the bound state of the molecular ion [see Fig. 1(b)]. By assuming two copropagating laser beams, negligible momentum is transferred to the atoms. In the limit of sufficiently slow excitation, the atom-laser field interaction is described by $H_{\rm int} = \hbar\Omega \int d^3 r \Psi_v(\vec{r})^* \Psi_0(\vec{r}) \hat{b}_v^\dagger \hat{b}_0 + \text{h.c.}$, where Ω is the Rabi frequency of the two-photon transition proportional to the laser intensity, and $\hat{b}_0$, $\hat{b}_v$ are bose annihilation operators for the BEC and bound atoms, respectively. The resulting dynamics corresponds approximately to that of a two-component BEC with effective coupling. The number of atoms stimulated into the bound state can be estimated by solving the equations of motion for $\hat{b}_0$, $\hat{b}_v$. For a sufficently short interaction time τ, assuming an undepleted BEC $\langle \hat{b}_0^\dagger \hat{b}_0 \rangle = N$, and neglecting spontaneous relaxation, we obtain for $N_v = \langle \hat{b}_v^\dagger \hat{b}_v \rangle$

$$N_v(\tau) \simeq |\Omega|^2 \tau^2 I_v(0) = 8\pi a_v^3 |\Omega|^2 \tau^2 N/V. \quad (9)$$

The rate of stimulation $W_v^{\rm st}$ is therefore proportional to the laser power with a characteristic frequency scale given by $W_v^{\rm st} \sim (\Omega\sqrt{a_v^3 N/V})$, which can be easily made larger than the spontaneous capture rate $W_v^{\rm cap}$.

In practice, very fast excitation by lasers might result in heating of the condensate. The heating rate $W_v^{\rm heat}$ is given by the transition probability from the bound state into the continuum [Fig. 1(c)]. This rate is also proportional to the laser power and can be estimated using Fermi's golden rule. Taking $1/W_v^{\rm st}$ as a characteristic transition time, we find that heating is negligible in a dilute condensate ($Na^3/V \ll 1$): $W_v^{\rm heat} \ll W_v^{\rm st}$ provided that

$$W_v^{\rm st} \ll \hbar/m(N/V)^{2/3}. \quad (10)$$

For realistic parameters, such as those of a sodium condensate used above, the stimulated rate $W_v^{\rm st}$ can be in the range of 100 KHz–1 MHz without substantial heating of the condensate. For weakly bound states, such rates are achievable with modest laser powers used, e.g., in Bragg spectroscopy experiments [6]. It is important to note that as the number of bound atoms increases, atom-atom interaction can shift the bound state energy out of resonance, thereby inhibiting coherent transitions. This is directly analogous to the recently proposed "dipole blockade" in Rydberg atoms [28], and can be potentially used for nontrivial manipulation of quantum states in a BEC.

In conclusion, we predicted the formation of large molecular ions in a BEC, analyzed the dynamics of their spontaneous growth and proposed a technique for their fast and coherent generation in specific states. Such mesoscopic molecular ions could be observed, e.g., in experiments involving weak ionization of BECs. Of further interest are experiments in which charged impurities are introduced via controlled processes involving either a combination of ion and atom traps, or using surface microtraps. The present work suggests that in such situations, ionic impurities can be used as a new tool for the accurate manipulation of BECs, including their quantum state. Finally, we note that one could manipulate, using external electric and magnetic fields, the position of the atomic cloud trapped around a charged particle.

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