Laser cooling down to molecular condensate

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On the repulsive side of the Feshbach resonance fermionic atoms can bind into diatomic molecules which at low enough temperature condense into molecular Bose-Einstein condensate. Sufficiently close to the resonance the molecules are bound so weakly that a significant fraction of unbound fermionic atoms survives even below the temperature of condensation. In this paper we show that laser cooling of the unbound fermionic atoms can reduce temperature of mixture of atoms and molecules below the condensation temperature. Numerical simulations for reasonable experimental parameters show that it is possible to achieve molecular condensate after a few seconds of laser cooling.

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Fermi superfluids are in the center of interest of recent studies in physics of ultracold quantum gases. They provide perhaps the most promising model system to study the superfluidity, and the Bardeen-Cooper-Schrieffer (BCS) theory in the limit of strong interactions, when the size of Cooper pairs becomes comparable or even smaller than average interparticle distance, and the mean field description breaks down [1]. In cold atom systems the use of Feshbach resonances permits to tune the *s*-wave scattering length between the atoms with opposite "spin" [2], assuring in this way a high flexibility of the systems. This technique allows in particular to drive the system through the BCS-BEC crossover when the scattering length diverges and changes sign from negative (attractive) to positive (repulsive). During this passage, the state of the system continuously evolves from the BCS superfluid of weakly correlated Cooper pairs, through the pseudogap regime, and then the unitarity limit, when the scattering length diverges, to the Bose-Einstein condensate (BEC) of diatomic molecules.

In practice, the realization of the BCS-BEC crossover in this direction, i.e., from Cooper pairs to molecules, is very difficult to achieve. The main reason is that just reaching the BCS regime requires very low temperatures [3,4]. The standard cooling method used for fermions with internal "spins" is a combination of evaporation and sympathetic cooling. This method suffers the effects of the, so-called, Pauli blocking at low temperatures, as well as heating due to the particle losses [5]. Perhpas the most spectacular results in this respect have been obtained in Ref. [6], where the comparison of data for ^{6}Li in terms of collisionless superfluid and collisional hydrodynamics was performed, and the first signatures of superfluidity were considered plausible. Apparently, an easier way to achieve superfluidity turned out to employ the Feschbach resonance technique, and to go first to the repulsive side of the resonance. On that side of the resonance pairs of fermions form large, but extremely stable bosonic molecules. These molecules have large cross sections for elastic scattering, and small for inelastic processes [7], making them essentially ideal subjects of evaporation cooling. By now several successful experiments with molecular condensates have been performed [8], and have opened a way to the first successful experiments with Fermi superfluids by going back to the attractive side of the Feschbach resonance [9].

A challenging alternative to collision based cooling methods is provided by the laser cooling. Earlier theoretical studies demonstrated that laser cooling allows to reach the degeneracy in Fermi systems [10], and to go all the way down to the Fermi superfluid [11] on the attractive side of the Feshbach resonance. The purpose of this paper is to show that laser cooling offers also an alternative technique to reach molecular condensate on the repulsive side of the resonance. Moreover, it allows to post-cool the condensate prepared via evaporative cooling down to even lower temperatures. We have developed here a fully self-consistent quantum kinetic theory of molecular condensate creation via laser cooling.[1] The results are very promising, despite the fact that condensate formation shifts the energies of quasiparticle excitations with respect to the equally spaced harmonic oscillator levels, and at the same time reduces fermionic population exposed to laser cooling, the cooling remains efficient well below the condensation temperature of $0.18T_F$ down to at least $0.06T_F$. Optimization of the cooling protocol could probably significantly reduce this temperature.

We consider system containing two species of fermions with "spin up and down," molecules which are in their bound states, and fermions in an excited state. We work on the repulsive side of the Feshbach resonance, where interactions strengths between spin up and/or down fermions and molecules, as well as the binding energy of the molecules are simple universal functions of the effective *s*-wave scattering length a between spin up and down fermions [7]. Close to the resonance the binding energy of molecules $\nu=-\hbar^2/ma^2$ can be made less than the condensation temperature k_BT_C for the molecules. As a result at all temperatures greater than T_C populations of molecules and fermions are comparable. Be-

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[1]Similar to self-consistent theories of the BEC growth.[12]

low T_C, a fraction of molecules condenses leaving room in molecular phase space for more fermions to bind into molecules. Fermionic population is then shrinking, but it remains still comparable to the shrinking population of noncondensed molecules all the way down to the dissociation temperature, $T_D=\nu/k_B$. Below T_D, fermionic population is suppressed, but at the same time almost all molecules are already condensed. Thus, close to the Feshbach resonance, when $T_D<T_C$, there are favorable conditions for efficient sympathetic cooling of molecules by collisions with spin up and/or down fermions at all temperatures of interest. The fermions in turn are subject to laser cooling, and provide the heat reservoir, which is comparable to the cooled system (noncondensed molecules). Populations of the spin up and down fermions are the same. Mutual s-wave interactions between these two populations and the population of molecules lead to thermalization on a time scale, which is faster than the rate of laser cooling. At the same time three-body collisions keep the system close to chemical equilibrium between molecules and fermions. The system remains thus in a state of quasiequilibrium with slowly decreasing temperature. This thermal state is described by the generalized BCS theory, or boson-fermion model [13] which includes BEC of molecules described by the Gross-Pitaevskii equation. The model takes into account coherent tunneling between molecules and pairs of spin up and down fermions. Thermal excitations of the molecular condensate are described by the bosonic Bogoliubov theory. Structure of the ground state of fermions and its fermionic excitations follows from the set of Bogoliubov-de Gennes equations together with self-consistent definitions of the gap function and the mean field potential. On the repulsive side of the Feshbach resonance the leading contribution to the gap function (order parameter) comes from the molecular BEC through the coherent coupling between molecules and pairs of fermions. In particular, the gap function experienced by fermions is proportional to the condensate wave function, and its fluctuations are in this regime very moderate.

Laser cooling of fermions was described in detail in Ref. [10]. Coherent laser excites atoms from, say, the spin down ground state to the excited state and spontaneous emission brings them back to the spin down ground state. Frequencies of the laser assure that the generalized Raman cooling takes place [14], laser detuning from the transition is such that by energy conservation the exitation of internal state is accompanied by decreasing energy of center of mass motion. On the other hand, large wavelength of the emitted radiation assures that in the spontaneous emission process an atom tends to stay in the same trap level. A net result is decreasing center of mass energy. A more detailed Λ-diagram of the transitions is shown in Fig. 1. Here $|g\rangle$ is the spin down ground state, $|e\rangle$ is the metastable excited state, and $|r\rangle$ is a third auxiliary fast-decaying state. Two lasers excite coherently the resonant Raman transition $|g\rangle \rightarrow |e\rangle$ with an associated Rabi frequency Ω. A repumping laser in or off resonance with the transition $|g\rangle \rightarrow |r\rangle$ pumps optically the atom into $|g\rangle$. With this three-level scheme one obtains an effective two-level system with an effective spontaneous emission rate γ which can be controlled by varying the intensity or the detuning of the repumping laser.

In this paper we take into account interactions between fermions and describe the cooling process in terms of instantaneous fermionic and bosonic (molecular) Bogoliubov quasiparticles, whose eigenfunctions and eigenenergies are self-consistently updated during the evolution. We work in the "festina lente" limit, when spontaneous emission rate γ is less than frequency of the harmonic confinement, to avoid reabsorbtion effects [16], and also employ spherical symmetry and ergodic approximations [11,12].

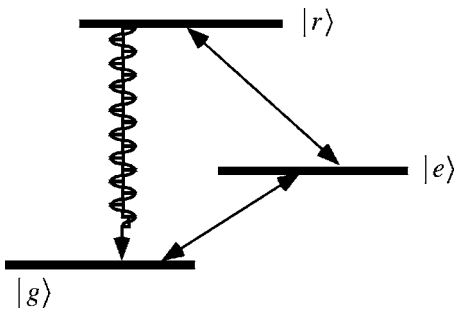

FIG. 1. A three-level Λ-scheme resulting in an effective two-level system ($|g\rangle$ and $|e\rangle$) with a Rabi frequency Ω and spontaneous emission rate γ. This figure originates from Ref. [11].

The boson-fermion model [13] is defined by the Hamiltonian

$$\hat{H} = \int d^3r \Bigg[\sum_{a=+,-,e,m} \hat{\psi}_a^\dagger \mathcal{H}_a \hat{\psi}_a + (\nu - \mu)\hat{\psi}_m^\dagger \hat{\psi}_m + \lambda \hat{\psi}_m^\dagger \hat{\psi}_+ \hat{\psi}_- + \text{h.c.} + g\hat{\psi}_+^\dagger \hat{\psi}_-^\dagger \hat{\psi}_- \hat{\psi}_+ + 0.3\, g\hat{\psi}_m^\dagger \hat{\psi}_m^\dagger \hat{\psi}_m \hat{\psi}_m + 1.2\, g\hat{\psi}_m^\dagger \hat{\psi}_m (\hat{\psi}_+^\dagger \hat{\psi}_+ + \hat{\psi}_-^\dagger \hat{\psi}_-) \Bigg], \tag{1}$$

where the fields $\hat{\psi}_+$ and $\hat{\psi}_-$ describe fermions with spin up and down, respectively, $\hat{\psi}_e$ corresponds to fermions in the excited state, and $\hat{\psi}_m$ is the bosonic molecular field. Here $\mathcal{H}_a = -\hbar^2/2m_a\nabla^2 + V_a(\vec{r}) - \mu$ is a single particle Hamiltonian, m_a is atomic mass m or molecular mass $2m$, $V_a(\vec{r}) = \frac{1}{2}m_a\Omega_a^2 r^2$ is the harmonic trap potential, μ is the chemical potential, $g = 4\pi\hbar^2 a/m$ is fermion-fermion interaction strength with a large positive s-wave scattering length a, λ is a coupling between pairs of atoms and molecules, and $\nu = -\hbar^2/ma^2$ is the molecular binding energy. Close to the Feshbach resonance, when g is large, we can neglect interactions with excited atoms.

In the mean field approximation, which closely follows the BCS theory, the quartic fermion-fermion interaction term in the Hamiltonian (1) is made quadratic by replacing products of operators by their averages in all possible ways. The averages are the mean field potential $W(\vec{r}) = 1.2\, g\langle\hat{\psi}_m^\dagger(\vec{r})\hat{\psi}_m(\vec{r})\rangle + g\langle\hat{\psi}_\pm^\dagger(\vec{r})\hat{\psi}_\pm(\vec{r})\rangle$, and the anomalous pairing potential $P(\vec{r}) = \langle\hat{\psi}_+(\vec{r})\hat{\psi}_-(\vec{r})\rangle$, which is mixing fermions with spin up and down. In the mean-field approximation the molecular field $\hat{\psi}_m$ is replaced by a c-number condensate amplitude ϕ that fulfills the stationary Gross-Pitaevskii equation

$$(2\mu - \nu)\phi = \mathcal{H}_m\phi + 0.6\ g|\phi|^2\phi + \lambda P. \quad (2)$$

Here ϕ is normalized to the number of condensed molecules. At low temperatures, when most atoms are bound into molecules and most molecules are condensed, physics is dominated by molecular condensate. The large molecular condensate is well described by the mean-field theory. The mean-field equations for fermions are

$$i\hbar\frac{d}{dt}\hat{\psi}_\pm = \mathcal{H}_\pm\hat{\psi}_\pm + W(\vec{r})\hat{\psi}_\pm \mp \Delta(\vec{r})\hat{\psi}^\dagger_\mp, \quad (3)$$

with an effective gap function $\Delta(\vec{r}) = gP(\vec{r}) + \lambda\phi(\vec{r})$.. These equations are mixing spin up and down components but they can be "diagonalized" by the Bogoliubov transformation $\hat{\psi}_\pm(\vec{r}) = \Sigma_m \hat{b}_{m,\pm}u_m(\vec{r}) \mp \hat{b}^\dagger_{m,\mp}v^*_m(\vec{r})$, with fermionic quasiparticle annihilation operators $\hat{b}_{m,\pm}$. The quasiparticle modes (u_m, v_m) fulfill the Bogoliubov-de Gennes equations with positive energies ω_m,

$$\omega_m u_m = +\mathcal{H}_\pm u_m + W\ u_m - \Delta\ v_m,$$

$$\omega_m v_m = -\mathcal{H}_\pm v_m - W\ v_m - \Delta^* u_m. \quad (4)$$

In a thermal state with inverse temperature β the average occupation numbers of quasiparticle states are $N_m = \langle\hat{b}^\dagger_{m,\pm}\hat{b}_{m,\pm}\rangle = [\exp(\beta\omega_m)+1]^{-1}$. Equations (4) are solved together with the self-consistency conditions,

$$\Delta(\vec{r}) = \lambda\phi(\vec{r}) + g\sum_m [1 - 2N_m]u_m(\vec{r})v^*_m(\vec{r}), \quad (5)$$

$$W(\vec{r}) = 1.2\ g\langle\hat{\psi}^\dagger_m(\vec{r})\hat{\psi}_m(\vec{r})\rangle + g\sum_m (1 - N_m)|v_m(\vec{r})|^2 + N_m|u_m(\vec{r})|^2, \quad (6)$$

by successive iterations with the chemical potential μ adjusted to keep the total number of atoms constant. In the low temperature limit, when the molecular condensate is large, the gap function in Eq. (5) is dominated by the $\lambda\phi$ contribution from the condensate. The same is true for W. As the molecular condensate is well described by the mean-field theory, we do not need to worry about fluctuations of Δ or W. The average molecular density $\langle\hat{\psi}^\dagger_m(\vec{r})\hat{\psi}_m(\vec{r})\rangle$ is a sum of the condensate density $|\phi(\vec{r})|^2$ plus density of noncondensed molecules. After every iteration the Gross-Pitaevskii equation (2) is solved by relaxation. The total number of atoms includes atoms bound into noncondensed molecules depleted from the condensate by thermal and quantum fluctuations. We estimate the number of noncondensed molecules using the approximate bosonic Bogoliubov modes in the Thomas-Fermi limit [17]. The ultraviolet divergence in Eq. (5) is regularized by the quickly convergent method of Ref. [18].

Excitation of atoms from the spin down state to the excited state is described by the Hamiltonian,

$$\hat{H}_{\rm las} = \int d^3r\left(\frac{1}{2}\Omega e^{i\vec{k}_L\vec{r}}\hat{\psi}^\dagger_e(\vec{r})\hat{\psi}_-(\vec{r}) + \text{h.c.} - \delta\hat{\psi}^\dagger_e\hat{\psi}_e\right), \quad (7)$$

driving coherent oscillations with Rabi frequency Ω and laser detuning δ. The excitation is accompanied by the spontaneous emission described by a superoperator,

$$\mathcal{L}\hat{\rho} = \gamma\sum_{mk} U_{mk}\mathcal{D}[\hat{b}^\dagger_{m,-}\hat{e}_k]\rho + V_{mk}\mathcal{D}[\hat{b}_{m,+}\hat{e}_k]\rho \quad (8)$$

with a spontaneous emission rate γ. Here, the Lindblad superoperator is $\mathcal{D}[\hat{A}]\rho = \hat{A}\rho\hat{A}^\dagger - \frac{1}{2}\hat{A}^\dagger\hat{A} - \frac{1}{2}\hat{A}^\dagger\hat{A}\rho$, and the matrix elements are, e.g., $U_{mk} = \int d\Omega_k \mathcal{W}(\Omega_k)|u_{mk}(\vec{k})|^2$ with the spontaneous emission pattern $\mathcal{W}(\Omega_k)$ and the generalized Frank-Condon factors $u_{mk}(\vec{k}) = \int d^3r\ e^{i\vec{k}\vec{r}}w^*_k(\vec{r})u_m(\vec{r})$. Here, $w_k(\vec{r})$ is the kth eigenstate of the harmonic oscillator and $\hat{e}_k$ is an annihilation operator of an excited atom in this state.

Adiabatic elimination of the excited state (cf. Ref. [19] also see Ref. [15]) leads to the kinetic equations for the occupation numbers taking into account the effects of Fermi statistics,

$$\frac{dN_{m,-}}{dt} = \sum_n \Gamma^{(-)}_{m\leftarrow n}(1 - N_{m,-})N_{n,-} - (n \leftrightarrow m) + C_{mn}(1 - N_{m,-}) \times (1 - N_{n,+}) - A_{nm}N_{m,-}N_{n,+},$$

$$\frac{dN_{m,+}}{dt} = \sum_n \Gamma^{(+)}_{m\leftarrow n}(1 - N_{m,+})N_{n,+} - (n \leftrightarrow m) + C_{nm}(1 - N_{m,+}) \times (1 - N_{n,-}) - A_{mn}N_{m,+}N_{n,-}.$$

They describe relaxation of $\pm$ quasiparticles ($\Gamma^{(\pm)}_{m\leftarrow n}$), and creation/annihilation of pairs of + and − quasiparticles (C_{nm} and A_{nm}). The transition rates are, e.g.,

$$\Gamma^{(-)}_{m\leftarrow n} = \frac{\Omega^2}{2\gamma}\sum_k \frac{\gamma^2 U_{mk}|u_{nk}(\vec{k}_L)|^2}{(\delta - \omega^e_k + \omega_n)^2 + \gamma^2_k}. \quad (9)$$

Here, ω^e_k is the energy of the kth harmonic oscillator state and γ_k is approximate spontaneous decay rate of an excited atom in this state, $\gamma_k = \gamma\Sigma_m U_{mk}(1 - N_{m,-}) + V_{mk}N_{m,+}$, see Appendix A.

Laser cooling drives average occupation numbers $N_{m,\pm}$ of fermionic quasiparticles out of thermal equilibrium. At the same time interactions between fermionic and bosonic quasiparticles drive the system towards thermal equilibrium. Numerical simulations in Ref. [10] show that thermal relaxation remains efficient all the way down to the temperature of $0.03\ T_F$. This justifies our assumption of fast equilibration to a quasiequilibrium state. After a short period of laser cooling dt the occupation numbers $N_{m,\pm}$ go out of equilibrium where $N_{m,\pm} = N_m = [\exp(\beta\omega_m)+1]^{-1}$. The initial total energy $E(T) = \Sigma_{m,\pm}\omega_m N_{m,\pm} + \Sigma_m\omega^B_m N^B_m$, including bosonic Bogoliubov quasiparticles with frequencies ω^B_m, and average occupation numbers $N^B_m = [\exp(\beta\omega^B_m) - 1]^{-1}$, changes by $dE = \Sigma_m\omega_m(N_{m,+} + N_{m,-} - 2N_m)$. The relaxation brings the occupation numbers $N_{m,\pm}$ to a new state of equilibrium at a temperature $T + dT$, but does not change the total energy. The energy of the system at the new temperature $E(T + dT)$ differs from

the initial $E(T)$ by $dE_T \approx (2dT/T^2)\Sigma_m \omega_m^2 N_m(1-N_m) + (dT/T^2)\Sigma_m(\omega_m^B)^2 N_m^B(1+N_m^B)$. Conservation of energy in thermal relaxation means that $dE=dE_T$. In our simulations we use this equality to find the new lower temperature $T+dT$, and then solve the Bogoliubov-de Gennes equations and the Gross-Pitaevskii equation self-consistently to adjust the Bogoliubov modes, the condensate wave function and the chemical potential to the new lower temperature. With the new Bogoliubov modes we calculate new transition rates, etc.

Calculation of the transition rates is the most time consuming part of the numerical simulation. This is why we were forced to assume spherical symmetry. For spherically symmetric $\Delta(r), W(r)$, and $\phi(r)$ the Bogoliubov modes are $u_{ln}(r)Y_{lm}(\theta,\varphi)$ and their energies ω_{ln} do not depend on m. The bosonic quasiparticle energies in the Thomas-Fermi limit [17] become $\omega_{ln}^B = \Omega_m(2n^2+2nl+3n+l)^{1/2}$. To be consistent with the spherical symmetry we also made ergodic approximation that quasiparticle occupation numbers $N_{ln,\pm}$ do not depend on m. In other words, we assume fast equilibration within each quasiparticle energy shell which keeps the system in a spherically symmetric state. See Appendix B for more on spherical symmetry.

In our simulations we use 81 harmonic oscillator levels, and assume that there are $N=10\,660$ atoms with spin up and down which in the noninteracting case at $T=0$ is equivalent to 39 filled energy levels. The frequency of the isotropic trap is $\Omega_a = 2\pi$ 2400 Hz, the same for all fermions and molecules. The scattering length for the interaction between the two species is $a=1200$ Å, a realistic value for interactions between two spin states $|F=9/2, m_F=9/2\rangle$ and $|F=9/2, m_F=7/2\rangle$ of ^{40}K near the Feshbach resonance. In the natural harmonic oscillator units this scattering length gives the interaction strength $g=4.7$, and the molecular binding energy $\nu=-7.3$. The wavelength of the cooling laser is $\lambda=720$ nm, and the laser detuning is $\delta=-12\hbar\Omega_a$. In all simulations the frequency of Rabi oscillations is much less than the spontaneous emission rate, $\Omega=0.1\gamma$, so that the average occupation of excited state remains small. In our simulations we neglect $W(r)$. The main effect of the mean field potential is an overall shift of energy levels which can be compensated by laser detuning δ. The coherent tunneling rate in trap units is $\lambda=1$.

In Fig. 2 we show temperature as a function of time for laser cooling with two values of γ. Similarly as on the BCS side of the Feshbach resonance the nonzero gap function $\Delta(r)$ induced by the molecular BEC does not affect much the efficiency of laser cooling. Moreover, the cooling remains efficient even below the dissociation temperature of $0.18T_F$. This is not quite surprising because even at such low temperature there is a nonzero fraction of fermions induced by coherent tunneling from the molecular BEC, which can be cooled by the laser. The initial cooling is faster for $\gamma=10$ but the final temperature of $0.06T_F$ is lower for $\gamma=1.25$. The best strategy to reach low temperatures in reasonable time is to do the cooling in a few stages starting with large γ and finishing with small γ, as shown in the inset in Fig. 2. In Fig. 3 we show the growth of the molecular condensate in the same simulations as in Fig. 2.

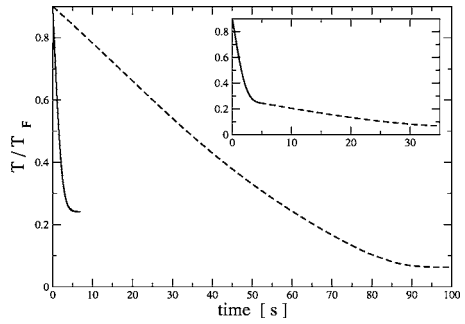

FIG. 2. Temperature T/T_F as a function of time for $\gamma=10$ (solid line) and $\gamma=1.25$ (dashed line). Here $T_F=40.5\hbar\Omega_a$, and the observed $T_C \approx 0.6T_F$. The cooling is faster for $\gamma=10$, but the lower final temperature of $0.06T_F$ is achieved for $\gamma=1.25$. The inset shows T/T_F in a two stage cooling process where γ is switched from 10 to 1.25 at $T=0.25T_F$.

To summarize, our numerical simulations show that for reasonable experimental parameters consistent with the requirements of the Festina lente regime, it is possible to laser cool fermions on the positive side of the Feshbach resonance all the way down to the molecular BEC. Despite the pairing effects, which produce a gap in the quasiparticle energy spectrum and the heat capacity of noncondensed molecules, it is possible to reach at least $0.06\,T_F$ in a time of a few seconds.

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APPENDIX A: TRANSITION RATES

Laser cooling is realized by the Hamiltonian,

$$\hat{H}_{\rm las} = \frac{1}{2}\Omega \int d^3r\, e^{i\vec{k}_L\vec{r}}\hat{\psi}_e^\dagger(\vec{r})\hat{\psi}_-(\vec{r}) + \text{h.c.}, \qquad \text{(A1)}$$

driving coherent oscillations with Rabi frequency Ω between the excited state e and the state $-$, together with the spontaneous emission superoperator,

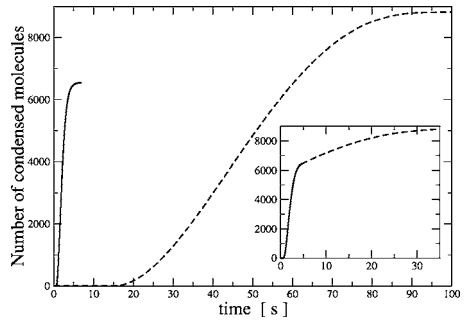

FIG. 3. The number of condensed molecules in the same simulations as in Fig. 1.

$$\mathcal{L}\hat{\rho} = \gamma \int d\varphi\, d\cos\theta\, \mathcal{W}(\theta,\varphi) \int d^3r_1 d^3r_2 e^{i\vec{k}(\vec{r}_1-\vec{r}_2)} \times [2\hat{\psi}_-^\dagger(\vec{r}_2)\hat{\psi}_e(\vec{r}_2)\hat{\rho}\hat{\psi}_e^\dagger(\vec{r}_1)\hat{\psi}_-(\vec{r}_1) - \hat{\psi}_e^\dagger(\vec{r}_1)\hat{\psi}_-(\vec{r}_1)\hat{\psi}_-^\dagger(\vec{r}_2)\hat{\psi}_e(\vec{r}_2)\hat{\rho} - \hat{\rho}\hat{\psi}_e^\dagger(\vec{r}_1)\hat{\psi}_-(\vec{r}_1)\hat{\psi}_-^\dagger(\vec{r}_2)\hat{\psi}_e(\vec{r}_2)], \quad \text{(A2)}$$

with an effective spontaneous emission rate of 2γ and the fluorescence dipole pattern $\mathcal{W}(\theta,\varphi)$. Expansion in the eigenmodes $w_l(\vec{r})$ of the harmonic trap

$$\hat{\psi}_e(\vec{r}) = \sum_l \hat{e}_l w_l(\vec{r}), \quad \text{(A3)}$$

and the Bogoliubov transformation (see main text) give

$$\hat{H}_{\text{las}} = \frac{1}{2}\Omega \sum_{ml} [\hat{b}_{m,-} u_{ml}(\vec{k}_L) + \hat{b}_{m,+}^\dagger v_{ml}^*(\vec{k}_L)]\hat{e}_l^\dagger + \text{h.c.} \quad \text{(A4)}$$

Here the (generalized) Frank-Condon factors are

$$u_{ml}(\vec{k}) = \int d^3r\, e^{i\vec{k}\vec{r}} w_l^*(\vec{r}) u_m(\vec{r}),$$

$$v_{ml}^*(\vec{k}) = \int d^3r\, e^{i\vec{k}\vec{r}} w_l^*(\vec{r}) v_m^*(\vec{r}). \quad \text{(A5)}$$

In a similar way, and after rotating wave approximation for e-atoms and the Bogoliubov quasiparticles, the spontaneous emission term becomes

$$\mathcal{L}\hat{\rho} = \gamma \sum_{ml} U_{ml}(2\hat{b}_{m,-}^\dagger \hat{e}_l \rho \hat{e}_l^\dagger \hat{b}_{m,-} - \hat{e}_l^\dagger \hat{e}_l \hat{b}_{m,-}\hat{b}_{m,-}^\dagger \rho - \rho \hat{e}_l^\dagger \hat{e}_l \hat{b}_{m,-}\hat{b}_{m,-}^\dagger) + \gamma \sum_{ml} V_{ml}(2\hat{b}_{m,+}\hat{e}_l \rho \hat{e}_l^\dagger \hat{b}_{m,+}^\dagger - \hat{e}_l^\dagger \hat{e}_l \hat{b}_{m,+}^\dagger \hat{b}_{m,+}\rho - \rho \hat{e}_l^\dagger \hat{e}_l \hat{b}_{m,+}^\dagger \hat{b}_{m,+}). \quad \text{(A6)}$$

Here the spontaneous emission rates are

$$U_{ml} = \int d\varphi d\cos\theta\, \mathcal{W}(\theta,\varphi)|u_{ml}(\vec{k})|^2,$$

$$V_{ml} = \int d\varphi d\cos\theta\, \mathcal{W}(\theta,\varphi)|v_{ml}(\vec{k})|^2. \quad \text{(A7)}$$

Adiabatic elimination of the excited state e, similar as in Ref. [19], results in the following evolution equations for the occupation numbers:

$$\frac{dN_{m,-}}{dt} = \sum_n \Gamma_{m\leftarrow n}^{(-)}(1-N_{m,-})N_{n,-} - \Gamma_{n\leftarrow m}^{(-)}(1-N_{n,-})N_{m,-} + C_{mn}(1-N_{m,-})(1-N_{n,+}) - A_{nm}N_{m,-}N_n, \quad \text{(A8)}$$

$$\frac{dN_{m,+}}{dt} = \sum_n \Gamma_{m\leftarrow n}^{(+)}(1-N_{m,+})N_{n,+} - \Gamma_{n\leftarrow m}^{(+)}(1-N_{n,+})N_{m,+} + C_{nm}(1-N_{m,+})(1-N_{n,-}) - A_{mn}N_{m,+}N_n. \quad \text{(A9)}$$

There are contributions from four different processes,

(i) relaxation of − quasiparticles,

$$\Gamma_{m\leftarrow n}^{(-)} = \frac{\Omega^2}{2\gamma}\sum_l \frac{\gamma^2 U_{ml}|u_{nl}(\vec{k}_L)|^2}{|\delta - \omega_l^e + \omega_n + i\gamma\sum_s U_{sl}(1-N_{s,-}+\delta_{s,n}) + V_{sl}N_{s,+}|^2}; \quad \text{(A10)}$$

(ii) relaxation of + quasiparticles,

$$\Gamma_{m\leftarrow n}^{(+)} = \frac{\Omega^2}{2\gamma}\sum_l \frac{\gamma^2 V_{nl}|v_{ml}(\vec{k}_L)|^2}{|\delta - \omega_l^e - \omega_m + i\gamma\sum_s U_{sl}(1-N_{s,-}) + V_{sl}(N_{s,+}+\delta_{m,s})|^2}; \quad \text{(A11)}$$

(iii) creation of pairs of + and − quasiparticles,

$$C_{mn} = \frac{\Omega^2}{2\gamma}\sum_l \frac{\gamma^2 U_{ml}|v_{nl}(\vec{k}_L)|^2}{|\delta - \omega_l^e - \omega_n + i\gamma\sum_s U_{sl}(1-N_{s,-}) + V_{sl}(N_{s,+}+\delta_{n,s})|^2}; \quad \text{(A12)}$$

(iv) annihilation of pairs of + and − quasiparticles,

$$A_{mn} = \frac{\Omega^2}{2\gamma}\sum_l \frac{\gamma^2 V_{ml}|u_{nl}(\vec{k}_L)|^2}{|\delta - \omega_l^e + \omega_n + i\gamma\sum_s U_{sl}(1-N_{s,-}+\delta_{n,s}) + V_{sl}N_{s,+}|^2}. \quad \text{(A13)}$$

APPENDIX B: SPHERICAL SYMMETRY

Simulation of laser cooling with a nonvanishing pairing function $P(\vec{r})$, and a Hartree potential $g_0\rho(\vec{r})$ is a numerically hard problem. The main difficulty is that after every short period of laser cooling it is necessary to reiterate Bogoliubov-de Gennes equations in order to obtain a self-consistent pairing function, Hartree potential, and the Bogoliubov modes (ω_m, u_m, v_m). With the new Bogoliubov modes, the transition rates $A_{mn}, C_{mn}, \Gamma^{\pm}_{m\leftarrow n}$ must be calculated again, and this is a very time-consuming operation. This numerical effort is reduced a lot with assumption of spherical symmetry. With spherically symmetric $\Delta(r)$ and $\rho(r)$ the Bogoliubov modes can be decomposed into spherical and radial parts,

$$u_{ln}(r)Y_{lm}(\theta,\varphi), \tag{B1}$$

$$v_{ln}(r)Y_{lm}(\theta,\varphi). \tag{B2}$$

In a similar way harmonic oscillator modes become $W_{ln}(r)Y_{lm}(\theta,\varphi)$. Quasiparticle energies ω_{ln} do not depend on m. We assume fast equilibration within each degenerate energy shell nl so that all occupation numbers in a given shell are the same $N_{ln,\pm}$. This assumption greatly simplifies the rate equations,

$$\begin{aligned}\frac{d}{dt}N_{l_1n_1,-} = \sum_{l_2n_2} & \gamma^{(-)}_{l_1n_1\leftarrow l_2n_2}(1-N_{l_1n_1,-})N_{l_2n_2,-} - \gamma^{(-)}_{l_2n_2\leftarrow l_1n_1} \\ & \times(1-N_{l_2n_2,-})N_{l_1n_1,-} + c_{l_1n_1l_2n_2}(1-N_{l_1n_1,-}) \\ & \times(1-N_{l_2n_2,+}) - a_{l_2n_2l_1n_1}N_{l_1n_1,-}N_{l_2n_2,+},\end{aligned} \tag{B3}$$

$$\begin{aligned}\frac{d}{dt}N_{l_1n_1,+} = \sum_{l_2n_2} & \gamma^{(+)}_{l_1n_1\leftarrow l_2n_2}(1-N_{l_1n_1,+})N_{l_2n_2,+} - \gamma^{(+)}_{l_2n_2\leftarrow l_1n_1} \\ & \times(1-N_{l_2n_2,+})N_{l_1n_1,+} + c_{l_2n_2l_1n_1}(1-N_{l_1n_1,+}) \\ & \times(1-N_{l_2n_2,-}) - a_{l_1n_1l_2n_2}N_{l_1n_1,+}N_{l_2n_2,-}.\end{aligned} \tag{B4}$$

Here $\gamma^{(\pm)}, a, c$ are averaged transition rates, for example,

$$\gamma^{(-)}_{l_1n_1\leftarrow l_2n_2} = \frac{1}{2l_1+1}\sum_{m_1m_2}\Gamma^{(-)}_{l_1n_1m_1\leftarrow l_2n_2m_2}. \tag{B5}$$

This equation combined together with the definition of $\Gamma^{(-)}$ gives

$$\gamma^{(-)}_{l_1n_1\leftarrow l_2n_2} = \frac{\gamma\Omega^2}{2(2l_1+1)}\sum_{m_1m_2}\sum_{l_en_em_e}\frac{U_{l_1n_1m_1l_en_em_e}\left|u_{l_2n_2m_2l_en_em_e}(\vec{k}_L)\right|^2}{[\delta-\omega(l_e+2n_e-\mu)+\omega_{l_2n_2}]^2+\gamma^2R^2_{l_en_e}}. \tag{B6}$$

Here the frequency of the intermediate excited state $\omega(l_e+2n_e-\mu)$ is measured with respect to the chemical potential μ because the quasiparticle energies $\omega_{l_2n_2}$ are also defined with respect to μ. For a laser beam with a $\vec{k}_L$ along the $\hat{z}$ axis the indices m_e and m_2 must be the same. Furthermore, it follows from the properties of Clebsch-Gordan coefficients that the sum $\Sigma_{m_1}U_{l_1n_1m_1l_en_em_e}$ does not depend on m_e so that we can set, e.g., $m_e=0$ and

$$\begin{aligned}\gamma^{(-)}_{l_1n_1\leftarrow l_2n_2} &= \frac{\gamma\Omega^2}{2(2l_1+1)} \\ &\times\sum_{l_en_e}\frac{\left(\sum_{m_1}U_{l_1n_1m_1l_en_e0}\right)\left(\sum_{m_2}\left|u_{l_2n_2m_2l_en_em_2}(\vec{k}_L)\right|^2\right)}{[\delta-\omega(l_e+2n_e-\mu)+\omega_n]^2+\gamma^2R^2_{l_en_e}} \\ &\equiv \frac{\gamma\Omega^2}{2(2l_1+1)} \\ &\times\sum_{l_en_e}\frac{\mathrm{SU}_{l_1n_1l_en_e}\mathrm{su}_{l_2n_2l_en_e}}{[\delta-\omega(l_e+2n_e-\mu)+\omega_n]^2+\gamma^2R^2_{l_en_e}}.\end{aligned} \tag{B7}$$

The last form shows that the transition rate $\gamma^{(-)}$ can be constructed out of matrices SU, su and a vector R. These elements can be expressed through even more elementary building blocks

$$\mathrm{SU}_{l_1n_1l_en_e} = \sum_l (jWu)^2_{ll_en_el_1n_1}S_{ll_el_1},$$

$$\mathrm{su}_{l_2n_2l_en_e} = \sum_m\left|\sum_l s_{ll_el_2m}(jWu)_{ll_en_el_2n_2}\right|^2.$$

The more elementary building blocks are

$$(jWu)_{ll_en_el_1n_1} = \int r^2dr\, j_l(kr)W_{l_en_e}(r)u_{l_1n_1}(r),$$

$$S_{ll_el_1} = (2l+1)\frac{2l_e+1}{2l_1+1}\sum_m\langle l,l_e,m,0|l_1,m^2\langle l,l_e,0,0|l_1,0^2,$$

$$s_{ll_el_2m} = i^l(2l+1)\sqrt{\frac{2l_e+1}{2l_2+1}}\langle l,l_e,0,m|l_2,m\langle l,l_e,0,0|l_2,0. \tag{B8}$$

The linewidth of an excited state l_e, n_e is approximately given by

$$R_{l_e n_e} = \sum_{ln} (1 - N_{ln}) \mathrm{SU}_{lnl_e n_e} + N_{ln} \mathrm{SV}_{lnl_e n_e}. \quad \text{(B9)}$$

The matrix SV is obtained from the matrix SU by a substitution $u \to v$. As the eigenfunction $u_{ln}(r)$ evolves in the process of laser cooling it is more efficient to express the matrix (jWu) through a static matrix (jWW) built out of harmonic oscillator modes,

$$(jWW)_{ll_e n_e l_1 n_1} = \int r^2 dr\, j_l(kr) W_{l_e n_e}(r) W_{l_1 n_1}(r). \quad \text{(B10)}$$

The static matrices s, S, jWW were prepared once, and stored then on a disk. They facilitate calculation of the temperature dependent matrices SU, su, R every time the wave functions u, v are updated.

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