Translational cooling of doped nanocrystals by Raman pulses: Towards macroscopic quantum state

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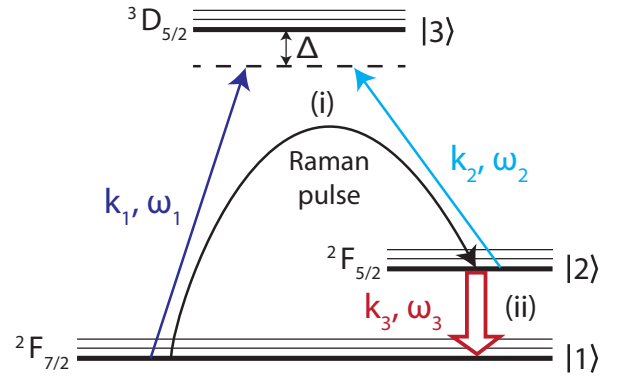
Here we present a novel method for deep optical cooling of CaF_2:Yb^{3+} nanocrystals, which is based on the coherent population transfer induced in the impurity ions by a sequence of UV Raman pulses [1]. A doped nanocrystal localized in a radio-frequency trap is cooled due to the strong photon recoil from the laser pulses of varied intensity and duration.



General energy scheme for cooling of CaF_2:Yb^{3+} nanocrystals. Stark split levels $^2F_{7/2}$, $^2F_{5/2}$, and $^3D_{5/2}$ in the energy structure of Yb ions form a three-level system. When the nanocrystal is exposed to the Raman pulses, the cooling occurs in two steps: (i) coherent population transfer from level 1 to level 2 without populating the excited state 3 and (ii) spontaneous decay from state 2 through the emission of an IR photon in a random direction

Consider the electric field of the Raman pulse:

$$\mathbf{E} = \mathbf{e}_1 E_1 \exp(-ik_1 z)\exp(i\omega_1 t) + \mathbf{e}_2 E_2 \exp(ik_2 z)\exp(i\omega_2 t) + \text{c.c.},$$

The ω_1 photon is absorbed by the doped nanocrystal, while the ω_2 photon propagating in the opposite direction induces emission of a similar photon. The resulting **total recoil momentum** Δp is additive and directed parallel to the laser beam of frequency ω_2 if the detuning Δ is positive (w is the coherent transfer probability, n_{Yb} is the number of Yb ions):

$$\Delta p = w n_{Yb} \hbar (k_1 + k_2)$$

Solving the optical Bloch equations yields the following populations of the ground states [2]:

$$A_1(p - \hbar k, t) = A_1^{(0)}(p)\left[1 - \frac{\alpha^2}{D^2}\sin^2(Dt)\right],$$

$$A_2(p + \hbar k, t) = A_1^{(0)}(p)\frac{\alpha^2}{D^2}\sin^2(Dt),$$

where $k = (k_1 + k_2)/2$, $A_1^{(0)}$ is the initial population of 1 level, $\alpha = g^2/\Delta$, $D^2 = (pk/M - \Delta)^2 + \alpha^2$, and Δ and Rabi frequency g are the same for the two beams. In the resonant case $\Delta = pk/M$ and the transfer probability is

$$w = \sin^2(g^2 t_R/\Delta),$$

where t_R is the duration of the Raman pulse. This allows the **deeper cooling** by effective change the total recoil of the nanocrystal by varying the pulse intensity or duration.

The motion of the nanocrystal inside the radio-frequency trap is described by the Mathieu equations [3, 4]:

$$\ddot{x} + (a + 2q\cos 2\tau)\, x = -f_R W_x(\tau),$$
$$\ddot{y} - (a + 2q\cos 2\tau)\, y = -f_R W_y(\tau),$$

where a and q are the DC and AC trap parameters, $f_R = 4\Delta p/(M\rho_0^2\Omega^2\tau_R)$ is the dimensionless force, Ω is the trap frequency, and $\tau_R = t_R\Omega/2$. $W_{x,y}(\tau)$ is the shape of the Raman pulses from corresponding directions:

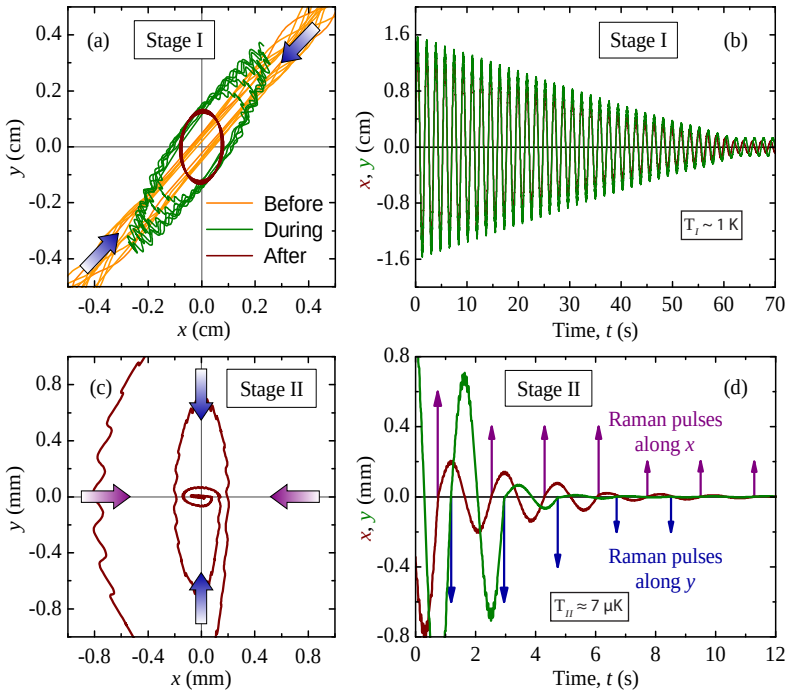
$$W_{x,y}(\tau) = \sum_n W_0[\tau - nT_R(\tau)]$$

Here $W_0(\tau)$ is the shape of a single pulse and $T_R(\tau)$ is the time-dependent period of the pulses, which has a lower limit determined by the lifetime of the level 2, ~10^{-3} s.

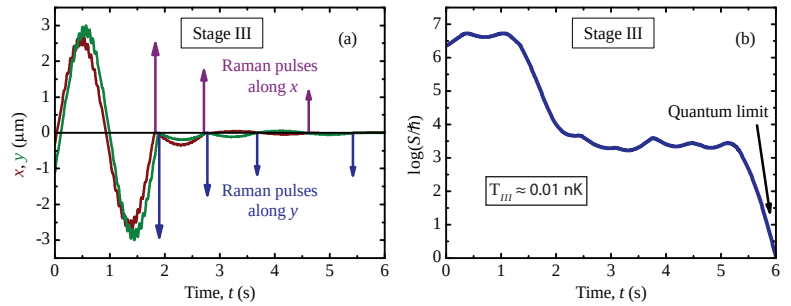
Note that we ignore the recoil from the spontaneous IR emission, because it is much smaller than the UV recoil. Still, it sets a **fundamental limit** on the external temperature of the nanocrystal during the cooling:

$$T_{IR} = \frac{\hbar^2 k_3^2}{2Mk_B} \sim 10^{-13}\ \text{K},$$

where k_3 ~ 10 μm^{-1} and M ~ 10^{-15} g. We do not reach this limit during the cooling, so that the influence of IR photons can be safely ignored.



Path functions (a, c) and time traces (b, d) of the doped nanocrystal during Stage I and II of optical cooling. $n_{Yb} = 10^4$ (doping degree $\varepsilon \approx 0.12\%$), trap parameters are $a = 0$ and $q = 0.1$, and the radio frequency is $\Omega = 100$ Hz. During Stage I Raman pulses have a constant period of 1.7 s, duration of 1 μs, and initial intensity corresponding to Rabi frequency of g = 15 MHz. In panel (d), Stage II, upward and downward arrows indicate aperiodic Raman pulses coming in the x- and y-directions: large, medium, and small arrows correspond to the Rabi frequencies of 2.5, 1.1, and 0.6 MHz.



Time trace (a) and normalized mechanical action S (b) of the nanocrystal at Stage III of the cooling. Trap frequency is linearly reduced from 100 to 20 Hz. In panel (a), large, medium, and small arrows correspond to the Rabi frequencies of the Raman pulses 845, 475, and 320 Hz, respectively.

When the **external temperature** of the nanocrystal is reduced to 0.01 nK at the end of Stage III, its de Broglie wavelength becomes comparable to its size. The nanocrystal can then enter a **macroscopic quantum state**, in which the whole nanocrystal moves as a single quantum particle. This process is well illustrated by the nanocrystal's mechanical action $S = M\,\Delta r\,\Delta v$, where Δr and Δv are the amplitudes of the coordinate and velocity oscillations. The action approaches the value of a single quantum of action, and the nanocrystal becomes an externally quantum object.

[1] Ivanov A., Rozhdestvensky Y., Perlin E. Vibronic model of laser cooling with stimulated Raman adiabatic passage pumping for Yb^{3+}-ion-doped crystals //J. Opt. Soc. Am. B. – 2016. – V. 33. – N. 8. – pp. 1564-1573
[2] Ivanov V. S., Rozhdestvensky Y. V., Suominen K. A. Theory of robust subrecoil cooling by stimulated Raman adiabatic passage //Phys. Rev. A. – 2012. – V. 85. – N. 3. – p. 033422
[3] Paul W. Electromagnetic traps for charged and neutral particles //Rev. Mod. Phys. – 1990. – V. 62. – N. 3. – p. 531.
[4] Rozhdestvensky Y. V., Rudyi S. S. Nonlinear ion dynamics in a radiofrequency multipole trap //Tech. Phys. Lett. – 2017. – V. 43. – N. 8. – pp. 748-752.

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