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Spatial separation of atomic states in a laser-cooled ion crystal

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We have produced a laser-cooled ion crystal in a linear Paul trap containing several hundred Ca^+ ions. A portion of the ions can be excited by an additional laser to a long-lived metastable energy level that decouples them from the cooling laser radiation. The light pressure acting upon the remaining laser-cooled ions forces them into the direction of the laser beam, while the ions in the metastable state drift to the crystal side that points toward the cooling laser. Depending on the number of ions in the crystal and on the fraction of the excited ions, the spatial separation of the ions in the metastable state and the ions in the cooling cycle can be as large as several hundred micrometers. [S1050-2947(98)50407-6]

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Ions, stored in Paul or Penning traps can exhibit phase transitions between a gaslike behavior and a crystalline structure if they are sufficiently cooled by laser radiation so that the Coulomb energy between adjacent particles is much stronger than the thermal kinetic energy. Such crystals have been observed in recent years in different laboratories [1–6]. In these experiments those ions that offer a two-level system and strong cooling transitions such as Mg^+ or Be^+ have been investigated. More recently, Ca^+, a heavier ion of the alkaline earth, has been successfully crystallized [7,8]. It exhibits a three-level system and requires two lasers for a continuous cooling cycle.

Various types of traps have been employed in the investigations of ionic crystals. Penning traps [4,5] with static electric and magnetic trapping fields require low cooling power because of the virtual absence of heating processes. Three-dimensional Paul traps [1,7] only allow rather small crystals to be formed, because heating from the rf trapping field increases with the distance from the trap center. Linear Paul traps [2,3,6,8] have the advantage of having a field-free region along the trap axis that favors the formation of a linear crystalline chain of ions. Recently the formation of large ion crystals in a linear trap containing several 10^4 ions has been reported [6].

The formation of ionic crystals can be described by molecular-dynamics simulations [9] that predict the existence of parallel shells. This has actually been observed in experiments [3,4] and one of the objectives of investigations of ionic crystals is to test such calculations. The results depend critically on details of the heating process and the numerical accuracy. A second interest in linear ion crystals arises from the fact that they are considered potential candidates for the experimental realization of quantum gates [10,11,8] that are based on the entanglement of internal and external degrees of freedom of the trapped ions. Finally, ion crystals promise further improvement in the precision of spectroscopic measurements [12,13].

In this Rapid Communcation we report on a novel effect on an ion crystal. We have observed a spatial separation of an ionic ground and a long-lived metastable state. This is caused by light-pressure-enforced drift in an ion crystal of moderate size. It requires the existence of a metastable state in the ion under investigation that is not coupled to the cooling transition. Such levels exist in some alkaline-earth ions such as Ca^+, Sr^+, and Ba^+, and also in Tl^+, In^+, Hg^+, and others. In our experiment we use a linear Paul trap made of four parallel cylindrical electrodes. Radial confinement is achieved by a rf quadrupole field. The electrodes are segmented into three parts so that a positive dc potential can be added to the outer segments to provide axial confinement.

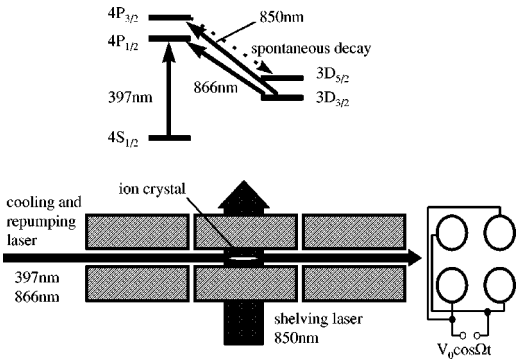

FIG. 1. Partial level scheme of Ca^+ and experimental setup showing the segmented trap electrodes, the cooling and repumping lasers parallel to the trap axis, and the shelving laser, which excites ions to the $4P_{3/2}$ level from where they decay into the metastable $3D_{5/2}$ state. The shelving laser may also be parallel to the trap axis.

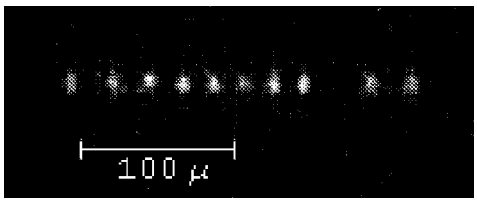

FIG. 2. Small linear ion crystal along the trap axis. The crystal contains 11 ions including an impurity ion that is not excited by any laser but cooled by Coulomb interaction with the remaining ions in the crystal. The impurity ion changes place at a typical rate of 1/4 sec^{-1}.

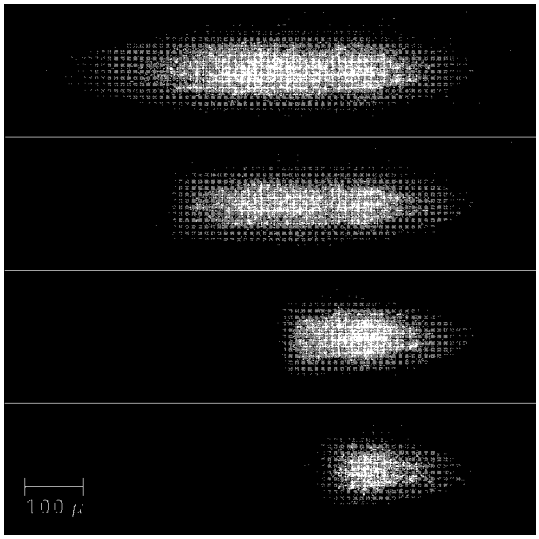

FIG. 3. Moderate size ion crystal containing about 400 ions showing a shell structure. In the picture at the top, only the cooling and repumping lasers are present. In the following pictures an additional shelving laser excites a fraction of the ions in the crystal to a long lived metastable state. Those ions aggregate at the left side of the crystal, which points in the opposite direction to the cooling and repumping lasers. From top to bottom the shelving laser power is increased (0.4, 0.8, and 1.3 μW, respectively).

The trap electrodes have a diameter of 6 mm and a closest distance from the center of 2.7 mm. The trap is driven by a 2-MHz rf voltage of typically 50 V amplitude. Figure 1 shows a scheme of our experimental setup. We use Ca^+ for our experiment. The ions are created by electroionization of an atomic beam inside the trap. Irradiation by a slightly red detuned laser at the $4S_{1/2}$-$4P_{1/2}$ resonance transition (397 nm) and a repumping laser at the $3D_{3/2}$-$4P_{1/2}$ transition (866 nm) cools the ions. At sufficiently high laser power, crystallization occurs that is detected either by a characteristic kink in the fluorescence intensity [14] or by direct observation with a charge-coupled-device (CCD) camera. The spatial resolution of our optical detection system allows direct observation of individual ions up to a number of about 50, while for higher ion numbers the interionic distance becomes too small to be resolved. The number of ions can then be estimated from the total amount of fluorescence. If we tune a third laser to the $3D_{3/2}$-$4P_{3/2}$ transition at 850 nm (see level scheme in Fig. 1) some ions may spontaneously decay from the $4P_{3/2}$ state into the metastable $3D_{5/2}$ level, whose lifetime has been determined to be about 1 sec [15]. During the time which the ions spend in the $3D_{5/2}$ state they are not subjected to any laser radiation and do not emit fluorescent light. They remain, however, in the crystal and are cooled by Coulomb interaction with the neighboring ions. Consequently, they would appear as dark spots inside the crystal. This is, in fact, the case in a small linear crystal that forms along the trap axis (Fig. 2). Diffusion of the dark spots inside the crystalline chain can be observed by the change of position to a neighboring ion. This takes place at a typical rate of 1/4 $\sec^{-1}$.

If, however, the crystal contains many ions, the diffusion rate is much faster due to the higher temperature in larger crystals and a lower two-ion exchange energy barrier. The drift is caused by the light pressure acting on those ions, that are subjected to the continuous cooling laser radiation. This force pushes the ions in the direction of the laser beam. The ions excited to the metastable $3D_{5/2}$ level, however, do not experience any light force and consequently aggregate at the side of the crystal that is directed against the laser. The spatial separation of the ions in the $3D_{5/2}$ state from those in the cooling cycle takes place in a time that is faster than the time resolution of our CCD camera (20 msec). A desirable consideration of the diffusion process would require experimental data on the drift rate that cannot be obtained by our present equipment. The number of ions that are driven to the $3D_{5/2}$ state can be varied by changing the power of the 850-nm laser. We observe a sharp boundary between the excited and the metastable parts of the crystal. Figure 3 illustrates this in a series of pictures of a crystal, containing about 400 ions, at different powers of the 850-nm laser. The direction of the laser beam is perpendicular to the trap axis and the cooling laser direction. Similar results have been obtained when all the lasers are directed colinear along the axis. The existence of a sharp boundary indicates that the nonvisible part of the ions remains crysallized, since the charge distribution of the bright parts does not change when the separation takes place. Symmetry considerations suggest that this would not be the case if the dark ions were to form a noncrystallized cloud.

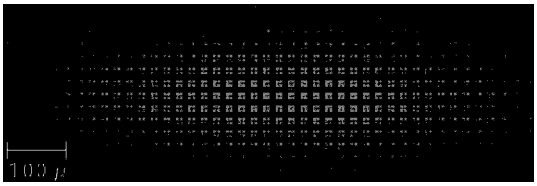

FIG. 4. Excitation of a cooled but not crystallized ion cloud by a shelving laser of the same intensity as in Fig. 3, second from bottom. No separation of bright and dark parts takes place. Instead, the whole crystal appears darker than it appears without the shelving laser.

With increasing power, the dark part of the crystal increases. If the number of cooled ions in the crystal becomes too small, the crystal melts because the cooling power is no longer strong enough to overcome heating effects that act on all of the ions in the crystal. Heating is caused by collisions with background gas atoms at our residual pressure of 2×10^{-10} mbar and a subsequent pickup of energy from the rf trapping field. Decreasing the 850-nm laser power reestablishes the crystal. The shift in the position of the boundary between the two parts is completely reversible.

When we perform a similar experiment at cooling laser powers that are not sufficient for crystallization, the observation is different: the thermal motion of the ions in the cooled but not crystallized cloud is much stronger than the light induced drift and no spatial separation takes place. Instead, the whole ion cloud appears darker, depending on the number of ions driven into the metastable $3D_{5/2}$ state (Fig. 4).

Since the separation between adjancent ions in the crystal is of the order of 10 μm, the separation between the centers of the dark and bright parts of the crystal is as large as several hundred μm. This allows us to easily visualize these parts by the detection optics. We have complete population

inversion in the dark part of the crystal, which could give rise to lasing if the trap is placed between high reflecting mirrors. It may also allow precision spectroscopy on the metastable state without perturbation from scattered light background and separation of different isotopes in the trap.

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