Laser cooling

The mechanical forces exerted by light can dramatically lower the temperature of a sample of atoms or ions, allowing very-high-resolution spectroscopic measurements and ultralow-temperature atomic physics experiments.

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In the photograph on the opposite page we see a single mercury ion held nearly at rest in an electromagnetic "trap." Physicists have seen individual atoms before, in arrays imaged by field ion microscopes and more recently by vacuum tunneling microscopes, but what we see here is different. It graphically demonstrates a physicist's ideal: holding a single isolated atom nearly at rest for careful examination.

A technique now commonly called laser cooling made the photograph possible by reducing the mercury ion's kinetic energy. This cooling not only limited the ion's movement, but also sharpened its spectral features by reducing Doppler broadening, enhancing the scattering of laser light tuned to one of its transitions. Several laboratories now use lasers to cool ions and neutral atoms to kinetic energies corresponding to temperatures near a millikelvin. (See PHYSICS TODAY, September 1986, page 17.)

As originally proposed in 1975 by Theodor Hänsch and Arthur Schawlow at Stanford University and independently by Wineland and Hans Dehmelt at the University of Washington, laser cooling can substantially reduce Doppler effects in high-resolution spectroscopy. The technique should eventually reduce inaccuracies in spectroscopy to 1 part in $10^{10}$ or better for single trapped ions. More accurate atomic clocks are an obvious prospect. Other applications include tests of gravitational interactions. Laser-cooled neutral atomic beams may finally allow realization of Jerrold Zacharias's 1953 proposal for an "atomic fountain" experiment. Here one would achieve long observation times by directing a slowed atomic beam upward and letting gravity return it to near its original position. Even with moderately slowed atomic beams, the velocity compression achieved from laser cooling will greatly reduce the uncertainty in the second-order, or time dilation, Doppler shift.

Laser cooling has a potential role in many other experiments. It may give us ways to:

- Study collisions between very cold atoms or ions. Such studies should give detailed information on interactions, with very high energy resolution.
- Study atom–surface collisions at low temperature. It may be that at sufficiently low temperature, atoms will "bounce" with minimal perturbation to their structure. This would allow the construction of nearly ideal boxes for storing atoms for use in spectroscopy and other experiments.
- Focus atomic beams. Laser beams directed transversely to an atomic beam act as lenses. These lenses are dissipative in the direction normal to the atomic beam, so beam focusing is not limited by Liouville's theorem. Laser cooling may also be used to advantage in ion storage rings.
- Manipulate antihydrogen, which, if produced, must be used efficiently.
- Obtain unique states of condensed matter. Observing liquid and solid plasmas is a possibility, as we discuss below.
- Observe Bose condensation of hydrogen or other atoms. This phenomenon may eventually be observed by cooling atoms held in a suitable "trap."

In many cases laser cooling may be the only practical way to control the velocity distribution of a sample of ions or neutral atoms.

We begin this article with an explanation of how light imparts mechanical forces on atoms. Because these forces are the same on both ions and neutral atoms, we will take the term "atom" to include both unless we specifically state otherwise. We then discuss how one can use these forces to reduce the
kinetic energy of a sample. Finally, we discuss what can be achieved in the laboratory.

Early history. The study of the mechanical forces that light exerts on matter has a long history. In 1873 James Clerk Maxwell used the theory of electromagnetism to calculate the force on a solid body due to the absorption or reflection of a beam of light. In the early 1900s quantitative measurements of the force exerted by light on solid bodies and gases verified Maxwell’s radiation pressure calculation. In 1917 Albert Einstein used quantum theory to calculate the influence of the electromagnetic radiation field on the motion of molecules. He showed that the light pressure causes molecules to come into thermal equilibrium with a radiation field if that field has the Planck spectrum. Aside from showing the consistency of quantum theory and statistical mechanics, this calculation was important in establishing the quantum nature of light, because it was necessary to assume that the molecule emits radiation as a discrete bundle with a definite energy and momentum, and not as a spherical wave.

In 1933 Otto Frisch did the first experiment to show directly the momentum transferred to an atom by the absorption of a photon. In this experiment, light from a sodium resonance lamp deflected a beam of sodium atoms. When tunable lasers became available in the 1970s, experiments of this sort were repeated. Due to the much higher spectral intensities available, an atom could be made to absorb many photons one at a time, but at a high rate, resulting in larger deflections. Around this time physicists made proposals to use intense, resonant optical fields to manipulate atoms in various ways, such as accelerating them or trapping them in optical potential wells. Among those who first recognized the possible applications of resonant laser radiation pressure on atoms were Arthur Ashkin in the United States and several scientists in the Soviet Union. In his 1950 paper on optical pumping, Alfred Kastler suggested some ways of using light to cool or heat atoms. These ideas are related to laser cooling but are difficult to realize in practice.

Optical forces on atoms
The force that light exerts on atoms is often conceptually divided into two parts. These are called the light pressure or scattering force and the gradient or dipole force. At least in some simple cases, one can distinguish these forces clearly. In the general case, and particularly for intense fields, the simple descriptions of the forces and their fluctuations break down, and a more fully quantum mechanical description is required.

We can understand the scattering force as the momentum transferred to the atom as it scatters a photon. The average scattering force is in the direction of propagation of the light and is equal to the product of the momentum $\hbar k$ per photon and the photon scattering rate. The photon wavevector $k$ has magnitude $\frac{2\pi}{\lambda}$, where $\lambda$ is the wavelength of the light. The average force reaches a maximum when the light is resonant with an atomic transition. The scattering force fluctuates because the photons scatter at random times and because the direction of the re-emitted photon, and hence the direction of the recoil momentum due to this re-emission, is also random.

To see how large the scattering force can be, let us consider a specific example. Assume that the light from a single laser beam is resonant with the lowest-frequency atomic transition and that the light is intense enough to saturate the transition, that is, that the rate for stimulated emission exceeds the spontaneous decay rate. When saturated, the atom spends about half of its time in the excited state. Therefore the average force on...
Laser cooling mechanism for free or weakly bound atoms or ions.

"Weakly bound" means that the oscillation period of the trapped atom or ion is longer than the lifetime of the upper state of the cooling transition. The frequency \( \omega_q \) of incident photons is assumed to be less than the rest frequency \( \omega_0 \) of the atomic transition. At a velocity where the atom's transition frequency \( \omega_q \) is equal to \( (1 - k \cdot v)\omega_0 \), the atom resonantly scatters photons. When the photon wavevector \( k \) is antiparallel to the atom's velocity \( v \), the average reduction in the atom's velocity is \( \hbar k / m \) per scattering event, where \( m \) is the atom's mass.

The origin of the gradient or dipole force has a strongly resonant character, but unlike the scattering force, it is dispersive in nature. Its sign is such that it attracts an atom to a region of high light intensity if the frequency of the light is below the atomic resonance and repels an atom if it is above. That is, the electric polarizability is positive below resonance and negative above resonance. To this extent, the atom is like a charged harmonic oscillator—an electron bound to a positive core by an external force if the frequency is below resonance, and 180° out of phase if the frequency is above. If the electric field is spatially inhomogeneous, then averaged over one cycle of the optical radiation a net force on the atom can result. Some proposals for trapping atoms with optical fields are based on the use of the dipole force.

Laser cooling of free atoms

Laser cooling based on the use of the scattering force can be explained as follows: Consider an atom with a strongly allowed resonance transition. For simplicity, let \( k \) be the lowest-frequency transition, so that if this transition is excited, the atom must decay to the ground state. A laser beam whose frequency is close to but lower than the atomic resonance frequency irradiates the atom. If the atom is moving against the laser beam, then the frequency of the light in the rest frame of the atom is Doppler shifted toward resonance. Hence, the scattering force is higher for an atom moving against the laser beam, and the atom's velocity is damped. In this way, the velocity of an atomic beam can be reduced substantially.

If only one laser beam is used and the atom is otherwise free, the atom will eventually turn around and move away parallel to the beam. Therefore, for a gas of atoms, if another laser beam of equal intensity and frequency but opposite direction is introduced, atoms with their velocities in the other direction also have their speeds reduced. The intensities must be equal for the average scattering force on a motionless atom to be zero; if the intensities are not equal, the atoms will have a net drift velocity. One can obtain cooling in all directions by using three orthogonal pairs of counterpropagating laser beams. However, because of the inherent fluctuations of the scattering force, the velocity is not damped to zero.

The theoretical minimum temperature that can be obtained is given by a balance between dissipation and fluctuations. Assume the laser intensity is below saturation, even when tuned to resonance. We also assume that the recoil energy \( R \), given by \( \hbar k v / 2m \), is less than \( k T \). The recoil energy is the kinetic energy that an initially motionless atom of mass \( m \) would have due to its recoil after emitting a photon of wavevector \( k \). The minimum temperature \( T_{\text{min}} \) is achieved when the laser frequency is tuned below the atomic resonance frequency by an amount equal to \( \gamma / 2 \), in which case

\[
k T_{\text{min}} = \frac{1}{2} \hbar \gamma \quad (1)
\]

For a decay rate \( \gamma \) of \( 10^8 / \text{sec} \), the minimum temperature is about 0.38 mK.
Absorption as a function of laser frequency. In the sideband cooling limit, the oscillation frequency \( \omega_c \) of the atom in the trap is assumed to be larger than the radiative decay linewidth \( \gamma \). Therefore the absorption spectrum consists of a “carrier,” or recoilless line, at the atom's rest frequency \( \omega_0 \) plus Doppler-effect sidebands separated by the frequency \( \omega_c \). If the laser is tuned to \( \omega_0 - \omega_c \), the atom absorbs photons of energy \( \hbar(\omega_0 - \omega_c) \) and emits photons of average energy \( \hbar(\omega_0 - \omega_c) = R \), where the recoil energy \( R \) is \( \frac{\hbar \omega_0^2}{2m} \). When \( R \) is much less than \( \hbar \omega_0 \), there is an energy deficit of approximately \( \hbar \omega_0 \) per scattering event, causing a decrease in the kinetic energy of the atom.

To cool a trapped atom, one tunes a narrow-band laser to a sideband on the low-frequency side of the unshifted resonance, for example, to a frequency \( \omega_0 - p \omega_c \), where \( p \) is an integer. The atom makes transitions to the upper electronic state, decreasing its vibrational energy, by absorbing photons of energy \( \hbar(\omega_0 - p \omega_c) \). When the laser makes a transition back to its ground electronic state, it may, in general, either increase or decrease its vibrational energy, but the average change in the vibrational energy is equal to the recoil energy \( R \). When \( R \) is less than \( \hbar \omega_0 \), cooling occurs.

Consider a particular case. A single ion is trapped in a nearly isotropic three-dimensional harmonic potential well, a situation that is approximated by an ion in an rf trap, or Paul trap, with normal-mode vibrational frequencies all approximately equal to \( \omega_c \). A quantum state of the atom in the well is identified by its internal quantum numbers and the set of harmonic oscillator quantum numbers \( \{ n_x, n_y, n_z \} \) corresponding to the well. Three laser beams propagate along the \( x, y \), and \( z \) axes, and each is tuned to the corresponding first lower sideband. We assume that the recoil energy \( R \) is much less than the energy \( \hbar \omega_0 \), a condition that is not hard to satisfy in practice. In the steady state, the mean values of the quantum numbers are

\[
\langle n_x \rangle = \langle n_y \rangle = \langle n_z \rangle = \langle n \rangle = \langle \frac{1}{2} \rangle \langle \gamma^2 / \omega_c \rangle \ll 1
\]

(2)

There are two reasons that the mean values do not go to zero with laser cooling. First, the average change \( R \) in the atom's vibrational energy after it
emits a photon is positive. Second, there is some probability of driving a transition that leads to an increase in vibrational energy on absorption rather than a decrease, because the neighboring sidebands, although far from resonance, still have finite intensities. The simple theory just outlined applies when the cooling transition is not saturated. Markus Lindberg at the University of Frankfurt and Juha Jaavanainen and Stig Stenholm at the University of Helsinki have calculated the steady state for an arbitrary ratio of natural linewidth to motional frequency and also for arbitrary laser intensity. As in the case of free atoms, the lowest temperatures are achieved in the limit of low intensity, so the simple theory is usually adequate.

Experiments on trapped ions

It is perhaps not surprising that the first laser cooling experiments were done in 1978. At the National Bureau of Standards in Boulder, Robert Drullinger, Fred Walls and Wine land demonstrated cooling by a direct observation of ion temperature, which they determined from the currents that ion motion induces in trap electrodes. They observed the cooling of Mg+ ions to 40 K in a Penning trap. At the same time in Heidelberg, Werner Neuhäuser, Martin Hohenstatt, Peter Toschek and Dehmelt demonstrated cooling of Ba+ ions in a Pen trap by observing the ions’ increased storage time in the trap.

In subsequent experiments at these and other laboratories, physicists have measured temperatures on the order of 10 mK or less. They have typically determined the temperatures by measuring the contribution of Doppler broadening to spectral lines. For strongly allowed electric dipole transitions, this method is not very sensitive at low temperatures because Doppler broadening contributes only a small fraction of the overall linewidth. For positive magnesium-24 ions at 1 mK, for example, the Doppler broadening of the first resonance line (at 280 nm) is only 0.5 MHz, while the natural radiative linewidth $\gamma/2\pi$ is 43 MHz. One realizes a more sensitive measurement of temperature by probing a transition with a linewidth $\gamma$ that is much less than the motional oscillation frequency $\omega_m$ of an ion in the trap. Here one can use the strength of the motional sidebands to determine the ion temperature. So far, only cooling in the limit $\gamma \ll \omega_m$ (equation 1) has been demonstrated for ions, but if cooling in the limit $\omega_m \gg \gamma$ is used, it should be possible to reduce the kinetic energy to nearly the zero-point energy (equation 2). Laser cooling of trapped ions is now done in laboratories at NBS in Boulder, Hamburg University, the University of Washington in Seattle, the University of Paris in Orsay, the National Physical Laboratory at Teddington in England and the Max Planck Institute at Garching in West Germany. Other laboratories are setting up similar experiments.

Single ions. Single, laser-cooled, trapped ions are interesting because they provide a simple system for study. First, the oscillation frequencies of an ion in a trap are nearly harmonic, which makes spectra of single ions particularly simple. In contrast, if two or more ions are stored together in a trap, the oscillation frequencies of the individual ions are dominated by ion-ion interactions at low temperature. Second, the lowest possible kinetic energies, given by equations 1 and 2, are obtained for single ions. This is because part of the motion in the Paul trap (the rf-driven micromotion) or in the Penning trap (the magnetron or rotation motion) is nonthermal and not affected in the same way by laser cooling. The kinetic energy in these nonthermal motions can be minimized for single ions. Finally, the simplicity of single trapped ions allows a straightforward comparison of laser cooling theory and experiment.

Single ions are also interesting from the standpoint of spectroscopy because the perturbations in spectral measurements can be extremely small or well controlled. Historically, the most difficult problem in high-accuracy ion trap spectroscopy has been minimizing the second-order Doppler frequency shift, which is due to relativistic time dilation. Reducing this systematic effect requires low temperatures. Experimenters have achieved temperatures of about 10 mK or less with single Ba+ ions at Hamburg University, with single Mg+ and Ba+ ions at the University of Washington, with single Mg+ and Hg+ ions at NBS in Boulder and with single Mg+ ions at Garching. At a temperature of 10 mK, an ion with mass 100 u has a fractional second-order Doppler shift of $-1.4 \times 10^{-17}$, which is almost negligible. Single la-
Laser-cooled ions have also allowed detailed studies to be made of the interaction between atoms and radiation, as seen, for example, in quantum "jumps" and in photon "antibunching," an effect in which the distribution of arrival times of fluorescence photons at a detector is nonclassical because atoms can emit only one photon at a time.

**Liquid and solid plasmas.** Large numbers, or "clouds," of trapped ions are more properly referred to as nonneutral ion plasmas.\(^{17}\) These plasmas are interesting because, unlike fusion plasmas, they can reach a global thermal equilibrium. When these plasmas can be laser cooled, the densities are high enough—above \(10^7/\text{cm}^3\)—and the temperatures are low enough—less than 10 mK—that the plasmas become strongly coupled and should show liquid and solid behavior.\(^{17}\) It should be possible to obtain non-neutral plasmas whose dynamics are dominated by quantum effects—for example, a positron plasma "sympathetically" cooled with laser-cooled ions of beryllium-9, as described below.

Even though single ions give the lowest temperatures, larger samples of laser-cooled trapped ions have already yielded interesting spectroscopic results. Spectroscopy\(^{11}\) in the rf and microwave regions has featured linewidths of 0.01 Hz and fractional inaccuracies as small as 1 part in \(10^{13}\).

**Sympathetic laser cooling.** Unfortunately, direct laser cooling of ions and neutral atoms is relatively easy only for a very few elements. Sodium is often the practical choice for neutral atom researchers; singly ionized magnesium, which is isoelectronic with sodium, is a favorite choice for ion trappers. So far both groups have avoided working with molecules.

One can extend laser cooling to other species of ions by storing two ion species in the same trap. The species that is easy to laser cool will cool the second species through Coulomb collisions, making the second species available for high-resolution spectroscopic investigation or other experiments. Under typical conditions, ion–ion thermalization by Coulomb coupling takes place in less than a second. As a demonstration of this technique, \(\text{He}^+\) ions have been cooled to less than 1 K by laser-cooled ions of beryllium-9 in a Penning trap.\(^{15}\) It should be possible to extend sympathetic cooling in order to cool neutral atoms or molecules by ion–atom or atom–atom collisions.

**Experiments on neutral atoms**

If neutral atoms could be trapped easily and held in thermal isolation from the surroundings, then laser cooling them would be similar to laser cooling ions. Unfortunately, neutral-atom traps with the required thermal isolation are not very deep. For example, magnetic traps, which convert the atom's kinetic energy into internal, Zeeman energy, at best have a depth of about \(5 \times 10^{-3}\) eV, or 8 K. This assumes that an atom with a magnetic moment equal to one Bohr magneton is captured in a magnetic well that is 10 T deep. Ion traps, in contrast, can be kilovolts deep.

Most neutral-atom laser cooling experiments start with an atomic beam of an alkali such as sodium and slow and cool the beam with a counterpropagating laser beam.\(^{16}\) Because sodium atoms are emitted from an oven source with a velocity of about 1000 m/sec, the slowing and cooling must be done very efficiently to stop the atoms before they strike some portion of the apparatus. Even at maximum cooling efficiency, this requires about 50 cm. The first neutral-atom cooling experiments were reported by S. V. Andreev, Victor Balykin, Vladilen Letokhov and Vladimir Minogin in Moscow and by William Phillips and Harold Metcalf at NBS in Gaithersburg, Maryland. The Moscow group saw beam slowing and velocity compression due to a counterpropagating fixed-frequency laser beam. If a fixed-frequency laser beam is used, atoms with velocities that put them in resonance with the laser through the first-order Doppler frequency shift are efficiently slowed. However, these atoms are soon slowed enough that they are Doppler shifted out of resonance with the laser beam and the slowing is greatly reduced. Similarly, faster atoms are slowed only very slightly. The result is that a hole is carved out of the atoms' velocity distribution at a velocity corresponding to the Doppler-shifted laser frequency, and the affected atoms tend to bunch at slightly lower velocity.

To make the cooling more efficient, one needs a way to keep the atoms in resonance with the laser while they are slowed down. The NBS group accom-
plished this by continuously tuning the atoms’ frequency. Investigators from the group directed the atomic beam down the bore of a solenoid whose magnetic field varied with position. The field varied in such a way as to keep the slowed atoms, whose Zeeman frequency shift depended on position in the magnet, continuously in resonance with the fixed-frequency laser beam. This enabled more of the atoms to be slowed to lower velocities. Recent experiments at MIT have also used this technique. Another way to keep the atoms tuned to the frequency of the laser is to sweep, or “chirp,” the frequency at an appropriate rate. The first demonstration of beam slowing by this technique was made in 1983 at NBS, Gaithersburg. Physicists at the Joint Institute for Laboratory Astrophysics later used the same technique to slow and stop atoms.19 Subsequent experiments at Bell Laboratories,20 Bonn University, the State University of New York at Stony Brook, the École Normale Supérieure in Paris and by another group at JILA21 have employed laser chipping for cooling. The latter experiments at JILA accomplished slowing and cooling using diode lasers, demonstrating that cooling at a relatively low cost is practical. With these techniques, atoms are now stopped (and even turned around) in several of the above laboratories. Temperatures of the stopped atoms are typically a few tens of millikelvins, and gravity starts to play a role because the slowed atoms are in the apparatus long enough to fall a significant distance.

The group at Bell Labs has demonstrated the lowest temperatures yet achieved through laser cooling.22 Recently enabled by a chirped laser,23 they precool sodium atoms from a pulsed laser ablation source. They then subject these atoms to three mutually orthogonal, intersecting pairs of counterpropagating laser beams of cross section about 1 cm2, tuned to achieve minimum temperature. The region of intersection features a laser cooling damping force in all directions. The accompanying recoil heating causes the atoms to undergo Brownian motion. The diffusion time for atoms to leave this “optical molasses” can be on the order of 1 sec, which is plenty of time for many experiments. By rapidly turning the molasses off and on, the Bell Labs group was able to measure the velocities of the atoms and determine that the temperature was 240 μK (with a probable range of 180–440 μK), which is in agreement with the limit implied by equation 1.

Now that sources of slow atoms exist, they can be used as injection sources for the relatively shallow neutral-atom traps. Phillips and his coworkers were the first experimenters to capture atoms in a magnetic trap.24 (Wolfgang Paul and his collaborators had held neutrons in a magnetic trap in 1978.) The MIT group recently used a different magnetic trap to hold atoms.25 The Bell Labs group has used their optical molasses to inject atoms into a gradient-force or dipole-force laser trap.26 In Moscow, Balakin and A. I. Sidorov have demonstrated cooling in two dimensions by collimating atomic beams.27 The Paris group recently demonstrated stimulated cooling by its effect on the collimation of atomic beams.28

In principle, one could apply laser cooling to normal solids. For example, crystals that are doped with impurity ions could be driven on the lower phonon sidebands of certain transitions. Nonradiative decay from the upper level of the cooling transition, which shows up as heat, may dominate the cooling process. It is interesting to examine the economics of large-scale laser cooling. If substantial cooling requires about 106 scattering events per atom, then cooling one mole of material will require more than 106 joules of laser energy. Therefore, laser cooling may not be practical on a large scale, but in many cases it may be the only way to lower or manipulate the velocities of atomic samples. Judging by the number of laboratories now using laser cooling or setting up experiments using laser cooling, it appears likely that the technique will have many interesting applications in the future.

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References

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