# Chapter 16 Laser Cooling of Atoms and Ions

#### 16.1 Introduction

As generators of intense, spectrally pure radiation at optical frequencies, lasers serve not only as stabilized sources for frequency standards in that region of the spectrum, but equally important, they have changed the entire embodiment of the *microwave* cesium standard. They provide a means of slowing down the thermal motion of atomic particles. This is critically important when the resonance frequency of a transition in an atom or ion is used as a reference, since it is essential that the Doppler frequency shifts due to the particle motion be eliminated as far as possible. This can be accomplished by what is now called *laser cooling*, a truly remarkable technique, which we take up in this chapter. It is a technique that has made it possible to reach particle velocities corresponding to temperatures only a small fraction of a degree above absolute zero, where all thermal motion ceases.

An understanding of how laser cooling of atomic particles is possible begins with the realization that the interaction of a light beam with such a particle cannot only affect the internal motions within the particle, but also its center-of-mass motion. This ultimately derives from the fact that a light wave not only conveys energy but also carries with it momentum, both linear and angular. Long before the famous  $E = mc^2$  formula of Einstein's theory of relativity was established, giving the equivalence of mass and energy, Maxwell's classical electromagnetic theory of light predicted that light falling, for example, upon a reflecting surface exerts a pressure on it, proportional to the intensity of the light. Even before that, the question of whether a light beam carried momentum was a subject of experimentation and philosophical debate for two centuries. The central issue was whether a light beam was a wave in the *ether* or a stream of minute particles, as held by the so-called *corpuscular theory*. Adherents of the latter, which incidentally included Isaac Newton, believed that the correctness of their theory would be confirmed if it could be shown that a light beam possesses momentum, as would a stream of any material particles. To resolve the question, many investigators directed powerful light beams at delicately suspended objects and looked for any minute movement in those objects that could be attributed to the impact of particles. In one such experiment carried out in mid-18<sup>th</sup> century, a very thin sheet of copper was delicately suspended and a beam of sunlight was directed at it by means of a mirror; a deflection of the copper sheet was observed, and although the presence of heating of the air near the surface of the copper was recognized as contributing to that deflection, there were some who believed that nevertheless the observations confirmed the mechanical impact of the light beam on the surface. Subsequent experiments by other investigators failed to show any movement in the irradiated object distinguishable from the effects of heat; this shows, it was concluded, that light cannot consist of particles and must therefore be vibrations in some medium. Among those taking the apparent absence of a deflection as an argument favoring the wave theory was Thomas Young, whose light interference experiments perhaps clouded his judgment. For it had previously been pointed out by Euler, the great Swiss mathematician, that light pressure might just as reasonably be expected from the wave theory as from the corpuscular theory. It is a remarkable historical fact that Euler not only correctly postulated the existence of light pressure but also, following a suggestion by Kepler, developed a theory to account for the tail of a comet based on the pressure of light from the sun acting on small dust particles in its head.

## 16.2 Light Pressure

An argument can readily be made for the existence of light pressure on the basis of Maxwell's electromagnetic wave theory of light. Thus a light beam falling perpendicularly, for example, on a reflecting metal surface must be pictured as subjecting the conducting surface to mutually perpendicular electric and magnetic fields oscillating at optical frequency in a plane parallel to the surface. The electric current produced in the metallic surface by the electric field will be acted on by the magnetic field, producing a Lorentz force perpendicular to both fields, that is, in the direction of the incident wave. The physical origin of this force is identical to the force on a current-carrying conductor in a magnetic field, as in an electric motor. The magnitude of the pressure this causes on the surface is derived in Maxwell's original theory in terms of the mechanical stress transmitted through the *ether* the postulated universal medium. However, with the abandonment of the concept of a universal medium, as required by Einstein's theory, and in order to preserve the conservation law of linear momentum, Maxwell's field equations are now interpreted as leading to the electromagnetic field *itself* carrying momentum. From this it follows that to deflect a light beam requires a force to be exerted, with an equal reaction on the object providing that force; hence the radiation pressure on lightscattering objects. The amount of momentum (per unit volume) represented by *M*,

which we must attribute to the field in a light beam of intensity *I* (watts · m<sup>-2</sup>), can be shown according to classical theory to be given by

$$
M = \frac{I}{c^2},\tag{16.1}
$$

where as usual, *c* is the velocity of light. We can interpret the intensity *I* as the flow at the velocity *c* of energy residing in the field and distributed with density  $\rho_F$ (joules· <sup>m</sup>−3), with a similar interpretation for the amount of momentum *<sup>M</sup>*. The classical result suggests that we must attribute to the field a mass density ρ given by the following:

$$
\rho = \frac{\rho_E}{c^2}.
$$

This is recognized as a special case of Einstein's  $E = mc^2$ , which applies to any form of energy. That it should appear in classical electromagnetic theory is not surprising, since Einstein's theory was constructed on the basis that the way Maxwell's equations transform from one coordinate reference frame to another should be true of all physical laws, including those of mechanics. It was this that necessitated the revolutionary changes in the concepts of space and time that are characteristic of Einstein's theory.

If a light beam of intensity *I* falls on a perfectly absorbing surface, so that the directed energy of the beam is converted to random thermal motion with zero net momentum, there is a continual change in momentum of the beam, which must be taken up by the absorber; hence by Newton's laws of motion there will be a pressure *P* exerted on the surface equal to the rate of change of momentum. Since the amount of momentum change that occurs per unit time over unit area is *Mc*, this being the amount of momentum carried by photons in a cylinder of unit cross section and length equal to the distance light travels in one second, the pressure is simply given by the following:

$$
P = Mc = \frac{I}{c},\tag{16.3}
$$

where *I* (watt · m<sup>-2</sup>) is the beam intensity and *c* is the velocity of light.

The same result immediately follows from the de Broglie formula, which applies to all particles, including photons, namely  $p = h/\lambda$ , where p is the linear momentum of the particle. If we suppose the light beam consists of a stream of photons with a flux density (number of photons crossing unit cross-sectional area per unit time) represented by *j*, then clearly we have  $I = jh\nu$ . Furthermore, the rate at which momentum is lost from the beam over unit area of the surface of an ideal absorber, which by Newton's law is the pressure, is given by  $P = jh/\lambda$ , and the relationship  $P = I/c$  follows.

Numerically, the size of radiation pressure is extremely small on the ordinary scale of things; for example, the pressure due to direct sunlight, whose intensity is on the order of 1000 watts  $\cdot$  m<sup>-2</sup>, is only about 3 × 10<sup>-6</sup> newton  $\cdot$  m<sup>-2</sup>, that is, about  $3 \times 10^{-11}$  times atmospheric pressure. It is little wonder that so much

experimental difficulty was encountered in finding unambiguous evidence of its existence. A sensitive device, now a scientific curiosity called "Crookes' radiometer," was devised to demonstrate radiation pressure by William Crookes in 1875, the year that Maxwell's treatise was published. It consists of light metallic vanes blackened on one face and shiny on the other, delicately balanced and free to rotate inside an evacuated tube. It does indeed spin when exposed to a sunbeam. However, it soon became evident that the observed spinning of the vanes had more to do with thermal effects due to the residual gas surrounding them than to any radiation pressure. Later experiments, however, did confirm the essential validity of the theory.

## 16.3 Scattering of Light from Small Particles

With the advent of the laser it became possible to exploit a different approach to studying light pressure, with far-reaching ramifications. The ability to achieve high intensities of monochromatic light by focusing a laser beam down to diameters on the order of microns meant that the motion of individual small particles could be observed. Moreover, this can be done in highly transparent nonabsorbing materials, simply relying on the reflection and refraction of light to bring about a change in the particle's momentum; this alleviates the problems associated with heating effects. In one such experiment, reported by Ashkin in 1970, small transparent spheres of latex having diameters on the order of one micron ( $10^{-6}$  m) were freely suspended in pure water. An  $Ar^+$  ion laser ion laser beam passed through a glass cell containing the suspension of latex spheres and was manipulated to converge to a radius of about 6 microns on an individual sphere, as shown in Figure 16.1. The strong scattering from the spheres enabled their motion to be observed visually by means of a microscope. It was found that not only will a sphere be driven in the direction of the beam by radiation pressure, but also, if the sphere is initially off the beam axis, it will be drawn toward it. This Ashkin explained using an argument based on the theory of light pressure and the approximate concept of light rays. This is valid to the extent that the wavelength of the light is very much smaller than the radius of the sphere; otherwise, a proper wave solution must be sought.



Figure 16.1 Ashkin's experiment to study forces on latex spheres due to a laser beam



Figure 16.2 The balance of radiation forces acting on a dielectric sphere according to Ashkin

The solution to the problem of the scattering of electromagnetic waves from small particles is of considerable importance in such fields as atmospheric optics and optical astronomy; the case of plane waves scattered by a homogeneous sphere permits a general solution, first given by Mie around 1908. For the purposes of an approximate, qualitative explanation of the behavior of the particles in the laser beam, there should be little error in assuming ray optics. First we note that the refractive index of latex at the wavelength of the laser light ( $n = 1.58$ ) is greater than that for water  $(n = 1.33)$ , and therefore the light falling on a sphere will be converged on the emergent side, as by a thick convex lens, as shown in Figure 16.2. This means that part of the linear momentum of the beam is deflected away from its axis and the remaining component along the axis reduced; therefore, to conserve momentum the latex sphere must experience a force along the axis in the direction of the initial beam. Moreover, if the sphere is off-axis and the intensity of the beam varies over its surface, there will be an asymmetry in the amount of momentum deflected toward the axis versus the amount away from the axis; the result is a net force on the sphere in the direction of increasing intensity, that is, toward the axis. The result would have been in the opposite direction had the sphere been of a lower refractive index than the medium, for example an air bubble.

#### 16.4 Scattering of Light by Atoms

In extending the idea of linear momentum exchange to that between a laser beam and free atoms or ions, we are faced with the need for an altogether different and more complicated quantum description of the interactions involved. One of the important processes that take place has already been met in the optical pumping

of free atoms and ions, namely resonance fluorescence. In this, the atomic particle will interact strongly only if the radiation has a frequency resonant with one of a set of discrete frequencies in its spectrum. Thus resonant absorption of a photon from a beam is accompanied by a quantum transition to a higher energy state, from which the particle ultimately falls back to the ground state, radiating its excitation energy in all directions in a definite angular pattern. It is as though photons were taken out of the beam and scattered in all directions away from the atom or ion. The net transfer of linear momentum resulting from this process is obtained by applying the law of conservation of linear momentum between the initial state of the beam–atom system prior to the interaction, and their final state, long after the interaction, including any photons radiated from the atom. The relative probability of the photon being radiated in the different directions, that is, in the language of antenna engineers, its radiation pattern, must conform to the symmetry laws governing the system. In particular, a free atom *spontaneously* emitting a photon does *not* do so preferentially along any one particular direction; this assumes that in the process of spontaneous emission the atomic particle has no "memory" of the excitation by the laser beam. This certainly would not be the case for stimulated emission.

It does not mean that the radiation pattern must be isotropic, with equal intensity in all directions; however, it does mean that for any given direction the intensity will be equal to that along the opposite direction, as shown in Figure 16.3. The detailed angular distribution of the emitted radiation is complicated, as we saw in the rubidium case, by the fact that it is determined by the change in the *angular* momentum state of the atom or ion accompanying the transition. We recall that a photon is endowed with an intrinsic spin of one unit of  $h/2\pi$ , and the photon– atom system exchanges angular momentum as well as linear momentum. If after the absorption and re-emission of a photon the atom is left in a different angular momentum state, then in order to conserve angular momentum, the radiation



Figure 16.3 The polar diagram giving the radiation pattern from an atom undergoing an electric dipole transition

pattern resulting from the photon–atom interaction must carry the balance of the angular momentum the photon had prior to the interaction. It is this type of exchange on which Kastler's optical pumping technique for magnetic resonance in free atoms is based. In any event, as already stated, there can be no preferred direction of spontaneous emission, and therefore the expected total linear momentum carried off by the *scattered* photon is zero. It follows that in order to conserve linear momentum, the linear momentum of the absorbed photon must be taken up by the atom or ion. Thus as photons are absorbed out of a beam by the atomic particle, its momentum changes incrementally in the direction of the beam as if it was impelled by a succession of impulses, producing an average force.

In order to establish the relative scale of these effects, a knowledge of which is essential to distinguish between what is observable in practice from what may be only of theoretical interest, let us assume that a laser beam of intensity *I* watts  $\cdot$  m<sup>-2</sup> is directed exactly in the direction opposite to the velocity of an otherwise free particle, so that the flux density of momentum carried by the photons is, as we have seen, *I*/*c*. If a given particle presents an absorption cross section to the beam of σ, then it will suffer, on the average, a change in momentum of (*I*/*c*)σ units per second, which by Newton's law is the measure of the force acting on it. Now, if we assume that the conditions are such that this force continues unchanged while the particle is slowed down (this assumes that the spectrum of the light is broad compared to the maximum Doppler shift of the particle resonance frequency), then the distance traveled by the particle before it is brought to rest can be obtained simply by using the conservation of energy: If the stopping distance is *D*, then the work done on the particle by the radiation pressure is  $F_{rad}D$ , and therefore  $F_{\text{rad}}D = \frac{1}{2}MV_z^2$ ; that is, finally we have the following:

$$
D = \frac{1}{2}MV_z^2 \frac{c}{I\sigma} = \frac{1}{2}kT\frac{c}{I\sigma},
$$

for particles in thermal equilibrium at temperature  $T$ . If we substitute realistic numerical values, for example  $I = 10^2$  watts·m<sup>-2</sup> and  $\sigma \approx 4 \times 10^{-14}$  m<sup>2</sup>, we find for  $T = 300$  K that  $D_{\text{stop}} \approx 50$  cm, a very practical figure. Of course, the beam can be focused down to a smaller cross section to increase the intensity at the ion, thereby reducing the braking distance. However, we should recall that it takes a finite time (the *radiative lifetime*) for the emission of the photon from the excited state, and therefore obviously the rate at which the absorption–emission cycle can be repeated is limited by this lifetime. As the intensity is raised to the point where the probability of absorption becomes comparable to that of spontaneous emission, the particle will be increasingly in the excited state, and stimulated emission will become significant. The radiation pattern from particles undergoing stimulated emission is radically different from that of spontaneous emission, ultimately because unlike the latter, stimulated photons are phase coherent with the photons in the beam. The photon resulting from stimulated emission in fact propagates in the same direction as the photon stimulating the emission; consequently, the atom momentum is unchanged in an absorption-stimulated emission cycle. The result is

that the simple description we have given for momentum exchange with spontaneous emission does not apply even approximately to the case where stimulated emission enters the picture.

### 16.5 Optical Field Gradient Force

There is another mechanism by which an atomic particle interacting with an optical field may experience a force affecting its center-of-mass motion; it is manifested only when the intensity of the optical field is strongly *inhomogeneous*. The physical origin of this force, sometimes called the *gradient force*, and some of its features can be understood in general terms using classical field concepts rather than the proper quantum description. We recall that the positive (nuclear) and negative (electronic) charges in an atomic particle are displaced relative to each other by the electric component of the optical field against their mutual attraction, thereby inducing an electric dipole moment. By virtue of its position in the electric field, this induced dipole possesses potential energy proportional to  $E^2$ , where *E* is the electric field component in the light wave. The dynamical response of the charges to the oscillating electric field is a quantum problem; but suffice it to say that it exhibits resonant behavior at certain frequencies. It is found that the dipole energy is positive or negative depending on whether the optical frequency is above or below a resonant frequency of the particle. This behavior can be understood in terms of the reversal in the phase of the charge displacement (and hence direction of the induced dipole) relative to the field, which occurs for field frequencies on opposite sides of the resonant frequency. Although the field oscillates at the optical frequency, the energy of the polarized atomic particle, being proportional to  $E^2$ , does not average to zero over time, as we saw in connection with the light shifts in the energy separation of the hyperfine states in Rb. As a consequence, the particle has an average potential energy that varies with the intensity of the field where it happens to be located. As with the AC Stark effect leading to the light shifts, this potential energy is positive or negative depending on the optical frequency in relation to the resonance. If the intensity of the optical field varies in space, as in a laser beam with a Gaussian intensity profile, there will be a strong gradient in the particle energy, with the result that, it tends to fall like a rock down a hill.

We will not pursue the gradient force any further and will limit ourselves in what follows to radiation pressure; later in the chapter we will learn of an exciting new mechanism involving *polarization gradients*.

## 16.6 Doppler Cooling

If an atomic particle had an initial velocity component in a direction opposite to a laser beam tuned to exact resonance, that component would be steadily reduced and ultimately pass through zero and increase in the reverse direction. That clearly would not lead to a steady state in which the particle motion has been slowed, corresponding to a lowering of temperature; rather we would have a propulsion mechanism, not a cooling one. To achieve a slowing down of the particle such as might be caused by a frictional force, it is necessary that the particle experience a force opposing its motion, no matter in what direction that motion happens to be. An obvious solution, one might think, is to have *two* laser beams, one resisting motion in one direction and the other in the opposite direction. Unfortunately, the effects of the two beams would cancel each other out, unless the radiation pressure exerted by a given beam depends on its direction relative to the motion of the atomic particle. For if we assume for simplicity that the particle experiences a force only when it is moving in the direction opposite to that of a given beam and no force otherwise, then it is clear that having two opposed laser beams will cause a retarding force to act on the particle by either one beam or the other, depending on which direction it is moving in. This crucial condition on the laser–particle interaction can be met if the *absorption* probability depends on whether a particle moves *in* the direction of the beam or *opposite* to it. But in fact, we know that this *is* in general the case: the Doppler effect shifts the frequency of the field seen by a moving particle, and therefore only if the the resonance line shape is symmetrical and the laser frequency is tuned precisely to the center of the resonance in the particle frame of reference will the probability of absorption be the same for either direction of motion.

Suppose then that an atomic particle is placed in two opposing collinear laser beams, and for simplicity we restrict our attention to the component of the particle motion along the common beam axis. The frequency profile of the atomic absorption will be assumed to have a certain natural line width and be symmetrical about the maximum at its center. Let us consider the interaction of the particle with the laser beam directed opposite to its velocity component, causing a Doppler displacement of the laser frequency seen by the atom towards a higher frequency. It is possible then that by tuning the laser so that its frequency would fall on the *low* side of the resonance profile if the particle were at rest, the Doppler shift would result in the absorption becoming *more* likely, because of the upward shift in frequency toward the maximum, than it would be if the particle were at rest. And similarly, the Doppler shift would cause the absorption to be *less* likely if the atom were moving in the same direction as the laser beam. The result is that such an atom experiences a much greater force opposing a component of its motion directed opposite to the beam direction than one accelerating a component in the same direction as the beam. The total effect in the presence of both laser beams is that there is a net force opposing the motion in either direction, as desired. Ideally, therefore, the continued absorption of photons from the laser beams, with the frequency offset we have described, will cause an atom to be slowed down much like the effect of a frictional force.

In order to make this last statement a little less confusing and a little more quantitative, let us assume that the atomic particle is free (aside from the laser



Figure 16.4 The net force on an atomic particle in two opposed laser beams tuned below resonance

beams of course) and therefore has a Lorentzian resonance line shape characteristic of natural broadening due to the finite lifetime of its excited state. If we represent the full width of the resonance line by  $\Delta v$  and assume that the two laser beams are tuned to the same frequency  $v_L = v_0 - \delta$ , that is, *below* the center of the line, then a plot of the relative strength of the *net* force exerted on the particle as a function of its velocity will be as shown in Figure 16.4. We note that for particle velocities *V* such that the Doppler shift  $(V/c)v<sub>L</sub>$  is less than  $\delta$ , the dependence of the force on velocity is nearly linear and can be approximated for those values of *V* by the equation

$$
F_{\rm rad} = -\alpha V, \qquad \qquad 16.5
$$

where  $\alpha$  is a positive constant analogous to the damping factor used to model friction in a mechanical system. The optimum choice of  $\delta$  to obtain the greatest damping factor is  $\delta \approx \Delta v/2$ . Thus an atomic particle whose initial velocity produces a Doppler shift in the range between  $+\Delta v/2$  and  $-\Delta v/2$  (called the capture range) will experience a viscous drag tending to slow down its motion along the beams' axis. For beam intensities below the extreme where the rate of absorption becomes comparable to the rate of spontaneous emission, the value of the coefficient  $\alpha$  is proportional to the intensity of the laser beams. However, for intensities beyond that point, the proportionality is no longer valid, and  $\alpha$  cannot be increased indefinitely. Now we can estimate the rate of loss of kinetic energy by the atomic particle due to radiation pressure by assuming that the force  $F$  is the resultant of the two forces exerted by the opposing beams. If as before, we assume that the beams are tuned to  $v_0 - \Delta v/2$ , and recall that the (linear) Doppler frequency shift can be

written  $(V/c)v = kV/2\pi$ , then for a scattering cross section having the natural Lorentzian line shape, we can write for the resultant force the following:

$$
\langle F \rangle = \frac{I}{c} \left( \sigma^+ - \sigma^- \right); \quad \sigma^{\pm} = \sigma_0 \left( \frac{\Delta \nu}{2} \right)^2 \frac{1}{\left( \frac{\Delta \nu}{2} \pm \frac{kV}{2\pi} \right)^2 + \left( \frac{\Delta}{2} \right)}, \quad 16.6
$$

where  $\sigma_0$  is the scattering cross section at resonance, and *k* here represents the wave vector  $(2\pi/\lambda)$ . In the range of small velocities such that  $kV \ll \Delta v$ , we can approximate the expressions for  $\sigma^{\pm}$  to obtain simply

$$
\sigma^- - \sigma^+ = \sigma_0 \frac{kV}{\pi \Delta v}.
$$

Hence the net force can be written

$$
\langle F \rangle = -2I\sigma_0 \left(\frac{\mathbf{v}}{c^2 \Delta \mathbf{v}}\right) V. \tag{16.8}
$$

Now, the average rate at which this force does work on the particle in resisting its motion is simply *FV*, which is therefore the rate of *loss* of kinetic energy by the particle. It follows that in time *t* the average loss of kinetic energy is given by

$$
E_k = 2I\sigma_0 \left(\frac{\nu}{\Delta \nu}\right) \frac{V^2}{c^2} t.
$$

#### 16.7 Theoretical Limit

It might be thought that the slowing-down process would continue until the particle came to complete rest; but this is not the case. As the particle is slowed down, the Doppler shift tends toward zero, and the differential force between particle motion in one direction and the other is diminished. But the fundamental limit is ultimately set by the discrete quantum nature of the momentum exchange attending the absorption and emission of photons: The time-dependence of these events is defined only statistically by certain probabilities; and the direction of spontaneous emission is also statistically defined. At each of these random events the atomic particle recoils with a finite jump in momentum in a random direction. The result is that the atomic particle has a residual random motion similar to the erratic zigzag motion, called *Brownian motion*, named for the English botanist Brown, who first observed the random motion of plant pollen suspended in a liquid.

Its existence is historically important in the kinetic theory of gases, since it demonstrated directly that thermal energy is, on a molecular scale, the kinetic energy of their random motion. Since the suspended particle may suffer collisions from any direction, the average of its displacement from a fixed point is zero; however, since it executes a "random walk" in the manner we discussed in a previous chapter, it diffuses out with a *mean square* displacement that increases linearly with the number of collisions, and therefore with time.

In our present one-dimensional case, where the particle is subjected to impulses randomly distributed in time and direction, each imparting  $(h/\lambda)$  units of momentum through photon scattering from two opposing laser beams, the mean square momentum  $\langle p^2 \rangle$  *increases* with time as follows:

$$
\langle p^2 \rangle = 2 \left(\frac{h}{\lambda}\right)^2 \frac{I}{h\nu} \sigma_0 t, \qquad 16.10
$$

where *I* (watts · m<sup>-2</sup>) is the intensity of *each* beam,  $\sigma_0$  the (resonance) atomic cross section, and *t* the time.

Finally, if we make the reasonable assumption that these processes will lead to an equilibrium where the gain and loss of energy exactly balance each other, then according to our model this occurs when the kinetic energy is the following:

$$
\frac{1}{2}MV_{\min}^2 = \frac{1}{4}h\Delta v.
$$
 16.11

Expressed in terms of an equilibrium temperature, this mean kinetic energy along one dimension must be equated to  $\frac{1}{2}kT$ , where *k* is the Boltzmann constant; thus we obtain the lowest temperature  $T_{\text{min}}$  attainable by this Doppler technique:

$$
T_{\min} = \frac{h \Delta \mathbf{v}}{2k} \tag{16.12}
$$

If we substitute for  $\Delta v$  the atomic resonance line width for cesium, that is, approximately  $5\times10^6$  Hz, we find that the lowest attainable temperature is on the order of  $120 \mu K$ , that is, 120 millionths of a degree above absolute zero! At this temperature the average linear momentum of a Cs atom reaches within an order of magnitude of the momentum of a *single* photon, the fundamental limit to cooling by this method. At such a super-cold temperature the second-order fractional Doppler shift  $(E_k/Mc^2)$  in the cesium resonance frequency would be entirely negligible at only one part in  $2.5 \times 10^{19}$ ! In practice, of course, this degree of accuracy is meaningful only if a multitude of other sources of systematic frequency shifts, which no doubt become significant at this level, could also be as elegantly reduced or taken into account.

## 16.8 Optical "Molasses" and the Magneto-Optical Trap

So far, the discussion has been limited to one dimension; clearly, to cool free particles in three dimensions, multiple sets of laser beams are required. A configuration consisting of three mutually perpendicular pairs of opposing lasers has been successfully used to cool a cloud of free neutral atoms. In one set of experiments at the Bell Telephone Laboratories, sodium atoms were observed not only to be cooled by the action of the lasers but also to manifest another effect of the viscous drag their motion is subjected to, namely a reduction in their diffusion rate. That is, a given group of atoms occupying a certain space takes very much longer to diffuse out into a larger volume and the atoms are in this sense "confined" in what was dubbed "optical molasses."

Used in conjunction with a specially tailored magnetic field, laser cooling can produce a so-called *magneto-optical trap* for neutral atoms. It relies on the shift produced by an external magnetic field in the quantum energy states of atoms, and hence their optical resonance frequencies, that is, the Zeeman effect. Since the radiation pressure depends on the rate of photon absorption and reemission, and hence the frequency detuning of the atomic resonance with respect to the laser frequency, it follows that the light pressure acting on an atom is a function not only of its velocity because of the Doppler effect, but also of the magnetic field acting on it. If this field varies from point to point in space, then the light pressure the atom experiences will vary as a function not only of its velocity, but also of its position in space. Now, we recall that with pure Doppler cooling we were able to produce a net radiation pressure that changed sign with the velocity vector by having two oppositely directed laser beams tuned below the peak resonance frequency. To achieve a similar reversal with respect to displacement from a fixed spatial position, we impose a magnetic field that reverses direction at that point, together with two oppositely directed laser beams whose circular polarizations are in opposite senses (clockwise or anticlockwise). We recall that in the Zeeman effect, there are *selection rules* that govern optical transitions between quantum states which impose an essential connection between the polarization of the radiation and the magnetic substates between which transitions are observed. This comes ultimately from the requirement that angular momentum of the photon–atom system be conserved in the process of emission or absorption.

Thus, for example, in Figure 16.5 we show the Zeeman effect in the socalled  $D_2$ -line of the principal resonance in the Cs atom, arising from the transition between the ground state, with its electron spin of 1/2 and zero orbital angular momentum, designated as  ${}^{2}S_{1/2}$ , and the first excited state with a combined (orbital + spin) angular momentum of  $3/2$ , designated as  ${}^{2}P_{3/2}$ . In the presence of an external magnetic field, the substates with different orientations of the angular momentum with respect to the direction of the magnetic field, designated by the quantum numbers *m*, will be separated in energy by an amount dependent on the strength of the field.

Now we recall that a photon carries *one* unit (*h*/2π) of angular momentum but has only *two* possible orientations with respect to a given axis, corresponding to the electric field in the light wave rotating clockwise or counterclockwise. Thus by absorbing a photon designated as having a  $\sigma^+$  circular polarization, an atom gains precisely one unit of angular momentum along the beam axis. Symbolically, we can write:

$$
m'-m=+1 \quad (\sigma^+ - radiation), \tag{16.13}
$$

where  $m'$  and  $m$  are the components (in units of  $h/2\pi$ ) of the atomic angular momentum along the fixed axis, before and after the transition respectively. Similarly, for the oppositely polarized  $\sigma^-$ -radiation we have

$$
m'-m=-1 \quad (\sigma^- - radiation). \tag{16.14}
$$



Figure 16.5 The Zeeman splitting of the  $D_2$  line in Cs, neglecting the hyperfine structure

From the figure we see that as the magnetic field strength increases and the *m*-substates are more widely separated in energy, the resonance frequency for excitation with  $\sigma^+$  radiation is shifted to a higher value, whereas resonance with σ− radiation is at a frequency shifted downward. Let us now limit our attention to how the resonance frequency of excitation by only one laser beam, having, say,  $\sigma^+$  polarization, changes when the direction of the magnetic field is reversed with respect to our fixed axis. We note that the energy displacement of the *m*-substates is proportional to the magnetic field, and that if the latter is reversed in direction, we must reverse the direction of the displacements, so that the order of the *m*-values on an energy level diagram is reversed, as shown in Figure 16.6.

It is clear that now excitation with the same  $\sigma^+$  radiation is resonant at a frequency that is shifted downward rather than upward as the magnetic field strength increases. By the same argument, excitation with σ− radiation will have an upward shift on reversal of the field. Thus we see that having two oppositely directed laser beams, circularly polarized in opposite senses, with a parallel magnetic field of uniform gradient in space, passing through zero and reversing at some point, can result in an atom experiencing a corresponding reversal in the radiation pressure acting on it as it passes through that point. All that is necessary is that the lasers be tuned *below* the zero-field frequency. Such an arrangement, consisting of three mutually perpendicular pairs of lasers, is therefore called a  $\sigma^+ - \sigma^-$  magneto-optical trap. The essential feature introduced by using σ-radiation in conjunction with a spatially varying magnetic field is that the simple "resistive" force  $F_{rad} = \alpha kV$  is replaced by  $F_{rad} = \alpha (kV + \beta B)$ , where α and β are constants. It can be shown that for a sufficiently intense laser beam, the atoms will follow trajectories with a



Figure 16.6 The effect of reversal of the magnetic field on the energy of m-sublevels

velocity whose Doppler shift compensates for the Zeeman shift in the resonance frequency. Stated mathematically, this amounts to the following:

$$
kV = \frac{2\pi g \mu_B}{h}B, \qquad 16.15
$$

where  $k = 2\pi/\lambda$ ; *g* is the *g*-factor, the measure of the magnetic moment of an atom accompanying its mechanical angular momentum;  $\mu_B$  is the Bohr magneton; and *B* is the magnetic field acting on the atom. Suppose that the magnetic field distribution is chosen so that its (cylindrical) components vary in space according to the following:

$$
B_r = -\frac{C}{a^2}r; \quad B_\phi = 0; \quad B_z = 2\frac{C}{a^2}z.
$$

Such a field is produced, for example, in the neighborhood of the midpoint (taken as the origin) between two parallel coaxial circular coils separated by a distance equal to their radius, the so-called *Helmholtz* configuration, but having currents circulating in *opposite* directions. This is seen to correspond to the "pure" quadrupole field distribution familiar in the electrostatic case. To the extent that the light pressure acting on the atom can be accurately modeled as a velocity-dependent term due to the Doppler effect and a term having the spatial dependence of the magnetic field due to the Zeeman effect, its motion, for example in the radial direction, will be governed by an equation of the following form:

$$
\frac{d^2r}{dt^2} + \alpha \frac{dr}{dt} = -\beta r,\tag{16.17}
$$

where  $\alpha$  and  $\beta$  are constants. This is recognized as the equation of motion for a damped harmonic oscillator, the form of whose solution for *r*, the radial distance of the atom from the axis  $(r = 0)$ , depends on the relative values of the damping coefficient α and the "restoring force" coefficient β. For  $\beta$  >  $(α/2)<sup>2</sup>$  the radial motion, if allowed to continue, is a damped oscillation with a decaying amplitude; otherwise, the particle is drawn toward the axis without oscillation. The atoms are in this sense trapped in the radial direction, their distance from the axis falling exponentially; that is, they are focused.

Initial attempts to study these effects quantitatively by measuring the temperature of the atoms in the "molasses," yielded results that seemed to be compatible with the theory as outlined. It was assumed therefore that the well-understood processes invoked in the theory were adequate to explain fully the phenomena involved. However, it soon developed that with the application of a more refined technique to measure the velocity distribution of the cooled atoms by W. Phillips and his group at the U.S. National Institute of Standards and Technology, there was revealed a conflict between theory and observation. The temperature was, in fact considerably lower than the lower limit predicted by the theory. Moreover, contrary to the theory, the lowest temperature was reached when the lasers were tuned not one half a line width, but several line widths from resonance. There was understandable skepticism at first about these contrary results; not only because of the well-established nature of the processes assumed in the theory, but also because experimental results generally fall short of theoretical limits and certainly should not exceed them! Nevertheless, further experiments on sodium and other atoms at the Ecole Normale Superieure, in Paris, and other laboratories in the U.S. con- ´ firmed that the low temperature limit of Doppler cooling had been broken and an explanation had to be sought elsewhere.

## 16.9 Polarization Gradient Cooling: The Sisyphus Effect

To find an explanation for the unexpectedly low temperatures, we must take a much more detailed view of the interaction of the sodium atoms with the laser beams. First, the alkali atom is not a two-level atom with a simple ground state and an excited state. Like the other alkali atoms, rubidium and cesium, which we discussed in earlier chapters, sodium's ground state has the spectroscopic designation  ${}^{2}S_{1/2}$  with a spin angular momentum  $J = 1/2$ , which couples to a nuclear spin  $I = 3/2$ , giving rise to a hyperfine frequency of 1.771 GHz, which is, incidentally, less than one-third the value for  $Rb^{87}$ . In a weak magnetic field it has Zeeman substates with magnetic quantum numbers  $m<sub>J</sub> = -2, -1, 0, +1, +2$  for the  $F = 2$ hyperfine state and  $m<sub>J</sub> = -1, 0, +1$  for the  $F = 1$  state. Further, we recall the vast improvement in the observation of magnetic resonance in free atoms made possible by an optical method called by its originator, Kastler, *optical pumping*, which involves the absorption of circularly polarized resonance light followed by spontaneous re-emission. This technique exploits the selection rules governing optical transitions imposed by the law of conservation of angular momentum applied to the photon–atom system. By continued repetition of the optical pumping cycle, atoms are transferred predominantly into that substate in which the angular momentum is oriented in the direction of that of the absorbed photon. In general, then, we can expect that the detailed distribution of atoms among the different magnetic substates will depend on the type of polarization of the absorbed resonance light, since that determines the component of its angular momentum along the beam axis.

Following Cohen-Tannoudji and Dalibard (Cohen-Tannoudji, 1989) we look to one of the subtle effects in optical pumping long studied theoretically and confirmed experimentally, namely the displacement of quantum energy levels by the action of the pumping light (Cohen-Tannoudji, 1962), as providing a new cooling mechanism. We have already met this before in connection with the optically pumped rubidium standard: *light shifts*. The precise energies of the quantum states of an atom interacting with the pumping light beam are displaced slightly with respect to a free atom, and the displacement is in proportion to the intensity of the beam. Moreover, the energy displacements appear to an extent dependent on the probability of the transitions, which occur according to the selection rules applied to the type of light polarization present.

According to Cohen-Tannoudji and Dalibard, we will find that under suitable conditions, rapid spatial variation in the polarization of the light can lead to a lower temperature limit than that of Doppler cooling. To do this, consider the idealized example of a one-dimensional "molasses" consisting of two opposing frequencyoffset laser beams having mutually perpendicular linear polarizations and equal intensity. Because each laser beam is spatially coherent, that is, there is a well defined phase relationship between the optical field at different points along the beam, the fields of the two laser beams will, in the span of half a wavelength, have a phase difference that varies from zero to 360 $^{\circ}$ . Thus if we let  $E_x$  and  $E_y$  represent the two counterpropagating light waves, we have the following:

$$
E_x = E_0 \sin(\omega t - kz); E_y = E_0 \sin(\omega t + kz), \qquad 16.18
$$

and therefore at a fixed point along the axis  $z<sub>0</sub>$ , the two components will have a difference of phase  $\Delta \phi = 2kz_0$ . This means that at points within each half wavelength along the common beam axis, the phase difference between the two equal and perpendicular field vectors will pass continuously through the values  $0^\circ$ ,  $90^\circ$ , 180◦, 270◦, 360◦. Now, phase differences of 0◦ and 180◦ simply yield combined fields that are linearly polarized along directions at 45◦ to the original directions, while phase differences of 90° and 270° yield circularly polarized fields rotating in opposite senses about the beam axis. This pattern is repeated every half wavelength along the beam axis, as shown in Figure 16.7.

In order to bring out the consequences of having such variation in the light polarization on the optical pumping of an atom and the attendant light shifts, let us consider the specific example of sodium. Let us ignore its nuclear spin and assume that the optical pumping cycle occurs between the ground state having the total angular momentum quantum number  $J = 1/2$  and the excited state having  $J = 3/2$ . We recall that space quantization of the angular momentum leads to a multiplicity of substates, so that the ground state is made up of substates in which the components of angular momentum along a given axis (chosen here to be in a fixed direction along the light axis) are  $m<sub>J</sub> = +1/2$  and  $-1/2$ , and



Figure 16.7 The polarization of the resultant of two counterpropagating beams with perpendicular polarizations



Figure 16.8 Light shifts in sodium atom in two opposed laser beams perpendicularly polarized

the excited state  $m'_J = +3/2, +1/2, -1/2$ , and  $-3/2$ . Now, if the atom happens to be at a point where the polarization is circular, with the field rotating in the positive sense (the rotation of a right-hand screw advancing in the direction of the light beam), designated by  $\sigma^+$ , it can absorb a photon only if the excitation results in an *increase* in the  $m<sub>J</sub>$  value by one unit. Of the two possible transitions,  $m_J = -1/2 \rightarrow m'_J = +1/2$  and  $m_J = +1/2 \rightarrow m'_J = +3/2$ , it can be shown that the former will occur at one-third the rate of the latter, and consequently, the light shift for the latter  $m<sub>J</sub> = +1/2$  is greater. Moreover, it also happens that the optical pumping cycle tends to concentrate the atoms in this light shifted  $m<sub>J</sub> = +1/2$  state; this, after all, was the whole point of the Kastler optical pumping technique. Conversely, if the atom happens to be where the polarization state of the field is circular in the opposite direction, designated as  $\sigma^-$ , the  $m<sub>J</sub> = -1/2$  substate will suffer the greater light shift and become more populated than the other. At points where the polarization is linear, the two substates are equally shifted. Figure 16.8 illustrates the variation in the light shifts of atoms in the two substates as a function of their position within a range of one half wavelength along the axis of the laser beams. We see therefore that if we had a set of stationary atoms strung along the beam axis, they would alternate between being predominantly in one substate and predominantly in the other, with their energy varying in step. This picture is really a static one of an equilibrium situation, achieved after a sufficient time has allowed the pumping cycle to be repeated many times. As such, the spatial variation in energy we have found cannot, for the purposes of predicting the forces acting on a moving atom, be used as a static potential energy distribution analogous to hills and valleys on the earth's surface. The dynamical problem is considerably more complicated; we must take into account the time development of the pumping cycle itself, simultaneous with the varying polarization of the light seen by the moving atom. The static energy distribution found above would apply only if the pumping cycle took place so rapidly that the atomic substate population was in quasi-equilibrium at every point as the atom moved along the beam. The word "population" can refer to a single particle; it is used as the relative probability. Let us imagine the system set in motion: The pumping cycle repeats itself on the average every  $\tau_p$  seconds, the mean pumping time, causing the populations to evolve in time, to alternate between one substate and the other, while the atom moves through the different polarization states of the light. Because of the delay in pumping the atoms into a particular substate, the population of that state reaches its maximum only after it has passed the point where the light shift is maximum, leading to an asymmetry in the population distribution relative to the periodic light shift, as shown in Figure 16.9. The result is that the atoms find themselves constantly being optically pumped into the substate whose energy is on the rising side of the energy curve, no matter in what direction they are moving. This brings to mind Sisyphus, the Greek mythological figure whose task in Hades was forever to push a rock uphill, only to have it roll back down; hence this method of atomic cooling has been dubbed *Sisyphus cooling*. Without entering into the difficult question as to how the photon–atom interaction leading to the light shifts can couple to the center-of-mass motion of the atom, we can use the law of conservation of energy merely to say that the rising light-shift energy leads to a corresponding loss of kinetic energy, that is, a cooling of the atom.



Figure 16.9 The asymmetry between light shift of energy and substate population distribution in sodium atoms (Cohen-Tannoudji, 1990)

The important question remains as to why Sisyphus cooling is effective at a much lower temperature than Doppler cooling. The answer must be sought in the essentially different physical processes involved in the cooling and the theoretical limits these processes set on the lowest achievable temperature. In the Sisyphus process it is the light shift in energy that plays a central role: If the potential well created by the light shift is designated as  $\Delta E_m$ , then each optical pumping cycle results in the atom losing an amount of energy on the order of  $\Delta E_m$ . After many such cycles, the atom will have lost so much energy that it no longer has enough energy to penetrate the adjacent regions, and it remains trapped. In this limit the thermal energy  $kT$  is on the order of  $\Delta E_m$ . If the pumping light intensity is reduced,  $\Delta E_m$  is reduced proportionately, and the limit can be far below the Doppler value of  $h \Delta v_n$ , where  $\Delta v_n$  is the natural line width of the optically excited state.

Theory shows that the dependence of the friction coefficient  $\alpha$  on the pumping light intensity is radically different for the two processes: In contrast with Doppler cooling, where  $\alpha$  decreases linearly with the intensity as it approaches zero, in the Sisyphus cooling mechanism  $\alpha$  remains essentially constant. To make this fact at least plausible, we note that although reducing the light intensity makes the light shift correspondingly smaller, this is offset by the lengthening of the pumping delay that enhances the population asymmetry, which, as explained above, is the root cause of the cooling effect. In Figure 16.10 are reproduced curves comparing the dependence of the average force experienced by the atom on the mean velocity for Doppler cooling and Sisyphus cooling. We see that in the latter, while the velocity capture range shrinks as the intensity of the pumping light is reduced, the slope of the linear portion of the curve where it crosses the origin (which is a measure of  $\alpha$ ) remains constant. It is apparent, then, that since the velocity capture range for Doppler cooling is greater, the initial cooling of an atom is most effectively done by



Figure 16.10 Comparison of forces due to Doppler cooling and polarization-gradient as a function of particle velocity (Cohen-Tannoudji, 1990)

that method; only when the cooling has progressed to a relatively low temperature would the Sisyphus mechanism become effective.

Finally, we note that the optimum detuning of the laser to obtain the most effective cooling is determined in the Sisyphus cooling technique by the need to maximize the light shift, which can occur at much greater frequency departures from resonance than is allowed in the Doppler technique.

All these features have been brilliantly confirmed by experiments, initially on sodium but more especially on cesium, whose energy level structure permitted a wider confirmation of the dependence of the cooling on laser detuning. Furthermore, the unexpected sensitivity of the molasses to magnetic fields follows logically from the known displacement and "mixing" that such fields cause in the magnetic substates. Detailed studies of the effect of magnetic fields confirmed that their influence is diminished, as expected, at higher pumping light intensities.

The extent of the cooling was determined, as in the Doppler technique, by a time-of-flight method, which yields the velocity distribution of the atoms in the molasses. As expected, a nonequilibrium double-peaked distribution was observed, indicating distinct velocity capture ranges for the two types of cooling mechanisms. Velocities corresponding to absolute temperatures around 2.5 μK have been achieved; this amounts to but a few times the recoil of an atom in scattering a single optical photon. More recently, special techniques are being explored to attain subrecoil energies; temperatures as low as 3 nanodegrees Kelvin (along one dimension) have been reported by Cohen-Tannoudji et al.

#### 16.10 Laser Cooling of Trapped Ions

Of particular interest to frequency standards is the cooling of isolated ions confined in the kinds of electric and magnetic field configurations described in an earlier chapter. We recall that the motion of ions in those fields, under conditions of stable confinement, is oscillatory with a discrete line spectrum. Thus for the Paul trap, the motion along each of the coordinate axes contains the following discrete frequencies:

$$
\omega_n = (2n \pm \beta) \frac{\Omega}{2}, n = 0, 1, 2, \dots,
$$

where  $\Omega$  is the frequency of the rf trapping field, and  $\beta$  lies in the range  $0 < \beta < 1$ and differs in general between the axial and radial motions. Under the conditions commonly chosen in practice, where the amplitude of the high-frequency ion jitter in response to the field is small,  $\beta \ll 1$ , and the higher frequencies with  $n > 1$  have a negligibly small amplitude.

The technique of laser cooling can be equally applied to an ion executing such an oscillatory motion as any neutral atom moving about randomly; in fact, it is somewhat simpler. First, because the motions along *all three dimensions* can be simultaneously slowed by using laser beams along a *single* axis, as long as it does *not* lie in the *x*-*y* symmetry plane of the coordinate system we have been using. In the absence of the cooling laser beams, our particular choice of axes, namely taking the *z*-axis to be along the axis of symmetry of the trapping field, ensures that the equations are "uncoupled"; that is, in the equation of motion for a particular coordinate, the other coordinates are not involved. However, the Doppler drag on the motion of an ion due to a beam directed at some angle to the mid-plane couples the motions, leading to 3-dimensional cooling. Also, in practice, departures from the assumed ideal field distribution, and to a lesser extent ion–ion collisions, could lead to exchange of energy between the different components of the ion motion. Second, the ions are thermally isolated in ultrahigh vacuum and therefore can gain heat from the environment (or through rf heating in the case of the Paul trap) only very slowly, making it easier to observe even a small rate of cooling.

The oscillatory ion motion does, however, have a distinctive effect on the optical absorption spectrum of the ions. If we imagine a plane monochromatic light wave of frequency ν falling on an ion executing simple harmonic motion of frequency  $v_m$  along the direction in which the wave is traveling, the ion will see, because of the Doppler effect, a wave whose *frequency* is modulated, rising and falling as the ion swings back and forth, first counter to the wave direction, then with it. But we recall from Chapter 7 that there is a curious mathematical fact that runs counter to intuition here: Except in the limit of infinitely slow modulation, the frequency of the wave does not actually pass continuously through all the values in the Doppler range. In fact, the spectrum of such a frequency-modulated (FM) wave is discrete, containing the undisplaced frequency ν and a series of "sidebands" spaced at equal intervals of  $v_m$  extending symmetrically to infinity above and below ν. The frequency spacing of the sidebands in this spectrum remains unchanged if the amplitude of oscillation, and therefore the Doppler frequency range of the ion, is changed; there is merely an increase in the amplitudes of sidebands at frequencies farther from the undisplaced center frequency. Quantitatively, the amplitude of the sideband at frequency ( $v \pm n v_m$ ) is proportional to  $J_n(2\pi a/\lambda)$ , where as usual,  $J_n$  represents a Bessel function of order *n*, and  $a/\lambda$  is the ratio of the amplitude of the ion oscillation to the wavelength of the light wave. The graph of the Bessel function  $J_n(2\pi a/\lambda)$  of the first few orders *n* was given in Chapter 7 in Figure 7.4. If the particle is constrained to oscillate over a range below one wavelength, that is, if  $a/\lambda$  < 1, then all the amplitudes rapidly approach zero for increasing *n* above zero. In this case the power resides principally in the undisplaced frequency, and the (first-order) Doppler effect is effectively absent. This is the Dicke effect we found to be so important in the hydrogen maser and the rubidium standard; however, there the wavelengths involved were in the centimeter range, larger than the mean free path at ordinary temperatures, rather than the submicron range that concerns us here.

The extent to which the discreteness of the spectrum seen by an oscillating ion alters the circumstances of laser Doppler cooling depends on whether the natural line width for resonance absorption is greater or less than the spacing of the Doppler sidebands. If, for example, the spacing is *larger* than the natural line width,

then for Doppler cooling it is clearly necessary that the laser be tuned to fall exactly on some sideband  $n v_m$  *below* the undisplaced resonance frequency; this ensures that the corresponding *upper* sideband seen by the ion will fall exactly on resonance. As the cooling proceeds and the amplitude of oscillation of the ion diminishes, the power in that upper sideband will also rapidly decrease, making further cooling less and less efficient.

The other extreme, in which the natural line width is much larger than the spacing of the Doppler sidebands, is expected to be more common, since ν*<sup>m</sup>* is typically not greater than a megahertz, whereas optical resonance line widths are perhaps ten times that. In this case we need the resonance line-shape function; for a free ion it is a Lorentzian function with a frequency width  $\Delta v$  determined by the radiative lifetime of the states involved in the optical transition. Since the external fields used to confine the ions are expected to produce a negligible distortion of the resonance line shape, the cross section for resonance scattering can be written as follows:

$$
\sigma(v) = \sigma_0 \left(\frac{\Delta v}{2}\right)^2 \frac{1}{(v_0 - v)^2 + \left(\frac{\Delta v}{2}\right)^2}.
$$

Now, as already pointed out, the spectrum that the oscillating ion sees depends very much on the value of  $2πa/λ$ . At one extreme,  $2πa/λ \gg 1$ , which is equivalent to the Doppler variation in the light frequency oscillating very slowly, that is, the oscillation frequency  $v_m$  being very small compared to the maximum Doppler shift. In this case the spectrum consists of a large number of closely spaced lines of significant amplitude for all harmonic numbers *n* up to  $(2\pi a/\lambda)$ . For a given maximum Doppler shift, if the ion oscillation is assumed to be very slow (with a correspondingly large amplitude to keep the Doppler shift constant) the spectrum is so closely spaced as to form a nearly continuous band of width about equal to the Doppler shift caused by the ion velocity at its maximum. In this limit it would be a valid approximation simply to take the light spectrum seen by the oscillating ion as a single frequency displaced according to the instantaneous velocity of the ion. However, we would then have to allow for the fact that the ion does not spend the same length of time at different phases in its oscillation. In this approximation it is as if the ion were not confined at all, but moving freely, just as was assumed for the neutral particles discussed earlier. The same description of the Doppler cooling should then apply here also.

In general, however, 2π*a*/λ may assume any value, and in the *ion* frame of reference, it is as if it were being irradiated, not by a monochromatic beam of frequency ν, but rather by light having independently the several discrete frequencies  $v \pm n v_m$ . Furthermore, when an ion absorbs a photon, it does so at the sideband frequency that it sees, in its own frame of reference, as being at its resonance frequency  $v_0$ . That is, the harmonic number *n* must satisfy  $v_0 = (v + n v_m)$ ; but in the *laboratory* frame this corresponds to a photon of energy *h*ν being absorbed. Now in this process of resonance absorption and re-emission, an atomic particle will, in its *own* frame of reference, re-emit radiation of very nearly the same energy

as it absorbs. (There is a recoil energy of the ion that comes at the expense of the emitted photon, but for the present purposes this is negligible). However, this radiation will have, in the *laboratory* frame with respect to which the ion is oscillating, Doppler sidebands symmetrical about the center frequency, so that on the average, the energy of the photon emitted will be just that of the photon absorbed in the ion frame of reference, that is,  $h(v + n v_m)$ . The result is that in the laboratory frame of reference a photon of energy *h*ν has been absorbed and a photon of average energy  $h(v + n v_m)$  has been emitted. If the laser frequency v is set *below* the frequency of maximum resonance absorption  $v_0$ , so that the absorption is more likely for the higher sidebands ( $n > 0$ ) than for the lower ones ( $n < 0$ ), there will be a net loss of energy by the ion, which must come from its center-of-mass motion.

This way of describing the cooling process is physically equivalent to the one given earlier in terms of light pressure; a quantitative comparison is of course meaningful only in the range where both approximate treatments are valid. If we limit ourselves to the case where  $2\pi a/\lambda \gg 1$ , the instantaneous velocity of the ion is related to *n* by equating the Doppler shifts:  $n v_m = (V/c)v$ . Now, we had previously found that a photon beam of intensity *I* exerted an average force  $F = I\sigma/c$ on an ion, and therefore the rate at which the ion energy is reduced (*FV*) is given by  $I\sigma(V/c)$ , that is,  $I\sigma n v_m/v$ , or  $h n v_m$  per photon absorbed. This is just the result obtained by balancing the energy between absorbed photons Doppler-shifted in frequency and the emitted photons.

While the limiting case of  $2\pi a/\lambda \gg 1$  is expected to be relevant in the initial stages of ion cooling, it is the opposite extreme, where this parameter approaches a value much less than unity that will determine the ultimate level of cooling that can be reached. As already pointed out, in this limit the amplitudes of the Doppler sidebands above  $n = 1$  become negligible, so that the spectrum seen by the oscillating ion consists of the center frequency ν and only one side-band on each side of it,  $(v + v_m)$  and  $(v - v_m)$ . In this case it is relatively easy to obtain an explicit expression for the rate of cooling, since the intensity of these sidebands,  $J_{\pm 1}^2 (2\pi a/\lambda)$ , is approximately  $(\pi a/\lambda)^2$ . If we again assume a Lorentzian function of width  $\Delta v$  for the resonance line shape with  $\Delta v \gg v_m$ , we find that there is not just the loss of energy as a result of absorbing a photon of energy *h*ν followed by emission of one of average energy  $h(v+v_m)$ , but also a *gain* resulting from the absorption/emission cycle at the other  $(v - v_m)$  sideband. The overall rate of cooling, after some simplification, is given by the following:

$$
\frac{dE}{dt} = -2\left(\frac{\pi a}{\lambda}\right)^2 I \sigma_0 \frac{v_m}{v} \frac{v_m}{\Delta v}.
$$

In arriving at this, we have taken into account the effect on an existing motion as manifested by the Doppler shift and based the derivation of the laser cooling on the assumption that in the frame of reference in which an ion is initially *at rest*, it emits photons of the same frequency/energy as it absorbs. This is a valid approximation as long as the energy of *recoil* of an ion in the act of absorbing or emitting a photon is negligibly small compared to the energy of any prior motion. This may give rise

to some confusion, since the Doppler cooling mechanism is also in a sense due to "recoil"; it is clear that a distinction must be made: They represent two levels of approximation in terms of the ratio between the photon and ion momenta. As the ion momentum is brought near zero by cooling, the higher-order approximation, which applies even to ions at rest, becomes necessary. It is precisely this ultimate recoil energy that will set the minimum ion energy attainable. It is the same recoil energy which accounts for the residual Brownian motion described earlier. Since the conservation of linear momentum requires that when a photon is absorbed, the ion recoil with a momentum  $h\nu_0/c$ , it follows that from a zero initial velocity the ion gains kinetic energy amounting to  $(hv_0/c)^2/2M$ . Now, under the present conditions  $J_0^2(2\pi a/\lambda) \approx 1$ , and thus nearly all the radiation the ion sees is at the center frequency  $v$ , which has been assumed to be set at  $v_0 - \Delta v/2$ . Since at this frequency  $\sigma = \sigma_0/2$  (by definition), it follows that the rate of energy gain by the ion is given by the following:

$$
\frac{dE}{dt} = \frac{I}{h\nu} \frac{\sigma_0}{2} \left(\frac{h\nu_0}{c}\right)^2 \frac{1}{2M}.
$$

Finally, in order to obtain the ultimate equilibrium value of the ion energy, we equate the rate of energy loss to energy gain and obtain, after some simplification,

$$
E_{\min} = \frac{1}{2}M(2\pi v_m a)^2 = \frac{h\Delta v}{4}.
$$

While this analysis does not pretend to be anything more than a rough quantitative sketch of the basic physical processes involved, it nevertheless leads to the correct dependence of the cooling on the physical parameters. However, it is flawed in one serious physical respect: At or near the minimum energy, the conditions may be approached where the ion motion can no longer be treated using classical mechanics; it becomes a problem in quantum mechanics. This is seen from the fact that it is possible that  $E_{\text{min}}$  may not be much larger than  $h\nu_m$ , the quantum of energy for a harmonic oscillator of frequency  $v_m$ . The energy of such an oscillator can assume only the following discrete values:  $h\nu_m(n + 1/2)$ , where *n* is a whole number, and  $\frac{1}{2}h\nu_m$  is the so-called *zero point energy*. The fact that the energy cannot reach precisely zero is, of course, a quantum phenomenon; the classical description is an approximation valid only for very large values of the quantum number *n*. To pursue the matter further would take us too far afield; suffice it to say that it is truly remarkable that the experimental implementation of the ideas we have described has been so successful as to reach the quantum level of oscillation in the trap. An ion harmonically bound in a trap with a frequency  $v_m = 10^5$  Hz and cooled down to the point where there is a 95% chance that it is in the state of zero point energy would have a temperature around  $0.8 \mu K$ . In the case of a Hg<sup>+</sup> ion it would have an amplitude of oscillation no more than 0.02  $\mu$ m, that is, on the order of  $\lambda/10$ , and a second-order Doppler shift of no more than one part in  $10^{21}$ !