COOLING OF GASES BY LASER RADIATION[1*]

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It is shown that a low-density gas can be cooled by illuminating it with intense, quasi-monochromatic light confined to the lower-frequency half of a resonance line's Doppler width. Translational kinetic energy can be transferred from the gas to the scattered light, until the atomic velocity is reduced by the ratio of the Doppler width to the natural line width.

It is well known that light exerts a radiation pressure on any substance which reflects or scatters it. It is also known that the scattering cross section of an atom can be quite large when the light frequency is in resonance with a sharp absorption line [1]. Thus the radiation pressure of laser light has been used to selectively deflect atoms of a chosen isotopic species from a beam.

We wish to point out that if the laser radiation is essentially isotropic, but confined to frequencies on the lower half of the Doppler-broadened absorption line of an atomic vapor, the gas can be cooled. That is, the average translational kinetic energy of the atoms can be reduced.

To understand why this occurs let us first consider the irradiation of the vapor by a single directed laser beam. Only those atoms which are moving towards the laser source will find the light Doppler-shifted up in frequency so as to have a large scattering cross section. To atoms moving away from the laser, the frequency of the light will appear lowered out of resonance with the scattering transition. Thus if the laser light is confined to the lower half of the Doppler line width, the atoms can only lose energy and momentum by scattering of the laser light, and never gain.

If the light comes from all directions, atoms will lose energy by scattering the oncoming light, while the Doppler-shift will detune any light wave traveling in the same direction as the atoms. In this way the translational temperature of the atoms can be reduced until ultimately the Doppler line width is as small as the natural line width[2*]

To estimate the size of the effect, consider a gas of magnesium atoms at a temperature of 600 K, illuminated by intense light on the low-frequency side of the singlet resonance line at 2852.1 Å. The Doppler width at this temperature is 3.8×10^9 Hz full width at half maximum, and the natural line width, determined by the 2 nsec radiative lifetime of the upper state, is 8×10^7 Hz. Thus radiation cooling could reduce the average atomic velocity by a factor of about 50, which is equivalent to a reduction in temperature to $600/(50)^2 = 0.24$ K.

Cooling of this order could be achieved quite quickly. When a photon of momentum $h\nu/c$ is scattered by an atom of mass M, moving towards it with a velocity v, the average change in velocity is

$$\Delta V = \frac{\Delta(Mv)}{M} - \frac{h\nu}{Mc}.$$

Thus at each scattering, the velocity of an atom will decrease, on the average, by about 6 cm/sec. Since the r.m.s. velocity $v_0 = (3kT_0/M)^{1/2}$ is, at $T_0 = 600$ K,

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2* A different mechanism of "cooling" with quasi-monochromatic light, the depletion of low, thermally populated molecular energy levels via optical pumping, has been discussed elsewhere [2].

initially 80,000 cm/sec, about $v_0/\Delta v \approx 13{,}000$ scattering events will substantially complete the velocity reduction. If the light intensity is comparable to that needed for saturation of the atomic absorption, a photon can be scattered essentially every $\tau = 2$ nsec, and the cooling process need only take about $t_0 = \tau v_0/\Delta v \approx 3 \times 10^{-5}$ sec. During this time, the average atom will move a path length

$$l_0 = \tfrac{1}{2} v_0 t_0 = 3kT_0 c\tau/2h\nu,$$

independent of the atomic mass. For the present example, this path is on the order of 1 cm, i.e. it is not necessary to illuminate a large volume, if the radiative lifetime τ is sufficiently short, and/or the light frequency ν is sufficiently high.

To saturate the Doppler-broadened magnesium resonance line requires a flux of about 1000 W/cm^2. Half of this would be needed to irradiate only the lower half of the Doppler profile. To irradiate a 1 cm cubic volume with six such beams directed along the six perpendicular directions would require a total power of 3000 W. Since the power would need to be applied for about 3×10^{-5} sec for complete cooling, a pulse energy of 0.1 J would be required. Such a pulse could be generated by harmonic generation from a flashlamp-pumped dye laser. The required power can be substantially reduced, if the six rays are generated by multiple reflections of the same laser beam. The limiting effect of power broadening of the atomic resonance can be avoided by using a lower light intensity towards the end of the cooling pulse.

Radiation cooling could be applied to provide slow-moving atoms which would remain for a long time in interaction with a weak optical or radiofrequency field. A particularly interesting case might be hydrogen, where experiments are under way to study the 1s → 2s two-photon absorption. The upper state has a lifetime of 1/7 sec, so that fractional line widths of 10^{-15} might eventually be obtained. However, at 300 K the r.m.s. velocity of hydrogen atoms is nearly 3×10^5 cm/sec. Thus transit time is likely to be an important source of line broadening unless the atomic velocity is reduced. Another possible application might be to improve the collimation of an atomic beam by reducing the transverse velocities through two-dimensional radiation cooling.

It is possible that radiation cooling might occur naturally in some astronomical objects. A continuous-spectrum light source will have little influence on atoms, except for the narrow band within the Doppler width of a resonance line. If just those frequencies on the upper side of the Doppler line are removed, cooling will occur. Such removal might come about by absorption from a volume of gas moving toward the region being cooled.

The process of cooling by narrow-band light requires that each atom scatters many photons. Thus the atom must have a high probability of returning to the original lower state so that it can scatter repeatedly. If strong monochromatic sources of X-rays become available, cooling of even complex atoms and molecules could occur by just a few scattering events, although the residual recoil momentum may then become an important limiting factor. It should also be noted that the cooled region should be optically thin enough so that most of the scattered light can escape to a distant absorber.

The possibility of cooling by nearly monochromatic light illustrates that such radiation is equivalent, in a thermodynamic sense, to mechanical work or electricity rather than heat energy [2], even though this particular process is envisioned as operating far from equilibrium. When monochromatic light of low entropy is scattered by a moving atom, the frequency of the scattered light and so the energy of the scattered photon, is higher by an amount depending on the recoil direction. Thus the bandwidth and so the disorder of the light is increased in the cooling process.

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