Optical pumping, light shifts and laser cooling

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S. PANCHARATNAM wrote a series of beautiful papers on optical pumping problems and related phenomena[1]. He was interested in interference effects which can be observed when several routes can be followed by an atomic system during a photon scattering process. Such interference effects can also be analysed in terms of off diagonal elements of the atomic density matrix, the so-called Zeeman coherences. A different way of representing population differences between Zeeman sublevels or Zeeman coherences between pairs of sublevels is to use vectors or ellipsoids for describing the corresponding anisotropy of the atomic state, more precisely its orientation or its alignment. Several papers of Pancharatnam are devoted to the analysis of the birefringence of an optically pumped vapour due to spin alignment. He interpreted the modulated signals which can be observed under magnetic resonance in terms of a precessional motion of the ellipsoid of alignment. Another problem investigated by Pancharatnam concerns light shifts, i.e. the fact that Zeeman sublevels in atomic ground states can be displaced by quasi-resonant light. He tried to interpret these light shifts with a semiclassical dispersion theory and establish a connection between light shifts and the dispersive part of the refractive index.

All Pancharatnam's works were done at a time where the only light sources available for optical pumping experiments were spectral lamps, excited by dc or microwave discharges and emitting a light with a broad spectral width and a weak intensity. The spectacular development of tunable laser sources, which started in the early seventies, stimulated several experimental and theoretical studies dealing with resonance fluorescence in intense resonant monochromatic fields. More attention was paid to the exchanges of linear momentum between atoms and photons and it became clear that it was also possible to manipulate with laser beams the translational degrees of freedom of an atom, i.e. its position and velocity. A new research field, called laser cooling and trapping of atoms, has appeared and is expanding very rapidly[2]. Surprisingly enough, it turns out that several of the important new developments in this field are based on physical effects, such as optical pumping and light shifts which were investigated in the early sixties[3]. In this special issue dedicated to the memory of S. Pancharatnam, I would like to briefly describe two examples of recent developments which, I am sure, would have pleased him, because they use concepts which were quite familiar to him.

Sisyphus cooling

When the atomic ground state has a non-zero angular momentum $J_g \neq 0$, i.e. when it has several Zeeman sublevels, these sublevels undergo in general different light shifts, which depend on the light polarization. In zero magnetic field, the Zeeman degeneracy can be removed by light. In non-zero magnetic field, the splittings between Zeeman sublevels are changed by light and magnetic resonance curves, which can be very narrow in atomic ground states, are light-shifted. Thus were the light shifts observed for the first time in the early sixties[4].

In laser cooling experiments, the laser frequency is slightly detuned from resonance. Light shifts, which are due to quasi-resonant light, are thus different from zero in the atomic ground state. Furthermore, in most experiments, which use three orthogonal pairs of counterpropagating waves, the polarization of the laser electric field varies in space. It follows that the light shifts of the various ground state sublevels, which vary from one sublevel to the other and depend on the light polarization, are spatially modulated. In addition to virtual absorptions and reemissions of photons by the atom, which are at the origin of light shifts, there are real absorption and emission processes, which modify the populations of the various ground state sublevels (and the Zeeman coherences). The corresponding optical pumping rates from one sublevel to the other also depend on the laser polarization and they must therefore exhibit a spatial modulation, which is correlated with the spatial modulation of light shifts, because both modulations have the same origin, which is the spatial modulation of the laser polarization.

We want to show now how such correlated spatial modulations of light shifts and optical pumping rates can give rise to *efficient cooling mechanisms*. For the sake of simplicity, we consider a one-dimensional laser configuration formed by two laser plane waves, propagating in opposite directions along the *z*-axis, with orthogonal linear polarizations, parallel to the *x*-axis and to the *y*-axis, respectively (lin ⊥ lin configuration). The phase difference between the two waves varies along the *z*-axis

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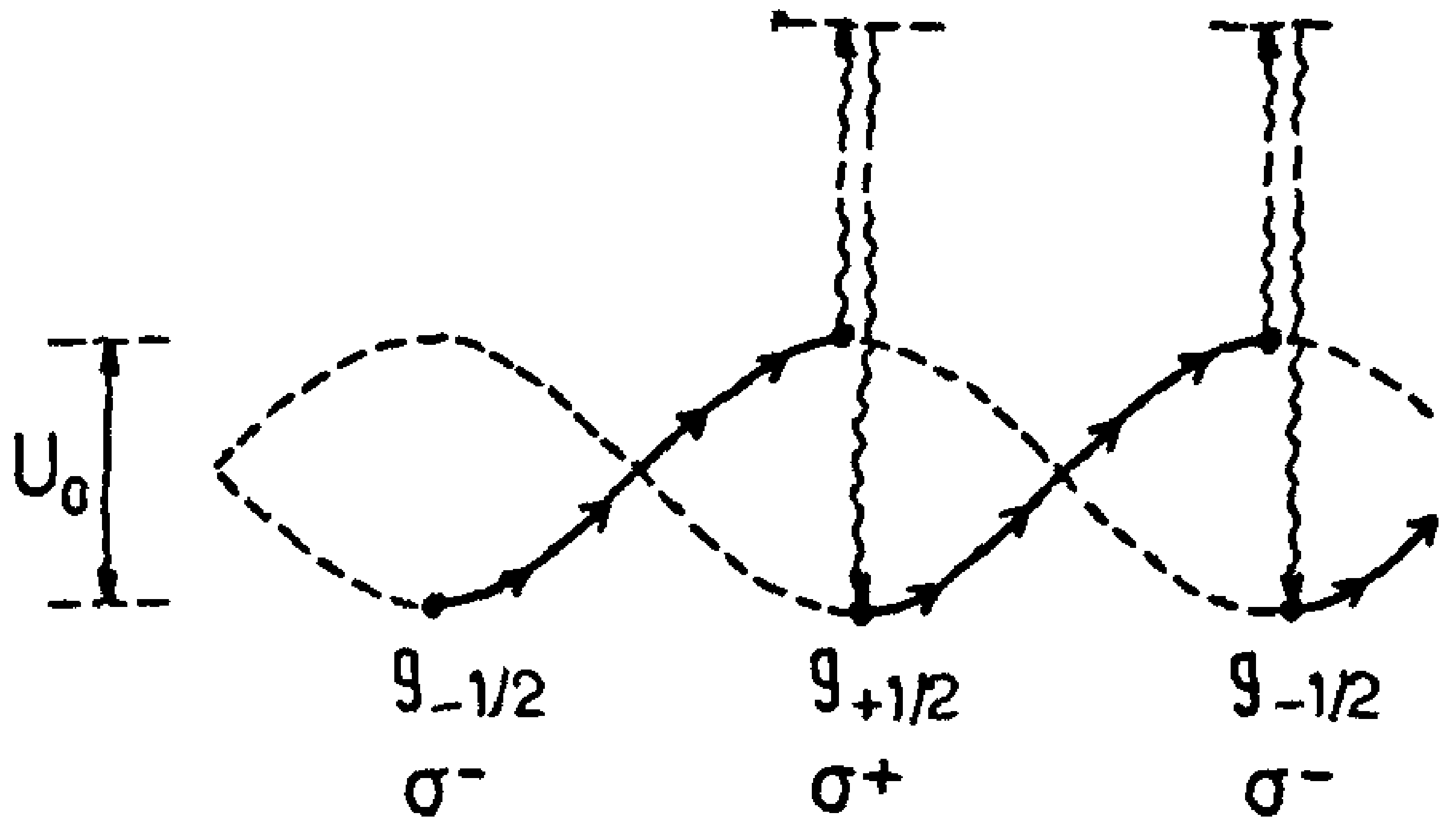

Figure 1. Correlated spatial modulations of light shifts and optical pumping rates for a $J_g = 1/2 \to J_e = 3/2$ transition in a one-dimensional lin ⊥ lin laser configuration. As a consequence of these correlations, the moving atom is running up the potential hills more often than down.

and the total field has a polarization which varies from σ^+ (right circular) to σ^- (left circular) every quarter wavelength $\lambda/4$, the field having in-between an elliptical polarization, which can become linear, with axes at 45° from the x and y-axis. We also suppose that such a laser configuration excites an atomic transition $g \leftrightarrow e$ with angular momenta $J_g = 1/2$ and $J_e = 3/2$. No magnetic field is applied, so that the two Zeeman sublevels $g_{\pm 1/2}$ of g are degenerate in the absence of light. If the laser frequency ω_L is detuned towards the red of the atomic frequency ω_A ($\omega_L < \omega_A$), light shifts are negative. In the places where the light polarization is σ^+, the sublevel $g_{+1/2}$ is shifted downwards three times more than $g_{-1/2}$, because the square of the σ^+ Clebsch–Gordan coefficient starting from $g_{+1/2}$ is three times larger than the square of the σ^+ Clebsch–Gordan coefficient starting from $g_{-1/2}$. A non-zero splitting U_0 appears between the two sublevels (Figure 1). The conclusions are reversed in the places where the polarization is σ^-, the sublevel $g_{-1/2}$ being shifted downwards three times more than $g_{+1/2}$. Figure 1 represents the corresponding spatial modulations of the light-shifted energies of $g_{\pm 1/2}$. The vertical arrows of this figure also show the optical pumping processes which transfer atoms from $g_{-1/2}$ to $g_{+1/2}$ in the places where the polarization is σ^+, and from $g_{+1/2}$ to $g_{-1/2}$ in the places where the polarization is σ^-. It clearly appears in Figure 1 that optical pumping rates and light shifts are correlated: the optical pumping rate is more important from the higher Zeeman sublevel to the lower one.

Consider now an atom moving to the right and starting from the bottom of a valley of the potential curve of the sublevel $g_{-1/2}$. If the optical pumping time is long enough, the atom has enough time to climb the potential hill and to reach the top of this hill where the probability for the atom to be transferred by optical pumping to the other sublevel $g_{+1/2}$, i.e. to the bottom of the next valley, is maximum. From there, the same sequence can be repeated (Figure 1). Such a situation, where the moving atom is running up potential hills more often than down, is quite similar to the situation of Sisyphus in Greek mythology. It gives rise to a very efficient and general cooling mechanism[5]. When the atom climbs up a potential hill, it is slowed down: part of its kinetic energy is transformed into potential energy which is then dissipated by spontaneous anti-Stokes Raman processes where the energy of the emitted photon is larger than the energy of the absorbed laser photon. It is not the place here for presenting a detailed theory of Sisyphus cooling and various subsequent developments,[1] such as the observation of quantization of atomic motion in an optical potential well or the realization of spatially ordered arrays of atoms, called now optical lattices[6]. Our motivation was just to show how well-known effects, such as optical pumping and light shifts, which were very familiar to Pancharatnam, could be combined in new ways, to give rise to new cooling mechanisms.

Subrecoil cooling using velocity selective dark states

In most cooling schemes, including Sisyphus cooling, fluorescence cycles never cease, giving rise to fluorescence photons which are spontaneously emitted in random directions and which communicate a random recoil to the atom. The atomic momentum spread Δp cannot, in these conditions, be smaller than the photon momentum $\hbar k$, and the corresponding temperature T, defined by $k_B T/2 = \Delta p^2/2M$, is always larger than the recoil limit T_R defined by $k_B T_R/2 = E_R$, where $E_R = \hbar^2 k^2/2M$ is the recoil kinetic energy of an atom absorbing or emitting a single photon. In order to overcome the recoil limit and to achieve $T < T_R$ one must inhibit the absorption of light for atoms with $p \simeq 0$. One must protect ultracold atoms from the 'bad' effects of light, i.e. from the random recoil due to fluorescence photons. That can be achieved by using a physical effect, called velocity selective coherent population trapping[7].

Coherent population trapping can be simply explained for an atom having two ground state sublevels g_1 and g_2 and one excited sublevel e_0 (Λ-type configuration of Figure 2 *a*). Two laser beams with frequencies ω_{L1} and ω_{L2} drive the two transitions $g_1 \leftrightarrow e_0$ and $g_2 \leftrightarrow e_0$, respectively. We call Δ the detuning from resonance for the stimulated Raman process consisting of the absorption of one ω_{L1} photon followed by the stimulated emission of one ω_{L2} photon (or vice versa). If the fluorescence rate R_F from such a system is plotted versus Δ, one finds that R_F strictly vanishes for $\Delta = 0$ (Figure 2 *b*). The dip of Figure 2 *b* around $p = 0$ has a width Γ', which is the radiative width of the ground state, much smaller than the natural width Γ of the excited state

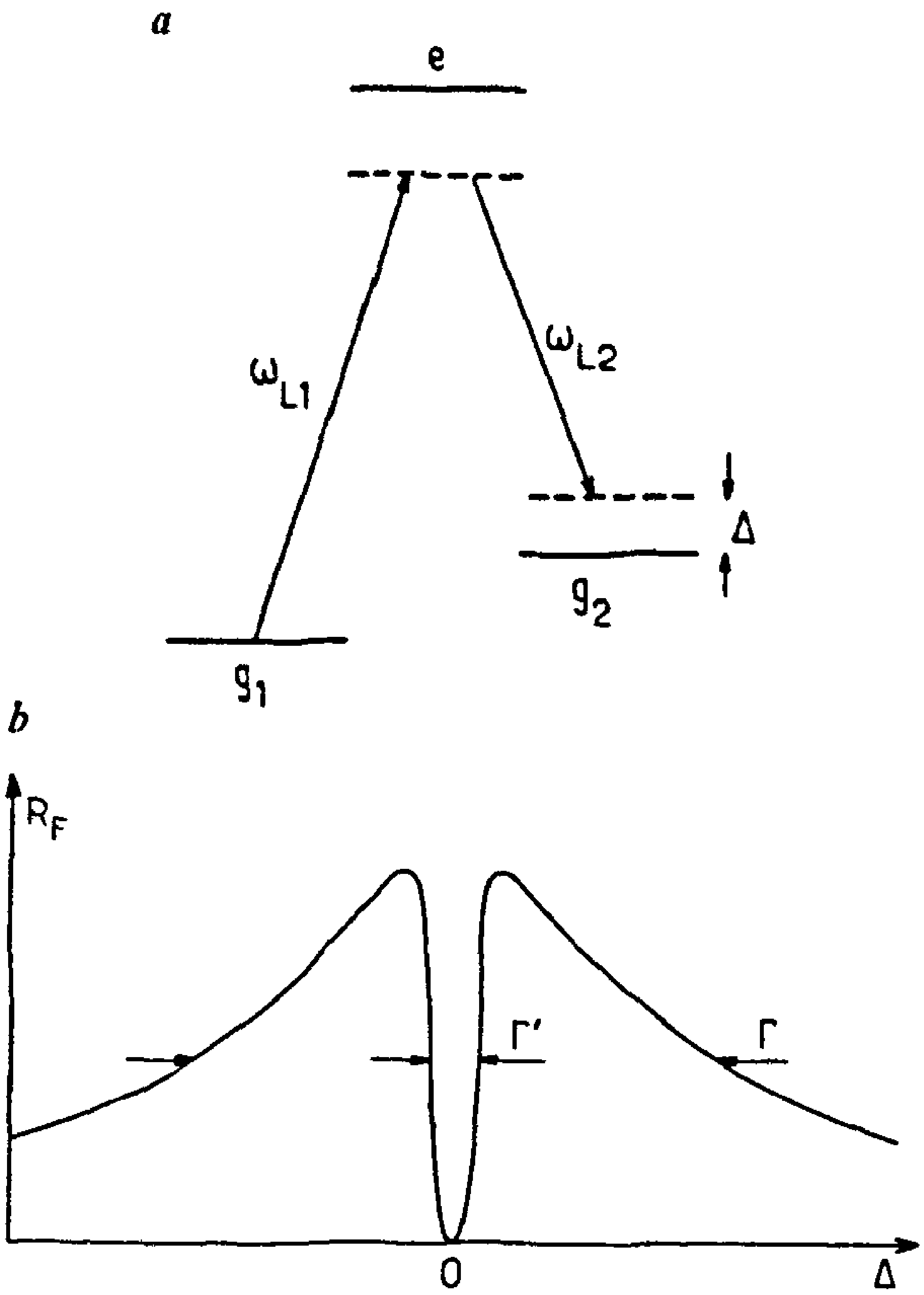

Figure 2. *a*, Three-level atom forming a Λ-configuration Δ is the detuning from resonance for the stimulated Raman process associated with the two laser fields ω_{L1} and ω_{L2} driving the transitions $g_1 \leftrightarrow e_0$ and $g_2 \leftrightarrow e_0$, respectively. *b*, Fluorescence rate R_F versus Δ. Γ and Γ' are the widths of the excited state *e* and ground state *g*, respectively.

which determines the rate of decrease of R_F at large Δ. The vanishing of R_F for $\Delta = 0$ is due to the fact that atoms are optically pumped in a linear superposition of g_1 and g_2, $|\psi_{NC}\rangle = c_1|g_1\rangle + c_2|g_2\rangle$, such that the two absorption amplitudes from g_1 to e_0 and from g_2 to e_0 interfere destructively. For $\Delta = 0$, such a state, which is not coupled to e_0 by the laser–atom interaction Hamiltonian, is in addition a stationary state, i.e. an eigenstate of the total Hamiltonian *H*. Atoms pumped in $|\psi_{NC}\rangle$ remain there forever, so that $|\psi_{NC}\rangle$ is a perfect trap, called also sometimes a 'dark state'. For $\Delta \neq 0$, $|\psi_{NC}\rangle$ is no longer an eigenstate of *H*. It is contaminated by other unstable states and the trap is no longer perfect. This explains why R_F increases with Δ around $\Delta = 0$.

To apply such a scheme to laser cooling, the key point is to make Δ proportional to the atomic velocity *v*. This can be achieved by choosing the wave vectors k_1 and k_2 of the two laser waves ω_{L1} and ω_{L2} in such a way that the stimulated Raman process between g_1 and g_2 is not Doppler free. For example, if k_1 and k_2 have opposite directions along the *z*-axis (and if $|k_1| \simeq |k_2| \simeq k$), the Doppler shift for the stimulated Raman process is $\Delta = 2kv$, because in the atom rest frame moving with a velocity *v* along the *z*-axis, the two laser waves have opposite Doppler shifts. In Figure 2 *b*, one can then replace Δ by $2kv$. Coherent population trapping becomes velocity selective; atoms with $v = 0$ no longer absorb light, and the smaller *v* is, the smaller is the absorption rate. In fact, velocity selection is not enough to achieve cooling. The density of atoms near $v = 0$ must also be increased. Such an increase is obtained through optical pumping in *v*-space. Atoms with $v \neq 0$ can absorb light and reemit fluorescence photons which communicate to them a random recoil. After a fluorescence cycle, the atomic velocity changes and it can happen that the new atomic velocity becomes closer to zero than the initial one, before the fluorescence cycle. Atoms can thus be transferred from the $v \neq 0$ absorbing states into the quasidark states $v \simeq 0$ where they remain trapped and accumulate. The longer the interaction Θ, the narrower the range δv around $v = 0$ in which the atoms can remain trapped during Θ. In fact, one can show that $\delta v \sim 1/\sqrt{\Theta}$, and consequently that there are no fundamental lower limits to the temperature *T* which can be achieved by such a scheme and which varies as $1/\Theta$.

The previous considerations are very qualitative. A more careful analysis is needed to evaluate the possibilities of such a subrecoil cooling scheme and to extend it to two and three dimensions. In particular, when Δp becomes smaller than $\hbar k$, the de Broglie wavelength $\lambda\!\!\!^{-}_{dB} = \hbar/\Delta p$ is larger than the laser wavelength $\lambda\!\!\!^{-}_{L} = 1/k$. This mean that atoms are delocalized in the laser wave and that their translational degrees of freedom must be treated quantum mechanically. It is not the place here to present these treatments and to describe the latest experimental developments in this field. I have mentioned this cooling scheme because it uses quantum interference effects between absorption amplitudes, a problem which also interested Pancharatnam in the early days of optical pumping[8].

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1. See papers 17 to 25 in *Collected Works of S Pancharatnam*, with a foreword by G W. Series, Oxford University Press, 1975.
2. See for example 'Laser manipulation of Atoms and Ions', Proceedings of the 1991 Varenna Summer School (eds. Arimondo, E., Phillips, W D. and Strumia, F), North-Holland, Amsterdam, 1992.
3. A simple review of these new developments using optical pumping and light-shifts can be found in Cohen-Tannoudji, C. and Phillips, W., *Phys. Today*, 1990, **43**, 33.
4. Cohen-Tannoudji, C., *C.R. Acad. Sci.*, 1961, **252**, 394.
5. Dalibard, J. and Cohen-Tannoudji, C., *J. Opt. Soc Am*, 1989, **B6**, 2023.
6. See for example the search and discovery paper of Collins, G P, in *Phys. Today*, June 1993, p. 17 and references therein
7. Aspect, A., Arimondo, E., Kaiser, R., Vansteenkiste, N and Cohen-Tannoudji, C., *Phys. Rev. Lett*, 1988, **61**, 826, and *J Opt. Soc Am.*, 1989, **B6**, 2112 and references therein to earlier work on coherent population trapping.
8. See for example paper 17 of Ref. 1.