

PATENT SPECIFICATION

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(54) SELECTIVE EXCITATION OF ATOMS OR MOLECULES TO HIGH-LYING STATES

(71) We, MASSACHUSETTS INSTITUTE OF TECHNOLOGY, a corporation organised and existing under the laws of the state of Massachusetts, United States of America, residing at 77 Massachusetts Avenue, Cambridge, Massachusetts 02139, United States of America, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to selective excitation of atoms or molecules to high lying states and more particularly to apparatus for separating and a method of separating different isotopes of the same element by selective excitation of the isotopes.

The literature on previous attempts at isotope separation is quite extensive. Present production methods largely involve the known, extremely costly and inefficient diffusion process. This process is only viable in very large scale installations.

Recently, proposals for using lasers to separate isotopes have appeared, and proposed techniques include frequency selective excitation of one isotope-shifted transition in an interaction region. The excited species can then be swept out of the interaction region by chemical or ionization processes. These techniques require narrow-banded lasers, stabilized to distinguish in frequency between very closely spaced levels. These requirements often prevent very high powered or efficient systems required for production applications.

Another suggested technique using lasers involves deflection of one isotope with respect to another isotope in an atomic or molecular beam using momentum transfer from resonantly absorbed laser light. Again, these techniques require a very narrow-banded laser, good stability, and, in addition, an atomic beam apparatus which generally means that one is working with a lower density of atoms than, say, with a gas cell,

and hence the production of separated isotopes in an atomic beam apparatus can be expected to be lower than in a gas cell. 50

It is an object of the present invention to provide apparatus and method for selective excitation of one particular isotope of a mixture of isotopes into a state where the excited species can readily be swept out and collected, for example by ionization. 55

According to one aspect of the present invention, there is provided a method of selectively populating high-lying excited states of atoms or molecules comprising (i) energizing the atoms or molecules with a pulse of a first circularly-polarized pulsed radiation, the coherent frequency components of the first pulsed radiation having frequencies corresponding to the energy difference between a lower energy level and the frequency split levels of an intermediate energy level, and the duration of the pulse being less than $2\pi/\Delta\omega$, where $\Delta\omega$ is the frequency difference of the split levels; and (ii) applying a pulse of a second circularly-polarized pulsed radiation to the atoms or molecules after the termination of the first pulse, so as to provide an upper energy level with a greater population than existed therein before the atoms or molecules were excited by the first and second radiations, the coherent frequency components of the second pulsed radiation having frequencies corresponding to the energy difference between the split levels of the intermediate energy level and the upper energy level, and the duration of the second pulse being less than $2\pi/\Delta\omega$; the first and second radiations having the same handedness of circular polarization. 60 65 70 75 80 85

The invention also provides apparatus for selectively populating high-lying excited states of atoms or molecules comprising (i) means for energizing the atoms or molecules with a pulse of a first circularly-polarized pulsed radiation, the coherent frequency components of the first pulsed radiation having frequencies corresponding to the energy difference between a lower energy level and 90 95

the frequency split levels of an intermediate energy level, and the duration of the pulse being less than $2\pi/\Delta\omega$, where $\Delta\omega$ is the frequency difference of the split levels; (ii) means for applying a pulse of a second circularly-polarized pulsed radiation to the atoms or molecules after the termination of the pulse of first radiation so as to provide an upper energy level with a greater population than existed therein before the atoms or molecules were excited by the first and second radiation pulses, the coherent frequency components of the second pulsed radiation having frequencies corresponding to the energy difference between the split levels of the intermediate energy level and the upper energy level, and the duration of the second pulse being less than $2\pi/\Delta\omega$, the first and second radiations having the same handedness of circular polarization.

By applying an electric field and by providing means to generate such a field, it is possible to ionize those atoms or molecules which are raised to the upper level. The ionized particles are then readily separable from the remaining un-ionized particles. The method and apparatus therefore afford a way of separating isotopes.

According to a second aspect of the invention, therefore, there is provided a method of separating isotopes of an element comprising (i) energizing atoms or molecules containing atoms of the element, isotopes of which are to be separated, with a first circularly-polarized pulsed radiation, the coherent frequency components of the first pulsed radiation having frequencies corresponding to the energy difference between a lower energy level and the frequency split levels of an intermediate energy level, and the duration of a pulse of the first radiation being less than $2\pi/\Delta\omega$ where $\Delta\omega$ is the frequency difference of the split levels; (ii) applying a second circularly-polarized pulsed radiation to the atoms or molecules after termination of the first pulse, the two radiations having the same handedness of circular polarization, the coherent frequency components of the second pulsed radiation having frequencies corresponding to the energy difference between the split levels of the intermediate energy level and an upper energy level, and the duration of the pulse of the second pulsed radiation being less than $2\pi/\Delta\omega$; (iii) applying an electric field to the energized atoms or molecules after application of the second radiation to ionize atoms or molecules raised to the upper level; and (iv) separating the ionized atoms or molecules of one isotope from the un-ionized atoms or molecules of the other isotope.

The invention furthermore provides apparatus for separating isotopes of an element, comprising (i) means for energizing atoms or molecules containing atoms of the

element, isotopes of which are to be separated, with a first circularly-polarized pulsed radiation, the coherent frequency components of the first pulsed radiation having frequencies corresponding to the energy difference between a lower energy level and the frequency split levels of an intermediate energy level, and the duration of the pulse being less than $2\pi/\Delta\omega$ where $\Delta\omega$ is the frequency difference of the split levels; (ii) means for applying a second circularly-polarized pulsed radiation to the atoms or molecules after the termination of the first pulse, the two radiations having the same handedness of circular polarization, the coherent frequency components of the second pulsed radiation having frequencies corresponding to the energy difference between the split levels of the intermediate energy level and an upper energy level, and the duration of the pulse of the second pulsed radiation being less than $2\pi/\Delta\omega$; (iii) means for applying an electric field to the energized atoms or molecules after application of the second radiation to ionize atoms or molecules raised to the upper level; and (iv) means for separating the ionized atoms or molecules of one isotope from the un-ionized atoms or molecules of the other isotope.

To practice the invention, pulsed lasers can be employed; the primary requirements are to have reasonably short pulses—microsecond or nanosecond regime depending upon the atomic or molecular species involved, some delay between pulses, and the same handedness of circular polarization. A comparatively gross timing and stability is all that is required of the lasers, and far less stringent constrictions apply than for prior laser techniques.

Typically, transition linewidths are tens to hundreds of MHz wide

$$(3 \times 10^4 \text{ MHz} = 1 \text{ cm}^{-1} \text{ in Energy}).$$

Isotope shifts of spectral lines are typically hundreds to about a thousand MHz. Thus the prior techniques require lasers having narrower bandwidths than these isotope shifts and stabilized to wander by not more than one linewidth. In the present technique, typical laser linewidths and jitters of tens of thousands of MHz (and sometimes more) are allowable. This permits more powerful laser systems to be used. The laser technology exists and is well-developed. The method is quite efficient and especially adaptable to large scale production because of its simplicity. It must be emphasized that this invention can be applied to either atomic or molecular species containing atoms of an element isotopes of which are to be separated, whichever is more efficient for the isotope to be separated.

The present invention will now be des-

cribed in more detail by way of example only with reference to the accompanying drawings, in which:

Fig. 1(a) shows schematically a system of sublevels in the (I, J, F, M_f) representation;

Fig. 1(b) gives the same levels as in Fig. 1(a) in the degenerate (I, J, M_i, M_j) representation;

Fig. 2(a) is a block diagram illustrating a preferred embodiment of this invention;

Fig. 2(b) is a detailed drawing of a variable delay unit shown in Fig. 2(a);

Fig. 2(c) is a timing diagram showing the sequence of laser pulses and an electric field pulse;

Fig. 3(a) shows schematically a system of sublevels in the (I, J, F, M_f) representation for relevant sodium energy levels; and

Fig. 3(b) shows the same levels as in Fig. 3(a) in the degenerate (I, J, M_i, M_j) representation.

The central aspect of the invention is the successive excitation of the atoms or molecules of an element consisting of or containing the isotope species to be separated by two pulsed lasers having the same sense or handedness of circular polarization. The first laser pulse creates a coherent superposition state in an intermediate level. The second laser pulse serves to excite selectively atoms or molecules prepared in this superposition state to a higher level. The critical conceptual aspect of this technique centres upon the first step i.e. the creation of the coherent superposition state. Therefore, a detailed description of this step is useful.

Fig. 1(a) shows schematically a particular system of sublevels in the (I, J, F, M_f) representation. Levels *a* and *b* are separated by a frequency split $\Delta\omega$ by the hyperfine interaction. Laser 1 of Fig. 2 excites a coherent superposition from level *g* to levels *a* and *b*; laser 2 causes transitions from this superposition to a higher level *k*.

Consider the first pulsed laser exciting an atomic or molecular level which has a frequency splitting $\Delta\omega$ (see Fig. 1(a)). For example, this splitting can be due to fine structure or hyperfine structure. The splitting could also be externally induced by electric or magnetic fields or other perturbations. If the laser pulse duration Δt is short enough, $\Delta t < 2\pi/\Delta\omega$ each photon has Fourier frequency components over a frequency range larger than $\Delta\omega$ and it creates an excited state that is a coherent superposition of states *a* and *b*. Optimally, the centre frequency of the laser should be within $2\pi/\Delta t$ of the frequencies corresponding to the energy differences of the average energy of level *a* and level *b*. When this optical frequency is used, the Fourier power spectrum is centred about the frequency separation of the levels to be excited. The centre frequency of the laser is not critical and it is apparent that photons centred

about frequencies greater than $2\pi/\Delta t$ can still contribute to creating coherent superposition states of the *a* and *b* levels because of the wide spectrum of a short pulse.

Fig. 1(b) gives the same levels in the degenerate (I, J, M_i, M_j) representation. Here U_1, V_1 and W_1 have $M_j = -\frac{1}{2}$; U_2, V_2 and W_2 have $M_j = +\frac{1}{2}$. The incoming photons are right-hand circularly polarized and according to the selection rules, each must cause M_i to increase by one. Thus excitation of the uppermost state is only possible when the intermediate state has some "V₁-like" character.

At this point, a brief discussion of these different ways of representing the same quantum states may be useful in understanding the principle of operation of this invention. In the case of hyperfine structure, the

total electron angular momentum \vec{J} , interacts

with the nuclear spin, \vec{I} , and splits a particular energy level into several levels each characterized by a different magnitude of the total

angular momentum $\vec{F} = \vec{I} + \vec{J}$. In the case of $J = \frac{1}{2}$, shown in Fig. 1(b), the quantum

number corresponding to \vec{F} can have the two values $F = I \pm \frac{1}{2}$. The states are described by the principal quantum number *n* and the quantum numbers *I*, *J*, *F* and m_F , where the

latter corresponds to the projection of \vec{F} onto a particular axis in space, F_z . However, the states can equally well be expressed in terms of *n*, *I*, *J*, m_I and m_J . Here the last two are the quantum numbers of I_z and J_z . These two different, but equivalent descrip-

tions are possible because \vec{F} is simply the vector sum of \vec{I} and \vec{J} .

For the $J = \frac{1}{2}$ case, level *a* corresponds to $F = I - \frac{1}{2}$ and *b* to $F = I + \frac{1}{2}$, each with the same m_F . Levels *a* and *b* are linear combinations of levels V_1 and V_2 . V_2 and V_1 have $m_j = -\frac{1}{2}$, $m_i = m_F - \frac{1}{2}$; and $m_j = -\frac{1}{2}$, $m_i = m_F + \frac{1}{2}$, respectively.

Similarly, the *g* level of Fig. 1(a) is a lower level with $m_F' = m_F - 1$, and *g* is a linear combination of degenerate levels U_1 and U_2 . U_1 has $m_j = -\frac{1}{2}$, $m_i = m_F' + \frac{1}{2}$, and U_2 has $m_j = +\frac{1}{2}$, $m_i = m_F' - \frac{1}{2}$.

If the laser is right-hand circularly polarized, the only permissible excitation between the lower level and the intermediate level occurs between U_1 and V_2 .

Fig. 1(b) shows that if we prepare the superposition states at $t=0$ with right-hand circularly polarized (rhcp) light, it is all "V₂-like" in character. This follows because absorption of rhcp light increases m_j by 1 according to selection rules. As the state evolves freely in time, after the laser pulse

has terminated, the V_1 component builds up by a transfer from level V_2 until it reaches a maximum at $t=\pi/\Delta\omega$. In fact, the amount of occupancy of V_1 oscillates with a period of $T=2\pi/\Delta\omega$, exactly the characteristic period of the frequency splitting $\Delta\omega$. This is called "resonance oscillation".

A second laser pulse, also with rhcp and incident on the system at $t=\pi/\Delta\omega$ can excite atoms out of the superposition state into a high-lying level k .

The level k of Fig. 1(a) is an upper level with $m_F''=m_F'-1$ and k is a linear combination of degenerate levels w_1 and w_2 . w_1 has $m_F=-\frac{1}{2}$, $m_I=m_F''+\frac{1}{2}$ and w_2 has $m_F=+\frac{1}{2}$, $m_I=m_F''-\frac{1}{2}$. As the laser is right-hand circularly polarized, the excitation occurs between the intermediate level v_1 , and the upper level w_2 . Atoms in this high-lying level can now be ionized easily (several hundred volts/cm electric fields), then swept out and collected efficiently by electric fields.

This laser excitation can take place in an atomic (or molecular) beam or a gas cell. The latter permits higher densities of atoms (or molecules). The only restriction on a cell are the destruction of high-lying states by collisions—this should only begin to be important at higher pressures than are typical in atomic beams.

An example of this phenomenon is in exciting $\text{Na}^{23}(I=3/2)$. Fig. 3 shows the relevant sodium energy-levels in the F representation (Fig. 3(a)) and schematically in the degenerate representation (Fig. 3(b)). Theoretical understanding of this real case lies in realizing that it consists of several $g \rightarrow a, b \rightarrow k$ sublevel systems as discussed above and as shown in Fig. 1. In the sodium case; the g levels are sublevels of the $3s_{1/2}$ ground state; the a and b levels are sublevels of the $F=2$ and $F=1$ levels of the $3p_{1/2}$ state respectively; and the k levels are sublevels of high-lying $ns_{1/2}$ levels, n being 4 or greater than 4. Since all levels involved have $J=\frac{1}{2}$, each sublevel system in the degenerate representation looks similar to Fig. 3(b).

In the case of the $3p_{1/2}$ level of sodium, the frequency splitting between the $F=2$ and $F=1$ states is 200 MHz. This means that the laser pulses should be shorter than 5n sec. and the time between pulses, $t=\pi/\Delta\omega$, should be approximately $2\frac{1}{2}n$ sec.

It must be emphasized that these examples where $J=\frac{1}{2}$ for all levels are just simple cases of the isotope separation method. The method is valid for a wide variety of more complex level structures. In particular, for example, any atom or molecule with hyperfine splitting (any I and J) will exhibit similar resonance oscillation and be subject to selective excitation versus an isotopically different species.

It must be noted that in the above discussion both lasers could have been left-hand

circularly polarized and the superposition state could have been all "V₁-like" at $t=0$. What is important is that both lasers have the same sense of circular polarization for maximum excitation of the upper-most level.

Typically atoms (or molecules containing such atoms) of odd atomic weight (as U^{235}) have hyperfine structure as discussed above. For the isotope (say U^{238}) of even atomic weight, where there is no hyperfine structure, there is no resonance oscillation, and hence the second rhcp laser pulse is much less likely to excite to the high-lying level. In the $J=\frac{1}{2}$ case, a high-lying state is not populated at all for the even isotope. Thus by simply practicing this invention, the odd isotope can be excited and separated from the even isotope.

The resonance oscillation method has been described for the case where hyperfine structure is exhibited by one isotope but not by the other isotope. In fact, the method is of more general application and can be used to separate isotopes even if both are excitable to levels which exhibit frequency splitting. In this latter case, the splitting of one isotope ($\Delta\omega_1$), is almost always different from that of another ($\Delta\omega_2$). Thus the time of application of the second laser pulse can be adjusted for maximum excitation of just one isotope. In particular, if isotope 1 is to be excited, the optimal time t of application of the second laser pulse is $t=\pi/\Delta\omega_1$. Similarly, for optimal excitation of the second isotope, $t=\pi/\Delta\omega_2$. In these cases selectivity in excitation occurs because the optimal excitation time for one isotope differs from that of the other—and thus they are excited to the higher state with different probabilities.

In all these cases, the desired isotope can either be the one selectively excited and swept out by an ion collector or the one remaining in the interaction region.

Figure 2 illustrates equipment comprising a preferred embodiment of the invention. Lasers 1 and 2 are pumped by the same laser 3—as for example two tunable pulsed dye lasers 1, 2 pumped by the same pulsed nitrogen laser 3. The output of laser 3 is divided by a beam splitter 8. One beam 9 pumps laser 1. The other beam 10 is reflected by mirror 11 before pumping laser 2. The output of laser 2 is delayed in a variable delay unit 4 with respect to the output of laser 1 by bouncing the first-mentioned output off movable mirrors 5, 6 of variable separation as shown by direction arrows 7 in Fig. 2(b). Mirror 12 reflects the output of laser 1 into variable delay unit 4 where it is merely redirected by mirror 13 to impinge upon the atoms or molecules 14 in a chamber 15. The outputs of lasers 1 and 2 are circularly polarized by polarizers 16. The polarizers 16 may be either right or left hand polarizers so long as both are the same.

The laser 3 is triggered in a conventional manner by a pulse source 17 which produces a continuous train of pulses. The pulses from source 17 are also fed to a delay unit 19 to provide a pulse delay t' which is greater than the delay $t = \pi/\Delta\omega$ of the pulse from laser 2. A delayed pulse from unit 19 is fed to and triggers a pulsed voltage source 20 which develops an ionizing voltage $+V$ between two spaced electrode plates 21, 22 within the chamber 15. The space between plates 21, 22 contains a gas which has been energized by lasers 1 and 2. The gas atoms are ionized and the resulting ions are also accelerated toward plate 22 by the ionizing voltage so that they pass through a screen 23 which forms a major portion of plate 22. A suitably biased ion collector 18, biased by connection e.g. to a negative voltage source 24, collects the ions which pass through the screen 23 to provide substantially only one isotope at outlet 25 whereas the remainder of the gas exits at outlet 26. Chamber 15 is furnished with gas to be treated at inlet 27.

Fig. 2(c) shows the timing sequence of the laser pulses and the ionizing electric field pulse. The pulse duration of laser 1 at time $t=0$ is optimally considerably shorter than $2\pi/\Delta\omega$. This assures a high degree of coherence in the superposition state. The duration of laser pulse 2 is also optimally much shorter than $2\pi/\Delta\omega$. This allows maximum selectivity of excitation from the superposition state at time $t=\pi/\Delta\omega$. The ionizing electric field pulse preferably closely follows the pulse from laser 2 in order to minimize the period and hence allow a higher repetition rate.

Although in the preferred embodiment the ion-creating electric field is established by a pulsed source energized after the second laser radiation was terminated, it is apparent that in an alternative embodiment of the invention, the electric field can be continuously applied. A feature of this embodiment is that the ionization voltage produces a change in the frequency separation of the levels and causes additional energy levels. The perturbations in the energy levels of the gas because of the continuously applied ionizing field can be avoided if instead a pulsed ionizing field is applied after the second laser pulse since there are no perturbations in the absence of the ionizing field.

Although the second laser pulse has been described as being applied at a time $\pi/\Delta\omega$ after the first laser pulse, it will be apparent to those skilled in the art that since the amount of the level that is V_1 in character oscillates with a period of $T=2\pi/\Delta\omega$, the second pulse could be applied at times $n\pi/\Delta\omega$ where $n=1, 3, 5, \dots$

Although the particular embodiment of the invention just described uses two separate excitation lasers, it will be apparent to those skilled in the art that if the frequency corres-

ponding to the difference of the first and second energy levels is close to that of difference of the second and third energy levels, then the two lasers may be replaced by only one laser whose circularly polarized beam is split in a beam splitter to provide two beams corresponding to the excitation output beams from lasers 1 and 2. One beam, corresponding to the output of laser 2, is delayed in variable delay unit 4 of Fig. 2. The apparatus otherwise is the same as that of Fig. 2.

The method and apparatus as explained in detail hereinbefore are employed specifically to effect a separation of one isotope from another isotope or other isotopes. The method and apparatus can, however, be used for achieving selectively-populated high level energy states of atoms or molecules. The present invention may find application in preparing a chemical reactant for readiness to participate in a chemical reaction. Moreover, the invention may be applicable in the preparation of isotopically-tagged compounds.

WHAT WE CLAIM IS:—

1. A method of selectively populating high-lying excited states of atoms or molecules comprising (i) energizing the atoms or molecules with a pulse of a first circularly-polarized pulsed radiation, the coherent frequency components of the first pulsed radiation having frequencies corresponding to the energy difference between a lower energy level and the frequency split levels of an intermediate energy level, and the duration of the pulse being less than $2\pi/\Delta\omega$, where $\Delta\omega$ is the frequency difference of the split levels; and (ii) applying a pulse of a second circularly-polarized pulsed radiation to the atoms or molecules after the termination of the first pulse, so as to provide an upper energy level with a greater population than existed therein before the atoms or molecules were excited by the first and second radiations, the coherent frequency components of the second pulsed radiation having frequencies corresponding to the energy difference between the split levels of the intermediate energy level and the upper energy level, and the duration of the second pulse being less than $2\pi/\Delta\omega$; the first and second radiations having the same handedness of circular polarization.

2. A method of separating isotopes of an element comprising (i) energizing atoms or molecules containing atoms of the element, isotopes of which are to be separated, with a first circularly-polarized pulsed radiation, the coherent frequency components of the first pulsed radiation having frequencies corresponding to the energy difference between a lower energy level and the frequency split levels of an intermediate energy level, and the duration of a pulse of the first radiation being less than $2\pi/\Delta\omega$ where $\Delta\omega$ is the frequency difference of the split levels;

- (ii) applying a second circularly-polarized pulsed radiation to the atoms or molecules after termination of the first pulse, the two radiations having the same handedness of circular polarization, the coherent frequency components of the second pulsed radiation having frequencies corresponding to the energy difference between the split levels of the intermediate energy level and an upper energy level, and the duration of the pulse of the second pulsed radiation being less than $2\pi/\Delta\omega$; (iii) applying an electric field to the energized atoms or molecules after application of the second radiation to ionize atoms or molecules raised to the upper level; and (iv) separating the ionized atoms or molecules of one isotope from the un-ionized atoms or molecules of the other isotope.
3. The method according to claim 1 or claim 2, wherein the first radiation is produced by a laser and the second radiation is produced by the same laser, and the second radiation is caused to travel over a longer path before reaching the atoms or molecules than the first radiation thereby to produce a time-delayed second radiation.
4. The method according to claim 1, 2 or 3, wherein the first pulsed radiation is produced by one laser, the second pulsed radiation is produced by a second laser, and it is arranged that the second radiation reaches the atoms or molecules after a predetermined delay following arrival of the first radiation.
5. The method according to any of claims 1 to 4, wherein the time at which the second radiation is applied is approximately $\pi/\Delta\omega$ seconds after said first radiation is applied.
6. The method according to any of claims 1 to 5, wherein the duration of the first and second pulses and the time interval between them are those values which result in the largest population of the high energy level.
7. A method according to claim 2 or any claim dependent thereon, wherein the said electric field applied to ionize the elevated isotopic species is employed to separate the elevated isotopic species from the remaining, un-elevated isotopic species.
8. A method of selectively populating high-lying excited states of atoms or molecules, substantially as herein described with reference to the accompanying drawings.
9. A method of separating isotopes of an element, substantially as herein described with reference to the accompanying drawings.
10. Isotopes when separated by the method claimed in claim 2, claim 7 or claim 9, or any of claims 3 to 6 when dependent on claim 2.
11. Apparatus for selectively populating high-lying excited states of atoms or molecules comprising (i) means for energizing the atoms or molecules with a pulse of a first circularly-polarized pulsed radiation, the coherent frequency components of the first pulsed radiation having frequencies corresponding to the energy difference between a lower energy level and the frequency split levels of an intermediate energy level, and the duration of the pulse being less than $2\pi/\Delta\omega$, where $\Delta\omega$ is the frequency difference of the split levels; (ii) means for applying a second circularly-polarized pulsed radiation to the atoms or molecules after the termination of the pulse of first radiation so as to provide an upper energy level with a greater population than existed therein before the atoms or molecules were excited by the first and second radiation pulses, the coherent frequency components of the second pulsed radiation having frequencies corresponding to the energy difference between the split levels of the intermediate energy level and the upper energy level, and the duration of the second pulse being less than $2\pi/\Delta\omega$, the first and second radiations having the same handedness of circular polarization.
12. Apparatus for separating isotopes of an element, comprising (i) means for energizing atoms or molecules containing atoms of the element, isotopes of which are to be separated, with a first circularly-polarized pulsed radiation, the coherent frequency components of the first pulsed radiation having frequencies corresponding to the energy difference between a lower energy level and the frequency split levels of an intermediate energy level, and the duration of the pulse being less than $2\pi/\Delta\omega$ where $\Delta\omega$ is the frequency difference of the split levels; (ii) means for applying a second circularly-polarized pulsed radiation to the atoms or molecules after the termination of the first pulse, the two radiations having the same handedness of circular polarization, the coherent frequency components of the second pulsed radiation having frequencies corresponding to the energy difference between the split levels of the intermediate energy level and an upper energy level, and the duration of the pulse of the second pulsed radiation being less than $2\pi/\Delta\omega$; (iii) means for applying an electric field to the energized atoms or molecules after application of the second radiation to ionize atoms or molecules raised to the upper level; and (iv) means for separating the ionized atoms or molecules of one isotope from the un-ionized atoms or molecules of the other isotope.
13. Apparatus according to claim 11 or claim 12, wherein the first energizing means is a laser and the second energizing means is the same laser, and means is provided to delay the arrival of the second radiation at the atoms or molecules with respect to the arrival of the first radiation.
14. Apparatus according to claim 13, wherein the said delaying means constrains the second radiation to follow a longer path

- to the atoms or molecules than the first radiation.
15. Apparatus according to claim 11 or claim 12, wherein the first pulsed radiation energizing means is one laser and the second pulsed radiation energizing means is a second laser.
16. Apparatus according to any of claims 11 to 15, wherein the time at which the second radiation is applied is approximately $\pi/\Delta\omega$ seconds after the first radiation is applied.
17. Apparatus according to any of claims 11 to 16, wherein the duration of the first and second pulses and the time interval between them are those values which result in the largest population of the high energy level.
18. Apparatus according to any of claims 11 to 17, wherein the energising radiations interact with the atoms or molecules in an interaction zone comprising a gas cell.
19. Apparatus according to claim 12, or any of claims 13 to 18 when dependent on claim 12, in which in an interaction zone wherein the radiations interact with the atoms or molecules, there is a pair of electrode plates between which the ionizing electric field is applied, and the said field is employed to separate the elevated isotopic species from the remaining, un-elevated isotopic species.
20. Apparatus according to claim 12 or any of claims 13 to 19 when dependent on claim 12, wherein means is provided to create the said electric field intermittently in timed relation to the second pulsed radiation.
21. Apparatus for elevating atoms or molecules to high-lying excited states substantially as hereinbefore described with reference to and as shown in the accompanying drawings.

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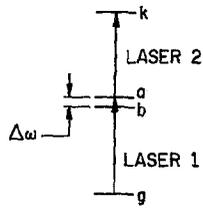


FIG. 1(a)

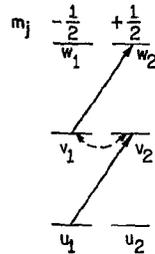


FIG. 1(b)

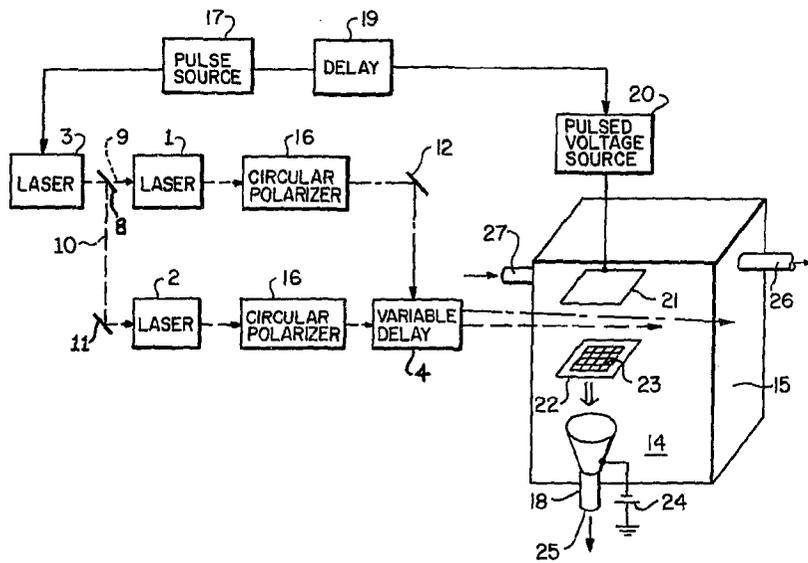


FIG. 2(a)

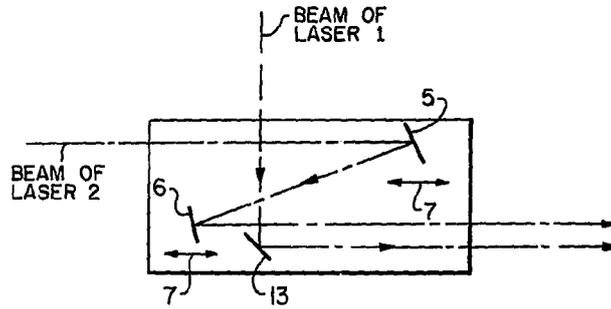


FIG. 2(b)

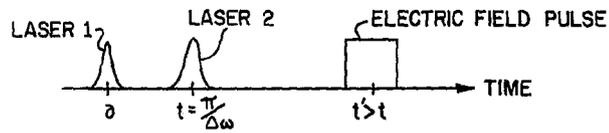


FIG. 2(c)

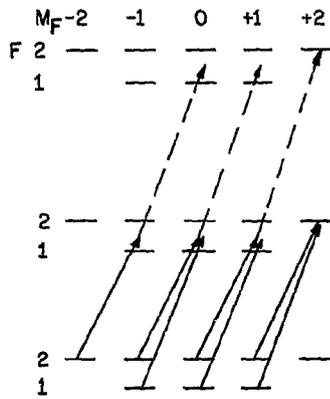


FIG. 3(a)

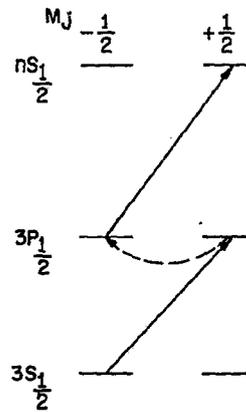


FIG. 3(b)