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Isotopes Separation by Laser Technique: A Review

Abstract

The separation of isotopes is one of the basic needs of science and industry. Various techniques of separation of isotopes have been developed over the years which have their own advantages and disadvantages. One of the most efficient technique is the "Laser Isotope separation" (LIS) technique. This paper reviews the various types of LIS, their advantages and effectiveness. Its wide applications in comparison to other techniques are highlighted.

Keywords: Isotopes; laser excitation; dissociation; ionization

Introduction

The development of tunable lasers have made it possible to excite selectively practically any quantum state of atoms and molecules with excitation energy in the range of 0.1 to 10 eV. With the advent of tunable lasers it is possible to generate coherent radiation of sufficient power in the wavelength range from 200 nm to 2000 nm. This can be used to excite a considerable proportion of atoms and molecules to a selected quantum state. This qualitative growth of quantum electronics has made it possible to start systematic investigations of the selective action of laser radiation on matter. One of the most useful and important achievements has been separation of isotopes using lasers[8].

The first large scale separation of stable and long lived isotope was attempted to produce nuclear weapon materials. This induced the fissile isotope of uranium and heavy hydrogen i.e. deuterium. Uranium isotopes were conventionally separated the gaseous diffusion process[2] and deuterium by electrolysis of water. Both are expensive processes requiring a great deal of electrical power. Both these isotopes are in great demand for use in fission power reactors.

Laser isotope separation has become a subject of considerable interest in past few years largely because of the necessity to provide fuel for nuclear reactors, which are expected to be an increasingly important source of energy. Separation of isotopic species using lasers particularly nitrogen, sulfur and carbon is important for providing non-radiative tracer in agriculture, environmental protection and production control[5].

In principle, separation of isotopes is straight-forward, given the tunability and narrow spectral bandwidths, attainable with presently available lasers. The first step of the process is simply to excite selectively an energy level of an isotopic species in a gas or solid containing the particular species in the normal isotopic abundance. The second step is to trap selectively the excited species or the products of its decomposition[9]. In the present paper a review of the principle of "Laser Isotope Separation" (LIS) and different methods used for isotope separation is presented.

Laser for Isotope Separation

The advent of lasers has opened new avenues in the field of isotope separation. Earlier, the major problem in developing laser isotope separation processes lied in the limitations of availability of tunable lasers. To be useful, laser must operate with a sharp frequency output and either be tunable in the region of interest or have frequencies which match precisely the absorption lines selected. Tunable lasers generally having limited average power have proved very useful in this field.

The essential pre-requisites for a scheme of isotope separation by lasers are:- (1) an absorption spectrum with well-resolved isotopic shift, (2) a laser sufficiently monochromatic and tunable to excite the absorption of one isotope and not the other, (3) a chemical or physical process which acts selectively on excited atoms or molecules and separates them from unexcited ones, but need not have any inherent isotopic selectivity and

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(4) a set of photochemical rate constants and physical conditions such that energy transfer from a laser excited species to an undesired species does not occur before the separation in the step (3)[6].

Principle of Laser Isotope Separation

Isotopes are atoms which have the same number of protons and electrons but differ in the number of neutrons. Since most chemical properties are determined by the electrons surrounding the nucleus, the isotopes of an element behave in almost indistinguishable manners. Thus the method of separating an element from a mixture becomes cumbersome when an isotope is to be separated from a mixture of isotopes of the same element.

Isotope separation using a laser is a fundamentally different technique where one of the slight differences in the energy levels of the isotope due to the difference in nuclear mass. This difference is termed as isotopic shift[4]. Thus light of a certain wavelength may be absorbed by one isotope, while the other isotope of the same element may not be absorbing it. Since light emerging from a laser is extremely monochromatic, one may shine laser light on a mixture of two isotopes and excite the atom of one of the isotopes, thus earmarking it for a subsequent separation.

In addition to the high monochromaticity, the high intensity of the laser is also responsible for its application for laser isotope separation (LIS) because with low intensity beams, the separation rate would be too small for practical pupose[9].

The basic principle behind the LIS process is to first selectively excite the atoms of the isotope by irradiating a stream of the atoms by laser beam and then separate the excited atoms from the mixture by the various existing techniques of isotope separation[9]. A necessary condition is that the isotopic shift must be larger than the actual line width, so the particular isotope can be separated. This means that the material to be separated must be in gaseous form, because the spectral lines in liquids are generally broaden into a smooth band contours. In solids, the isotopic shift can be observed in some crystals. Isotope separation cannot however be achieved, because the separation processes which have to follow the optical excitation, are not applicable[1]. The atoms or the molecules in the gaseous form are randomly oriented and so their spectral band width is broadened. In order to obtain to a sharp and narrow band, a supersonic nozzle is introduced. This nozzle not only forces the atoms or molecules to move in unidirectional beam but also reduces their temperature to 1 to 10 k. at this stage even though the temperature is very low, yet the atoms or molecules remain in gaseous form. The translational temperature of the system is reduced making the absorption lines very narrow so that individual isotopic species could be selectively excited to higher electronic or vibrational states[9].

Methods of Isotope Separation

Isotopes can be separated using laser radiation by following methods (1) Photophysical process (2) Photochemical process (3) Separation by radiation pressure[8,9].
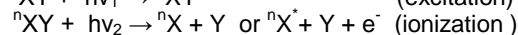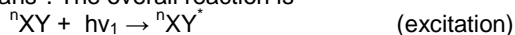
Photophysical Process

Here the separation may in principle be accomplished by laser excitation as two step photoionisation, two step photodissociation and photo-predissociation.

Two step photoionization

Consider two atomic isotopes ^nX and ^mX, a tunable laser can be used to generate a photon pulse at an energy $h\nu_1$ that corresponds to the transition from ground state to the second excited state of ^nX but out of resonance with respect to the same transition in the isotope ^mX. This will selectively pump the ^nX isotope. If the sample is now exposed to a second photon flux at an energy $h\nu_2$, which can cause photoionization from the second excited state of ^nX but is not absorbed by ^mX, then $^nX^+$ ions are produced. These ions may be extracted by the application of an accelerating potential. Alternatively, two photon ionization with the third excited state as the intermediate step and photons of identical energy $h\nu_3$ could be used to produce $^nX^+$ ions[7]. This method can be used to separate ^{235}U by excitation with a dye laser of a resonance transition of neutral uranium atomic beam[8].

Two-step photodissociation

Selective two step photodissociation may be used to reach molecular dissociation continua as well as ionization continua. The selective step $h\nu_1$ may be either a vibrational or on electronic transition. The level excited must be sufficiently high that its thermal population is negligible and that its absorption co-efficient for $h\nu_2$ is substantial, while that of the thermally populated states is negligible[6]. The product of dissociation can be removed by chemical reaction or in the case of ionization, by electric and magnetic means[3]. The overall reaction is

$$^nXY + h\nu_1 \rightarrow {}^nXY^* \qquad \text{(excitation)}$$
$$^nXY + h\nu_2 \rightarrow {}^nX + Y \text{ or } {}^nX^* + Y + e^- \text{ (ionization)}$$

while $h\nu_1$ is tuned such that ^mXY is not affected. Generally $h\nu_1$ and $h\nu_2$ each has to be several electron volts for the photoionization process, since ionization potentials of most of the molecules are usually ≥ 8 eV.

The energy of the first quantum $h\nu_1$ should be absorbed in two steps photodissociation should be much larger than kT (~0.025 eV at room temperature), if the process is to be efficient, where T is the gas vibrational temperature. When $^mXY \leq kT$, the thermal population of the upper level involved in this transition may be appreciable in both ^nXY and ^mXY. In this case, $h\nu_2$ may excite ^mXY as well, since the transition is into continuum. Therefore, $h\nu_1$ optically separates the different isotopes into two groups; those that can absorb $h\nu_1$ and those that cannot. $h\nu_2$ may be absorbed by both isotopes but only produce dissociation in the tagged group[3].

Photo-predissociation

A much simple though less general method of isotope separation is photo-predissociation. If the excited state of a molecule is sufficiently long-lived, may be longer than 10^{-10} sec., it will exhibit sharp, well-resolved absorption spectra. In the simplest case,

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the excited state will decay by spontaneous emission of fluorescence. These radiative life times generally fall within the range of 10^{-8} sec. to 10^{-5} sec. If the molecular excited state is quasi-bound (metastable) with respect to dissociation and if dissociation occurs with a life time between 10^{-10} sec. and the radiative life time, the selective optical excitation can yield isotopically enriched dissociation products. If these products are stable molecules or if they may be chemically trapped, then an isotope separation can be performed. This method is simple and efficient. The process of photo-predissociation has been investigated in great detail for formaldehydes[6].

Photochemical Process

The possibility of photochemical separation of isotopes by excitation of the electronic state of atoms and molecules and of the vibrational levels of molecules has been demonstrated successfully. However, inspite of the considerable optimism of the early investigations the actual progress has been much less remarkable than that achieved by the photophysical method[8].

Electronic photochemistry

Chemical reactions of electronically excited atoms and molecules have been the subject of active research for many years, mainly before the appearance of the first lasers. Several successful photochemical laser separation schemes have been reported recently. The most interesting results were obtained in photochemical separation of the ^{35}Cl and ^{37}Cl isotopes by selective excitation of the $I^{37}Cl$ molecules with radiation of a cw (continuous wave) dye laser, the laser radiation excites only the $I^{37}Cl$ molecule to a state below the predissociation limit. These excited molecules participate in two types 0f reactions. In one case, the $I^{37}Cl$ molecule reacts with trans-ClHC=CHCl, forming cis- ClHC=CHCl, enriched by 10% with the $I^{37}Cl$ isotope. In the other case, they react with 1,2-Dibromoethylene forming trans-ICIC=CHCl, enriched by 50% with the ^{37}Cl isotopes. The highest pressure in this scheme is 7.5 torr in the mixture[8].

A mixture of Cl_2CS molecules are excited with diethxyethylene and achieve enrichment of the ^{37}Cl isotopes. A mass spectroscopic analysis of the residual Cl_2CS molecules obtained after irradiation with an argon ion laser or by dye laser indicated that the concentration of the ^{35}Cl changed from the natural value (75%) to 64% or 80%, depending on the isotopic molecule selected for irradiation[8].

Separation by Radiation Pressure

Isotopes can also be separated by another method by deflecting the free atoms or molecules by radiation pressure. A photon of energy hv carries with it a momentum of hv/c. when this photon is absorbed by an atom, conservation of momentum require that the atom acquire this momentum. Thus the absorption tends to push the atom in the direction of the incident photon. The momentum acquired in a single absorption is very small; hence for the atom to gain sufficient momentum it must absorb many photons. This requires that atoms have a short life time in the excited state. Every time when an atom emits a photon, it acquires a momentum equal and opposite to that it gained during absorption. Since the emissions occur in all random directions, the net effect of many absorptions and emissions is to push the atom along the laser beam. In this scheme a laser beam is allowed to impinge on an atomic beam at right angles which selectively excite a particular isotope. Thus the atoms of the isotope are deflected by the laser beam[9].

Conclusion

From the various methods discussed, those based on pre-dissociation and two-step ionization and dissociation seems to be quite straight forward. Also pre-dissociation is a very elegant method but it is obviously limited to only few special cases. The use of photochemical processes, without dissociation, entails the problem that the chemical reaction must take place within the life time of the excited state. This requires under certain circumstances, high particle densities within the reaction chamber. Such densities in turn, induce various disturbing processes, such as non-radiative transitions to the ground state, which result in thermal heating of the molecular ensemble. As a result, especially in conjunction with the use of vibrational excitation, non-selective population of the excited states is observed. Furthermore, an excitation transfer may take place during collisions, so that the excitation energy is also transferred to isotopes, which are not affected by the selective excitation. It is expected that this important application of laser will considerably promote the development of lasers, especially of frequency-tunable ones.

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