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(i) INFRARED LASER ISOTOPE SEPARATION IN A VAN DER WAALS CLUSTER BEAM

(ii) LASER ISOTOPE SEPARATION BY COMBINING ISOTOPICALLY SELECTIVE CONDENSATION WITH INFRARED VIBRATIONAL PREDISSOCIATION

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INFRARED LASER ISOTOPE SEPARATION IN A VAN DER WAALS CLUSTER BEAM

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Abstract : Efficient laser isotope separation is demonstrated in two distinct schemes. In the first, termed "laser enhanced gas dynamic isotope separation", irradiation of the collisional zone of the gas expansion provokes an isotopically selective condensation, as well as changes in the velocity distributions of the free jet. The resulting large changes in particle mass and/or changes in beam geometry, lead to separation of $^{32}SF_6$, $^{33}SF_6$ and $^{34}SF_6$ in an SF_6/Ar mixture and enrichment factors in excess of 2.0. In similar experiments we have separated bromine isotopes in CF_3Br/Ar mixtures and silicon isotopes in SiF_4/Ar mixtures. In the second scheme, irradiation of the collision free part of the free jet induces single photon infrared vibrational predissociation of van der Waals clusters. Thus at selected wavelengths clusters containing one isotopomer can be made to recoil out of the beam preferentially, resulting in the separation of isotopes. In both schemes a tunable IR laser of only a few watts is used.

I. Laser effects in the collisional part of the free jet.

Mass separation effects in a free jet expansion of a mixture of gases (or isotopomers) are well established [1]. Among these effects are pressure diffusion, centrifugal separation in a curved nozzle, velocity slip, background invasion, Mach-disc and skimmer interference, and Mach number focussing. The efforts to separate isotopes by these effects have mostly been abandoned (the curved nozzle being a notable exception [2]) as the relative mass differences $\left(\frac{\Delta m}{m}\right)$ between isotopomers of interest were generally too small which resulted in enrichment factors too close to unity.

In the first part of this paper we report on large increases in $\frac{\Delta m}{m}$ which are induced in mixtures of isotopomers by selective condensation. This implies that the onset of condensation in a rapidly cooled gas containing several isotopomers is inhibited for one of the isotopomers by "heating" this isotopomer with an IR laser. This may be envisaged as follows in a mixture of SF_6 diluted with Ar, which is the model system chosen. Condensation with a large excess of Ar

$$SF_6 + Ar \xrightleftharpoons{Ar} SF_6\,Ar \qquad (1)$$

$$SF_6\,Ar_{<m-1>} + Ar \xrightleftharpoons{Ar} SF_6\,Ar_{<m>} \qquad (2)$$

which takes places for all isotopomers of $^{i}SF_6$. However when we "heat" for instance the $^{32}SF_6$ isotopomer by irradiating the gas with an IR laser near 10.59 µm, condensation (i.e. the formation of $^{32}SF_6\,Ar_{<m>}$) will be strongly inhibited [3,4], i.e.

$$^{32}SF_6 + h\nu \longrightarrow {}^{32}SF_6^{*} \qquad (3)$$

$$^{32}SF_6^{*} + Ar \xrightleftharpoons{M} {}^{32}SF_6\,Ar^{*} \qquad (4)$$

$$^{32}SF\,Ar^{*} \longrightarrow {}^{32}SF_6 + Ar + \Delta E \qquad (5)$$

The rapid decomposition of $^{32}SF_6\,Ar^{*}$ by reaction (5) results in a strong decrease of $^{32}SF_6\,Ar$, and hence also in a decrease of $^{32}SF_6Ar_{<m>}$ which is formed from $^{32}SF_6\,Ar$. The isotopic selectivity of this process will only be retained if the SF_6 is diluted sufficiently in Ar so as to avoid scrambling of the vibrational excitation between SF_6 isotopomers, by rapid near resonant V-V energy transfer.

The resulting selective condensation is shown in Fig. 1. Fig. 1 a shows the relative intensity of the $^{i}SF_5^{+}Ar$ signal, which is related to the SF_6Ar concen-

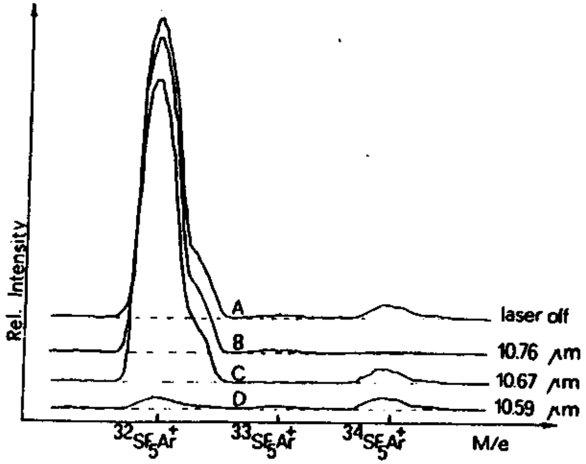

Figure 1 : The isotopically selective condensation 0.5 % SF_6 in Ar. T_0 = 225 K. P_0 = 1.5 bar. Laser power about 20 kW cm^{-2}.
a) no laser, b) irradiation of $^{34}SF_6$ at 10.76 µm,
c) irradiation of $^{33}SF_6$ at 10.67 µm,
d) irradiation of $^{32}SF_6$ at 10.59 µm.

tration. Here, ${}^{i}SF_6$ with i = 32, 33 and 34 are at natural abundance (95.0 : 0.76 : 4.22 %). In fig. 1 d the ${}^{32}SF_6$ monomer is excited at 10.59 µm which results in nearly complete disappearance of the ${}^{32}SF_5^{+}Ar$ peak, i.e. the condensation of ${}^{32}SF_6$ is essentially completely inhibited. The peak due to ${}^{34}SF_6Ar$ however, remains essentially unchanged indicating little or no scrambling.

To avoid sampling errors in our quadrupole mass spectrometer the center part of the molecular beam which passes through the skimmer (the apparatus has been described in detail recently [4]) has been condensed on a zeolite at 77 K and transferred to a cell for IR absorption spectroscopy. The resulting spectrum, which shows the effect of the laser upon the isotopomer ratio, is shown in Fig. 2. Clearly the laser has caused a significant increase in the fraction of ${}^{33}SF_6$ and ${}^{34}SF_6$ near the centroid of the free jet. An enrichment factor α of about 1.7 is found ($\alpha = \{\bar{x}_E/(1-\bar{x}_E)\}/\{\bar{x}_D/(1-\bar{x}_D)\}$ where the subscripts E and D refer to the enriched and depleted fraction of the component of mole fraction $\bar{x}$). This factor increases rapidly with decreasing SF_6 content of the beam [5].

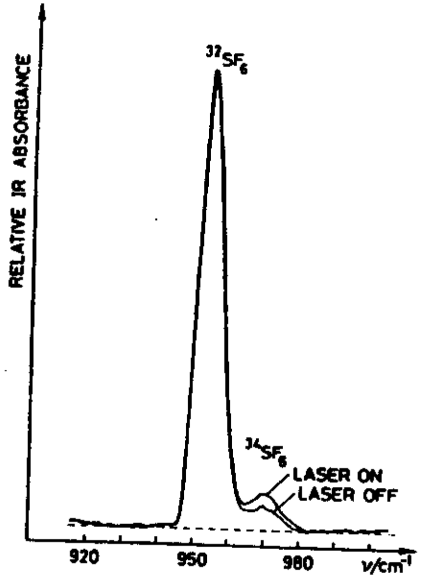

Figure 2 : IR absorption spectra of two samples collected from the molecular beam in a cell, in one case with the laser on, in the other case with the laser off.
0.25 % SF_6 in Ar. T = 240 K, p_0 = 1.3 bar. Laser power about 20 kW cm⁻².

Fig. 3 shows the relative isotope enrichment factor (arbitrary scale) as a function of the wavelength of the IR laser. The large enrichment in ${}^{34}SF_6$ and ${}^{33}SF_6$ over ${}^{32}SF_6$, observed between 942 and 950 cm⁻¹ is due to irradiation of the ${}^{32}SF_6$ monomer. Between 935 and 940 cm⁻¹ the ${}^{33}SF_6$ monomer is irradiated resulting in a relative depletion of ${}^{33}SF_6$. Between 925 and 931 cm⁻¹ the ${}^{34}SF_6$ is irradiated resulting also mainly in its own depletion. Thus choosing the wavelength allows us to either enrich or deplete a particular isotope in the beam.

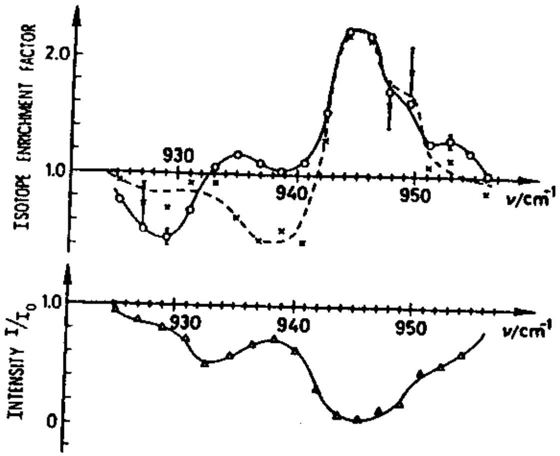

Figure 3 : a) Isotope enrichment factor as a function of the laser wavelength for ${}^{34}SF_6$ (—o—) and for ${}^{33}SF_6$ (--x--). 0.25 % SF_6 in Ar. T_0 = 225 °K, p_0 = 1.6 bar. Max laser power about 20 kW cm⁻².

b) Relative beam attenuation (measured on SF_5^{+} ion mass) due to laser irradiation.

Fig. 3 b shows the relative decrease in total SF_6 concentration caused by the laser at the same wavelength, where the enrichment was measured. This decrease in molecular beam intensity near the beam centroid may be explained as follows : Following the vibrational excitation of SF_6, the excess energy stored in the ν_3 mode relaxes in part by V-T energy transfer. This additional translational energy causes a significant increase in the mean off axis translational energy of the beam. This causes the molecular beam to widen and thus the intensity near the beam centroid drops. If beam widening can be carried out selectively for one isotope, it will also lead to depletion of this isotope near the beam centroid. The relative contributions of selective condensation and selective beam widening to the measured enrichment have not yet been established unambiguously.

Fig. 4 shows the relative isotope enrichment factor as a function of the gas pressure of Ar added in the first chamber. In one possible explanation of the observed phenomena, we assume selective condensation to be an important mechanism in our separation, then the results of Fig. 4 may be explained by a background invasion mechanism in which the heavier ${}^{34}SF_6Ar_{<m>}$ are knocked out of the beam center less easily than the lighter ${}^{32}SF_6$. Thus as the pressure in the first chamber increases, background invasion of the beam increases, and the enrichment in ${}^{34}SF_6$ near the beam centroid also increases. Finally, at pressures above 0.25 mbar, the selectivity decreases again. At these pressures most clusters are destroyed by collisions with Ar, for instance

$$SF_6Ar + Ar \longrightarrow SF_6 + 2\,Ar \qquad (6)$$

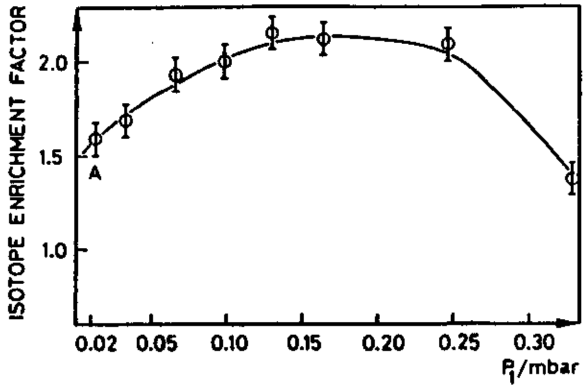

Figure 4 : Effect of the background pressure in the first chamber on the enrichment of $^{34}SF_6$ isotopomer. Conditions like Fig. 1.

so that the condensation becomes effectively non existent, and hence cannot be selective. Consequently any separation of isotopomers due to mass differences induced by selective condensation must also disappear.

II. Laser effects in the collision free part of the free jet.

If a van der Waals molecule absorbs energy from an IR laser beam in the collision free part of the free jet, the resulting vibrational excitation can lead to decomposition of the cluster. In such an IR vibrational predissociation the exothermicity of the reaction will partially end up in relative translational energy of the fragments. These fragments may then recoil out of the beam. The consequent decrease in the intensity of the photodissociated cluster near the beam centroid can be measured by a mass spectrometer set to monitor appropriate mass. If the decrease in measured intensity at this mass is measured as a function of the laser wavelength, we can actually measure the action spectrum of the infrared vibrational predissociation [6]. Such spectra are not always trivial to interpret due to cluster fragmentation in the ion source.

In an imaginary beam containing exclusively SF_6Ar one can imagine that the vibrational predissociation spectra of $^{32}SF_6Ar$, $^{33}SF_6Ar$ and $^{34}SF_6Ar$ are shifted by about 8 cm^{-1}. Thus if the IR vibrational predissociation spectra are not too wide, enrichment in $^{34}SF_6$ and $^{33}SF_6$ may be induced near the center of the beam by selective recoil of the $^{32}SF_6$ and Ar fragments of $^{32}SF_6Ar$. Such IR photon induced recoil is effective only if a significant fraction of the beam is present in the form of van der Waals clusters. This condition generally implies the presence of many different types of clusters (SF_{6_m}, $SF_{6_m}Ar_n$, etc.), so that the action spectrum for isotope separation would be hard to predict. Thus we have simply measured the isotope enrichment of a strongly clustered beam as a function of the wavelength and stagnation conditions in a search for optimal isotope separation.

Fig. 5 shows one result of the enrichment factor obtained in this way for different wavelengths. It is shown here that quite effective isotope separation is possible, and that both enrichment and depletion of certain species can be attained upon variation of the wavelength.

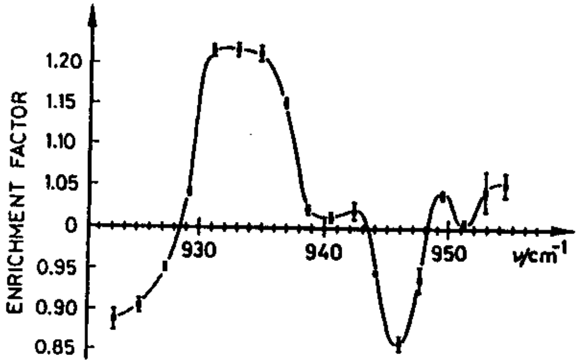

Figure 5 : Laser wavelength dependence of $^{34}SF_6$ isotopomer enrichment due to cluster vibrational predissociation. Laser power max about 20 kW cm^{-2}. 0.5 % SF_6 in Ar. T_0 = 240 °K, p_0 = 1.6 bar.

Acknowledgment : The authors are grateful to P. Melinon for help with some preliminary experiments and to the Swiss Fonds National for financial support.

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Laser Isotope Separation by Combining Isotopically Selective Condensation with Infrared Vibrational Predissociation.

by

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Abstract: The feasibility of a new, two-laser, isotope separation scheme is tested. The first laser induces isotopically selective condensation in the collisional region of a free jet of SF_6 diluted in Ar. Thus a molecular beam containing non-clustered $^{32}SF_6$ and clustered $^{34}SF_{6<m>}Ar_{<n>}$ is produced. The second laser, which acts in a second chamber in the collision free part of the molecular beam, then separates the SF_6 isotopomers in space by infrared vibrational predissociation of the clusters, which is followed by the recoil of the fragments containing $^{34}SF_6$ away from the beam centroid. The magnitude of the fragment recoil is measured, as well as the effect of SF_6 dilution on the vibrational predissociation spectrum in the 9-11 µm region.

Introduction: Effective laser isotope separation in van der Waals cluster beams has recently been demonstrated by two distinct processes [1,2]. The first process is based on the well established aerodynamic mass separation effects in a free jet, which concentrate the heavier species of a gas mixture near the beam centroid [3]. Much of the classical work on such aerodynamic separation of isotopes was abandoned due to the small enrichment factors inherent in the small relative mass differences ΔM/M between isotopomers of interest. In our laboratory we have attempted to improve upon the classical aerodynamic separations, by artificially increasing ΔM/M, using a method called isotopically selective condensation [1]. This selective condensation works as follows: In a beam of SF_6 diluted in Ar, stagnation conditions are chosen at which all SF_6 isotopomers start condensing to form van der Waals clusters $(SF_6)_{<m>}Ar_{<n>}$. A tunable cw IR laser is then used to selectively excite the ν_3 vibrational mode of $^{32}SF_6$ at the nozzle exit, in the collisional region of the free jet expansion. This "hot" isotopomer then does not condense, thus remaining in the form $^{32}SF_6$, whereas the non excited isotopomers continue to condense, and form clusters $(^{33,34}SF_6)_{<m>}Ar_{<n>}$. The resulting large mass differences between the clustered isotopomers and the unclustered $^{32}SF_6$, can lead to a more than two orders of magnitude increase in the measured enrichment factors [1]. Laboratory scale separation of isotopes has been carried out using this method. Samples enriched in ^{33} and $^{34}SF_6$ have been collected near the beam centroid, after passing through a skimmer, and were analysed by IR and MS techniques to confirm their isotopic content. This isotope separation method based on selective condensation was shown to be quite generally applicable. Besides the separation of sulphur isotopes in SF_6 and Si isotopes in SiF_4, Br isotopes in CF_3Br have also been separated.

The second process for isotope separation in a free jet is based on selective infrared vibrational predissociation (IRVP) of van der Waals clusters in the collision free part of the gas expansion [2,4]. Van der Waals clusters can undergo vibrational predissociation following the absorption of IR radiation [5]. In such experiments the cluster fragments will recoil away from the center of the molecular beam. Furthermore, clusters containing different isotopomers, such as for instance $^{32}SF_6Ar$ and $^{34}SF_6Ar$ will exhibit isotope shifts in their IRVP spectra, and may thus be photodissociated selectively. Such a selective IRVP, followed by the recoil of one of the isotopomers away from the beam centroid, leads to a separation of isotopes in space. In this way a tunable cw CO_2 laser has been used for isotope enrichment by IRVP in a cluster beam of SF_6 and Ar, and we have reported overall enrichment factors in excess of 1.1.

In reference 2 we proposed a third process for separating isotopes which combines processes 1 and 2. Fig. 1 demonstrates the principle. A first laser induces isotopically selective condensation, which causes some degree of isotope enrichment in the beam passing through the skimmer. This enrichment however is not the main purpose of the isotopically selective condensation. The important point is to create a molecular beam in which one or more of the isotopomers (here $^{34}SF_6$) are present essentially only in the form of van der Waals clusters $^{34}SF_6Ar_{<m>}$, whereas the other isotopomer ($^{32}SF_6$) is present mainly in unclustered form. IR excitation of the clustered isotopomers will cause their IRVP followed by the recoil of the $^{34}SF_6$ fragments away from the beam centroid. The center of the beam is thus diluted in $^{34}SF_6$. The unclustered $^{32}SF_6$, even if it would absorb IR radiation will hardly recoil at all, and thus stay in the molecular beam. The two isotopomers may then be separated in space by a collimator, as shown in Fig. 1. Two important points should be made: First, the excitation by the second laser need not be narrow band in order to be selective. Even if all species $^{32}SF_6$ and $^{34}SF_6Ar_{<m>}$ absorb IR radiation, isotopically selective recoil of $^{34}SF_6$ out of the beam center will follow. Second, the separation efficiency depends on the fraction of the excess energy ($E_{h\nu}$-BDE) going into relative translational energy of the fragments. As method 2 described above has been shown to work [2], and as it depends also on sufficient recoil of the cluster fragments, it was thought that the combination of methods 1 and 2 might be worth trying. Below we report on several experiments in which this two-laser method is tested.

Results and discussion: The experimental setup has been described in detail elsewhere [1,2,6]. Fig. 2 shows the effect of combining selective condensation (laser 1) with IRVP (laser 2). The full curve shows an "IRVP enrichment spectrum" using only the second laser. The relative enrichment or depletion in $^{34}SF_6$ is measured near the beam center. The enrichment near 950 cm^{-1} and 933 cm^{-1} is due to the recoil of fragments containing $^{32}SF_6$ following the IRVP of $(^{32}SF_6)_2Ar_{<m>}$ [7]. The enrichment in $^{34}SF_6$ near 942 cm^{-1} is due to the IRVP of $^{32}SF_6Ar_{<m>}$. The relative depletion in $^{34}SF_6$ near 946 cm^{-1} and 927 cm^{-1} is due to the IRVP of respectively $^{32}SF_6{}^{34}SF_6Ar_{<m>}$ and $^{34}SF_6Ar_{<m>}$ [7]. The dotted curve shows the same IRVP enrichment spectrum, however now with the first CO_2 laser at the 10P16 line inhibiting condensation of $^{32}SF_6$. One expects all clusters containing $^{32}SF_6$ to decrease in abundance, as is actually observed. This decrease appears to be stronger for the clusters containing two $^{32}SF_6$ molecules than for the clusters containing only one $^{32}SF_6$. The concentration of $^{34}SF_6Ar_{<m>}$ is virtually unchanged. Thus irradiation with laser 1 at 946 cm^{-1} followed by IRVP in the region between 923 and 928 cm^{-1} will result in a separation of isotopes such as described in Fig. 1.

The efficiency of the 2-laser method depends on the amount of fragment recoil in IRVP. That such recoil can cause effective separation has already been demonstrated in the enrichment experiments of reference 2. We have measured this recoil directly in an experimental setup similar to that of Fig. 1. Two consecutive skimmers are used rather than the one indicated, to obtain an improved beam collimation prior to the laser excitation (only laser 2). The collimator of Fig. 1 in front of the MS is removed, and the QMS can be moved up and down accross the molecular beam to monitor its profile. This beam profile is measured once with the No. 2 laser "on" at the 10P34 line, and once with the laser "off". At this wavelength (931 cm^{-1}) we dissociate mainly $(^{32}SF_6)_2Ar_{<m>}$. The difference between the measurements with the laser on and off is shown in Fig. 3. Clearly the center of the beam (laboratory angle 0 degrees) is depleted in SF_6 due to the IRVP, whereas the recoiling fragments enhance the SF_6 concentration away from the beam center. A rough deconvolution (lab. to c.m.) of these measurements indicates that of the order of 1% of the available excess energy goes into translation (recoil of the fragments). This order of magnitude did not appear to vary strongly with the types of clusters $SF_{6<m>}Ar_{<n>}$ dissociated. In these measurements the distance between the 2 mm diameter QMS ioniser entrance, and the point at which the laser intersects the molecular beam, is 20 cm.

Fig. 4 shows the effect of SF_6 dilution on the IRVP enrichment spectra. The full line is for 0.15% of SF_6 in Ar, the dotted line for 0.3% SF_6 in Ar. As expected the dilution in SF_6 causes a relative decrease in $(SF_6)_2Ar_{<m>}$ as can be seen near 933, 946 and 950 cm^{-1}. Simultaneously one observes a relative increase in $SF_6Ar_{<n>}$ as shown by the increases in IRVP efficiency near 926 and 942 cm^{-1}.

Conclusions: The feasibility of a new 2-laser isotope separation method is demonstrated. Isotopically selective condensation is effectively combined with IR vibrational predissociation (IRVP). The fraction of the available excess energy in IRVP which goes into recoil of the fragments is only of the order of 1% for those of the clusters $(SF_6)_{<m>}Ar_{<n>}$ tested. The latter measurements may help to shed some light on the mechanism of IRVP.

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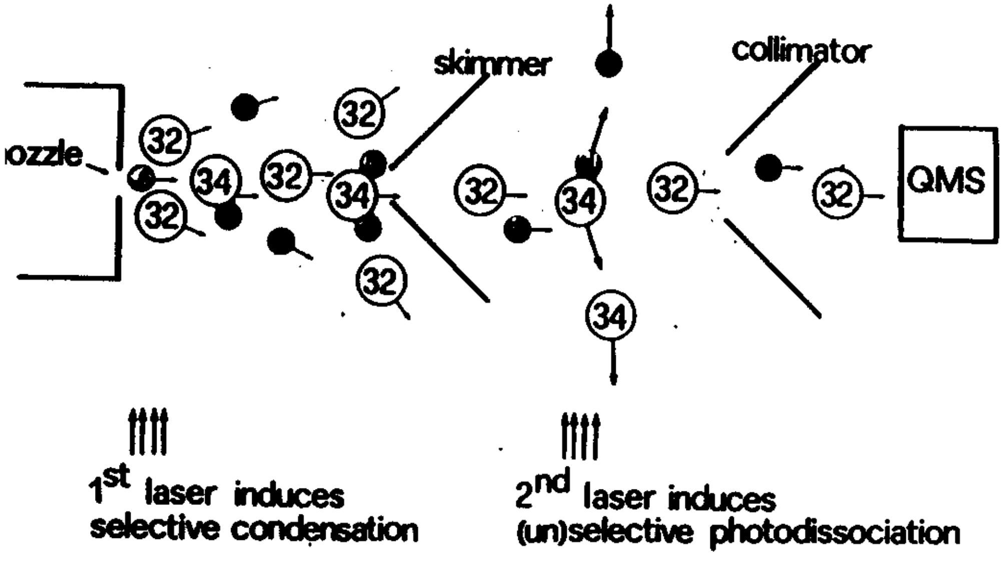
Figure captions:

Fig. 1: A schematic representation of the 2-laser isotope separation method. The first IR laser selectively excites $^{32}SF_6$ in the collisional part of the free jet, thus inhibiting the formation of clusters containing $^{32}SF_6$. The $^{34}SF_6$ condenses to $^{34}SF_6Ar_m$ clusters. A second laser then removes the $^{34}SF_6$ from the center of the molecular beam by IR vibrational predissociation followed by recoil of the fragments. The latter takes place in the collision free part of the gas expansion. ㉜ represents a $^{32}SF_6$ molecule, ●㉞● represents a $^{34}SF_6Ar_2$ cluster, and ● is an Ar atom. QMS is a quadrupole mass spectrometer.

Fig. 2: The overall enrichment factor α [1)] for enrichment in $^{34}SF_6$ by IR vibrational predissociation. The results represented by the solid line are taken only with laser 2, the dotted line gives results obtained with laser 2 together with laser 1, the latter inhibiting condensation of $^{32}SF_6$. Laser 1 with 10 W at the 10P16 line. Laser 2 with about 15 W. 0.15% SF_6 in Ar. P_o=1.3 bar, T_o=235 K.

Fig. 3: The changes in SF_6 beam profile due to infrared vibrational predissociation and the consecutive fragment recoil. The laser power is 15 W at the 10P34 line where mainly $(^{32}SF_6)_2Ar_{<m>}$ clusters are dissociated. T_o=235 K, P_o=1.3 bar. 0.15% SF_6 in Ar. The recoil of the IRVP fragments causes a decrease in SF_6 intensity near the beam centroid (0^o lab. angle) and increases the SF_6 intensity away from the beam centroid ($\theta_{lab.} > 1.5^o$).

Fig. 4: The effect of dilution of SF_6 in Ar on the IRVP enrichment spectrum of $^{34}SF_6$. The full line is with 0.15% SF_6 in Ar, the dotted line with 0.3% SF_6 in Ar. T_o=235 K, P_o=1.3 bar. The laser power is about 15 W, applied 4 mm downstream from the nozzle. Increased dilution decreases the relative concentration of clusters containing SF_6 dimers and increases the relative concentration of clusters containing one SF_6 molecule.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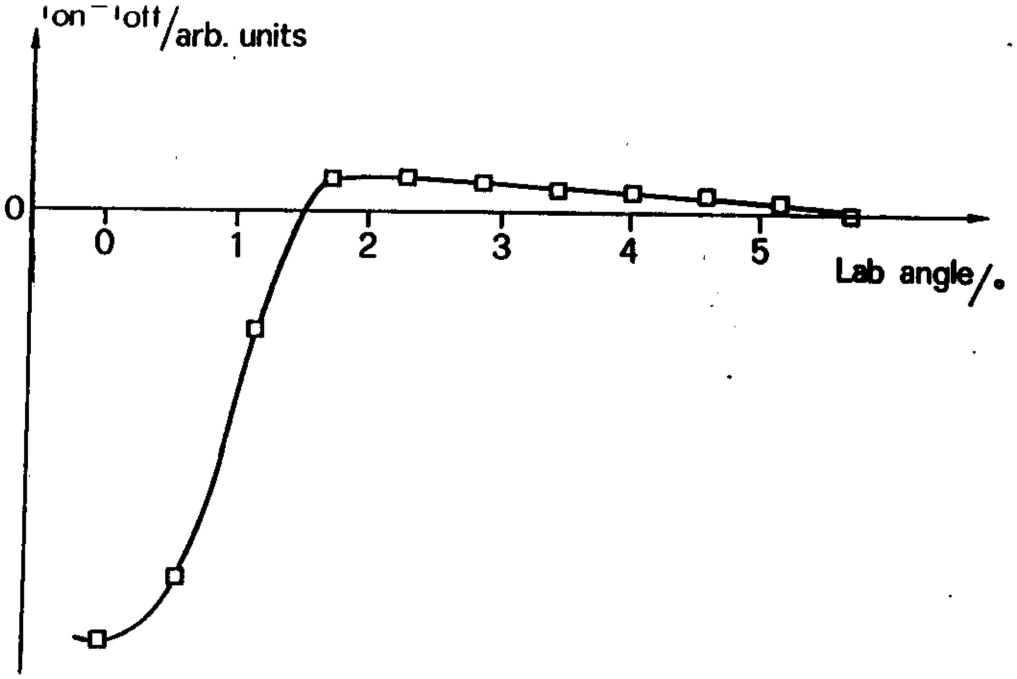
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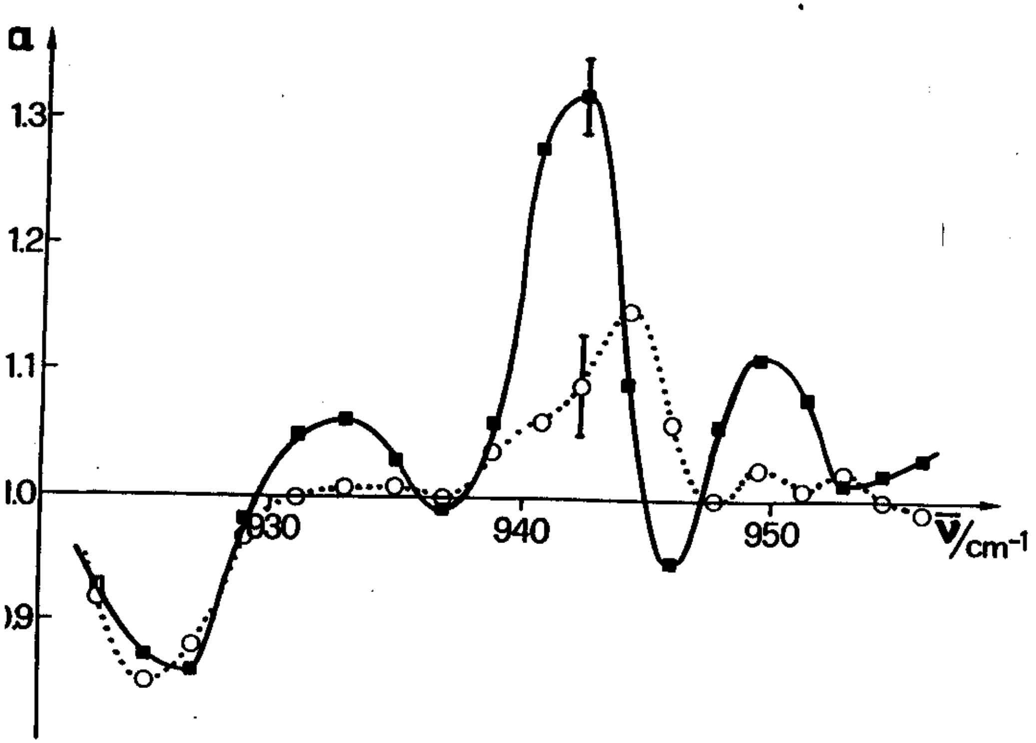
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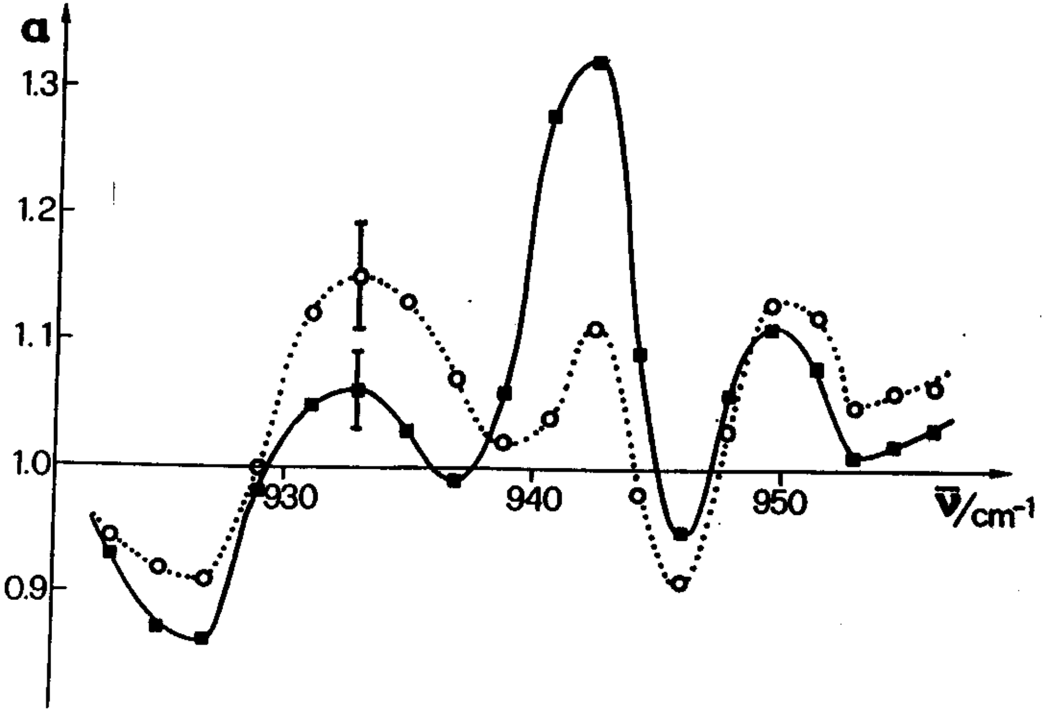
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