PHOTO-INDUCED CATAPHORETIC ISOTOPE SEPARATION

MASTER

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Abstract

This report is submitted in support of a one-year continuation proposal. A more comprehensive report to include the work of the other principal investigators will be submitted in September 1979.

That part of the studies on radiation-induced cataphoretic isotope separation which has been undertaken by this principal investigator has been associated with the use of a 0.63μ He:Ne laser as the radiation source in an attempt to obtain separation of Ne^{20} and Ne^{22} isotopes. A cataphoretic discharge tube containing mixed neon isotopes is within the laser resonator, whereas the laser tube contains only one neon isotope.

Cataphoretic separation of the neon isotopes has not been observed, although selective excitation of one isotope has been demonstrated and a very sensitive technique for detecting cataphoretic isotope separation has been developed. Also, it is found that the exchange rate between the two neon isotopes in the excited laser levels is not great enough to significantly reduce the effect of selective excitation. Measurements of the current changes induced by the radiation indicate that the populations of those levels with which the laser radiation interacts is too small. It is therefore planned to undertake double-pumping from the metastable $1s$ levels, employing neon isotope radiation lamps to pump from the $1s$ to the $2p$ levels, and the laser from the $2p_4$ to the $3s_2$ level. These experiments appear preferable at this stage to the much more elaborate and expensive experiments involving double pumping with tunable dye lasers.

1. Introduction

A study of radiation-induced cataphoretic separation of isotopes in a gaseous discharge was started in June 1976 under Contract No. E(11-1)2961. Progress reports have been submitted in February 1977, and February 1978 as report numbers COO-2961-1, and COO-2961-2, by J. A. Carruthers, L. M. Chanin, and H. J. Oskam. This report is submitted by J. A. Carruthers covering the last year's effort, and is submitted at this time in support of a one-year continuation proposal. A more comprehensive report which includes the work of the other two principal investigators will be submitted in September, 1979.

The basic concept of radiation-induced cataphoretic isotope separation is based on the preferential excitation of one isotope by the radiation with the result that one isotope is more readily ionized, and relatively more of its ions should move toward the cathode in the dc discharge. In the work discussed here the radiation source is a He-Ne 0.63 μ laser, containing only the isotope Ne²⁰. A cataphoresis discharge tube containing He³, Ne²⁰, and Ne²² is also within the laser resonator and is subjected to the laser radiation. Preferential depletion of the upper (3s₂) level should occur for the Ne²⁰ isotope, and hence preferential ionization of the Ne²² isotope should be observed, hopefully leading to cataphoretic separation of the Ne²⁰ and Ne²² isotopes in the dc discharge.

The isotope shift in Ne²⁰ and Ne²² is about 875 MHz, with Ne²² at the higher frequency. The half-power full width of the Doppler-broadened line is about 1400 MHz. Hence the isotope shift is hardly enough for preferential pumping of one isotope,

and we have had to make use of a moving-mirror technique for the laser in order to obtain an additional shift in the laser radiation. With this technique preferential excitation of the one isotope has been demonstrated, an important result.

The 0.63μ Ne laser line ($3s_2 - 2p_4$) has the upper $3s_2$ level at about 0.9 eV below the ionization level, whereas the lower $2p_4$ level is an additional 2 eV lower (see Fig. 1). One would therefore expect the ionization probability to be higher for atoms in the upper $3s_2$ state. If the population in the cataphoresis tube is inverted, with that of the $3s_2$ level greater than that of the $2p_4$, the laser radiation due to the He-Ne²⁰ laser should reduce the population of the upper Ne²⁰ level, leaving that of the Ne²² isotope relatively unchanged. The current in the cataphoresis tube should decrease when the laser radiation is present due to a decreased ionization rate for the Ne²⁰ isotope. Observations would appear to support this expectation, but the change in current due to the laser radiation is small, generally less than one percent, depending on discharge conditions, and even the detailed mechanisms leading to the current change are not entirely clear. A number of experiments have been undertaken in the hope of finding the conditions for maximum current change, as well as to help clarify the mechanisms involved. We have not been able to observe actual cataphoretic separation of the Ne²⁰ and Ne²² isotopes. Thus, although some additional experiments along this line are of interest, it appears essential to get a higher proportion of the excited Ne atoms in the higher energy levels, such as the $3s_2$ level, in order to have sufficient effect on the current and on the ionization probability due to

FIGURE 1. Helium and neon energy levels.

the laser radiation. It is because we believe that it may be possible to accomplish this by a relatively simple technique that a continuation proposal is being submitted.

In brief, it is believed essential to carry out experiments based on a double radiation-pumping approach, using Ne isotopic discharge lamps to pump from the 1s to the 2p levels, and a 0.63 μ laser to pump from the 2p₄ to 3s₂ level. Our extremely high sensitivity for detecting changes in the Ne²⁰:Ne²² ratio may be sufficient to indicate whether double-pumping techniques are going to work before investing the effort and cost for a double tunable dye-laser system. The confocal scanning Fabry-Perot spectrometer used to observe changes in the Ne²⁰:Ne²² ratio is capable of detecting well under 1% change in the isotope ratio.

2. Neon Laser Studies

The basic apparatus for studying laser-induced cataphoretic isotope separation at 0.63μ has been described in the previous progress reports, (COO-2961-1, COO-2961-2). The apparatus as shown in Fig. 2 has been changed only to allow measurements of the laser-induced current change in the cataphoresis tube. A beam chopper is placed between the two tubes to periodically interrupt the laser, and the resulting periodic current changes in the cataphoresis tube are monitored on an oscilloscope. The part of the apparatus which warrants further discussion (section 2.1) is the scanning Fabry-Perot spectrometer, which, because of its special characteristic, permits detecting changes in the $\text{Ne}^{20}:\text{Ne}^{22}$ isotope ratio of much less than 1%, and this is important to a continuation proposal.

Work continued on a systematic search through a range of He-Ne mixtures for detectable changes in the $\text{Ne}^{20}:\text{Ne}^{22}$ ratio at one end of the cataphoresis tube. No cataphoretic separation was observed. Effort was then concentrated on measurements of the so-called opto-galvanic effect, or changes in discharge current (or voltage) in the cataphoresis tube induced by the laser radiation. These experiments were undertaken in the hope of learning more about the detailed mechanisms by which the laser radiation affects the neon ionization rate. The experiments and results are discussed in sections 2.2 and 2.3.

The small magnitude of the opto-galvanic effect for the 0.63μ laser radiation is discouraging, and it is further noted that the effect decreases with increasing current. Thus, for the high currents and the high He:Ne ratios generally desirable for

Neon Isotope Separation Schematic
Figure 2

cataphoretic separation¹, the effect of the laser radiation on the neon ionization rate is small. Therefore, for the primary objective of demonstrating laser-induced cataphoretic isotope separation, it is essential to get a higher fraction of the excited atoms to occupy the higher energy levels, instead of the metastable $1s$ levels, for example.

Instead of initiating studies on a variety of neon transitions by setting up a tunable dye laser, it appears that the next set of experiments to be undertaken should combine isotope radiation lamps and a 0.63μ laser for double radiation pumping from the metastable $1s$ levels using comparatively simple apparatus. Considerable experience has been obtained under the contract on both pumping techniques and the next step would be to combine them. If the $3s_2$ population can be increased by a significant factor using this technique, the high detection sensitivity available in our system may allow us to observe cataphoretic isotope separation. Only if some indication of isotope separation is observed, or some other new information comes to light, would the use of tunable dye lasers be justified in a double pumping scheme.

One important result, discussed further in sections 2.2 and 2.3, is that it has been shown that the laser radiation can be shifted enough by means of the moving-mirror technique to selectively excite the one neon isotope. Further, it has been found that excitation exchange between the isotopes in the $3s_2$ level is not occurring in significant amounts. This result is encouraging.

2.1 Scanning Fabry-Perot Spectrometer

There are two similar versions of the instrument available, one with 3.75 cm spacing and a free spectral range of 2.0 GHz, the other with spacing of 2.0 cm, and free spectral range of 3.75 GHz. These instruments are used to make measurements on the fluorescent lineshape and when two neon isotopes are present, to detect small changes in the relative amounts of the isotopes.

Because the free spectral range of the instruments are comparable with the linewidth when two isotopes are present, the lineshape is not properly reproduced, even though the linewidth of the spectrometer is much less than that of the fluorescent line. But the sensitivity for observing small changes in the ratios of the two isotopes is thereby improved. This somewhat fortuitous result has been important to the laser-induced isotope separation studies because only small changes in the neon isotope ratios can be expected to result from the laser pumping. Double-pumping combined with this high detection sensitivity should offer an improved prospect of observing cataphoretic isotope separation.

Figures 3(a) and 3(b) are reproduced from a more detailed account of calibration tests of the sensitivity of one of the scanning Fabry-Perot spectrometers, when used to monitor the $\text{Ne}^{20}:\text{Ne}^{22}$ isotope ratio⁷. Even when the fluorescence sampling tube is cooled to reduce the Doppler linewidth the two isotopes are barely resolved. The very shallow minimum is extremely sensitive to small changes in the isotope ratio. Figure 3(b) is a plot of the slope of the curve of Fig. 3(a), obtained using synchronous detection etc. The two central peaks, the one negative and the other positive, are carefully measured when known

(a) Spectrum for $\text{Ne}^{20}:\text{Ne}^{22} = 1$.

(b) Slope of the spectrum curve.

FIGURE 3. Typical lineshape data from the Fabry-Perot spectrometer. (a) The partially resolved 0.63μ line for equal Ne^{20} and Ne^{22} mixture. (b) The slope of the spectrum curve.

quantities of one isotope are added. For example, in a 10:1:1 mixture of $\text{He}^3:\text{Ne}^{20}:\text{Ne}^{22}$ the ratio of the amplitudes of these peaks changed by 11% for each 1% change in the $\text{Ne}^{20}:\text{Ne}^{22}$ ratio. Under the best conditions 1/10% change in the $\text{Ne}^{20}:\text{Ne}^{22}$ ratio is detectable. Traces of the type shown in Figs. 3(a) and 3(b) have been obtained routinely under a variety of conditions of pressure, He:Ne ratio, and current, in the cataphoresis tube, but cataphoretic separation of the neon isotopes has not been observed.

2.2 The Opto-Galvanic Effect

When the attempts to observe cataphoretic separation of the neon isotopes induced by the 0.63μ laser radiation produced negative results, it was decided to examine the laser-induced current (or voltage) changes in the cataphoresis tube²⁻⁶, with the hope of explaining our negative result, or of improving our prospect for obtaining measurable isotope separation.

In Fig. 4 some typical i-v characteristic curves and measured values of the opto-galvanic effect for $\text{He}:\text{Ne}^{20}$ in the cataphoresis tube, with the laser tube also containing $\text{He}:\text{Ne}^{20}$, are shown. The changes in current, Δi , induced by the laser radiation are given as a function of the discharge current and discharge voltage for two gas mixtures. The resistance connecting the discharge tube to the variable regulated voltage supply is 61 k. The values of Δi can readily be converted to change in voltage, Δv , by multiplying by 61 k, but the increments are of opposite signs since an increase in current corresponds to a decrease in voltage across the discharge. Because the laser radiation has a significant effect on the shape of the electron-energy distribution curve³, it is not clear what represents the

FIGURE 4. Discharge parameters for the cataphoresis tube for two He:Ne ratios, 10:1 and 27:1. The opto-galvanic current changes, Δi , are shown. The variation in laser power is also recorded.

best way to report the data. We prefer to report the raw data, noting that in general an increase in current (or decrease in voltage) indicates an increase in the ionization rate.

For an inverted population, with the population of the upper $3s_2$ level greater than that of the lower $2p_4$ level, the laser radiation induces a negative value of Δi , a decrease in discharge current. The effect is reduced for larger values of the dc discharge current. The result would be explainable in terms of a decreased ionization rate for neon when the population of the upper level is reduced by the radiation, but the reduction in the effect as the discharge current is increased indicates that the mechanism is more complicated, and that the $1s$ and $2p$ populations are also significantly involved³. It has even been suggested that the opto-galvanic effect may be related to the population of the helium metastable levels⁴. Our data is not inconsistent with the suggestion by Garscadden et al. that at low currents the contribution to Δi due to the $3s_2$ level dominates, whereas at higher current the contributions from the $2p$ and $1s$ levels, which are of opposite sign, are increasingly important. Further detailed experiments with a tunable dye laser would be needed to more completely describe the detailed mechanisms, but it is believed that these experiments are not the more important ones in respect to the basic goal, radiation-induced cataphoretic isotope separation.

It is seen from Fig. 4 that not only does the opto-galvanic effect for 0.63μ laser radiation decrease at high currents, but also decreases as the He-Ne ratio is increased. But cataphoretic separation is generally favored by large currents and a high

proportion of the buffer gas¹. The problem seems to be related to the relatively low population of the high-lying $3s_2$ level compared with that for the metastable $1s$ population.

In Fig. 4 it is seen that the 0.63μ laser power also varies with the dc discharge current in the cataphoresis tube. This power change is due to the additional gain provided by the cataphoresis tube. The laser tube current is held constant, but the added gain by the cataphoresis tube depends on its current.

From the data of Fig. 5 it is seen that the opto-galvanic current changes, Δi , are proportional to laser power. If the Δi values of Fig. 4 are recalculated for a constant magnitude of laser power, these values would show a much greater reduction as the dc discharge current is increased. The linear relationship of Fig. 5, however, shows that we may need to have higher laser power capability since we are apparently not saturating the transition. On the other hand, if we increase the power by too large a factor, we will encounter more difficulty in being able to selectively excite the one isotope (section 2.3). That is, as the laser power supplied mainly by the He:Ne²⁰ laser tube is increased so as to saturate the Ne²⁰ in the cataphoresis tube, the Ne²⁰ population in the $3s_2$ level is decreased, and with further increase in laser power there is unwanted saturation of the Ne²² population. However, we anticipate wanting to increase the laser power, and a folded-cavity arrangement using a longer laser tube has been tested, as reported in the 1978 progress report C00-2961-2.

2.3 Selective Isotope Excitation

As first described in the proposal in 1976 the use of a moving-mirror laser resonator has been an integral part of the

FIGURE 5. Opto-galvanic effect current versus laser power in He:Ne mixture of 28:1 at 2.7 Torr.

apparatus for cataphoresis studies in neon using 0.63 μ neon laser line as the radiation source. Because of the rather broad Doppler linewidth of 1400 MHz, it had been anticipated that the 875 MHz isotope shift between Ne^{20} and Ne^{22} might not be enough to insure that the laser radiation would selectively excite the one isotope. The measurements on the opto-galvanic effect in the cataphoresis tube have served to illustrate that the moving-mirror arrangement is in fact essential in order to obtain selective excitation of one isotope.

Results obtained here several years ago showed that when one mirror of a laser resonator is moved continuously at a velocity of several cm/sec the multi-mode laser output shifts to one side of the Doppler linewidth, to high frequencies when the laser resonator is being shortened and to lower frequencies when the laser is being lengthened⁸. In the apparatus for cataphoretic studies, shown in Fig. 2, the laser tube contains $\text{He}^3\text{:Ne}^{20}$, while the cataphoresis tube usually contains He^3 and equal amounts of Ne^{20} and Ne^{22} . The cataphoresis tube provides some gain, and hence the laser modes are shifted slightly to high frequencies because of the Ne^{22} that is present. Because the isotope separation is less than the Doppler linewidth of either isotope, and because of the image-mode nature of the interaction of the radiation in a laser resonator, the result is there is not much difference in the excitation of the two isotopes unless the moving-mirror technique is employed. It is found that a simple but effective technique can be developed with practice where the mirror is moved out for about 10 cm by hand, is quickly moved back while tilting out of alignment, and then moved outward again, etc. A motor-driven

system can probably be developed, but such attempts to date have not been effective.

The data of Fig. 4 is for the opto-galvanic effect in the cataphoresis tube when Ne^{20} is in both the laser tube and the cataphoresis tube. When the neon in the cataphoresis tube is changed to equal proportions of Ne^{20} and Ne^{22} at the same total pressure, the opto-galvanic effect is reduced, but only by about 15% if the laser mirrors are stationary. The opto-galvanic effect should be reduced by a factor of two if only the Ne^{20} isotope were interacting with the laser radiation.

The effect of moving the laser mirror to shift the laser radiation to lower frequencies, so as to reduce the interaction with the Ne^{22} isotope, is illustrated in Fig. 6. These are plots of the changing magnitude of the opto-galvanic effect as the laser mirror is moved outward to shift the laser line. Even with practice it is not possible to move the mirror at constant velocity, so the changes are not seen as rectangular waveforms. In Fig. 6(a) there is a plot of the change in opto-galvanic effect for Ne^{20} and Ne^{22} in the cataphoresis tube in equal proportion. The opto-galvanic effect is reduced by about 25% due to the frequency shift. In Fig. 6(b) there is a plot of the change in opto-galvanic effect when the single isotope of neon, Ne^{22} , is in the cataphoresis effect. The opto-galvanic effect is reduced to about 1/3, although not eliminated when the frequency is shifted.

These results are consistent with the following relationships. With no frequency shift the laser radiation interacts with the Ne^{20} and Ne^{22} in the proportion of 0.5 to 0.35, respectively. When the frequency is shifted by moving the mirror, the laser

(a) Ne^{20} and Ne^{22} in equal mixture.

(b) Ne^{22} only.

FIGURE 6. Change in opto-galvanic effect, ΔI , as mirror is being moved to shift laser to lower frequency.
(a) $\text{Ne}^{20}:\text{Ne}^{22} = 1$. (b) Ne^{22} only.

radiation interacts with Ne^{20} and Ne^{22} in the proportion of 0.5 to 0.12, respectively. It is seen that it is essential to make use of the moving-mirror techniques, and to try to improve it so as to obtain even better selectivity in exciting the one isotope.

These results are important not only in showing that the moving-mirror technique is necessary to improve the selectivity of the excitation process, but also show that excitation exchange rate between the two isotopes is not so rapid as to seriously alter the excited populations of the two species.

3. References

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