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PHOTO-INDUCED CATAPHORETIC ISOTOPE SEPARATION

Final Report

for Period June 15, 1976 - June 15, 1981

MASTER

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Abstract

This final report describes studies performed under contract No. E(11-1)2961 with the Department of Energy during the period of June 15, 1976 to February 28, 1981. The studies were performed by students Robert Nelson, Bruce Land and Jim Herrmann with supervision by Dr. J. A. Carruthers.

The original studies were undertaken to study the feasibility of radiation-induced cataphoretic separation, by J. A. Carruthers, L. M. Chanin and H. J. Oskam. This part of the work is concerned with laser-induced cataphoretic separation in neon using a He-Ne 6328A° laser. The basic concept of radiation-induced cataphoretic isotope separation is based on the preferential excitation of one isotope with the result that one isotope is more readily ionized, and relatively more of its ions move toward the cathode in the dc discharge. For the later part of the work a second radiation source was added, a helical Ne^{20} radiation lamp.

Radiation-induced cataphoretic isotope separtion has not been observed. Selective excitation has been achieved by both the $He\text{-}Ne^{20}$ 6328A° laser and the Ne^{20} helical radiation lamp in spite of the fact that the isotope shift is comparable with Doppler-broadened linewidths. Collisional excitation exchange between the Ne^{20} and Ne^{22} atoms does not appear to be a problem for the neon partial pressure range involved.

The population of the $3s_2$ and $2p_4$ laser levels (6328A°) are apparently too low to offer reasonable expectation of inducing

observable cataphoretic isotope separation by means of the 6328A° laser radiation, even with the high detection sensitivity of the scanning Fabry-Perot spectrometer system. The use of the additional radiation source in the form of a helical Ne^{20} radiation lamp has not improved the effectiveness of the laser 6328A° sufficiently to justify further experiments with the 6328A° laser. It has become clear from these experiments, however, that for isotope separation in neon it is well to concentrate on using radiation sources that interact mainly with the 1s population.

Experiments which appear justifiable at this stage require a turnable dye laser, but it appears that a highly stable narrow-linewidth laser is not required in the initial experiments. For this reason an experiment is being considered using a relatively simple argon-pumped dye laser which is available, so that outside support is unnecessary. If this experiment gives very encouraging results, as discussed in Section 4, further experiments with highly stable ion lasers for selective isotope pumping could be considered.

I. 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 reports COO-2961-1 and COO-2961-2 by J. A. Carruthers, L. M. Chanin, and H. J. Oskam. Other reports were COO-2961-3 by J. A. Carruthers in June 1979, and a final report by L. M. Chanin and H. J. Oskam in January 1980 as COO-2961-4. This is a final report by J. A. Carruthers.

The basic concept of radiation-induced cataphoretic isotope separation is based on the preferential excitation of one isotope by 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.(1) For most of the work discussed here the radiation source is a He-Ne 6328A° laser, containing only the isotope Ne^{20}. A cataphoresis discharge tube containing He^{3}, Ne^{20}, and Ne^{22} is also within the laser resonator and is subjected to the laser radiation. Also, for much of the work during the final year a second radiation source was added in the form of a spiral Ne^{20} monoisotopic fluorescent lamp mounted around the cataphoresis tube, as discussed in section 2.5. Because of the interaction with the laser radiation, preferential depletion of the upper ($3s_2$) level should occur for the Ne^{20} isotope, and hence preferential ionization of the Ne^{22} isotope should be observed, hopefully leading to cataphoretic separation of the Ne^{20} and Ne^{22} isotopes in the dc discharge.

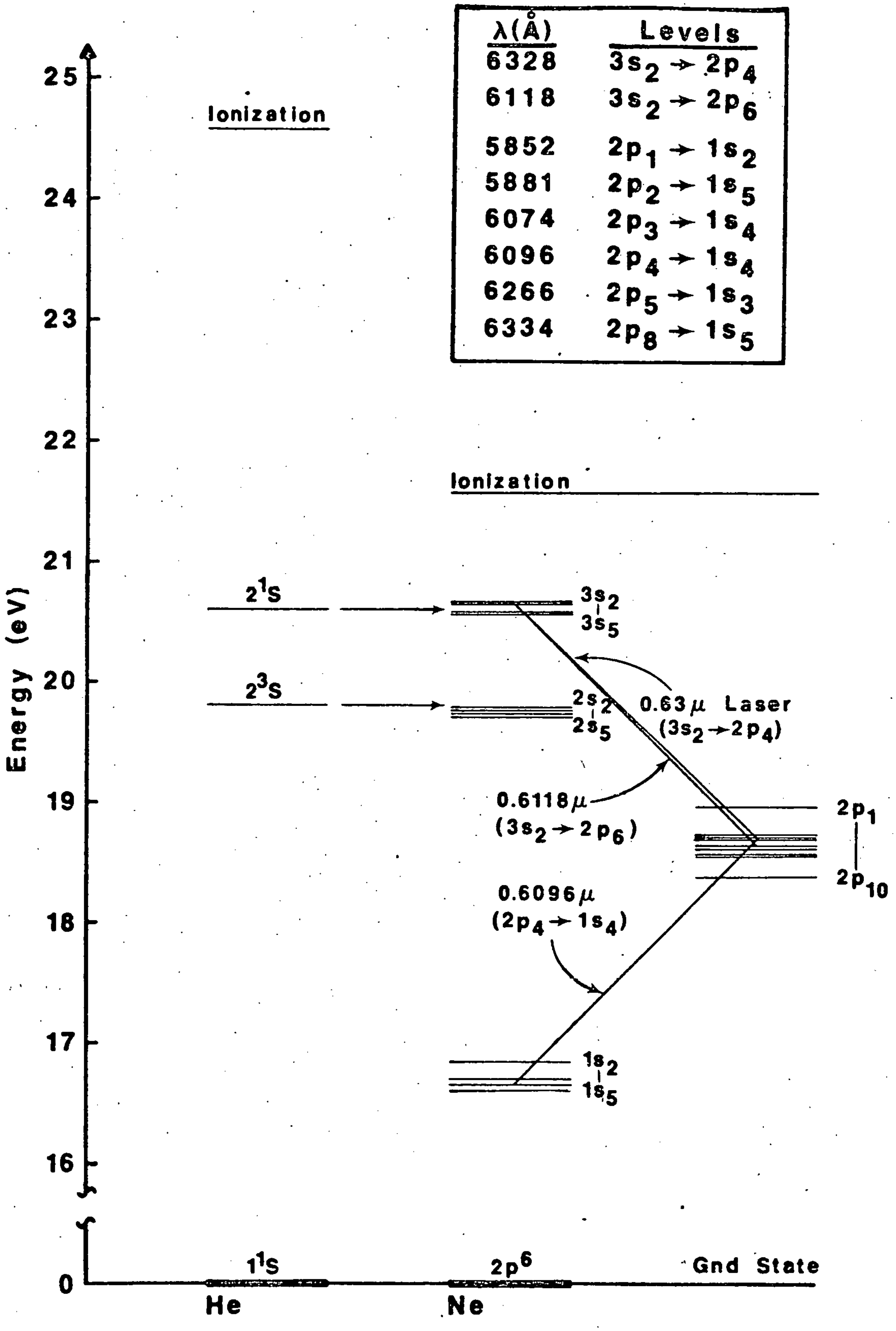
The isotope shift for Ne^{20} and Ne^{22} is about 875 MHz(2), with Ne^{22} at the higher frequency. The half-power full width of the Doppler-broadened line is about 1400 MHz. Hence the isotope shift is hardly enough for preferential pumping of one isotope. A method for operating the laser to improve the selectivity of excitation of the one neon isotope had been developed in the course of research on self-locking in multimode lasers.(3) The technique is preferable to operating the laser in a single mode since higher power is achieved and ultra-sensitive control systems are not required to control cavity tuning. The technique depends on having one mirror of the laser resonator moving at a constant velocity of about 10 cm per sec. If the mirror is moving to shorten the cavity, the laser modes are confined to the upper half of the Doppler linewidth, while if the mirror motion lengthens the cavity the modes are restricted to the lower half. There is no appreciable loss of laser power since the laser beam traverses the gain medium in both directions and, because of image hole-burning, interacts with all of the atoms of the one isotope.

The 6328A° 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-Ne^{20} laser should reduce the population of the upper

Ne^{20} level, leaving that of the Ne^{22} 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 Ne^{20} 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.

A spectroscopic technique for observing isotopic separation from the change in fluourscent line shape has been used throughout. Using a specialized scanning Fabry-Perot spectrometer a very sensitive technique has been developed, capable of detecting a change in isotope ratio of less than 1%.

Although the system has a high detection sensitivity for isotope separation all attempts to achieve measurable separation of the neon isotopes were unsuccessful. It was believed that one reason for the negative result was that the relative population of the laser $3s_2$ and $2p_4$ levels was too low. Therefore during the final year it was decided to attempt experiments involving two radiation sources, one a monoisotopic neon radiation source to selectively pump from the 1s levels to the 2p levels, the other the 0.63 μ laser source to pump from the $2p_4$ to the $3s_2$ level. This experiment has also produced a negative result. Some of the observations during the experiment suggested that very slight



λ(Å)	Levels
6328	$3s_2 \rightarrow 2p_4$
6118	$3s_2 \rightarrow 2p_6$
5852	$2p_1 \rightarrow 1s_2$
5881	$2p_2 \rightarrow 1s_5$
6074	$2p_3 \rightarrow 1s_4$
6096	$2p_4 \rightarrow 1s_4$
6266	$2p_5 \rightarrow 1s_3$
6334	$2p_8 \rightarrow 1s_5$

Fig. 1. Helium and Neon energy levels. A list of some of the lines monitored is also shown.

cataphoretic separation might be occuring, both these effects were eventually shown to be extraneous. The high intensity fluorescent radiation lamp was found to affect the temperature of the cataphoresis tube, and therefore to cause an observable change in line shape, but a change which was not consistent with a change in isotope ratio. The general theory of radiation-induced cataphoretic isotope separation has been covered in the final report by Chanin and Oskam(4), January 1980. Their results using monoisotopic radiation lamps were also negative. Their conclusions are also pertinent.

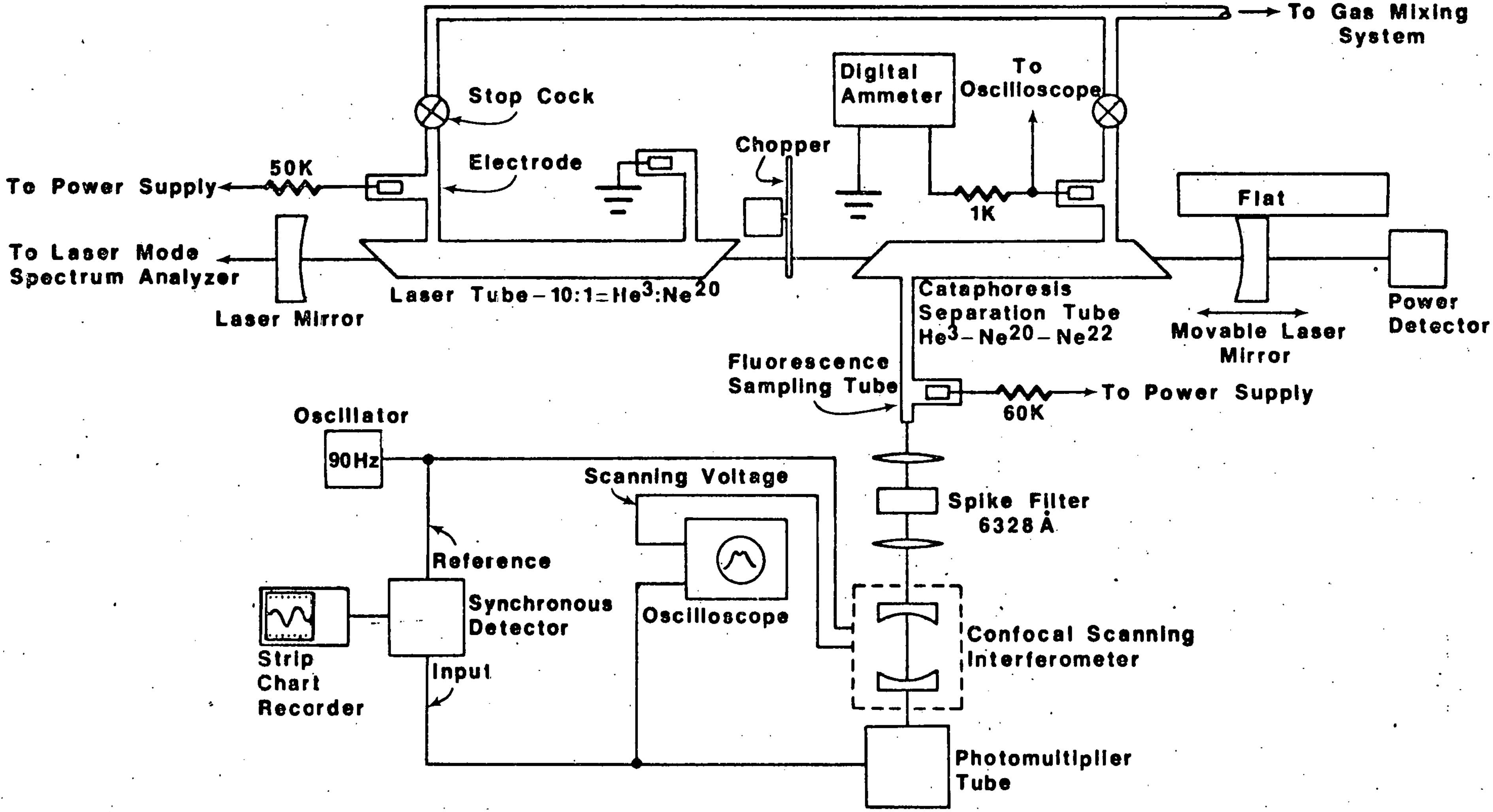
2. Apparatus

2.1 The Basic System

The basic apparatus for studying laser-induced cataphoretic isotope separation at 6328A° is shown in Fig. 2. For observing the laser-induced current changes 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 pump and filling station was assembled from an existing system, with a Granville-Phillips differential capacitive manometer and calibrated leak valves added. Both the laser and cataphoresis tubes are attached to the pumping station. The laser cavity is about 1.2 m long, and uses high-reflectivity mirrors of 1.47 and 10.0 meters radii of curvature. The laser operates with high-order off-axial modes, and the cavity is designed for the radiation to fill the bore of the cataphoresis tube as completely as possible.

The shape of the cataphoresis tube has evolved considerably. It is now about 50 cm long, with a bore diameter of 6 mm. Observation of the spontaneous fluorescence is made using a side tube of about 5 inches in length, which is in the discharge path but not in the path of the laser beam. Thus one is able to observe the relative fluorescent intensity of the Ne^{20} and Ne^{22} isotopes near one end of the discharge, and to note any change in the relative populations of the two isotopes due to cataphoretic separation. The fluorescence sampling tube produces a relatively intense fluorescent beam of small source area which is fed through spike filters to a wide-angle broad-band confocal



Neon Isotope Separation Schematic
Figure 2

spectrometer. The spectrometer is scanned repetitively through its free spectral range, and the double-isotope fluorescent line can be displayed on an oscilloscope. A small AC scan at 90 Hz, with narrow-band signal amplification and synchronous detection also permits the slope of the line shape to be simultaneously traced out on a strip chart recorder with a much better signal-to-noise ratio. This slope tracing is very sensitive to small changes in the isotope ratio.

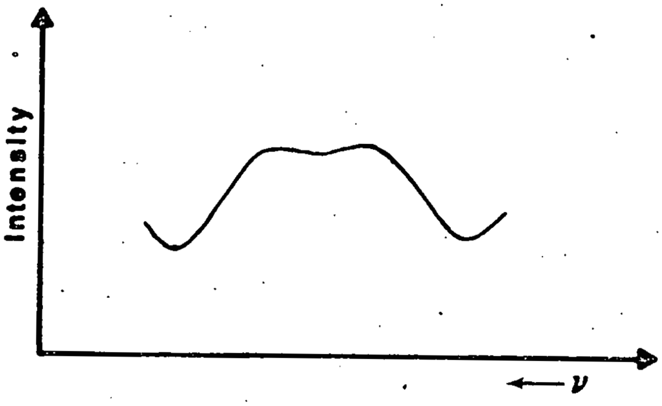
The isotope splitting at 6328 A° for Ne^{20} and Ne^{22} is about 875 MHz. At the temperature of an ordinary discharge tube the Doppler broadening is appreciably greater than this, and is sufficient to keep the two isotope lines from being properly resolved. But by cooling the fluorescence sampling tube by means of liquid nitrogen (the main laser-irradiated section of the cataphoresis tube is not cooled) the Doppler width is reduced and the isotope lines are better resolved.

2.2 The Scanning Fabry-Perot Spectrometer

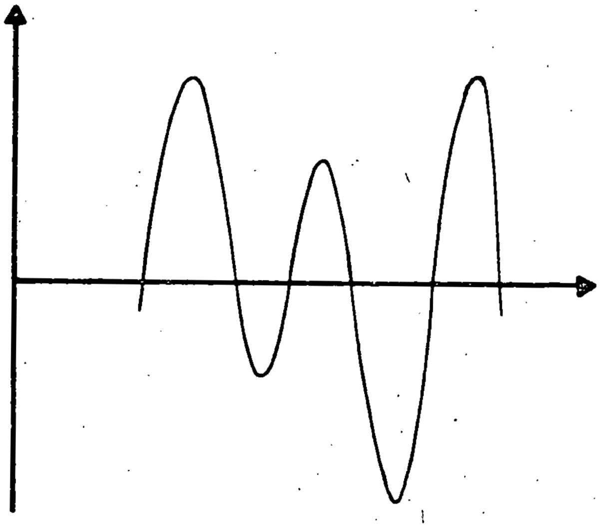
There are two similar versions of the instrument available, one with 3.75 cm spacing and a free spectral range of 2.0 GHz, the other with spacing of 2.0 cm, and free spectral range of 3.75 GHz. Because the free spectral range of the instruments are comparable with the linewidth when two isotopes are present, the line shape is not properly reproduced, even though the linewidth of the spectrometer is less than that of the fluorescent line. But the sensitivity for observing small changes in the ratios of the two isotopes is thereby improved. That is, if the ratio of the two isotopes is initially equal to unity, a small change in the isotope ratio produces relatively large changes in the slope of the line shape curve near the central minimum.

Figures 3(a) and 3(b) show typical results of calibration tests of the sensitivity of one of the scanning Fabry-Perot spectrometers, when used to monitor the $Ne^{20}:Ne^{22}$ isotope ratio.(5) Even when the fluorescence sampling tube is cooled to reduce the Doppler linewidth the two isotopes are only partially 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 quantities of one isotope are added. For example, in a 10:1:1 mixture of $He^3:Ne^{20}:Ne^{22}$ the ratio of the amplitudes of these peaks changed by 11% for each 1% change in the $Ne^{20}:Ne^{22}$ ratio. Under the very best conditions 1/10% change in the $Ne^{20}:Ne^{22}$ ratio is detectable, and a 1% change is readily observable under virtually all conditions.

The high sensitivity of the Fabry-Perot system for detecting changes in the isotope ratio has had a significant influence on the program. The relatively small population of the neon $3s_2$ and $2p_4$ levels associated with the 0.63 μ laser has meant that the cataphoretic separation of the neon isotopes induced by laser radiation would be relatively small. That is, most of the neon ions would arise from the highly populated 1s levels, not the 3s or 2p levels. But because of the high sensitivity for observing cataphoretic separation it had been anticipated that some cataphoretic separation would be observable. However, to date, this expectation has not been fulfilled.



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



(b) Slope of the spectrum curve.

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

2.3 The Opto-Galvanic Effect

When early attempts to observe cataphoretic separation of the neon isotopes induced by the 6328A° laser radiation produced negative results, it was decided to examine the laser-induced current (or voltage) changes in the cataphoresis tube[6-8], with the hope of improving the prospect for obtaining measurable isotope separation.

In Fig. 4 some typical i-v characteristic curves and measured values of the opto-galvanic effect for He:Ne^{20} in the cataphoresis tube, with the laser tube also containing He: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 kΩ. The values of Δi can readily be converted to changes in voltage, Δv, by multiplying by 61 k, but the increments are of opposite sign 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 curve, it is not clear what represents the best way to report the data. The raw data is shown here, noting that in general an increase in current (or decrease in voltage) indicates an increase in the ionization rate. The data is discussed further in Section 3.1.

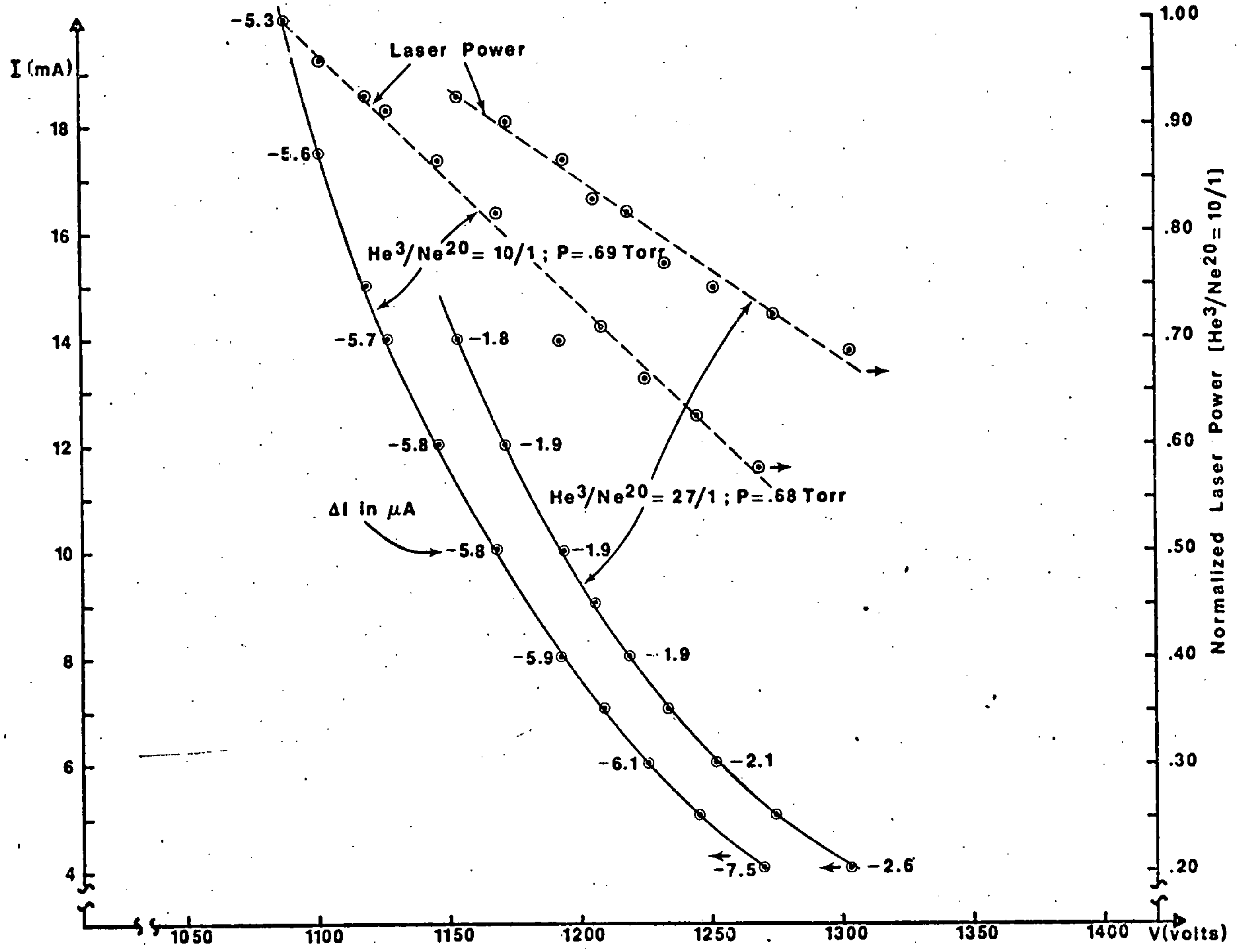


Fig. 4. Discharge parameters for the cataphoresis tube for two He:Ne ratios, 10:1 and 27:1. The opto-galvanic current changes, Δi, are shown. the variation in laser power is also recorded.

2.4 Moving-Mirror Laser for Selective Excitation

Previous work has shown[3] 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 higher frequencies when the laser resonator is being shortened and to lower frequencies when the laser is being lengthened. In the apparatus shown in Fig. 2, the laser tube contains He^3: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 as expected 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 Ne20 isotope were interacting with the laser radiation because the Ne^{20} concentration is reduced by a factor of 2. But when the mirror is moved outards to reduce the laser frequency and thereby reduce the interaction with the Ne^{22} atoms the opto-galvanic is further reduced by about 25%. Similarly, if the cataphoresis tube contains He^{3}-Ne^{22} the opto-galvanic effect due to the laser is still quite large. But if the laser frequency is down-shifted by moving the mirror the opto-galvanic effect is reduced by about two thirds.

These observations show the importance of shifting the laser radiation by as much as possible toward the one isotope line if pumping is to be selective. This is especially true if the cataphoresis tube provides gain, but is not so important if the cataphoresis tube produces loss.

2.5 Monoisotopic Radiation Source

In order to improve the prospect of observing cataphoretic isotope separation it was decided to use a combination of laser radiation and a monoisotopic fluoresencent radiation source. the latter would primarily affect the 1s and 2p populations, while the former would mainly alter the $2p_4$ and $3s_2$ populations.

Two spiral tubes of 7 turns each were installed around the cataphoresis tube. The inner bore diameter was 0.8 cm, and the overall length was about 30 cm. A reflective shield was placed outside the lamps, with aluminum foil on the inner surface. The tubes were filled with Ne^{20} at about 0.7 Torr. The maximum current is about 70 mA.

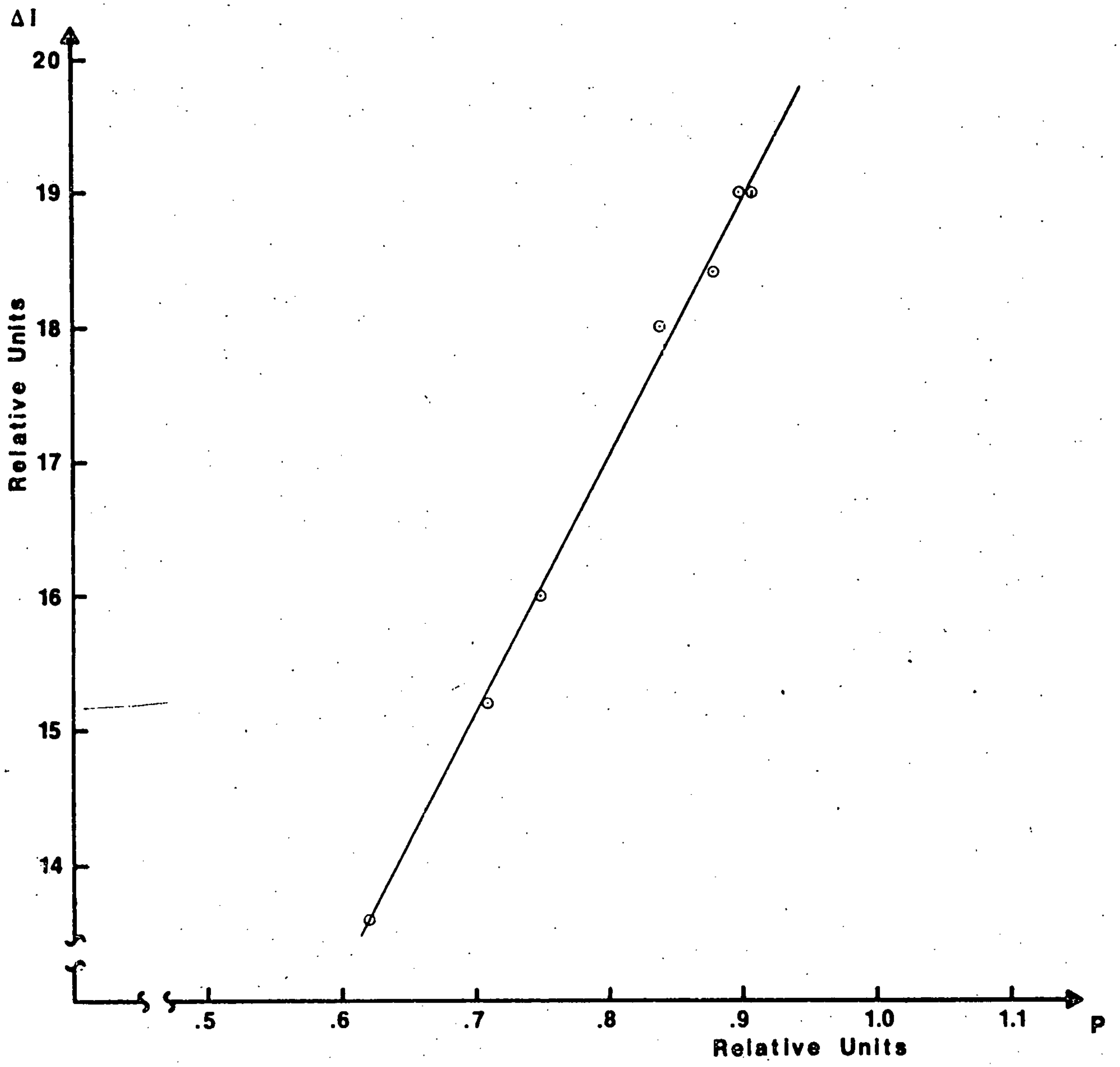


Fig. 5. Opto-galvanic effect current versus laser power in He:Ne mixture of 28:1 at 2.7 Torr.

3. Results and Discussion

3.1 Opto-Galvanic Effect.

It is seen from Fig. 4 that not only does the opto-galvanic effect for 6328A° laser radiation decrease for high currents in the cataphoresis tube, 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 gas. This observation appears to be related to the relatively low population of the high-lying $3s_2$ and $2p_4$ levels compared with those of metastable 1s levels. It is also seen in Fig. 4 that the 6328A° laser power 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 and if the Δi values of Fig. 4 are recalculated for a constant magnitude of laser power, these values would show an even greater reduction as the dc discharge current in the cataphoresis tube is increased. The linear relationship of Fig. 5, however, shows that it would be better to have a higher laser power since the transition is apparently not saturated. On the other hand, if the power is increased too much some difficulty will be experienced in being able to selectively excite the one isotope (section 2.3). That is, as the laser power supplied mainly by the He:Ne^{20} laser tube is increased so as to saturate the Ne^{20} in the cataphoresis tube, the Ne^{20} population in the $3s_2$ level is decreased, and there will be unwanted interaction with the Ne^{22} atoms. Thus with the small isotope shift encountered here optimum power level for selective excitation is fairly important.

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 is 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 involved (9). Our data is consistant 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(10). However, there is still the possibility that the opto-galvanic effect is partly attributable to the helium buffer gas(6) at least over some of the current range.

A full explanation of the opto-galvanic effect in a He-Ne over a wide current remains an interesting problem which it is hoped can be pursued further. However, the small magnitude of the effect is discouraging in regard to laser-induced cataphoretic isotope separation. For the high currents and high He-Ne ratios generally desirable for cataphoretic separation(1), the effect of the laser radiation on the ionization probability is apparently very small.

In an effort to improve the prospects for inducing cataphoretic isotope separation experiments were undertaken with the two radiation sources, a helical monoisotopic radiation lamp as well as the laser. Mixtures of helium and neon as well as neon alone were used in the cataphoresis tube, with pressures ranging from 0.2 Torr to 3 Torr. In all cases the opto-galvanic current change when the laser radiation was chopped was disappointingly small, and in many cases narrow band amplifier had to be employed in order to get the fluctuating current signal above background noise. The laser-induced opto-galvanic current charges were generally in the range 0.1% or less.

It is therefore concluded that the effect of the 6328A° laser radiation on the ionization rate is too small to induce measurable cataphoretic separation, even with the high sensitivity for detecting such separation which the system possesses. The relative population of the $3s_2$ and $2p_4$ levels is apparently much too small in comparison with that of the 1s levels.

The opto-galvanic fractional current changes in the cataphoresis tube produced by the Ne^{20} helical radiation lamp was much greater than for the laser radiation, being as high as 20-30%. Fig. 6 shows a typical i-v curve for the cataphoresis tube, with the opto-galvanic current change Δi induced by the helical radiation lamp recorded for a 61 kΩ load. In this case the gas mixture was 2 Torr of Ne^{20} and Ne^{22} in equal proportions. Although the opto-galvanic effect is appreciably larger than for the laser radiation the fractional current changes still decrease rapidly for the high currents which are desirable for cataphoresis. Still, it is disappointing that no cataphoretic isotopic separation has been observable when pumping with the Ne^{20} helical radiation lamp even with the high detection sensitivity of the system.

3.2 Selective Excitation

Because cataphoretic separation has not been observable one area of concern has been whether the excitation by the He-Ne^{20} laser and the Ne^{20} helical radiation lamp has been sufficiently selective, particularly as the isotope shift is of the same order as the doppler bandwidth for most of the lines. Of equal concern is whether the collision rate is high enough to cause significant excitation exchange between the isotopes, thus counteracting the selective excitation. Experiments have been performed to examine these questions, mainly by observing the fluorescence from 2p levels involving the 6096A°, 6074A° and 5882A° lines. It appears that selective excitation is achieved by both the laser (with moving mirror) and by the Ne^{20} helical radiation lamp. Also, significant excitation exchange between the Ne^{20} and Ne^{22} isotope is not occurring up to a partial pressure of 2 Torr of Ne, at least for the 2p levels.

3.2.1 Laser Pumping

One of the experiments involved observing the change in 6096A° fluorescence induced by the He^3-Ne^{20} laser when the cataphoretic tube contained He^3-Ne^{20}-Ne^{22} at 2 Torr in a 10-1-1 mixtures. Fig. 7 shows photographs of the double-peaked fluorescence lineshapes from the scanning Fabry-Perot spectrometers. Ne^{20} corresponds to the peak on the right side.

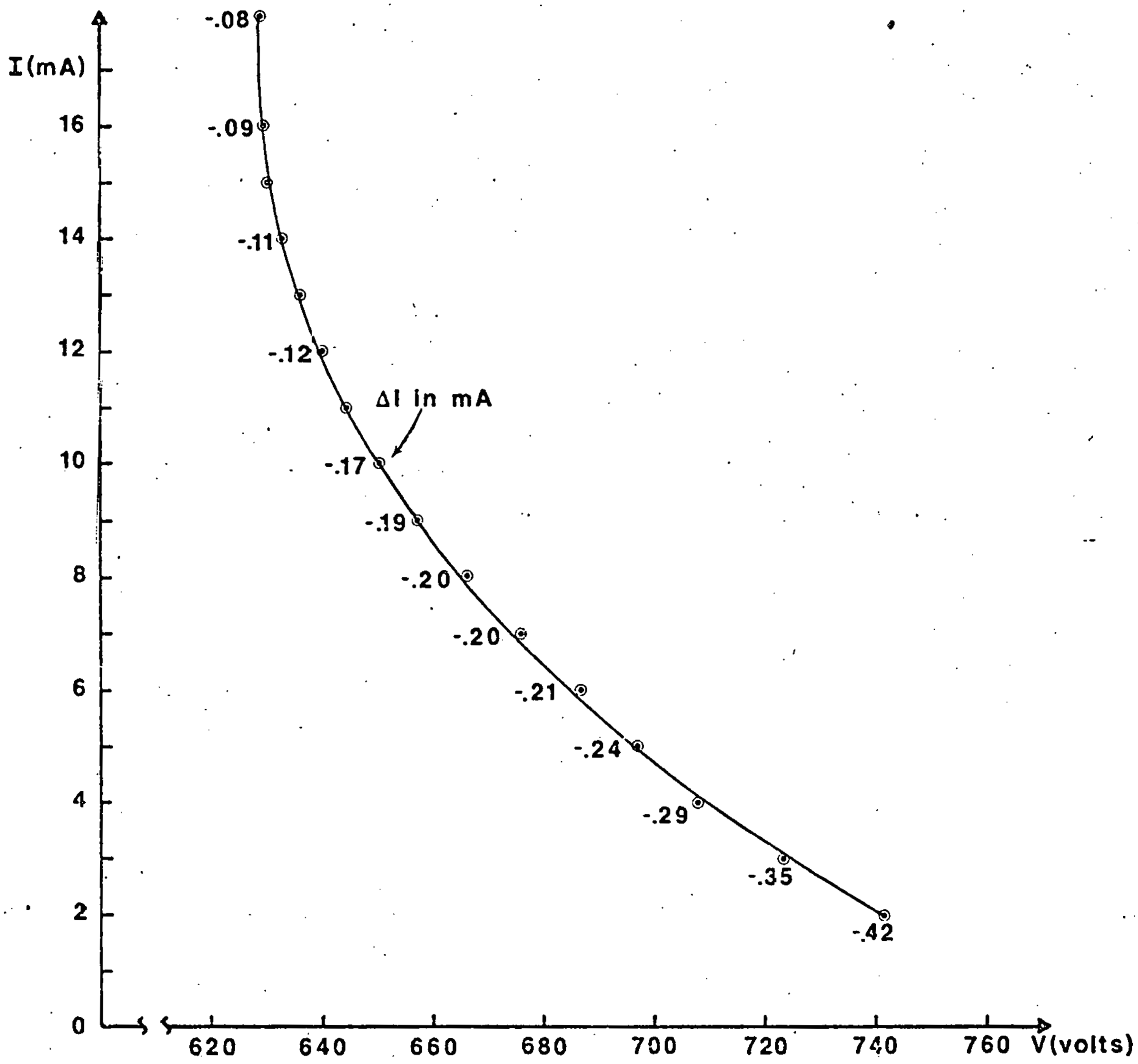
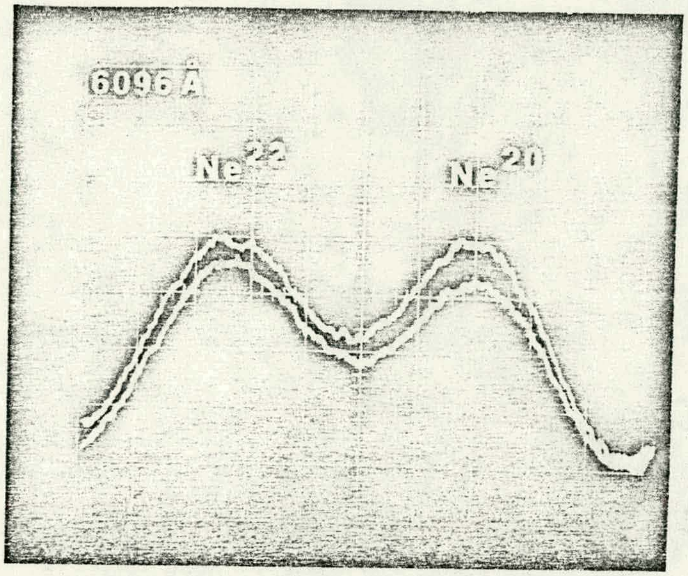


Fig. 6 Discharge parameters for the cataphoresis tube with a gas mixture of Ne^{20}:Ne^{22} = 1 at a total pressure of 2 Torr. The opto-galvanic current changes, Δi, shown are induced by the helical radiation lamp.

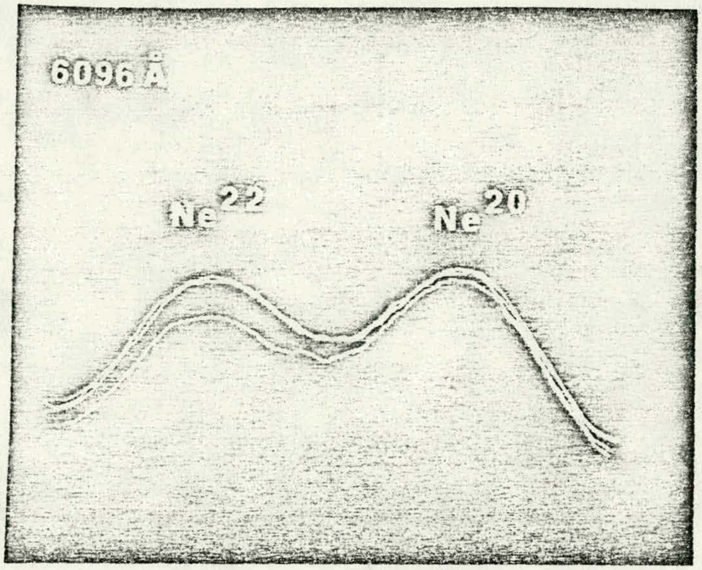
In Fig. 7(a) there are scans for the laser off (lower curve) and for the laser on (upper curve), but when the laser mirror is stationary. The effect of laser pumping on the $2p_4$ level is apparent, showing an increase in the 6096A° radiation and in the $2p_4$ population. But the effect is not confined to the Ne^{20} peak, and is quite appreciable on the Ne^{22} peak as well, showing either that the laser radiation is interacting significantly with the Ne^{22} atoms or that there is appreciable excitation exchange between the Ne^{20} and Ne^{22} atoms. But by using the moving mirror technique to shift the laser radiation to lower frequencies, and away from the Ne^{22} line it is possible to resolve the question.

In Fig. 7(b) the effect of the moving mirror is observed. The reduction in the laser interation with the Ne^{22} atoms while the laser is moving is evidenced in the lower trace for the left-hand peak. Taking Figs. 7(a) and 7(b) together, it can be seen that the population of the $2p_4$ level of the Ne^{20} atoms is being selectively increased by the laser, and that this selective pumping is not undone by collisional excitation exchange between the isotopes.

Hence it can be concluded that the moving-mirror laser provides a means of obtaining selective isotope excitation, and that the selective population change is not destroyed by collisional excitation exchange. Observation on selective pumping by the laser have been confined to the lower $2p_4$ laser level, and attempt to do equivalent observations on the upper $3s_2$ laser level have not been successful. The difficulty is that there is interference from the laser radiation and it has not been possible to complete- ly overcome the difficulty. However, since



7(a): Shows the increase in population of the 2_{p4} level with stimulated emission from the 6328A° laser $(3s_2 \rightarrow 2p_4)$. The laser mirror is stationary.



7(b): Shows the increase in selective pumping of the Ne^{20} $2p_4$ level by moving the laser mirror.

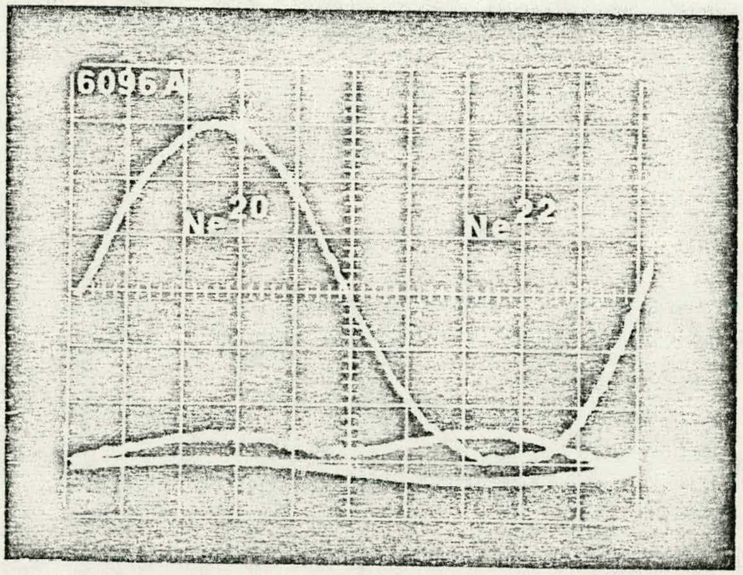


Fig. 8. Demonstrates selective excitation of the Ne^{20} $2p_4$ level by the helical radiation lamp (upper trace). The middle trace shows the normal fluorescent levels.

the isotope shift for both of the laser levels is very small compared with kT it is not likely that excitation exchange occurs between the isotopes at the $3s_2$ level when it does not occur at the $2p_4$ level.

3.2.2 Ne^{20} Helical Radiation Lamp Pumping

Selective pumping to the 2p levels by the Ne^{20} helical radiation lamp has been demonstrated by observing the fluorescence from the 2p levels. Selective pumping of the Ne^{20} atoms from the 1s levels to the $2p_2$, $2p_3$ and the $2p_4$ levels have been observed using narrow band spike filters in combination with the scanning Fabry-Perot spectrometer to display the 5882A° line, the 6074A° line and the 6096A° line. A photograph of the 6096A° line is shown in Fig. 8 for 0.4 Torr of Ne^{20}, Ne^{22} in equal proportion. The lowest trace merely shows the extraneous coupling from the helical pump lamp to the spectrometer when the cataphoresis discharge tube is off. The middle trace shows the double-peaked fluorescence from the cataphoresis tube when the pump tube is off. The upper peak shows the large increase in the Ne^{20} peak due to pumping from the $1s_4$ level. It appears that the population of the $2p_4$ level can be increased by about a factor of four. Again it is apparent that excitation exchange due to collisions between the Ne atoms is not significant.

A population increase in the $3s_2$ level due to pumping by the Ne^{20} helical radiation lamp has also been observed using the 6118A° line. But the pumping was comparatively small, appreciably less than the effect produced by the laser radiation.

4. Conclusions

Because the opto-galvanic current changes in the cataphoresis tube observed for the 6328A° laser radiation is very small under all conditions it is concluded that the possiblilty of 6328A° laser-induced cataphoretic separation is extremely small. This conclusion is reinforced by the observaton that the opto-galvanic effect decreases as the current in the cataphoresis tube increases. Whereas high discharge currents are required for cataphoretic separation$^{(1)}$.

Pumping from the 1s levels by means of the helical Ne^{20} radiation lamp has not sufficiently increased the efficiency of the laser radiation in inducing cataphoretic separation to alter the conclusion of the proceeding paragraph. The $3s_2$ and $2p_4$ laser level populations remain too low compared with those of the 1s levels.

The opto-galvanic effect produced by the helical Ne^{20} radiation lamp is considerably greater than for the 6328A° laser radiation. Since the 1s populations are so large and play such a dominant role in the two-step ionization process in neon it appears that to be successful any scheme for radiation-induced cataphoretic separation in neon must involve pumping from these levels.

Selective pumping of the one isotope by both the He-Ne^{20} laser and by the Ne^{20} helical radiation lamp has been demonstrated, even though the isotope shift is small and is comparable with the half-power half-width of the Doppler-broadened lines.

Excitation exchange between the Ne^{20} and Ne^{22} atoms does not appear to be a significant factor up to partial pressures of Ne of about 2 Torr. As discussed by Chanin and Oskam(4) the difficulty in obtaining cataphoretic separation in neon appears to stem from the competing effect of reducing the 1s populations for one isotope while simultaneously increasing the populations of higher levels for the same isotope. The result is that as the current increases the net influence by the radiation on the ionization rate is diminished. This conclusion is supported by the observation that the opto-galvanic effect decreases for higher currents.

Although there appears to be no basis for continued attempts to induce cataphoretic separation in neon using the He-Ne 6328A° laser, there may be value in studies designed to help understand the basic mechanism for the opto-galvanic effect in a He-Ne discharge produced by 6328A° radiation. Such studies may help explain why the opto-galvanic effect is current dependent, to what extent the 1s populations are involved, and whether the helium ionization plays a significant role(6).

Because of the high sensitivity of the present scanning Fabry-Perot system for detecting small changes in the isotope ratio in neon, it may be of value to undertake some additional studies using the helical Ne^{20} radiation lamp before the system is dismantled. If increased pumping by the Ne^{20} helical lamp can be obtained by improved design there is the possibility of observing cataphoretic separation. Consideration will be given to continuing such experiments, at low priority.

The most worthwhile experiments at this stage would involve a tunable dye laser(12,13), but would not require the dye laser to have the extremely narrow linewidth and high stability required for selective excitation of one isotope. First, as discussed by Chanin and Oskam(4), the He-Ne cataphoresis tube should be operated under conditions where cataphoretic separation of the helium and neon occurs, and a neon gradient is observable. This will probably require a high He-Ne ratio, and a high discharge current. The discharge tube would contain helium and a single neon isotope, probably Ne^{20}. Then a tunable dye laser with relatively broad linewidth can be used to pump from the 1s levels on selected lines. Only if a fairly large opto-galvanic effect is observed for the dye laser radiation would there be justification for considering experiments involving pumping of one isotope selectively. The first choices for the transitions to be explored with dye laser pumping are 6267A° ($1s_3$ - $2p_5$) and 6334A° ($1s_5$ - $2p_8$). Narrow band spike filters are on hand for these lines, and they are of relatively long wavelength so that reasonable high power should be available from the dye laser. The $1s_3$ and $1s_5$ are true metastables, but both are thermally coupled to the $1s_4$ to some extent(11). The hope would be that by selectively pumping for the Ne^{20} isotope from either of the $1s_3$ or $1s_5$ metastable levels one can decrease the Ne^{20} population in al three $1s_3$, $1s_4$ and $1s_5$ levels more than the resulting increase in Ne^{20} populations for the $1s_2$ and 2p levels. It is noted that for the low Ne partial pressures which will probably be used in the cataphoresis cell it is likely that the absorption coefficient is sufficiently low that the dye laser beam can be directed down the bore of the cataphoresis tube. Observations made recently on the effects of radiation trapping on line shape tend to support this belief. Consideration is being given to the possibility of undertaking these experiments within the limitations of equipment and resources available at present.

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