

SPECTRAL LINE BROADENING IN NEON DUE TO THE RESONANCE INTERACTION*

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The self-broadening of the 585.2 nm line in neon has been studied using a scanning Fabry-Perot interferometer. Experiments were performed in emission at 77K and 273K. The broadening constant was found to be the same at the two temperatures to within the experimental uncertainty, and equal to $(6.14 \pm 0.05) \times 10^{-20} \text{ cm}^{-1} \text{ atom}^{-1} \text{ cm}^3$.

1. Introduction

The broadening of spectral lines is of interest as it gives information on the collision processes in a gas and the interactions between atoms. The case of resonance broadening is important as it is caused by a particularly simple and strong interaction for which calculations may be readily performed (see, for example, [1-3]). The case therefore provides a critical test of line broadening theories; in the so-called "impact regime" these predict a broadening given by [4]

$$\Delta\tilde{\nu}_{1/2} = k_{JJ'} \frac{e^2 f}{8mc^2 \tilde{\nu}_0} N \text{ cm}^{-1}.$$

Here $\Delta\tilde{\nu}_{1/2}$ is the Lorentzian halfwidth (full width at half maximum), f and $\tilde{\nu}_0$ are respectively the absorption oscillator strength and the wavenumber of the resonance transition, and N is the number density of perturbers. $k_{JJ'}$ is a numerical factor depending only on the total angular momenta (J and J' respectively) of the upper and lower levels of the resonance transition.

Minor modifications to the conventional theory have been proposed (see, for example, [5]) but these leave largely unaffected the essential properties of resonance broadening, (i) proportionality to number density of perturbers and to the oscillator strength of the resonance transition, and (ii) absence of temperature dependence and pressure shift.

* Dedicated to Professor Aleksander Jabłoński on the occasion of his 80th birthday.

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An experimental programme to test these predictions has been in progress for a number of years [6–12]. The work has concentrated on the rare gases, since it is then simple to find the number density of perturbers. The broadening of an emission line terminating on the resonance level is measured rather than that of the resonance line itself (which is in the far ultra-violet region of the spectrum). It is then possible to choose conditions such that there is little self-absorption for the line studied. The profile of the line is measured as a function of temperature and pressure and this enables the resonance broadening constant to be determined.

The element most thoroughly studied has been helium [6, 8, 12], since an accurate value for the f -value is available. However, helium presents special difficulties due to its low mass; the Doppler effect is large, and any excitation mechanisms which distort the velocity distribution can lead to appreciable systematic errors in profile measurements [12]. Neon was therefore chosen for the present study; although the f -values for the resonance transitions are not known precisely, velocity effects are less important. One also has the advantage that use may be made of a tuneable dye laser to measure the instrumental profile for the lines of interest, and to study resonance broadening in absorption from the resonance level.

2. Apparatus

2.1. The light source

This was similar to that described by Vaughan [8]. Light was collected from the positive column of a weak DC discharge run at pressures in the range 0.6 — 9 torr. The region of the discharge studied was constructed, to minimize self-absorption, and immersed in a constant temperature bath (either liquid nitrogen or ice). The current used was always in the range 0.1–0.5 mA.

2.2. The high resolution system

High resolution was obtained with a plane Fabry-Perot etalon (spacer 1.51 cm, free spectral range 331.0 mK) which was pressure scanned by leaking in CO₂ through a constant flow valve. A 3m Littrow mounted diffraction grating (600 lines/mm) provided auxiliary dispersion and the intensity at the centre of the etalon ring pattern was monitored using a photomultiplier in the photon counting mode. For some of the runs two different lines close in frequency were recorded simultaneously. The integration time was 1 second, and the data were recorded on paper tape for subsequent computer analysis.

2.3. Analysis of data

This was essentially as described by Silver and Stacey [13]. Firstly a polynomial was fitted to the centres of gravity of the peaks in the trace, which was typically over five or six orders of the etalon. This eliminated non-linearities in the scan, typically 1 % per order. Then the trace was superposed into one order with 128 data points as in figure 1. This superposed profile was then fitted to a convolution of an Airy function with a Gaussian distribution by a least squares minimization routine. This theoretical profile comprised

two components, each with the same Airy width but with differing Gaussian widths (corresponding to the different masses of ^{20}Ne and ^{22}Ne). We thus assume that the pressure broadening of the two isotopes is the same, i. e. that the isotopes act as equivalent perturbers for each other. This is expected to be justified since the isotope shift in the resonance line is less than the energy uncertainty during a collision. There was also a contribution

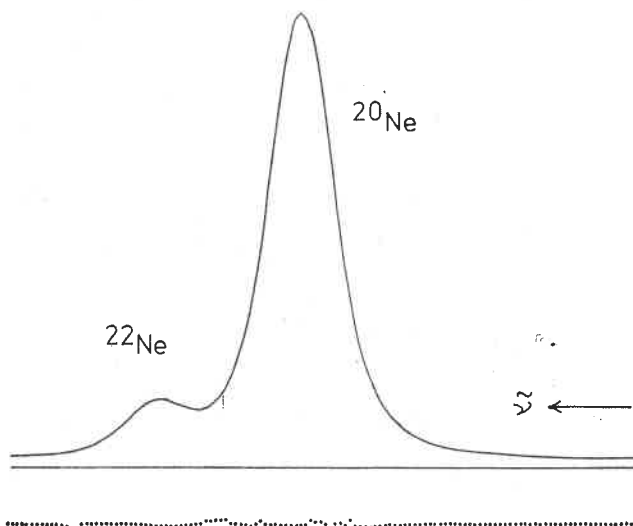


Fig. 1. The spectral profile of the transition 585.2 nm in neon, in the form of a superposition (see text). The discharge pressure and temperature were 1.51 torr and 77 K respectively. One order of interference (331 mK) is shown. The dotted line shows the difference, magnified by a factor of 6, between the superposed data and the best-fit theoretical profile

to the theoretical profile from the photomultiplier dark current, which was measured for each run.

The Gaussian width of the line is assumed to be due to the thermal motion of the atoms (with a contribution from small effects such as the finite size of the scanning aperture). The Airy width has three contributions: the instrumental width of the etalon, the natural width of the line being studied and the pressure broadening of the line. It can be shown that the convolution of an Airy distribution with any Lorentzian profile is an Airy distribution of increased width. Therefore the natural width and the pressure broadening contribute solely to the analysed Airy width. This Airy width may then be converted back to an equivalent Lorentzian width [14] which is given simply by the sum of the three contributing widths.

3. Experiments

The majority of the experiments were performed on the line 585.2 nm, which terminates on the strong resonance level $1s_2$. However, some tests were performed on other lines to make independent measurements of the isotope ratio.

3.1. Preliminary experiments

These were performed to investigate various possible sources of error. In particular, tests were made to ensure that the amount of scattered light in the spectrograph was at an acceptable level. Typically the peak count rate was $40\,000\text{ s}^{-1}$ with a photomultiplier dark count of 70 s^{-1} and an estimated amount of scattered light less than 20 counts s^{-1} .

To test for self-absorption profiles were recorded under the same conditions of temperature and pressure in the discharge but at different currents (0.1–0.5 mA). No systematic change in the analysed widths or the isotope ratio was detected in these experiments. This also served to show that the photon counting system was linear, as the count rate varied by a large factor in these experiments.

As in recent work on helium, computer simulation tests were performed to check the effect on the analysis of various small effects including saturation of the detection system, slight misalignment of the scanning aperture, and the finite sampling time. All these effects were found to change the analysis only by small amounts; in particular, the analysed Airy width never changed by more than 0.4 mK.

3.2. Experiments at 77 K

Figure 2 shows the results of experiments at 77 K for pressures up to 8 torr. These results were obtained with a discharge current of 0.1 mA. Within the experimental uncertainty, the Lorentzian width of the line varies linearly over this range, with a slope of

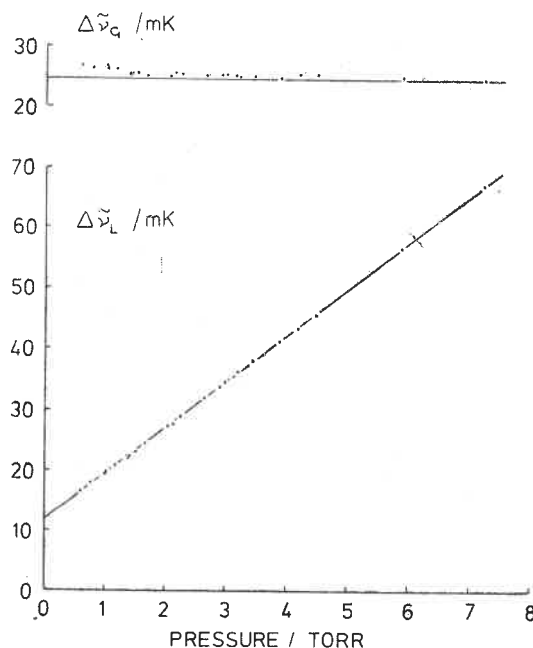


Fig. 2. Contributions to the line-width of 585.2 nm at 77 K. The lower plot shows the variation of the analysed Lorentzian width with pressure, the upper plot shows the variation of the analysed Gaussian width. The horizontal line corresponds to the bath temperature

$7.65 \pm 0.03 \text{ mK torr}^{-1}$. The Gaussian width is close to the bath temperature value of 24.1 mK over most of the range, thus indicating the absence of the distorting effects encountered in helium [12]. The analysed value of the isotope ratio varies slightly from the natural abundance value of 0.097, depending on the origin of the sample used.

3.3. Experiments at 273 K

Figure 3 shows the results of experiments at 273 K for pressures up to 9 torr with a current of 0.15 mA. The Gaussian width is again close to the bath temperature value except at very low pressure, and we take this to be due to the atoms not being thermalised properly before emission of a photon at these low number densities. Taking the points above 2 torr, the slope of the graph of Lorentzian width is $2.18 \pm 0.02 \text{ mK torr}^{-1}$.

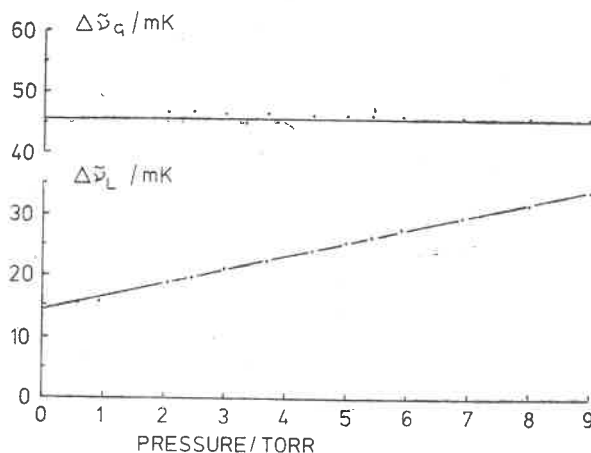


Fig. 3. Contributions to the line-width of 585.2 nm at 273 K. The lower plot shows the variation of the analysed Lorentzian width with pressure, with a straight line drawn through the points above 2 torr. The upper plot shows the variation of the analysed Gaussian width. The horizontal line corresponds to the bath temperature

3.4. Measurement of the instrumental profile

The instrumental profile was studied by tuning a dye laser of bandwidth 0.1 mK to the wavelength 585.2 nm and using it to illuminate the high resolution system. It then acted effectively as a monochromatic source and when the etalon was scanned as usual the instrumental profile was recorded. These experiments showed that the instrumental profile is very sensitive to the alignment of the apparatus and that it is not well described by the convolution of an Airy distribution and a Gaussian function. The instrumental profile was seen to improve substantially in quality when the etalon was stopped down so that only the central region was used.

Experiments are now in progress to measure the broadening at low densities accurately by simultaneously recording the 585.2 nm line and the instrumental profile as measured by the laser, the wavelength of the latter being tuned close to 585.2 nm. In this way we hope to study the so-called "extrapolation anomaly" [6].

3.5. Experiments on other lines

Independent checks were made on the isotope ratio of the gas by studying other lines not terminating on the strong resonance level. Generally, at low densities and currents the analysed isotope ratios agreed with that found for the line 585.2 nm, to within the experimental error. These experiments were performed at 77 K. At higher densities and currents there was disagreement, due probably to the onset of self-absorption in the non-resonance broadened lines.

4. Discussion

From the above results, we obtain values for the broadening of 585.2 nm of $(6.13 \pm 0.03) \times 10^{-20}$ and $(6.16 \pm 0.25) \times 10^{-20} \text{ cm}^{-1} \text{ atom}^{-1} \text{ cm}^3$ at 77 K and 273 K respectively. There is good agreement between the present work and that of Kuhn and Lewis [9], though it must be remembered that these authors used an enriched isotopic sample. We also note that our calculation of the perturber density is based on the measured pressure and the bath temperature; this gives results consistently higher (several per cent at 77 K) than those obtained for the same physical conditions using the method of Kuhn and Lewis.

The absence of temperature dependence noted by Kuhn and Lewis for this and other lines connecting to the $1s_2$ level is thus confirmed in the present work, but to higher precision. This result, taken together with the much smaller broadening found in transitions between levels not subject to the resonance interaction [9] leads us, following Lewis [15], to ascribe the entire broadening in 585.2 nm to the resonance interaction. The error involved in this assumption is difficult to assess, but is unlikely to exceed a few per cent.

In order to compare with theory, we take $k_{JJ'} = 1.53$, the value given by several authors [2, 3], and use the present results to find the f -value of the strong resonance transition. This gives $f_{736} = 0.155$, with an error of 0.002 due to the measurement, but the additional uncertainties arising from any non-resonant contribution to the observed broadening and approximations in the theory have to be taken into account. Using the usual simple relationship, our result implies a lifetime for the strong resonance level of 1.57 ns. Other experimental values are 1.54 ± 0.06 (beam-foil [16]), 1.65 ± 0.16 (cascade level crossing [17]) and 1.87 ± 0.18 ns (pulse excitation [18]). The evidence in neon therefore confirms the theoretical value of $k_{JJ'}$ to within about 10%, but more accurate values of the resonance oscillator strength would make the comparison more critical. There is some hope that such data may become available in the near future [17].

5. Conclusion

We have made measurements of self-broadening in the 585.2 nm line of neon; the broadening is ascribed to the resonance interaction in view of its temperature independence and the evidence from other lines that higher order effects are negligible. The broadening is consistent with the prediction of the currently accepted theory, though the comparatively poor accuracy with which the oscillator strength of the strong resonance transition in neon is known prevents a critical test.

However, the remarkably high precision now possible in the study of spectral line profiles as demonstrated in the present work opens new possibilities for theoretical advance. The study of small effects on the line profile due to the breakdown of approximations in the simple theory can be investigated, and experimental and theoretical work with this aim is in progress.

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