

*EFFECT OF COLLISIONS ON THE SHAPE OF THE NEON SPECTRAL LINES*

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A number of experiments are described in which impact self-broadening of spectral lines in neon is investigated. In particular, the Doppler contour of a series of neon lines corresponding to transitions between levels of the  $2p^5ns - 2p^53p$  configurations,  $n = 3-7$  and narrow resonances for the  $1.15 \mu$  ( $4s'[1/2]_1^0 - 3p'[1 1/2]_2$ ) and  $0.63 \mu$  ( $5s'[1/2]_1^0 - 3p'[1 1/2]_2$ ) lines are investigated. Lines which originate or terminate at resonance levels are mainly studied. The form of the interaction between the colliding atoms is determined by the analysis of the dependence of collision broadening on the principal quantum number of the resonance level. It is found that the predominant type of interaction between atoms excited to the  $n = 3$  level is resonance interaction. Exchange and short-range Coulomb forces are predominant in atoms with  $n = 6$  and  $n = 7$ . For  $n = 5$ , van der Waals and exchange interactions should be taken into account. All three types of interaction may be important for  $n = 4$ . The contribution of small angle scattering in elastic collisions to the broadening of narrow resonances is estimated. The effect exerted on the shape of the Doppler contour by the dependence of line broadening and shift on the velocity of the colliding atoms is analyzed.

**INTRODUCTION**

**I**MPORTANT information can be obtained on the interaction of colliding particles by a study of the shapes of the emission lines of atoms and molecules. A large number of experimental and theoretical researches have been devoted to such investigations (see the review<sup>[1]</sup>). Early investigations touched on the broadening and shift of the lines upon collision. They were usually carried out on the resonance absorption lines of vapors of the alkali metals.<sup>[1]</sup> In order to increase the accuracy of the measurements and to exclude the masking effect of Doppler broadening, the investigations were made at high gas pressures, when the broadening from collisions was at least comparable with the Doppler broadening.

An impact model has usually been employed for analysis of the line shape.<sup>[1,2]</sup> Under such experimental conditions, the angular scattering of the atoms does not play any role whatever in the line broadening and was therefore not taken into account, both in the theory of broadening and in the analysis of the experimental data. Problems of the scattering of atoms and molecules were based on the theory and practice of atomic beams,<sup>[3]</sup> where, naturally, questions connected with the emission line shape were not considered. A new situation arose when methods of laser spectroscopy came into use for the investigation of atomic collisions.<sup>[4-6]</sup> The majority of these methods were based on the study of the narrow resonances which form on the Doppler contour of the lines upon resonant interaction of the strong monochromatic field with the moving atoms. The high resolving power of the new methods has made it possible to make measurements with high accuracy at very low pressures, when the collision broadening is much smaller than the Doppler broadening and the impact model is clearly valid.

At the same time, analysis of the effect of the collisions on the shape of the narrow resonances has shown

that the role of collisions here is more many-sided than in the consideration of collision broadening of the Doppler contour. Along with quenching collisions that randomize the phase, the shapes of the narrow resonances can be affected by the angular scattering of the atoms in the collisions and by the dependence of the broadening and the line shift on the velocity of the atoms. In some cases, the resonance radiation is captured by an adjacent transition.

The new possibilities of high-resolution laser spectroscopy have stimulated theoretical investigations.<sup>[7-12]</sup> In the works of Rautian and Sobel'man,<sup>[7,8]</sup> the fundamental constants which characterize the frequencies of "strong" and "weak" elastic collisions, and collisions which randomize the phase of the radiation, were formally introduced into the density-matrix equations that describe the interaction of the atoms with the electromagnetic field. The researches of<sup>[14-16]</sup> have an important significance for understanding the physics of the collisions and the analysis of the experimental data. In these researches the impact parameters were determined from the microscopic theory.

**1. STATEMENT OF THE PROBLEM**

In spite of the broad possibilities of laser spectroscopy, the analysis of the experimental data with a view toward the establishment of the elementary processes has turned out to be difficult. At the same time, the obtaining of data which formally determine the broadening of the Lamb dip, for example, does not present any special difficulties. Comprehensive investigations and a comparison of the results of physically different experiments can give an answer as to the character of the collisions and their effect on the emission line shape. It is important to compare the experimental results obtained by different methods, and the direct methods of observation of some phenomenon or other take on special significance. In our opinion, the following experimental investigations are of interest.

1. The study of the shape of narrow resonances as a function of the pressure. This allows us to determine the pressure broadening of the lines and also the natural line width. Careful analysis of the shape of the narrow resonances can give direct information on the character of the elastic scattering of atoms in collisions.

2. Joint measurement of the collision broadening of the narrow resonances and of the Doppler contour of the line. A comparison of these data makes it possible to estimate the role of elastic collisions, inasmuch as the elastic collisions lead to the broadening of the narrow resonances and, in the absence of phase randomization, can lead to a narrowing of the Doppler contour.<sup>[7]</sup> Analysis of the Doppler profile of the line can give further information on the dependence of the broadening and line shift on the velocity of the atoms.

3. Investigation of "strong" collisions. By "strong" collisions, we mean collisions in which the velocity of the atoms changes within the limits of the mean thermal velocity. It must be expected that in many cases such collisions are connected with the resonance transfer of the excitation. The effect of these collisions on the shape of the line has been well studied theoretically.<sup>[14-16]</sup>

4. The investigation of the shift and the asymmetry of the line. The comparison of these data with data on impact broadening can indicate the relative significance of the role of collisions that randomize the phase of the radiation and also give information on the interaction potential of the colliding particles and the characteristics of the differential scattering cross section.<sup>[19]</sup>

5. The investigation of the temperature dependence of the broadening and shift. These investigations give important information on the type of interaction in the collisions.

6. The investigation of the dependence of the line broadening on the principal quantum number. Analysis of such a dependence allows us to determine the applicability range for any impact model. As is shown below, this dependence is very sensitive to the type of interaction of the colliding particles.

In the present work, we have generalized the results of experiments carried out by us in recent years, in which the self-broadening of the lines of neon has been studied. They include studies of the Doppler contour of a series of lines corresponding to transitions between the levels of the  $2p^5ns - 2p^33p$  configurations, and narrow resonances on the  $1.15 \mu$  line ( $4s'[1/2]_0^0 - 3p'[1/2]_2$ ) and the  $0.63 \mu$  line ( $5s'[1/2]_1^0 - 3p'[1/2]_2$ ). We centered our principal attention on the lines which begin or end on a resonance level. In this case one can expect that the main contribution to the line broadening will be made by processes of resonance transfer of the excitation. Our investigations included five resonance levels with principal quantum numbers 3-7. The results of theoretical researches<sup>[14-16]</sup> make it possible to calculate the contribution of resonance collisions to the line broadening. A comparison of the theoretical results with the experimental data make it possible to ascertain the region of applicability of the model of resonance collisions as a function of the value of the principal quantum number  $n$ . Preliminary results of our investigations<sup>[20]</sup> have already shown significant differences in the line broadening for large  $n$  in comparison with the results which theory predicts, and also a difference

in the broadening of the resonances and the Doppler contour. The present work is devoted to a detailed exposition and discussion of these results.

## 2. EXPERIMENTAL RESULTS

1. Choice of the transitions. About ten neon lines were investigated. These correspond to transitions between the  $2p^5ns$  and  $2p^33p$  levels (Fig. 1). In what follows we shall for simplicity use the generally accepted Paschen designations of the levels. We have studied the strongest lines, which are connected with the resonance levels. In addition, for the  $1s - 2p$  transitions, we carried out experiments on the collision broadening of lines which end on the metastable levels  $1s_3$  and  $1s_5$ . The choice of the specified lines was connected with the possibilities of the experimental apparatus. In working with the Fabry-Perot interferometer, we attempted to use lines lying in a narrow range of wavelengths. The pressure broadening of all lines, with the exception of the  $1.15 \mu$  line, was studied using the Doppler contour of the emission or absorption by means of the Fabry-Perot interferometer. Furthermore, pressure broadening of the  $0.63$  and  $1.15 \mu$  lines was studied by means of laser spectroscopy.

2. Investigation of collision broadening of the Doppler contour of the lines. Under the assumption of statistical independence of the broadening due to collisions and the Doppler effect of the line contours, the radiation has the Voigt form:

$$I(\Omega) = I_0 \frac{a}{\pi} \int_{-\infty}^{\infty} \frac{e^{-y^2} dy}{(\Omega - y)^2 + a^2}, \quad (1)$$

where  $\Omega = (\omega_0 - \omega)/\Delta\omega_D$ ;  $a = \Gamma/\Delta\omega_D$ ;  $\omega_0$  is the frequency corresponding to the line center;  $2\Gamma$  is the dispersion width, consisting of the natural and collision widths of the lines;  $\Delta\omega_D$  is the Doppler parameter;  $\Delta\omega_D = kv$ ,  $k = 2\pi/\lambda$  is the wave number;  $v = (2kT/m)^{1/2}$  is the mean thermal velocity.

The integral (1) is well known and tabulated. For  $a \ll 1$ , one can use the expansions of (1) for small and for large  $\Omega$ , which are given in the book of Mitchell and

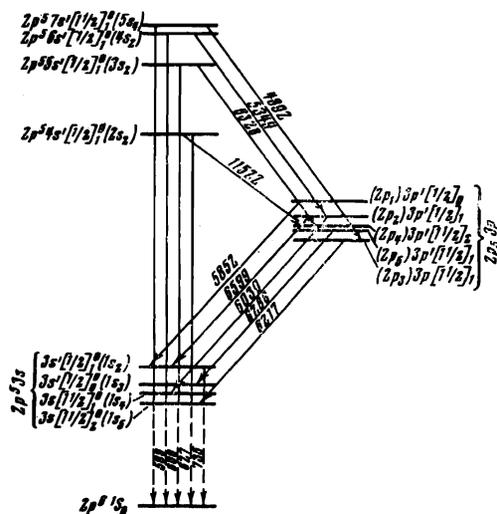


FIG. 1. Scheme of the levels and studied transitions of neon. The levels should be read: upper left—7s (and not 7s'), lower right—2p (and not 2p<sub>3</sub>).

Zemansky.<sup>[21]</sup> A whole series of methods is known, according to which one can carry out the separation of its Gaussian and dispersion parts. Thus, measuring the total width of the contour at the half maximum,  $\Delta\omega_{1/2}$ , and knowing the ratio  $\Delta\omega_D/\Delta\omega_{1/2}$  from the tables given in the work of Davies and Vaughan,<sup>[22]</sup> we can find the ratio  $\Gamma/\Delta\omega_D$  or  $\Gamma/\Delta\omega_{1/2}$ .

In the study of strong transitions between the contours, the spontaneous emission, line can be distorted as a consequence of radiation reabsorption.<sup>[23]</sup> The reabsorption lines, which are not so important at small absorptions in a discharge tube, can lead to a significant difference between the studied contour and the Voigt contour at appreciable absorptions. Large absorption is observed for lines which end on metastable levels. It is virtually impossible to avoid the effect of reabsorption here. In such cases, the absorption method is more reliable. The method of measurement of the shape of the absorption line has been described previously.<sup>[24]</sup> The experimental setup consisted of two discharge tubes, an optical circuit, a photodetector and an electronic recording system (Fig. 2). The tube 1 (diameter 2.2 mm, length 60 mm) served as the source of the reference signal. Tube 2 (diameter 10 mm) serves as the absorption cell. The construction of the tube permitted us to change the discharge length from 40 to 170 mm, because of which we could choose the necessary absorption level. A liquid-nitrogen cooled FEU-15 photomultiplier served as the photodetector. The optical system consisted of a tunable Fabry-Perot interferometer, a set of diaphragms, lenses, an SPM-1 monochromator and a PGS-2 spectrograph. The recording system consisted of a tuned amplifier, a synchronous detector, and an x-y recorder.

The construction of the Fabry-Perot interferometer has been described by Lisitsin.<sup>[25]</sup> The mirrors had a silver coating and were mounted on a piezoceramic at a distance of 22.5 mm from one another. The reflection coefficient for  $\lambda = 0.63 \mu$  was equal to 0.91 and did not change by more than 0.8% within the range  $\Delta\lambda = \pm 500 \text{ \AA}$ . The scanning of the interferometer and the horizontal sweep of the recorder were effected simultaneously by a sawtooth voltage of amplitude 400 V. The sweep time could be changed within the range 10–100 sec. The apparatus function of the interferometer was recorded by means of a single-mode He-Ne laser, and its width at a wavelength  $0.63 \mu$  amounted to 250 MHz. Measurement of the width of the neon line in the  $1s - 2p$  transitions was made from the recording of the contours of the ab-

sorption lines at different pressures of the gas under study. The pressure in the absorption cell changed from 0.2 to 12 Torr. The separation of the Gaussian and dispersion components of the contours was made by means of the tables from<sup>[22]</sup>. The Doppler width was determined from the temperature of the discharge. The neon lines with wavelengths 5852, 6599, 6266, 6217 and 6030 Å were investigated by this method. All these lines terminate on levels having the principal quantum number 3. The 5852 and 6599 Å lines have the resonance lower level  $1s_2$ .

A characteristic feature of the results obtained is the sharp difference in the value of the broadening for lines which terminate on a resonance level and on metastable levels. The width of the 5852 Å line amounts to  $2\theta\Gamma/\theta p = 70 \pm 10 \text{ MHz/Torr}$ , at a discharge current of 10 mA and a gas temperature of 300°K, while the width of the lines which terminate on the  $1s_3, 1s_4, 1s_5$  levels is less than 10 MHz/Torr. The results of our measurements are in excellent agreement with the results obtained by Kuhn and Levis.<sup>[26]</sup> The broadening of the 5852 Å line measured in this research amounted to  $66 \pm 4.6 \text{ MHz/Torr}$ , while the width of the lines that terminate on the  $1s_3, 1s_4, 1s_5$  levels was in the range 6–12 MHz/Torr.

The investigation of the broadening of lines that begin from a highly excited level with principal quantum numbers 5, 6, and 7 was made by recording the spontaneous emission contours. The lines 6328, 5349, 4892 and 5433 Å were studied in this way. The diameter of discharge tube was 4.5 mm, the discharge current 25 mA, the gas temperature  $T = 320^\circ\text{K}$ . In the study of the 5842 Å line by this method, the discharge current was 2 mA, the tube diameter 10 mm, and  $T = 300^\circ\text{K}$ . To eliminate the effect of reabsorption on the shape of the spontaneous emission lines (this applied especially to  $\lambda = 5862 \text{ \AA}$ ), the discharge conditions in the tube studied were so chosen that the absorption on the studied lines did not exceed 3%. In order to reduce to a minimum the effect of reabsorption on the results of the study of collision broadening, all the measurements for each line at the various gas pressures were made at a constant value of the absorption at the center of the line. This was achieved by a small variation of the discharge current. In total, some 1000 interferograms were obtained. For the reduction of the experimental curves, we used the method proposed by Ballik.<sup>[27]</sup> The ratio of the width of the line contour to the distance between orders of the interferometer was found from the experimental recordings, as well as the ratio of the intensities at the center of the line and at the minimum of the signal. By using these data, the Lorentz and Doppler line widths were obtained from the curves given in<sup>[27]</sup>.

The broadening of the 5852 Å line, determined from the contour of the spontaneous emission line, agreed within the limits of accuracy with the value obtained by use of the absorption method. The test for the absence of significant systematic errors in our results lay in the fact that the values found for the natural line widths of the lines 5852 Å and 6328 Å (these data were obtained by extrapolation of the curve of the dependence of the dispersion part of the contour to zero pressure) agreed well with the data from other measurements. The experimental recordings of the contours of spon-

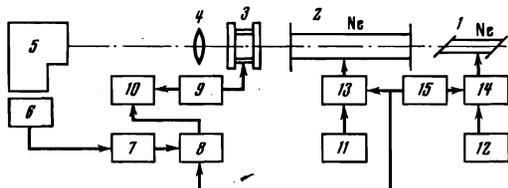


FIG. 2. Circuit arrangement for the recording of the shape of the lines of absorption and spontaneous emission: 1, 2—discharge tubes with neon, 3—Fabry-Perot interferometer, 4—lens, 5—PGS-2 spectrograph, SPM-1 monochromator, 6—Photoreceiver, 7—resonance amplifier, 8—synchronous detector, 9—sawtooth voltage generator, 10—recorder, 11, 12—voltage sources, 13, 14—current modulators, 15—sound generator.

taneous emission of the lines 5852, 6328, 5433 Å were treated by the method which was used by us for analysis of the contours of the absorption lines with use of the tables of Davies and Vaughan.<sup>[22]</sup>

We emphasize that in this case we are directly measuring the broadening of the Doppler contour at the half maximum. For the 5852 Å line, the results of treatment by the Ballik method<sup>[27]</sup> and by the use of the tables from<sup>[22]</sup> were in agreement. But for the lines 6328 and 5433 Å, the results of treatment of the data by these two methods differ somewhat.

3. Investigation of the collision broadening of the line 1.15. The first results on the collision broadening of resonances on lines from the 2s levels were obtained in the investigation of the shape of the Lamb dip at  $\lambda = 1.15 \mu$  in an He-Ne laser.<sup>[5]</sup> Lisitsyn and one of the authors<sup>[26]</sup> measured the broadening of the 1.5  $\mu$  line ( $2s_2 - 2p_1$ ) by means of the internal absorption cell. Data had been obtained previously<sup>[12,29]</sup> on the broadening of the Lamb dip in a neon laser. These data were used for the analysis of the effect of the capture of resonance radiation on the shape of narrow resonances. Recently, the broadening of the 1.15  $\mu$  line was measured by Holt.<sup>[30]</sup> The data of<sup>[12,28,29]</sup> differ by one order of magnitude from the data in<sup>[5,30]</sup>. In this connection, we shall set forth in detail the method of our experiment and, in contrast with previous researches<sup>[12,29]</sup>, we shall concentrate our main attention on the problems of collision broadening of the Lamb dip in a neon laser ( $\lambda = 1.15 \mu$ ).

Our studies showed that in tubes of small diameter ( $\sim 2$  mm) and at small discharge currents, generation is possible at neon pressures to 2 Torr. Such a range of pressures is sufficient to study the effect of the collisions on the shape of the Lamb dip. For analysis of the dependence of the generation power on the frequency at  $\lambda = 1.15 \mu$ , it is necessary to take into account the effect of capture of the resonance radiation and of "strong collisions,"<sup>[12,29]</sup> which leads to homogeneity of saturation and by the same token decreases the depth of the dip. The dependence of the generation power on the frequency, with account of these effects, is given by the expression<sup>[12]</sup>

$$P(\Omega) \propto \frac{\eta - \exp[(\Omega/k\bar{v})^2]}{1 + \Gamma^2/(\Gamma^2 + \Omega^2) + \alpha}. \quad (2)$$

Here  $\Omega$  is the frequency detuning relative to the center of the line,  $\eta$  is the ratio of the unsaturated gain at the center of the line to the losses, and  $\alpha$  is a parameter that takes into account the homogeneity of the saturation and depends on the relaxation constants of the upper and lower levels of the transition. The expression (2) is valid for  $\eta - 1 \ll 1$  and  $\Gamma \ll k\bar{v}$ .

Reduction of the experimental data reduces to the selection of such values of the parameters entering into Eq. (2) that the computed curve coincides with the experimental. For convenience in the analysis of the experimental curves, the expression (2) was transformed to

$$P = A \frac{\eta - \exp[(\Omega/k\bar{v})^2]}{1 + a\Gamma^2/(\Omega^2 + \Gamma^2)}. \quad (3)$$

Formally, Eq. (3) is identical with the expression used by Javan and Szoke<sup>[5]</sup> for analysis of the shape of the Lamb dip in an He-Ne laser. Our  $\Gamma$  and  $a\Gamma$  correspond

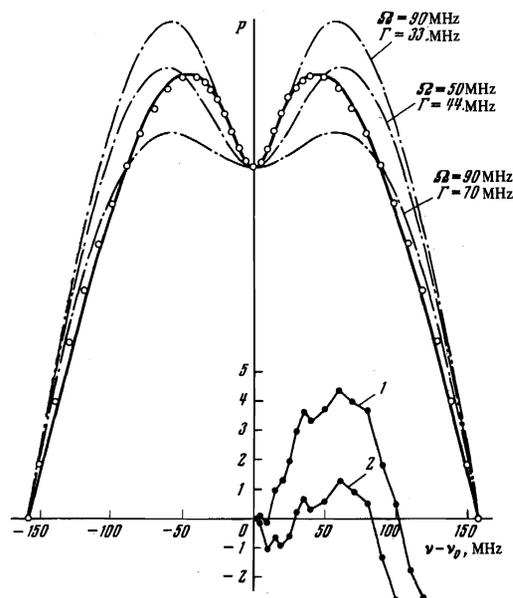


FIG. 3. Experimental and calculated dependences of the generated power on the frequency of the laser in neon ( $\lambda = 1.15 \mu$ ). The continuous curve is the experimental, neon pressure 0.7 Torr, discharge current 6.4 mA, open circles—calculated values for the parameters determined from the experimental curve,  $a = 0.5$ ,  $\Gamma = 23$  MHz,  $k\bar{v} = 435$  MHz,  $\eta = 1.14$ , the dashed curves are calculated for the parameters  $a = 1$ ,  $k\bar{v} = 435$  MHz,  $\eta = 1.14$  (for the upper curve,  $\Omega = 20$  MHz). Below are drawn the deviations the calculated values of  $P_D$  from the exponential  $P_e$ ,  $((P_D - P_e)/P_e) \cdot 100\%$ ; 1—calculated values correspond to the parameters of the zeroth approximation ( $a^{(0)} = 0.45$ ,  $\Gamma^{(0)} = 24.5$  MHz), 2—calculated values correspond to the parameters of the first approximation ( $a^{(1)} = 0.5$ ,  $\Gamma^{(1)} = 23$  MHz).

to the constants  $\gamma'_{ab}$  and  $\gamma_{ab}$  from<sup>[5]</sup>; however, they have an entirely different physical meaning.<sup>[12,29]</sup>

Five parameters enter into (3); these must be determined from the experimental curves. The parameter  $A$  is a scaling coefficient, which does not depend on the frequency and is not of interest. We have determined the Doppler parameter from independent measurements. For this purpose, we established the generation threshold by varying the discharge current in the amplifier tube of the laser. Then, by varying the length of the discharge and establishing the previous current, we measured the frequency detuning  $\Omega_R$  for which quenching of the generation takes place. The ratio of the corresponding lengths of the discharge is equal to the ratio of the amplification to the losses  $\eta = l_2/l_1$ . Then  $k\bar{v} = \Omega_D \ln^{-1/2}(l_2/l_1)$ . The line width measured in this fashion amounted to  $k\bar{v} = 440 \pm 10$  MHz and the corresponding temperature was  $T = 306 \pm 14^\circ$  K.

The frequency scale was determined from the distance between the orders of the laser interferometer, which was equal to 765 MHz in our case. Two or three orders were recorded to determine the frequency scale.

The parameters  $a$  and  $\Gamma$  were chosen by the method of successive approximations.<sup>1)</sup> In Fig. 3, the solid line is the experimental curve and the points are the results of calculation from Eq. (3) for the parameters determined from the experimental curve. The deviation of

<sup>1)</sup>The method of calculation of the parameters  $a$  and  $\Gamma$  is described in our preprint. [19]

the calculated values from the experimental close to the dip did not exceed 2%. The dependence of the relative error on the detuning for two choices of the parameters  $a$  and  $\Gamma$  is shown on this same drawing, at the bottom. Curve 1 corresponds to the zeroth approximation of the parameters  $a$  and  $\Gamma$ , and curve 2 to the first approximation. The relative error increases somewhat near the threshold. Here the error in the measurements of  $\eta$  and  $k\nu$  has the strongest effect.

For comparison, the figure shows curves calculated from the formula of Lamb,<sup>[31]</sup> which did not take into account the homogeneity of the saturation ( $a = 1$ ). Three values of  $\Gamma$  correspond to the three curves; these were determined for detuning by 20, 50 and 90 MHz. As we see, it has not been possible to obtain agreement between the calculated and experimental curves for a single value of the parameter  $\Gamma$ . Consequently, the shape of the Lamb dip in the laser at  $\lambda = 1.15 \mu$  cannot be described by the Lamb formula without account of the homogeneity of the saturation.

The experimental setup was developed in accord with the analysis just given. The laser resonator had a length of 19 cm and was made up of plane and spherical mirrors. Thanks to the excellent elimination of vibrations and to the stability of the discharge, it was possible to make high-quality recordings with very low noise levels (less than 1%) at a comparatively slow variation of the laser frequency. The laser frequency was changed by displacement of one of the mirrors, which was attached to a piezoceramic cylinder. A voltage was applied to this cylinder which changed linearly with time. The discharge tube, with a diameter of 2 mm, had two sections of length 89 and 118 mm. The level of the excess of the gain over the threshold was usually established within the range 1.15–1.1. No dependence of the width of the dip on the discharge current was observed.

Reduction of the results of the measurements gave for the half-width of the line in the range of pressures 0.3–1.6 Torr the value  $\Gamma = (17 \pm 1) + (6.6 \pm 1)p$  MHz, where  $p$  is the neon pressure (in Torr). Similar results on the broadening of the  $1.15 \mu$  line in neon were obtained in another of our experiments,<sup>[32]</sup> which consisted of the study of the shape of the narrow peak in the  $1.15 \mu$  gain line, produced by a strong field acting on the adjacent transition  $2s_2 = 2p_1$ ,  $\lambda = 1.52 \mu$ . At the same time, our results differ by more than an order of magnitude from the results obtained in the work of Holt,<sup>[30]</sup> where the shape of the Lamb dip was also studied in a laser operating with pure neon. The broadening in pure neon observed in this case amounted to  $107 \pm 35$  MHz/Torr. Such a difference in the results could evidently be explained by the fact that Holt<sup>[30]</sup> was used for the treatment of the experimental curves a theoretical model which did not take into account the effect of capture of radiation on the frequency dependence of the generation power. The line width was determined from the agreement of the experimental and theoretical curves of the dependence of the ratio of the maximum power to the power at the center of the dip on the excitation parameter. Calculations showed that this ratio also depends in large degree on the capture parameter.

As we have already pointed out, our data differ from the results obtained by Javan and Szoke,<sup>[5]</sup> where the Lamb dip was studied in a laser operating on the He-Ne

mixture. It is possible that this difference is due to the specific effect of elastic scattering in collisions with neon atoms in the Ne-He mixture in the absence of line-broadening collisions with He atoms. We shall discuss this in Sec. 3.

4. Investigation of collision broadening of the  $0.63 \mu$  line. A large number of researches have been devoted to this problem. However, in most cases, the investigations were carried out for He-Ne mixtures, where the broadening collisions of neon with the helium atoms largely mask the collision broadening with atoms of neon. Moreover, by virtue of the specifics of the helium-neon laser, studies on collisions with neon atoms can be carried out over a very limited range of pressures. Only comparatively recently have studies of collision broadening of narrow resonances been carried out in pure neon in a laser with nonlinear absorption<sup>[33,34]</sup> and for a nonlinear interaction of the radiation of two lasers with an absorbing medium in the inner cell.<sup>[35,40]</sup> The results of the latter measurements differ both from one another and from the results of measurement of collision broadening in collisions with atoms of neon in the He-Ne mixture.<sup>[36]</sup> The method of internal saturation of the absorption cell with neon, in spite of a number of virtues, also has notable shortcomings, which prevent the obtaining of data with high accuracy. As a basic weakness, we can mention the following: the range of studied pressures is limited, the shape of the peak depends on the parameters of the laser in a complicated way, which makes difficult the treatment of the results, the shift in the gain line leads to additional distortions of the peak of generation power, it is difficult to determine the parameter of saturation in the absorbing medium.

In this connection, we undertook new detailed investigations of impact self-broadening of the narrow resonances of the neon line  $\lambda = 0.63 \mu$ , using the effect of saturation in an external absorption cell.<sup>[37]</sup> The method used in the present work is based on the investigation of narrow resonances which develop in the nonlinear interaction of strong and weak opposing waves in a magnetic field.<sup>[38]</sup> The theory of the method has been developed by Baklanov and one of the authors.<sup>[39]</sup> For strong fields, the shape of the line depends on the value of the field and on the ratio of the constants of the level relaxation. For differing constants of the level relaxation and weak fields, the absorption coefficient of a weak opposing wave is given by the expression<sup>[39]</sup>

$$K = K_0 \left( 1 - \frac{\kappa}{1 + \kappa + \sqrt{1 + \kappa}} \frac{\Gamma_1^2}{\Omega^2 + \Gamma_1^2} \right) \exp \left[ - \left( \frac{\Omega}{k\nu} \right)^2 \right] \quad (4)$$

$$\Gamma_1 = \frac{1}{2} \Gamma (1 + \sqrt{1 + \kappa}). \quad (5)$$

Here  $K$  is the absorption coefficient of the weak field in the presence of the strong one,  $K_0$  is the unsaturated absorption coefficient at the center of the line, and  $\kappa$  is the parameter of inhomogeneous saturation.

In this research our principal problem lay in the determination of the line width; therefore, all the basic measurements were made in relatively weak fields, when the line shape can be described by Eq. (4). The measurements were performed on the apparatus described previously<sup>[38,40]</sup> and used for the stabilization of the frequency of a helium-neon laser with  $\lambda = 0.63 \mu$ .

Figure 4 shows a variant of the apparatus, used for the study of the line shape. The radiation of a powerful tunable single-frequency laser,<sup>[41]</sup> which has linear polarization, was passed through a polarization prism, transformed by a quarter-wave plate into radiation with circular polarization and focused by a lens on the center of the outer absorption cell (length 50 mm, diameter 2.5 mm). A weakly reflecting mirror placed behind the absorbing cell produced a beam of weak intensity which passed through the cell in the reverse direction and was brought out by the polarization prism to the photodetector. Simultaneously, the polarization prism, in combination with the quarter-wave plate, guaranteed decoupling between the mirror and the resonator of the laser. Part of the radiation which passes through the cell in the forward direction is directed onto a second photodetector, connected in opposition to the first to a common load. By adjusting the signals at the output of the photodetectors such that in the absence of absorption they were equal and opposite in phase, we could decrease the noise in the recording system by more than an order of magnitude. The difference in the readings of these photodetectors in the presence of absorption corresponds to absorption of the studied opposing wave. All the measurements were made for the center of the beam.

The signal from the load was fed to a recording system that consisted of a selective amplifier, a synchronous detector and an automatic recorder. While varying the laser frequency, we plotted the shape of the absorption line of the weak wave in the presence of the strong one and also the dispersion curve of the absorption line (Fig. 5). The dispersion curve was obtained with modulation of the magnetic field in the cell. In a longitudinal magnetic field, the absorption line was split into two Zeeman components with right and left circular polarizations. The radiation field in the cell has a circular polarization and interacts only with one of the Zeeman components of the line. Scanning of the positions of the components by a magnetic field leads to modulation of the absorption of a weak field wave. At small scanning amplitudes, the dispersion curve represents the first derivative of the absorption line, given by Eq. (4). As a consequence of this, the distance between the maxima on the curve of Fig. 5 is equal to  $2\Delta\Omega = 2\Gamma_1/\sqrt{3}$ . Using this relation, we determined  $\Gamma_1$  from curves of the type of Fig. 5.

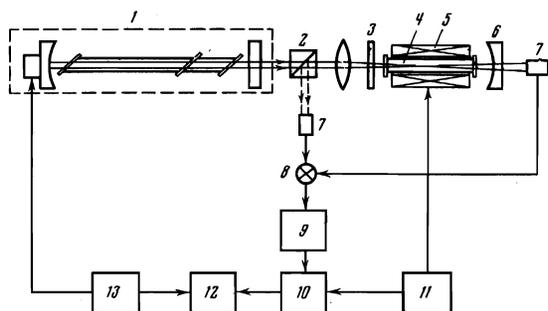


FIG. 4. Scheme of experimental arrangement: 1—high-power single-mode laser,  $\lambda = 0.63\mu$ , 2—polarization prism, 3—quarterwave plate, 4—external absorption cell with neon, 5—solenoid, 6—weakly reflecting mirror, 7—photodetector, 8—common load (sumerator), 9—filter amplifier, 10—synchronous detector, 11—audio generator, 12—recording instrument, 13—generator of a linearly varying voltage.

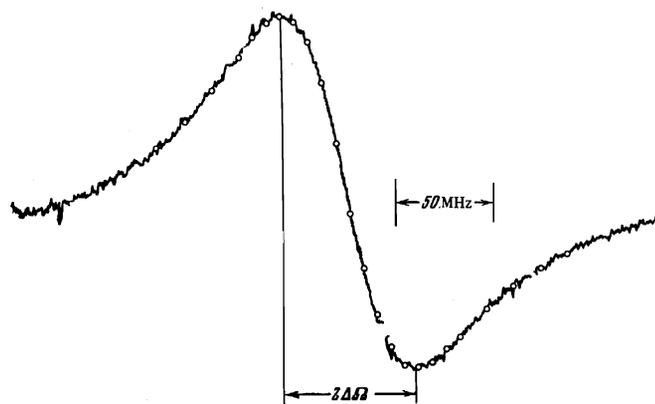


FIG. 5. Experimental plot of the dispersion curve of the absorption line of a weak wave. The points of the curve correspond to the computed values, calculated for the parameter  $\Gamma$  determined from this curve.  $p_{Ne} = 1$  Torr,  $I_{dis} = 20$  mA.

The strong field of the forward wave gives rise to dip broadening, which must be taken into account in the determination of the Lorentz width. This has been done by two different methods. In the first method the dependence of  $\Gamma$  on the intensity of the radiation passing through the cell was established for each pressure, and the parameter  $\Gamma$  was determined by extrapolation to zero field. Inasmuch as the method of measurement used was based on the saturation effect, it was impossible to reduce the field in the cell to a very small value. Therefore, in obtaining the true value of the width, extrapolated to zero field, the form of the approximating function is of importance. We used the relation (5) as the approximating function. When the dependence is approximated by a straight line, the sum of the squares of the errors exceeds the value obtained by using the function (5).

The second method of determination of  $\Gamma$  was as follows. The absorption coefficient was measured in the cell after one pass through the center of the line for the strong field (the saturated absorption coefficient  $K_S$ ) and for the weak field (the unsaturated absorption coefficient  $K_0$ ). In the measurement of  $K_0$ , the radiation intensity was attenuated by a factor greater than 100, so that one could assume approximately that  $\kappa \approx 0$  for the weak field. Using the known relation  $K_S = K_0/\sqrt{1 + \kappa}$ , we determined the saturation parameter for the strong field. We determined  $\Gamma$  by substituting the resultant value of  $\Gamma$  in Eq. (5).

The values of  $\Gamma$  found by the two different methods were approximately the same at each pressure. The results given in Fig. 6 correspond to a discharge current of 20 mA in the cell. The approximating straight line was constructed by the least squares method from the total results obtained by the different methods. Similar measurements were made for discharge currents of 30, 40, 50 mA. A small increase in the line width with the discharge current was observed with a rate of about  $\partial\Gamma/\partial I = (0.15-0.17)$  MHz/mA. The slope of the curve of the dependence of  $\Gamma$  on pressures remained constant here within the limits of accuracy of the measurements and was equal to  $\partial\Gamma/\partial p = 24 \pm 2$  MHz/Torr. (The external cell was water-cooled. The gas temperature in the discharge channel was  $320 \pm 10^\circ$  K and changed little with the discharge current.) The natural line width  $\Gamma_0$

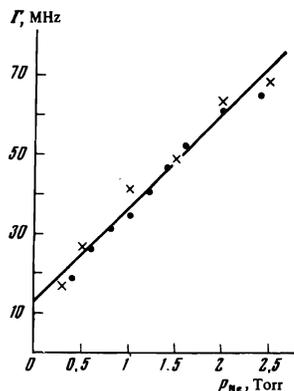


FIG. 6

FIG. 6. Impact self-broadening for the  $0.63\mu$  line of neon: X—values obtained by extrapolation to zero field, ●—values calculated from the measured saturation parameter.

FIG. 7. Dependence of the broadening of the neon line on the principal quantum number  $n$ . 1—measured values, 2—calculated values of resonance broadening.

$= 11 \pm 3$  MHz was obtained by extrapolations to zero pressure and current.

For comparison, the results of other experiments are given in Table I. As we see, there is a great deal of scatter of the value of the broadening, and the scatter is large even for the data of the same authors.<sup>[34,42,43]</sup> These authors used slightly different methods of treatment of the data on the generation peak in the laser with nonlinear absorption. Both the analysis of the data of the treatment of the Lamb dip in a neon laser ( $\lambda = 1.15 \mu$ ) and the present case demonstrate the importance of the choice of the model which is used as the basis of the experimental data reduction.

The results of our measurements differ most strongly from the data of Mikhnenko and Protsenko.<sup>[36]</sup> Their results on the broadening of the  $0.63\text{-}\mu$  line by neon were obtained in a study of broadening in the He-Ne mixture by investigating the dependence of the broadening on the ratio of the components of the system. Their data also differ significantly from the results of other measurements carried out in a discharge of pure neon. Our results are closest to the results of<sup>[33,42,44]</sup>.

### 3. DISCUSSION OF THE RESULTS

1. The results of all the measurements are shown in Table II. Figure 7 shows the dependence of the collision broadening on the principal quantum number  $n$  of a single level. Values are given for the lines  $5852 \text{ \AA}$  ( $n = 3$ ),  $11522 \text{ \AA}$  ( $n = 4$ ),  $6328 \text{ \AA}$  ( $n = 5$ ),  $5349 \text{ \AA}$  ( $n = 6$ ), and  $4892 \text{ \AA}$  ( $n = 7$ ). The characteristic feature of the resultant dependence is the presence of a clearly marked minimum corresponding to  $n = 4$ .

In order to understand this dependence, we consider the character of the interaction of the colliding atoms. We estimate the contribution of the resonance collisions to the line broadening. For transitions between two ex-

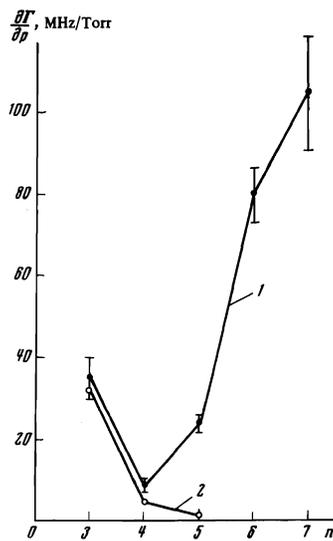


FIG. 7

Table I. Self-broadening of the  $6328 \text{ \AA}$  line in  $\text{Ne}^{20}$ 

Experimental research	MHz/Torr	Method of measurement
V. N. Lisitsyn and V. P. Chebotaev [33]	$15 \pm 10$	Peak power in the laser with internal absorption cell
V. M. Tatarsenko et al. [34]	$7 \pm 1.5$	Peak power in the laser with internal absorption cell
The same [42]	$23 \pm 2.5$	
The same [43]	$35 \pm 2.5$	
C. V. Shank and S. E. Schwarz [35]	$\sim 41$	Nonlinear interaction of the radiation of two lasers with the absorbing medium in the external cell. Single-direction waves
P. W. Smith and T. Hänsch [44]	$\sim 25$	Countering waves
G. A. Mikhnenko and E. D. Protsenko	$70 \pm 10$	Lamb dip in He-Ne laser for variation of the He-Ne ratio
Our measurements	$24 \pm 2$	

Table II

$\lambda, \text{ \AA}$	Transition		MHz/Torr	Method of measurement
	Racah notation	Paschen notation		
5852	$3s'[1/2]_0^0 - 3p'[1/2]_0$	$1s_2 - 2p_1$	$35 \pm 5$	Absorption
			$41 \pm 5$	Spontaneous emission Reduction of interferograms by method of [22]
6599	$3s'[1/2]_1^0 - 3p'[1/2]_1$	$1s_2 - 2p_2$	$40 \pm 5$	Spontaneous emission Reduction of interferograms by method of [27]
			$40 \pm 7$	Absorption
6266	$3s'[1/2]_0^0 - 3p'[1/2]_1$	$1s_2 - 2p_5$	$0 \pm 5$	Absorption
6030	$3s[1/2]_1^0 - 3p[1/2]_1$	$1s_4 - 2p_2$	$0 \pm 5$	Absorption
6247	$3s[1/2]_0^0 - 3p[1/2]_1$	$1s_5 - 2p_7$	$0 \pm 5$	Absorption
11522	$4s'[1/2]_1^0 - 3p'[1/2]_2$	$2s_2 - 2p_4$	$8,8 \pm 1$	Lamb dip
			$24 \pm 2$	Dip in absorption line
6328	$5s'[1/2]_1^0 - 3p'[1/2]_2$	$3s_2 - 2p_4$	$30 \pm 5$	Spontaneous emission Reduction of interferograms by method of [22]
			$42 \pm 5$	Spontaneous emission Reduction of interferograms by method of [27]
5433	$5s'[1/2]_1^0 - 3p'[1/2]_1$	$3s_2 - 2p_{10}$	$34 \pm 7$	Spontaneous emission Reduction of interferograms by method of [22]
			$45 \pm 10$	Spontaneous emission Reduction of interferograms by method of [27]
5349	$6s'[1/2]_1^0 - 3p'[1/2]_1$	$4s_2 - 2p_2$	$80 \pm 7$	Spontaneous emission Reduction of interferograms by method of [27]
4892	$7s[1/2]_1^0 - 3p[1/2]_1$	$5s_4 - 2p_7$	$105 \pm 35$	Spontaneous emission Reduction of interferograms by method of [27]

cited levels, one of which is optically coupled to the ground state, while the moments of the ground and excited states are equal to zero and unity, we have the relation<sup>[16]</sup>

$$\Gamma = 5.7 N_0 \bar{\lambda}^3 A_0 = 3.8 N_0 r_0 c \bar{\lambda} f, \quad (6)$$

where  $2\Gamma$  is the resonance collision width at the half-maximum;  $N_0$  is the concentration of atoms in the ground state;  $2\pi\lambda$  is the wavelength corresponding to the transition to the ground state,  $A_0$  is the probability of a radiative transition to the ground state,  $f$  is the oscillator strength of the ground state, and  $r_0$  is the classical radius of the electron.

It is known that the oscillator strength falls off rapidly with increase in the principal quantum number, approximately in proportion to  $1/n^3$ , which leads to a rapid decrease in the contribution of the resonance collisions to the line broadening. In Fig. 7, curve 2 shows the change in the resonance broadening with increase in  $n$ . The broadening was calculated from Eq. (6). Here the following values of parameters were used:  $f(1s_2 - 1S_0) = 0.16 \pm 0.014$ ,<sup>[45]</sup>  $A(2s_2 - 1S_0) = 1.52$

$\times 10^8 \text{ sec}^{-1}$ ;  $A(3s_2 - {}^1S_0) = 0.55 \times 10^8 \text{ sec}^{-1}$ . We determined the parameters  $A(2s_2 - {}^1S_0)$  and  $A(3s_2 - {}^1S_0)$  by using the known values of the natural broadening of the lines for  $\lambda = 1.15 \mu$  and  $\lambda = 0.63 \mu$  and the published data for the rate of radiative decay of the levels:  $A(2p_4) = 0.523 \times 10^8 \text{ sec}^{-1}$ ,<sup>[46]</sup>  $A(2s_2) - A(2s_2 - {}^1S_0) = 1.0 \times 10^7 \text{ sec}^{-1}$ ,<sup>[46]</sup> and  $A(3s_2) - A(3s_2 - {}^1S_0) = 3.1 \times 10^7 \text{ sec}^{-1}$ .<sup>[47]</sup>

The measured and calculated values of the broadening for the quantum number 3 (level  $1s_2$ ) are identical. This indicates that the resonance broadening of the line is dominant in this case. The broadening of the lines which terminate on the metastable levels  $1s_3$  and  $1s_5$  is at least an order of magnitude smaller.

The contribution of resonance broadening for the  $1.15 \mu$  line, which corresponds to the transition from the level with quantum number 4, amounts to about half the total value of the broadening, in accord with the data of Fig. 7. The additional broadening evidently should be attributed to excitation of the  $2p_4$  level.

For the most highly excited levels with quantum numbers 5 and 6, the value of the resonance broadening becomes small and the impact broadening of the lines increases appreciably. The nonresonant interaction forces begin to play an important role. At large distances between the colliding atoms, the Van der Waals attractive force act on them:  $U(R) \sim -C_6/R^6$ . At small distances, when overlap of the electron shells of the colliding atoms occurs, short-range forces begin to play the principal role. These fall off according to an exponential law. They result from a combination of the Coulomb forces, averaged over the charge distribution, with the forces which appear as a result of the exchange of electrons between the atoms.

We now estimate the order of the cross sections for the Van der Waals and exchange interactions. The Van der Waals interaction constant  $C_6$  can be estimated by using the expression<sup>[48]</sup>

$$C_6 = e^2 \beta (\bar{r}_k^2 - \bar{r}_i^2) \text{erg-cm}^6 \quad (7)$$

where  $\beta$  is the polarizability of the exciting atom,  $\bar{r}_k^2$  and  $\bar{r}_i^2$  are the mean square values of the coordinate of the valence electron in the upper and lower states of the transition under study. The expression is valid for the case in which the energy interval between the excited levels  $k$  and  $i$  is much less than that between the ground state and the other levels of the perturbing atom. This condition is well satisfied for inert gases. To find  $r^2$ , we can use the approximate expression<sup>[49]</sup>

$$\bar{r}^2 = \frac{1}{2} a_0^2 (n^*)^2 [5(n^*)^2 + 1 - 3l(l+1)], \quad (8)$$

where  $n^*$  is the effective quantum number,  $l$  the orbital quantum number, and  $a_0$  the Bohr radius. The effective quantum number  $n^*$  was determined from the known numerical values of the neon terms.<sup>[50]</sup> The polarizability for neon was taken to be  $0.41 \times 10^{-24} \text{ cm}^3$ .<sup>[51]</sup> The found values of  $C_6$  were used for the calculation of the impact line broadening due to Van der Waals interaction:

$$2\Gamma_B = 1.3C_6^{1/3} \bar{v}^{1/2} N_0. \quad (9)$$

The dependence of  $\Gamma_V$  on  $n^*$  can be obtained from the relations (7)–(9). For large quantum numbers  $n^*$  we have  $\Gamma_V \propto (n^*)^{1.6}$ .

The calculated dependence of  $\partial\Gamma_V/\partial p$  on the effective principal quantum number is shown in Fig. 8 by the dashed line. The points denote the measured values of the broadening of the lines. From a comparison of the calculated values of  $\partial\Gamma_V/\partial p$  with the experimental data, it is seen that the measured value of the broadening increases with increase in the quantum number much more rapidly than the broadening due to the Van der Waals interaction. From this it can be concluded that for large  $n$  the collision broadening is due to an interaction that is different from the Van der Waals broadening. An additional proof of this fact is the ratio of the shift and broadening of the  $0.63 \mu$  line. The very small shift of the  $0.63 \mu$  line, which, according to the data of<sup>[33,43]</sup>, lies in the range 0–5 MHz/Torr, indicates the presence of other types of interactions. Among these, interest attaches to a study of the exchange interaction.

Inasmuch as the exchange interaction is effective at distances when overlap of the electron shells of the colliding atoms takes place, it is of interest to compare the observed impact broadening cross section with the dimensions of the electron shell of the excited atom. With this aim, we compared the values of the mean distance  $\bar{r}$  of the excited atom from the nucleus for the different levels and the optical radius of interaction of the atom for lines which include the given levels. The values of  $\bar{r}$  were determined from the approximate formula<sup>[49]</sup>

$$\bar{r} = a_0 (n^*)^2 \left[ 1 + \frac{1}{2} \left( 1 - \frac{l(l+1)}{(n^*)^2} \right) \right], \quad (10)$$

and  $r_{\text{opt}}$  from the experimental values of  $\Gamma_{\text{imp}}$  with use of the relation  $\Gamma_{\text{imp}} = \pi \bar{r}_{\text{opt}}^2 \bar{v} N_0$ . Comparison of the values of  $\bar{r}$  and  $r_{\text{opt}}$  showed that, beginning as early as quantum number  $n = 4$  ( $n^* = 2.76$ ), these values are approximately equal, i.e., the atoms effectively interact at distances such that overlap of the electron shells takes place and, consequently, exchange of the electrons can play a role.

Impact broadening with exchange interaction for large  $n = 20$ – $30$  has been studied theoretically in the work of Alekseev and Sobel'man.<sup>[52]</sup> For the case of small  $n$ , the calculation of the impact broadening was performed by Kazantsev.<sup>[53]</sup> In his work, on the basis of the theoretical analysis of the collision of two hydrogen-like atoms, one of which is strongly excited, it has been shown that in the case of overlap of the electron shells of the colliding atoms, the effective interaction potential has the form

$$U(R) \sim \begin{cases} 1/R^2 R_0 & R \leq R_0 \\ 0 & R > R_0 \end{cases},$$

( $R_0$  are the turning points, at which the classical momentum of the electron vanishes). Here the cross section of the broadening collision coincides at  $n^* < 6$ – $7$  with the diameter of the exciting atom:

$$\frac{\partial\Gamma}{\partial p} \propto \sigma \sim R_0^2 \sim (n^*)^4.$$

Figure 8 shows the functions  $\alpha_1/n^{*3}$  and  $\alpha_2 n^{*4}$ , which represent the variation of the resonance broadening and the broadening due to exchange interaction. The coefficients  $\alpha_1$  and  $\alpha_2$  were so chosen that the function  $\alpha_1/n^{*3}$  coincides with the broadening at  $n = 3$  and  $\alpha_2 n^{*4}$  coin-

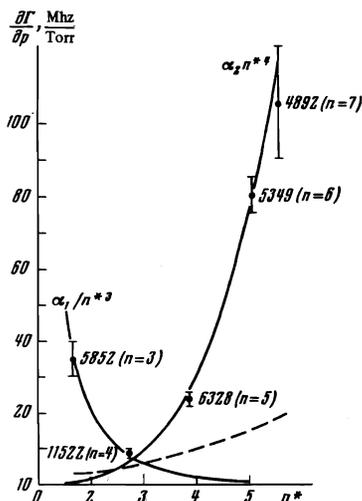


FIG. 8. Comparison of the experimental and theoretical dependences of the broadening on the principal quantum number. The value of the coefficients  $\alpha_1$  and  $\alpha_2$  are so chosen that the values of the functions are identical with the experimental points for  $n = 3$  and  $n = 6$ , respectively. ●—experimental points, the dashed line, the broadening due to Van der Waals type of interaction.

cides with the broadening at  $n = 6$ . As we see, the curves reflect accurately the experimental dependence of the value of the broadening on the principal quantum number.

In noting the excellent qualitative agreement of the dependence of the impact broadening on  $n$  for  $n \sim 5, 6, 7, \dots$ , it should also be noted that a similar dependence should be expected because of the presence of short-range attraction forces, which arise upon averaging the Coulomb forces over the volume charge distribution of the colliding atoms. It is evident that in the range of small  $n$  of interest to us, these forces appear only at distances comparable with the dimensions of the electron shell. The repulsion forces can be taken into consideration if we use a potential of the Lennard-Jones type for the description of the interaction:  $U(R) = C_{12}/R^{12} - C_6/R^6$ , as has been done in<sup>[53-56]</sup>. The coefficients  $C_{12}$  and  $C_6$  can be found from the experimental data on the broadening and shift of the lines and the dependence of these quantities on the temperature.<sup>[55]</sup> For the analysis of the dependence of the broadening on the principal quantum number, it is more convenient to use a potential of the Buckingham type:  $U(R) = a_n \exp(-b_n R) - C_6/R^6$ ; here  $b_n$  depends on  $n$  and is of the order of the reciprocal of the shell radius.

Although the determination of the specific type of interaction requires additional investigation, it can be regarded as reliably established that the interaction potential differs from the Van der Waals potential in the collision of excited atoms of neon ( $n = 6, 7$ ) with atoms in the ground state. It can be concluded from the analysis of the dependence of the impact broadening of the neon line on the principal quantum number that resonance interaction is the dominant form of interaction of atoms that have been excited to the level  $n = 3$ ; for atoms with  $n = 6$  or  $7$ , exchange and short-range Coulomb forces predominate; the case of  $n = 4$  or  $5$  is intermediate. For  $n = 5$ , it is necessary to take into account the Van der Waals and exchange interactions,

while for  $n = 4$  all three types of interaction can be important.

2. It was indicated above that a comparison of the collision shift and the line broadening can be used for the determination of the constants in the interaction potential. In the general case, it is necessary to do this with great care, especially when the collision broadening is determined from an analysis of the Lamb dip. Here an important contribution to the broadening can be made by elastic scattering of the atoms at small angles. When the excitation of both levels is the same, the broadening and the shift due to randomization of the phase are absent, and the principal process of broadening of the Lamb dip is the angular scattering of the atoms.<sup>[59]</sup> In this connection, a comparison of the broadening of the Lamb dip and the Doppler contour is very important. In the absence of randomization of the phase, the angular scattering of the atoms leads to a decrease in the effective vector velocity of the atoms and to a narrowing of the Doppler contour.<sup>[7]</sup> Therefore, the differences in the broadening of the two contours can indicate the contribution of elastic scattering to the line shape. A comparison of the experimental data for the  $0.63 \mu$  line shows that the collision broadening of the Doppler contour is not less than the broadening of the narrow resonance. In this case, the angular scattering plays a small role in the broadening of the Lamb dip.

The value of the angular scattering can be estimated by using the following considerations. The total cross section of elastic scattering in the quasiclassical approximation for the potential  $U = C_n/R^n$  is given by the formula (see Sec. 126 in<sup>[49]</sup>)

$$\sigma = 2\pi^{n/(n-1)} \sin \left[ \frac{\pi}{2} \left( \frac{n-3}{n-1} \right) \right] \Gamma \left( \frac{n-3}{n-1} \right) \times \left[ \Gamma \left( \frac{n-1}{2} \right) / \Gamma \left( \frac{n}{2} \right) \right]^{2/(n-1)} \left( \frac{C_n}{\hbar v} \right)^{2/(n-1)}$$

In the case of excitation of one of the levels, this cross section is identical with the cross section of the broadening due to randomization of phase in the classical theory of Lindholm (see Sec. 36 in<sup>[2]</sup>). From an analysis of the data on the broadening of lines which terminate or begin with the  $2p$  level, and also from experiments on the observation of the shape of the stimulated Raman scattering resonance line in neon<sup>[57]</sup>, it follows that the principal contribution to the broadening in collisions is due to excitation of the  $2p^5ns$  levels. Consequently, one can immediately estimate the contribution of diffusion of the atoms in the velocity space, due to angular scattering by collisions. According to Massey and Burhop,<sup>[3]</sup> the characteristic angle of scattering in the hard-sphere model is given by the relation  $\theta = \hbar/mvr$ , where  $r$  is the radius of the sphere, and  $m$  and  $v$  are the mass and velocity of the atom. Since the broadening of the  $0.63 \mu$  line is due to excitation of a single level,  $r$  can be set equal to the optical radius (which is close to the radius of the electronic shell). We then find  $\theta \sim 0.01$  rad.

Inasmuch as in the analysis of the Lamb dip we consider atoms with zero projections of the velocity on the axis of observation, the angular scattering of these atoms leads to the appearance of a  $z$  component of the velocity. The width of the distribution of the scattered

atoms with respect to  $v_z$  for a single scattering act will be equal to approximately  $\theta \bar{v}$ . This leads to an additional broadening of the Lamb dip by  $\theta k \bar{v}$ . For the  $0.63 \mu$  line, the scattering contribution to the broadening observed in this fashion amounts to 10 MHz, i.e., it is significantly less than the width of the dip. For pressures of the order of several Torr, the contribution of the diffusion of the atoms in the scattering at small angles is very small, which is confirmed by the linear dependence of the dip broadening on the pressure.

Another situation exists for the  $1.15 \mu$  line. Here, as estimates show, the Doppler shift in the scattering of atoms is several times that of the collision broadening. Therefore the small-angle scattering is similar in its manifestation to "strong" collisions, since, in a single collision act, the atoms leaves the range of velocities in which it interacts resonantly with the field. In this connection, the shape of the Lamb dip can be complicated, consisting of base of width  $\theta k \bar{v}$ , and a very narrow resonance with width  $2\Gamma$ . The ratio of the amplitudes of the base and the narrow resonance is equal to  $\sim \Gamma/\theta k \bar{v}$ .

It is interesting that the addition of another gas, for example He, can have a significant effect on the results of measurements of the broadening for Ne\* - Ne collisions. Actually, the scattering angle of the neon atoms colliding with the He atoms is much less than in collisions with Ne atoms, because of the difference in the masses of the atoms. As a consequence of this, such collisions will not make a contribution to the base, but will broaden the narrow resonance and decrease its amplitude. At the same time, the change in the partial pressure of the neon, while not affecting the width of the narrow peak to any extent, can significantly change the value of the base. If it is not possible to separate the peak and the base in the experimental study of the Lamb dip, then a change in the ratio of their amplitudes can become manifest in the change of the width of the complex contour. It is possible that the difference noted above in the results on the broadening of the  $1.15 \mu$  line, obtained in pure neon<sup>[12,29]</sup> and in the He-Ne mixture<sup>[5]</sup> is due to the considered effect of elastic scattering.

Recognizing that the scattering angle  $\theta$  is inversely proportional to the diameter of the atom, one can expect that the relative role of impact broadening, due to the phase randomization will increase with increase in the principal quantum number, and the broadening of the narrow resonances, which is associated with the diffusion of atoms in velocity space in small-angle scattering, will decrease.

3. We pause to note some qualitative features which were discovered in the comparison of the collision b broadening of the Doppler contour and the narrow resonance at  $\lambda = 0.63 \mu$ . Table II gives the results of the reduction of the contours of spontaneous emission of the neon lines with wavelengths 5852, 5433, and 6328 Å. The experimental data were reduced by two methods, which are based on the fact that the line contour has a Voigt form. The first of these<sup>[27]</sup> is sensitive to the wings of the line; in the second<sup>[2]</sup> the value of the width of the contour at half maximum is used. For the 5852 Å line, both methods gave practically the same result, while the discrepancy of the results for the 5433 and 6328 Å lines did not affect the accuracy of the experiment.

This discrepancy can be explained qualitatively if we recognize that the broadening and line shift depend on the velocity of the radiating atom. In this case the line contour will have a complicated asymmetric profile, which is different from the Voigt form.<sup>[20]</sup> The asymmetry of the contour follows from the fact that the center of the line connected with radiation of atoms whose velocity  $v < \bar{v}$  will undergo a smaller shift than the wing, which is due to the radiation of atoms with velocities  $v > \bar{v}$ . From this it is also understood that if the reduction of the contours is carried out by the method based on analysis of the wings of the line, then the value of the Lorentzian width found by such a method will be larger than that found from the width of the contour at half maximum. The impact-broadening constant, determined from the Lamb dip should be even smaller in such a case, since the principal contribution to the Lamb dip is made by atoms with a zero velocity projection on the observation axis.

The arguments set forth above are in qualitative agreement with the experimental results (Table II). The validity of the approach selected for the explanation of the results is confirmed by the fact that measurements for 5852 Å (made under similar conditions) gave excellent agreement in the treatment by the two methods. The fundamental mechanism of broadening of the  $1s_2$  line of neon is, as we have already established, a resonant exchange of excitation in the collisions. The cross section  $\sigma$  of this process is inversely proportional to  $v$ , and since the line width of the radiation is proportional to  $Nv\sigma$ , it will not depend on the velocity, and the resultant contour of the radiation line will have a Voigt form. We also note that, in spite of qualitative agreement of these representations, and also the conclusions of<sup>[18]</sup> with the results of experiment, the final solution of the problem of the dependence of the width and line shift on the velocity requires additional detailed investigations.

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Note added in proof (21 November 1972). Data for the impact broadening of the 6328 Å line of neon; presented in a recent dissertation by Knutson, agree well with our results

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