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Linewidth Studies on the Kr^+ 469.4 nm and 473.9 nm Lines Excited in a Helium-Krypton Hollow Cathode Discharge

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Abstract. Collisional and Doppler linewidths ($\Delta\nu_C$ and $\Delta\nu_D$) of the 469.4 nm and 473.9 nm Kr ion lines were measured in a He-Kr hollow cathode discharge using Fabry-Perot technique. A linear dependence of $\Delta\nu_C$ on He pressure was found for both lines. Significant differences were found between the temperature values deduced from the $\Delta\nu_D$-s of the two lines, and an unexpected temperature dependence of the broadening parameter for the Kr^+ 469 nm line was also observed. The temperature difference between the two lines is explained by excitation of the upper level of the 469 nm line by second kind collisions between metastable He atoms and ground-state Kr ions, while the temperature dependence of the broadening parameter of the Kr^+ 469 nm line is suggested to be due to the inverse process.

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Hollow cathode excited (HC) gas lasers usually oscillate in a single longitudinal mode without using any mode selection. Single mode operation is characteristic of laser systems, where the gain curve is homogeneously broadened, i.e. all atoms excited to the upper laser level interact in the same manner with the radiation field. Line broadening due to the finite lifetime of the participating energy levels or broadening due to collisions with other atoms are homogeneous, while Doppler broadening due to the thermal motion of atoms is inhomogeneous. In the visible and UV spectral region at low pressures (several mbars) the homogeneous broadening is small compared to the Doppler broadening, but by increasing the gas pressure the collisional linewidth increases and can be comparable to the Doppler linewidth. In HC lasers the actual working gas pressure amounts to several tens of mbars which is high enough for homogenizing the whole gain curve which results in single mode operation.

Noble gas mixture HC lasers, among them the blue He-Kr^+ laser ($\lambda = 469.4$ nm) shows this behaviour.

It was demonstrated by Jánossy et al. [1, 2] that at a working pressure of about 30 mbar, only one longitudinal mode exists. The frequency of this mode can be tuned – by changing the resonator-length – over a frequency range of about 450 MHz. This tuning range is significantly larger, than the longitudinal mode separation of 144 MHz which can be calculated from the given length of the resonator. The nearly constant mode intensity during the tuning process also indicates that the homogeneous linewidth appears to be in this order [2].

In this paper we present direct linewidth measurements under conditions typical for laser operation, using a HC light source with similar geometry, current and pressure values. Besides the expected linear increase of the collisional linewidth with the He pressure, we found an unexpected temperature dependence of the broadening parameter and large differences between the temperature values deduced from the Doppler linewidths of the 469 nm and 474 nm Kr^+ lines. These features can be explained by taking into account the specific excitation mechanism of the upper laser level.

The Kr^+ 469.4 nm line arises from the $5p^4D^0_{7/2} \leftarrow 6s^4P_{5/2}$ transition. In a helium-krypton mixture excited in a DC hollow cathode discharge, the upper level is believed to be populated by second kind collisions between metastable He atoms and ground-state Kr ions [3]:

$$\mathrm{He}(2^3S_1) + \mathrm{Kr}^+(4^2P^0_{3/2}) \rightarrow \mathrm{He}(1^1S_0) + \mathrm{Kr}^+(6s^4P_{5/2}) + 0.35\,\mathrm{eV}. \quad (1)$$

Ground-state Kr ions are formed mainly by collisions with fast electrons. The $Kr^+(6s^4P_{5/2})$ level (19.47 eV) lies 0.35 eV lower than the $He(2^3S_1)$ level (19.82 eV) and the excess energy is taken away as kinetic energy by the colliding particles. Typical conditions for laser operation are 30–40 mbar He pressure with 0.1 mbar Kr partial pressure and 2–4 A exciting current for a laser having 40 cm total cathode length and 3 mm cathode bore diameter.

The Kr$^+$ 473.9 nm line, which was also investigated, arises from the $5s^4P_{5/2} \leftarrow 5p^4P^0_{5/2}$ transition. Here the upper level (16.6 eV) lies significantly lower than the He 2^3S_1 level, therefore it is believed to be populated mainly by electron impact. Above a He pressure of 20 mbar and at a Kr partial pressure of 0.1 mbar this line is significantly weaker than the Kr$^+$ 469 nm line.

1. Experimental Setup and Evaluation Method

The light source was similar to that used in earlier laser experiments [2] (Fig. 1). Spontaneous light emitted from a 3 cm long water cooled Al-cathode was measured: the middle of the discharge region inside the cathode was imaged by a lens on the 100 µm slit of a small monochromator (Jobin-Yvonne Model HUV-20, resolution 0.5 nm) which selected the desired wavelength. The exit slit of the monochromator was then imaged by a second lens to the ∅150 µm input diaphragm of a piezoelectrically scanned triple-pass Fabry-Perot interferometer (FPI, Tropel Model 350). The partial pressure of Kr was always 0.1 mbar, the He pressure was varied from 10 to 40 mbar. The intensity of the Kr$^+$ 469 nm and Kr$^+$ 474 nm lines was rather weak, especially outside the 20–30 mbar region, therefore photon counting was applied. FP interferograms were recorded by a standard multichannel analyzer and X–Y recorder. Typical measuring times were 500–1000 s. The apparative profile and finesse of the FPI was measured simultaneously with the Kr$^+$ lines using the scattered light from the FPI input diaphragm illuminated with the $\lambda = 488$ nm radiation of a single mode Ar$^+$ laser (Spectra Physics Model 164 with internal etalon). The actual working finesse of the FPI proved to be about 35 at mirror distances of 10–15 mm (free spectral range 10–15 GHz) which were generally used in the measurements.

The discharge inside the cathode was sustained by a current stabilized power supply. After an initial cleaning period of several hours using low currents (30–50 mA), the discharge proved to be stable and free from arcs during the whole measuring time. The spatial intensity distribution changed with changing the pressure and current as well, due to the changes of the size of the negative glow. These changes could differ significantly for the different spectral lines due to the different excitation mechanisms as it was shown by Kuen et al. [4] and Rózsa [5]. This problem is complicated further by the fact, that the 3 cm long light source was observed "end on" collecting the light by a lens within a cone angle of about 10° resulting in a rather indefinite spatial average. At low pressures, when the middle of the discharge is far more the brightest, this averaging causes no serious problem. At larger pressures (above 35 mbar) when the size of the negative glow decreases, the middle of the discharge becomes dark (Fig. 4). Significant changes in the parameters can take place here which should be kept in mind by evaluation of the data.

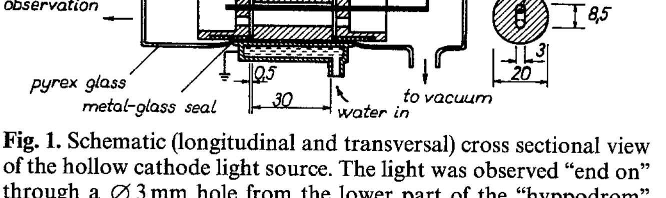

Fig. 1. Schematic (longitudinal and transversal) cross sectional view of the hollow cathode light source. The light was observed "end on" through a ∅3 mm hole from the lower part of the "hyppodrom" shaped cathode hollow

For evaluation of the interferograms a rather simple deconvolution method was applied. In the first step, from the observed FP interferogram the halfwidths of the Lorentzian and Gaussian components ($\Delta\nu_L$ and $\Delta\nu_G$, full widths at half maxima) were determined assuming that the line has a true Voigt-profile. Davies and Vaughan [6] have published tabulated data on the frequency intervals between symmetrical points of given intensities of a Voigt profile. Six points were chosen by us in the intensity range 0.3–0.05 (compared to the intensity maximum), from which an average value for $\Delta\nu_L$ and $\Delta\nu_G$ could be easily determined. A small correction due to the overlapping of the FPI orders and the constant background was calculated here using the method of Bradberry and Vaughan [7].

In the second step correction due to the apparatus function of the FPI was performed. Corrected Lorentzian linewidths (i.e., collisional linewidths $\Delta\nu_C$) and corrected Gaussian linewidths (i.e., Doppler linewidths $\Delta\nu_D$) could be calculated using the correction curve published by Lindsay et al. [8] for the triple pass FPI. They assumed, that the apparatus function $T(\nu-\nu_i)=[1+K^2(\nu-\nu_i)^2]^{-3}$ where $(\nu-\nu_i)$ denotes the frequency difference from the transmission peak ν_i of the FPI, and K is an instrumental parameter. In our case, however, the measured apparatus function had this shape only at small ($d=0.3$ mm) mirror distances, while at the generally used distances of 10–15 mm, a modified apparatus function was observed with a halfwidth $\Delta\nu_a^{\text{exp}}$ larger than the theoretically expected $\Delta\nu_a^{\text{theor}}=(c/2d)(0.51/F_1)$. Here F_1 is the FPI finesse for single pass, c the light velocity and d the mirror distance [9]. (In our case, for $F_1=27$ and $d=10$ mm, $\Delta\nu_a^{\text{theor}}=0.28$ GHz.) This extra broadening could be attributed to mechanical vibrations and had a Gaussian character. Therefore the correction for $\Delta\nu_L$ was determined from $\Delta\nu_a^{\text{theor}}$, while the correction for $\Delta\nu_G$ was made with $\Delta\nu_a^g$, the Gaussian part of $\Delta\nu_a^{\text{exp}}$ in the following way: $(\Delta\nu_D)^2=(\Delta\nu_G)^2-(\Delta\nu_a^g)^2$, where $(\Delta\nu_a^g)^2=(\Delta\nu_a^{\text{exp}})^2-(\Delta\nu_a^{\text{theor}})^2$.

2. Results and Discussion

2.1. Collisional and Doppler Linewidths in the 13–37 mbar He Pressure Region

Two series of measurements were made: one with 100 mA (approximately the laser threshold current) and the other with 200 mA (typical laser current). The results are summarized in Figs. 2 and 3. For the Kr^+ 469 nm line (Fig. 2) points for $\Delta\nu_C$ in the 13–30 mbar pressure region fit well to straight lines intersecting each other close to zero pressure at about $\Delta\nu_0 \approx 41$ MHz, in accordance with the expected natural linewidth of (43 ± 3) MHz, calculated from the lifetimes of the upper and lower levels $\{\tau_{\text{upper}} = (6.5 \pm 0.6)$ ns, $\tau_{\text{lower}} = (8.8 \pm 0.8)$ ns [10]$\}$. Least square fitting resulted for zero pressure linewidths (46 ± 5) MHz and (35 ± 4) MHz, for the corresponding slopes (19.9 ± 0.2) MHz/mbar and (22.8 ± 0.1) MHz/mbar, respectively. The two values were obtained at 100 and 200 mA currents.

In connection with the problem of single-mode laser operation at 469 nm, from the linewidth data of Fig. 2 it can be stated, that in the 30–40 mbar He pressure region $\Delta\nu_C = 700$–800 MHz. This is indeed much larger than the typical mode separation and therefore the collisional line broadening in these lasers can result in single mode operation.

For the Kr^+ 474 nm line (Fig. 3) the measured $\Delta\nu_C$ values in the 13–30 mbar pressure region fit also quite well to straight lines intersecting each other close to zero pressure, at about $\Delta\nu_0 \approx 230$ MHz, in good agreement with the value of $\Delta\nu_0 = (200 \pm 50)$ MHz, measured earlier by Korolev et al. [11]. Calculation gives here zero pressure values of (220 ± 50) MHz and (235 ± 15) MHz, slopes (13.0 ± 1.0) MHz/mbar and 11.5 ± 0.5) MHz/mbar, respectively. The two values belong again to 100 and 200 mA currents.

In Figs. 2 and 3 the Doppler linewidths ($\Delta\nu_D$) are also shown. As can be seen, at a given current in the 13–30 mbar pressure region they are practically constant; $\Delta\nu_D(469\text{ nm}) = 1.21$–$1.33$ GHz, $\Delta\nu_D(474\text{ nm}) = 1.05$–$1.16$ GHz, the two different values corresponding to 100 and 200 mA, respectively.

The mean statistical error (standard deviation) of the linewidth data, deduced from 2–4 measurements for each point, amounts to about ± 30 MHz, while that of the slope data is less than ± 1 MHz/mbar. These error limits can be taken as the measure of accuracy concerning their relative changes with the current (or temperature), but it does not reflect the possible systematic errors due to the spatial average taken in the light source or due to the deconvulation process. These systematic errors could affect the results without distorting the linearity. Therefore the absolute values of the Lorentzian and Doppler linewidths (given in MHz) and the values of the broadening constants (given in MHz/mbar) are not more reliable than ± 50 MHz and ± 3 MHz/mbar, respectively.

In the $p > 30$ mbar pressure region the Kr^+ 474 nm line was too weak to be measured. For Kr^+ 469 nm the measured $\Delta\nu_C$ values at $p \approx 37$ mbar lie *below* the straight lines and the corresponding $\Delta\nu_D$ values are *smaller* than those for lower pressures (Fig. 2). This deviation can be explained by the change of the spatial intensity distribution of the Kr^+ 469 nm line inside the cathode. At 37 mbar He pressure the light intensity starts to decrease in the centre of the cathode hollow. In this case more light is collected by the imaging lens from the negative glow near the water cooled cathode wall and therefore a lower gas temperature is observed. The decrease of the intensity

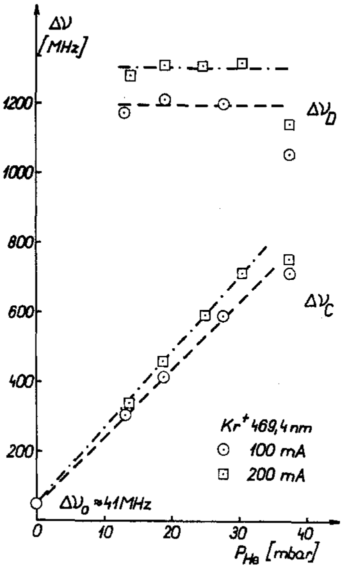

Fig. 2. Observed collisional ($\Delta\nu_C$) and Doppler ($\Delta\nu_D$) linewidths (full widths at half maxima) for the Kr^+ 469 nm transition at different He pressures. Kr partial pressure 0.1 mbar

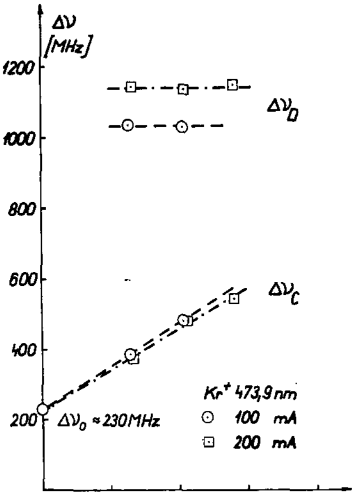

Fig. 3. Observed collisional ($\Delta\nu_C$) and Doppler ($\Delta\nu_D$) linewidths (full widths at half maxima) for the Kr^+ 474 nm transition at different He pressures. Kr partial pressure 0.1 mbar

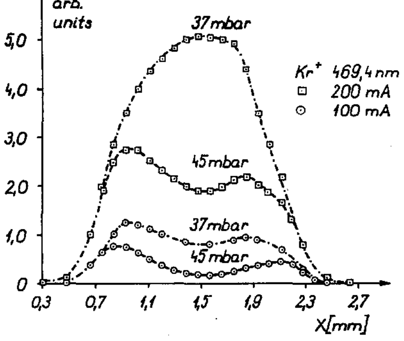

Fig. 4. Transverse intensity distribution of the Kr^+ 469 nm line at 37 mbar and 45 mbar pressures and at 100 mA and 200 mA currents. x: transverse position inside the cathode

in the centre is more pronounced at 100 mA current, but for larger pressures it can be clearly seen at 200 mA as well. This is illustrated in Fig. 4, where the radial intensity distribution of the Kr^+ 469 nm line is shown at 37 and 45 mbar pressures and at 100 and 200 mA exciting currents, respectively. The radial intensity distribution was measured by transversal movement of the focusing lens.

2.2. Temperature Data Deduced from the Doppler Linewidths

For determining the dependence of the collisional linewidth $\Delta\nu_C$ on the He atom density N_{He} we have to determine the gas temperature T. Temperature values can be deduced from the Doppler linewidths of different spectral lines, however they may differ significantly from each other. In the 13–30 mbar pressure region, where at a given current the measured Doppler linewidths for the Kr^+ 469 nm and Kr^+ 474 nm lines were nearly constant (Figs. 2 and 3), an average temperature could be determined, but the temperature values for Kr^+ 469 nm proved to be about 160 K higher, than those for Kr^+ 474 nm, at both current values (Table 1).

We tried to determine the gas temperatures for the two excitation currents from the Doppler linewidth of the He 502 nm line, too. Temperature differences can also be observed here, but the temperature values showed a monoton decrease with increasing pressure. The reliability of the He 502 nm temperature data is questionable, however, because the lower level of this transition is the metastable $\mathrm{He}(2^1S_0)$ level. This level is highly populated in the given pressure and current region and can cause serious deformation of the line profile. Therefore these data were not taken into account.

The situation is better with the Kr^+ 474 nm line, where the population of the lower level can be neglected and the upper level is assumed to be excited by electron impact. Thus the temperatures deduced from the Doppler widths of this line can be accepted as true gas temperature values. The mean statistical error (standard deviation) of the temperature data is about ±25 K, which can be taken again as the measure of accuracy concerning the relative changes of the temperature with the exciting current or with the He pressure. The accuracy in the determination of the real gas temperature however – due to the possible systematic errors – is estimated to be ±50 K.

The difference of about 160 K between Kr^+ 469 nm and Kr^+ 474 nm temperatures can be explained by the difference of their excitation mechanisms. Calculations based on a simple model using average thermal velocities for the colliding particles and taking into account the conservation of momentum and energy during the collision show that at the excitation of the upper level of the Kr^+ 469 nm line by second kind collisions, see (1), the excess energy of 0.35 eV causes a temperature rise of about 200 K for the colliding Kr^+ ions. This is in quite good agreement with our measured value. Therefore the observed higher temperature for the Kr^+ 469 line can be taken as a further support of the assumption, that the dominant excitation mechanism for the Kr^+ 469 nm laser transition is second kind collisions of type (1).

Table 1. Average temperature values deduced from the Doppler linewidths of the Kr^+ 469 nm and Kr^+ 474 nm lines, at different currents (pressure region 13–30 mbar, estimated error ±50 K, see text)

Current Line	100 mA	200 mA
Kr^+ 469 nm	600 K	710 K
Kr^+ 474 nm	444 K	545 K

2.3. Temperature Dependence of the Broadening Parameters of the Kr^+ 469 nm and Kr^+ 474 nm Lines

Assuming that the Kr^+ 474 nm temperatures give the true gas temperatures, the collisional linewidth data $\Delta\nu_C$ for Kr^+ 469 nm can be expressed in terms of the He atom density N_{He}. In the density range corresponding to the 13–30 mbar pressure region, where the temperature was nearly constant, the dependence of $\Delta\nu_C$ on N_{He} is also linear: the two straight lines corresponding to the two temperature (and current) values ($\bar{T}_1 = 545$ K for 200 mA; $\bar{T}_2 = 444$ K for 100 mA) intersect each other again close to $N_{\mathrm{He}} = 0$, and $\Delta\nu_0 \approx 41$ MHz as before, thus it can be written:

$$\Delta\nu_C(N_{\mathrm{He}}, T) = \Delta\nu_0 + a(T, \lambda) N_{\mathrm{He}} . \tag{2}$$

The broadening parameter $a(T, \lambda)$ for the two temperatures is:

$$a(545\,\mathrm{K}, 469\,\mathrm{nm}) = (171 \pm 1)\,\mathrm{MHz}/10^{17}\ \mathrm{atoms} \cdot \mathrm{cm}^{-3},$$

$$a(444\,\mathrm{K}, 469\,\mathrm{nm}) = (122 \pm 1)\,\mathrm{MHz}/10^{17}\ \mathrm{atoms} \cdot \mathrm{cm}^{-3}.$$

Here mean statistical errors (standard deviations) are given. The reliability of "a" – due to the possible systematic errors in the determination of $\Delta\nu_C$ and T – is estimated not to be better than $\pm 30\,\mathrm{MHz}/10^{17}$ atoms $\cdot\,\mathrm{cm}^{-3}$.

The temperature dependence of $a(469\,\mathrm{nm})$ is surprising because – if the dominant broadening mechanism is phase interrupting collisions between different atoms – a rather weak temperature dependence is expected [12]:

$$a = 2\pi \langle \sigma v_r \rangle \sim T^{0.3} . \tag{3}$$

(Here σ denotes the collision cross section and v_r the relative velocity of the colliding atoms.) In our case, however

$$a(545\,\mathrm{K}, 496\,\mathrm{nm}) / a(444\,\mathrm{K}, 469\,\mathrm{nm}) = 1.40 = (\bar{T}_2/\bar{T}_1)^{1.65 \pm 0.40} . \tag{4}$$

The error limit in the exponent is calculated from the statistical errors of $\bar{T}_2$ and $\bar{T}_1$.

It is interesting to compare the broadening parameter data of the Kr^+ 469 nm line with that of the Kr^+ 474 nm line. They can be determined – in terms of N_{He} – in the same way as before, resulting:

$$a(545\,\mathrm{K}, 474\,\mathrm{nm}) = (87 \pm 8)\,\mathrm{MHz}/10^{17}\ \text{atoms} \cdot \mathrm{cm}^{-3},$$

$$a(444\,\mathrm{K}, 474\,\mathrm{nm}) = (79 \pm 4)\,\mathrm{MHz}/10^{17}\ \text{atoms} \cdot \mathrm{cm}^{-3}.$$

As before, mean statistical errors (standard deviations) are given here. The reliability of the parameter data – due to systematic errors – is not better than $\pm 30\,\mathrm{MHz}/10^{17}$ atoms $\cdot\,\mathrm{cm}^{-3}$. It is clear that the temperature dependence of $a(474\,\mathrm{nm})$ is significantly weaker, than that of $a(469\,\mathrm{nm})$. The actual change of $a(474)$ with T corresponds – within the measuring error – to the theoretically expected $T^{0.3}$ – dependence of (3):

$$a(545\,\mathrm{K}, 474\,\mathrm{nm})/a(444\,\mathrm{K}, 474\,\mathrm{nm}) = 87/79 = 1.10 \pm 0.10$$

while

$$(T_2/T_1)^{0.3} = (545/444)^{0.3} = 1.06 \pm 0.02$$

indicating, that the main physical process in the pressure broadening of the Kr^+ 474 nm line is phase interrupting collisions between ground state He atoms and excited Kr ions.

The origin of the observed temperature dependence of $a(469\,\mathrm{nm})$ is not clear. The difference in the temperature dependence of a for the two Kr^+ lines suggests that in the broadening of the Kr^+ 469 nm line some other physical process or processes can also take part along with the phase interrupting collisions. The most plausible assumption would be that it is the second-kind collision of type (1) but in the opposite direction: inelastic collisions between excited Kr^+ ions and ground state He atoms would contribute to the broadening of the Kr^+ 469 nm line. This is, however, not the usual case, because it is generally accepted, that inelastic collision cross sections for different atoms are one or two orders of magnitude smaller than those for phase interrupting collisions and therefore they can be neglected in the line broadening mechanisms. (See, e.g., the monography of Bennett [12] on the physics of gas lasers.) On the other hand, however, there is an indication that He-Kr^+ collisions, (1), would be an exception from this rule. Analyzing their gain-measurement on the He-Kr^+ 469 nm laser, Solanki et al. [13] have stated, that the cross section for the second-kind collision of type (1) should be $10^{-14}\,\mathrm{cm}^2$ or even larger; i.e. it would be comparable with that of the phase interruption collisions. If their statement is correct, this process cannot be neglected any more. Therefore it seemed interesting for us to analyze our results from this point of view as well.

It was shown by Jones and Robertson [14, 15] that the temperature dependence of the second-kind collision cross section (σ) can be given as

$$\sigma(T) = \sigma_0 \exp(-E_a/kT), \tag{5}$$

where E_a has the form of an activation or dissociation energy characterizing the height of the potential barrier of the quasi-molecule, formed by the colliding atoms, σ_0 is a constant of cm^2 dimension and k is the Boltzmann-constant.

Applying (3) and (5) to our case, it can be assumed that the broadening parameter $a(T)$ for Kr^+ 469 nm can be written as the sum of two components $a_1(T)$ and $a_2(T)$, the first one describing the phase interrupting collision broadening, the second one the inelastic collision broadening:

$$\begin{aligned} a(T) &= a_1(T) + a_2(T) \\ &= \alpha T^{0.3} + \beta T^{0.5} \exp(-\gamma/T), \end{aligned} \tag{6}$$

where α, β, and γ are constants; $\gamma = E_a/k$.

Unfortunately our data are insufficient to determine the values of α, β and γ. But assuming that near 500 K the second term in (6) becomes dominant, the observed temperature dependence of "a", (4), can be explained. Namely, the exponential function in (6) can be approximated with a linear one and by this way the second term in (6) will be proportional to $T^{1.5}$, which is close to the value found by us. This result makes it likely that inelastic collisions do indeed play a role in the broadening of the Kr^+ 469 nm line.

Further investigations on the He-Kr^+ system and also on other noble gas mixture are needed to find additional support to the interpretations outlined and to clarify further details of the process involved in the excitation and broadening processes.

3. Summary

It was found that the collisional linewidth for the Kr^+ 469 nm laser transition – at optimal laser parameters – amounts to about 800 MHz. This large homogeneous broadening explains the single mode operation property of the He-Kr^+ laser.

A difference of about 160 K was observed between the temperatures deduced from the Kr^+ 469 nm and Kr^+ 474 nm Doppler linewidths. We explained this fact by the second-kind collision excitation mechanism of the upper laser level.

An unexpected temperature dependence of the broadening parameter for Kr^+ 469 nm was observed, which has been explained by quenching collisions between ground state He atoms and excited Kr ions.

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