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On the mechanism of populating 3p levels of neon under pumping by a hard ioniser

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***Abstract*. The effect of quenching additives on the luminescence properties of helium – neon mixtures under pumping by α particles emitted from ^{210}Po atoms is considered. It is concluded that, under excitation by a heavy charged particle, the population of the $3p'[1/2]_0$ level of neon is not related to the dissociative recombination of molecular ions. It is suggested that the most likely channels for populating the 3p level are the excitation transfer from metastable helium atoms to neon atoms and direct excitation of neon by nuclear particles and secondary electrons.**

Keywords: *laser, nuclear pumping, neon, population mechanism, metastable atom, cascade transitions.*

Lasing in the visible range, absence of nondegrading and chemically inactive working gas mixture, and high efficiency are undoubted advantages of high-pressure lasers on 3p – 3s transitions in neon pumped by ionising radiation [1, 2]. 585-nm lasers have a low lasing threshold. The possibility of designing a He – Ne – H_2 laser pumped by α particles was considered theoretically in [3]. In this study we investigated the dependence of the line intensity at $\lambda = 585$ nm on the concentration of quenching additives (H_2, Ar, Kr, D_2) in He – Ne mixtures under excitation by α particles from ^{210}Po.

The system for measuring spectra was described in [4]. A stainless steel chamber contained 18 ^{210}Po sources. The excitation region was 25 mm in diameter and 70 mm long. The maximum range of 5-MeV α particles in helium under normal conditions is 183 mm [5]. Before mounting the sources the chamber was heated and degassed at a pressure of $\sim 10^{-5}$ Torr. After mounting, the sources were evacuated without heating for two to three weeks to obtain reproducible (within 3 % – 7 % intensity for different gases) luminescence spectra.

The gas pressure was measured by a standard pressure-and-vacuum gauge and a VDG-1 pressure gauge; the gas purities were (Ne) 99.996 %, (He) 99.99 %, (Ar) 99.992 %, and (Kr) 99.999 %. Technical hydrogen and deuterium [enriched in D_2 to 99%, with nitrogen (about 0.1 %) and oxygen (about 0.05 %) impurities] were purified by infiltrating through silica gel and active copper. The emission spectrum was analysed using an SPM-2 monochromator with a quartz prism and FEU-106 photoelectron multiplier, operating in the photon-counting regime. The activity of sources was 9.6 GBq; at a helium pressure of 2 atm this value corresponds to an average energy contribution $W \sim 3 \times 10^{-5}$ W cm^{-3} and gas-volume-averaged ionisation rate $S \sim 4 \times 10^{12}$ cm^{-3} s^{-1}.

The measured dependences of the luminescence intensity at $\lambda = 585$ nm on the pressure of quenching additives are shown in Fig. 1. We also performed measurements with admixtures of technical nitrogen containing ~ 2 % oxygen.

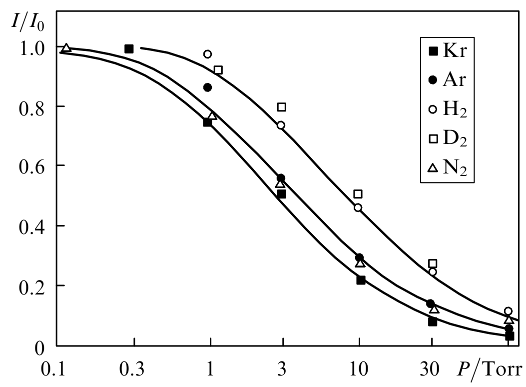

Figure 1. Dependences of the luminescence intensity of neon atoms at a wavelength of 585 nm on the pressure of additives to the He (2 atm) + Ne (50 Torr) mixture; I_0 is the luminescence intensity in the mixture without additives.

The processes in the Ne atom in active media of lasers on 3p – 3s transitions have been well-studied [1]: the upper laser level is occupied mainly due to the dissociative recombination of Ne_2^+ and $HeNe^+$ molecular ions. At relatively weak pumping $HeNe^+$ ions form also Ne_2^+ ions in the substitution reaction

$$HeNe^+ + Ne \to Ne_2^+ + He.$$

On the assumption that the luminescence intensity at the line with $\lambda = 585$ nm is determined by the competition between the charge exchange in Ne_2^+ ions at the quenching admixture and recombination of electrons with Ne_2^+, we obtain the following expression for the additive pressure at which the luminescence intensity decreases by half:

$$P = \sqrt{\beta S}/k,$$

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where k is the charge exchange coefficient of Ne_2^+ ions at additive particles and β is the recombination coefficient of Ne_2^+ with electrons.

For hydrogen $k = 1.1 \times 10^{-10}$ cm^3 s^{-1} [6]; then, at an 'average' ionisation rate, the pressure $P \sim 2 \times 10^{-4}$ Torr, a value corresponding to a hydrogen concentration of 8×10^{12} cm^{-3}. The measured P values are 2.8 Torr for Kr, 3.8 Torr for Ar, and 8–9 Torr for H_2 and D_2 in mixtures with helium (2 atm) and neon (50 Torr) (see Fig. 1). The high luminescence intensity of neon in mixtures with argon or krypton at their pressure of several Torr may be related to small charge exchange coefficients of Ne_2^+ at Ar and Kr [6]. In [1] the charge exchange rate constant $k = 1.3 \times 10^{-13}$ cm^3 s^{-1} for Ne_2^+ at H_2 was used to match the computational model with the experimental data on electronic-beam and nuclear pumpings. This k value could partially explain the results for hydrogen mixtures (Fig. 1). The k values for nitrogen were reported to be 9.1×10^{-10} [6] and 8.6×10^{-10} cm^3 s^{-1} [7]; the decrease in the intensity by half in mixtures with nitrogen (3.5 Torr) are inconsistent with the large charge exchange coefficient.

The effective luminescence with $\lambda = 585$ nm at quenching-additive pressures of a few Torr can be caused by the inhomogeneous track structure of the plasma formed. The range of 5-MeV α particles in helium at a pressure of 2 atm is 9 cm; the track radius ($\sim 5 \times 10^{-4}$ cm) is determined by the free path of secondary electrons with energy of ~ 100 eV, and the track region volume is $\sim 7 \times 10^{-6}$ cm^3. The formation energy of an electron–ion pair in helium is 45 eV, a passage of an α particle leads to the formation of 10^5 electrons, and the initial electron concentration in the track is $n_e \sim 10^{10}$ cm^{-3}. At this concentration the characteristic recombination time of electrons and Ne_2^+ ions is $\tau = 1/\beta n_e \sim 10^{-3}$ s. The track lifetime, which is determined by ambipolar diffusion, is several tens of nanoseconds [8]. The track diffusion time over the gas volume is much shorter than the characteristic recombination time, and the weak dependence of the luminescence intensity on the pressure of additives is not related to the track plasma structure. The dependences of the luminescence intensity on the krypton pressure, measured at different helium pressures (Fig. 2), confirm this conclusion. With a change in the mixture pressure from 1 to 6 atm the initial electron concentration in the track increases by a factor of 200,

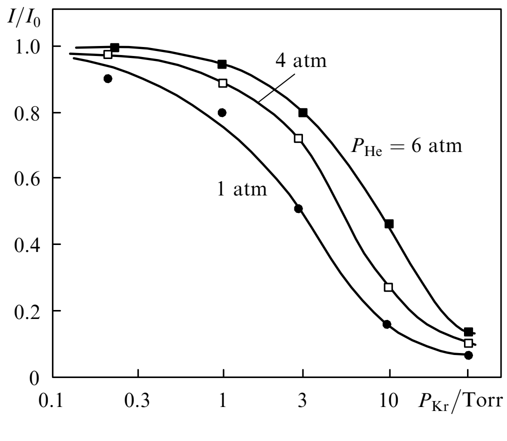

Figure 2. Dependences of the luminescence intensity of neon atoms on the krypton pressure in the He + Ne (40 Torr) + Kr mixture at helium pressures of 1, 4, and 6 atm.

and the krypton pressure at which the luminescence intensity decreases by half increases by a factor of only three, which can be explained by the efficient quenching of the $3p'[1/2]_0$ state of neon by He atoms at high helium pressures.

Apparently, upon excitation by a heavy particle, the $3p'[1/2]_0$ level of neon is populated not during the dissociative recombination of molecular ions, as in the case of Hg levels (under nuclear pumping of mercury-containing mixtures HgI levels are populated during dissociative recombination of Hg_2^+ ions [9]). Adding oxygen (0.1 Torr) to the $^3He + Hg$ mixture reduces the intensity of the triplet lines and the resonant mercury line by a factor of about 500 [10], which is related to the attachment of electrons to O_2 molecules. Apparently, the charge exchange in He_2^+ ions at O_2, which competes with the exchange at mercury atoms, is insignificant in this case, because O_2^+ ions should also undergo charge exchange at Hg atoms. An addition of technical nitrogen (to 50 Torr) with an oxygen impurity (~ 2 %) to the He (2 atm) + Ne (50 Torr) mixture led to the same decrease in the luminescence intensity as the addition of pure Ar and Kr; i.e., the attachment of electrons to electronegative impurity does not affect the occupation of the $3p'[1/2]_0$ level of neon.

Poletaev et al. [11], who studied the spectrotemporal characteristics of the luminescence of pure neon under pumping by heavy charged particles, concluded that neon levels are populated via direct excitation by nuclear particles and secondary delta electrons; in He–Ne mixtures population occurs during excitation transfer from helium metastable atoms:

$$He^m + Ne + He \rightarrow Ne(3p) + 2He.$$

In our opinion, a more likely channel for populating the 3p levels of neon in a mixture with helium is the cascade transitions from 4s levels:

$$He^m + Ne \rightarrow Ne(4s) + He,$$

$$Ne(4s) \rightarrow Ne(3p) + h\nu.$$

The energies of Ne (4s) and He (2^3S_1) levels are known to be similar: the operation of He–Ne laser ($\lambda = 1.15$ μm) is based on the excitation transfer from He(2^3S_1) to neon atoms. The 4s–3p transition lines were not observed for a high-pressure mixture [11] because these transitions lie in the IR spectral region beyond the FEU sensitivity limit. Abramov et al. [12], who recorded spectra up to 1100 nm, observed the 966.5-nm line (which corresponds to the $4s[3/2]_2 - 3p[1/2]_1$ transition) upon excitation of neon and He–Ne mixture by uranium fission fragments. In addition, they identified more than ten lines of the 3d–3p transitions with a total specific emission power of about 32 mW cm^{-3} in neon (64 kPa) and 7 mW cm^{-3} in a He:Ne = 179:1 mixture (180 kPa). These transitions lead mainly to the population of the three lowest 3p levels. The emission power at the $3p[1/2]_1 - 3s'[1/2]_0$ transition (743.9 nm) is 5 mW in neon and 2.8 mW in the He–Ne mixture. Hence, knowing the level lifetime (25.4 ns) and probability of this transition (2.4×10^6 s^{-1}) [13], one can determine the total emission power from the $3p[1/2]_1$ level, which is 82 mW for neon and 46 mW for the He–Ne mixture. The emission intensity from the $3p[1/2]_1$ level is about 50 % of the total emission intensity at the 3p – 3s transition [14]; thus, the 3d – 3p transitions provide ~ 20 % of emission intensity at the 3p – 3s transitions in neon and ~ 7 % in the He – Ne mixture. The emission spectrum may also contain some other 3d – 3p transitions, which were not indicated in [12], because their long-wavelength edge extends to 1169 nm [13].

The population of 3p levels as a result of recombination of Ne_2^+ ions could be impeded by the Ne_2^+ exchange at impurities in the He – Ne mixture without additives. Table 1 contains the measurement results that allow one to estimate this possibility. The luminescence intensity at 585 nm in a mixture with high-purity ^{3}He (the content of nitrogen, hydrogen, and hydrocarbons is below 0.0001 %) did not differ from the intensity in the mixture with helium of grade B. It contains also the intensities I for other mixtures, correlated to the spectral sensitivity of the system and the energy contribution to the gas, I^*. For the mercury triplet line with $\lambda = 546$ nm the branching ratio is 0.53 [13], and the pumping selectivity for the 7^3S_1 level is ~ 0.8 [9]. The excitation selectivity of the $B^2\Sigma_u^+$ state in the He – N_2 mixture is ~ 0.75 [15]. Having compared the luminescence intensity in these mixtures and taken into account that the emission intensity at $\lambda = 585$ nm is 15 % – 20 % of the intensity of all lines of the 3p – 3s transitions in neon [1], one can conclude that the main (which may clearly dominate) channels for populating 3p levels of neon under pumping by a hard ioniser are the processes that are not related to the dissociative recombination of molecular ions.

Table 1. Luminescence intensities for gas mixtures excited by α particles.

Mixture composition	I (rel. units)	λ/ nm	Activity/ GBq	I^* (rel. units)
^{4}He (2 atm) + Ne (50 Torr)	5.69	585	9.6	5.69
^{3}He (2 atm) + Ne (50 Torr)	5.72	585	9.6	5.72
^{3}He (2 atm) + Hg (1.5 mTorr)	15.7	546	16	8.4
Xe (1 atm) + Hg (1.5 mTorr)	79	546	22	13.2
He (4 atm) + N_2 (0.3 Torr)	20.6	391	5.4	13.7
	6.0	427		4.0
He (1 atm) + Ne (1 atm)	7.2	585	9.6	4.6
	3.7	703		(2.4)
Ne (1.3 atm) + Ar (50 Torr)	3.1	585	6.8	2.5
Ne (3 atm) + Ar (50 Torr)	4.3	585	6.8	2.4
	1.5	703		(0.8)

The changes in the luminescence intensity at $\lambda = 585$ nm are related to not only the quenching of the $3p'[1/2]_0$ level by additives (the quenching rate constant is 4.6×10^{-11} cm^3 s^{-1} for H_2 and 5.3×10^{-11} cm^3 s^{-1} for Ar [16]). Taking into account the level lifetime (14.3 ns [13]), we obtain the H_2 or Ar pressure at which the quenching rate becomes equal to the spontaneous decay rate of this level (~ 40 Torr). Apparently, the decrease in the luminescence intensity with an increase in the partial pressure of quenching additive is mainly related to the Penning process [ionisation of additive atoms in collisions with metastable helium atoms (Table 2)].

On the assumption that the neon luminescence intensity at $\lambda = 585$ nm in the He – Ne mixture (50 Torr) with H_2, Ar, Kr, or N_2 additives is determined by the competition between the nonresonant excitation transfer from He (2^3S_1) to neon atoms and the Penning process, one can estimate the quenching additive pressure at which the intensity decreases by half. The satisfactory agreement between the estimated and measured P values suggests that under ionising pumping of He – Ne mixtures with a high helium content 3p levels are populated as a result of excitation transfer from He (2^3S_1) atoms to neon atoms and subsequent 4s – 3p cascade transitions.

Table 2. Quenching parameters for He (2^3S_1) atoms.

Atom, molecule	Process	$k/10^{-11}$cm^3 s^{-1} [17]	$k/10^{-11}$cm^3 s^{-1} [18]	P/Torr estimate	P/Torr experiment
Ne	He(2^3S_1) + Ne → Ne(4s) + He	0.45	0.36		
H_2	Penning	5 ± 3	2.9	~ 7	8
D_2	Penning		2.6	~ 8	9
Ar	Penning	9 ± 5	7.1	~ 3	3.8
Kr	Penning	11		~ 2	2.8
N_2	Penning	7.2 ± 1.4	7.1	~ 3	3.5

An addition of argon (50 Torr) to neon causes a decrease in the luminescence intensity by half at $\lambda = 585$ nm and by a factor of three at $\lambda = 703$ nm (the data for $\lambda = 703$ nm are listed in Table 1 without a correction to the instrumental spectral sensitivity), which approximately corresponds to the quenching of 3p levels by argon. The intense emission at the 3p – 3s transitions upon neon excitation by α particles [14] and spontaneous fission fragments of ^{252}Cf ($W \sim 10^{-8}$ W cm^{-3}) [11] suggests that in pure neon 3p levels are also populated in the processes that are not related to the dissociative recombination of Ne_2^+ ions. When pumping by uranium fission fragments [19, 20], the luminescence intensity of the Ne (0.64 atm) + NF_3 (1.5 Torr) mixture is approximately equal to the emission intensity at the 3p – 3s transitions in neon with an Ar, Kr, or Xe additive (8 Torr). The presence of electronegative impurity in neon, as well as for the He – Ne mixture, does not lead to a sharp decrease in the emission intensity at the 3p – 3s transitions.

Thus, the results of this study indicate that the mechanism of excitation of 3p – 3s transitions in neon during dissociative recombination of molecular neon ions is inconsistent with the experimental data for the case of weak pumping by ionising radiation:

(i) efficient luminescence is observed when quenching additives with a pressure of a few Torr are added to the mixtures studied [in the case of recombination-induced population of levels the emission intensity at $W \sim 3 \times 10^{-5}$ W cm^{-3} would sharply decrease at an H_2 (N_2) pressure of $\sim 10^{-4} - 10^{-3}$ Torr];

(ii) the attachment of electrons to electronegative impurity does not affect the population of the Ne $3p'[1/2]_0$ level.

The most likely population channel for the Ne 3p level appears to be the transfer excitation from metastable helium atoms to neon atoms and direct neon excitation by nuclear particles and secondary electrons (Fig. 3). In neon secondary electrons excite the 3d, 4s, and 5s levels, and the 3p levels are populated via cascade transitions from these levels. The 3d, 4s, and 5s levels, which are radiatively coupled with the ground state, should be efficiently excited by an electron impact. The absence of lines due to the transitions from the 5s levels in [11, 14] is explained by the fact that the probabilities of the 5s – 3p transitions are much lower than the probabilities of the 5s – 4p transitions in the IR spectral region. The 5s – 4p, 4p – 4s, and 4s – 3p cascade

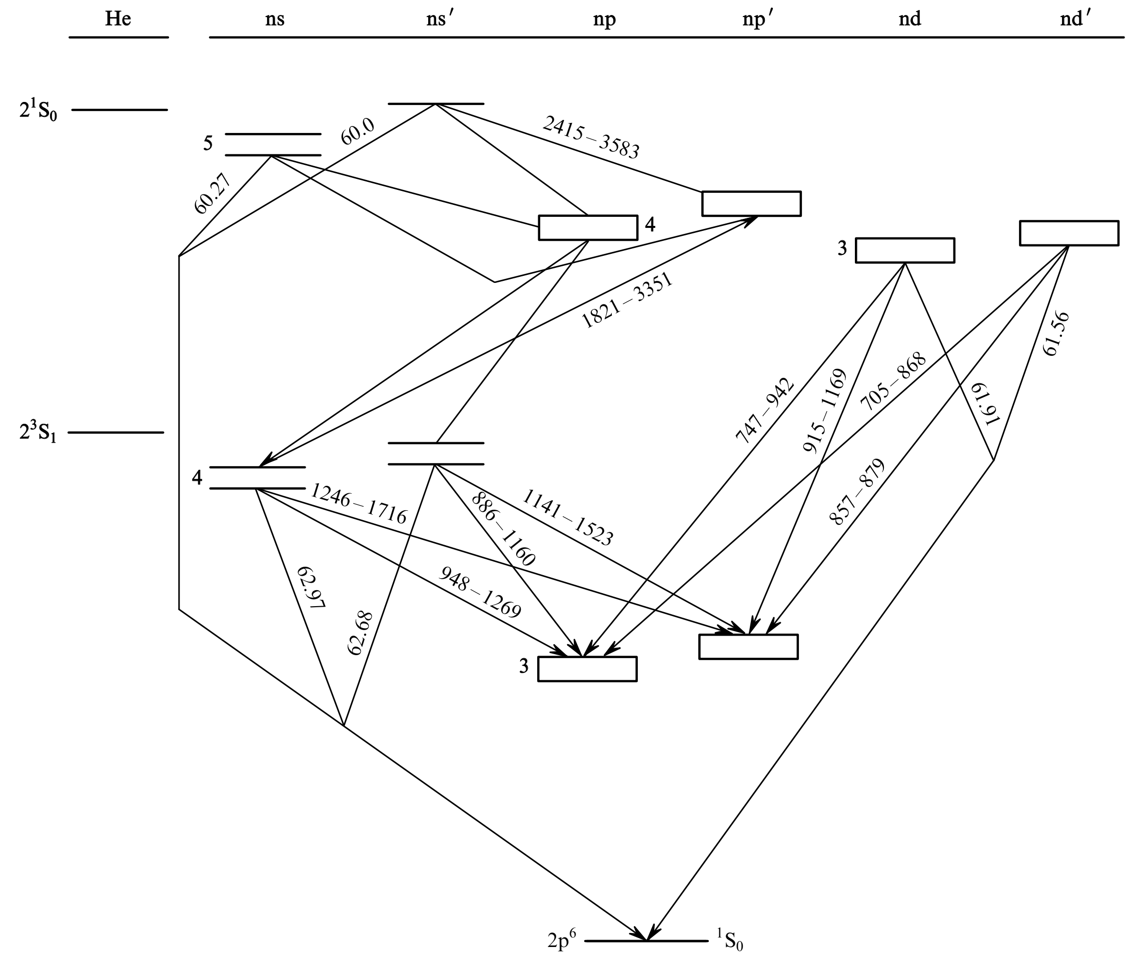

Figure 3. Schematic diagram of population of the 3p levels of neon (the transition wavelengths are in nanometers).

transitions can also contribute to the population of 3p levels of neon. In helium–neon mixtures 3p levels are also populated during nonresonant transfer excitation from metastable He (2^3S_1) and He (2^1S_0) atoms to the Ne 4s and 5s levels and subsequent cascade transitions.

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