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THE POSSIBILITY OF EMPLOYING OPTICAL PUMPING FOR EXCITATION OF NOBLE GAS ATOMS

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To produce an inverted population in the levels of noble gas molecules (Xe_2, Kr_2, etc), it is suggested that an optical excitation method be applied in which use is made of the transitions that occur in triple collisions between a photon and two unexcited atoms. The parameters of the pumping source required for appreciable amplification are estimated.

IN[1] there was considered the possibility of obtaining generation in the vacuum ultraviolet by exciting helium, neon, argon, krypton, and xenon in the liquid and aggregate states with an electron beam. It turns out that the inverted population can be obtained either between the conduction and valence bands, or by using localized exciton states. In[2] it was shown that the excitons produce localized complexes of the type of excited diatomic molecules of a noble gas. Therefore the inversion mechanism proposed in[1] can be readily extended to molecules of noble elements in the gas phase.

These molecules are of interest because their lower energy state is unstable. The lifetime of the molecule in this state is equal to the time of separation of the atoms. The lifetime of the excited molecule is determined by the probability of radiative and nonradiative transitions to the lower electronic state. The transition with emission of a quantum of light leads to the occurrence of continuous-spectrum bands[3], and the molecule dissociates into two atoms in the ground state $^1S_0$.

The emission spectra of noble gases, obtained by excitation with electric or microwave discharges, have been thoroughly investigated in a number of papers (see, for example,[3,4]). An essential feature of the spectra is the presence of two emission bands, one located in the region of the first resonance line of the atom, and the second in the longer-wave region. For example in the case of xenon one band has a maximum near 1540 Å and the second at 1750 Å. These bands correspond to transitions from two different excited electronic states of the molecule to the ground state[3]. The shorter wavelength band corresponds to the transition between the levels $^1\Sigma_u^+$ and $^1\Sigma_g^+$ ($^1\Sigma_g^+$ is the ground state), while the long-wavelength band corresponds to the transition $^3\Sigma_u^+ \to {}^1\Sigma_g^+$. Similar emission spectra were observed also in the bombardment of noble gases by α particles[2].

In view of the fact that the lifetime of the molecule of the noble gas at the lower electronic level is small, it is possible in principle to obtain inverted population in the system of levels of the molecule. The gain will be determined by the radiative lifetime of the molecule, by the width of the emission band, by the rate of formation of the excited molecules, and by the value of the losses.

In the gas phase, as will be shown below, it is advantageous to excite the molecules by an optical method that employs the transitions $^1\Sigma_g^+ \to {}^1\Sigma_u^+$. Such transitions take place in triple collisions between one photon and two unexcited atoms of the noble gas. This phenomenon was observed in[5] in the study of absorption in xenon. Recently, molecular absorption was investigated in argon[6], krypton[7], and xenon[8,9]. A feature of the considered process is the quadratic (and at high pressures even stronger) dependence of the gain on the pressure. This is connected with the fact that at each instant of time the density of the dimers of the noble gases (Xe_2, Kr_2, etc.) is equal to $n_0 = (1/2)N^2\langle\sigma_{\text{coll}}v\rangle\tau_{\text{coll}}$, where N is the number of atoms per unit volume, σ_{coll} is the collision cross section, v is the average velocity, and τ_{coll} is the collision time, equal to $\sim\sqrt{\sigma_{\text{coll}}}/v$. The widths of the absorption band are quite appreciable even at pressures of several hundred Torr (2000–3000 cm^{-1} for xenon in the 1470 Å region at a pressure of 400 Torr[5]).

The luminescence of optically-excited xenon was investigated in[10]. It was shown that when light is absorbed at a wavelength 1470 Å, only one symmetrical band is emitted, with a center at 1715 Å. The emission intensity increases with increasing xenon pressure, reaching saturation (the pressure rose to several hundred Torr). This result apparently indicates that all the molecules excited in the state $^1\Sigma_u^+$ go over very rapidly (within a time shorter than the radiative lifetime in the state $^1\Sigma_u^+$) into a state corresponding to a smaller internuclear distance than in the unexcited molecule, i.e., a process perfectly analogous to the formation of the localized exciton in the liquid[2] takes place. The growth of the intensity with increasing pressure is due to the absence or quenching of excited molecules in collision with xenon atoms and to the decrease of the influence of the cell walls. Analogous conclusions can be reached by investigating the spectra of the luminescence of noble gases bombarded by α particles[2] and the results of[11], where light flashes in Geiger counters filled with argon were investigated.

The main source of losses in noble gases is apparently photoionization of the excited molecule[1]. It may turn out that the photoionization cross section at the frequency of the working transition is larger than the amplification cross section. In this case no lasing can be achieved. In view of the lack of exact data on the cross sections, we shall estimate the parameters of the pump source without taking photoionization into account.

The rate of formation of the excited molecule is $w = \sigma_p n_0 I$, where σ_p is the cross section for the absorption of the pump photons, and I is the intensity of light in photons/cm^{2}sec. We shall assume that all the excited molecules relax very rapidly to the level $^3\Sigma_u^+$, the lifetime τ of which is determined only by the emission. Then, after a time τ, there is established at this level an equilibrium molecule concentration equal to $n^* = w\tau$. According to the Franck-Condon principle, the molecules emit at a constant internuclear distance corresponding to the configuration of the excited molecule. The appreciable value of the Stokes frequency shift of the luminescence relative to the pump frequency indicates that the potential energy has a large slope in the lower electronic state. This gives grounds for assuming that there are no particles at the lower level at the internuclear distance determined by the dimensions of the excited molecule. Therefore the gain k is equal to[12]

$$k = \frac{1}{4}\frac{\lambda^2 n^*}{\tau\Delta\omega} = \frac{1}{4}\frac{w}{\Delta\omega}\lambda^2,$$

where $\Delta\omega$ is the width of the emission band at half the height and λ is the emission wavelength. We note that the amplification of the light by centers having Stokes shifts of the absorption and emission spectra was considered in[13].

The expression for k does not contain the lifetime of the excited molecule. It must be pointed out, however, that the amplification cross section is larger than the photoionization cross section σ_{ph} only when $\tau < \lambda^2/4\sigma_{ph}\Delta\omega$. The values of the lifetime for molecules of noble gases were determined experimentally only for He_2 ($\tau = 0.05$ sec[14]) and Ar_2 ($\tau = 3.4 \times 10^{-6}$ sec[11]). From a comparison of the conditions for the excitation of the luminescence spectra of noble gases in electric discharges[3] it follows that for such molecules (Kr_2, Xe_2) the lifetimes will apparently be shorter (10^{-6}–10^{-8} sec). We shall make the numerical estimates for Xe_2.

The width of the emission band of Xe_2 in the 1715 Å region is 5×10^3 cm^{-1} [10]. In order for k to equal 10^{-3} cm^{-1} (corresponding to a gain of 10% per meter), it is necessary that the rate of formation of the excited molecules w be equal to 10^{22} sec^{-1} cm^{-3}. This value of w corresponds to $n^* = 10^{14}$–10^{16} cm^{-3} at $\tau = 10^{-8}$–10^{-6} sec, or to an amplification cross section σ ($k = \sigma n^*$) equal to $\sigma = 10^{-17}$–10^{-19} cm^2.

If we choose the gas pressure in such a way that the depth of penetration of the pump light $(\sigma_p n_0)^{-1}$ is equal to 0.1–1 cm, then to obtain a gain $k = 10^{-3}$ cm^{-1} it is necessary to have a source with intensity $I = 10^{21}$–10^{22} photon/cm^{2}sec. Such an intensity is obtained in the xenon absorption band from a black body radiating at a temperature 10,000–20,000°. At the present time it is not difficult in principle to produce such light sources with dimensions ~ 1 m[15].

The foregoing inversion mechanism may turn out to be useful not only in noble gases. It is known that collisions of excited and unexcited atoms produce molecules in vapors of certain metals (mercury, cadmium, etc.)[16]. In some organic substances and gases, formation of entire complexes (excimers) have been observed when excited and unexcited molecules are joined together[16]. All these processes greatly influence the absorption and emission spectra. The use of transitions in systems in which the ground state is unstable can greatly extend both the class of substances used in quantum radiophysics and the spectral range of the working devices.

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