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Emission of the ^{3}He – Xe – Cd mixture in the active zone of a nuclear reactor

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***Abstract*. The luminescence spectrum of the ^{3}He – Xe – Cd mixture in the wavelength range 350 – 830 nm is studied at a gas temperature 250 – 600 °C. The mixture was excited by the products of the nuclear reaction ^{3}He(n, p)T. The mechanism of population of CdI levels is discussed and the rate constant of the charge exchange of Xe_2^+ ions with cadmium atoms is estimated as $k \sim 10^{-13}$ cm^3 s^{-1}.**

Keywords: *luminescence, cadmium, xenon, nuclear pumping, laser.*

1. Introduction

Mixtures of inert gases with vapour of group II metals are promising active media for developing high-power efficient lasers pumped by a hard ioniser [1 – 3]. The use of (He) – Xe – Hg – H_2 for obtaining lasing at the triplet lines of mercury was proposed in Refs [3 – 5]. Pumping of the $7\,^3S_1$ state of HgI occurs upon dissociative recombination of Hg_2^+ ions. The rate constant of deactivation of the lower laser level by hydrogen is an order of magnitude higher than for the upper level [3]. The use of xenon as a buffer gas ensures ionisation of mercury atoms (without charge exchange with H_2). Using such a setup, quasicontinuous generation was realised at the mercury atom $7\,^3S_1 - 6\,^3P_2$ transition in a pulsed nuclear reactor [6].

A similar scheme was used for exciting mixtures of mercury with inert gases by using an electron beam [7]. In this study, a mixture of He – Ne – Ar a total pressure of 2300 Torr was used as the buffer gas. Citing a publication more recent than [3, 8], the authors of [7] indicate that the main channel of population of the $7\,^3S_1$ state is the recombination of Hg_2^+. Attempts to use H_2 for depopulating the $6\,^3P_2$ level were not successful, and generation was quenched at a hydrogen pressure of 20 Torr [7]. In our opinion, this is due to the fact that the molecular helium ions formed in the plasma effectively exchange charge with Ar and Ne atoms; in turn, Ar_2^+ and Ne_2^+ ions exchange charge with H_2 molecules [9], and the rate constants of charge exchange of Ar_2^+ and Ne_2^+ ions with Hg are low (5×10^{-11} cm^3 s^{-1} for Ne_2^+ and less than 10^{-12} cm^3 s^{-1} for Ar_2^+ [10]). Since the mercury vapour pressure was ~ 1 Torr, the beam energy was spent on ionising H_2 molecules rather than the mercury atoms. The main channel of ionisation of mercury in such a mixture is the Penning process involving excited argon atoms.

The energy level diagram for cadmium atoms is analogous to the energy level diagram for mercury atoms. Hence, it should be interesting to study the possibility of quasicontinuous lasing at the $6s-5p$ transitions of Cd. The quantum efficiency of such a system (ratio of the photon energy to the energy of electron – ion pair formation in Xe) exceeds 11 %, which is much higher than for the cadmium ion transitions in the He – Cd mixture. In this work, we shall study the emission spectrum of the ^{3}He – Xe – Cd mixture in the active zone of a nuclear reactor.

2. Experimental

The setup for spectral measurements in a reactor was described in Refs [4, 5]. A glass ampoule of length 60 mm and diameter 30 mm with a radiation-resistant cerium window at the end face was heated for several hours in a vacuum of $\sim 10^{-5}$ Torr. After this, the ampoule containing a piece of Cd-116 was filled with ^{3}He under a pressure of 600 Torr, or with a mixture of ^{3}He (200 Torr) and Xe (400 Torr), and placed in the central channel of the active zone of a nuclear VVR-K reactor in steady state. The lower part of the experimental channel of height 700 mm was made of two concentric tubes of X18H10T steel with a gap of about 2 mm between them. The temperature of the ampoule could be varied by changing the pressure of ^{4}He in the gap. During spectral measurements, the temperature of the bottom of the experimental channel was measured first, after which a test ampoule with a thermocouple was used for temperature calibration of the ampoule containing Cd-116. The error in measuring the temperature of the ampoule (in degrees Celsius) was ~ 10 %. The contents of the ampoule were excited by the products of nuclear reaction ^{3}He(n, p)T, the thermal neutron flux density was 1.0×10^{13} neutrons cm^{-2} s^{-1}. Light from the active zone of the reactor was extracted through a 6m-long tube and was detected by a SPM-2 monochromator with a quartz prism and a FEU-106 photomultiplier operating in the photon counting mode. The spectral range (350 – 830 nm) was determined by the transmission of the ampoule window and the photomultiplier sensitivity.

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3. Results and discussion

The emission spectrum of the ^{3}He – Cd mixture mainly coincides with the one described in Refs [11, 12] and differs from it only in the ratio of the intensities of certain lines. The continuous spectrum is formed by the radiation emitted by Cd_2^* molecules. The dependence of the intensity of atomic lines at 508.6 nm ($6^3S_1 - 5^3P_2$) and 643.8 nm ($5^1D_2 - 5^1P_1$) on the temperature of the ampoule containing Cd-116 is shown in Fig. 1 (hereafter, the line intensities are uncorrected for the spectral sensitivity of the setup, 1 unit = 1000 pulses s^{-1}). In addition to the cadmium triplet lines at 467.8, 480 and 508.6 nm and the line at 643.8 nm, the atomic lines also include the line at 361.0 nm ($5^3D_3 - 5^3P_2$) with a maximum intensity of about 5.5 units at a temperature of 350 °C. For comparison, the maximum intensity of the brightest ionic lines at 441.6 nm under our conditions was 110 units at a temperature of 360 °C.

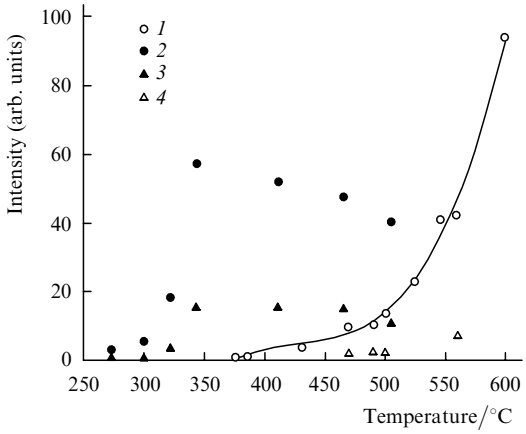

Figure 1. Dependence of the intensity of the 508.6-nm (*1*, *2*) and 643.8-nm lines (*3*, *4*) in the ^{3}He – Cd (*2*, *3*) and ^{3}He – Xe – Cd (*1*, *4*) mixtures on temperature of the ampoule with cadmium-116.

The intensity of the 361.0-nm line is below the sensitivity threshold of the setup (~ 0.1 units) in the ^{3}He–Xe–Cd mixture. As expected, there are no CdII lines. Similarly to mercury containing mixtures, cadmium energy levels are apparently excited due to the following processes (M is the third particle):

$$Xe + (p, T) \rightarrow Xe^+ + (p, T) + e, \quad (1)$$

$$Xe^+ + Xe + M \rightarrow Xe_2^+ + M, \quad (2)$$

$$Xe_2^+ + Cd \rightarrow Cd^+ + 2Xe, \quad (3)$$

$$Cd^+ + Xe + M \rightarrow CdXe^+ + M, \quad (4)$$

$$CdXe^+ + Cd \rightarrow Cd_2^+ + Xe, \quad (5)$$

$$Cd_2^+ + e \rightarrow Cd^* + Cd \quad (6)$$

(it is assumed that, as in the Xe–Hg mixture [10], the rate constant for Xe^+ charge exchange with cadmium atoms is $k_a < 10^{-10}$ cm^3 s^{-1}).

The population of the 6^3S_1 and 5^1D_2 levels of cadmium may occur directly in the course of reaction (6) or through a cascade from higher levels. The wavelengths of such transitions lie outside the sensitivity range of the photomultiplier. Lasing at transitions $6^3P_1 - 6^3S_1$, and $4^3F_2 - 5^3D_3$ in the He–Cd mixture was reported in Ref. [13]. The absence of the 361.0-nm line in the emission spectrum indicates that either the population density of the 4^3F_2 level is insignificant in the He–Xe–Cd mixture, or the levels 4^3F_2, and 5^3D_3 are strongly quenched by xenon. The highest intensity of the 508.6-nm line in the ^{3}He – Xe – Cd mixture is attained at a much higher temperature than in the ^{3}He–Cd mixture (see Fig. 1). Apparently, this is due to the fact that the charge exchange of Xe_2^+ with Cd occurs slowly. Figure 2 shows the dependence of the radiation intensity at 508.6 nm on the density N of cadmium atoms. The density of cadmium atoms (in cm^{-3}) was calculated by the formula [14]

$$N = \frac{1.41 \times 10^{27}}{T_g} \exp\left(-\frac{12550}{T_g}\right). \quad (7)$$

Here, T_g is the absolute temperature of the gas in kelvins.

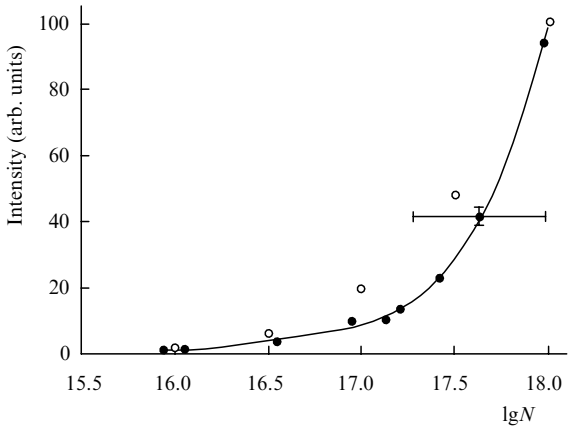

Figure 2. Dependence of the intensity of the 508.6-nm line (●) on the density of cadmium vapour in the ^{3}He–Xe–Cd mixture. Light circles correspond to calculations by expression (8) for $I_\infty = 200$ units, $k = 10^{-13}$ cm^3 s^{-1}.

It can be shown [4] that if the population of cadmium levels occurs due to processes (1)–(6), the dependence of the intensity on N can be described by the expression

$$I = \frac{kN}{kN + (\alpha S)^{1/2}} I_\infty, \quad (8)$$

where I_∞ is the intensity for high density of cadmium atoms; k is the rate constant of process (3); and $\alpha \approx 10^{-6}$ cm^3 s^{-1} is the electron–ion recombination constant, which is assumed to be identical for the primary ions. The gas ionisation rate ($S \approx 10^{16}$ cm^{-3} s^{-1}) was estimated from known values of the nuclear reaction cross section and the mean free path of proton and triton in the gas. Fitting of the experimental dependence by expression (8) leads to the estimates of I_∞ (~ 200 units) and k (~ 10^{-13} cm^3 s^{-1}).

Thus, the rate constant of charge exchange of Xe_2^+ with cadmium atoms is small in contrast to the rate constant of

charge exchange of Xe_2^+ with mercury atoms. A very high density of cadmium vapour ($\sim 3 \times 10^{18}$ cm^{-3}) is realised at a temperature of about 700 °C. For such a high density, the quenching of the $6\,^3S_1$ state by its own atoms must be taken into account, although no noticeable quenching was observed in the range of densities investigated by us. The dependence of the intensity on N is described quite well by expression (8) (Fig. 2). It is possible that charge exchange of krypton with Cd will be faster, and cadmium atoms can also be ionised during the Penning process.

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