AN EXPERIMENTAL STUDY OF THE CORRELATION BETWEEN ION AND ATOMIC SPECTRA OF Eu IN THE He-Eu MIXTURE

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A correlation between the ^7D_j *states of* Eu$^+$ *and the atomic transition system* $z^{10}D_j \to a^{10}D_j^0$ *in a gas discharge has been revealed by the method of selective perturbation of populations of the* ^7D_j *states of* Eu$^+$*. A scheme is proposed for the production of an inverted medium in a* He–Eu$^+$ *laser. A feature of this scheme is the relaxation of the metastable* $^7D_j^0$ *states during the process of electron capture and excitation of* Eu *atoms to field ionization states.*

The mixture of europium and helium vapors in which oscillations through the transitions from the $^{7,9}P_j$ resonance levels of the Eu ion to its $^{7,9}D_j^0$ metastable levels occur on excitation by a gas discharge is an unique laser medium. The uniqueness lies in the fact that under steady-state excitation conditions the $^7D_j^0$ metastable states relax for about 10^{-9} s. This provides steady-state oscillations restricted only by the discharge constriction. An important peculiarity of this mixture is the high helium pressure. The best pressure should be much higher than the atmospheric pressure, and the threshold He pressure should be higher than 180 Torr to initiate oscillations at the wavelength $\lambda = 1.002$ μm through the $^7P_4 \to {}^7D_5^0$ transition.

Despite the fact that this mixture was investigated in a number of experiments,[1–3] an adequate mechanism of relaxation of the $^7D_j^0$ metastable states was not found. At present there are at least two versions of this mechanism.[1] The first version implies the relaxation which occurs due to binary collisions of helium atoms with excited ion states of europium in intersecting the terms of the (EuHe)$^{+*}$ quasimolecules. The second version proposes recombination of the europium ion, i.e., the relaxation is assumed to proceed through the shifted states of the Eu atom created in the course of electron capture by europium ions and at the stabilization of this process due to the collisions with the helium atoms. In the shifted spectrum the Eu atom can be ionized by radiation into excited states or into the ground state Eu$^+$, or it can go through collisions into an ordinary atomic spectrum. The main argument in favor of this mechanism which was never observed in gas lasers is a very complex, almost quasicontinuous spectrum of the shifted states of Eu atoms in the range 3-4 eV being over the ionization threshold of the atom.[4]

Therefore, further investigation of the physical processes occurring in the He–Eu mixture is of interest. This paper describes an experimental study of the perturbations in the populations of the Eu ionic and atomic states, caused by resonance radiation on its action on one of the optical transitions in the ion spectrum.

Let us consider the structure of the Eu$^+$ lower excited states shown in Fig. 1, to understand the essence and the results of the experiment. This structure consists of two weakly coupled systems, one having a multiplicity of 7 and the other of 9. Typical "self-limited" oscillations are observed in each system on pulsed gas discharge excitation. A steady-state inversion limited in time by the discharge constriction is readily attained through the $^7P_4 \to {}^7D_5^0$ transitions at an elevated He pressure.

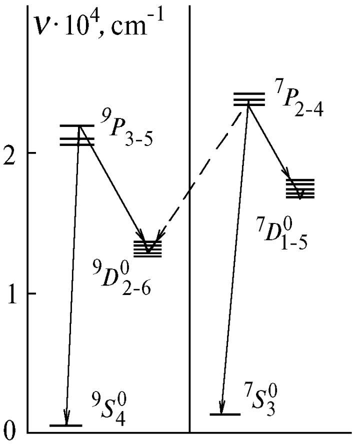
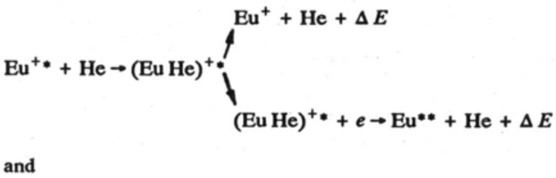


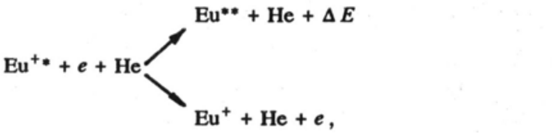
FIG. 1. Simplified scheme of levels and optical transitions in the spectrum of the europium ion.

As can be seen from Fig. 1, the $^7D_j^0$ metastable states are in fact isolated. Therefore, the simple way of relaxation through nonadiabatic collisions with He with energy transfer to the lower terms is impossible. Two of the above-mentioned processes



$$\mathrm{Eu}^{+*} + \mathrm{He} \to (\mathrm{Eu\,He})^{+*} \begin{cases} \nearrow \mathrm{Eu}^+ + \mathrm{He} + \Delta E \\ \searrow (\mathrm{Eu\,He})^{+*} + e \to \mathrm{Eu}^{**} + \mathrm{He} + \Delta E \end{cases} \quad (1)$$

and



$$\mathrm{Eu}^{+*} + e + \mathrm{He} \begin{cases} \nearrow \mathrm{Eu}^{**} + \mathrm{He} + \Delta E \\ \searrow \mathrm{Eu}^{+} + \mathrm{He} + e, \end{cases} \quad (2)$$

where (EuHe)$^{+*}$ is the excited molecular ion and Eu** the shifted state of the europium atom can be found by a

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response in the atomic spectrum if one periodically acts on the concentration of any component of the mixture. The simplest way to introduce a periodical perturbation into the Eu^{+}* ($^7D^0_j$) state density is to use the intrinsic laser field of the He–Eu laser to transfer some part of the population from the $^{7,9}P_j$ resonance to $^7D^0_j$ metastable states.

For the oscillating mode the initial perturbation in the pumping rate of the $^7D^0_j$ state can readily be found from the output energy of the laser. Its typical value is $\Delta\beta = 2 \cdot 10^{12}$ particle $\cdot$ cm^{-3} per excitation pulse of duration 1 μs. This substantial value of perturbation was good reason to hope that it could be found in experiment as a growth of intensity of spectral lines, if only a small portion of the perturbation reached the atomic spectrum.

The experimental setup shown in Fig. 2 was used to investigate the optical response in the atomic spectrum to perturbation in populations in the ion spectrum.

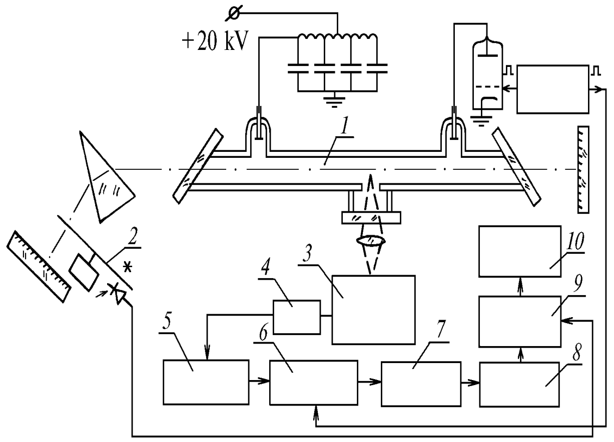

FIG. 2. Block diagram of the setup.

The setup included the He–Eu laser *1* with a selective resonator tuned to λ = 1.002 μm. The shutter *2*, which periodically interrupts the resonator with a frequency of 1 kHz, was placed into the laser resonator. The discharge tube had a side window to observe the spontaneous emission from the discharge. The discharge was initiated at a frequency of 10 kHz by pulses of duration 1 μs and the required temperature regime of the tube (~ 600° C) was provided. The radiation of the tube passed through the monochromator *3* and was recorded with the PMT *4*. The high repetition rate of initiating pulses made it possible to use a wideband system for separation of signals from noise based on the gating followed by transformation of the signals into the low-frequency region. To this end, the PMT signals, – a 10 kHz pulse sequence amplitude modulated with a frequency of 1 kHz – was amplified with the help of a wideband amplifier whose output was connected to the peak voltmeter *7* by the electronic switch *6* for 0.5 μs in synchronism with initiating pulses. The peak voltmeter had a time constant of 10^{-3} s. The 1 kHz variable component was amplified at the peak voltmeter output using the selective amplifier *8*. It was detected by the synchronizing detector *9* and recorded by the recorder *10*.

This design of the recording system allowed the amplitude and phase of the perturbations in line intensity to be recorded with the laser field being present or absent in the resonator.

When the shutter was placed in front of the monochromator, the setup recorded the total lateral radiation of the discharge. The records of the entire spectrum are needed to identify the lines on which the intensity modulation occurs and to estimate the modulation percentage.

Figure 3 shows one of the signal record fragments corresponding to the green region of the spectrum. In this region, the atomic lines of Eu essentially radiate. Their density is not large, so that the spectrum is resolved using an MDR–3 monochromator. As can be seen from the record, modulation signals of different sign take place for almost all the lines on a level close to the noise level. However, there are some lines on which the modulation signal is substantial. These lines are listed in Table I. All the lines belong to the transitions $z^{10}D_j \rightarrow a^{10}D^0_j$ (32117–30945 cm^{-1}) → (12923–13778 cm^{-1}) of the configurations $4f^7(^8D^0)5d(^9D^0)6p$ and $4f^7(8s^0)5d(^9D^0)6s$, respectively. Thus the correlation between the ion and atomic spectra is readily found. Table I lists the lines on which a substantial modulation of the populations was observed.

TABLE I. Atomic spectrum lines showing the modulation percentage of intensities more man 1%.

Wavelength, nm	Upper state $z^{10}D_j$; E, cm^{-1}	Lower state $a^{10}D^0_j$; E, cm^{-1}
545.1		13/2, 13779
535.7	13/2, 32117	11/2, 13547
540.2		9/2, 13322
547.2	11/2, 312726	11/2, 13457
557.0		13/2, 13779
545.2		7/2, 13049
557.7	9/2, 31382	11/2, 13457
548.8		5/2, 12923
552.6	7/2, 31138	7/2, 13049
558.0		9/2, 13222
554.7		5/2, 13049
558.6	5/2, 30945	7/2, 13049

Investigations of the shorter wavelength range of the spectrum were unsuccessful, since the discharge spectrum in this range failed to be resolved because of the high line density.

In analyzing the recorded response in the atomic spectrum to the perturbations in the particle flux in the ion spectrum, some peculiarities should be noted. First, taking into account the Eu spectrum well developed due to the competition between the outer and inner shells, it should be noted that considerable perturbations in population (about 8%) show up only in the above-mentioned transition system with a multiplicity of 10. Second, there exists a correlation between the line intensity and its modulation: at least, in the shown fragment the more intense lines belong to this $z^{10}D_j \rightarrow a^{10}D^0_j$ system. Furthermore, the fragment involves the resonance line λ = 576.6 nm and some lines on which no modulation occurs or the modulation is not in excess of the noise level. And, finally, there are (~ 1 %) weakly modulated lines with the modulation phase being opposite to the set of lines shown in Fig. 3.

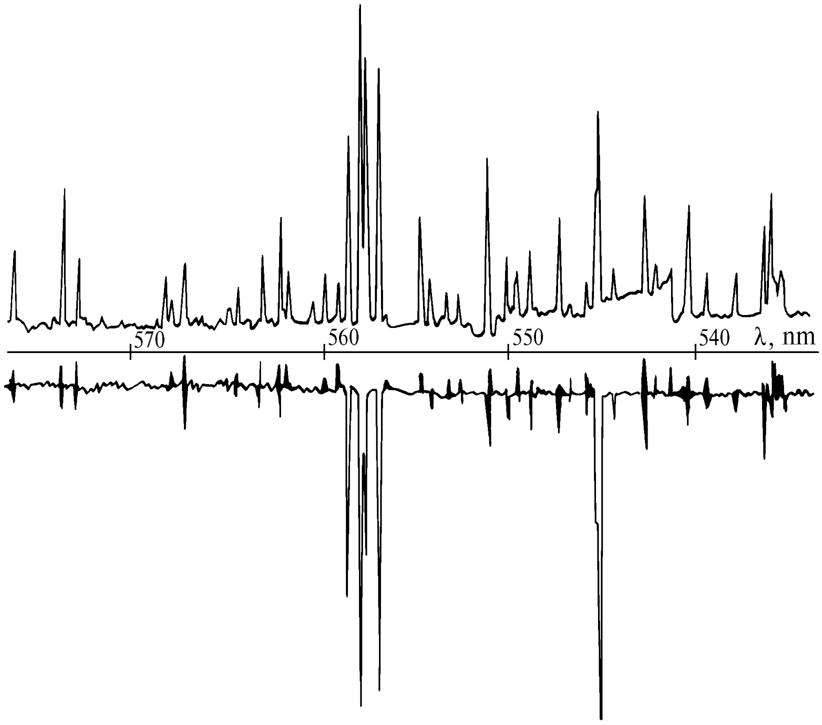

FIG. 3. Fragments of records of the complete spectrwn (at the top) and of the spectrum recorded mm a modulator present in the laser resonator (at the bottom).

The above noted spectral selectivity of line modulation requires an analysis of the possibility for exciting the z^{10}D? states through some resonance processes, such as direct ion—atomic collisions involving the excited states of both the atom and ion. Such an analysis has shown that in this case an energy resonance is actually observed for the reaction

$$\mathrm{Eu}^*(a^{10}D_j^0)+\mathrm{Eu}^+(^7D_j^0)\leftrightarrows\mathrm{Eu}^*(z^{10}D_j)+\mathrm{Eu}^+(^{7,9}S_{3,4}^0)+\Delta E, \quad (3)$$

However, estimations of the excitation rates 0 of the $z^{10}D_j^0$ states from the ground state of the atom 0_O and through reaction (3), gives $\beta_3 \simeq (10^{-3}–10^{-4}) \cdot \beta_0$ owing to the small concentration of the particles involved in reaction (3). If the energy resonance for reactions of type (3) is taken into account alone, then the modulation could also be expected for some lines in the given spectrum fragment. This effect, however, has not been observed.

The lines listed in Table I have rather small (10^{-19} cm^2) cross sections for the electronic excitation from the ground state. In this connection the correlation between the intensities of lines and the observed modulation is an indication of the predominant "from–above" excitation from the ionization state.

Analysis of spectral lines with small modulation factors and the modulation phase being opposite to that for the $z^{10}D_j \to a^{10}D_j^0$ transitions has shown that the lines with the outlined modulation characteristics have lower levels common to the $z^{10}D_j \to a^{10}D_j^0$ system.

Taken together, the described facts unambiguously point to the fact that the laser excitation channel for the $^7D_j^0$ states in the Eu ion additionally (by about 8%) increases the population of the $z^{10}D_j$ and $a^{10}D_j^0$ states in the atomic spectrum. Comparison of the absolute relaxation rates of the $^7D_j^0$ states in the ion spectrum with the excitation rates of the $z^{10}D_j$ state from the atomic ground level using the electron excitation cross sections given in Refs. 5 and 6 demonstrates that the particle flux observed by us and providing the growth in line intensity in the atomic spectrum equal 8% corresponds to 1% of the particles involved in the oscillations at the wavelength λ equal 1.002 μm. Possibly, there exist some other channels for the relaxation in the atomic spectrum, but it is most likely that the majority of the particles reverts to the first ionization state due to field ionization.

Based on the results, we propose a scheme for the production of an inverted medium through Eu ion transitions shown in Fig. 4.

Within the framework of the scheme, the He-Eu laser should possess, on the one hand, some features of the lasers operating through the transitions from resonance to metastable levels and, on the other hand, some features of the lasers operating by a recombination mechanism. As an illustration of the manifestations of the recombination mechanism in Fig. 5 we show the temporal behavior of the oscillations at the wavelength λ = 1.002 μm, the spontaneous radiation at λ = 617.3 nm controlling the population of the $^7P_4$ upper operating state, the discharge current, and the absorbed radiation at λ = 1.002 μm. As follows from this figure, the upper-level population decreases in the late phase of the discharge, while, the medium, on the contrary, bleaches and oscillations at λ = 1.0019 μm occur. But it is possible if the relaxation rate of the lower (and, likely, upper) $^7D_j^0$ level increases towards the end of the current pulse in synchronism with increase in electron density. In this connection the complicated transient processes occurring in the initial stage of the discharge on varying the Eu vapor density engage our attention. At elevated vapor densities (~ 10^{-1} Torr), oscillations occur only at the initial stage of the discharge and typically last for 200 ns. With decrease of the Eu concentration and for all other factors being equal, oscillations appear in the mid-phase of the discharge. Finally, under low (~ 10^{-2} Torr) Eu pressures, oscillations occur in the late phase of the current pulse.

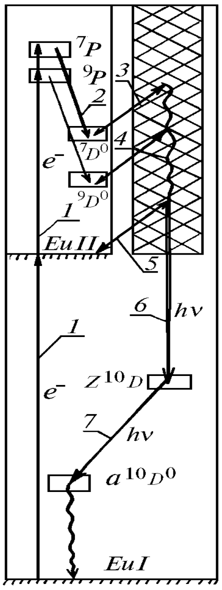

*FIG. 4. Scheme of excitation and relaxation of the He-Eu laser operating states (*λ = 1.002 μm*): 1) electron impact ionization and excitation, 2) laser transition, 3) recombination into shifted atomic states, 4 and 6) collisional and radiative stabilization of recombination, 5) field ionization, and 7) one of the ways for radiative relaxation.*

The metallic conductivity in the discharge converts to the conductivity connected with the buffer gas ionization within the above-mentioned Eu concentration range, the E/p_i parameter and the characteristic times of discharge development are changed correspondingly. On this basis, it is clear that oscillations only occur when the Eu ions reside in a sufficiently dense matrix of "hot" electrons. This is the salient feature of lasers operating by the transitions from resonant to metastable levels. In the first case such conditions are created due to the ionization of europium itself but for a short time. In the latter case these conditions are steady-state but they exist due to the ionization of helium.

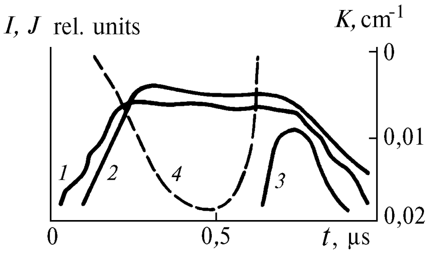

FIG. 5. Wave forms of current I (1), spontaneous radiation J at λ = 617.3 nm *(2), oscillations (3), and radiation absorption* κ *at* λ = 1.002 μm *(4) at* P_{He} = 180 Torr *in the* He-Eu *mixture.*

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