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Characteristics and Parameters of Nanosecond Air Discharge Plasma between Chalcopyrite Electrodes

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Abstract—Characteristics and parameters of nanosecond-discharge plasma in a system with sharply inhomogeneous distribution of the electric-field strength between $CuInSe_2$ electrodes in atmospheric-pressure air are presented. The time dependence of the electron temperature is determined by analyzing the emission dynamics of electrode-erosion products from chalcopyrite at an interelectrode distance of 2 mm.

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Chalcopyrites of the $CuInS_2(Se_2)$ and $CuSbS_2(Se_2)$ type are among the most promising materials for use in photovoltaic devices (solar cells), which is due to the large absorption coefficient in the visible and near-IR spectral ranges and their high light stability [1]. It is of importance for practical use of these materials to obtain high-quality thin films of the corresponding stoichiometric composition or nanostructures based on them.

In [2, 3], the temperature and density of electrons, motion velocities of indium and copper atoms, and recombination rates of metal ions in laser erosion plasma from a polycrystalline $CuInSe_2$ target in a vacuum were studied by time-resolved emission spectroscopy. Nanostructures based on the compound $CuInSe_2$ have been synthesized and their basic characteristics studied also in spark and pulsed arc discharges in ethanol containing a stoichiometric mixture of powdered copper, indium, and selenium [4]. Synthesis of nanostructures of chalcopyrites $CuFS_2$ in a laser torch and in a spark discharge in distilled water and a study of their characteristics were reported in [5], and synthesis of aerosol metal nanoparticles with sizes of 8–75 nm in an air flow excited in a multiple-gap gas-discharge generator was examined in [6].

Recently, considerable attention has been given to analysis of the physics and characteristics of processes in nanosecond discharges with runaway electrons ignited in systems with an inhomogeneous electric-field strength distribution in nitrogen or atmospheric air [7–9]. All these studies used simple metallic electrodes unsuitable for syntheses of complex-composition nanostructures from materials of chalcopyrite type.

The present communication reports on a study of characteristics and parameters of a nanosecond discharge with runaway electrons between chalcopyrite electrodes, which can be used to synthesize the corresponding nanostructures.

Cylindrical and rough electrodes with a diameter of 6 mm made of the polycrystalline $CuInSe_2$ material and mounted in a stainless-steel chamber had a curvature radius of their working-edge surfaces of approximately 1–2 mm. The interelectrode spacing was 2 mm. The air pressure in the chamber was 1 atm.

The discharge was ignited using a generator of high-voltage bipolar nanosecond pulses with resonant recharging of a pulse storage capacitor (1.54 nF) and TGI-I-1000/25 hydrogen discharge valve as a switch. The voltage pulse of single polarity at the modulator output had an amplitude that could be varied within the range of 10–25 kV with a pulsed cable transformer and a width of 20–30 ns. The pulse repetition frequency could be varied within the range of 40–1000 Hz, but was 50–100 Hz in most cases.

The emission was analyzed with MDR-2 (200–650 nm) and DFS-12 (360–650 nm) monochromators. The emission intensity oscillograms of the laser-plasma spectral lines were measured with an ELU-14 FS linear electron multiplier and 6-LOR 04 oscilloscope. The time resolution of the system was approximately 2–3 ns. The oscillograms are presented with consideration for the spectral sensitivity coefficient of the ELU-14 FS + MDR-2 system. The experimental setup was described in more detail in [10].

The time-averaged emission spectra of the discharge were presented in [10, 11]. In emission analysis with the MDR-2 monochromator, the highest intensity spectral lines were those of the erosion products of chalcopyrite: Cu I: 324.7, 327.4, 400.3, 407.3, 427.5, 464.3, 510.6, 515.3, 521.8, 570.0, and 573.2 nm; In I: 283.7, 293.3, 303.9, 325.6, 410.2, and 451.1 nm. No spectral lines of atoms and atomic selenium ions were found. At the same time, analysis of the plasma emission in the spectral range of 200–250 nm revealed

Fig. 1. Photograph of the glow of a nanosecond-discharge plasma in atmospheric-pressure air between electrodes made of polycrystalline chalcopyrite $CuInSe_2$ (the vertical size of the glow region is approximately equal to the inter-electrode spacing of 2 mm).

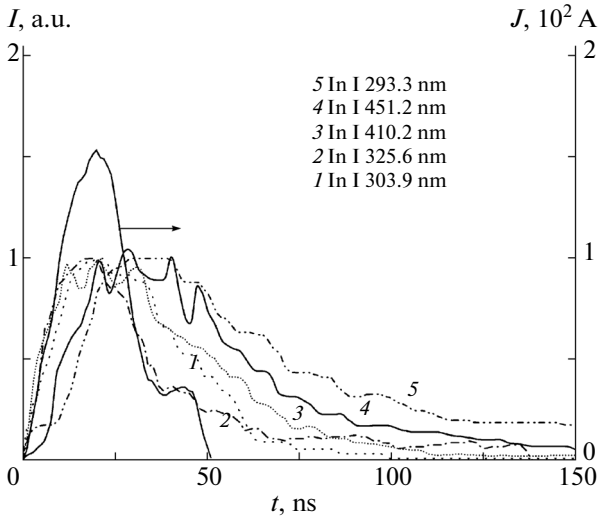

Fig. 2. Oscillograms of the nanosecond-discharge current and emission associated with transitions in the indium atom in a nanosecond discharge with chalcopyrite $CuInSe_2$ electrodes.

a group of high-intensity spectral lines of copper atoms and ions, which is characteristic of a nanosecond discharge with copper electrodes in atmospheric air and is of interest for application in a miniature lamp with short-duration UV emission [12, 13].

A photograph of the discharge (Fig. 1) shows its bulk nature. This kind of discharge in a "tip–tip" system of electrodes under atmospheric air pressure is possible without any separate pre-ionization system only in the case of pre-ionization by a short pulse of high-energy runaway electrons and the accompanying X-ray radiation [14–16]. The homogeneity of the discharge under study exceeds that of discharges with runaway electrons in a system of metallic electrodes [16, 17], because these latter also show a bright near-cathode glow.

It follows from the discharge-current oscillograms and highest intensity spectral lines of the indium atom that, during the first 50 ns, the lines are correlated with the current pulse (Fig. 2). However, a high-intensity afterglow also occurs, with its duration exceeding 150 ns for some of the spectral lines. The longest duration was observed for the emission associated with spectral transitions in the indium atom with the lowest upper-level energy, which may be due to the recombination mechanism of their population at the trailing edge of the current pulse.

Because the time in which equilibrium distribution is attained in atmospheric-pressure pulse discharges does not exceed 1 ps [17], which is substantially shorter than the current pulse duration, it is possible to determine in this case the electron temperature by the formula [1, 19]:

$$kT_e = \frac{E_2 - E_1}{\ln\left(\frac{I_1 A_2 g_2 \lambda_1}{I_2 A_1 g_1 \lambda_2}\right)}, \quad (1)$$

where k is Boltzmann's constant, E_1 and E_2 are the atomic-level energies, I_1 and I_2 and λ_1 and λ_2 are the emission intensities of the spectral lines and their emission intensities, and A_1 and A_2 and g_1 and g_2 are the spontaneous transition probabilities and the statistical weights of the energy levels.

The intensity oscillograms of the spectral lines were used to calculate the time dependences of the occupancies of the excited states of indium atoms (with a common upper energy level $6s^2S_{1/2}$, $E_{ex} = 3.022$ eV (spectral lines at 410.2 and 451.1 nm) and $7s^2S_{1/2}$, $E_{ex} = 4.50$ eV (293.3 nm) (Fig. 3), where E_{ex} is the energy of the upper energy level). The time dependence of the electron temperature was determined by an energy-distribution analysis for the occupancies of excited states of indium atoms at certain instants of time.

The maximum electron temperature of 1.6 eV was reached at the leading edge of the current pulse. The time dependence of the electron temperature and the maximum T_e in the discharge under study are qualitatively correlated with the corresponding dependence $T_e(t)$ for a discharge with a beam of runaway electrons in atmospheric-pressure nitrogen, which was determined from the band intensity ratio of the nitrogen molecule (394.3 nm) and the molecular nitrogen ion (391.4 nm) [20].

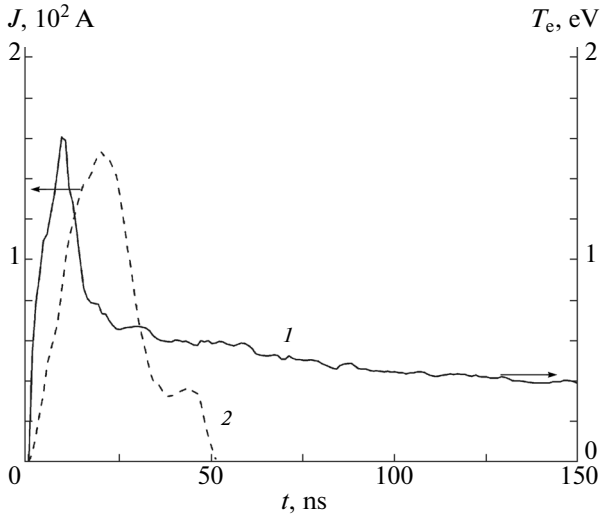

Fig. 3. Electron temperature in plasma determined from the emission oscillograms of chalcopyrite erosion products vs. time and the current oscillogram of the nanosecond discharge at $d = 2$ mm.

Thus, it was demonstrated that a point-source UV lamp with composite chalcopyrite electrodes can be developed. Its emission is based on a group of spectral lines of the copper atom in the spectral range of 200–230 nm; the maximum electron temperature of 1.6 eV is reached at the leading edge of the current pulse and decreases in the afterglow (at $t > 50$ ns) to 0.4–0.6 eV.

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