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# COPPER DIMERS IN THE VAPOR FLOW IN ELECTRON-BEAM VAPORIZATION

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Electron-beam heating is widely used in the industry. In particular, electron-beam vaporization is used for depositing coatings and thin films in a vacuum [1]. This heating method has been used lately for producing ultrafine powders, which are formed when the vapor is mixed with a gas [2]. The characteristics of the coatings and powders are determined to a considerable extent by the physical processes occurring in the vapor flow. The formation of gaseous-phase fluxes in electron-beam vaporization has not been investigated to a sufficient extent.

The contactless methods are the preferred ones among the known diagnostics methods. One of them is the electron-beam method, which is widely used in experimental investigations of rarefied gas dynamics [3, 4].

In our experiments, the electron beam was used both as a means of vaporization and as a probe. We have investigated the luminescence spectra of copper vapor, excited by an electron beam, with the aim of devising diagnostics methods for both the vapor and the condensed phases. The presence of the copper dimer in vapor flow was the subject of a special investigation.

The $Cu_2$ spectrum was first detected in the luminescence of copper vapor under thermal excitation in a King furnace [5]. Two systems of molecular bands corresponding to the $B^1\Sigma_u^+ - X^1\Sigma_g^+$, and $A^1\Pi_u - X^1\Sigma_g^+$ electron transitions were identified. Subsequently, they were registered during both radiation and absorption in a King furnace [6], in a supersonic vapor jet [7], in gaseous-phase production of clusters [8], and in pulsed laser vaporization of copper into a pulsed He jet or a steady-state jet of cold He [9-11]. Besides these band systems, the C—X system [9, 10] and many others [12] have been recorded.

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As it interacts with the metal surface, a sufficiently powerful electron beam causes intensive metal vaporization. As a result, a peculiar vapor jet is formed, where the zone of interaction between the electron beam and the surface plays the role of the nozzle.

The source of the copper-vapor jet which we used has been described in [13]. The experimental arrangement is shown in Fig. 1. The device described in [14] was used in experiments. The device includes an electron gun with a thermionic cathode. The electron beam parameters are the following: accelerating voltage, 5-25 kV; current, up to 70 mA; power, up to 1.5 kW. The gun is provided with a magnetic lens for electron beam focusing. The focused beam diameter is equal to $\approx$ 3 mm. The differential pumping system for gun evacuation makes it possible to raise the pressure in the operating chamber to 10 Pa while maintaining a pressure of $10^{-3}$ Pa in the cathode region. The source of the copper-vapor jet consists of a graphite crucible 1, which is surrounded by screens, and a wire feed system. The feed system is necessary for periodically replenishing the metal supply in the crucible. The following quantities are monitored during the operation of the source: the accelerating voltage U, the crucible current $I_{cru}$, the magnetic lens focusing current If, and the pressure $p_H$ in the operating chamber.

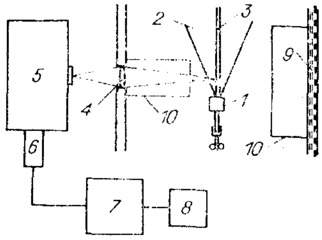


Fig. 1

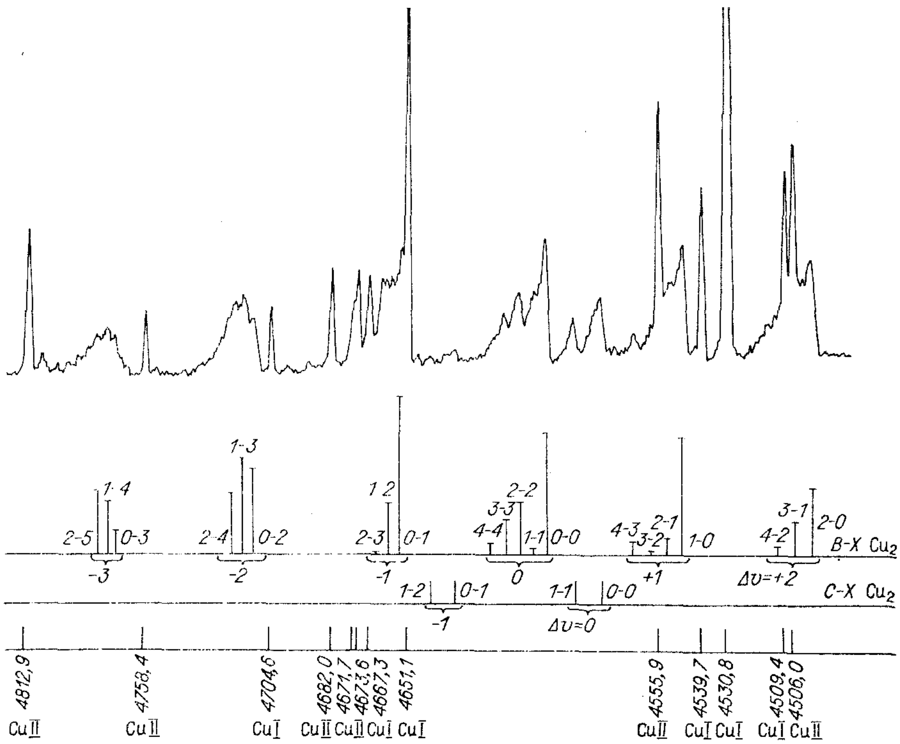


Fig. 2

The jet of copper vapor 2 is observed visually through a window 9 with respect to the characteristic green luminescence in the zone of the electron beam 3. The radiation in the axial region of the jet is collected by means of lens 4 and directed to the entrance slit of an SPM-2 monochromator 5. An FEU-39A photomultiplier 6 is provided at the exit slit. After passage through an IMT-05 amplifier 7, the electric signal from the photomultiplier is recorded by a KSP-4 self-recorder 8. Deposition of metal dust on the lens and the window is prevented by pressure-feeding a gas (usually $N_2$) into the chambers 10. The spectra are recorded under the following conditions: $I_{cru}$ = 10-30 mA; U = 14 kV; $I_f$ = 95 mA; the distance between the melt surface and the measuring point is equal to 6 mm; an atmosphere of $N_2$ and He at the pressure $p_H$ = 10 Pa is used in the chamber. Moreover, measurements were also performed in an atmosphere of $N_2$ in a higher vacuum, $p_H = 10^{-2}$ Pa.

Depending on the intensity, up to 48 lines were recorded in the wavelength range from 350 to 600 nm. These were identified according to tables [15] as the lines of copper atoms and ions. Gas luminescence is recorded as the vapor expands in a vacuum chamber filled with $N_2$ or He in the jet region under investigation. This indicates that the background gas penetrates the jet core, which is especially characteristic for the scattering operating conditions [16]. Luminescence of the possible products of chemical interaction between the metal vapor and the background gas was not observed in the spectra.

A band system whose appearance did not depend on the pressure and composition of the atmosphere was recorded for a certain electron beam power in the $\Delta\lambda$ = 450-481.2-nm range of the luminescence spectrum of the copper-vapor jet. Figure 2 shows such a spectrum, obtained during the jet outflow into an atmosphere of $N_2$ at $p_H = 10^{-2}$ Pa and $I_{cru}$ = 22 mA. The positions of the edges of bands the B–X $Cu_2$ system, based on the data from [5], and of certain bands of the C–X $Cu_2$ system, based on the data from [9], as well as the wavelengths of the lines of copper atoms and ions [15] are given below the spectrum. The good agreement between the edges of the recorded bands and the results obtained in [5, 9] enables us to ascribe the spectrum obtained to $Cu_2$. In particular, we identified the Deslandres oblique band series with $\Delta\nu$ = + 2, +1, 0, −1, −2, −3 of the B–X system and $\Delta\nu$ = 0, −1 of the C–X $Cu_2$ system.

The intensity of $Cu_2$ bands depends heavily on the beam current; as $I_{cru}$ varies from 13 to 21 mA in an atmosphere of $N_2$ at $p_H = 10^{-2}$ Pa, the intensity increases by a factor of 20. Changes in pressure in the operating chamber and in the atmosphere composition affect noticeably the dimer signal. For the same power supplied, the bands are most intensive at $p_H = 10^{-2}$ Pa; at $p_H$ = 10 Pa in a He atmosphere, the signal is noticeably weaker, while it is weaker still at $p_H$ = 10 Pa in $N_2$. Such signal behavior does not agree with the expected one. The cooling of the metal vapor after it is mixed with a gas should intensify the condensation process; this procedure is fairly often used for producing clusters [17]. Thus, the dimer signal should become stronger with an increase in pressure within the operating chamber. However, it has been found that the vaporization rate depends heavily on the pressure above the evaporation surface. This phenomenon is explained in [1] by the return of copper atoms to the surface as a result of collisions with background gas particles. The rate of copper vaporization by a constant-power electron beam drops considerably even as the pressure increases to 1 Pa [1].

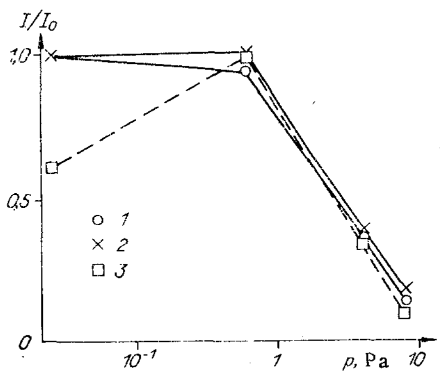


Fig. 3

In order to verify this, we plotted the intensities of the lines CuI $\lambda$ = 448.04 nm and CuI $\lambda$ = 453.08 nm, as well as CuII $\lambda$ = 455.59 nm, as functions of the $N_2$ pressure in the operating chamber for $I_{cru}$ = 12 mA (Fig. 3, point 1-3, respectively). There is evidently good agreement with the data given in [1].

In order to estimate the temperature at the measuring point, we compared the experimental and theoretical distributions of band intensity for the B–X $Cu_2$ system. The calculations were performed by using the equation

$$\frac{I_{v'v''}}{I_{00}} = \left(\frac{\nu_{v'v''}}{\nu_{00}}\right)^4 \frac{q_{v'v''}}{q_{00}} \frac{\sum\limits_{v''} q_{v'v''} \exp\left[-\frac{G(v'')hc}{kT}\right] \sum\limits_{v''} \nu^3_{0v''} q_{0v''}}{\sum\limits_{v''} q_{0v''} \exp\left[-\frac{G(v'')hc}{kT}\right] \sum\limits_{v''} \nu^3_{v'v''} q_{v'v''}}. \quad (1)$$

Here, $I_{v'v''}$ is the intensity of the (v', v") band of the B–X $Cu_2$; system $q_{v'v''}$ is the corresponding Franck–Condon factor, $\nu_{v'v''}$ is the transition frequency, G(v") is the value of the term with the vibrational number v" in the $X^1\Sigma_u^+$ state, $G(v'') = \omega_e(v'' + 1/2) - \omega_e x_e(v''+1/2)^2$: and T is the vibrational temperature in the $X^1\Sigma_u^+$ state. The values of the Franck–Condon factor were borrowed from [18], while the values of $\omega_e$ and $\omega_e x_e$ were borrowed from [5]. This expression holds under the following assumptions: excitation of the $B^1\Sigma_g^+$ state occurs as a result of direct electron impact, the Franck–Condon principle holds for the excitation, and the population distribution with respect to vibrational levels in the $X^1\Sigma_u^+$ state is a Boltzmann distribution with the temperature T. The calculation results based on (1) for T = 2300 K are given below the spectrum in Fig. 2. The heights of the lines marking the positions of the band edges of the B–X system correspond to the $I_{v'v''}/I_{00}$ ratio. It is evident from Fig. 2 that the theoretical intensity distribution is close to the measured distribution. Band overlapping by lines of copper atoms and ions, changes in the photomultiplier spectral sensitivity, and the reduction in band intensity due to a lowering of the metal level in the crucible were neglected in scanning the spectrum. The estimated temperature is roughly equal to the surface temperature in the zone of interaction between the electron beam and the metal, which has been measured by means of a pyrometer in [13] for similar beam parameters. The vapor density at the measuring point was estimated by means of the expression for isentropic expansion of a monatomic gas,

$$\frac{n}{n_0} = \frac{B}{(x/d_*)^2}, \quad (2)$$

where x = 6 mm, $d_*$ = 3 mm, and B = 0.16. The value of $n_0$ was estimated by means of the method described in [1]. We can find the vapor pressure if we know the approximate dimensions of the meniscus in the liquid metal. In our case, the meniscus diameter was ≈3 mm, while its depth was ≈ 2 mm, which yielded $p_0 \approx 1000$ and $n_0 \approx 3 \cdot 10^{16}$ cm$^{-3}$. Then, expression (2) yields $n \approx 10^{15}$ cm$^{-3}$.

Thus, we have investigated the spectrum of luminescence excited by electron impact in a copper-vapor jet produced by electron-beam vaporization. The copper dimer emission was observed. Two band systems, B–X and C–X $Cu_2$, were recorded. Comparison with the spectrum calculated under the assumption of validity of the Franck–Condon principle has shown satisfactory agreement, which makes it possbile to use electron-beam diagnostics for measuring the density and the vibrational and rotational temperatures of dimers.

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