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Time-Resolved Optical Spectroscopy of Lead at Near Critical-Point States[1]

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Experiments were performed to investigate the behavior of lead near its critical point. Emission spectra of shocked lead samples during unloading into helium at different initial pressures were measured by an optical multichannel analyzer (OMA), as well as by a fast optical pyrometer. To describe the obtained experimental data, a model of a thin mixture layer was suggested, in which helium emits due to the presence of external electrons from lead.

KEY WORDS: critical state; electron concentration; lead; shock compression; spectroscopy; thermal conductivity.

1. INTRODUCTION

Usage of the method of fast multichannel optical pyrometry to study isentropic expansion of shocked samples allows one to determine pressure of entrance of isentrope into the two-phase region [1], to trace the boiling curve, and to evaluate parameters of the critical point [2]. At the same time, the presence of the metal–dielectric transition in this region of phase diagram leads to an increase in emissivity of lead from values of 0.3 to 0.4, usual for liquid metals, up to 1 which is usual for gases. Heat transfer on the lead–gas boundary also can result in spectral peculiarities and in different values of temperature in volume and on the boundary. Previous

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pyrometry experiments gave a brightness temperature for the 585.5-nm channel different from the other channels (650, 810, 880, and 975 nm). Current spectral measurements were done to investigate spectral peculiarities with the aim to improve the pyrometer measurement reliability.

2. MEASUREMENTS

In this work, optical emission from the free surface of shocked and expanded lead samples was measured by means of optical spectroscopy, in the spectral range from 460 to 650 nm, with a time resolution of 20 to 40 ns, and simultaneously by pyrometry at 585.5, 650, 810, 880, and 975 nm, with a time resolution of 3 to 10 ns. The experimental setup was analogous to those in Refs. 1 and 3. Lead samples, shocked up to a pressure of 265 GPa in the first series of experiments and up to 223 GPa in the second, were unloaded into helium at different initial pressures (Fig. 1). Pyrometric measurements with high time resolution were used to determine the temperature of lead in the final state of expansion without heat transfer, by the intensity of emission at the moment after the sharp rise in the beginning of expansion. Quartz fibers were used to transfer emission from the experimental setup to the detection system. The velocity of the shock wave in helium was determined by an optical baseline method permitting the determination of the final pressure of the lead expansion by a known helium equation of state. A xenon flash lamp was used to determine the spectral sensitivity of a spectrometer detector assembly. The

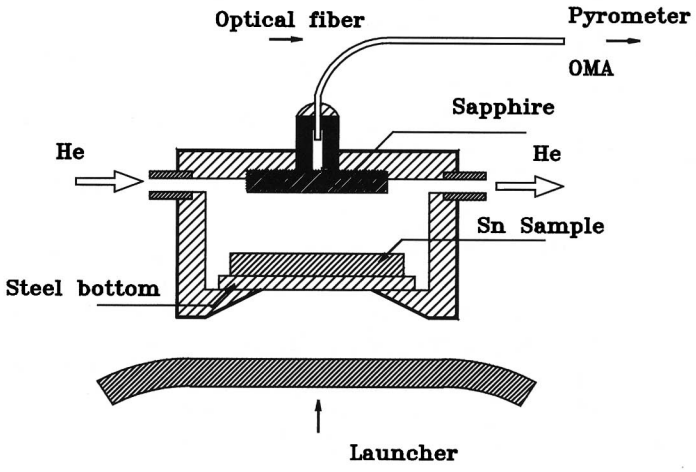

Fig. 1. Experimental setup.

brightness temperature of the flash lamp was determined by a pyrometer. The pyrometer was calibrated using a tungsten ribbon lamp as a standard source of light.

3. DISCUSSION

The states of lead generated on the expansion isentrope in the first series of experiments correspond to the nonideal plasma state and to the two-phase region state generated after condensation [2]. In the second series of experiments; two-phase region states are generated due to an evaporation of the liquid metal on a free surface [3]. Entropy of lead in the first series of experiments is more than critical point entropy, and in the second the entropy of lead is less then the entropy of critical point, so there is a gas lead–helium boundary in the first series of experiments and a liquid

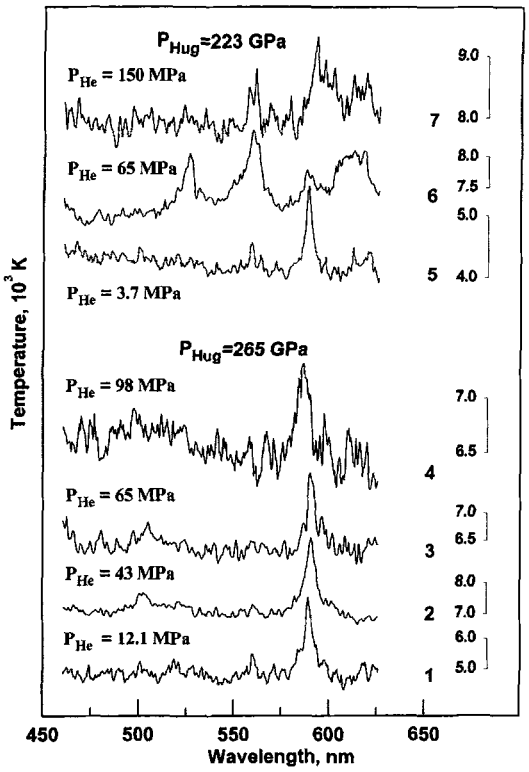

Fig. 2. Spectral intensity of emission 200 ns after exiting of the shock wave into helium.

lead–helium boundary in the second. The emission spectra, registered 200 ns after the exit of the shock wave from the free surface of lead, are shown in Fig. 2. The fact that lead–helium boundary conditions are different leads to the difference of the spectrum registered for the same final pressure (65 MPa). Lead has a lot of emission lines of similar intensity in the investigated spectral range according to spectral data. The line with the wavelength of ~ 590 nm can be clearly seen up to the maximum investigated pressures in the first series of experiments and at low pressure in the experiments of the second series. This line was not registered in the analogous experiments with lead unloading into hydrogen, but when tin was unloading into helium, the same line was observed. So, in further analysis it was assumed that this line near 590 nm corresponds to the helium line of 587.6 nm.

The time dependence of this line width, its center position, and its integral intensity (product of width and intensity of line – parameter proportional to the full probability of line emission) was analyzed. For this analysis the line shape was approximated by a Lorentz profile. The offset of position of the center of the line was about 0.5 nm. The line width was determined with an accuracy of $\sim 50\%$.

The time and spectral dependence of the brightness temperature are shown in Fig. 3. The integral intensity of this line remains constant for 1 μs, the time needed for a shock wave to travel through the helium base, with an accuracy of about 10% from the initial value.

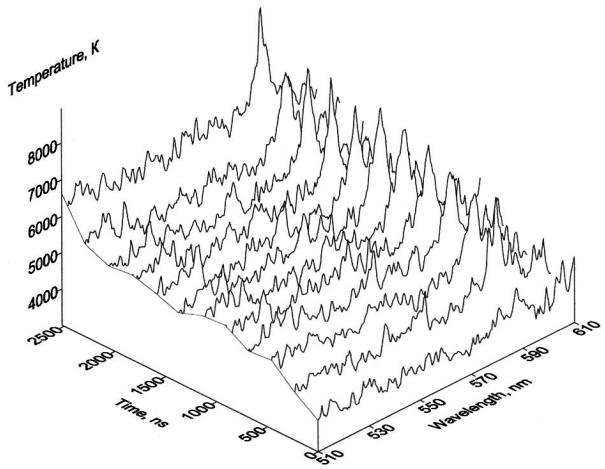

Fig. 3. Dependence of brightness temperature versus wavelength and time.

The increase in the final pressure leads to the increase in the line width and the background emission noise. For the highest pressure achieved in the first series of experiments (98 MPa), the line near 590 nm cannot be described by one Lorentz line.

The time dependence of the line position is shown in Fig. 4. It can be seen that the position of the line and its width remain constant in the initial period of time. After 400 to 800 ns, the position of the line rapidly changes during the period of time comparable with apparatus resolution.

The shift of the helium line from its free atom value was described by the model of electron impact broadening [4]: $\Delta L = \alpha N_e$, where ΔL is the shift of the line position from its value for a free atom, N_e is the electron concentration, and α is the proportionality coefficient. For the helium line at 587.6 nm the theoretical value for α is $\alpha = -0.8 \times 10^{-18}$ nm $\cdot$ cm^3. In Ref. 5, as well as in our work, an opposite sign of this constant was experimentally observed. To estimate the electron concentration, the theoretical value of α was used, but with the opposite sign. The experimentally measured width of line and its position gave $N_e \sim 10^{18}$ cm^{-3}, changing from 10^{18} up to 5×10^{18} cm^{-3} with an increase in the final pressure of expansion. Experimental values of N_e from this model are presented in Fig. 4 (right axis). The experimental results were compared with theoretical calculations of the properties of helium and lead performed according to the chemical model of plasma [6]. The velocity of the shock wave in

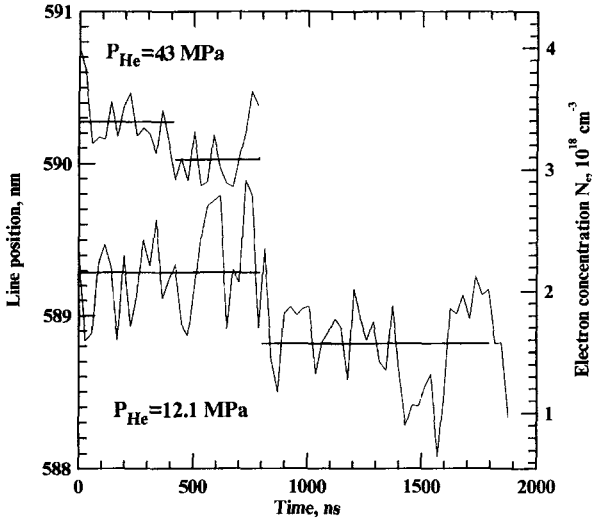

Fig. 4. Position of He (587.6 nm) line center (left axis) and electron concentration (right axis) versus time.

helium with the given initial temperature and pressure was taken as initial data for the calculations. Parameters of shocked lead were determined in a separate series of experiments. The shock wave compresses helium up to the state of nonideal plasma, with a mean temperature $\sim 10,000$ K and concentration of the electrons $N_e < 10^{16}\,\mathrm{cm}^{-3}$. The calculations for lead resulted in a value of $N_e \sim 10^{18}\,\mathrm{cm}^{-3}$.

To explain the existence of line on the wavelength 590 nm, it was suggested that helium emits in a very thin mixture layer, due to the presence of external electrons. These electrons are penetrating into helium from lead. As additional confirmation of this model, the fact may be used that the integral line intensity is constant in time. Under the assumption that helium radiates out of the volume, the integral intensity must increase in time due to the increase in shocked helium layer width. The fact that the mean position of the helium line changes from one value to another may be explained by arrival of the reflected expansion wave from the inner boundary of lead. The thickness of the radiating layer can be estimated from the duration of this transition. In the present work this time is limited by the time resolution of the apparatus (20 ns). The sound velocity of the shocked helium is about $\sim 5\,\mathrm{km}\cdot\mathrm{s}^{-1}$, so the thickness of the radiating layer is less then 0.1 mm, the result being diminished by the order of magnitude, if lead sound velocity is used for estimation. The time between the exit of the shock wave on the free surface and the moment of the break of line mean position can be used to determine the sound velocity in expanded metal.

The temperature of the radiating layer was evaluated by numerical simulation of heat transfer between two layers (in our case for helium and lead) with given initial temperatures in volume [7, 8]. The volume temperature of lead was obtained from pyrometry measurements and the volume temperature of helium was calculated, using the chemical plasma model [6]. Heat transfer coefficients were calculated in the first approximation of Chappman–Enskog for the monatomic gas [9]. The electron component of heat transfer was calculated by the equation $\chi_e = \chi_a (N_e/N_a)\sqrt{(M/m)}$, where N_e and N_a are the concentrations of the electrons and atoms, respectively and M and m are the masses of atom and electron. The values of N_e, C_v, and χ_a necessary for the modeling were obtained from the chemical plasma model. It was assumed that electrons and atoms have equal mean free paths [9]. Calculations of temperature on the metal–helium interface were made using the standard conductive heat transfer model [10], taking into account the temperature dependence of the thermal conductivity coefficient.

Results obtained from theoretical model and experimental data for four pressures are presented in Fig. 5. Curve 1 is the brightness temperature

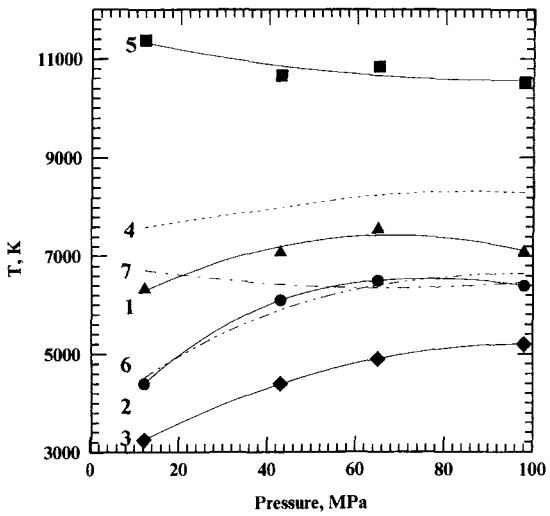

Fig. 5. Experimental and theoretical temperatures of the helium–lead boundary for different final pressures of expansion. Curve 1, the experimental brightness temperature of helium on the center of line 587.6 nm (filled triangles, experimental points; smooth line, polynomial approximation). Curve 2, the mean spectral temperature of the radiating layer (without radiation in lines) (filled circles, experimental points). Curve 3, the temperature of lead obtained by pyrometric measurements (filled squares, experimental points). Curve 4, the temperature on the helium–lead boundary, calculated from the model of heat transfer. Curve 5, the temperature of shocked helium (filled squares, experimental points). Curve 6, the temperature on the helium–lead boundary, calculated from the model of turbulent mixing layers. Curve 7, the lead temperature calculated for the given pressure and concentration of electrons according to the chemical plasma model [6].

of helium on the center of the line at 587.6 nm; curve 2, the mean spectral temperature of the radiating layer (without radiation in lines); curve 3, the temperature of lead obtained by pyrometric measurements; curve 4, the temperature on the helium–lead boundary, calculated from the model of heat transfer; and curve 5, the temperature of shocked helium. One can see that the calculated temperature on the helium–lead boundary obtained from the model of heat transfer does not correspond to the experimental data.

For better agreement with the experimental data, another model was proposed, in which it was assumed that helium and lead are mixed together

due to the hydrodynamic instability. As a first approximation it may be assumed that, in this region, equal volumes of lead and helium are mixing, and then their temperatures adiabatically equalize. This model results are presented in Fig. 5 (curve 6) and, within the experimental error of 300 K, agree with experimental points of curve 2. The lead electron concentration in the mixture layer was calculated according to Griem's dependence of line position as a function of electron concentration. Then the lead temperature was calculated for the given pressure and concentration of electrons according to the chemical plasma model [6] (curve 7, Fig. 5). The resulting temperatures agree with experimental data in the limits of experimental error (300 K) for mean spectral temperature at high pressures. It is not so at low pressures. It may be supposed that in this case hydrodynamic instabilities at the boundary layer increase. An increase in surface instability can result in lead entering into deeper helium layers, with higher temperatures, so that lead will be ionized at higher initial temperatures.

4. RESULTS

A model of heat transfer was proposed for the boundary between gas and near-critical point state metal. This model allows a calculation of the volume temperature of expanded metal from the known resultant temperature on the gas–metal boundary. This permits an investigation of the thermodynamic properties in the two-phase region by pyrometers with less time resolution. Also, it was shown that pyrometric measurements have to be performed in the spectral intervals away from the lines of the investigated materials. Spectroscopic measurements also provide more information on the sound velocities in the dense nonideal plasma state, and its composition.

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