Properties of liquid argon at shock compression in pressure range of 125-530 GPa

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Summary. With use of semispherical generators of shock waves (SWG) for the basic Hugoniot adiabat, we measured density of shock-compressed liquid argon of \(\approx 5\text{g/cm}^3\), temperature of \(\approx 31300\text{K}\), and coefficients of light absorption up to \(\approx 180\text{cm}^{-1}\) at pressures up to \(\approx 230\text{GPa}\). Densities of compressed argon of \(\approx 5.8\) and \(7.1\text{g/cm}^3\) were recorded at pressures of \(\approx 300\) and \(530\text{ GPa}\), respectively, during reflection of shock wave from sapphire window. The obtained data were analyzed basing on various theoretical models.

Introduction

Study of thermodynamic and optical properties of shock-compressed condensed noble gases is of interest, because they have symmetric electronic structure in atom and dense packing in crystalline state. So, these substances are convenient for theoretical description of their properties followed by comparison with test.

In this work, comparing to the previous works, we extended the area of study of liquid argon properties at shock compression with use of experimental constructions consisting of cryogenic cell and semi-spherical generators of shock waves MZ-4 and MZ-8 [1]. We obtained experimental data on density, temperature and light absorption in earlier-unstudied range of pressures reaching 230 GPa at the basic Hugoniot adiabat and reaching 530 GPa in shock wave reflected from sapphire window.

1. Measurement of dynamic characteristics

For shock-wave tests with liquid argon at pressures higher 100 GPa, experimental devices were developed. They consist of semi-spherical generators of shock waves of the type MZ-4 and MZ-8 [1] and cryogenic cuvettes. Similar design with generator MZ-4 was earlier used when studying properties of shock-compressed liquid xenon [16].

The experimental device is schematically presented in fig. 1. At plate (1), from one side, unit of high explosive (HE) (2) and steel semi-spherical impactor (3) are symmetrically fastened, and from the other side – cylindrical casing (4), which contains flange (5) fastened with semi-spherical screen (6) and holder (7) with gauges intended to measure velocities of shock waves in the screen and liquid argon (fig.1(a)). The internal space between the shell and the semispherical screen of the experimental device was pumped out up to residual pressure of air of \(\sim 10^{-2}\text{torr}\) or lower. After HE initiation by explosion products, impactor (3) was accelerated through air gap (8). During impact against screen (6) made of aluminum AD-1, the impactor forms a shock wave in it, which later enters liquid argon (9).
Fig. 1. Semispherical experimental device to study properties of shock-compressed liquid argon

The measuring unit, which consists of screen (6) and holder (7) with gauges, is presented in increased scale in fig. 1(b). Sapphire window (1) is glued in the central hole of holder (7) with light diameter $\varnothing 5$ mm. Radiation of front of shock wave was recorded by optical pyrometer through that sapphire window. Velocity of shock wave in liquid argon was measured by the electrocontact and optical basis methods. In the first case, to measure time of shock wave motion, we used from 6 to 12 couples of contacts located at the known basis. In the second case, data were processed using oscillograms of radiation of shock wave front.

Liquid argon was produced in a separate liquefier and poured into a cryostat directly before the experiment. Air around the cell with liquid nitrogen was pumped out up to residual pressure of $\sim 10^{-2}$ torr or lower. The initial temperature of liquid argon $T_0 = 87$ K was controlled by calibrated platinum thermometer. Density of liquid argon $\rho_0 = 1.4$ g/cm$^3$ corresponds to the measured temperature $T_0$.

In accordance with the reflection method for determination of pressures and particle velocities in direct and reflected wave basing on the measured value of shock wave velocity in argon, it is necessary to solve the problem on decay of random breaks, which are formed at shock wave arrival to the aluminum-argon and argon-sapphire interfaces. It is simple to solve this problem in experiments with shock wave generators of planar geometry. It requires to measure velocities of shock waves in studied substance, screen and sapphire, and to know the equation of state for the screen and the sapphire. Peculiarity of experiments with spherical generators is growth of shock wave velocity in studied sample and elements of experimental device during wave motion towards the center. As an example, fig.2 presents results of the experiment with shock wave generator MZ-4 together with calculation data. Numerical simulation of the experiments was performed by the one-dimensional gasdynamic code of VNIIEF. In the calculations, the equations of state of materials of the experimental device in Mie-Gruneisen form were used. Cooling down of elements of the experimental device to temperature of liquid argon $T_0 = 87$ K was taken into account.
Fig. 2. Dependence of shock wave velocity on radius in semispherical experimental device with generator MZ-4

Parameters of the shock adiabat for aluminum were chosen basing on data from [2]; the $D-U$ relation from [3] as $D = 8.74 + 0.957U$ (1) was used for sapphire.

The values of average velocities of shock waves have meaning only for radii $R_{\text{meas}}$, which are located in the middle of measurement bases in corresponding elements of the construction (see fig. 2). Due to this reason, for determination of thermodynamic parameters of shock-compressed substance, average values of velocities of shock waves, which were measured in elements of the experimental device, should be corrected at transition to a single boundary of decay of random break. Parameters of shock-compressed liquid argon were determined using the conservation laws at the break decay boundary, which was formally transmitted to the radius of measurement of shock wave velocity in argon $R_{\text{meas. Ar}}$ (see fig. 2). For this purpose, alongside with measurement of shock wave velocity in argon, we measured velocity of shock wave in the screen of the experimental device. This value was further corrected by the calculation method regarding to the value at radius $R_{\text{meas. Ar}}$. This variant of data processing seems to be reasonable, since the results for liquid argon are obtained in the earlier-unstudied area of parameters, and the state in the screen (aluminum AD-1) in this area of pressures is well-known. Table 1 presents the measured values of velocities of shock waves in the screens and the parameters of shock waves in aluminum corrected regarding to radius $R_{\text{meas. Ar}}$, which are used for determination of parameters of shock waves in liquid argon by the reflection method.

Table 1 – Parameters at basic shock wave

<table>
<thead>
<tr>
<th>SWG</th>
<th>Measured velocity of shock wave in screen</th>
<th>Corrected parameters of shock wave in screen at radius $R_{\text{meas. Ar}}$</th>
<th>Parameters of shock wave in liquid argon</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$R_{\text{meas. Ar}}$, mm</td>
<td>$D$, km/s</td>
<td>$D$, km/s</td>
</tr>
<tr>
<td>MZ-4</td>
<td>32.75</td>
<td>12.91 ±0.26</td>
<td>13.65</td>
</tr>
<tr>
<td>MZ-8</td>
<td>21.65</td>
<td>14.90 ±0.44</td>
<td>16.58</td>
</tr>
</tbody>
</table>
The $D-U$ parameters, which were obtained by formal transmitting of the screen boundary to the radius of measurement of average velocity of shock wave in liquid argon, are presented in fig. 4 together with data [4-7]. All results are approximated by two dependences:

$$D = 0.971 + 1.922U - 0.079U^2 \quad U \leq 3.84 \text{ km/s}$$

and

$$D = 2.82 + 1.137U \quad \text{at} \quad U > 3.84 \text{ km/s.}$$

During reflection of the shock wave, which passes in argon, from the sapphire window having initial density $\rho_0 = 4.02 \text{ g/cm}^3$, a reflected shock wave is formed, which propagates back to the argon area (Ar1). Pressure in the reflected wave $P_2$ is equal to pressure $P_{\text{Sap}}$ in sapphire, and it can be determined with use of the measured value of shock wave velocity and the known adiabat of sapphire.

**Table 2 – Parameters of directed and reflected shock waves in liquid argon**

<table>
<thead>
<tr>
<th>SWG</th>
<th>Parameters of shock waves in sapphire</th>
<th>Initial parameters of shock waves in liquid argon</th>
<th>Parameters of reflected shock waves in liquid argon</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$R_{\text{meas}}$, mm</td>
<td>$D_1$, km/s</td>
<td>$D_{\text{corr}}$, km/s</td>
</tr>
<tr>
<td>MZ-4</td>
<td>25.6</td>
<td>14.09</td>
<td>13.86</td>
</tr>
<tr>
<td>MZ-8</td>
<td>14.34</td>
<td>16.89</td>
<td>16.43</td>
</tr>
</tbody>
</table>

2. **Measurement of optical characteristics**

For measurement of temperatures, a high-speed pyrometer of visible range of spectrum was used. Radiation of shock wave front in liquid argon was recorded by photomultipliers (FEU) at wavelengths $\lambda = 600, 550, 498, 450, \text{ and } 406 \text{ nm}$, which were picked out by interferential light filters with filtration of $\sim 50\%$ and the band $\Delta \lambda \approx 10 \text{ nm}$ at the level of half of the maximum
value. To weaken radiation, neutral light filters were used. Their filtration was experimentally measured by spectrophotometer DR-4000U. The problem of determination of the temperature and radiating capacity using spectral fluxes measured in the experiment was solved by the non-linear least-squares method for the model with two parameters: $T$ and $\varepsilon$. The thermal radiation flux for the measured spectral temperatures was calculated by the Planck's formula:

$$N(\lambda, T) = \varepsilon \cdot C_1 \lambda^{-5} \left[ \exp\left(\frac{C_2}{\lambda T}\right) - 1 \right]^{-1} = C_1 \lambda^{-5} \left[ \exp\left(\frac{C_2}{\lambda T_b}\right) - 1 \right]^{-1}$$

Here, $\varepsilon$ is the radiating capacity of the body, $\lambda$ is the wavelength, $T$ is the actual temperature, $T_b$ is the brightness temperature, and the constants are $C_1 = 1.19 \cdot 10^{-16}$ Wm$^{-2}$/sr and $C_2 = 0.0144$ mK. Table 3 presents values of spectral temperatures measured in two experiments with liquid argon with use of generators MZ-4 and MZ-8. This table gives also values of actual temperatures $T$ and values of radiating capacity $\varepsilon$, which are obtained by approximations of experimental values by the least-squares method.

Table 3 – Temperature of shock-compressed liquid argon

<table>
<thead>
<tr>
<th>$\lambda$, nm</th>
<th>MZ-4</th>
<th>MZ-8</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_s$, K</td>
<td>$T$, K</td>
<td>$\varepsilon$</td>
</tr>
<tr>
<td>406</td>
<td>18125</td>
<td>24580±1700</td>
</tr>
<tr>
<td>450</td>
<td>-</td>
<td>17815</td>
</tr>
<tr>
<td>498</td>
<td>17626</td>
<td>550</td>
</tr>
<tr>
<td>600</td>
<td>16433</td>
<td>600</td>
</tr>
</tbody>
</table>

$P^*$ - values of pressures related to boundaries of screens

During experimental measurement of the coefficient of light absorption, the basic attention was paid to the initial area of radiation intensity growth, which had duration of $\sim$ 100 ns. As in [8], the rise of the shock-front glow was associated with the increase in the thickness of the shock-compressed layer, which had (averaged) coefficient of light absorption $\alpha$. Neglecting the reflection and using the Bouguer-Lambert-Beer formula for transmittance,

$$\tau = \exp(-\alpha \cdot l),$$

where $\alpha$ is the coefficient of light absorption of a layer having thickness $l$, one can write the rise of radiation intensity in the normal direction as:

$$N = N_0 [1 - \exp(-\alpha \cdot l)] = N_0 [1 - \exp(-\alpha (D - U) \cdot t)]$$

where $N_0$ is the radiation intensity of an optically dense layer, $l = (D - U) \cdot t$ is the thickness of a shock-compressed substance, $t$ is the shock-wave passage time in the substance. It is easy to obtain the relation for evaluation of $\alpha$ from (9):

$$\alpha = -\left[1 / (D - U) \cdot t \right] \ln\left(1 - N_0 / N_0\right).$$

According to the experiments, the coefficient of light absorption is changed in the range from $\approx 3$ cm$^{-1}$ to 180 cm$^{-1}$ in liquid argon at pressures up to $\approx 250$ GPa. The approximation dependence of these data is the following:

$$\ln \alpha = (5.71 \pm 0.10) - (1.70 \pm 0.07) \cdot \left[10^4 / T\right].$$

3. **Comparison with results of theoretical calculations**

The new experimental results are presented in Fig. 5 in the pressure-density coordinates and, in the temperature-pressure coordinates, in Fig. 6, together with the available experimental data and the results of theoretical calculations.
Experiment:

- this work – at basic adiabat;
- this work at reflected adiabat;
- [5]; - [6]; ▽ - [6] – at reflected adiabat;

Calculation: 1,2,3,4 – [6]; 5,6,7- this work

Fig. 5. Shock adiabat of liquid argon

Calculation [8]: 1- without account for excitation of electrons, 2 – with account for, 3 – effective temperature in red area of spectrum (λ = 670 nm); 4 – model SAHA IV [this work]

Fig. 6. Temperature in shock-compressed liquid argon

In [6], the form of the equation of state of argon in solid and liquid phases was taken similar to the earlier-developed semi-empirical EOSs of metals. And the contribution of zero oscillations to pressure and energy was not separated. Our calculations was performed in the frames of the modified chemical model of plasma with use of the universal code SAHA-IV
In the frames of that model, argon was calculated as strongly non-ideal mixture of ions, electrons and atoms. When calculating the equilibrium plasma composition and its thermodynamic properties, the partial degeneracy of the electronic component and the interactions between all sorts of particles were taken into account. To describe the coulombic nonideality, an improved modification of the pseudopotential approach suggested in was used. With this modification, the effective electron-ion interaction was described by the short-range-corrected Coulomb potential (Glauberman-Yukhnovskiy potential). The effective depth of this potential was taken as equal to the interaction energy of an electron-ion pair at the average distance between heavy particles (ions and atoms). This corresponds to the cutoff energy adopted in this model for separating the free and bound (intra-atomic) states in the calculation of atomic partition functions. Apart from the contribution from the Coulomb interaction between charged particles, the strong repulsion of heavy particles at close distances was taken into account. This was accomplished using the approximate equation of state for "soft spheres", modified to a mixture of particles with different diameters. The degree of softness of the intermolecular repulsion potential was chosen from the requirement of the best description of the experimentally measured equation of state for condensed argon at room temperature.

As one can see in fig. 4, the semi-empirical equation of state of argon [6] does not describe results of this work at pressures higher 100 GPa. The calculations, which are performed with use of the universal code SAHA IV, give satisfactory description of data for both the main and the reflected Hugoniot adiabates up to pressures of 530 GPa.

The temperature-pressure relations, which are calculated by the models [6] and SAHA IV, are in good agreement in the range up to 100 GPa (fig. 5). Even with account for the "gray" character of shock wave front radiation, new experimental \( T(P) \) data are, probably, located below the calculated dependences. It can be associated with screening of the radiation by cold electrons.

References