ELECTRICAL CONDUCTIVITY OF SHOCK COMPRESSED CONDENSED ARGON AT PRESSURES FROM 20 to 70 GPa

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It was shown in [1] that upon compression of liquid argon to a volume $V < 13.5$ cm^3·mole^{-1} thermal excitation of electrons in the conduction zone markedly lowers the temperature T and pressure p behind a shock wave front. Agreement between calculated and experimental Hugoniot adiabats was achieved in [1] at $p > 40$ GPa with consideration of narrowing of the forbidden zone with decrease in V. Measurements of conductivity σ of liquid argon behind a shock wave front permit a better understanding of its state as well as determination of the forbidden zone width W(V, T) using the relationship $\sigma = \sigma_0 \exp(W/2kT)$ (where σ_0 is the minimum metallic conductivity), valid for liquid semiconductors with charge carriers excited in nonlocalized states [2].

The distribution of σ in liquid argon behind a plane steadystate shock wave was measured using the two-probe method described in [3]. A diagram of the experimental equipment is shown in Fig. 1, where 1 is a plane shock wave generator; 2, explosive charge; 3, aluminum of iron striker plate driven to known velocity within a vacuum [4]; 4, screen of same metal as striker plate; 5, dielectric insert (Teflon-4 for $p < 40$ GPa; Caprolon for $p = 41.6$ GPa, Al_2O_3-based ceramic for higher pressures); 6, copper current lead with inner diameter of 39 mm, wall thickness 0.5 mm, height $h = 5$ mm; 7, central electrode of 1 mm diameter copper wire; 8, steel cylinder, filled with reduced argon, 99.9% purity; 9, 20-cm connecting cable. The 0.4-mm-diameter copper wire probes are located at distances $r_1 = 5$ mm, $r_2 = 15$ mm. The electrical circuits used for the measurements are the same as those of [3]. The experimental arrangement insured the absence of argon boiling within the experimental chamber. The initial argon temperature was 87.4°K. Pressure was determined from the known state in the screens and the Hugoniot adiabats of liquid argon [5, 6] and the dielectrics [7-9]. Cooling of the screens and dielectrics to T = 87.4°K was considered. Asymmetry of the shock wave entry into the liquid volume studied did not exceed 0.04 μsec.

*Deceased.

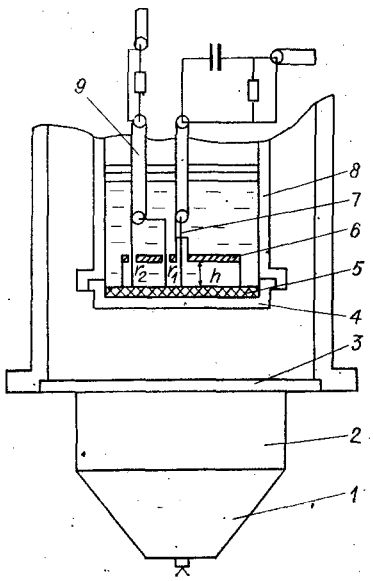

Fig. 1

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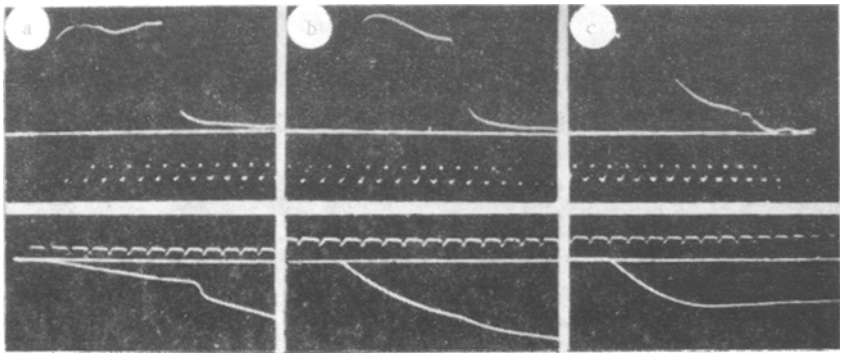

Fig. 2

Figure 2a-c shows typical oscillograms of voltage (top) and current for argon pressures of 23.2, 36.9, and 68.6 GPa, respectively. The time marker frequency is 10 MHz. Figure 3 shows the experimental results in the form of curves of σ as a function of the time the material has been in the compressed state behind the shock wave for pressures of: 1, 23.2; 2, 27.4; 3, 32.7; 4, 36.9; 5, 41.6; 6, 49.5; 7, 68.6. Each curve was obtained by averaging 4-6 experiments. The uncertainty levels were determined for a 0.9 confidence level. The measurement times were limited to the moment of arrival at the argon–dielectric boundary of the rarefaction wave from the free surface of the striker, or the wave reflected from the striker–screen temperature boundary. Possible bending and twisting of the striker plate causes the shock wave to envelop the electrodes after the probes, which distorts the conductivity profile at $t \lesssim 0.08$ μsec. In this interval data were obtained by interpolating $\int_0^t \sigma dt$ assuming monotonicity of the change in σ, and is shown by dashed lines. The values shown are somewhat elevated above the true σ values, the elevation being greater the smaller t, since it was not considered that at the moment when current appears in the circuit t = 0 there may already be a nonzero thickness layer of compressed argon between the probes.

The change in σ with duration of the material's stay in the compressed state indicates the absence of thermodynamic equilibrium in the argon near the shock wave front. The form of $\sigma(t)$ changes near p = 40 GPa, which is probably caused by an electron level transition — intersection of the 3d and 4s levels [10], with corresponding appearance of localized states at the edges of the conduction and valent zones [2]. For argon at p = 36 GPa [6] recorded a decrease in the slope of the dependence of shock wave velocity on mass velocity, which is apparently also explainable by the appearance of localized states, the filling of which leads to reduction in T and p: we note that a change in the form of $\sigma(t)$ due to fusion and metallization has been observed previously in shock-compressed CsI [11].

Concerning the weak growth in σ at $t \geqslant 0.08$ μsec at $p < 40$ GPa, which apparently corresponds to weak nonequilibrium of the thermodynamic state, and the reduction in relaxation time to ≈ 0.2 μsec for $p > 40$ GPa, it can be proposed that the conductivity values $\sigma_k(p)$ at the end of the measurement time are close to equilibrium. Then the slope of $\log \sigma_k$ as a function of inverse temperature T^{-1} will correspond to the conduction activation energy E. A graph of the function $\log \sigma_k(T^{-1})$ is shown in Fig. 4. Temperatures on the Hugoniot adiabat were determined by interpolation in the p–T plane of data from [1, 12], calculation with an exp-6 potential and consideration of conduction zone narrowing at $p > 40$ GPa. Since the curve $\log \sigma_k(T^{-1})$ is close to linear, the increase in σ_k with growth in T can be described by the method of least squares using the expression $\sigma_k = C \exp(-E/kT)$, where $C = (4.6 \pm 1.0) \cdot 10^5 \cdot \Omega^{-1} \cdot m^{-1}$, $E = 7.2 \pm 0.2$ eV. Such a temperature dependence of conductivity and the large value of C, exceeding the minimum metallic conductivity σ_0, are characteristic of many amorphous and liquid semiconductors, and can be explained by the linear dependence of forbidden zone width on temperature, $W(V, T) = W(V, 0) - 2\gamma T$ [2]. It can be proposed that a nearly linear decrease in W with increase in T is satisfied for $P < 40$ GPa in our case also. Then at T = 0 and $V > 13.5$ $cm^3 \cdot mole^{-1}$ $W \approx$ const ≈ 2 and $E = 14.4 \pm 0.4$ eV, which agrees with the calculations of [1]. Narrowing of the forbidden zone with decrease in volume for $p > 40$ GPa apparently maximizes the increase with temperature of the coefficient γ in the temperature dependence of conduction activation energy E. The increasing rate of forbidden zone width (or more accurately, mobility gap) narrowing due to partial destruction of close order is characteristic of such classical liquid semiconductors as Se, As_2Se_3 [13].

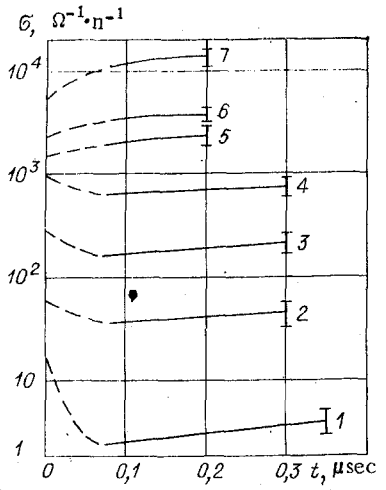

Fig. 3

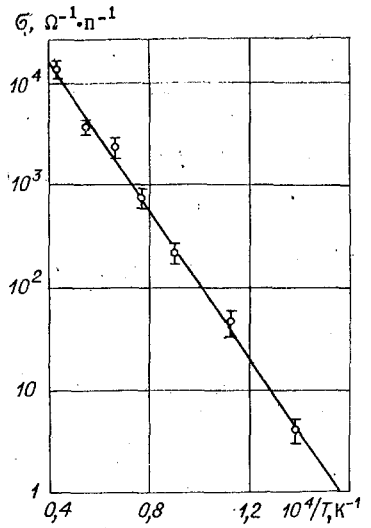

Fig. 4

Possible inaccuracy in the T determination at p = 68.6 GPa has no significant effect on the conclusion of reduction in W at p > 40 GPa, caused by the combined action of temperature and pressure, or the conclusion of absence of liquid argon metallization in the pressure range studied. In fact, in order to obtain a significant decrease in the slope of $\log \sigma_k(T^{-1})$ at p = 68.6 GPa, indicating metallization, it would be necessary to increase the calculation temperature, which is only possible if it is assumed that the forbidden zone width does not increase. On the other hand, the absence of decrease in forbidden zone width assumes an increase in temperature and reduction in activation energy, which contradicts the original assumption. It can be expected that metallization of liquid argon in a shock wave will be caused primarily by an increase in temperature and corresponding collapse of the mobility gap, as in the case of metallization of liquid Se, As_2Se_3.

The relatively long time required for establishment of thermodynamic equilibrium (>0.1 μsec) indicates a process of structural disordering behind the shock wave front, which probably occurs with increase in temperature. Data obtained by the nonequilibrium molecular dynamics method [14] indicate structural relaxation in the shock front. However the relaxation time in the case of shock compression of liquid argon does not exceed $\sim 10^{-12}$ sec in [14]. It is possible that the long relaxation time in the present case is related to the not completely homogeneous structure of the argon before the arrival of the shock wave. In a liquid located in a state close to the liquid-vapor equilibrium line, homophase and heterophase density fluctuations exist [15]. Before arrival of the shock wave, in the absence of boiling, the argon is in a thermodynamically stable state, characterized by a steady state distribution of density fluctuations over number and size. The action of the shock wave leads to an initially inhomogeneous temperature distribution. It is possible that the observed relaxation time is caused by a relatively slow, diffusionlike, character of structural readjustment of the material near the shock-wave front.

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