Electrical conductivity of adiabatically compressed alkali metal vapor

I. M. Isakov, A. A. Likal'ter, B. N. Lomakin, A. D. Lopatin, V. E. Fortov

Branch of the Institute of Chemical Physics, Academy of Sciences of the USSR
(Submitted 29 March 1984)
Zh. Eksp. Teor. Fiz. **87**, 832–839 (September 1984)

The electrical conductivity of adiabatically compressed, highly nonideal cesium and potassium plasmas has been measured at $P \approx 2$–10 MPa and $T \approx 2200$–4300 K. The experiments were carried out in a heated ballistic apparatus in which a range of high plasma densities lying between those of static and shock-wave experiments can be achieved by adiabatic compression of the saturated vapor. The extent to which the measured conductivity exceeds the calculated conductivity conforms to the argument that ion clusters strongly affect the ionizational equilibrium.

I. INTRODUCTION

The physical properties of dense, cool alkali metal plasmas with strong interparticle interactions are presently being studied by two approaches which have access to greatly different parameter ranges[1] (Fig. 1). First, the methods of shock-wave compression[1–3] and pulsed discharges[4,5] can produce states with high temperatures, $T \gg 4\cdot 10^3$ K, far from the saturation curve. In this region there is typically a strong Coulomb interaction in a dense plasma with well-developed ionization. Second, the methods of static (oven) experiments[6,7] can heat plasmas to temperatures no higher than 2300 K in the vicinity of the boiling curve, where the interaction of neutral atoms with charged particles and with each other is important because of the low degree of ionization. It turns out that both the level of the electrical conductivity and its temperature dependence are quite different in these two ranges.

The results of the "high-temperature measurements" of Refs. 1, 2, and 4 ($T \gg 4\cdot 10^3$ K) are described by the conventional models of electron transport in a dense plasma with screening, ion correlations, and electronic excitation.[3]

The results of the "low-temperature measurements" (of which there have been few so far) near the saturation curve[6,7] imply an anomalously high conductivity of a dense, weakly ionized plasma—several orders of magnitude higher than the usual plasma estimates. In Refs. 8–10 the anomalous conductivity was attributed to the formation of heavy ion clusters containing tens of atoms in a dense plasma and to a pronounced shift of the ionization equilibrium toward an increase in the density of conduction electrons. Biberman *et al.*[11] have suggested making use of this effect to develop a high-power MHD generator using a nonideal plasma of alkali metals. It would thus obviously be desirable to carry out measurements in temperature and density ranges lying between those of the static and dynamic approaches.

This range of plasma states presently poses severe difficulties both theoretically and experimentally. Here there are strong and structurally complicated interparticle interactions involving electrons, ions, and neutral atoms, which distort the electron energy spectrum, lead to the production of ion clusters, shift the ionization equilibrium, and lead to a metal-insulator transition.[1] A systematic calculation of the physical properties of a dense, highly nonideal plasma in this region runs into serious complications, which make it necessary to appeal to models based on experimental data.[3]

So far, no experiments have been carried out on the intermediate range of parameters between "cluster plasmas" and "gaseous plasmas." The temperature range $T \gtrsim 2\cdot 10^3$ K is inaccessible to steady-state experiments involving heating with ovens.[6,7] The particular features of the dynamic methods[3] of compressing and heating plasmas in shock waves, on the other hand, restrict these methods to a limited density range and cause a pronounced heating of the plasmas ($T \gtrsim 4\cdot 10^3$ K).

In the experiments reported here we use the adiabatic compression of saturated vapor in a ballistic apparatus to produce nonideal plasmas with temperatures and densities in the intermediate ranges.[12–15] The measurements are carried out in a region of the phase diagram (Fig. 1) in which the thermodynamic manifestations of the nonideal nature of the plasma are slight, while the strong interparticle interaction affects the electrical conductivity and other electronic kinet-

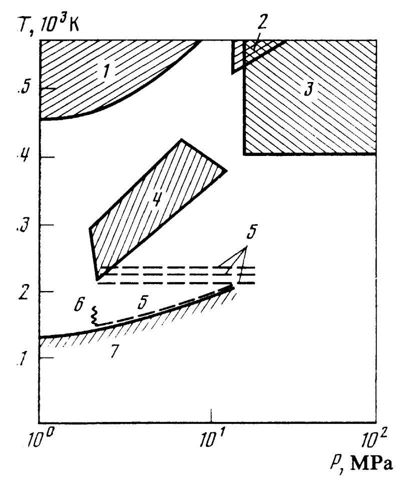

FIG. 1. Regions on the T–P diagram of cesium in which the electrical conductivity has been measured. 1—Ref. 2; 2—Ref. 4; 3—Ref. 5; 4—present experiments; 5—Ref. 6; 6—Ref. 7; 7—phase equilibrium curve.

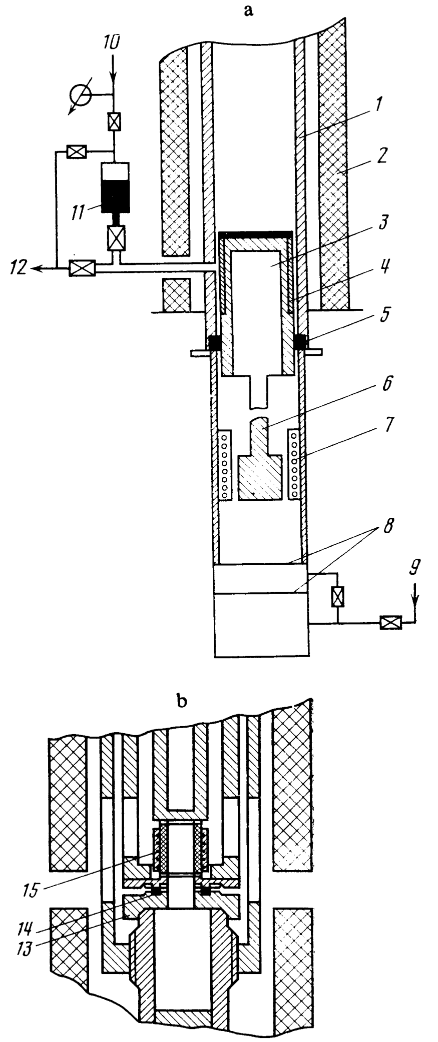

FIG. 2. The adiabatic compression tube. 1—Tube; 2—oven; 3—piston; 4—copper cap; 5—ring of a material similar to Teflon; 6—back of piston; 7—displacement pickup; 8—membranes; 9—pusher-gas inlet; 10—argon inlet; 11—cesium; 12—to vacuum system; 13—collar with beryllium washer 14; 15—device which measures the electrical conductivity.

ic properties of the plasma.

2. EXPERIMENTAL APPARATUS AND DIAGNOSTICS

The experiments are carried out in an adiabatic compression apparatus which is heated to $T = 1150$–1170 K to produce a high initial vapor density of the cesium (up to 0.3 MPa) or the potassium (up to 0.2 MPa) [Ref. 12].

The experimental apparatus is shown in Fig. 2a; Fig. 2b shows separately the upper part of the heated compression tube. Inside the high-temperature steel tube, with an inside diameter of 30 mm, is a closely ground hollow piston of stainless steel (or of titanium for lower temperatures) with a cylindrical copper cap. A copper disk mounted above the piston provides an additional seal and also serves as a shock absorber when the piston strikes the end of the tube. A two-section steel collar with a beryllium washer and a ceramic cylinder makes a butt joint with the upper part of the tube. The ceramic is one of various modifications with an aluminum oxide base; the inner and end surfaces are polished. Independent adjustments ensure a hermetic seal of the collar and the ceramic with the tube. The tube is filled with cesium or potassium vapor after the appropriate argon pressure above the surface of the liquid metal is produced by means of vacuum pumps. The piston in the tube is positioned above the inlet aperture to prevent liquid metal from reaching the surface of the piston (Fig. 2a).

To determine the initial state of the cesium or potassium vapor we measure the temperature of the tube (near the piston, at the center, and at the ceramic) within 2% with a thermocouple, and we measure the vapor density from the absorption of 14-keV x radiation. In addition, we measure the argon pressure above the surface of the liquid metal in the supply system. These measurements are carried as an indirect check, to make sure that there is no vapor condensation in the upper part of the tube, including the surface of the beryllium washer through which the x-ray measurements are carried out. For this purpose, we carried out a special series of experiments to determine the relationship between the vapor density of the alkali metals and the argon pressure. We found that the two are in correspondence over the range $P_0 \approx 0.08$–0.35 MPa for cesium and over the range $P_0 \approx 0.08$–0.2 MPa for potassium.

The vapor density during the compression is determined from the absorption of pulsed x radiation with an energy of 23–25 keV from which the long-wavelength part of the spectrum has been sliced off by additional absorbers. Calibration measurements were carried out with xenon and argon, which are neighbors of cesium and potassium along the atomic number scale. Figure 3 shows an oscilloscope trace of the x radiation corresponding to the last 5 cm of motion of the piston. We simultaneously determine the position of the piston (the volume of compressed vapor) from the change in the signal from an inductive pickup around the back part of the piston. The results of these measurements

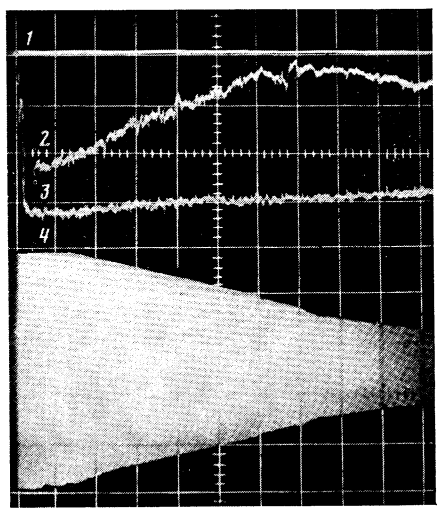

FIG. 3. Oscilloscope trace of the measurements of the x-ray absorption (2) and of the electrical conductivity (4). The sweep time is 0.5 ms/div. 1—Zero line; 3—reference signal corresponding to the initial cesium vapor density.

agree within 10%, and this is the figure which we adopted as the error in the determination of the density on the compression isentrope.

The electrical conductivity is measured by an induction method from the change in the amplitude of a signal in a parallel oscillator circuit whose inductance is a copper wire winding on the surface of the ceramic cylinder. Most of the measurements were taken at the frequency $f = 1.5$ MHz with a conductivity pickup 15 mm in diameter. The circuit was calibrated with samples of graphite and electrolytic solutions with values of σ in the range 20-3000 S/m. The range of measured values corresponds to a ratio of the radius of the pickup to the thickness of the skin layer between 0.1 and 1. Figure 3 shows a typical oscilloscope trace from the measurements of σ, along with a trace of the x radiation. The error in the measurements, which consists of the uncertainty in the calibration and the error in the reading of the oscilloscope traces, is estimated to be 30–50% (the accuracy worsens with decreasing σ). The experimental data may be distorted by the formation of a conducting film on the surface of the ceramic because of vapor condensation or an interaction of the cesium or potassium with impurities in the ceramic. To determine where this effect is influential, we carried out measurements at various frequencies and at various pickup diameters with an admixture of argon.

The temperature and the pressure during the compression were calculated from the measured parameters (the initial density and temperature and the density during the compression) under the assumption of an isentropic process. A special thermodynamic analysis showed that the error of this method of calculating the pressure and the temperature on the isentrope does not exceed 10% over these parameter ranges.

In experiments with adiabatic tubes it is necessary to ensure that the compression is isentropic in order to obtain correct results. During the compression of a condensing medium, a deviation from an isentropic behavior may result from not only heat loss, leakage of gas from the test volume, and the formation of shock waves (on the one hand) but also the condensation of the medium on the tube wall (on the other). The tube wall is cooler than the vapor, which is heated during the compression. Methodological experiments carried out to determine the compression isentropes of mixtures of cesium vapor and argon during each direction of the piston movement[14] demonstrated that dissipative processes are of minor importance. This conclusion agrees with conclusions reached previously from experiments with an artificially developed heat-transfer surface and also with estimates of molecular, radiative, and convective heat transfer.[15] In contrast, direct measurements of the density of the compressed plasma by the x-ray method and the agreement of these measurements with the densities found from the position of the piston allow us to rule out any possible leakage of material through the moving piston in interpreting the experiments. This agreement also indicates that the condensation level of the cesium or potassium in our experiments is the limiting possible level (within the measurement errors).

Although experience with heated adiabatic tubes[15] and shock tubes[16] shows that the condensation is evidently suppressed by the formation of a sheath of uncondensed impurities at the wall, it is not obvious that condensation should have no effect on the readings of the conductivity pickup. For an experimental test of the effect of a conducting film on the measurements we used the following methods: We varied the dimensions of the conductivity pickup, varied the frequency of the circuit over the range 0.5–8 MHz, and added up to 30% argon to the cesium vapor. We used the results of these experiments to selected the pickup dimensions and the frequency for the oscillator circuit for the bulk of the conductivity measurements.

3. EXPERIMENTAL RESULTS AND THEIR DISCUSSION

The experimental results are shown in Figs. 4–6 along with corresponding calculations of the conductivity from Ref. 17. We found values of the electrical conductivity along 13 cesium isentropes with $P_0 = 0.09$–0.31 MPa and $T_0 = 1150 \pm 20$ K and along two potassium isentropes with $P_0 = 0.1$–0.2 MPa and $T_0 = 1170 \pm 20$ K. We see a tendency toward a discrepancy between the experimental and calculated values at high temperatures and again at low temperatures at elevated pressures.

Of particular interest is the behavior of the electrical conductivity of cesium in the vicinity of the liquid-vapor boundary. Figure 6 shows a plot of σ on two isobars according to the measurements of σ along the isentropes; shown for comparison are the results of static experiments.[6,7]

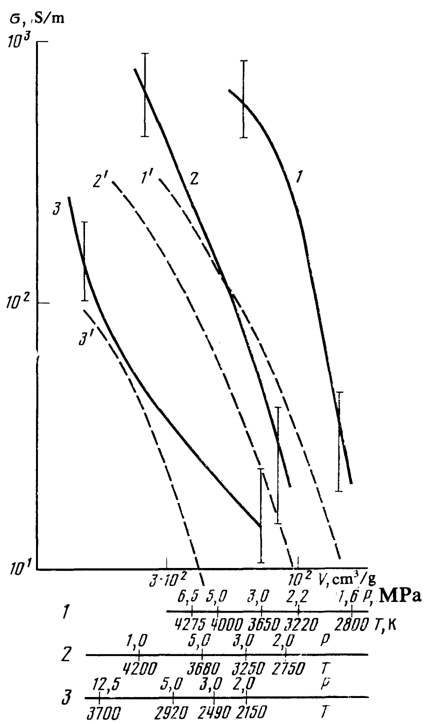

FIG. 4. Electrical conductivity of cesium on several isentropes. 1—$P = 0.11$; 2—0.16 MPa; 3—0.31 MPa. $T = 1150 \pm 20$ K. The dashed lines are calculations from Ref. 17.

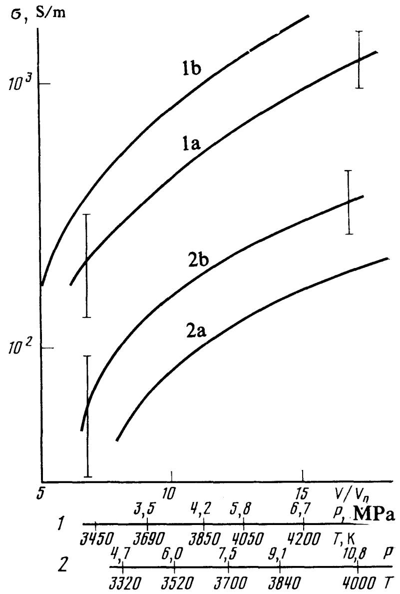

FIG. 5. Electrical conductivity of potassium (a) and of cesium (b) on several isentropes. 1a—$P = 0.11$ MPa; 1b—$P = 0.12$ MPa; 2a, 2b—0.18 MPa.

At a pressure of 2 MPa, our data agree with the results of Ref. 7, within the scatter in the experimental points. On the other hand, two of the points from Ref. 6 lie about an order of magnitude above our experimental data. The probable reason for this discrepancy is that the experimental error is greatest at low conductivities. At a pressure of 5 MPa (Fig. 6b), we have considerably fewer experimental results, but the temperature dependence of the conductivity is apparently of the same nature as at $P = 2$ MPa.

The picture of the electrical conductivity of cesium vapor as a function of the temperature and pressure which emerges from these experimental results is qualitatively different from the usual picture for a weakly ionized ideal gas. The deviation from an ideal behavior is seen in two ways under our conditions. First, the Coulomb and polarization interactions between free electrons, ions, and atoms lead to an effective lowering of the ionization potential. Second (but no less important), the interparticle interaction leads to the formation of ion clusters, in each of which several atoms are bound. It is the conversion of positive atomic ions into cluster ions which is responsible for the anomalously high density of conduction electrons and thus the high electrical conductivity of a vapor near saturation.[8–10]

Since the electrical conductivity of a weakly ionized plasma is determined primarily by the number of conduction electrons, an interpretation of the experimental results requires a calculation of the degree of ionization, which depends on the ion composition of the plasma. If the ion mass spectrum is restricted to diatomic and triatomic ions, the composition can be calculated easily from the available information on the constants of these molecular ions. Working in this manner we can obtain a rather good description of the high-temperature branch of the dependence (Fig. 6), but we cannot describe the low-temperature branch. At low temperatures, we must take into account the far heavier ion clusters, which contain up to 20 atoms. These clusters are treated as nonrigid molecules in which internal rotations can be excited easily (liquid-drop models have also been proposed for very large clusters[9,10]). The simplest model (a self-consistent central field independent of the number of atoms in the cluster) contains two parameters, which turn out to be quite close to the corresponding parameters of a diatomic ion. As a consequence we have some good interpolation properties, so that this model can be applied over a broad range of parameters.

The ion composition is described by a Poisson distribution with the following average number of atoms bound to an ion:

$$S = \frac{1}{2} n\lambda^3 \frac{T^2}{B\omega} \exp\left(\frac{q-\Delta}{T}\right). \tag{1}$$

Here n is the density of atoms, $\lambda = (4\pi h^2/mT)^{1/2}$, m is the mass of an atom, T is the temperature, the parameter $B\omega$ is equal to the product of the rotational constant and the frequency of the radial vibrations, q is the binding energy of the atom, and Δ is the threshold for the excitation of rotations

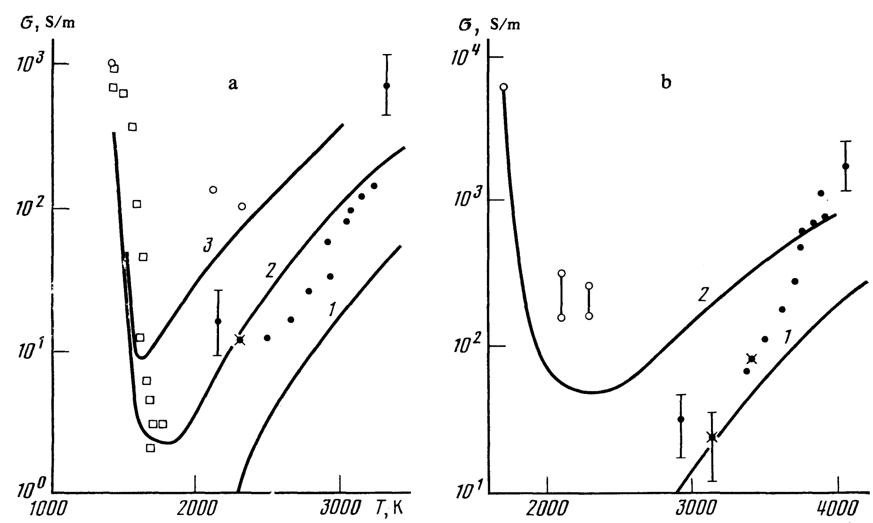

FIG. 6. Electrical conductivity of cesium on isobars: a—2.0 MPa; b—5 MPa. ○—Data from Ref. 6; □—data from Ref. 7; ●—present experiments (the crosses are points found in measurements with an argon admixture). Calculated curves: 1—According to Ref. 17; 2—the present study; 3—Ref. 9.

(this threshold is inconsequential at temperatures $T \gtrsim 5T_m$, where T_m is the melting point of the metal). The total density of positive ions is $n_i \exp S$, where n_i is the density of atomic ions.

The degree of ionization of the plasma is given by

$$n_e/n = (n\lambda_e^3)^{-1/2} \exp(-I/2T + S/2 + \Delta I_p/2T)(1+\gamma_0/2), \quad (2)$$

where n_e is the electron density, λ_e is the thermal wavelength of an electron, I is the ionization potential of the atom, and ΔI_p is the lowering of this ionization potential caused by the polarization interaction of electrons and ions with atoms. The last factor in (2) is a Debye correction calculated in a grand canonical ensemble [γ_0 is the Debye interaction parameter corresponding to the electron density and found from (2) without the Debye correction]. It is also a simple matter to correct (2) for negative ions.

The temperature dependence of the electrical conductivity shown in Figs. 6a and 6b and corresponding to Eq. (2) agrees qualitatively with the experimental data both in the region in which complex ions play a dominant role and in the region in which these ions are not very important. Another possible reason for the quantitative discrepancies would be the experimental errors and the uncertainties in the calculation factors.

At high temperatures the experimental results also agree qualitatively with the calculations of Ref. 17, which focused on the Coulomb interaction. There, the ion clusters play essentially no role because of dissociation.

We wish to thank V. E. Bespalov for assistance is designing the experiment and V. I. Kucherenko for calculating the electrical conductivity on the compression isentropes.

[1]V. A. Alekseev, I. T. Yakubov, and V. E. Fortov, Usp. Fiz. Nauk **139**, 193 (1983) [Sov. Phys. Usp. **26**, 99 (1983)].
[2]V. A. Sechenov, É. V. Son, and O. V. Shchekotov, Pis'ma Zh. Tekh. Fiz. **1**, 891 (1975) [Sov. Tech. Phys. Lett. **1**, 388 (1975)].
[3]V. E. Fortov, Usp. Fiz. Nauk **137**, 361 (1982) [Sov. Phys. Usp. **25**, 448 (1982)].
[4]I. Ya. Dikhter and V. A. Zeĭgarnik, Dokl. Akad. Nauk SSSR **227**, 656 (1976).
[5]N. V. Ermokhin, B. M. Kovalev, P. P. Kulik, and V. A. Ryabyĭ, Teplofiz. Vys. Temp. **15**, 695 (1977).
[6]H. Renkert, F. Hensel, and E. Franck, Ber. Bunsenges. Phys. Chem. **B75**, 507 (1971); F. Hensel, in: Proceedings of the Eighth Symposium on Thermophysical Properties, Gaithersburg, USA, 1981.
[7]V. A. Alekseev, A. A. Vedenov, V. G. Ovcharenko, *et al.*, High Temp. High Press. **7**, 676 (1975).
[8]A. A. Likal'ter, Teplofiz. Vys. Temp. **16**, 1219 (1978); **19**, 746 (1981); **21**, 249 (1983).
[9]A. N. Lagar'kov and A. K. Sarychev, Teplofiz. Vys. Temp. **17**, 466 (1979).
[10]I. T. Yakubov, Dokl. Akad. Nauk SSSR **247**, 841 (1979) [Sov. Phys. Dokl. **24**, 634 (1979)].
[11]L. M. Biberman, A. A. Likal'ter, and I. T. Yakubov, Teplofiz. Vys. Temp. **20**, 565 (1982).
[12]I. M. Isakov and B. N. Lomakin, Teplofiz. Vys. Temp. **17**, 262 (1979).
[13]B. N. Lomakin and A. D. Lopatin, Teplofiz. Vys. Temp. **21**, 190 (1983).
[14]A. A. Borzhievskiĭ, V. A. Sechenov, and V. I. Khorunzhenko, Teplofiz. Vys. Temp. **21**, 181 (1983).
[15]A. T. Kunavin, A. V. Kirillin, and Yu. S. Korshunov, Teplofiz. Vys. Temp. **11**, 261 (1973); **13**, 1304 (1974).
[16]B. N. Lomakin and V. E. Fortov, Zh. Eksp. Teor. Fiz. **63**, 92 (1972) [Sov. Phys. JETP **48** (1973)].
[17]V. K. Gryaznov, I. L. Iosilevskiĭ, Yu. G. Krasnikov, *et al.*, Teplofizicheskie svoĭstva rabochikh sred razofaznogo yaderpogo reaktora (Therophysical Properties of the Working Media of Gas-Phase Nuclear Reactors) (ed. V. M. Ievlev), Atomizdat, Moscow, 1980.

Translated by Dave Parsons