Permittivity measurement at high dynamic pressures

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A method is proposed for measuring the permittivity ϵ at high dynamic pressures with a time resolution of $\sim 10^{-7}$ sec and an error of 1–2%. The permittivity ϵ of benzene was determined at 9 MHz as a function of compression σ up to $\sigma = 1.79$. It was found that, within the range of validity of the Lorentz-Lorenz formula, the polarizability of benzene molecules falls with rising pressure.

Dielectric measurements at high and ultrahigh pressures can give valuable information on the nature of intermolecular interactions and the structure of molecules under these extremal conditions. However, methodological difficulties do not permit experiments to be carried out under static pressures exceeding several gigapascals (GPa). For this reason use is made of dynamic compression methods. The absence of restrictions associated with the strength of the container around a sample makes it possible to extend considerably the range of pressures in investigations of the properties of dielectrics under dynamic conditions. The extremely short compression and heating times in a shock wave, amounting to $\sim 10^{-12}$ sec for a liquid,[1] provide an opportunity to investigate, in principle, metastable states of matter and the kinetics of fast phase transitions such as melting and solidification.

The present paper describes a system for the determination of the permittivity ϵ with a time resolution of $\sim 10^{-7}$ sec behind a shock-wave front and it reports the dependence of ϵ of benzene on the degree of compression σ up to $\sigma = 1.79$, which corresponds to dynamic pressures of ~ 13 GPa. The error in the determination of ϵ in a single experiment is estimated to be 1–2%.

The basic idea of this measurement method is the determination of the modulus of the complex resistance of a cell compressed by a shock wave at a fixed frequency and the calculation of ϵ from the known parameters of the cell.

The apparatus is shown schematically in Fig. 1. A measuring cell was a parallel-plate capacitor, formed by an electrode 1 and a screen 4. The electrode was made of aluminum foil, 0.01 mm thick, which was bonded to a Teflon film 2. This film was stretched by rings 3 parallel to the screen at a distance of $x_0 = 3$ mm from the latter.

A plane shock-wave front, formed by an explosive generator (80 mm in diameter and 120 mm high), passed through the screen and the sample with hardly any reflection from the thin electrode. The matter between the screen and the electrode was subjected for about 10^{-6} sec to a high and slowly falling dynamic pressure. The values of the pressure and the degree of compression were calculated from the known state of the screen material and from the shock adiabat of benzene.[2]

The duration of application of the pressure indicated that a probing frequency f should exceed several megahertz. In our experiments the value of f was 9–10 MHz. A current of this frequency was supplied from an oscillator 6 to a tuned amplifier 7, which was based on a GU-34B vacuum tube with an amplitude stabilization

circuit. The anode voltage supplied to the tube was 1.5 kV. The need to use a power amplifier was dictated by the stringent requirements for suppression of electrical interference in the system because the expansion of an ionized cloud of detonation products created a considerable electrical noise.

The sensitivity of the system was increased by selecting the differential measurement principle. The output end of the amplifier 7 was connected to a 5-m long coaxial cable with a wave impedance of 150 Ω. The high-frequency signal passed through this cable and through identical capacitors C_1 and C_2 to a measuring channel including the cell and a reference channel with a balancing capacitor C_5. These capacitors, as well as capacitors C_3 and C_4, were attached to a Textolite plate in the direct vicinity of the cell.

The output of a broadband differential amplifier 8 produced a voltage difference between the reference and measuring channels and this difference was recorded with a fast-response oscilloscope 9. The voltage phases reaching the inputs of the amplifier 8 were made identical by ensuring that the system was rigorously symmetrical and by including controlled phase shifters.

The oscilloscope scan was triggered 1–2 μsec before the shock compression of the cell and this was done by an electric contact device which produced a signal applied through a shaper 10 and a controlled delay unit to the input of the lock-in amplifier of the OK-33 oscillograph.

Special experiments demonstrated that in the case of a sudden change in the capacitance equivalent to the cell, the transient processes ended during one oscillation, i.e., approximately in 10^{-7} sec.

Before each experiment we plotted the dependence of the amplitude of the deflection h of the oscilloscope
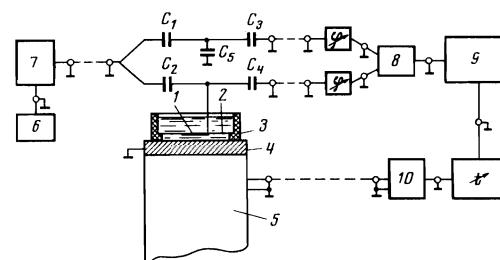

FIG. 1. Schematic diagram of the apparatus: 1) electrode of 30 mm in diameter; 2) Teflon film 0.015 mm thick; 3) ebonite rings of 50 mm in diameter; 4) copper or aluminum screen, 80 mm in diameter and 8 mm thick; 5) explosive substance; 6) GZ-7A oscillator; 7) tuned amplifier; 8) differential amplifier; 9) OK-33 oscilloscope; 10) trigger pulse shaper; $C_1 = C_2 = C_3 = C_4 = 1$–2 pF.
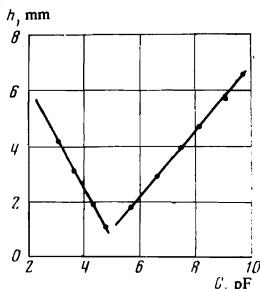
FIG. 2. Calibration graph.

FIG. 4. Dependences of the value of $\sqrt{\epsilon}$ of benzene on the degree of compression: 1) experimental results; 2) values calculated from the Lorentz-Lorenz formula; •) dielectric measurements; ■) refractive index measurements [3].
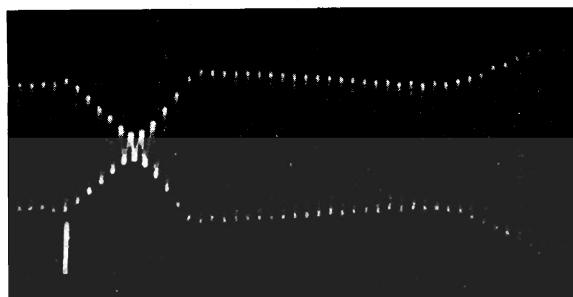

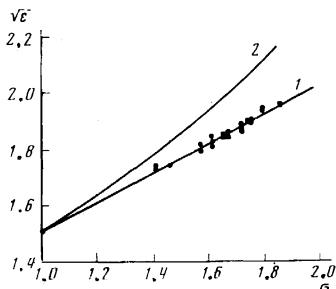FIG. 3. Experimentally obtained oscillogram, f=9.2 MHz.

beam recorded photographically on the capacitance of standard capacitors connected in parallel with the empty cell. One of such graphs is shown in Fig. 2. We can see that the sensitivity of the system was ~1 mm/pF for a linear dependence h(C).

A typical oscillogram obtained in our experiments is shown in Fig. 3, where the arrow identifies the moment of arrival of a shock wave in the sample. In this experiment the capacitance C_5 was selected to be slightly greater than the initial cell capacitance but smaller than the capacitance of the cell in the compressed state. The cell capacitance increased during the motion of the shock-wave front between the screen and the electrode. The voltage drop across the input of the amplifier 8 first fell, passed through a minimum, and began to rise. Some increase in the oscillation amplitude at the end of the oscillogram corresponded to the arrival of a second shock wave in the sample from the screen.

The oscillograms obtained and the calibration graphs were used to find the difference between the cell capacitances at a high pressure and in initial state. Then, the known initial parameters of the cell were used to calculate its capacitance in the compressed state. This procedure allowed us to eliminate stray capacitances of the assembly.

The permittivity of the compressed matter was determined by calibrating the cell with liquids of known permittivities ϵ. A liquid layer above the electrode was replaced with an insulator disk whose permittivity was identical with ϵ of the investigated sample under normal conditions. For example, in the case of benzene we used polyethylene as the insulator. A micrometer screw set the disk with the electrode at a distance from the screen equal to x_0/σ and the cell capacitance was de-

termined as a function of ϵ of the calibrating liquid, and then ϵ was found behind the shock-wave front.

The main error in the measurement of ϵ by the above method was due to electrical noise, which resulted in slight random variations of the high-frequency voltage at the output of the amplifier 8. This error was estimated to be 1—2%.

The experimental dependence of the value of $\sqrt{\epsilon}$ of benzene on the degree of compression σ is plotted in Fig. 4, where each point represents a separate experiment. Throughout the investigated range of densities this dependence is described satisfactorily by the straight line $\sqrt{\epsilon} = 0.985 + 0.525\sigma$.

Figure 4 includes also the dependence of $\sqrt{\epsilon}$ on the degree of compression calculated from the Lorentz-Lorenz formula on the simplest assumption that the polarizability of the benzene molecule α remains constant in spite of changes in its volume. We can see that the calculated curve deviates considerably from the experimental dependence. Thus, we may conclude that, according to the Lorentz-Lorenz theory, the polarizability α of benzene molecules decreases as a result of compression.

It is interesting to compare these results with those reported by Yushko et al.[3] on the dependence of the refractive index of benzene on the dynamic pressure, calculated using Fresnel formulas on the basis of the measured coefficients of reflection of light from a shock front. We can see from Fig. 4 that the results of the optical measurements of Yushko et al. agree with our values of $\sqrt{\epsilon}$. This means that, throughout the investigated range of pressures, we find that, firstly, the contribution of the atomic polarizability to the permittivity of benzene is negligible, and secondly, during the time intervals used in our study no significant physical or chemical changes take place in benzene behind a shock-wave front.

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[2] R. D. Dick, J. Chem. Phys. 52, 6021 (1970).
[3] K. B. Yushko, G. V. Krishkevich, and S. B. Kormer, Pis'ma Zh. Eksp. Teor. Fiz. 7, 12 (1968) [JETP Lett. 7, 7 (1968)].

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