

Metal-to-Nonmetal Transitions

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The Metal–Nonmetal Transition in Fluid Mercury: Landau–Zeldovich Revisited

Friedrich Hensel

Abstract. The paper reviews recent experimental results, which show that the formation of a mixed phase of metallic and nonmetallic domains following the putative ‘Landau–Zeldovich first-order metal–nonmetal transition’ noticeably influences the liquid–vapor transition of fluid mercury. An attempt is made to connect the observable consequences of the mixed phase existence with the scenario of noncongruent evaporation.

2.1 Introduction

An important open problem in the statistical mechanics of fluids is an understanding of the interrelation of the metal–nonmetal transition and the liquid–vapor phase transition in metallic systems. It is now 50 years since Landau and Zeldovich [1] first called attention to this question, specifically in relation to liquid mercury. While emphasizing that one cannot distinguish metal (M) from nonmetal (NM) above the absolute zero of temperature, they nevertheless proposed that electronic transitions could introduce additional lines of first-order, that is, discontinuous electronic transitions in the phase diagram of the fluid state. They suggested the three possible diagrams shown in Fig. 2.1. Lines of first-order metal–nonmetal transitions might occur wholly in the liquid (Fig. 2.1b) or vapor state (Fig. 2.1c) or, as a third possibility, the transition might be an extension of the liquid–vapor pressure curve (Fig. 2.1a) beyond the critical point into the supercritical state. Specifically for mercury, Landau and Zeldovich [1] propose Fig. 2.1b with a triple point at the intersection of the liquid–vapor and the metal–nonmetal transition.

The problem that throughout the fluid range of mercury the electronic structure can change discontinuously with temperature and pressure has stimulated extensive experimental and theoretical efforts and the general subject has been repeatedly reviewed in the literature [2–10]. A comprehensive review of the available experimental and theoretical results is therefore unnecessary, and I shall refrain from any attempt to cover the entire field. Instead, I have

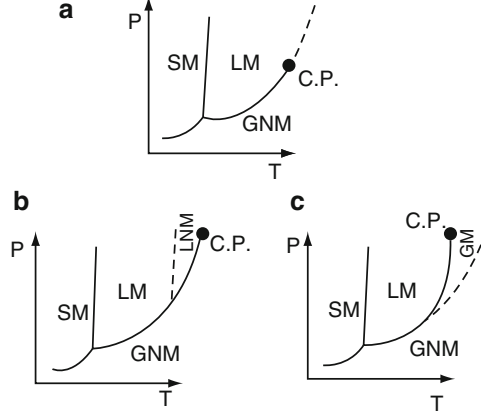


Fig. 2.1. Schematic pressure-temperature (p-T) phase diagrams proposed by Landau and Zeldovich [1]: S = solid, L = liquid, G = gas, M = metal, NM = non-metal. The critical point C.P. terminates the line of liquid-gas equilibrium. *Dashed curves* indicate lines of proposed first-order M-NM transitions

selected for attention recent, partly unpublished, experimental results which evidently show that the apparently continuous changes in the electric structure and transport properties of fluid mercury observed during the course of the metal-nonmetal (M-NM) transition are connected with local microscopic inhomogeneities in the electronic structure.

The renewed interest in this problem is mainly motivated by the exciting progress in the search for density fluctuations inherent to the M-NM transition in expanded fluid mercury, which has come from the recent small angle X-ray scattering (SAXS) measurements of Inui et al. [11–13] and from very accurate ultrasound velocity [14] and absorption [15, 16] measurements by Yao and coworkers and by Kozhevnikov [17].

2.2 The Liquid-Vapor Phase Boundary of Mercury

The SAXS experiments on mercury under extreme conditions of temperature and pressure in the vicinity of the critical point and the M-NM transition were carried out on the high-energy X-ray diffraction beam line (BL04B2) at SPring-8 in Japan [13]. Inui et al., were able to observe a change in the character of the SAXS-intensity as the density of liquid mercury increases from the critical region to higher densities where a continuous change from nonmetallic to metallic behavior has been predicted. Analysis of the data employing the usual Ornstein-Zernike plots shows that the long wavelength limit of the scattering function $S(0)$ and the correlation length ξ follow approximately through the peak around the critical point, but with increasing density $S(0)$

continues to decrease while ξ falls to a constant value, independent of temperature, of about 5–6 Å, indicating fluctuations of different character, which Inui et al. [13] ascribed correctly to fluctuations between insulating and metallic regions on a length scale of intermediate-range order. However, the Ornstein–Zernike analysis does not permit to truly separate the additional scattering related to the M–NM transition from the critical scattering. Be that as it may, the exciting observation of Inui et al. [13] stimulated Maruyama et al. [18] to apply the reverse Monte Carlo [19] method and the Voronoi–Delaunay analysis [20] on the basis of the structure factors determined by X-ray diffraction experiments by Tamura and Hosokawa [21] to characterize the intermediate-range fluctuations in the M–NM-range. The model structure resulting from the void analysis shows a well defined binary mixture of M- and NM-domains with arbitrary sizes and shapes. The concentration of the M-domains increases with increasing density and the spatial distribution of these domains resembles micro- or better nanoemulsions, that is the system can be treated as a two-density model. The assumption is then, that there is a matrix of density ρ_1 in which are embedded particles or domains of density ρ_2 . More generally, the density in the system is either ρ_1 or ρ_2 . If the two phases have sharp boundaries, there are general rules called the Porod law and the Porod invariant [22]. And as a matter of fact, a new inspection of the SAXS-data [23] over the density range from 3.5 g cm^{-3} to 11 g cm^{-3} covered by the experiment [11, 12] shows that the data are consistent with Porod’s theory of two-phase systems [22]. In particular, in the liquid mercury density range between about 11 and 9 g cm^{-3} , the coexistence of M nanodomains with a density of 10.7 g cm^{-3} and NM-nanodomains with a density of 8.3 g cm^{-3} is obtained.

As pointed out by Landau and Zeldovich [1], a salient feature concerning the M–NM transition in mercury is the possible occurrence of a first order phase transition, characterized by a discontinuous change in density from ρ_M to ρ_{NM} , that is there exists an interval in which the equilibrium state is macroscopically phase-separated into regions of higher and lower than average density. However, when long range Coulomb forces are taken into account, this instability with macroscopic separation is frustrated due to electrostatic energy cost. A Coulomb interaction precludes macroscopic phase separation: consequently, the system can form intermediate phases ‘electronic micro- or nanoemulsions,’ where domains of one phase (M) are embedded into the other phase (NM). A large number of small domains would minimize the Coulomb energy, but they cost too much surface energy. The distance between the domains and their size are determined by minimizing a free energy, which takes into account both the effects [24, 25].

It goes without saying that the finding of the formation of a nanoemulsion in fluid mercury for densities smaller than about 11 g cm^{-3} has implications for the interpretation of the phase behavior and the electronic transport properties. In particular, liquid mercury can no longer be considered as a homogeneous one-component fluid for which the liquid–vapor phase boundary in the pressure (p)–temperature (T) plane is represented by a single line, the

vapor pressure curve. If it forms a binary mixture of M- and NM domains, the general features of its liquid–vapor transition are known from the thermodynamics of mixtures; that is, the two-phase region in the p – T -plane is no longer a single line but a two-dimensional domain, whose boundary parameters depend on the $[NM]/[M]$ concentration ratio. Before turning to the discussion of new results, it is inevitable that some steps of the previous reviews will be retraced in order that the present account should not be unsystematic.

There used to be general agreement that the most significant experiments relevant to the exploration of the relationship between the liquid–vapor and metal–nonmetal transitions in fluid mercury are direct measurements of electrical properties that signal the transformation from a metallic to a non-metallic state. Data such as those of the electrical conductivity (see Fig. 2.2) [26–30] clearly demonstrated that for fluid mercury there is no sharp (first-order) electronic transition except across the apparent liquid–vapor phase boundary, that is, the liquid–vapor phase separation tends to separate the nonmetallic and metallic fluids. Near the apparent critical point the conductivity drops sharply, thus showing a strong effect of the incipient phase transition on the electronic structure. The close correlation between the behavior of the density and that of the electrical conductivity (Fig. 2.2) convincingly shows that the variation of elemental density is the dominant factor governing the metal–nonmetal transition. However, it has to be emphasized that in practice very careful measurements are required to separate the ever present effects of density and temperature in the apparent critical region. Part of the difficulty arises because both the compressibility and also the pressure derivative of the electrical conductivity become very large in the critical region. This means that small errors in pressure measurement will cause large density and conductivity errors. Consequently, for a reliable correlation of the conductivity and density, precise temperature and pressure control is essential. This is not easily achievable, because precise measurements of the properties of fluid metals are notoriously difficult. This is immediately evident from the fact that the critical point of mercury is near a temperature $T = 1,478^\circ\text{C}$ and a pressure $p = 1,673$ bar well beyond the range of standard experimental techniques of condensed matter physics.

A serious additional experimental problem related to the determination of the liquid–vapor phase boundary in mercury is that nearly all investigations – including the experimental data in Fig. 2.2 – are not concerned with saturation conditions where both liquid and vapor are present in equilibrium. Instead, the method usually employed was to heat a sample continuously at a constant pressure until an abrupt change of the measured property (e.g., the conductivity) signals that apparently the liquid boiled out of the cell at the vaporization point. This is a very efficient method and the vapor pressure data obtained in this way are very accurate and reproducible. However, it goes without saying that the employment of this method by nearly all experimentalists working on the determination of the liquid–vapor phase boundary of mercury is based on the assumption that mercury forms a homogeneous one-component

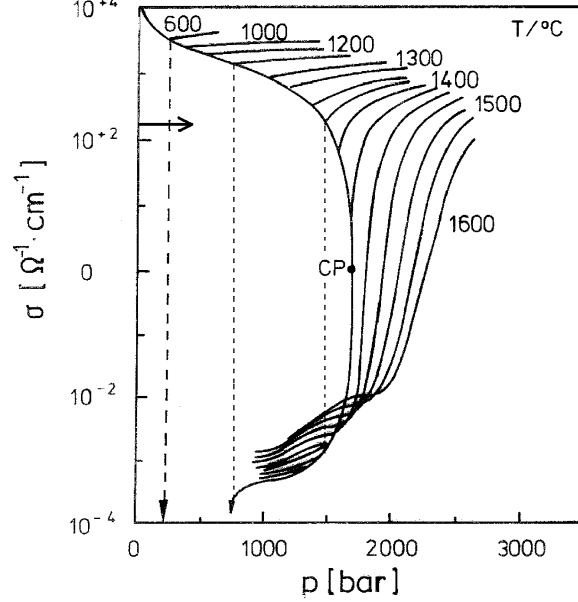


Fig. 2.2. DC electrical conductivity data for mercury at sub- and supercritical temperatures as a function of pressure [26–29]

liquid; thus, ignoring the fact that the thermal equilibrium times in a mixture liquid–vapor two-phase region are much longer than those of pure substances for a given temperature distance ΔT from the transition [31]. There is only one experiment described in the literature that was mainly concerned with saturation conditions where both liquid and vapor were present in equilibrium [32,33]. The authors of this work – also assuming that mercury is a homogeneous one-component liquid over the whole liquid range – pointed out that their vapor pressure curve $p(T)$ of mercury – measured under true equilibrium conditions – has an unusual and probably unique form in that the logarithm of the pressure (p) against the reciprocal temperature ($1/T$) plot showed a relatively sharp change of slope at a temperature of 1,088°C (1,361 K) and a saturated liquid density $\rho_L = 10.7 \text{ g cm}^{-3}$. As mentioned earlier, the latter is about the density at which the inspection [23] of the SAXS-data [11] shows that liquid mercury starts to form a nanoemulsion.

Seen at glance from Fig. 2.3, the two experimentally determined curves BC and SC (solid lines) do not intersect. The dashed line (see *inset*) is only an attempt (assuming that the two curves approach each other) to compare the phase behavior of mercury with well-known features from the theory of fluid binary mixtures [34], that is cricondentherm T_{max} , cricondenbar p_{max} , and the critical point C.P., for which the composition of the two coexisting phases is equal. The total vapor pressure within the two-phase region of mercury is plotted as a function of density in form of isobars or isotherms, respectively, in

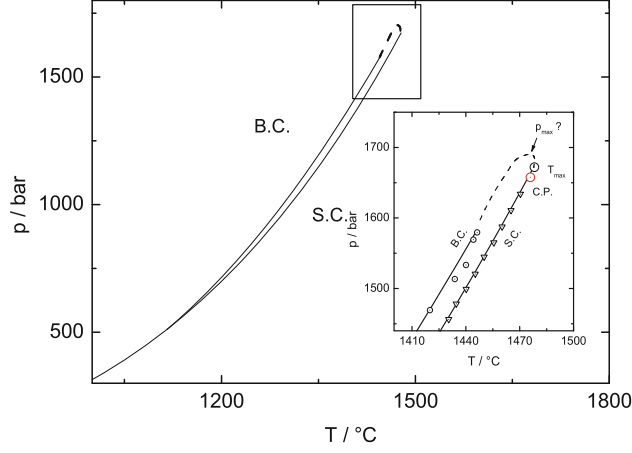


Fig. 2.3. Pressure-temperature diagram of fluid mercury: two-dimensional two-phase region for temperatures higher than $T = 1,088^\circ\text{C}$ is observed instead of the standard p - T saturation curve; BC -boiling (liquid) conditions, $C.P.$ critical point; $T_{\max}$ = cricondenthem, $p_{\max}$ = cricondenbar. As the critical point of binary systems with variable composition is difficult to locate, we choose the location at $([NM]/[M])_{BC} = ([NM]/[M])_{SC}$

Figs. 2.4 and 2.5. The isothermic phase transition starts and finishes at different pressures, while the isobaric phase transition starts and finishes at different temperatures. The curves inside the two phase regions are not really straight lines, and they should be simply considered as a guide to the eyes. They are horizontal only for temperatures smaller than $1,088^\circ\text{C}$ or pressures smaller than 458 bar, respectively.

In Fig. 2.3, we displayed both vapor pressure branches: the pressure obtained for slow evaporation [32,33] under true equilibrium conditions resulting in the bubble or boiling curve, which we designate BC together with the pressure obtained for nonequilibrium conditions when temperature or pressure are changed with a finite rate [26]. The latter results in the saturation curve, which we designate SC. The unusual form of the phase boundary has implications for the behavior of nearly all properties of mercury, which have not been recognized in the past. The essential difficulty is that the effect is small. This is immediately evident from the effect of isothermal evaporation on the DC electrical conductivity σ , which has been measured at BC condition [32] and SC-condition [26]. Figure 2.6 displays the effects of evaporation for the two isotherms $T = 1,350^\circ\text{C}$, and $T = 1,400^\circ\text{C}$. The differences in pressure for SC- and BC-conditions are not large enough to be seen in a diagram as that shown in Fig. 2.2. As the electrical conductivity under these conditions can be well described [36], by the effective medium approach [35] we are able to calculate the volume fraction ϕ of the metallic component at liquid BC- and SC conditions. The values of ϕ are displayed in the plot of Fig. 2.6. Similar effects are observed for evaporation at the isobar of 1,400 bar (see Fig. 2.7).

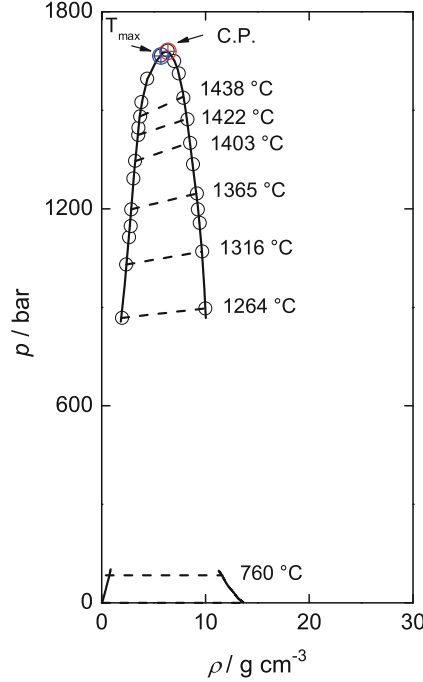


Fig. 2.4. Non-congruent pressure density diagram. For temperatures higher than 1,088°C, the isothermal phase transition starts and finishes at different pressures. The dashed lines are not measured curves but serve only as a guide for the eyes. *BC* = boiling curve, *SC* = saturation curve; $T_{\max}$ = cricondentherm. C.P. critical point where $([NM]/[M])_{BC} = ([NM]/[M])_{SC}$

The availability of electrical conductivity data at BC- and SC-conditions permits us to calculate the volume fractions of the components by employing the effective medium theory. Data for the volume fraction of the metallic component are presented in Fig. 2.8 in the form of isobars as a function of temperature. The difference in the volume fraction at liquid BC- and liquid SC-condition is relatively large. The dashed line is based on the assumption that the volume fractions for SC- and BC-condition is equal at the ‘apparent’ critical point.

The scenario is thus very similar to that of noncongruent phase transitions highlighted by Iosilevskiy and colleagues [37–39] in relation to the evaporation of uranium dioxide. The effect of the noncongruent phase transition is most vividly seen in the shape of the liquid vapor coexisting curve. Figure 2.9 shows an extension of the density–temperature plot of Fig. 2.5 over the whole liquid–vapor coexisting range. The most remarkable feature is the strong deviation from the empirical rule of rectilinear diameters of Cailletet and Mathias [40].

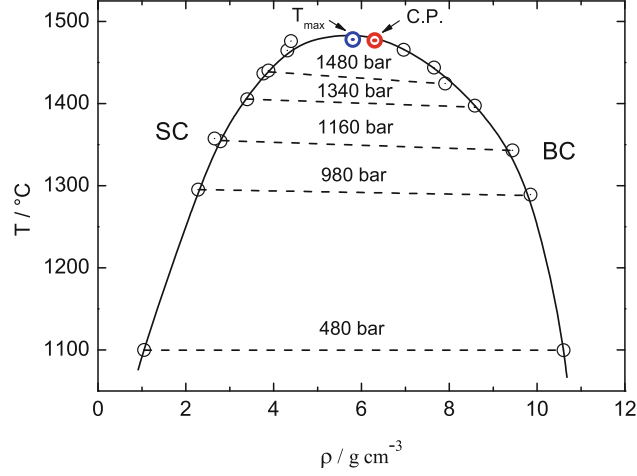


Fig. 2.5. Noncongruent temperature density diagram. For pressures higher than about 450 bar, the isobaric phase transition starts and finishes at different temperatures. The *dashed lines* are not measured curves but serve only as a guide for the eyes. *BC* = boiling curve, *SC* = saturation curve, $T_{\max}$ = cricondentherm

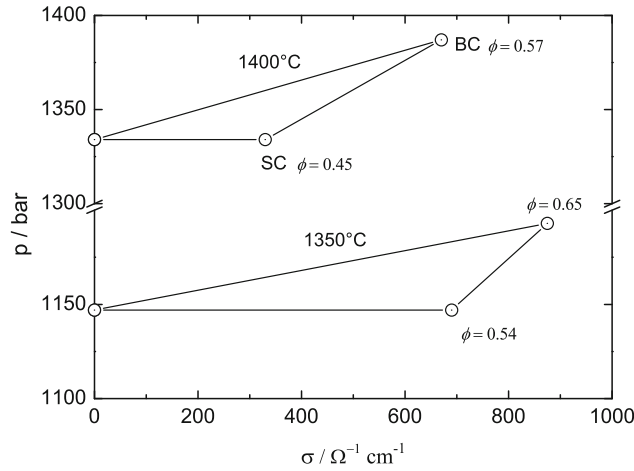


Fig. 2.6. Noncongruent pressure–electrical conductivity σ diagram for $T = 1,350^\circ\text{C}$ and $T = 1,400^\circ\text{C}$, ϕ is the volume fraction of the metallic component. The conductivity of the coexisting vapor is smaller than $10^{-3} \text{ ohm}^{-1} \text{ cm}^{-1}$

A remarkable wiggle is observed at about a temperature of $1,088^\circ\text{C}$. It is obvious that a law of corresponding states is not valid.

With the knowledge of the data in Figs. 2.4, 2.5 and 2.8, it is easy to calculate different properties that are effected by noncongruent evaporation. As an

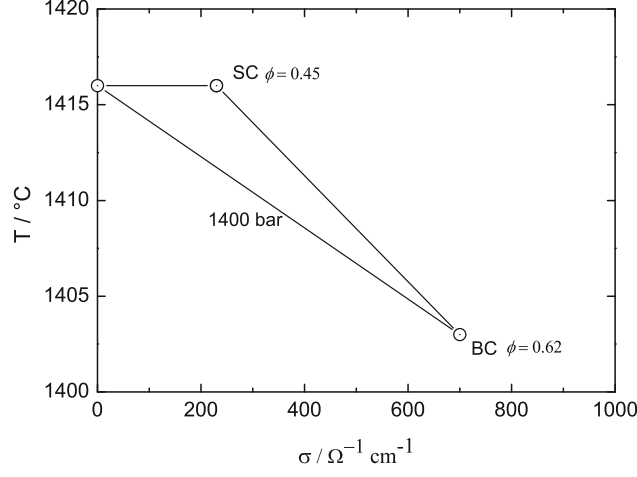


Fig. 2.7. Noncongruent temperature–electrical conductivity σ diagram for $p = 1,400$ bar, ϕ is the volume fraction of the metallic component. The conductivity of the coexisting vapor is smaller than $10^{-3} \text{ ohm}^{-1} \text{ cm}^{-1}$

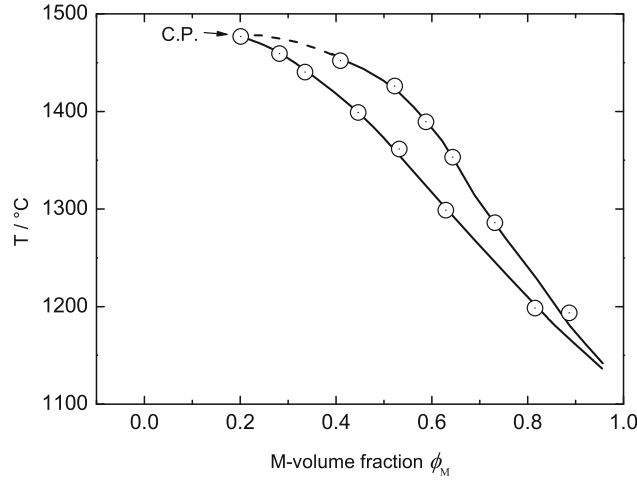


Fig. 2.8. Volume fraction ϕ of the metallic component at BC liquid and SC liquid conditions. ϕ is calculated from the measured electrical conductivity employing the effective medium theory [35]. The *dotted line* is an extrapolation assuming that ϕ along the BC- and SC- curves becomes equal at C.P

example, we calculate for the density of 9.45 g cm^{-3} at the liquid BC-branch different derivatives

$$\left(\frac{\partial \rho}{\partial T}\right)_{\text{BC}} = 0.007 \frac{\text{g}}{\text{cm}^3 \text{K}}; \left(\frac{\partial p}{\partial \phi}\right)_{\rho, T} = 669 \text{ bar}; \left(\frac{\partial \phi}{\partial \rho}\right)_{p, T} = -0.4 \frac{\text{cm}^3}{\text{g}}$$

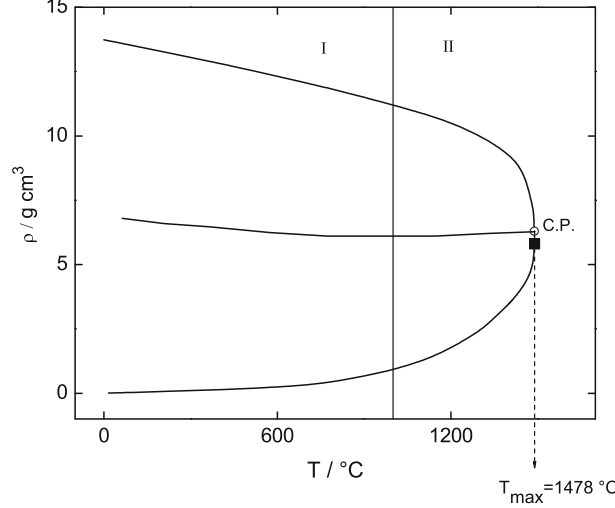


Fig. 2.9. Noncongruent evaporation of liquid mercury causes a pronounced wiggle of the diameter $\rho_d = (\rho_l + \rho_v)/2$ at a temperature of about 1,088° C with a skewing toward higher densities

from which we now evaluate the pressure derivative $\left(\frac{\partial p}{\partial T}\right)_{BC,\rho}$ of the 9.45 g cm^{-3} isochore close to BC-condition permitting variation of the composition ϕ as it occurs during bubble formation and the $\left(\frac{\partial p}{\partial T}\right)_{BC,\phi,\rho}$, keeping now the composition constant (the nonequilibrium fast measurement) from the thermodynamic relation:

$$\left(\frac{\partial p}{\partial T}\right)_{BC,\phi,\rho} - \left(\frac{\partial p}{\partial T}\right)_{BC,\rho} = \left(\frac{\partial \phi}{\partial \rho}\right)_{p,T} \left(\frac{\partial p}{\partial \phi}\right)_{\rho,T} \left(\frac{\partial \rho}{\partial T}\right)_{BC}.$$

The calculated value for this difference is about 1.85 bar K^{-1} .

This value is in very close agreement with the experimentally observed result [26, 32] displayed in Fig. 2.10, which is about 1.8 bar K^{-1} . Another effect that most probably can be explained in terms of “noncongruent” evaporation is the observation of “anomalous” wiggles in the ultrasound velocity c as a function of pressure at constant temperature or alternatively as a function of temperature at constant pressure by Kobayashi et al. [14] at BC- and SC-condition. Figures 2.11 and 2.12 display the effect of isobaric and isothermal “noncongruent” evaporation for the experimentally determined adiabatic compressibility $\beta_S = 1/\rho \cdot c^2$. The p–T coordinates observed by Kobayashi et al., for BC- and SC-conditions are included in the inset of Fig. 2.3 (the open circles and open triangles). They are surprisingly close to the vapor pressure curves found in [26, 32].

In conclusion, we regard the experimental observations presented above as compelling evidence that the features of the liquid mercury evaporation for

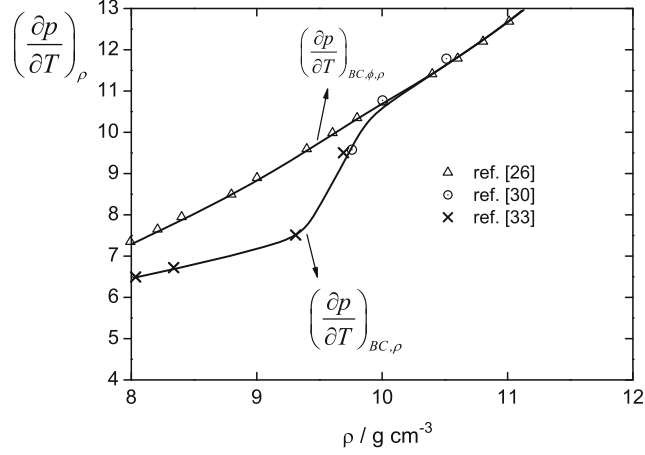


Fig. 2.10. Comparison between the measured isochoric pressure derivatives $(\frac{\partial p}{\partial T})_{BC,\phi,\rho}$ (lower curve) and $(\frac{\partial p}{\partial T})_{BC,\rho}$ (upper curve) asymptotically close to BC. The two curves split into two branches for densities lower than 10.7 g cm^{-3} (see text)

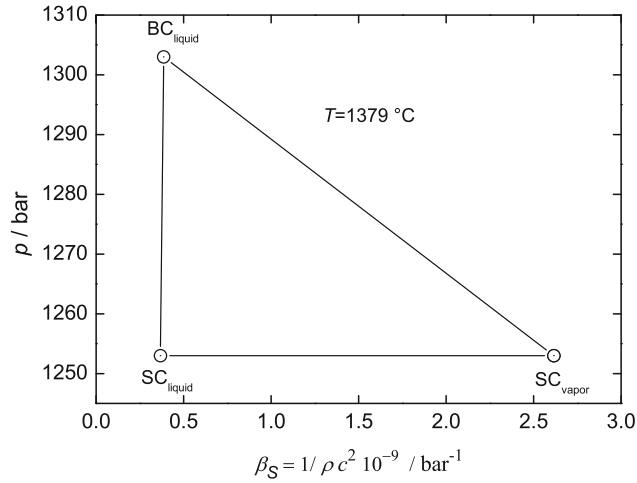


Fig. 2.11. Figure displays the effect of noncongruent evaporation at constant temperature on the adiabatic compressibility $\beta_S = 1/\rho \cdot c^2$ at BC- and SC-conditions

temperatures higher than $1,088^\circ\text{C}$ resemble the “noncongruent-evaporation” discussed by Iosilevskiy and colleagues [37–39]. This phenomenon may play an important role for many scenarios of postulated first-order phase transitions as those of the liquid–vapor transition in the high temperature uranium-oxide system [37], the postulated hypothetical plasma phase transition in giant planets [41], the phase separation in complex dusty plasmas which are

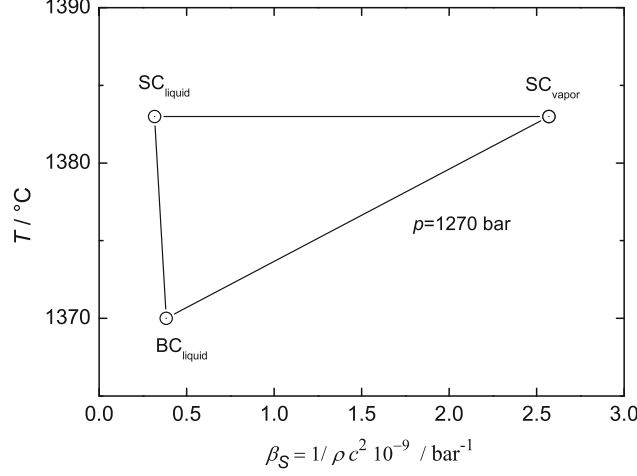


Fig. 2.12. Figure displays the effect of “noncongruent” evaporation at constant pressure on the adiabatic compressibility $\beta_S = 1/\rho \cdot c^2$ at BC- and SC-conditions.

predicted to show gas, fluid, and mixed phases of dust grains [42, 43], the formation of “pasta” structures in compact stars following an exotic first-order-phase transition [44], and the exotic phase transitions in neutron stars [45–47]. To our knowledge, the postulated noncongruence of hypothetical transitions in dense ionized systems at extreme conditions of temperature and pressure has never before been seen experimentally. If mercury is the first example for which the long-sought-for effect has been experimentally studied, it may serve as a model systems for simulating the transitions mentioned above for which the outlook for reliable measurements is unfavorable.

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