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# Turbidity measurements in xenon reanalyzed using the master crossover functions

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The turbidity measurements of Güttinger and Cannell [Phys. Rev. A **24**, 3188 (1981)] in the temperature range  $20 \text{ mK} \leq T - T_c \leq 29.5 \text{ K}$  along the critical isochore of homogeneous xenon are reanalyzed using the master crossover functions for the isothermal compressibility  $\kappa_T$  and the correlation length  $\xi$ , without adjustable parameter. We show that the turbidity data are well represented by the Ornstein-Zernike theory, within 1% precision. This excellent agreement confirms that the Ising-like critical behavior of xenon can be described in conformity with the universal features estimated by the massive renormalization scheme, only knowing the four critical coordinates of the vapor-liquid critical point in the (pressure, temperature, molecular volume) phase surface of the monoatomic xenon particle.

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In their experimental paper [1] devoted to a first unambiguous evaluation of the confluent corrections to scaling for the susceptibility of xenon, Güttinger and Cannell conclude to the needed theoretical progress in order to reduce the number of effectively free parameters, which would be extremely valuable. Now it is well-established [2], especially from the field theory framework [3], that the asymptotic singular properties of the one-component fluids can be described by the classical-to-critical crossover functions of universal features predicted for the complete  $\{d = 3, n = 1\}$  universality class of the three-dimensional (3D) Ising like systems.  $d$  is the space dimension, while  $n$  is the dimension of the order parameter (OP) density. Some recent theoretical improvements [4, 5] have extended the applicability range of the crossover functions far from the critical point, maintaining their fundamental interest in conformity with the accurate Ising-like universal values of the critical (leading and lowest confluent) exponents and (leading and first confluent) amplitude combinations and ratios [6]. Consequently, only the description of the Ising-like preasymptotic domain can be precisely characterized by a limited number of free (system-dependent) parameters. Moreover, this description needs to introduce the physical critical parameters such as the critical temperature  $T_c$ , the critical pressure  $p_c$ , the critical density  $\rho_c$ , etc., and to define a single length scale unit to make any physical properties in a dimensionless form. In that dimensionless scheme with length unit uniqueness [7], the singular behaviors of the selected one-component fluid are then characterized by two leading amplitudes and one-confluent amplitude of the two-term Wegner like expansions valid asymptotically close to the Ising-like critical point. Unfortunately, as a renormalizable theory cannot localize the real critical point, only experiments remain able to estimate the Ising-like nature and the limited number of readily independent amplitudes which characterize the critical point phenomena.

The above irreducible challenge introduces a true difficulty to analyze the experimental data obtained at “finite” distance to the critical point, where undetermined number and undetermined origin of nonuniversal corrections terms can contribute to a Wegner-like expansion [8]. To overcome this difficulty consists in only using the thermodynamic information which is contained in the related experimental localization of the isolated liquid-vapor critical point itself on a phase surface of equation of state  $p = \left(\frac{\partial A}{\partial V}\right)_{T,N}$ , as proposed in Ref. [9, 10].  $A(T, V, N)$  is the total Helmholtz free energy expressed in terms of its three natural variables, i.e., the temperature  $T$ , the total volume  $V$ , and the total number of particles  $N$ . After the normalization of this fluid description per constitutive particle, it is then thermodynamically equivalent to define a limited number (four) of finite critical coordinates of the critical point and to introduce a limited number (five) of asymptotic system-dependent parameters in a renormalizable theory. The term to term identification of the physical and theoretical singular behaviors within the preasymptotic domain closes the critical description by introducing three well-defined master (i.e., constant) numbers for all the one-component fluids. By application of the master crossover functions given in Ref. [11], we are then able to validate the real extension of the Ising-like asymptotic behavior of all the critical fluids, only using four well-defined critical coordinates. In such a procedure, xenon acts as a standard (non-quantum) Ising-like fluid [12, 13]. Accordingly, the analyses of the singular behaviors of physical properties of xenon are of basic interest. Hereafter we provide the results obtained for the case of the light transmission measurements of Güttinger and Cannell [1] on near-critical xenon in the temperature range  $20 \text{ mK} \leq T - T_c \leq 29.5 \text{ K}$ . In the Güttinger and Cannell experiment, the relative determination of the critical temperature and critical density of the fluid cell was locally controlled under the field acceleration due to Earth’s gravity. The related turbidity [ $\tau$ ]

data source used in the present study are given in Table I of Ref. [1], as a function of  $\Delta\tau^* = \frac{T-T_c}{T_c}$ , with  $T_{c,G_C} = (289.790 \pm 0.0005)$  K.

Turbidity of a transparent fluid close to its liquid-gas critical point is most essentially due to Rayleigh light scattering by local density fluctuations. According to Ornstein-Zernike (OZ) [14], the scattered light intensity can be written [15] as :

$$I(q) = \frac{A\kappa_T \sin^2 \Phi}{1 + (q\xi)^2} \quad (1)$$

where  $q$  represents the amplitude in the fluid of the transfer wave vector between the incident and scattered light wave vectors :  $q = \frac{4\pi n}{\lambda_0} \sin \frac{\theta}{2}$ .  $n$  is the refractive index of the fluid, and  $\theta$  is the scattering angle.  $\Phi$  is the angle between the polarization wave vector of the incident beam and the scattering wave vector.  $\kappa_T$  is the isothermal compressibility and  $\xi$  is the correlation length of the long-range fluctuating order parameter density, proportional to the local density difference  $\rho - \rho_c$ . The prefactor  $A$  is given by the following equation

$$\begin{aligned} A &= \frac{\pi^2}{\lambda_0^4} \left[ \rho \left( \frac{\partial(n^2)}{\partial\rho} \right)_T \right]^2 k_B \\ &= \frac{\pi^2}{\lambda_0^4} \left[ \frac{(n^2-1)(n^2+2)}{3} \right]^2 k_B \end{aligned} \quad (2)$$

which accounts for geometrical factors of the scattering configuration, laser optical wavelength in vacuum ( $\lambda_0$ ), light-fluid scattering cross section calculated from the Lorentz-Lorenz approximation of the effective local field, i.e., introducing the related critical value  $LL_c = \frac{M_{\text{mol}}}{\rho_c} \frac{n_c^2-1}{n_c^2+2}$  of the density expansion of the Lorentz-Lorenz function.  $M_{\text{mol}}$  is the molar mass of the fluid and  $k_B$  is the Boltzmann constant. Turbidity corresponds to the integral of Eq. (1) over all the scattering angles and writes [15] :

$$\tau_{OZ} = AT_c \kappa_T (1 + \Delta\tau^*) F(a), \quad (3)$$

where the dimensionless function  $F(a)$  is given by the following equation

$$F(a) = \left( \frac{2a^2 + 2a + 1}{a^3} \right) \ln(1 + 2a) - 2 \left( \frac{1 + a}{a^2} \right) \quad (4)$$

with  $a = 2(k_0\xi)^2$  and  $k_0 = \frac{2\pi n}{\lambda_0}$  ( $k_0$  is the amplitude of the incident light wave vector). All the needed experimental information to perform the OZ-analysis of the turbidity data can be found in the Güttinger and Cannell's paper [1] and references therein. In our present application of Eq. (3) we use the well-controlled values of the critical parameters of xenon which are defined in Table I (see also Refs. [13, 16–18] given in last column of

Parameter		Ref.
$m_{\bar{p}}$	$2.1805 \times 10^{-25}$ kg	
$T_c$	$289.733 \pm 0.015$ K	[13, 16]
$p_c$	$5.84007 \pm 0.00050$ MPa	[13]
$\rho_c$	$1113 \pm 3$ kg m <sup>-3</sup>	"
$\gamma'_c$	$0.1197 \pm 0.0006$ MPa K <sup>-1</sup> .	"
$(\beta_c)^{-1} = k_B T_c$	$4.0002 \times 10^{-21}$ J	"
$\alpha_c = \left( \frac{k_B T_c}{p_c} \right)^{\frac{1}{d}}$	$8.81501 \times 10^{-10}$ m	"
$Y_c = \gamma'_c \frac{T_c}{p_c} - 1$	4.9173	"
$Z_c = \frac{p_c m_{\bar{p}}}{\rho_c k_B T_c}$	0.286017	"
$n_c$	1.1375	[17, 18]
$LL_c$	$10.5271$ cm <sup>3</sup> mole <sup>-1</sup>	"
$A$	$(88.4943 \pm 0.35)$ J <sup>-1</sup> m <sup>-4</sup>	"
$k_0$	$1.12944 \times 10^7$ m <sup>-1</sup>	[1]

Table I: Critical parameters for xenon, with  $M_{\text{mol}} = 0.131313$  g mole<sup>-1</sup> and  $N_A = 6.0224179 \times 10^{23}$  (see text).

Table I). Since our present estimation of the singular behaviors of the isothermal compressibility [ $\kappa_T(\Delta\tau^*)$ ] and the correlation length [ $\xi(\Delta\tau^*)$ ] only use these critical parameters (see the detail below), the only differences with the Güttinger and Cannell's analysis originate from the critical coordinates of xenon.

The four values of the critical temperature  $T_c$ , the critical pressure  $p_c$ , the critical density  $\rho_c$ , and the common critical slope  $\gamma'_c = \left[ \left( \frac{\partial p}{\partial T} \right)_{\rho=\rho_c} = \frac{dp_{\text{sat}}}{dT} \right]_{T \rightarrow T_c^\pm}$  of the critical isochore ( $T \rightarrow T_c^+$ ) and the saturation pressure  $p_{\text{sat}}(T)$  curve ( $T \rightarrow T_c^-$ ) which are reported in Table I localize the vapor-liquid critical point on the  $p, v_{\bar{p}} = \frac{m_{\bar{p}}}{\rho}, T$  phase surface of xenon ( $v_{\bar{p}}$  is the xenon particle volume,  $\rho$  is the mass density and  $m_{\bar{p}}$  is the xenon atom mass; subscript  $\bar{p}$  refers to a particle quantity). From the critical coordinates we have then calculated the corresponding values of i) the energy unit  $(\beta_c)^{-1} = k_B T_c$ , ii) the length unit  $\alpha_c = \left( \frac{k_B T_c}{p_c} \right)^{\frac{1}{d}}$ , iii) the non-dimensional scale factor  $Y_c = \gamma'_c \frac{T_c}{p_c} - 1$  of the dimensionless thermal field  $\Delta\tau^*$ , and iv) the non-dimensional scale factor  $Z_c = \frac{p_c m_{\bar{p}}}{\rho_c k_B T_c}$  of the dimensionless ordering field conjugated of dimensionless order parameter density [11, 13].

We have then calculated the physical singular behavior of  $\xi(\Delta\tau^*)$  and  $\kappa_T(\Delta\tau^*)$  as proposed in Ref. [11], by using the following equations where quantum effects [19, 20] are neglected

$$\left( \frac{1}{\alpha_c} \right) \times \xi_{\text{cal}}(\Delta\tau^*) = \ell^*(T^*) = \frac{1}{Z_{\xi}^{\{1f\}} [\ell_{th}(t)]^{-1}} \quad (5)$$

$$(Z_c p_c) \times \kappa_{T,\text{cal}}(\Delta\tau^*) = \chi^*(T^*) = \frac{1}{Z_{\chi}^{\{1f\}} [\chi_{th}(t)]^{-1}} \quad (6)$$

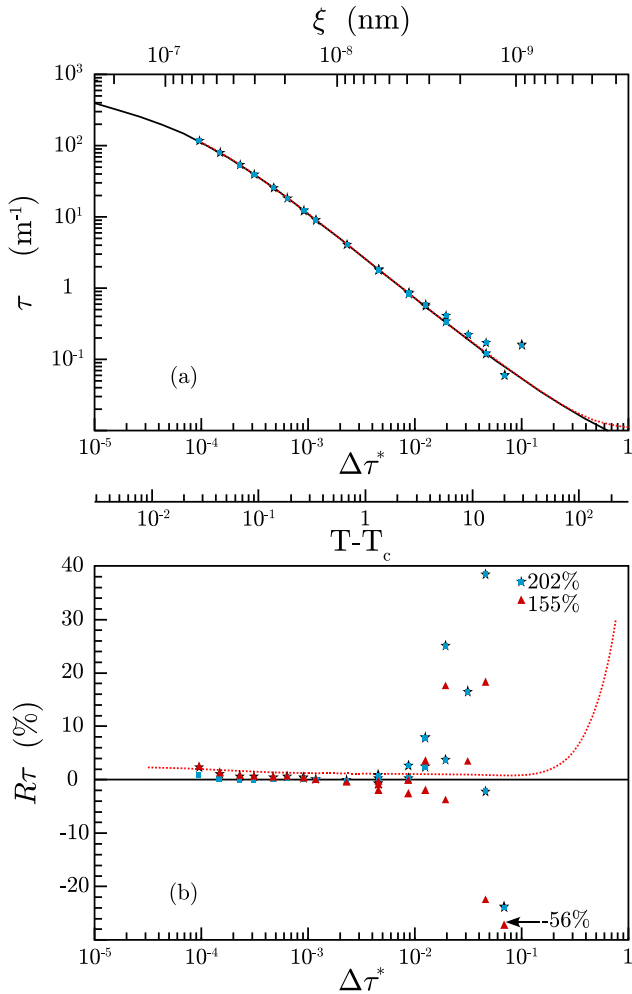


Figure 1: (a) log-log plot of the turbidity  $\tau$  (expressed in  $\text{m}^{-1}$ ) of xenon as a function of  $\Delta\tau^*$  along the critical isochore. Full (black) stars: from light scattering measurements of Güttinger and Cannell [1]. Continuous (black) curve: from Eq. (3). Dotted (red) curve: from Ref. [1]. Upper horizontal axis:  $\xi$  (expressed in nm). Lower horizontal axis:  $T - T_c$  (expressed in K), calculated using with  $T_{c,\text{GC}} = 289.79 \text{ K}$  ( $16.64^\circ\text{C}$ ) [1]. (b) log-lin plot of the related residuals  $R\tau = 100 \left[ \frac{\tau}{\tau_{\text{OZ}}} - 1 \right]$  (expressed in %) from reference to the calculated turbidity  $\tau_{\text{OZ}}$  of Eq. (3). Symbols and curve: same as in (a); full (blue) squares: with a  $T_{c,\text{GC}}$ -change of  $0.5 \text{ mK}$  (see text); full (red) triangles: with a  $\tau(T_{\text{or}})$ -change of  $-0.00025 \text{ cm}^{-1}$  (see text).

with  $t = \Theta^{\{1f\}} \mathcal{T}^* = \Theta^{\{1f\}} Y_c \Delta\tau^* = \vartheta \Delta\tau^*$ , i.e.,  $\mathcal{T}^* = Y_c \Delta\tau^*$  and  $\vartheta = \Theta^{\{1f\}} Y_c$ . In Eqs. (5) and (6), the master crossover functions  $\ell^*(\mathcal{T}^*)$  and  $\chi^*(\mathcal{T}^*)$  correspond to the modifications of the theoretical crossover functions  $[\ell_{th}(t)]^{-1}$  and  $[\chi_{th}(t)]^{-1}$  defined in Refs. [4, 5], introducing the three scale factors  $\Theta^{\{1f\}} = 4.288 \cdot 10^{-3}$ ,  $\Psi^{\{1f\}} = 1.74 \cdot 10^{-4}$  and  $\mathbb{L}_f = 25.6988$ , which characterize the one-component fluid subclass. Accordingly, the master prefactor values appearing in Eqs. (5) and (6) are  $\mathbb{Z}_\xi^{\{1f\}} \equiv \mathbb{L}_f = 25.6988$  and  $\mathbb{Z}_\chi^{\{1f\}} = (\mathbb{L}_f)^d (\Psi^{\{1f\}})^2 =$

$1950.70$ . The corresponding singular behaviors of  $\xi(\Delta\tau^*)$  and  $\kappa_T(\Delta\tau^*)$  for xenon are then in conformity with the universal features of the three-dimensional Ising-like universality class. The related values of the leading amplitudes of the Ising-like power law terms are  $\xi_0^+ = 0.184531 \text{ nm}$  and  $\Gamma^+ = 0.0578238$ , respectively (in standard notations).

The Güttinger and Cannell data points  $\tau_{\text{expt}}$  of xenon turbidity are plotted (full black stars) in Fig. 1(a) as a function of  $\Delta\tau^*$  in log-log scale. Our theoretical estimation of  $\tau_{\text{OZ}}$  using Eqs. (3) to (6) and parameters of Table I corresponds to the continuous black curve in Fig. 1(a), which appears in agreement with the experimental results. The Ornstein-Zernike fitting analysis of xenon turbidity initially performed by Güttinger and Cannell in Ref. [1] is also given as the dotted red curve in Fig. 1(a). The corresponding residuals (expressed in %) are given in the log-lin plot of Fig. 1(b). In this Fig. 1(b), we note that our estimation of  $\tau_{\text{OZ}}$  without adjustable parameter is in excellent agreement with the experimental measurements (full black stars), as well as the initial Ornstein-Zernike fitting analysis (dashed red curve) of Güttinger and Cannell. In order to illustrate the effect of the uncertainty ( $\simeq 0.5 \text{ mK}$ , [1]) in the experimental determination of  $T_{c,\text{GC}}$  approaching the critical temperature, the full blue squares in Fig. 1(b) correspond to the related behaviors of the residuals due to a  $T_{c,\text{GC}}$ -change of  $0.5 \text{ mK}$ . Similarly, in this asymptotic domain where  $T \rightarrow T_c$ , the very small increase of the residuals with the initial fitting analysis of Güttinger and Cannell, which are illustrated by the dotted red curve in Fig. 1(b), is mainly due to the small difference in the respective values of the leading critical exponents  $\nu$  and  $\gamma$  of  $\xi$  and  $\kappa_T$ , respectively. In the temperature range  $T - T_c \gtrsim 5 \text{ K}$  of Fig. 1(b), the increase of the residuals is due to the significant decrease of the xenon turbidity when  $T - T_c$  increases, which needs to have a very precise calibration of the turbidity measurements at these large temperature distance to  $T_c$ . To illustrate this latter remark, we have reported the new residuals with the experimental data points (ful red triangles) that are obtained after a change of  $-0.00025 \text{ cm}^{-1}$  in the initial calibration value [ $\tau(T_{\text{or}}) = 0.041 \text{ cm}^{-1}$ , see Ref. [1]] of the xenon turbidity at the reference temperature  $T_{\text{or}}$ . In the temperature range  $T - T_c \gtrsim 5 \text{ K}$ , accurate measurements of xenon turbidity then needs to use preferably a xenon sample cell of optical path larger than a few centimeters. Nevertheless, we believe that our above Ornstein-Zernike approach of xenon turbidity with Eq. (3), incorporating the  $\xi$  and  $\kappa_T$  values obtained from theoretical Eqs. (5) and (6) without adjustable parameters, is adequate in the  $T - T_c$  range covered by the Güttinger and Cannell measurements.

For the singular behavior of the correlation length as a function of  $\Delta\tau^*$ , we have also reported in the log-lin plots of Figs. 2(a) and (b) the residuals  $R\xi = 100 \left( \frac{\xi}{\xi_{\text{cal}}} - 1 \right)$

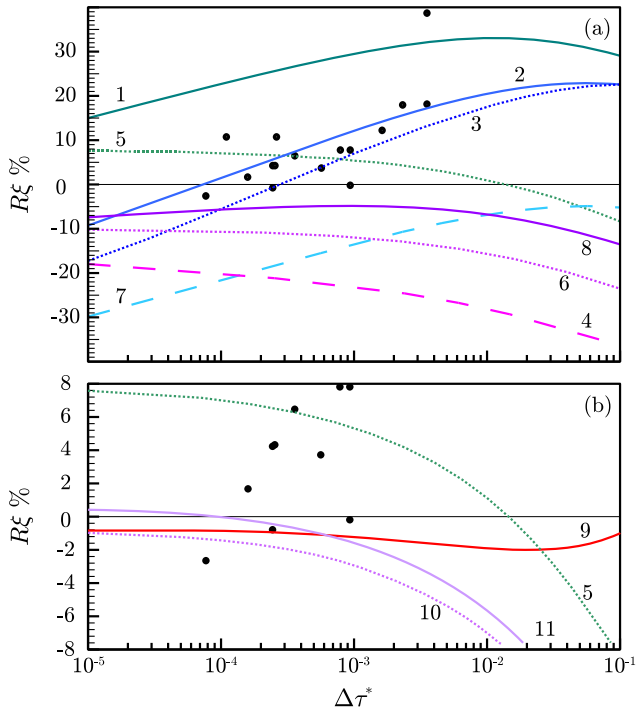


Figure 2: log-lin scale: residuals  $R\xi = 100 \left( \frac{\xi}{\xi_{\text{cal}}} - 1 \right)$  (expressed in %) for the values of the xenon correlation length  $\xi$  as a function of  $\Delta\tau^*$ , from reference to the calculated value  $\xi_{\text{cal}}$  using Eqs. (5). (a) Full black circles: experimental data of Ref [22]. Curves labeled 1 to 8: pure power law  $\xi = \xi_0^+ (\Delta\tau^*)^{-\nu}$  with adjustable exponent and amplitude values given in lines #1 to #8 of Table II, respectively. (b): Curves labeled 5, 9 to 11: two-term power law  $\xi = \xi_0^+ (\Delta\tau^*)^{-\nu} \left[ 1 + a_\xi (\Delta\tau^*)^\Delta \right]$  (red line) and equivalent pure power law  $\xi = \xi_0^+ (\Delta\tau^*)^{-\nu}$  with Ising-like exponent values fixed (see lines #5 and #9 to #11 of Table II, respectively).

#	$\nu$	$\xi_0^+$ (nm)	$a_\xi^{1,+} (\Delta\tau^*)^\Delta$	Ref.
1	0.60	0.302	n.u.	[21]
2	$0.58 \pm 0.05$	$0.3 \pm 0.01$	n.u.	[22]
3	$0.57 \pm 0.03$	0.307	n.u.	[23]
4	$0.64 \pm 0.02$	$0.136 \pm 0.006$	n.u.	[24]
5	0.63	0.2	n.u.	[22, 25]
6	0.63	0.167	n.u.	[25]
7	0.58	0.232	n.u.	[25]
8	0.62	0.193	n.u.	[26]
9	0.63	$0.184 \pm 0.009$	$0.55 (\Delta\tau^*)^{0.5}$	[1]
10	0.63	$0.184 \pm 0.009$	n.u.	[1, 27]
11	0.63	$0.1866 \pm 0.0010$	n.u.	[1, 16, 28]

Table II: Literature exponent and amplitude values of the power law  $\xi = \xi_0^+ (\Delta\tau^*)^{-\nu} \left[ 1 + a_\xi^{1,+} (\Delta\tau^*)^\Delta \right]$  for the xenon correlation length along the critical isochore (n.u.: not used). The corresponding leading values used in Eq. (5) are  $\nu = 0.6303875$  and  $\xi_0^+ = 0.184531$  nm

(expressed in %) of  $\xi$ -data published in the literature [21–28], from reference to  $\xi_{\text{cal}}$  here calculated using Eq. (5). In part (a), we have illustrated by the curves labeled 1 to 8 the dispersion (larger than 10%) of the indirect fitting analysis with a pure power law  $\xi = \xi_0^+ (\Delta\tau^*)^{-\nu}$  of the results provided by using (static and dynamic) Rayleigh-Brillouin light scattering methods, as reported in lines labeled #1 to #8 of Table II. In addition the residuals with the data measurements performed by the full red circles. This Fig. 2(a) gives clear evidence that the highly correlated values of  $\nu$  and  $\xi_0^+$  are not obtained with the required precision [29] from all these optical measurements performed at finite distance to  $T_c$ , which include the first fitting analysis [26] of the correlation length measurements made by using the precise differential technique and apparatus of Güttinger and Cannell. In part (b), the continuous red line labeled 9 corresponds to the residuals (then lowered at the 1%-scale) with the Güttinger and Cannell fitting analysis of their turbidity data that uses the two-terms power law  $\xi = \xi_0^+ (\Delta\tau^*)^{-\nu} \left[ 1 + a_\xi^{1,+} (\Delta\tau^*)^\Delta \right]$  with fixed values of the exponents (see the line labeled #9 of Table II). In addition, we have also given the residuals with two referring pure power laws (see the lines labeled #10 and #11 of Table II). Such pure power laws are used in the fitting analyses of the shear viscosity [27], the thermoacoustic boundary layers [16], and the bulk viscosity [28] of xenon near the critical point, i.e., a correlation length fit which covers the complete temperature range  $5 \times 10^{-7} \leq \Delta\tau^* \leq 10^{-1}$ . These applications then extend significantly the Güttinger and Cannell temperature range, especially approaching the critical temperature by two supplementary decades. Nevertheless, in spite of the questionable justification of the universal features related to the lowest order of the Ising-like Wegner expansions at large distance from  $T_c$ , we note a satisfactory agreement (of the order of the experimental uncertainty of 10%) in the reduced temperature range  $7 \times 10^{-5} \leq \Delta\tau^* \leq 2 \times 10^{-2}$  covered by the Güttinger and Cannell experiment.

A complementary analysis of the  $\kappa_T (\Delta\tau^*)$  measurements can be found in Ref. [13]. Here, through the amplitude-exponent values of Ref. [1, 12, 13, 30–34] reported in lines labeled #1 and #8 of Table III, we only underline the excellent agreement between our present leading power law (line labeled #6) of the isothermal compressibility calculated by Eq. (6) and the related leading power laws published in the literature (see Refs. in last column), when the Ising value of the exponent covers the small range  $1.230 \leq \gamma \leq 1.242$ . It is then interesting to note that the scaled forms of the equation of state [35, 36] can be controlled to provide asymptotic singular behaviors of the fluid properties which follow as close as possible the Ising-like singular behaviors calculated using the master crossover functions of the fluid subclass [13].

#	$\gamma$	$\Gamma^+$	Ref.
1	1.241	$0.0577 \pm 0.001$	[1]
2	1.24194	$0.057 \pm 0.001$	[1, 12]
3	1.2396	$0.0587 \pm 0.0040$	[1, 30]
4	1.239	n.a.	[1, 31]
5	1.240	0.0594	[1, 32]
6	1.2395935	0.0578238	[1, 13]
7	1.23	0.056	[33]
8	1.24	$0.058 \pm 0.002$	[34]
9	1.24	0.5779	[35, 36]

Table III: Amplitude-exponent results of the leading power law term of the isothermal compressibility data along the critical isochore of xenon. Lines #1 and #6: from turbidity measurements of Güttinger and Cannell [1]. Lines #7 and #8: from Fraunhofer optical measurements of density profiles of [33]. n.a.: non available. Line #6 corresponds to the leading values used in Eq. (6). Line #9 corresponds to the leading values calculated using the restricted cubic model of the scaled equation of state [35, 36].

As a conclusion, only using the four critical coordinates of the xenon critical point to calculate the xenon parameters needed by the master crossover functions for isothermal compressibility  $\kappa_T$  and correlation length  $\xi$ , we have estimated the singular behavior of xenon turbidity in agreement with the Güttinger and Cannell data, in the temperature range  $20 \text{ mK} \leq T - T_c \leq 29.5 \text{ K}$ , along the critical isochore.

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