COMMENTS

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Comment on "Equation of state and phase diagrams for dense multi-ionic mixture plasmas"

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An equation of state for dense fluid binary-ionic mixture (BIM) plasmas was derived recently by Ogata, Iyetomi, Ichimaru, and Van Horn [Phys. Rev. E 48, 1344 (1993)] from extensive Monte Carlo simulations data. They emphasized in particular the significance of finding *negative* deviation from the linearmixing approximation. In this Comment, I show that the important features of their results are the outcome of their particular choice for the equation of state of the one-component plasma, which has relatively large error bars that have not been properly taken into account and are not necessarily borne out by their data for the binary mixtures. I repeated their analysis of their same data for the bindary mixtures, but used another, comparable, one-component plasma fluid equation of state to find always *positive* deviations from linear mixing, in contradiction also with the sum-rule analysis of their free energy with respect to the zero separation of the screening potentials.

PACS number(s): 52.25.Kn, 05.70.Ce, 64.10.+h

I. INTRODUCTION

Classical plasmas, namely, positive ions in a uniform neutralizing background of electrons, make important basic models for dense stellar materials, and provide a most useful reference system in condensed-matter physics [1]. In a recent large effort by Ogata, Iyetomi, Ichimaru, and Van Horn [2] (OIIVH), the equation of state and phase diagrams for dense multi-ionic mixture plasmas were calculated on the basis of extensive Monte Carlo (MC) simulations. A major part of this work by OIIVH is the evaluation of the equation of state for dense binary-ionic mixture (BIM) plasmas, in the *fluid* phase, with charge ratios $R_Z \leq 5$, on the basis of internal energies calculated by the MC method. The result was represented in the form of deviation from the so called "linear-mixing" approximation, which is calculated by using a given fit to the equation of state of the onecomponent plasma. It was first checked for consistency with the zero-separation sum rule for the screening potential, and then extensively applied for calculating phase diagrams for mixtures. As stressed by OIIVH, their results are significantly different from other calculations in the literature, and crucially depend on finding negative deviation from the linear-mixing approximation, for relatively small concentrations of the larger charges.

In this Comment I show that the most important features of the BIM equation of state derived by OIIVH are not necessarily borne out by the MC data for the BIM internal energies themselves, but are the outcome of the particular fit used by OIIVH for the equation of state of the one-component plasma (OCP). I repeat the analysis of their same data, using their method but with another fit for the OCP energies, due to DeWitt, Slattery, and Stringfellow [3] (DWSS), and reach significantly different conclusions from those arrived at by OIIVH. A variant of the analytic variational hard-sphere model for the free energy of multi-ionic plasmas is presented, which is in good agreement with the data of OIIVH as reduced by the DWSS equation of state for the OCP.

II. DEVIATIONS FROM LINEAR MIXING

The system under consideration is a binary-ionic mixture consisting of N_i particles of species *i* with charge $Z_i e$ (i=1,2), in a uniform background of neutralizing electrons of volume *V*, at temperature *T*. The total number of the particles is $N=N_1+N_2$, it is assumed that $Z_2 > Z_1$, and the charge ratio $R_Z = Z_2/Z_1 > 1$, and the molar fractions x_1 and $x_2=1-x_1\equiv x$ are thus defined. The thermodynamic state of such a mixture is specified by the Coulomb coupling parameter

$$\Gamma_e = \frac{e^2}{a_e k_B T} , \qquad (1)$$

where

$$a_e = \left[\frac{3}{4\pi n_e}\right]^{1/3} \tag{2}$$

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$$a_i = a_e Z_i^{1/3}, \quad \Gamma_i = \Gamma_e Z_i^{5/3}, \quad i = 1, 2$$
, (3)

so that $\Gamma_2 = \Gamma_1 R_Z^{5/3}$.

The method used by OIIVH is the following. The bare MC data are given in the form of the excess internal energy normalized by Nk_BT , $u_{ex}^{BIM}(R_Z, x, \Gamma_1)$, for different values of R_Z , x, and Γ_1 . Consider first the result of the linear mixing (LM) "rule" [4],

$$u_{\rm LM}(R_Z, x, \Gamma_1) = (1-x) u_{\rm ex}^{\rm OCP}(\Gamma_1) + x u_{\rm ex}^{\rm OCP}(\Gamma_2 = \Gamma_1 R_Z^{5/3}) .$$
(4)

 $u_{ex}^{OCP}(\Gamma)$ represents the internal energy for the onecomponent plasma, and u_{LM} is expected to be a good approximation for u_{ex}^{BIM} . OIIVH perform the data analysis for u_{ex}^{BIM} by considering the difference

$$\Delta u_{\rm ex}^{\rm BIM} = u_{\rm ex}^{\rm BIM} - u_{\rm LM} \tag{5}$$

where $u_{ex}^{OCP}(\Gamma)$ is represented by a fixed function $u_0(\Gamma)$, which is obtained separately from a fit of only the OCP energies. The fitting function for the BIM data is thus required to yield the corresponding OCP limit, i.e., $\Delta u_{ex}^{BIM} = 0$ for $R_Z = 1$ or x = 0 or 1. After fitting Δu_{ex}^{BIM} by an appropriate function they integrate it analytically,

$$\Delta f_{\text{ex}}^{\text{BIM}}(R_Z, x, \Gamma_1 \ge 1) = \int_1^{\Gamma_1} \frac{d\Gamma'}{\Gamma'} \Delta u_{\text{ex}}^{\text{BIM}}(R_Z, x, \Gamma') + \Delta f_{\text{ex}}^{\text{BIM}}(R_Z, x, \Gamma_1 = 1) , \qquad (6)$$

in order to obtain the corresponding deviation of the excess free energy normalized by Nk_BT , f_{ex}^{BIM} , from its linear-mixing value, namely, $\Delta f_{ex}^{BIM} = f_{ex}^{BIM} - f_{LM}$. The excess free energies f_{ex}^{BIM} and f_{LM} are thus obtained by the corresponding integration of u_{ex}^{BIM} and u_{LM} . In particular,

$$f_{\rm LM}(R_Z, x, \Gamma_1) = (1 - x) f_{\rm ex}^{\rm OCP}(\Gamma_1) + x f_{\rm ex}^{\rm OCP}(\Gamma_2 = \Gamma_1 R_Z^{5/3}) .$$
(7)

The values of these functions at $\Gamma_1 = 1$ are obtained by the hypernetted chain (HNC) approximation which is of reliable accuracy [5] for $\Gamma_1 \leq 1$.

reliable accuracy [5] for $\Gamma_1 \leq 1$. The resulting Δf_{ex}^{BIM} is eventually used by OIIVH for the analysis of the phase diagrams, after being checked through an exact "zero-separation" relation. The OCP screening potential at the origin equals the excess free energy difference before and after a nuclear fusion reaction between two identical ions which form a single doublecharge ion [6]. In terms of the quantities defined above it takes the form [2]

$$h_{0} \equiv \frac{H(r=0)}{k_{B}T}$$

=2 $f_{ex}^{OCP}(\Gamma) - f_{ex}^{OCP}(2^{5/3}\Gamma)$
 $-\left[\frac{\partial}{\partial x}\Delta f_{ex}^{BIM}(R_{Z}=2,x,\Gamma_{1}=\Gamma)\right]_{x\to 0},$ (8)

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where the derivative of Δf_{ex}^{BIM} gives the deviation from the LM approximation

$$h_0^{\rm LM} = 2f_{\rm ex}^{\rm OCP}(\Gamma) - f_{\rm ex}^{\rm OCP}(2^{5/3}\Gamma)$$
(9)

of Jancovici [6]. The results thus obtained for h_0 from Eq. (8) are tested for consistency with the corresponding result as obtained from an extrapolation towards r = 0 of the MC data for the pair radial distribution function g(r) of the OCP, using the relation $H(r)/k_BT = \ln g(r) + Z_1^2 e^2/r$.

III. REDUCTION OF THE MC DATA FOR THE BIM: DEPENDENCE ON THE OCP EQUATION OF STATE

OIIVH make the choice [2]

$$u_{ex}^{OCP}(\Gamma \ge 1) = u_0^{OIVH}(\Gamma)$$

= -0.898 004\Gamma + 0.967 86\Gamma^{0.25}
+ 0.220 703\Gamma^{-0.25} - 0.860 97 (10)

and indeed find that the LM rule reproduces the u_{ex}^{BIM} data with relative errors less than 1% for all cases. Correspondingly [2],

$$f_{ex}^{OCP}(\Gamma \ge 1) = f_0^{OIVH}(\Gamma)$$

= -0.898004\Gamma + 3.87144\Gamma^{0.25}
-0.882812\Gamma^{-0.25}
-0.86097\ln\Gamma - 2.52692. (11)

The results for h_0 thus obtained by OIIVH from Eq. (8) agree well with the values given by Ogata, Iyetomi, and Ichimaru [7] (OII) by extrapolating g(r) data, in the form

$$h_0^{\text{OII}} = 1.148 - 0.0094 \ln \Gamma - 0.000 \, 17 (\ln \Gamma)^2$$
 (12)

As stressed by OIIVH, this agreement for h_0 , as well as important features of their phase diagrams, crucially depend on the negative sign and on the relatively large magnitude of their derivative of Δf_{ex}^{BIM} at $x \rightarrow 0$. The extrapolation method of OII was recently criticized [8], however. A reanalysis [8] of the OII data led to values for h_0 in good agreement with the analysis of Alastuey and Jancovici [6], and well represented by h_0^{LM} without the derivative correction. This situation, in view of the OIIVH agreement with OII, thus requires a closer look at the derivation of Δf_{ex}^{BIM} .

It is clear that even though the magnitude of the fitting function, Δu_{ex}^{BIM} , over the space of its independent variables is controlled by the data points which it fits, its derivatives strongly depend on the functional form of the fit, which is crucially affected by the choice of $u_0(\Gamma)$. To demonstrate the point, I repeated the whole procedure as performed by OIIVH and outlined above, but using the following DWSS representation [3]:

$$u_{\text{ex}}^{\text{OCP}}(\Gamma \ge 1) = u_0^{\text{DWSS}}(\Gamma)$$

= -0.8992\Gamma + 0.596\Gamma^{0.3253} - 0.268 (13)

for the MC data for the OCP. This is the latest fit of

DWSS for their long simulation runs [3]. Correspondingly [3],

$$f_{ex}^{OCP}(\Gamma \ge 1) = f_0^{DWSS}(\Gamma)$$

= -0.8992\Gamma + 1.8322\Gamma^{0.3253}
-0.268 \ln\Gamma - 1.3693. (14)

The OIIVH data for u_{ex}^{BIM} as reduced via the linearmixing approximation are presented in Table I, where $\Delta u_{ex}^{\text{BIM}}$ as a function of x for different values of R_Z and Γ_1 is compared for the two alternative choices for u_0 . Contrary to the OIIVH results, $\Delta u_{ex}^{\text{BIM},\text{OIIVH}}$, the function $\Delta u_{ex}^{\text{BIM}}$ as obtained from $u_0^{\text{DWSS}}, \Delta u_{ex}^{\text{BIM},\text{DWSS}}$, is always positive and its slope near x = 0 is also always positive. Also shown are the results of an analytic variational model (to be described below), $\Delta u_{ex}^{\text{BIM}, \text{var}}$ which agree well with $\Delta u_{ex}^{\text{BIM},\text{DWSS}}$. The particularly good agreement of $\Delta u_{ex}^{\text{BIM},\text{DWSS}}$ with the model for $x \le 0.1$ enables us to use the model (in the role of a judicious fit) to obtain reliable estimates of the derivative which corrects the h_0^{LM} . The various results for h_0/Γ are compared in Fig. 1. Note in particular that on the scale of Fig. 1 the LM cannot tell the difference between the u_0^{DWSS} and u_0^{OIIVH} , and only one is presented. The situation as described in Table I

TABLE I. Deviations from linear mixing: MC results for the BIM (from Ref. [2]) as reduced by Eq. (5) using the OIIVH and the DWSS fits for the OCP, $\Delta u_{ex}^{BIM,OIIVH}$ and $\Delta u_{ex}^{BIM,DWSS}$ (with same error bars from Ref. [2]) compared with the results of the variational model, $\Delta u_{ex}^{BIM,var}$. Note the excellent agreement between $\Delta u_{ex}^{BIM,DWSS}$ and $\Delta u_{ex}^{BIM,var}$ for $x \le 0.1$. See the text. The data are from Table I in Ref. [2] except for the $R_Z = 2$ point which is from Table IV in Ref. [2].

R_{Z}	x	Γ_1	$\Delta u_{ex}^{BIM,OIIVH}$	\pm Error	$\Delta u_{ex}^{BIM, DWSS}$	$\Delta u_{ex}^{BIM, var}$
3	0.01	10	-0.004	±0.001	0.000	0.001
3	0.05	10	0.000	± 0.001	0.004	0.004
3	0.10	10	0.004	± 0.001	0.007	0.006
3	0.20	10	0.009	± 0.001	0.012	0.011
3	0.50	10	0.013	± 0.002	0.013	0.014
3	0.01	20	-0.002	± 0.001	0.002	0.001
3	0.05	20	-0.001	± 0.001	0.002	0.003
3	0.10	20	0.004	± 0.001	0.007	0.006
3	0.20	20	0.003	± 0.002	0.005	0.010
3	0.50	20	0.008	± 0.002	0.007	0.013
5	0.01	5	0.001	± 0.001	0.001	0.001
5	0.05	5	0.007	± 0.001	0.009	0.009
5	0.10	5	0.015	± 0.001	0.017	0.016
5	0.20	5	0.023	± 0.001	0.024	0.025
5	0.50	5	0.027	± 0.002	0.025	0.030
5	0.01	10	-0.002	±0.001	0.002	0.002
5	0.05	10	0.004	± 0.001	0.008	0.009
5	0.10	10	0.011	± 0.001	0.015	0.015
5	0.20	10	0.015	± 0.002	0.018	0.024
5	0.50	10	0.015	± 0.003	0.016	0.028
2	0.50	20	0.007	± 0.001	0.006	0.006



FIG. 1. Consistency with respect to the zero-separation sum rule. $h_0 = H(0)/(k_BT/\Gamma)$. OII [Eq. (12)], LM [Eqs. (9) and (14)], LM+VAR [Eqs. (8), (11), and (18)], and OIIVH [Eq. (8) and the fit of [2]]. LM+VAR represents the present results for the deviations from linear mixing, while OIIVH are the corresponding results from Ref. [2] (see the text).

and Fig. 1 can be summarized as follows.

Given the MC data of OIIVH and of OII for the BIM energies and radial distribution functions, they lead to different conclusions depending on the analysis of these data: (1) Negative and relatively large deviations from the linear-mixing approximation for small x, by using u_0^{OIIVH} , in agreement with the OII extrapolation result h_0^{OII} , vs (2) positive and relatively small deviations from the linearmixing approximation for small x, by using u_0^{DWSS} , in agreement with the Jancovici-Alastuey extrapolation and the LM result h_0^{LM} .

These are numerically relatively small differences, but they lead to important qualitative implications [1,2,8], e.g., for the phase diagrams, for the enhancement factors for nuclear reaction rates in massive accreting white dwarfs, and in fundamental liquid state theory. The OII extrapolation has already been criticized [8], and one issue at hand is the differences between u_{ex}^{OCP} and f_{ex}^{OCP} as obtained from u_0^{OIVH} and from u_0^{DWSS} . These are compared in Fig. 2, where it should be remembered that all these functions are of the order of Γ . The accuracy of the DWSS and OIIVH fits to the MC data (Table 3 in Ref. [3]) is about the same with standard deviation



FIG. 2. Comparison of different fits for the OCP excess energies: $Du(DWSS) = u_0^{OIIVH} - u_0^{DWSS}, Df(DWSS) = f_0^{OIIVH} - f_0^{DWSS}$, and $Du(SDD) = u_0^{OIIVH} - u_0^{DDD}$. See the text.

 $\sigma \sim \pm 0.003$. Yet the OIIVH representation cannot be accepted as an equally good fit to the DWSS data in the region around $\Gamma = 10$ which is particularly relevant for Table I. The fit (10) used by OIIVH was derived by Ogata and Ichimaru [9] partly on the basis of the older data of Slattery, Doolen, and DeWitt [10] (SDD). Indeed, note (in Fig. 2) that in the region $1 < \Gamma < 50$ the SDD fit [10]

$$u_0^{\text{SDD}}(\Gamma) = -0.897744\Gamma + 0.95043\Gamma^{0.25} + 0.18956\Gamma^{-0.25} - 0.81487$$
(15)

is in excellent agreement with the fit (10) used by OIIVH. The DWSS fit [3] improves on the older SDD fit [10].

A second issue concerning the data reduction by OIIVH is their methodology for obtaining Δf_{ex}^{BIM} from which they also calculate quite peculiar phase diagrams. With inherent uncertainties of about ± 0.001 for the MC energies, then in view of the deviations of u_0^{OIIVH} from u_{ex}^{MC} , possible errors for u_{LM}^{OIIV} are about ± 0.004 . This relatively large uncertainty in the fits makes the whole issue of negative deviations from linear mixing rather spurious: With errors of about ± 0.004 for u_{LM} OIIVH are still able to cite errors of about ± 0.001 for $\Delta u_{ex}^{BIM,OIIVH}$ in their Fig. 2 and for the accuracy of their fit for $\Delta f_{ex}^{BIM,OIIVH}$. From this fit they subsequently obtain a large negative result for the derivative at x = 0, which turns out to be precisely what is needed in order that the result from Eq. (8) compare well with h_0^{OII} . With correct error bars in their Fig. 2, the negative deviations of OIIVH from linear mixing fade away.

Similar possible errors apply also to u_{LM}^{DWSS} . Note, however, that other calculations were reported [11] in which both the relevant OCP and BIM energies were obtained from extra-long MC samplings, always finding positive deviations from linear mixing.

IV. MODEL FOR THE DEVIATIONS FROM LINEAR MIXING

As mentioned above (Table I), the MC data of OIIVH for the BIM, $\Delta u_{ex}^{BIM, DWSS}$, as reduced by the DWSS equation of state for the OCP, agree well with an analytic model, especially for small concentrations of the larger charge ($x \le 0.1$). The model thus provides a convenient representation for the equation of state of the mixture, and is expected to be more reliable than a plain fit for purposes of interpolations and extrapolations of the data. The model is based on the idea of considering the well known variational hard-sphere model [12,13] separately for the mixture and for the one-component systems with Γ_1 and Γ_2 , and to calculate the deviation from linear mixing entirely within the model. The expectation is that inaccuracies of the model will cancel out between the results for the mixture and for linear mixing, to yield accurate values for Δu_{ex}^{BIM} .

The model excess free energy (in Nk_BT units) is written in the form

$$f_{ex}(\eta,\lambda,\Gamma_e,Z_1,Z_2,x) = f_{ex}^{PYV}(\eta,\lambda,x) + u_{PY}(\eta,\lambda,\Gamma_e,Z_1,Z_2,x) \quad (16)$$

where f_{ex}^{PYV} is the excess free energy for the hard-sphere binary mixture as obtained from the Percus-Yevick "virial" (PYV) equation of state [14], and u_{PY} is the excess energy obtained by the standard expression but using the PY radial distribution functions [14] in the energy integral, and can be expressed analytically. The variational parameters are the total hard-sphere packing fraction η and the ratio between the two hard-sphere diameters, λ . The optimal values of the parameters η_0 and λ_0 are obtained by minimizing f_{ex} . The reason for choosing the PY virial entropy is that for the OCP it provides the paradigm [13] for the functional form used to fit the MC data [see Eqs. (10) and (15) above], and yields [13] the following asymptotic large Γ expansion:

$$u_{\rm PY}(\eta_0(\Gamma),\lambda,\Gamma,Z_1=1,Z_2,x=0) = -0.9\Gamma + 0.971\Gamma^{0.25} - 0.5 + 0.180\Gamma^{-0.25} + \cdots$$
(17)

It is known [15] that the leading asymptotic large Γ term of this variational model for plasma mixtures obeys the linear-mixing rule, and in view of the nature of the asymptotic expansion the deviations from linear mixing are expected to be relatively small.

The Coulomb potential is a special limit of the more general Yukawa potential. The variational model can be expressed analytically also for Yukawa charges replacing the Coulomb charges, and thus represents electron screening corrections. For the screened Coulomb interaction, $(Z_i Z_j e^2 / r) e^{-\alpha r}$, the generalization of the Coulomb linear-mixing rule was derived recently in the form of the Yukawa linear-mixing rule [15], which involves the solution of nonlinear algebraic equations for the optimal ion-sphere radii R_i . In the limit $\alpha \rightarrow 0$, however, these yield [15] $R_i(\alpha=0)=a_i$, and the linear rule for Coulomb charges is recovered. Extensive data for Yukawa mixtures are not yet available, but our calculations predict that the deviations from the Yukawa linear mixing are always positive, and change very slowly with α . It is also interesting to note here that the deviations from the Yukawa linear mixing within the hypernettedchain approximation [15] are similar in magnitude to those obtained by the variational model, and are always positive.

The following model for the equation of state for fluid multicomponent plasmas is thus proposed. Combine (a) the linear-mixing approximation calculated with an accurate equation of state for the OCP, together with (b) the deviations from linear mixing calculated from the variational hard-sphere model (with the Percus-Yevick pair correlation functions, and the Percus-Yevick "virial" entropy):

$$f_{\rm ex}^{\rm mix} = (f_{\rm LM})_{\rm fit \ for \ OCP} + (\Delta f_{\rm ex}^{\rm mix})_{\rm variational \ HS \ model} .$$
(18)

This model agrees well with the OIIVH data when the DWSS fit for the OCP is used, and probably provides a more accurate BIM equation of state than the fit by OIIVH. Given a good representation for the potential energy of the one-component Yukawa system, the model, in its general form (18), is applicable to arbitrary fluid multicomponent Yukawa mixtures, and supplements the Yukawa linear-mixing rule developed [15] on the basis of the asymptotic strong coupling properties.

V. CONCLUSION

The present analysis finds that the negative deviations from linear mixing are obtained by OIIVH from underestimated error bars in their Fig. 2. Using the DWSS fit [3] to the OCP energies and repeating the OIIVH procedure [2] I obtain significantly different results, and thus demonstrate directly that the special features of Δf_{ex}^{BIM} , as emphasized by OIIVH, are the result of their choice of $u_0^{OIIVH}(\Gamma)$ and are not necessarily the reflection of their bare u_{ex}^{BIM} MC data. The analytic variational model, which provides the paradigm for the form of $u_0^{OIIVH}(\Gamma)$ itself, is in good agreement with the fluid binary-ionic mixture data of OIIVH, if the DWSS equation of state for the OCP is used to evaluate the deviations from the linear-mixing approximation.

The present analysis leads to a contradiction between (1) the OIIVH result [2] of *negative* and relatively *large* deviations from the linear-mixing approximation for small x, by using u_0^{OIIVH} in agreement with the OII extrapolation result [7] for the screening potential h_0^{OII} vs (2) *positive* and relatively *small* deviations from the linear-mixing approximation for small x, by using u_0^{DWSS} , in agreement with the Jancovici-Alastuey extrapolation [6] and the LM result h_0^{DN} , and in agreement with the previous criticism [8] of the OII extrapolation method.

ACKNOWLEDGMENTS

I thank Gilles Chabrier and Hugh DeWitt for interesting correspondence. This work was supported by The Basic Research Foundation administered by The Israel Academy of Sciences and Humanities.

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