CORRECTION

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Correction: The Lennard-Jones potential: when (not) to use it

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Correction for 'The Lennard-Jones potential: when (not) to use it' by Xipeng Wang *et al.*, *Phys. Chem. Chem. Phys.*, 2020, **22**, 10624–10633, **https://doi.org/10.1039/C9CP05445F**.

In a recent article,[1] Moro *et al.* presented evidence that, with increasing density, the Lennard-Jones-like (LJL) model with a cut-off length r_c = 2.0, proposed by Wang *et al.* in the published paper (**https://doi.org/10.1039/C9CP05445F**), goes through a sequence of hcp–fcc phase transitions. In their work, Moro *et al.* point out that their estimates of the fcc and hcp free-energies of the LJL model deviate, sometimes significantly, from the values in the Supporting Information of our published paper. We have checked our earlier calculations and found that there was indeed a problem: the free-energy calculations in the published paper were carried out using an MD implementation of our original (and current) MC program.[2] However, during this conversion, an error was introduced in the Gauss–Legendre quadrature. As a consequence, the solid free energies that we reported in the published paper deviated from those of Moro *et al.*,[1] in particular at higher temperatures. The aim of the present erratum is not to correct our earlier data: Moro *et al.*[1] already did that, and our current results agree with theirs for all temperatures and densities that we tested. Rather, this erratum aims to show that the problem was indeed due to the typo in the MD implementation, rather than to some intrinsic weakness of the MC-based Einstein-crystal method[2] that we described in our 2020 paper.

Here we report free-energy calculations using our original Monte-Carlo approach to compute the free energy of crystalline solids. The method used is as described in ref. 2. We point out that, in our MC calculations, we prepare an Einstein crystal where the mean-squared fluctuation of the atoms around their lattice sites is approximately equal to that of the interacting crystal. This is different from the thermodynamic integration method of ref. 1. Moreover, the free-energy difference between the pure Einstein crystal and models with a coupling parameter λ = 0.001 (*i.e.*, 99.9% Einstein Hamiltonian, 0.1% LJL Hamiltonian) was computed using a perturbation expression, as described in the published paper and ref. 2.

We carried out simulations for an almost-cubic fcc or hcp system, consisting of $N = n_x \times n_y \times n_z = 10 \times 12 \times 12 = 1440$ particles. We used a 10-point Gauss–Legendre quadrature to compute the free-energy difference between the (fixed-center-of-mass) Einstein crystal and interacting crystal. Every simulation took 10 000 MC cycles per quadrature point, excluding 2000 cycles equilibration (again, for every quadrature point). We always used the same parameters for fcc and hcp. With our Monte-Carlo code, we obtain perfect agreement with the Lattice-Switch MC (LSMC) results of ref. 1, and with the same accuracy. We note that our Einstein integration method (as described in the published paper) yields more accurate results than the Thermodynamic Integration method of ref. 1. This is presumably because our "integration path" is shorter. There was no problem with our earlier estimates of the pressure P, and the internal energy E. We refitted our data for the excess free energy per particle of the fcc and hcp solids (see Tables 1 and 2) using the functional form:

$$\beta \rho a_{exc}^S = \frac{3}{2} \rho \ln \beta + \sum_{n=n_{min}}^{n_{max}} \sum_{m=m_{min}}^{m_{max}} a_{n,m}^S \rho^n \beta^m, \quad (1)$$

where S stands for the fcc or hcp solid, and we recomputed the phase diagram (see Fig. 1). The computed location of the fcc–hcp transition (blue crosses) agrees with the values reported in ref. 1. We recomputed the solid–liquid coexistence curve, and note that they have shifted slightly (blue squares) with respect to our earlier calculations (red diamonds). The liquid–vapor data are not

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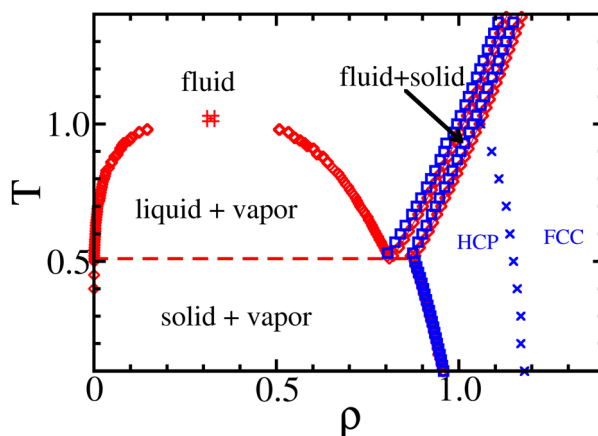[c] *Songshan Lake Materials Laboratory, Dongguan, Guangdong 523808, China*

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Table 1 Fit coefficients for the expression for excess free-energy density of the fcc phase for the LJL model with r_c = 2.0

n	$m = -2$	$m = -1$	$m = 0$	$m = 1$
0	149.1308	33.8178	−240.4856	78.0513
1	−608.8291	−320.4488	1197.4936	−365.0467
2	972.2152	891.8801	−2358.2015	679.6738
3	−755.6205	−1092.2398	2315.5287	−6530.0127
4	283.5720	623.0436	−1125.2783	309.9234
5	−40.5266	−135.7611	216.8797	−56.1099

Table 2 Fit coefficients for the expression for excess free-energy density of the hcp phase for the LJL model with r_c = 2.0

n	$m = -2$	$m = -1$	$m = 0$	$m = 1$
0	169.0558	−31.4967	−169.1413	63.9203
1	−730.4751	43.8471	833.2766	−299.1304
2	1251.9549	103.8821	−1623.9599	558.4164
3	−1064.7889	−259.4623	1583.1171	−542.7220
4	449.6473	190.5701	−763.0209	260.0897
5	−75.4508	−47.2149	145.6879	−47.1012

Fig. 1 Computed phase diagram of the potential given by eqn (1) for a cut-off distance r_c = 2.0 ("Lennard-Jones-like"). The computed location of the fcc–hcp transition (blue crosses) agrees with the values reported in ref. 1. We recomputed the solid–liquid coexistence curve, which has shifted slightly (blue squares) with respect to our earlier calculations (red diamonds). The liquid–vapor data are not affected.

affected. The figure does not show the width of the fcc–hcp two-phase region, as $\Delta\rho_{\text{fcc–hcp}}$ is of the order of 10^{-3}. Please refer to revised version of the ESI (**https://doi.org/10.1039/C9CP05445F**) for the updated calculation files.

The Royal Society of Chemistry apologises for these errors and any consequent inconvenience to authors and readers.

Notes and references

1 O. S. Moro, V. Ballenegger, T. L. Underwood and N. B. Wilding, *Phys. Chem. Chem. Phys.*, 2024, **26**, 7573–7579.
2 D. Frenkel and B. Smit, *Understanding molecular simulation: from algorithms to applications*, Elsevier, 2023.

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