Jarić and Mohanty Reply: We recently developed a density-functional formalism for calculating elastic moduli of crystals[1] and quasicrystals.[2] This formalism was illustrated in a recent Letter where we reported finding an elastic instability of a hypothetical icosahedral Co quasicrystal and the negative Poisson ratio of the hard-sphere (HS) crystal at melting.[3] Although our formalism is more general, these results were obtained from the second-order Ramakrishnan-Yussouff density-functional expansion.[4] Consequently, we suggested that third-order terms, which arise from the three-point correlations in the reference liquid phase, might be significant.

Our surprising negative-Poisson-ratio result especially stimulated much interest. Subsequent molecular-dynamics[5] and Monte Carlo[6] calculations, as well as a more general density-functional calculation which includes some higher-order correlations,[7] found that the Poisson ratio [defined as $\nu = C_{12}/(C_{11}+C_{12})$] is positive. They obtained $\nu = 0.347$, 0.343, and 0.674, respectively.

Such calculations do not account for the presence of an equilibrium density of vacancies and, therefore, cannot be compared with experiments or results of theories which take this effect into account. However, even if vacancies are excluded in the second-order Ramakrishnan-Yussouff density-functional theory, the HS Poisson ratio remains negative[1] ($\nu = -0.61$). Therefore, we shall explore the role of the neglected three-point correlations.

We shall focus on the uniform part $C_l^{(3)}(0,0)$ of the direct three-point correlation which has been recently calculated for the HS liquid.[8] This correlation contributes a term

$$-(1/6!)C_l^{(3)}(0,0)(n_s/n_l-1)^3$$

to the density functional, where n_s/n_l is ratio of the solid and liquid densities. At freezing, we have $n_s/n_l \approx 1.19$ and $C_l^{(3)}(0,0) \approx -300$ so that the above correction term changes ν to 0.06. This value of ν, although positive and remarkably close to the free-volume value[6] ($\gamma = 0.077$), is still too low. The correction term also affects the bulk modulus leaving the other eigenmoduli unchanged and too high. The corrected bulk modulus is within 10% of the more reliable values.[5-7]

Therefore, we conclude that inclusion of the three-point correlations beyond the uniform term is necessary and probably sufficient in order to calculate the elastic moduli with a similar degree of accuracy with which the thermodynamic potential can be calculated using only two-point correlations. It is hardly surprising that the second-order theory would yield accurate predictions of the melting transition (determined by the value of the thermodynamic potential) while at the same time leading to incorrect elastic moduli (determined by the derivatives of the potential). Undoubtedly, notwithstanding recent observations of quasicrystal distortions compatible with our results,[9] more sophisticated density-functional calculations, with more realistic two- and three-point correlations, are desirable.

Marko V. Jarić
Center for Theoretical Physics
Texas A&M University
College Station, Texas 77843

Udayan Mohanty
Department of Chemistry
Boston College
Chestnut Hill, Massachusetts 02167

Received 22 July 1987
PACS numbers: 62.20.Dc, 64.10.+h, 81.30.Kf

[1]M. V. Jarić and U. Mohanty, Phys. Rev. B, to be published.
[2]M. V. Jarić and U. Mohanty, to be published.
[3]M. V. Jarić and U. Mohanty, Phys. Rev. Lett. **58**, 230 (1987).
[4]T. V. Ramakrishnan and M. Yussouff, Phys. Rev. B **19**, 2775 (1979).
[5]D. Frenkel and A. J. C. Ladd, preceding Comment [Phys. Rev. Lett. **59**, 1169 (1987)].
[6]K. J. Runge and G. V. Chester, Phys. Rev. A (to be published).
[7]E. Velasco and P. Tarazona, Phys. Rev. A **36**, 979 (1987).
[8]B. B. Laird, J. D. McCoy, and A. D. J. Haymet, J. Chem. Phys. (to be published).
[9]D. Follstaedt, to be published.