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Comments

Comments are short papers which criticize or correct papers of other authors previously published in the **Physical Review**. *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Comment on "Quadrupolar glass freezing in solid hydrogen: Distribution functions for the orientational order parameters"

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It has been shown that in the above-mentioned paper the author starts from the orientational Hamiltonian of solid hydrogen containing an unphysical term leading, in general, to the nonvanishing average dipolar momentum of hydrogen molecules.

In Ref. 1, Kopeć has proposed the microscopic model of solid hydrogen in the quadrupolar glass (QG) state. It will be shown in the present Comment that this model contains an unphysical term and, in general, cannot be used to descibe the system under consideration.

The author starts from the Hamiltonian[1]

$$H = H_{QQ} + V \; , \tag{1}$$

where

$$H_{QQ} = -\sum_{i,j} J_{i,j} Q_{zi} Q_{zj} \; , \tag{2}$$

$$V = -\Omega \sum_{i} J_{xi} \; . \tag{3}$$

Here $Q_{zi} = 3J_{zi}^2 - 3$ and J_{zi}, J_{xi} denote the components in the local coordinate system of the angular momentum operator $\mathbf{J}_i$ related to the orthohydrogen molecule in the rotational state with $J = 1$ located at the ith site.

The orientable orthohydrogen molecules are randomly distributed in a matrix of spherical parahydrogen species and interact via long-ranged *exchange* (as has been written by the author) couplings J_{ij} obeying a Gaussian probability distribution with a zero mean. For the physical meaning of V [Eq. (3)] the author writes that this term "describes the quantum-mechanical tunneling between different quadrupole orientation, with Ω being tunneling frequency, in analogy to quantum dipolar case."

The model with the Hamiltonian H_{QQ} (2) has been studied in much detail in Ref. 2 with the difference that J_{ij} is understood as the electric quadrupole-quadrupole (EQQ) coupling. Though the use of the Hamiltonian (2), which is called a truncated EQQ Hamiltonian,[3] leads rather to a crude approximation, it gives some limited insight into the collective nature of the local freezing in the QG state (cf. Ref. 4 for a discussion).

On the contrary, the term V [Eq. (3)] has no physical justification. It was realized by Nakamura[5] a number of years ago that EQQ interactions are the most important orientational interactions between molecules in solid hydrogen. Moreover they are Lennard-Jones and Margenau–de Boer interactions, which are of secondary importance.[3] All orientational interactions lead to a coupling between quadrupolar momenta of molecules (the hydrogen molecule has no permanent dipolar momentum). A more thorough analysis of intermolecular interactions by A. B. Harris[6] leads to similar conclusions.

The presence in (1) of the term V [Eq. (3)] is in the contradiction with the physical picture since it leads to the nonvanishing thermal average $\langle \mathbf{J}_i \rangle$ which is proportional to the molecular dipolar momentum.[3] It has been many times stressed in the literature that for all components $\langle \mathbf{J}_i \rangle = 0$.[7,8]

In addition in Ref. 1, the lack of a well-defined glass transition in the system is explained by the breaking of spin reversal symmetry. The author writes that this symmetry is broken "even in the high temperature phase without applied field." This is a misinterpretation since spin reversal symmetry is understood in the usual sense, i.e., $\mathbf{J}_i \rightarrow -\mathbf{J}_i$, and its possible breaking is relevant for spin or pseudospin (as dipolar) systems and has no meaning for the quadrupolar ones. The true physical reason for the suppressing of the sharp phase transition in a solid ortho-parahydrogen mixture in the QG state is local symmetry breaking by dilution as is discussed in Refs. 4 and 8.

In conclusion, in Ref. 1 the author has inserted quite formally some elements of the theory of dipolar glasses into the theory of QG's, obtaining unphysical results for the orientational parameter distribution function when $\Omega \neq 0$.

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[1] T. K. Kopeć, Phys. Rev. B **48**, 3698 (1993).

[2] E. A. Luchinskaya, V. N. Ryzhov, and E. E. Tareeva, J. Phys. C **17**, L665 (1984); Theor. Math. Phys. **67**, 623 (1986); E. A. Luchinskaya, *ibid.* **67**, 524 (1986).

[3] A. B. Harris, Phys. Rev. B **1**, 1881 (1970).

[4] N. S. Sullivan, Can. J. Chem. **66**, 908 (1988).

[5] T. Nakamura, Prog. Theor. Phys. **14**, 135 (1955).

[6] A. B. Harris, Phys. Rev. B **1**, 1881 (1970).

[7] N. S. Sullivan, M. Devoret, B. P. Cowan, and C. Urbina, Phys. Rev. B **17**, 5016 (1978); N. S. Sullivan, M. Devoret, and D. Estève, *ibid.* **30**, 4935 (1984); Y. Lin and N. S. Sullivan, Mol. Cryst. **142**, 141 (1981); X. Li and H. Meyer, Phys. Rev. B **37**, 3216 (1988).

[8] N. S. Sullivan, C. M. Edwards, Y. Lin, and D. Zhou, Can. J. Phys. **65**, 1463 (1987).