Comment on "Low-Temperature Specific Heat of C_{60}"

In their Letter, Beyermann *et al.* [1] reported their measurement of the specific heat of solid C_{60} in the low-temperature simple-cubic (sc) phase. The C_{60} molecules in the solid are expected to vibrate as well as librate, and both types of motion should contribute to the specific heat. Since the solid has a sc structure with four molecules per primitive unit cell, there are twelve vibrational and twelve rotational excitation branches. In order to fit their experimental data, the authors proposed a model for the specific heat, which takes into account both vibrational and rotational contributions. In the model the authors assume that among the twelve vibrational modes, three modes are acoustic and they can be modeled by a Debye term in the specific heat, and the other nine modes are optical and they can be modeled by an Einstein term. The authors also make the *same* assumptions for the *rotational* modes.

I would like to point out that, otherwise a plausible model, this model contains an unfortunate mistake which can be easily corrected. The mistake is in the assumption that there are three acoustic rotational modes. In fact there do not exist any acoustic rotational modes.

The existence of acoustic modes is a result of invariance properties in the system. For translational motion, the solid is invariant if all molecules in the solid are translated by the same displacement vector. As a result, in the long-wavelength limit, modes that correspond to uniform translations have zero energy, and there are three such acoustic vibrational modes in a three-dimensional solid. However, for rotational motion, the situation is different. In solid C_{60}, each molecule rotates around an axis centered on itself. Consider an operation in which all the rotation axes for different molecules are aligned in the same direction, and all molecules are rotated by the same angle. Such an operation does not leave the sc solid C_{60} invariant, because the C_{60} molecule is anisotropic and is not rotating freely; in fact the anisotropic aspect of the C_{60} molecule is directly related to the librational motion of C_{60} molecules in the sc solid C_{60}. The lack of invariance with respect to the above rotational operation shows that there should not exist any acoustic rotational modes. Several authors have calculated the librational frequencies of sc solid C_{60}. Indeed, no acoustic libron modes are found, and the frequencies vary between 7–24 cm^{-1}.

The model used by the authors can still be a useful model, if the above mistake is corrected. To do so, the parameters n_{DR} and n_{ER} in the model should be fixed at 0 and 1, respectively. This corrected model is much more physical than the other second model used in their paper. I suggest that the authors carry out a new fit of their data with the corrected model. The result can be used to compare to the calculated libron frequencies, and help improve the intermolecular model potential. Since the calculated lowest and highest libron frequencies differ by a factor of 10, a model with more than a single rotational Einstein contribution might be even better.

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