Comment on "Lattice Phonon Modes in Solid C_{60} Studied by Far-Infrared Spectroscopy"

Recently, Huant *et al.* [1] have claimed to observe the two allowed zone-center far-infrared active translational modes of T_u symmetry for the simple cubic phase of solid C_{60}. In Fig. 3 of their Letter they show a transmission spectrum containing five distinct lines with frequencies less than 60 cm^{-1}. They assign the two strongest of these lines at 26.6 and 58.5 cm^{-1} to be the two T_u modes. In this Comment we demonstrate that their assignment is not correct.

Our samples, like theirs, consist of C_{60} powder compressed under high pressure to form a dense pellet. The C_{60} material studied was obtained from two sources, commercially obtained pure C_{60} and C_{60}-C_{70} extract, which we purified by means of column chromatography. In both cases the pellets were heat treated under vacuum to remove any trace of solvent as determined by infrared reflectivity. Room temperature x-ray characterization verified that the samples possessed a face centered cubic structure.

The dashed curve in Fig. 1 shows our low temperature spectrum for a sample which has been exposed to air. This spectrum is very similar to that in Fig. 3 of Huant *et al.* [1] and contains five low temperature absorption lines. However, when the sample is placed in an optically accessible vacuum cell, pumped for several hours at pressure $< 10^{-6}$ torr and temperatures of 100 °C, and then cooled to 1.5 K, the solid curve in Fig. 1 is obtained. The strong lines at 26 and 58 cm^{-1} have vanished and only two lines remain, at 40.9 and 54.7 cm^{-1}. It is these two lines which must therefore be identified with the two T_u zone-center modes associated with the low-temperature simple cubic phase of C_{60}.

Temperature dependent measurements between 300 and 1.5 K confirm this assignment. These two absorption lines first appear at 260 K (the accepted phase transition temperature to the simple cubic structure [2]) and grow in amplitude as the temperature is lowered. Both modes shift to higher frequency by a few wave numbers on cooling to 1.5 K. In contrast, the other modes in Fig. 1 (dashed) only appear below about 40 K.

When a degassed sample is exposed to air for an hour or so at room temperature and then cooled down, its spectrum once again looks like the dashed curve in Fig. 1.

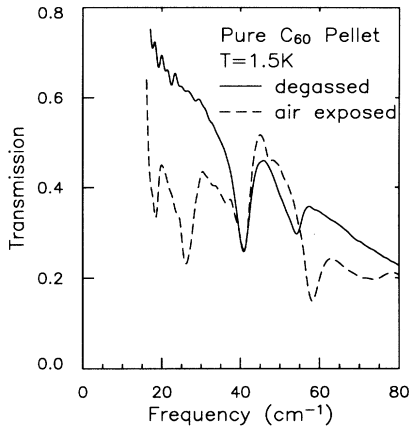

FIG. 1. Low temperature far-infrared transmission spectra of a C_{60} pressed pellet. The spectral resolution is 1.5 cm^{-1}.

This effect is completely reversible and one can monitor the growth of these additional absorption lines as a function of the time of exposure to air. Selectively filling the cell with an atmosphere of either pure O_2, N_2, or CO_2 does not reproduce this (dashed) spectrum. However, when water vapor is introduced into the cell the result is virtually identical to the air exposed case except that the impurity modes are now even stronger. Further details of these results will be presented in a future publication.

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[1] S. Huant, J. B. Robert, G. Chouteau, P. Bernier, C. Fabre, and A. Rassat, Phys. Rev. Lett. **69**, 2666 (1992).
[2] Paul A. Heiney *et al.*, Phys. Rev. Lett. **66**, 2911 (1991).