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Comment on "New Orientationally Ordered Low-Temperature Superstructure in High-Purity C_{60}"

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Abstract

A Comment on the Letter by G. Van Tendeloo *et al.* Phys. Rev. Lett. *69*, 1065 (1992).

Disciplines

Physics | Quantum Physics

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In an interesting Letter [1] Van Tendeloo et al. have reported the observation of superlattice reflections in electron diffraction from crystalline C_{60}. They interpret their data to indicate a doubling of the unit cell relative to the heretofore accepted [2–6] $Pa\overline{3}$ structure in which a unit cell contains four orientationally inequivalent molecules, each rotated (relative to a standard orientation [6]) through an angle ϕ about its local threefold axis. Van Tendeloo et al. propose a structure in which the unit cell contains eight molecules, four of which are rotated through an angle ϕ and four through an angle $-\phi$. The purpose of this Comment is twofold: first, we show that this proposed stucture leads to an x-ray powder diffraction spectrum which contradicts existing data; and second, that although cubic structures with eight molecules per unit cell on fcc lattice sites are possible, the proposed structure is actually trigonal, and not cubic as claimed in Ref. [1].

First, to test their proposed structure we compare the powder diffraction spectrum it gives, in Fig. 1, to that observed by Heiney et al. [7]. Clearly, the fit is much inferior to that based on the $Pa\overline{3}$ structure. Thus, either the proposed structure is incorrect, or the samples studied by Van Tendeloo et al. are qualitatively different from those of Refs. [4], [5], and [7].

The second point is that the only allowed cubic space groups for icosahedral molecules statically ordered on an fcc lattice [8], with a unit cell containing eight molecules, are quite different from $Pa\overline{3}$: $Fm\overline{3}$ (similar to [6] $Pm\overline{3}$), $Fd\overline{3}$, and $Fd\overline{3}c$. The last two are similar to [3] $Pn\overline{3}$.

To understand the observations of Ref. [1] (and similar observations of Ref. [9]) without abandoning the data of Refs. [4], [5], and [7], we consider a structure in which the angles ϕ and $-\phi$ in the model of Ref. [1] are replaced by $\phi + \delta$ and $\phi - \delta$. Such structures are not cubic: doubling the unit cell leaves only one of the (1,1,1) axes as a threefold axis [8]. For small δ, this trigonal structure, space group $R\overline{3}$, may be obtained from $Pa\overline{3}$ by condensing a zone-corner librational phonon. Although there is no coupling between librational and translational phonons at the zone corner [10], there is a cubic coupling energy of order $e\delta^2$, where e is the magnitude of the shear strain [8], all three of whose components have the same magnitude. This shear strain will cause the powder diffraction line indexed by (H, K, L) to split into components (depending on the choices of signs of H, K, and L). Both the superlattice diffraction intensity and the size of the powder diffraction splitting are proportional to δ^2. A key experiment would be to detect this splitting in the powder diffraction spectrum. (The size of the splittings is
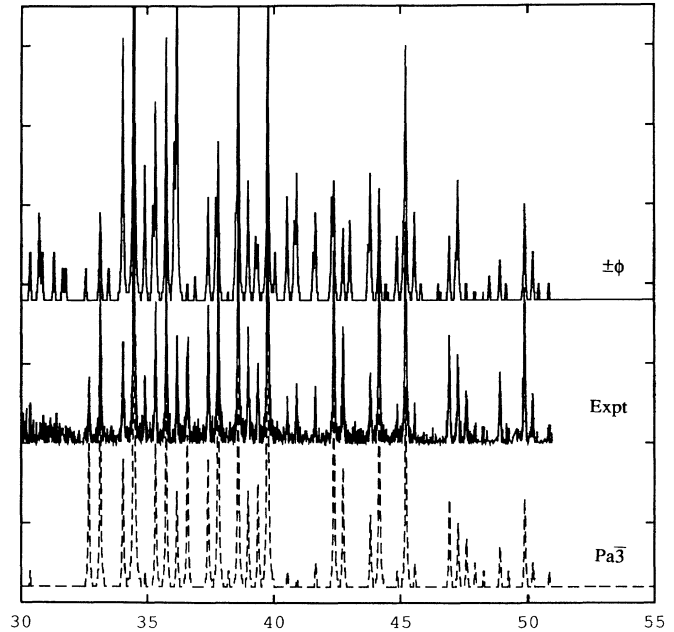

FIG. 1. Powder x-ray diffraction spectra (intensity vs scattering angle in degrees): (bottom) calculated (after D. E. Cox, see Ref. [6]) for $Pa\overline{3}$ with $\phi = 26.5°$; (middle) observed [7]; (top) for the structure of Ref. [1] for optimal $\phi = 22.4°$. The wider lines here are due to superlattice components.

probably below the resolution of Refs. [1] and [9].)

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