

**van Loosdrecht, van Bentum, and Meijer Reply:** Hadjiev and Thomsen express a legitimate concern about the possible effects of impurities on the Raman spectra of fullerenes [1]. It is well known that remains from solvents, oxygen, or the presence of  $C_{70}$ , can drastically change the vibrational properties of solid  $C_{60}$ . More specifically, in the presence of these impurities the rotational ordering transition extends over a much larger temperature range. In our experiments we tried to avoid pollution as much as possible. First, mass spectroscopic analysis of the chromatographically purified starting material indicates a  $C_{70}$  content less than 0.5%. The multiple sublimation growth technique [2] in vacuum provides a further purification. Second, morphological study, x-ray diffraction, electron diffraction, and high-resolution electron microscopy crystal analysis all indicate a nearly perfect crystal quality and no sign of  $C_{70}$ . Third, to exclude possible surface pollution from the growth method the crystals were cleaved to expose a clean fresh surface. To prevent surface deterioration, the crystals are mounted in an optical cryostat which is immediately evacuated. Fourth, some crystals were checked by mass spectroscopic analysis which showed no detectable traces of  $C_{70}$ . Finally, the Raman spectra themselves were checked for indications of  $C_{70}$ . In the low-temperature phase a total of 49 modes were observed, some of which are close to the known  $C_{70}$  modes. However, no features were observed near 1183 and  $1228\text{ cm}^{-1}$  (see the inset in Fig. 1), showing the absence of  $C_{70}$ . (Hadjiev and Thomsen refer to a peak near  $1450\text{ cm}^{-1}$ , which was in fact a spike in our raw data.)

In Fig. 1, we show the spectra of the highest  $H_2$  mode in the  $2a_0$  fcc phase (lower curve, 40 K) and the fcc phase (upper curve, 260 K). A comparison with curves (c) and (d) in Fig. 1 of Hadjiev and Thomsen clearly shows that the peak shift and broadening observed in our spectra cannot be explained by the presence of a small amount of  $C_{70}$  impurities. Note that the displacement of the  $1566\text{-cm}^{-1}$  mode to  $1575\text{ cm}^{-1}$  in the results of Hadjiev and Thomsen is a well-known effect of air-exposed  $C_{60}$  [3,4].

It remains surprising that such a large frequency shift is observed for this mode. Even though a quite large volume change is associated with the rotational ordering transition, it is not very likely that the intermolecular interactions alone can account for this effect. We propose that, in addition, the very strong electronic resonance conditions for 514-nm excitation change. Even very low irradiances of the order of  $5\text{ W/cm}^2$  can lead to an electronic excitation into the metastable triplet state and a corresponding softening of the phonon in the high-

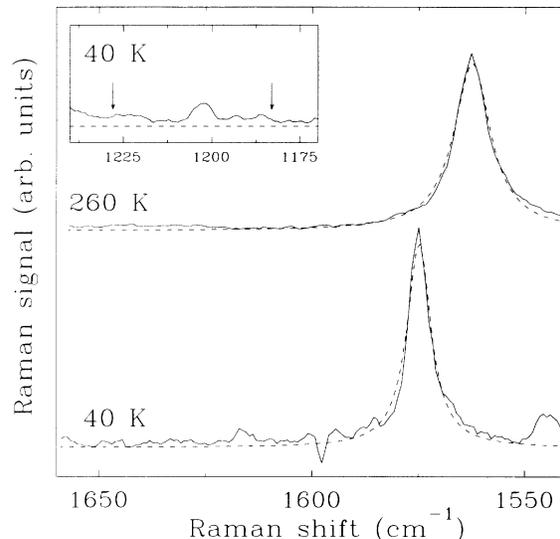


FIG. 1. Unpolarized Raman spectra of the highest  $H_g$  mode in single crystal  $C_{60}$  at  $T=40\text{ K}$  (lower curve) and  $T=260\text{ K}$  (upper curve). The dashed lines are fits of a single Lorentzian curve. Inset: The absence of the  $1183\text{-}$  and  $1228\text{-cm}^{-1}$   $C_{70}$  modes (indicated by arrows) in the  $T=40\text{ K}$   $C_{60}$  Raman spectrum.

temperature phase [3]. The presence of oxygen blocks the rotational motion and simultaneously quenches the triplet state [5], thus preventing the anomalous softening.

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