Moncton *et al.* Respond: Marti and Ceva[1] propose that the width of the incommensurate peaks reported by us in Ref. 2 is due to a distribution of critical points throughout the sample. The exfoliated graphite and Kr dose were identical to the lattice-gas melting experiment,[3] for which we observed a rounding of the critical point of 0.07 K (standard deviation). Consequently, we expected a similar broadening of the commensurate-incommensurate transition. Such a rounding would have two observable effects: It would induce a coexistence of commensurate and incommensurate phases in the vicinity of the average critical point, and it would broaden the incommensurate peaks. We have quantitatively analyzed both regimes and found that interpretations in terms of a rounded transition are incompatible with the data.

The controlling variable in this transition is the chemical potential of the adsorbate. A distribution of crystallite sizes, defects, edge energies, etc., would give rise to a distribution of critical chemical potentials. The experiment was carried out as a function of temperature in a closed cell so that the vapor pressure varied with temperature T. However, the relatively large cell acted as a reservoir, stabilizing the pressure, so that the chemical potential was roughly proportional to the temperature. It is therefore meaningful to analyze the transition as a function of T.

We first consider the influence of rounding on the commensurate peak intensity. A distribution of critical chemical potentials would produce a range of thermodynamic states throughout the sample at a specified temperature and pressure. If we imagine that the commensurate peak intensity due to one crystallite remains constant up to its critical point, then the total diffracted intensity is proportional to the number of crystallites in the commensurate phase. The composite line shape is seen in the range $T = 97.24$–97.55 K, where the peak intensities are 0.27–0.83 of the saturated value, respectively. Under the assumption of a Gaussian distribution of critical temperatures, the corresponding half width at half maximum (HWHM) of the critical temperature distribution is $\delta T = 0.23$ K. This analysis ignores any intrinsic evolution of the commensurate phase near the transition and so it produces an upper limit to δT which is almost certainly an extreme overestimate due to critical scattering.

It is straightforward to show that the half-width of the powder-averaged Lorentzian function due to δT is given by

$$\kappa/\epsilon = [\sqrt{3}\,(\epsilon/\epsilon_0)^{1/\beta} T/(\beta\,\delta T)]^{-1} \quad (1)$$

in Marti and Ceva's notation. The factor $\sqrt{3}$ arises from the azimuthal integral. Using our upper limit on δT of 0.23 K we obtain an upper limit for κ/ϵ attributable to rounding which is 4–5 times smaller than the measured values reported in Ref. 1 for $\epsilon > 0.02$ Å^{-1}.

The rounding model also has observable consequences for the peak shape. The presumed Gaussian distribution of energies would produce a Gaussian diffraction peak when ϵ is somewhat larger than κ. Marti and Ceva are correct in stating that the published data cannot distinguish the line shape. However, stimulated by the interest in these issues, we have performed another run with much better statistics, which clearly shows that the incommensurate peaks above 0.02 Å^{-1} misfit are not consistent with a Gaussian profile.

When $\epsilon < 0.02$ Å^{-1}, one cannot replace differentials with finite dispersions. We have explicitly calculated the expected distribution of ϵ under these conditions, and find that the experimental peak width is an order of magnitude broader than can be accounted for by the upper limit of δT. Furthermore, in the presumed coexistence regime, the distribution of incommensurabilities would peak at a finite value of ϵ because of the fact that there is a negligible number of crystallites having ϵ close to zero. If this mechanism were operative, then composite-line-shape data such as the 97.42-K scan in our Fig. 2 would have a relatively sharp peak centered at 1.72 Å^{-1}. Details of these calculations will be presented in a forthcoming publication.

For the above reasons, we concluded in Ref. 1 that the width of these peaks is an intrinsic feature of the incommensurate phase close to commensurability.

D. E. Moncton
P. W. Stephens[a]
R. J. Birgeneau
P. M. Horn
G. S. Brown

Received 13 August 1982
PACS numbers: 68.55.+b, 61.10.-i, 64.70.Dv

[a]Address for correspondence: Department of Physics, Brookhaven National Laboratory, Upton, N. Y. 11973.

[1]C. Marti and T. Ceva, preceding Comment [Phys. Rev. Lett. 49, 1678 (1982)].

[2]D. E. Moncton, P. W. Stephens, R. J. Birgeneau, P. M. Horn, and G. S. Brown, Phys. Rev. Lett. 46, 1533 (1981).

[3]R. J. Birgeneau, G. S. Brown, P. M. Horn, D. E. Moncton, and P. W. Stephens, J. Phys. C 14, L49 (1981).