COMMENTS

Comments are short papers which criticize or correct papers of other authors previously published in **Physical Review B.** *Each Comment should state clearly to which paper it refers and must be accompanied by a brief abstract. The same publication schedule as for regular articles is followed, and page proofs are sent to authors.*

Comment on ''Fluctuations during freezing and melting at the solid-liquid interface of xenon''

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Di Nardo and Bilgram have recently reported observations of anomalous diffusive light scattering at the crystal-melt interface of xenon [Phys. Rev. B **51**, 8012 (1995)]. They interpret their data as evidence for a crystal-like preordered layer at the liquid side of the interface which they associate with the crystallization process. We discuss an alternative explanation for their observations. [S0163-1829(96)04226-9]

Di Nardo and Bilgram have observed strong quasielastic light scattering at the growing crystal-melt interface of xenon.[1] Their report fails to mention either the extensive experimental literature on this phenomenon or the alternative explanation for it that many of these publications support. Unfortunately, their presentation leaves the reader with a biased view of this interesting but controversial area.

In 1978, Bilgram, Guttinger, and Kanzig reported a light-scattering phenomenon observed at the crystal-melt interface of a growing ice crystal. After growing for a time on the order of one hour, strong quasielastic scattering was observed at the interface.[2] By 1990 this effect had been reported to occur in eight different materials by groups in five different laboratories. (For a review, see Ref. 3.)

In Ref. 2 the authors proposed that the light scattering was caused by overdamped capillary waves on the crystal surface. However, it was later found that the strong quasielastic scattering cannot be observed on the crystal side of the interface, and is thus characteristic of the fluid layer adjacent to the interface.[4] Bilgram then proposed a ''mesophase'' model in which the fluid boundary layer constitutes a new state, characterized by a high isothermal compressibility.[5] The strong quasielectric scattering was then attributed to ordinary density fluctuations in this boundary layer.

An alternative explanation for the strong quasielastic scattering was suggested in 1986, based on the presence of residual dissolved gas in the fluid.[6] As solidification proceeds, rejected solute builds up as a ''spike'' ahead of the advancing interface. Eventually, the concentration becomes sufficiently high to nucleate small gas bubbles which then diffuse in the interface producing the observed scattering. The delay for onset of the scattering is then explained by the time required for the solute concentration to reach the level necessary for microbubble nucleation to begin. The microbubble model was put on a more quantitative basis in 1988,[7] and was shown to explain a number of experimental observations including those of Bilgram on the ice-water interface.

Additional support for this microbubble explanation came from experimental observations. (1) Laherrere *et al.*[8] observed scattering at the crystal-melt interface of succinonitrile. Repeated melting and recrystallization followed by pumping gradually reduced the intensity of the scattering. (2) Williams, Srinivasan, and Cummins[9] observed light scattering from the crystal-melt interface of salol growing in a very long sample tube. After several days the bubble size increased significantly, eventually becoming macroscopic so that the individual bubbles were visible. (3) Vesenka and Yeh[10] observed that once the scattering layer had formed, if the growth was stopped and immediately reversed, the scattering layer ''lifted off'' the crystal surface. (A more detailed review of these experiments can be found in Ref. 3.)

Bilgram[5] has criticized the microbubble model on the basis that homogeneous nucleation theory implies that bubble nucleation requires an enormous gas supersaturation. However, as is evident in opening a bottle of seltzer, even small supersaturation can produce profuse bubble nucleation via heterogeneous nucleation at the surfaces. In the solidification experiments, the crystal-melt interface provides the required surface.

While there may be more than one mechanism responsible for quasielastic interfacial light scattering during crystal growth, the microbubble mechanism is the simplest and, in our opinion, the most likely explanation for the observations reported by Di Nardo and Bilgram.

[1] S. Di Nardo and J. H. Bilgram, Phys. Rev. B **51**, 8012 (1995).
[2] J. H. Bilgram, H. Guttinger, and W. Kanzig, Phys. Rev. Lett. **40**, 1394 (1978).
[3] H. Z. Cummins and L. M. Williams, in *Light Scattering by Liquid Surfaces and Complimentary Techniques*, edited by D. Langevin (Marcel Dekker, New York, 1992), p. 287.
[4] O. N. Mesquita, D. G. Neal, M. Copic, and H. Z. Cummins, Phys. Rev. B **29**, 2846 (1984).
[5] J. H. Bilgram, Phys. Rep. **153**, 1 (1987).
[6] H. Z. Cummins. G. Livescu, H. Chou, and M. R. Srinivasan, Solid State Commun. **60**, 857 (1986).
[7] O. N. Mesquita, L. O. Ladeira, I. Gontijo, A. G. Oliveira, and G. A. Barbosa, Phys. Rev. B **38**, 1550 (1988).
[8] J. M. Laherrere, H. Savary, R. Mellet, and J. C. Toledano, Phys. Rev. A **42**, 1142 (1990); **42**, 2534 (1990).
[9] L. Williams, M. R. Srinivasan, and H. Z. Cummins, Phys. Rev. Lett. **64**, 1526 (1990).
[10] J. P. Vesenka and Y. Yeh, Bull. Am. Phys. Soc. **32**, 711 (1987); **33**, 781 (1988).