Comment on "Direct Measurement of the Percolation Probability in Carbon Nanofiber-Polyimide Nanocomposites"

In their Letter, Trionfi *et al.* [1] claimed to derive percolation critical exponents for a carbon nanofiber-polyimide (PCNF) nanocomposite. They suggested there that the latter system "belongs to a different universality class than the 3D lattice percolation model." In this Comment we intend to point out that their experimental results hardly support such an interpretation and that the tunneling-hopping-like approach can better account for their results.

In Ref. [1] six points data fitting for the dependencies of the percolation cluster probability, θ_∞, and the conductivity, σ, on the volume content of the "conducting phase" ,p, were concluded to yield mean field, Bethe latticelike, exponents. This interpretation in terms of a percolation phase transition has two difficulties. First, the percolation thresholds, p_c, of 0.002 ± 0.002 and 0.001 ± 0.001, may suggest that $p_c = 0$, and thus the whole premise of that argument and the meaning of the critical exponents is questionable [2]. Second, and more severe, the data were taken far away from the claimed p_c [as far as $(p - p_c)/p_c = 35$ for $\sigma(p)$ and as far as $(p - p_c)/p_c = 17$ for $\theta_\infty(p)$]. This is quite critical since it is well established that "when p_l is appreciably larger than $p_{lc} \ldots \Sigma$ as well as $P \ldots$ increase roughly linearly with the concentration p_l" [3], where the quantities Σ, P, and p_l here are the lattice counterparts of σ, θ_∞, and p. In view of the above their $\beta = 1.1 \pm 0.3$ value is more reliably accounted for by the above $\theta_\infty \propto p$ (or $P \propto p_l$ [3]) expectation. Hence, the interpretation of such (far from the apparent p_c) data by critical exponents, such as $\beta = 0.4$, or $\beta = 1$, is not justified and the $\theta_\infty \propto p$ dependence simply suggests that the data are associated with a homogeneous system.

In an attempt to pursue their "percolation model" in terms of a Bethe lattice, the authors of Ref. [1] apply the well known $p_{lc} = 1/(Z - 1)$ relation where Z is the site coordination in the Bethe lattice [3]. However, in doing so they mix p_{1c} (the critical occupation probability on a lattice) with the critical volume fraction p_c which are two different quantities. In particular, the value of p_l is not defined in the continuum, while in lattices, for a given p_l, the value of p depends on the volume and shape of the individual *impenetrable* particle that is attached to a site [4]. Hence, the derivation of the $Z = 500$ value by replacing p_l by the measurable p in Ref. [1], is simply wrong.

In view of the above let us suggest an alternative interpretation of the data of Ref. [1] by considering the $\sigma(p)$ dependence as shown there in Fig. 3 for PCNF and as given by the authors on a similar (FCNF) system in Ref. [5]. Following hopping [6,7] or other tunneling related mechanisms [8] one obtains that, depending on the shape and size of the particles, $\sigma = \sigma_\gamma \exp(-a_\gamma/p^\gamma)$, where, σ_γ and a_γ, are constants of the system. Indeed, by analyzing the $\sigma(p)$ data of Refs. [1,5] we found that the quality of the fits of the PCNF [1] and the FCNF [5] data to the latter dependence with $\gamma = 1$ and $\gamma = 1/3$, respectively, are at least as good as the fits to the $\sigma(p) \propto (p - p_c)^t$ percolation dependence proposed in Ref. [1]. In fact for such systems (depending on the density) the tunneling-hopping models can be shown to yield γ values in the $1/3$ to 1 range. Indeed, such equal quality fits have already been interpreted within the framework of a tunneling transport mechanism [8,9]. Note, however, that no critical region is involved in the tunneling-hopping interpretation of the conductivity and therefore the critical region restrictions do not apply. On the other hand, the corresponding models are consistent with random homogeneous systems [6].

In conclusion, considering that the data of Ref. [1] were obtained far away from the claimed p_c (that can be taken as 0) and that $\theta_\infty \propto p$, the observations of Ref. [1] (in contrast with the unfounded claims there) can be self consistently interpreted as due to a dilute homogeneous system in which a hoppinglike transport takes place.

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