Was Superlocalization Observed in Carbon-Black–Polymer Composites?

In a recent Letter [1], van der Putten *et al.* fitted the conductivity of carbon-black (CB)-polymer composites to the three-dimensional (3D) percolation form $\sigma=\sigma_T(p-p_c)^t$, where p was the concentration of CB and $t\approx 2$. The temperature-dependent coefficient σ_T was fitted to the variable range hopping (VRH) form, $\sigma_T=\sigma_0\times\exp[-(T_0/T)^\gamma]$, with $T_0=112$ K and $\gamma=0.65$. This value of γ was then interpreted as indicating superlocalization of the electronic states on the presumed fractal CB structure. For a fractal, these would have the form $\psi(\mathbf{r})\propto\exp[-(r/L)^\zeta]$. Reference [1] deduced the value $\zeta=1.94$, in apparent agreement with Lévy and Souillard [2], who predicted that $\zeta=d_w/2$, where d_w is the fractal dimension of a random walk on the percolating cluster.

Contrary to the statement in Ref. [1], the theoretical value of ζ is no longer "open to controversy." For typical percolation clusters, and for localized energies far away from the conductance band, strong analytic arguments showed [3] that $\psi\propto\exp(-Al)$, where $l\propto r^{d_{\min}}$ is the minimal path on the fractal. Thus $\zeta=d_{\min}$ (≈ 1.34 in 3D) and *not* $d_w/2$. This has been confirmed numerically, even for states near the band [4,5]. The only "controversy" left concerns Ref. [2], which used a relation between $\psi(\mathbf{r})$ and the distribution function for random walks, $P(\mathbf{r},t)=P(0,t)\exp[-c(r^{d_w}/t)^\alpha]$, with $\alpha=1$. As was explained [3], the correct value of α for typical realizations is $\alpha=d_{\min}/(d_w-d_{\min})$, equivalent to $\zeta=d_{\min}$.

Not only does theory not support the interpretation of Ref. [1], but also the parameters deduced there imply hopping distances R_h in the range $(0.4\text{–}1.8)L$, where L is the localization length, while VRH theory applies when $R_h\gg L$. Further, Ref. [1] finds that $R_h=O(L)=O(R_0)$, where R_0 is the size of a single CB sphere, while to observe fractal scaling (i.e., superlocalization) one would need to have $R_h\gg R_0$.

How then does one explain the data of Ref. [1]? Generally VRH should yield [6]

$$\sigma_T=\sigma_0(T_0/T)^s\exp[-(T_0/T)^\gamma]\,, \qquad (1)$$

with the temperature-dependent prefactor T^{-s}. Given the uncertainty in the model-dependent value of s, we present in Fig. 1 a plot of Eq. (1) for $s=2$, $\gamma=0.33$, and $T_0=42600$ K. For $4<T<100$ K, and even for s as large as 2, one can hardly distinguish between the two curves. They do differ significantly for $T<4$ K, and new data in that regime would be useful. Our curve has a minimum near 200 K, indicating that the exponential factor becomes dominated by the prefactor and implying that VRH theory is no longer appropriate at these high temperatures. Interestingly, the data in Fig. 2 of Ref. [1] show similar deviations from pure exponential at high T. According to Ref. [7], our value of T_0 seems more plausible than that ($T_0=112$ K) of Ref. [1]. In fact, in our fit, $(R_h/L)^\zeta$ is of order 5–15. In Ref. [1], the limited range of T and the fact that the deduced R_h is of order L and R_0 prevent a decisive interpretation of the data. The value $s=2$ is somewhat extreme and we used it to illustrate our points. Fits progressively better than that in Fig. 1 are obtained as s is decreased towards zero. In particular, a Coulomb gap with $\zeta=d_{\min}=1.34$, yielding $\gamma=0.57$, could certainly be fitted with a small value of s.

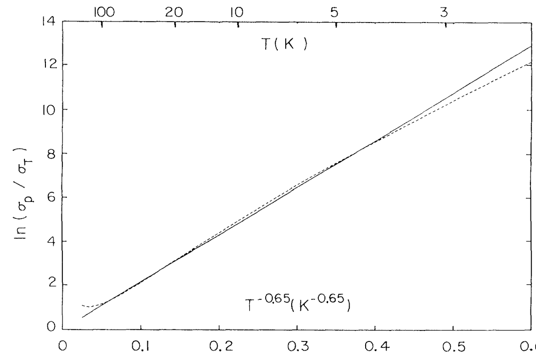

FIG. 1. Different fits to σ_0/σ_T. Full line: $\sigma_0/\sigma_T=\exp\{[112/T(K)]^{0.65}\}$, as in Ref. [1]. Dashed line: $\sigma_0/\sigma_T=364(T/T_0)^2\times\exp[(T_0/T)^{0.33}]$, with $T_0=42600$ K.

In conclusion, since the value of γ depends strongly on the analysis, the data of Ref. [1] have more than one explanation, and therefore confirm no theory. Measurements at lower T may present tests of our Eq. (1). Many other experimental studies also invoke too small values of T_0 and R_h. In such cases the inclusion of prefactors as in Eq. (1) may lead to more satisfactory interpretations.

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