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Temperature–dependent conduction in composites: a percolative approach

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Abstract: Granular composite systems show very interesting temperature-dependent conduction properties particularly in the low temperature regime. Some recent experiments on them tend to show Mott variable range hopping behaviour alongwith dilution dependent exponents. Here we propose a semi-classical percolative picture and a model random resistor cum tunneling-bond network (RRTN) for the composite systems in general to understand their low-temperature behaviour.

Keywords: variable range hopping, composites, percolation, random resistor cum tunneling network (RRTN)

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1. Introduction

The temperature (T) dependence of the conductivity $\sigma(T)$ of disordered systems has long been a subject of great interest, particularly in the low temperature region, because one expects the effects of quantum fluctuations due to disorder and/ or other types of scattering mechanisms (say, due to Coulomb interaction between charged carriers) to be very prominent there. For example, for an Anderson insulator at low temperature, Mott proposed his variable-range hopping (VRH) law [1],

$$\sigma(T) \propto \exp[-(T_0/T)^\gamma], \qquad (1)$$

where T_0 is a characteristic temperature (or, energy) scale that determines the domain in which phonon-assisted hopping among localized states at different energies contribute significantly. For a homogeneous d-dimensional solid $\gamma = 1/(d+1)$; γ=1/4 in 3D. For a pure system with Coulomb interaction, Efros and Shklovskii [2] found that $\gamma = 1/2$ which is true in any dimension. Aharony *et al.* [3] argued that random walk of a quantum particle on fractal-like structures should give rise to a generalized VRH formula:

$$\sigma(T) = \sigma_0(T_0/T)^s \exp[-(T_0/T)^\gamma], \qquad (2)$$

with the temperature dependent prefactor T^{-s} where $s > 0$ [4].

In a recent experiment [5] on a carbon-black–polymer composite, evidence of superlocalized states due to Coulomb interaction was reported, and fitting with eqn.(1) gives a value of $\gamma = 0.66 \pm 0.02$. Superlocalized states are such that their wave functions decay faster than exponential, namely $\psi(r) \sim exp(-\kappa L^\mu)$, with $\mu > 1$. In another experiment [6] with proton-doped polyaniline networks, it was found that the exponent γ increases (VRH, eqn. (1)) systematically from 0.25 to 1 upon decreasing the volume fraction p of the conducting component. They did also try the eqn.(2) to fit the low temperature data but that resulted in a large uncertainty in the exponent s. Such large values of $\gamma > 1/2$ gives us the first clue that just dilution or percolative aspects of a system may give rise to a continuously changing exponent with dilution.

In this peliminary work, our attempt would be to understand the behaviour of conductivity against temperature only from a percolative aspect. Some indications of dilution-dependence is also reported here. We use our recently proposed model [7] of lattice-based random resistor cum tunneling-bond network (RRTN) and examine whether the VRH [eqn.(1)] or its generalized form [eqn.(2)] may be fitted, given each that microscopic conducting/ tunneling element follow a known temperature behaviour.

2. The Model

The kind of RRTN we consider has proved to be very appropriate in studying the nonlinearity and the associated physics. We take a square lattice in 2D. Conducting bonds are thrown at random at a certain volume fraction p. The rest $(1-p)$ fractions are insulators. Now we allow tunneling through the nearest-neighbour (nn) gaps of two conducting bonds if a voltage is applied externally across the two opposite ends of such a network through two electrodes. As we treat the problem semiclassically, we may think of a tunneling bond (t-bond) sitting at

each such *nn* gap (unit lattice spacing) whose $i - v$ characteristic was considered to be piecewise linear for simplicity.

To evaluate the temperature-dependent conductivity of such a network we assume that all the t-bonds are active and that the whole macroscopic system is in an Ohmic regime. Given that all the t-bonds are conducting, we find that for infinite size systems the percolation threshold comes down from $p_c = 0.5$ for a RRN to $p_{ct} \sim 0.181$ for a RRTN [7] in 2D. We consider the volume fractions (p) which are between p_{ct} and p_c. The temperature (T) behaviour of each conducting (ohmic) bond is simply assumed to be $\sigma_{ohm} = a/T$, where a is constant. The t-bonds are assumed to have an activated behaviour: $\sigma_t = c \exp(-b/T)$, where b and c are also constants.

3. Results and Discussions

In all the works we consider below, we have chosen a=100, b=100, c=10 and several values of p from 0.18 to 0.5. As the system has dilution (and so disorder), the resistor elements corresponding to the ohmic and the tunneling bonds combine in a complex way in the tenuous current carrying portion of the network resulting in a non-trivial, non-monotonic conductance (in 2D this is also conductivity) as a function of the temperature T. As T increases, the conductance for the ohmic bonds decreases and that for the tunneling bonds increases. When the strength (conductance) of the two types of elements are comparable, the conductivity attains a peak value of σ_m at a $T = T_m$. A typical such behaviour is shown in Fig.1(a) for $p = 0.18$ and for different sizes of the squares from $L = 20$ to $L = 100$. In Fig.1(b), we show the same curves after scaling each axes by the peak values and find that all the graphs fall on top of each other. Because there are two opposing types of temperature behaviours in the two different types of bonds (ohmic and tunneling), it is certainly expected that there will be a crossover from some kind of exponential (in the low-T regime) to some kind of power law behaviour (in the asymptotic high-T regime) in the macroscopic sample. We had tried several plausible functional forms including both the VRH and the generalised VRH, but the generalized VRH law showed least fitting errors. This is consistent with the Aharony *et al.* [3] work. Thus we restrict ourselves to least-square fittings with eqn.(2).

First we found that it is necessary to fit the data for quite low temperatures compared to T_m. The overall fitting of the curves in Fig.1 upto about the peak temperature gives a $\gamma \simeq 1.2$. But, fitting only with the low temperature (for $7 \leq T \leq 20$) data shown in Fig.2 for $L = 20$ clearly indicates that $\gamma \simeq 0.64$ gives an excellent fit whereas $\gamma = 1.2$ gives a systematically bad fit to this section of the data. The low temperature fit further gives $s = 4.11$ and $T_0 = 1355$. Since the value of s seems to

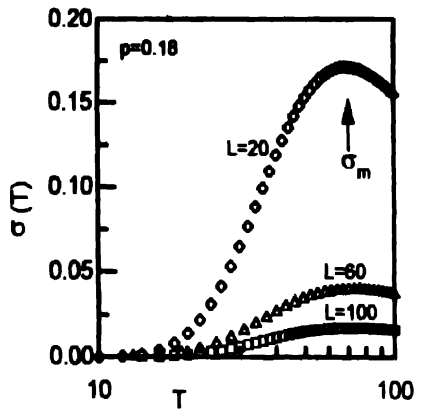

Fig. 1(a)

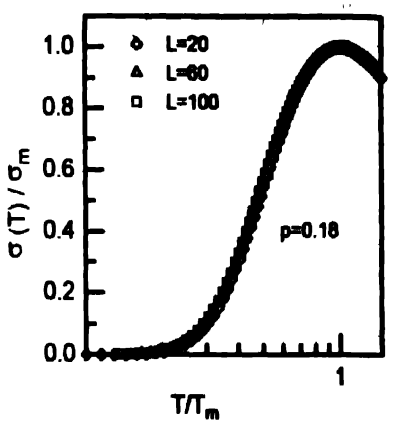

Fig. 1(b)

Fig.1(a): Behaviour of conductance ($\sigma(T)$) against temperature (T) for different system sizes (L) and for a fixed volume fraction (p). **(b)**: This figure shows the scaled conductance (σ/σ_m) against scaled temperature (T/T_m) for all curves in Fig.1(a). All the data points are shown to collapse for three different system sizes (L).

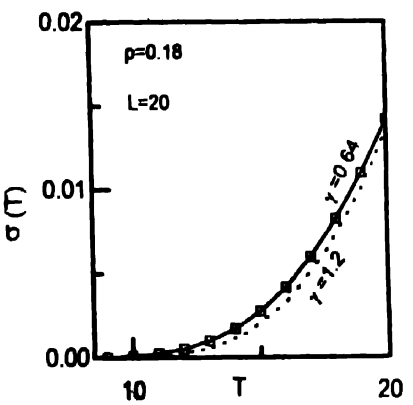

Fig. 2

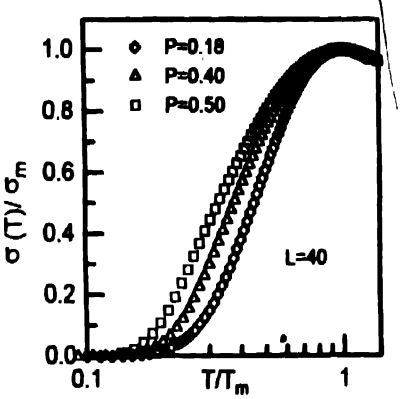

Fig. 3

Fig.2: Fitting of data is shown (for a limited region) for a typical $\sigma(T)$ against T curve by generalized VRH formula (eqn.(2) in the text). The solid line shows a better fit where the fitting is done for a small range of data in the low temperature (T) regime. The dotted line is a bad fit where the fitting is done for a large range of T (upto around the peak of the curve); only a portion of that is shown here.

Fig.3 This figure shows the scaled conductance (σ/σ_m) against scaled temperature (T/T_m) for three different volume fractions as indicated in the figure.

be somewhat large, we tried a replacement of the algebraic function in the prefactor by $\exp(T_0/T)^a$. But it did not work out as well as eqn.(2).

Next we looked at the results for different volume fractions. In Fig.3, we have shown the scaled conductivity versus temperature graphs for three different volume fractions p=0.18, 0.4 and 0.5. That the curves donot fall on top of each other indicate that the exponents are p-dependent. Indeed the trend of the graphs indicate that γ seems to be increasing with p. Further work in this direction is under progress.

Some words of caution may be appropriate here. If the fitting function employed is a perfect representation of the data, one expects a relation between T_0 and T_m which should also involve the volume fraction p and possibly other microstructural parameters. We did not find any clear relation between these quantities (even at a fixed p), by scanning all the cases we studied so far. This may imply that the function given in eqn.(2) does not seem to be an unique representation of the data. To check this we generated more data at as low temperatures as we can reach ($5 \leq T \leq 7$ for the parameter range chosen above) without compromising on our convergence factor for relaxation and remaining within reasonable amount of computer times. Our results show that inclusion of the lowest attainable temperature range, tends to change the exponents s and γ. Thus, a more complicated function than the one in eqn.(2) may be more appropriate. But in keeping with the literature, we restricted ourselves currently to the most general function proposed so far by Aharony *et al.*, as given in eqn.(2). We would like to point out here that it is not that only our results does not seem to be represented uniquely at all the temperature ranges by eqn.(2). Indeed, recent experiments [6, 8] cannot also fit this function uniquely; they seem to find large uncertainties in the exponent s and hence on γ, and this will certainly make the relation between T_0 and T_m very murky.

In summary then we have studied the low temperature behaviour in a random resistor cum tunneling-bond network (RRTN) and observed non-trivial generalized Mott-like variable range hopping behaviour whose characteristics seem to change with dilution. Further one needs to include the very low temperature regions of the data to obtain the idea for a reasonable fitting function for temperature- dependent hopping conductivity.

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