with the voids although with much larger scatter from sample to sample.[15]

In conclusion, we have shown that artificial cylindrical voids in a simple metal can induce a large linear magnetoresistance in quantitative agreement with classcial theory. The directional dependence shows clearly that indeed the influence of the voids has been studied, and not parasitic effects due to lattice imperfections or distortions.

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Fluctuation-Induced Tunneling Conduction in Carbon-Polyvinylchloride Composites

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We present evidence that in carbon-polyvinylchloride composites, consisting of aggregates of carbon spheres (100–400 Å) dispersed in the insulating matrix, the electrical conductivity can be ascribed to a novel mechanism of tunneling with potential-barrier modulation by thermal fluctuations. Theoretical consideration of the tunneling-probability modification by thermal fluctuating electric field across tunnel junctions yields expressions for the temperature and the field dependences of the conductivity in excellent accord with experimental results.

In recent years hopping conduction in disordered materials has recieved considerable attention as the mechanism responsible for the characteristic $\exp(-b/T^{\alpha})$ form of temperature dependence of conductivity observed in amorphous semiconductors[1] ($\alpha=\frac{1}{4}$) and sputtered granular metal films[2] ($\alpha=\frac{1}{2}$). There are, however, materials such as some conductor-insulator composites[3] and disordered semiconductors[4] for which no definitive theory has been proposed to explain their transport properties. In this Letter we report the observation of a new tunneling conduction mechanism for disordered materials in which the modulation of tunneling barriers by thermal fluctuations plays an important role in determining the dependence of the conductivity on temperature and electric field. In the following, the experimental results for the carbon-polyvinylchloride (PVC) composites are presented and compared with theoretical predictions based on the above mechanism. Application of the theory to other disordered systems will be published subsequently.

Carbon-PVC is a conductor-insulator composite consisting of carbon particles embedded in the insulating PVC matrix. Samples of this composite material were prepared by milling the compound together and injection molding. Three types of carbon were used in this study: (1) Ketjenblack[5] is a pigment composed of hollow spheres approximately 350 Å in diameter with 10–15-Å-thick walls[6]; (2) Columbia carbon[5] SA40-220 is similar to Ketjenblack but has a sphere diameter of 140 Å; (3) Mogul-L carbon[5] is a pigment containing carbon in the form of solid spheres with a diameter of about 200 Å. Transmission electron microscopy studies of all the specimens show the carbon to be highly structured.[6] That is, a carbon network is usually observed which is composed of touching or nearly touching micron-sized aggregates of graphitic spheres. Of the three types of carbon studied, Mogul-L showed some difference from the other two in exhibiting less of the tendency to form clumps and chains. As a result, samples with Mogul-L carbon generally show much higher resistivity than those with Ketjenblack or SA40-220.

To measure the resistivity ρ, samples were cut into disk-shaped pellets ~2 mm thick and 4 mm in diameter. Gold, evaporated on the faces, provided the electrical contacts. Both two-lead and four-lead resistivity measurements were performed. The results showed no perceivable difference between the two cases, indicating that contact resistance was not a problem and the two methods could be used interchangeably. The $\rho(T)$ data, plotted in Fig. 1, were taken at sufficiently low electric field that the samples were Ohmic. At higher voltages, self-heating dictated the use of voltage pulses. The high-field behavior of the current density j, shown in Fig. 2, was measured with 50- and 150-ns pulses at 60-Hz repetition rate.

Electrical conductivity of carbon-PVC composites results from percolation of electrons in the carbon networks. When the concentration of the carbon is large, the material exhibits graphitic conductivity,[7] indicating that the conduction network is continuous. As the concentration is decreased, conduction is dominated by electron tunneling across small barriers separating large conducting regions. A direct consequence of the large size of the conducting carbon aggregates is that the charging energy required to remove an electron from a neutral aggregate is completely negligible, in sharp distinction to conduction in granular metals[2,8,9] where charging energy plays the dominant role. Therefore, in the present case tunneling can be essentially regarded as between two bulk conductors. Let us consider one

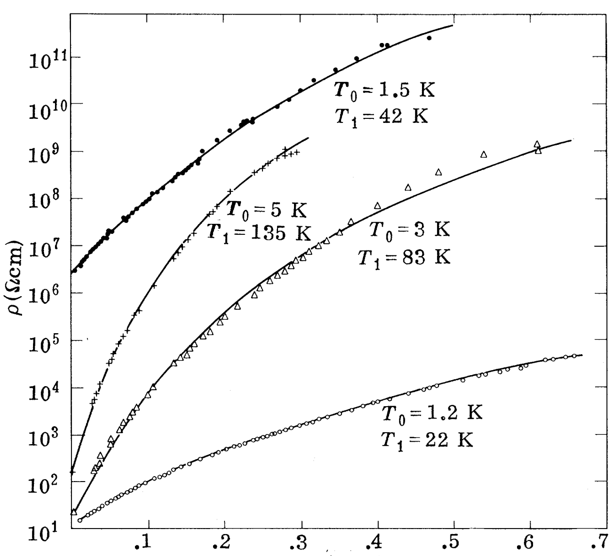

FIG. 1. Temperature dependence of the resistivity in carbon-PVC composites. Solid circles, Mogul-L, 45 wt.%; plusses, SA40-220, 15 wt.%; triangles, SA40-220, 20 wt.%; open circles, Ketjenblack, 16 wt.%. Solid curves are calculated from Eq. (6) with the values of T_0 and T_1 given beside each curve.

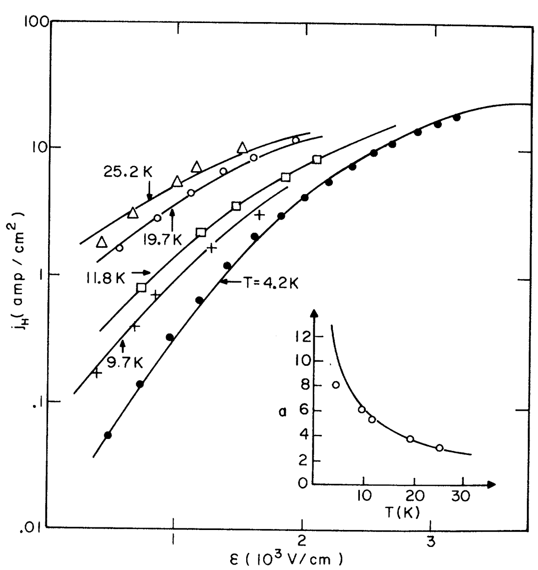

FIG. 2. High-field behavior of the current density in carbon-PVC composites. In this case the carbon is SA40-220, 20 wt.%. Solid lines are calculated from Eq. (7) with values of $a(T)$ plotted in the inset. The solid curve of the inset is given by Eq. (7b) with values of $T_0 = 3$ K and $T_1 = 83$ K obtained from the $\rho(T)$ data. Similar agreements between the theory and the experimental results are obtained for other samples.

of the tunnel barriers in the form of a plane parallel junction of area A and separation w. The barrier potential will be taken to be symmetrical (such as the image-force-corrected rectangular barrier) and therefore can be written in the form $V(x) = V_0 - \beta_2 x^2 - \beta_4 x^4 \cdots$, where $x = 0$ is defined as the center of the junction. For simplicity, in this Letter we will adopt the approximation of retaining only the x^2 term[10]:

$$V(x) = V_0 - (4V_0/w^2)x^2. \tag{1}$$

When an electric field ϵ is applied to the junction, the potential barrier is both lowered and narrowed. It is easily shown that in the WKB approximation the tunneling current density j has the form

$$j(\epsilon) = j_0 \exp\left[-\frac{\pi\chi w}{2}\left(\frac{|\epsilon|}{\epsilon_0} - 1\right)^2\right], \quad |\epsilon| < \epsilon_0, \tag{2}$$

where we have neglected the small backflow current; $\chi \equiv (2mV_0/\hbar^2)^{1/2}$ is the tunneling constant, m is the electron mass, $\epsilon_0 \equiv 4V_0/ew$, and j_0 is the pre-exponential factor which contains all the weak (nonexponential) temperature and electric field variations of j. It should be noted that for $|\epsilon| > \epsilon_0$, j is expected to have a slower, nonexponential dependence on ϵ.

There are two possible sources of the electric field. One is obviously the externally applied field ϵ_A. As for the second source, we observe that since the tunnel junction is situated in a thermal bath, there will inevitably be thermal fluctuations leading to voltage difference across the junction. In fact, it is well known[11] that the rms thermal fluctuation voltage of a junction is given by $(kT/C)^{1/2}$, where k is Boltzmann's constant and $C = A/4\pi w$ is the capacitance of the junction. When C is small, which is the case for the internal tunnel junctions of carbon-PVC composites, the thermal voltage fluctuations can be appreciable, resulting in large fluctuating electric field ϵ_T across the junction. Since the external field only has the effect of shifting the zero point of the thermal voltage fluctuations,[11] the total electric field across the junction at any given instant can be written as $\epsilon = \epsilon_A + \epsilon_T$.

A thermal fluctuating field can be either in the forward or the reverse direction. Therefore, for a fixed value of $|\epsilon_T|$ and a small applied field $\epsilon_A < |\epsilon_T|$, the net forward tunneling current is given by

$$\Delta j = j(\epsilon_A + \epsilon_T) - j(\epsilon_A - \epsilon_T). \tag{3}$$

A partial conductivity $\Sigma(\epsilon_T)$ can be defined in terms of Δj as

$$\Sigma(\epsilon_T) = \lim_{\epsilon_A \to 0} \frac{\Delta j}{\epsilon_A} = 2\,\frac{dj(\epsilon_T)}{d\epsilon_T}. \tag{4}$$

From $\Sigma(\epsilon_T)$ the conductivity of the junction σ is obtained by thermal averaging:

$$\sigma = \int_0^\infty P(\epsilon_T)\Sigma(\epsilon_T)\, d\epsilon_T. \tag{5}$$

Here the probability $P(\epsilon_T)$ for observing the field ϵ_T due to thermal fluctuations is given by[11] the Boltzmann factor $\exp(-u\epsilon_T^2/kT)$, and $u = wA/8\pi$ is a measure of the junction volume. Substitution of Eqs. (2) and (4) into Eq. (5) and the use of saddle-point integration yield

$$\sigma = \sigma_0 \exp[-T_1/(T + T_0)], \tag{6a}$$

$$T_1 = u\epsilon_0^2/k, \tag{6b}$$

$$T_0 = 2u\epsilon_0^2/\pi\chi w k, \tag{6c}$$

where in Eq. (6a) we will neglect the slowly varying temperature dependence of the pre-exponential factor and treat σ_0 as a constant. Equation (6) states that at $T \ll T_0$ the conductivity is temperature independent and reduces to the expected form $\sigma_0 \exp(-\pi\chi w/2)$ for tunneling through a parabolic barrier. However, at high temperatures the behavior becomes that of thermal activation[12] with activation energy $u\epsilon_0^2$. The high-field behavior of the junction can be similarly obtained. Neglecting the small flow of electrons counter to the applied field and using the saddle-point integration again, we have

$$j_H = \int_{-\epsilon_A}^{\infty} j(\epsilon_A + \epsilon_T)P(\epsilon_T)\, d\epsilon_T$$
$$\simeq j_0 \exp\{-a(T)[(\epsilon_A/\epsilon_0) - 1]^2\}, \tag{7a}$$

$$a(T) = T_1/(T + T_0). \tag{7b}$$

So far we have only been concerned with the behaviors of a single junction. In order to obtain the conductivity of the composite, it is necessary to consider a network of independently fluctuating[13] tunnel junctions with different values of T_1 and T_0. However, it can be shown[14] that with reasonable distribution functions for T_1 and T_0, the conductivity of the network is well described by single-junction characteristics. Therefore, in Figs. 1 and 2 we will directly compare the experimental results with the predictions of Eqs. (6) and (7). Figure 1 gives the temperature-dependent resistivity ρ for four samples. The $\rho(T)$ data are seen to be in excellent agreement with the behavior predicted by Eq. (6). The values of T_1 and T_0 used are given beside each curve. The knowledge of T_1 and T_0, though insufficient for the unique determination of V_0, w, and A, can nevertheless provide plausible estimates of their values. For example, a possible set of values for the junction parameters consistent with $T_1 = 83$ K and $T_0 = 3$ K is $V_0 \simeq 0.2$ eV, $w \simeq 75$ Å, and $A \simeq 250$ Å^2, where we have used the free-electron mass in the calculation of χ. In Fig. 1 we also note that two samples with different weight fractions of SA40-220 carbon have different values of T_1 and T_0. Such difference reflects the expected (and as-yet-undetermined) dependence of w and A on the composition of the composite. The high-field behavior is shown in Fig. 2 for the sample with 20 wt.% of SA40-220 carbon. The solid lines are calculated from Eq. (7) with $j_0 = 25$ A/cm^2 and $\epsilon_0 = 3750$ V/cm. It is seen that the experimental values of j at different temperatures tend to saturate at some fixed value as predicted by the theory. The ϵ dependence of j is well described by the form $\exp[-a(T)(\epsilon_A/\epsilon_0 - 1)^2]$ given by Eq. (7). The value of a (T) used in each fit to the experimental data is plotted in the inset. The solid curve in the inset is calculated from Eq. (7b) with the values of $T_1 = 83$ K and $T_0 = 3$ K obtained from the temperature dependence of the resistivity as shown in Fig. 1. The good agreement between the experimental values of $a(T)$ and the calculated curve with no adjustable parameter demonstrates the underlying unity of the $\rho(T)$ and the $j(\epsilon)$ behaviors.

It should be remarked that in Fig. 2 the non-Ohmic behavior for the carbon-PVC composites occurs at the very low field strength of 500–4000 V/cm. This is consistent with the material picture mentioned earlier that the tunnel junctions are separated by large conducting regions. Since the applied voltage will be mainly concentrated at the junctions, the actual field strength at the junction is expected to be larger than the applied value by a factor M, where M can be interpreted as the ratio between the average size of the conducting aggregate and the average junction width. By applying the definition of $\epsilon_0 = 4V_0/ew$ and taking $V_0 = 0.2$ eV and $w = 75$ Å, we get $\epsilon_0 \cong 10^6$ V/cm. When this value is compared to $\epsilon_0 = 3750$ V/cm used in fitting the experimental results, we get $M \simeq 300$. Therefore, the size of the conducting aggregate is about $300w \simeq 2$ μm, in order-of-magnitude agreement with the value observed in transmission electron micrographs.

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[12]Thermal activation over the potential barrier is a conduction process parallel to tunneling through the barrier. It can be shown that thermal fluctuations have the effect of lowering the activation energy from V_0 to $V_0' = V_0/[1 + (V_0/u\epsilon_0^2)]$. When $V_0 \gg u\epsilon_0^2$, we have $V_0' \simeq u\epsilon_0^2$. In this Letter the contribution of thermal activation conduction is disregarded since it is negligible at low temperatures and gives behavior indistinguishable from that of Eq. (6) at high temperatures.

[13]Fluctuations at different junctions should be independent since the correlation length for local electron-density fluctuations in the conducting region is expected to be much smaller than the separations between different junctions.

[14]By regarding each tunnel junctions as a conductor, the conductivity of the network can be obtained by the effective-medium theory [see S. Kirkpatrick, Rev. Mod. Phys. 45, 574 (1973); J. Bernasconi, Phys. Rev. B 1, 2252 (1973)]. Results of the effective-medium analysis will be published elsewhere.