# Metal-insulator transition and effects of localization and correlation in amorphous $Fe_xSb_{100-x}$ alloys

Gang Xiao and C. L. Chien
*Department of Physics and Astronomy, The Johns Hopkins University, Baltimore, Maryland 21218*
(Received 3 July 1986)

Study of the electrical resistivity of amorphous $Fe_xSb_{100-x}$ alloys with a very wide composition range of $0 \leq x \leq 85$ reveals a metal-insulator transition near $x=14$. Conduction via variable-range hopping takes place on the insulating side. The low-temperature resistivity on the metallic side has a $\sqrt{T}$ dependence, in agreement with recent scaling theories. In some metallic samples, the effects of localization and correlation dominate the electrical conductivity up to 30 K. The correlation gap, which vanishes at the metal-insulator transition, increases monotonically on both the metallic and the insulating side with a change of five orders of magnitude over the entire composition range.

The electrical resistivity of amorphous metallic alloys[1] is generally characterized by high values of resistivity ($>100\ \mu\Omega$ cm), small fractional changes over a large temperature range, negative temperature coefficients, and low-temperature anomalies such as resistivity minimum and divergent behavior. Although various models have been proposed to account for the transport properties, the validity of some has been seriously questioned, particularly those that implicitly assume a Boltzmann transport mechanism.[2] Recent theories using localization treatment represent an important approach which offers more insights into the transport properties of amorphous materials. A closely related subject is the occurrence of metal-insulator ($M$-$I$) transition in amorphous systems involving metallic and nonmetallic species. McMillan[3] has developed a scaling theory of metal-insulator transition in amorphous materials, which incorporated the concepts of disorder-induced localization, electron-electron interaction, and screening effects. There are specific predictions for the temperature dependence of resistivity on the metallic side, density of states, electron correlation gap, and other aspects.[3,4] The ideal amorphous materials suitable for testing the theory are those systems with wide composition range and having a $M$-$I$ transition. So far, the amorphous systems which have been used to study $M$-$I$ transition and effects of localization and correlation are mainly conventional semiconducting elements of Si or Ge alloying with metallic elements (Au, Nb, and Al) in a narrow composition range (10—20 %) around the $M$-$I$ transition, and most studies have been concentrated on the metallic side.

In this work, we present the study of electrical resistivity in the amorphous $Fe_xSb_{100-x}$ system over a wide composition range of $0 \leq x \leq 85$ in which a $M$-$I$ transition has been observed. Systematic investigations were carried out on both the insulating and metallic sides. The effects of localization and correlation are clearly distinguishable and can be followed as the composition is continuously varied. In some metallic samples, such effects dominate the electrical conductivity to as high as 30 K, which, to our best knowledge, is the widest temperature range ever reported for the effects of localization and correlation to appear in amorphous alloys. One important result is that the correlation gap, which opens in the one-electron density of states, occurrs on *both* the metallic and the insulating sides, but tends to vanish at the $M$-$I$ transition.

While crystalline pure antimony ($c$-Sb) is a semimetal with a small band overlap, amorphous pure antimony ($a$-Sb), as made by vapor quenching with a 77 K substrate temperature, turns out to be a semiconductor[5] with a pseudogap as large as that of $a$-Ge. This is solely caused by the structural difference between $c$-Sb and $a$-Sb. The conductivity of $a$-Sb can be described by Mott's theory of variable-range hopping,[5] where the phonon-assisted quantum tunneling takes place between the localized states at the Fermi level ($E_F$). When metallic elements are gradually introduced into an amorphous semiconductor, the pseudogap will eventually disappear. The electronic states become extended as the alloy enters into the metallic region. Due to the highly disordered nature of the metallic glass, localization[6,7] and electron correlation effects[6,8] emerge at low temperatures even in the metallic region.

The Fe-Sb alloy samples, of thicknesses about 1—3 $\mu$m, were all obtained by sputtering from composite targets onto liquid-nitrogen-cooled substrates. The background vacuum pressure was in the ($10^{-7}$—$10^{-8}$)-Torr range and the Ar pressure during the sputtering was 4 mTorr. Such vapor-quench techniques are capable of making amorphous alloys over wide composition range which is essential in our study. The amorphous nature of each sample was confirmed by x-ray diffraction. The resistance of the samples was determined by a standard four-probe method via a computer-controlled data-acquisition system. The high resolution in resistivity and temperature of the system yields very compact data, forming quasicontinuous curves. Detailed studies of the hyperfine interactions and magnetic properties of the $a$-Fe-Sb system have been described elsewhere.[9]

The temperature variation of the normalized resistivity in the Sb-rich region ($x \leq 20$) is shown in Fig. 1(a). It is immediately apparent that a $M$-$I$ transition occurs in the narrow range of $12 < x < 15$. The resistivity of the samples with $x \leq 12$ diverges as $T$ approaches zero, whereas those with $x \geq 15$ appear to have a finite resistivity to-

 

wards $T=0$. The critical metal composition $x_c=14\%$, at which the $M$-$I$ transition occurs in $a$-Fe-Sb, is slightly higher than $x_c=8\%$ for $Au_xGe_{100-x}$ (Ref. 10) and $x_c=11.5\%$ for $Nb_xSi_{100-x}$ (Ref. 11), but much lower than $x_c=50\%$ claimed for $a$-$Al_xGe_{100-x}$.[12] On the insulating side, variable-range hopping occurs. The metal-insulator transition here is, in the conventional sense, an Anderson transition[6,7] in which the disorder-induced localization occurs below $E_F$.

According to Mott *et al.*[13–15] the variable-range hopping resistivity has the following temperature dependence

$$\rho=\rho_0 e^{(T_0/T)^{1/4}}, \tag{1}$$

where

$$T_0=16\alpha^3/N(E_F)k_B\ . \tag{2}$$

In relation (2), $k_B$ is Boltzmann's constant, $\alpha$ is the coefficient of the exponential decay of the localized state, and $N(E_F)$ is the density of localized states at the Fermi level. Our data of various insulating samples are shown in Fig. 2(a) where plots of $\ln R$ versus $T^{-1/4}$ are displayed. The data can be well described by relation (1) from the lowest temperature of about 2.3 K to a crossover temperature ($T_{cr}$), indicated by the arrows, where the resistance deviates from Eq. (1). The slope of the linear portion of Fig. 2(a) is simply the value of $T_0^{1/4}$, which decreases rapidly with the addition of Fe. The compositional dependence of the constant $T_0$ is presented in Fig. 2(b). The decrease of $T_0$ is due primarily to the increase of $N(E_F)$ as the system approaches the $M$-$I$ transition. The localization

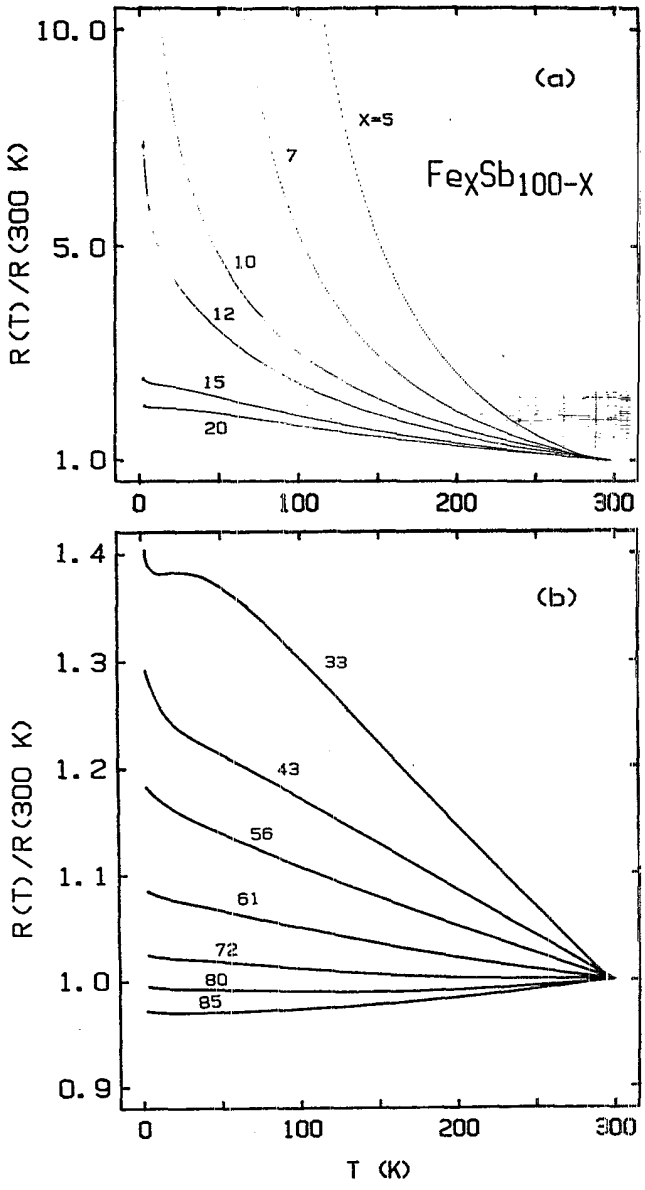


FIG. 1. Normalized resistivity of $a$-$Fe_xSb_{100-x}$ with temperature. (a) $5\leq x\leq 20$, (b) $33\leq x\leq 85$.

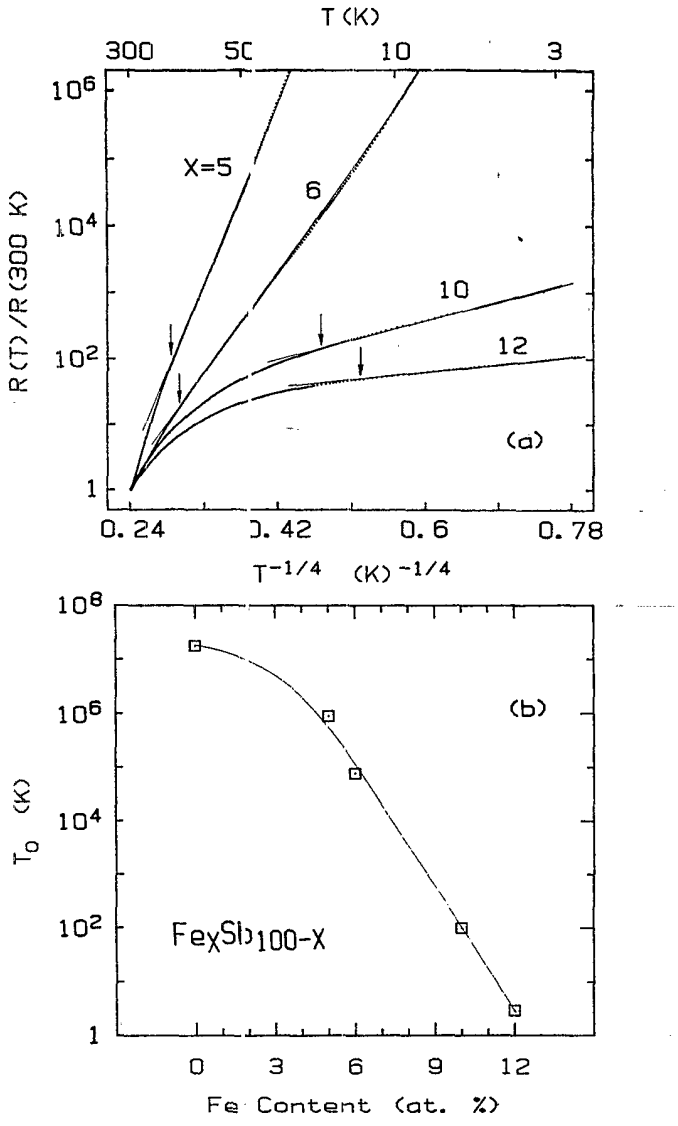


FIG. 2. (a) Temperature dependence of the insulating $a$-$Fe_xSb_{100-x}$ samples ($5\leq x\leq 12$), shown as $\ln R$ vs $T^{-1/4}$. The arrows indicate the crossover temperatures ($T_{cr}$ see text). (b) Compositional dependence of the constant $T_0$ defined in text.

length ($\alpha^{-1}$) is fairly small (e.g., $\sim 10$ Å and slowly varying at the insulating end. According to McMillan's scaling theory,[3] $\alpha^{-1}$ is expected to increase near the $M$-$I$ transition, further reducing $T_0$, and diverge at the transition. Our results in the insulating region can be summarized as follows. In $a$-Sb there exists a large pseudogap in which impurities such as Fe produce deep energy levels. These states are localized near the $E_F$ as evident from the $T^{-1/4}$ dependence. However, the pseudogap in $a$-Sb gradually disappears as Fe concentration increases. Near the $M$-$I$ transition in Fe-Sb, the density of localized states at $E_F$ is so high that the Anderson localization collapses, since generally only the lower density-of-states region is more susceptible to localization.[16]

Mott's variable-range hopping model incorporates the disorder effects on the electrical resistivity of amorphous insulator, and the dependence of $\exp(T^{-1/4})$ was derived assuming that the density of states is constant in the vicinity of the Fermi level. However, according to some theoretical[3,4,17,18] and experimental[19] observations, electron correlation effects also exist in amorphous insulator. Efros and Shklovskii[17,18] have studied the Coulomb interaction between localized electrons, and concluded that a Coulomb gap will appear in the density of states. This leads to a modification of the temperature dependence of resistivity in amorphous insulator to $\exp(T^{-1/2})$. We have also tried to fit our data to the $\exp(T^{-1/2})$ dependence, but overall, the Mott model provides far better fitting. At low Fe concentrations, the data can be fitted by the Mott model over a large temperature range. Even as the Fe concentration approaches the $M$-$I$ transition, where the fitting is limited to low temperatures, the applicable temperature range is still much wider for the Mott model. For example, we observed the $\exp(T^{-1/4})$ dependence in $Fe_{10}Sb_{90}$ below $T=20$ K, while the $\exp(T^{-1/2})$ fitting can at best be marginally argued at below $T=6$ K. The Coulomb gap in the density of states in the Efros-Shklovskii model is probably not realized in our system. Instead we expect a band tail below the Coulomb gap,[3] which makes the Mott model more appropriate.

We now turn to the metallic region for which the temperature dependence of the normalized resistivity is shown in Fig. 1(b). All metallic samples except $x=85$ show a negative temperature coefficient of resistivity, typical of many metallic glasses with high resistivity [$\rho(300\ \text{K}) \sim 150\ \mu\Omega\,\text{cm}$].[20] Most significantly, there appears a resistivity upturn in every metallic sample at low temperatures. In many metallic glasses, particularly those containing magnetic species, the low-temperature resistivity often shows a minimum and a $\ln T$ dependence. This has often been attributed to the Kondo effect.[21] In contradiction, however, in almost all amorphous systems the resistivity minimum and the $\ln T$ slope are independent of the applied magnetic field and insensitive to ferromagnetic orderings.[1,22] An alternative model has been proposed by Cochrane *et al.*[22] using the "two-level system" as a source of electron scattering. However this model suffers no less special difficulties than the Kondo model.[23] Overall, no consistent description is offered by either of these two models. In our Fe-Sb system $\ln T$ behavior is absent (see the inset of Fig. 3).

Electron localization occurs in systems with sufficient amount of disorder. Treating both localization and correlation effects on equal footing, McMillan's scaling theory of $M$-$I$ transition in amorphous materials predicts the density of states on the metallic side of the $M$-$I$ transition as

$$N(E)=N(0)[1+(E/\Delta)^{1/2}]\,, \tag{3}$$

where $\Delta$ is the electron correlation gap. The conductivity mechanism at low temperatures is the quantum diffusion of quasiparticles, leading to

$$\frac{\rho(T)-\rho(0)}{\rho(0)} = -\left[\frac{k_B T}{\Delta}\right]^{1/2}. \tag{4}$$

The correlation gap $\Delta$ shows a universal quadratic power-law dependence on resistivity

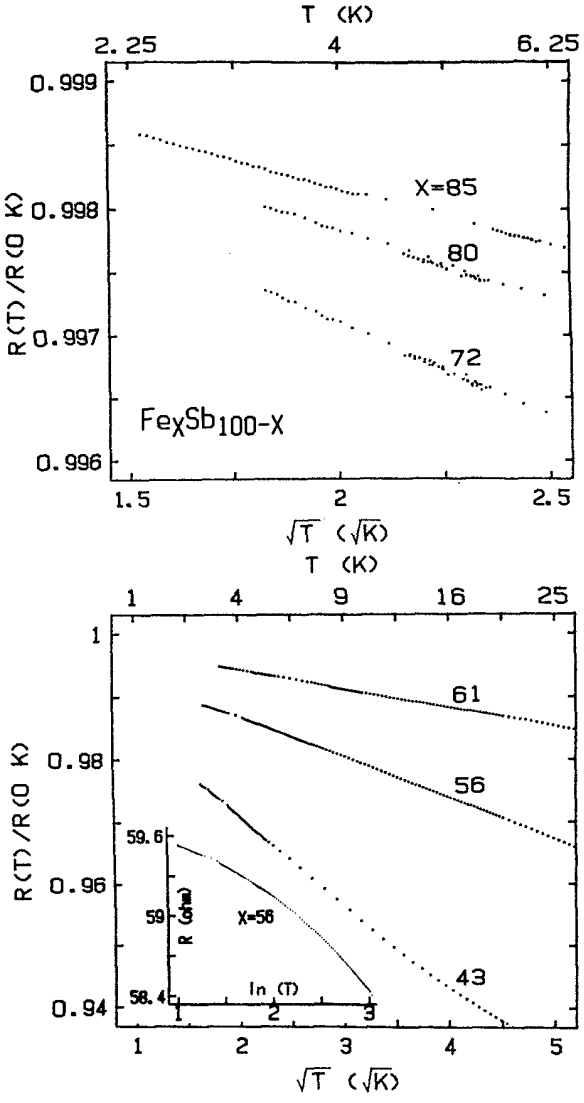


FIG. 3. Low-temperature resistivity of the metallic $a$-$Fe_xSb_{100-x}$ samples ($43 \le x \le 85$) as a function of $\sqrt{T}$. The inset shows the resistivity of $a$-$Fe_{56}Sb_{44}$ as a function of $\ln T$.

$$\Delta \sim [\rho(0)]^{-2} . \quad (5)$$

The $\sqrt{T}$ dependence of resistivity was first observed and explained in Si:P by Rosenbaum, Andres, Thomas, and Lee,[24] and in amorphous metal ($Fe_{40}Ni_{40}P_{14}B_6$) by Rapp, Bhagat, and Gudmundsson.[25] In our $a$-Fe-Sb system such dependence of resistivity is shown in Fig. 3. The $\sqrt{T}$ dependence is found over a wide composition range ($33 \leq x \leq 85$) and in some samples up to about 30 K. The appearance of localization and correlation effects over such a wide metallic range and temperature range is rather unique to the $a$-Fe-Sb system. In other amorphous materials reported, the $\sqrt{T}$ dependence of resistivity was generally observed at very low temperatures (below 10 K). A wide temperature range over which the effects of localization and correlation appear will facilitate the study of the interplay between localization and other physical mechanism such as superconductivity. Amorphous Sb alloyed with other suitable metallic elements may very well be good candidates for such study. For the metallic samples of $a$-Fe-Sb ($x < 33$) near the $M$-$I$ transition, the $\sqrt{T}$ dependence is not readily evident in our temperature range, and much lower temperatures are needed to observe the effects. As shown in the inset of Fig. 3, $\ln T$ gives a poor description of the data.

The determined correlation gaps ($\Delta$) are plotted against the Fe concentration in Fig. 4. There is a three orders of magnitude change in the correlation gap from $10^{-1}$ eV to 100 eV in the metallic region of $33 \leq x \leq 85$. Near the $M$-$I$ transition ($x < 33$), $\Delta$ is supposedly much smaller but cannot be determined in our measuring temperature range as mentioned above. A large spectrum of correlation gaps was also uncovered from recent tunnelling measurements in three other systems [granular Al (Ref. 26), $a$-NbSi (Ref. 11), and $a$-AlGe (Ref. 12)]. Cochrane and Strom-Olsen[27] reviewed a dozen individual metallic glasses and found $\Delta$ ranging from 50 to 1000 eV.

On the insulator side of the $M$-$I$ transition the correlation gap *also* exists, but it is more difficult to determine unambiguously. McMillan[3] suggested that the crossover temperature ($T_{cr}$) from hopping mechanism to other algebraic dependence at high temperature is the characteristic energy of the correlation gap, i.e., $k_B T_{cr} \sim \Delta$. Accordingly, $T_{cr}$ and therefore $\Delta$ can be obtained from Fig. 1. This enables us to obtain $\Delta$ over the *entire* composition range, metallic as well as insulating. The value of $\Delta$ spans five orders of magnitude. The decrease of $\Delta$ as the $M$-$I$ transition is approached from both sides is in good agreement with the predictions of McMillan's theory; that is, $\Delta$ collapses at the $M$-$I$ transition as the screening length diverges, and on both sides of the $M$-$I$ transition a correlation gap opens up in the one-electron density of states. Since Mott transition is characterized by a correlation gap, such a $M$-$I$ transition is therefore of the character of Mott transition as well as Anderson transition. McMillan's equal treatment of localization and correlation is well justified.

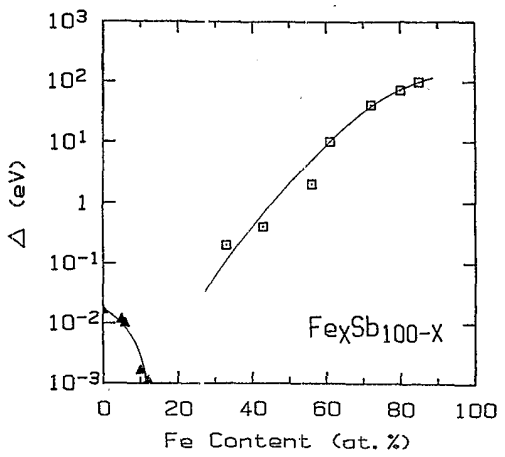


FIG. 4. Correlation gap ($\Delta$) of $a$-$Fe_xSb_{100-x}$ as a function of Fe content. Triangles and squares denote, respectively, insulating and metallic samples.

In conclusion, a metal-insulator transition has been observed in the amorphous Fe-Sb alloy system. In the insulating region, variable-range hopping is dominant at low temperatures. Going into the metallic region, the low-temperature resistivity shows a $\sqrt{T}$ dependence. Localization and correlation effects, varying in degree, appear throughout the entire composition range of the amorphous alloys. The correlation gap $\Delta$, which vanishes at the metal-insulator transition, increases monotonically on both the metallic and the insulating side. These are in agreement with the theoretical predictions, particularly those of McMillan's theory.

This work is supported by National Science Foundation (NSF) Grant No. DMR-86-07150.

[1]For reviews of electrical transport properties of amorphous materials, see, e.g., D. G. Naugle, J. Phys. Chem. Solids **45**, 367 (1984); K. V. Rao, in *Amorphous Metallic Alloys,* edited by F. E. Luborsky (Butterworths, London, 1983), pp. 401—431; R. Harris and J. O. Strom-Olsen, in *Glassy Metal II,* Vol. 53 of *Topics in Applied Physics,* edited by H. Beck and H. J. Guntherodt (Springer, Berlin, 1983), pp. 325—342.

[2]P. A. Lee and T. V. Ramakrishnan Rev. Mod. Phys. **57**, 287 (1985).

[3]W. L. McMillan, Phys. Rev. B **24**, 2739 (1981).

[4]B. L. Al'tschuler and A. G. Aronov, Zh. Eksp. Teor. Fiz. **77**, 2028 (1979) [Sov. Phys.—JETP **50**, 968 (1979)].

[5]J. J. Hauser, Phys. Rev. B **9**, 2623 (1974); **11**, 738 (1975).

[6]N. F. Mott, *Metal-Insulator Transitions* (Taylor and Francis, London, 1974).

[7]P. W. Anderson, Phys. Rev. **109**, 1492 (1958).

[8]N. F. Mott, Proc. Phys. Soc. London, Sect. A **62**, 416 (1949).

[9]C. L. Chien, Gang Xiao, and Karl Unruh, Phys. Rev. B **32**, 5582 (1985); Gang Xiao and C. L. Chien, J. Magn. Magn. Mater. **54-57**, 241 (1986).

[10]B. W. Dodson, W. L. McMillan, J. M. Mochel, and R. C. Dynes, Phys. Rev. Lett. **46**, 46 (1981).

[11]G. Hertel, D. J. Bishop, E. G. Spencer, J. M. Rowell, and R. C. Dynes, Phys. Rev. Lett. **50**, 743 (1983).
[12]J. Lesueur, L. Dumoulin, and P. Nedellec, Phys. Rev. Lett. **55**, 2355 (1985).
[13]N. F. Mott, J. Non-Cryst. Solids **1**, 1 (1968).
[14]N. F. Mott, Philos. Mag. **19**, 835 (1969).
[15]N. F. Mott and E. A. Davis, *Electronic Processes in Non-Crystalline Materials* (Clarendon, Oxford, 1979).
[16]D. J. Thouless, Phys. Rep. **13C**, 93 (1974).
[17]A. L. Efros and B. I. Shklovskii, J. Phys. C **8**, L49 (1975).
[18]A. L. Efros, J. Phys. C **9**, 2021 (1976).
[19]W. L. McMillan and Jack Mochel, Phys. Rev. Lett. **46**, 556 (1981).
[20]J. H. Mooij, Phys. Status. Solidi A **17**, 521 (1973).
[21]G. S. Grest and S. R. Nagel, Phys. Rev. B **19**, 3571 (1979).
[22]R. W. Cochrane, R. Harris, J. O. Strom-Olsen, and M. J. Zuckermann, Phys. Rev. Lett. **35**, 676 (1975).
[23]J. L. Black and B. L. Gyorffy, Phys. Rev. Lett. **41**, 1595 (1978); J. L. Black, K. Vladar, and A. Zawadowski, Phys. Rev. B **26**, 1559 (1982).
[24]T. F. Rosenbaum, K. Andres, G. A. Thomas, and P. A. Lee, Phys. Rev. Lett. **46**, 568 (1981).
[25]O. Rapp, S. M. Bhagat, and H. Gudmundsson, Solid State Commun. **42**, 741 (1982).
[26]R. C. Dynes, and J. P. Garno, Phys. Rev. Lett. **46**, 137 (1981).
[27]R. W. Cochrane and J. O. Strom-Olsen, Phys. Rev. B **29**, 1088 (1984).