

Metal-insulator transitions

Changes in temperature, pressure, magnetic field or alloy composition can affect the electronic band structure of substances, in some cases leading to dramatic changes in conductivity.

Sir Nevill Mott

Research on metal-insulator transitions is at the moment quite fashionable. In many materials the electrical behavior changes from metallic to nonmetallic when the pressure, temperature or magnetic field is varied or (as in alloys) when the composition is varied, and the theoretical description of these processes is quite complicated. The interest of the problem lies perhaps mainly in our imperfect understanding of the nature of a metal. In the days before quantum mechanics, when I first attended undergraduate lectures on the electron theory of solids, it was taught that in metals one or more atoms from each electron were free, whereas in nonmetals they were somehow fixed to the atoms or ions or to the chemical bonds. The long mean free paths of electrons in metals extending over hundreds or thousands of atomic spacings were not understood, and neither was the absence of any large contribution from the electrons to the specific heat.

With the discovery of quantum mechanics and Fermi-Dirac statistics the problem of the specific heat was almost immediately cleared up. As for the mean free path, it was shown that an electron wave could propagate without scattering in a perfect rigid lattice and the free path was therefore a consequence of impurities, defects or lattice vibrations. Then came the theories of Felix Bloch, Rudolf Peierls and Alan H. Wilson, which showed the origin of the difference between insulators, metals and semiconductors and have so greatly influenced the development of solid-state electronics. According to these theories, particularly in the papers of Wilson, the electrons in an insulator are not "stuck" but are considered in a first approximation as free. However, their

energy states in a periodic lattice fall into bands, and in an insulator the highest occupied band (the valence band) is completely full, so that (in the absence of thermal or optical excitation) exactly as many electrons are moving in one direction as in the opposite one. The valence band is separated from the empty conduction band by a "gap," small in intrinsic semiconductors so that electrons can be thermally excited across it, but greater than about 3 eV in a transparent material.

This model has proved itself extremely satisfactory for the description of most crystalline semiconductors and metals, for many of which the band forms are known in detail. It is perhaps remarkable that this is so, because the model neglects the interaction between electrons, except in the sense of the Hartree-Fock average, and this cannot be a small perturbation. Of course this interaction has been taken into account in more sophisticated work, particularly in describing magnetic properties and, as we shall see, it leads to one form of metal-insulator transition.

A branch of physics, however, in which the Wilson formulation has proved less satisfactory is that of the behavior of electrons in noncrystalline materials, particularly glasses. Since the band-gap itself depends on Bragg reflection of an electron wave from a crystalline lattice, which cannot occur in glasses, it is not clear how the gap occurs, but the mere fact that many glasses are transparent shows that it is present. In fact, for understanding some of the properties of glasses, it has proved convenient to go back to pre-quantum mechanical ideas and to say that the electrons are stuck in bonds. This is because, as first shown by B. T. Kolomiets's school in Leningrad some 20 years ago, the semiconducting glass such as arsenic telluride (As_2Te_3) cannot be doped with impurities. If

germanium is added it does not go into a substitutional position replacing three-fold coordinated arsenic with one electron loosely bound, as must probably occur in a crystal; on the contrary it forms four bonds with its neighbors, so that no electron can escape easily into the conduction band. For this reason most amorphous semiconductors show a striking increase in the conductivity on crystallization.

These facts suggest that an account of metal-insulator transitions should include noncrystalline as well as crystalline systems, and this will be attempted here. We shall not follow the historical development of the subject, but will rather start with the behavior of electrons in noncrystalline systems. Our understanding of the subject goes back to Philip Anderson's¹ paper, published in 1958, on "Absence of diffusion in certain random lattices" and to earlier experimental work by Hellmut Fritzsche and others on impurity conduction. Our problem is that of a *degenerate* electron gas, with states filled up to a limiting Fermi energy E_F , which does not lie in a gap but on the contrary is at an energy where the density of states, denoted by $N(E_F)$, is finite. In a crystalline system such a material would be metallic, by which we mean that the conductivity σ would tend to a finite value (dependent on purity) as the temperature tends to zero. In a noncrystalline material this is not necessarily so, and σ may tend to zero, which is what we mean by "insulating behavior." An advantage of treating transitions in nonperiodic fields first here is that a qualitatively correct description can be given in terms of non-interacting electrons, with models exactly parallel to those of the 1930's. The electron gas is described by one-electron wave functions ψ_E , solutions of the Schrödinger equation

$$\nabla^2\psi + \frac{2m}{\hbar^2}[E - V(x,y,z)]\psi = 0 \quad (1)$$

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with electron states filled up to the energy E_F and $V(x,y,z)$ the non-periodic potential. For crystalline materials, on the other hand, electron-electron interaction plays an essential role in all kinds of metal-insulator transitions.

Noncrystalline systems

Anderson considered the solutions of equation 1 for a crystalline array of potential wells, in any one of which (denoted by n) the lowest 1s state is written $\phi_n(r)$. If all the wells had the same depth, and the wells were far enough apart for the tight-binding approximation to be used, an electron could move freely through the array with a wave number k , with a wave function of the type

$$\psi_k = e^{ikx} u_k(x,y,z) \quad (2)$$

and a bandwidth B given by

$$B = 2zI, \quad (3)$$

where z is the co-ordination number and I the energy transfer integral

$$I = \int \phi_n H \phi_{n+1} d^3x$$

Anderson however considered wells with a range of depth ΔV . If $\Delta V/B$ is small, the effect is to introduce, for energies near the middle of the band, a finite mean free path L given (in perturbation theory) by

$$a/L = 0.7(\Delta V/B)^2 \quad (4)$$

where a is the distance between the wells. So if $\Delta V/B$ approximately equals 1, L is of order a and, according to a principle enunciated by Ioffe and Regel, it cannot be shorter than that. If $\Delta V/B$ is greater than a certain quantity, which is difficult to calculate but later work shows to be probably about 2 (for $z = 6$), Anderson showed that all states in the band become localized, with quantized energy values. No diffusion or conductivity is therefore

possible without thermal activation.

In 1967 I first pointed out that if $\Delta V/B$ is less than this critical value, an energy E_c will exist such that states below it are localized and states above it "extended." The energy E_c is often called a "mobility edge." The expected density of states is shown in figure 1. For energies E just above E_c , the "tight binding" wave function is thought to have the form

$$\psi = \sum e^{i\eta_n} \phi_n \quad (5)$$

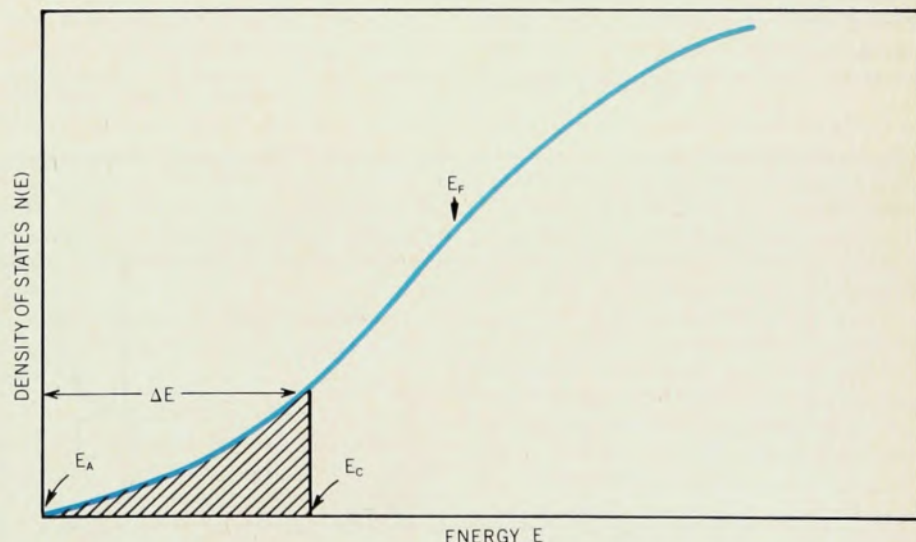
where the η_n are random phases. For energies just below it,

$$\psi = e^{-\alpha(r-r_0)} \sum e^{i\eta_n} \phi_n \quad (6)$$

where r_0 is the point in space where the wave function is centered and tends to zero as E approaches E_c , probably as $\alpha = \text{const}(E_c - E)^{2/3}$. There has been some

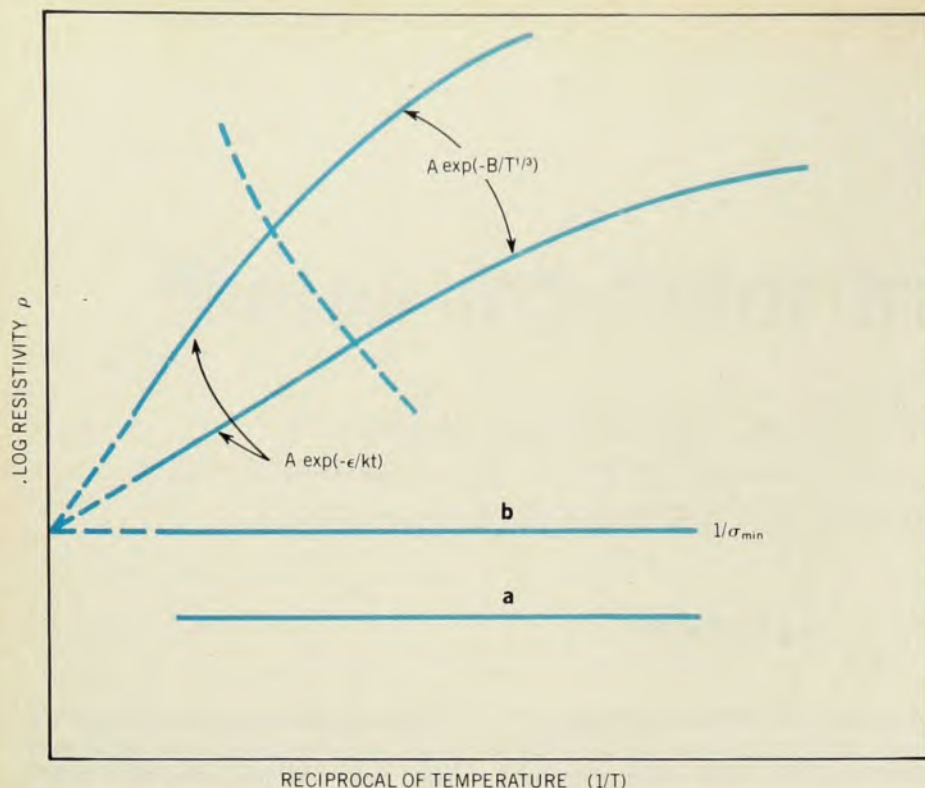
dispute as to whether localized and non-localized states can exist in the same energy range; in my view they cannot. Moreover, although calculations of the mobility edge have been made for only a few kinds of random potential, it seems a reasonable assumption that the behavior illustrated in figure 1 is quite general—although in the conduction bands of some glasses such as vitreous SiO_2 the magnitude of ΔE seems too small to affect appreciably the mobility of electrons there.

If a conduction band containing a degenerate electron gas has the form of figure 1, and if either the number of electrons (determining the Fermi energy) or the mobility edge or both can be varied, a "metal-insulator transition" may occur. If E_F lies above E_c , as in figure 1, the material is metallic, in the sense that σ tends



Density of states $N(E)$ in a noncrystalline material. Here E_c is the mobility edge, and energies for which states are localized are shaded. The Fermi energy E_F is marked for the case where conduction is metallic; if E_F lies below E_c , the conductivity tends to zero with T .

Figure 1



Resistivity of a system whose conduction band is illustrated in figure 1 as the Fermi energy moves across the mobility edge. The metal-insulator transition occurring has been called an "Anderson transition." Here ϵ denotes $E_c - E_F$. The factor $T^{1/3}$ is for two-dimensional systems; the dependence is $T^{1/4}$ in three dimensions.

Figure 2

to a finite value as $T \rightarrow 0$; it will probably vary little with temperature, because the scattering due to disorder will be greater than that due to phonons. If E_F lies below E_c , σ tends to zero with T .

The behavior for the two cases is illustrated in figure 2. In curve a, E_F lies above E_c . In curve b it lies at E_c , and in the other two curves below E_c . The conductivity in case b, in the limit of low temperatures, has the value that I have called the "minimum metallic conductivity" and denoted by $\sigma_{\min}$. This is, in principle, the value of the conductivity of a metal when the mean free path L has its minimum value, namely the distance a between the "wells" of the Anderson

model or more generally the scale of the fluctuations. There is a good deal more to it than that, for example, the Anderson localization criterion, but in three dimensions $\sigma_{\min}$ comes out to roughly $0.05 e^2/h a$, and in two dimensions $0.1 e^2/h$. In both cases there is considerable uncertainty in the numerical factor. When a is $3 \text{ \AA}$, $\sigma_{\min}$ is about $500 \text{ ohms}^{-1} \text{ cm}^{-1}$, though for impurity bands where a is much greater it is correspondingly smaller.

Temperature dependence

If E_F lies below E_c , two forms of conduction occur. At high temperatures current will be carried by electrons ex-

cited to the mobility edge E_c , so that

$$\sigma = \sigma_{\min} \exp [-(E_c - E_F)/kT] \quad (7)$$

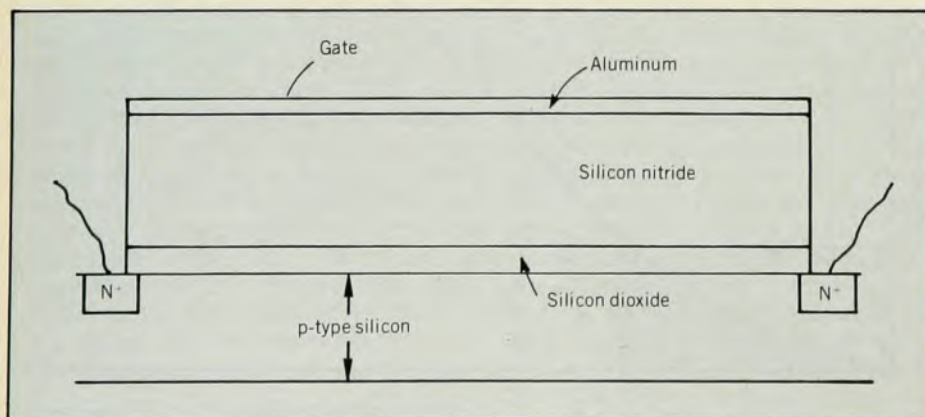
while at low temperatures, electrons near the Fermi energy determine the current—as in a metal. Exchanging energy with phonons, electrons hop from one localized state to another, the conductivity varying with temperature according to the law

$$\sigma = A \exp (-BT^{1/4}) \quad (8)$$

or, in two dimensions $1/3$ replacing $1/4$.

This kind of behavior has been observed in a wide variety of systems. In transition metal compounds of mixed valency such as $\text{La}_{1-x}\text{Sr}_x\text{VO}_3$ the number of electrons in the vanadium 3d band varies with x , as does the random field due to the random positions of the ions La^{3+} and Sr^{2+} . In a compensated semiconductor such as silicon doped with phosphorus and compensated by boron the number of electrons in the impurity band will depend on the degree of compensation, and the random element then arises both from the field of the charged boron acceptors (so that an impurity band is very close to Anderson's model) apart from the random positions of the donors. But perhaps the simplest case of all is provided by the inversion layer in a MOSFET device. Such a device, illustrated in figure 3, measures the current between the source and drain electrodes and its dependence on the voltage between a metal "gate," across a thermally-grown layer of SiO_2 , and the p-type silicon. The current flows in an inversion layer; at low temperatures the electrons in the inversion layer form a two-dimensional degenerate gas, two-dimensional in the sense that the wave functions perpendicular to the interface are in the lowest quantized state as shown. The potential energy in which the electrons move contains a random element, because in the layer of vitreous silica there are normally random charges; so a mobility edge is expected in the range of energies that can be occupied by electrons in the inversion layer. Moreover their Fermi energy can be varied and moved from below to above the mobility edge, as in figure 1, simply by changing the gate voltage. The dependence of current on temperature appears very much as in figure 2; in particular, a variation as $\exp(-B/T^{1/3})$ is observed² over many orders of magnitude, as are reasonable values of $\sigma_{\min}$.

On the whole, then, the model outlined here, in which there is no interaction between electrons, gives a satisfactory qualitative description of the electrical properties both of compensated impurity bands and inversion layers, except in one respect. In the range of temperature where the current is carried by electrons at a mobility edge one would expect the Hall coefficient to be activated and the Hall mobility to be independent of T ; a



A MOSFET device. Here the current flows in an inversion layer. The device measures the current between source and drain electrodes.

Figure 3

theory dating from 1971 due to Lionel Friedman suggests that the Hall mobility should be

$$\mu_H = 0.1ea^2/h \tag{9}$$

and sometimes this behavior is observed. But in other cases measurements of the Hall coefficient R_H give a temperature-independent value, $1/nec$, where n denotes all the electrons in the conduction band or inversion layer. This is the same value as one finds when E_F lies above E_c , when conduction is metallic.

Naturally, in trying to understand this discrepancy, various workers have noticed that the interaction between electrons is large, the average of $e^2/\kappa r_{12}$ over a volume a^3 being as large or larger than the random field V that produces Anderson localization. It has been proposed that, owing to this interaction, some kind of crystallization or ordering of the electrons may occur and that charge transport can occur by a mechanism similar to the flow of a liquid.³ In my opinion, it is likely that in inversion layers and compensated impurity bands the correlation term $\langle e^2/\kappa r_{12} \rangle$ can be taken care of within the Hartree-Fock approximation, and the difficulty is our imperfect understanding of the Hall effect. Next we shall discuss phenomena in which correlation does play an essential role, and shall give some indication of what happens when correlation and disorder affect the behavior.

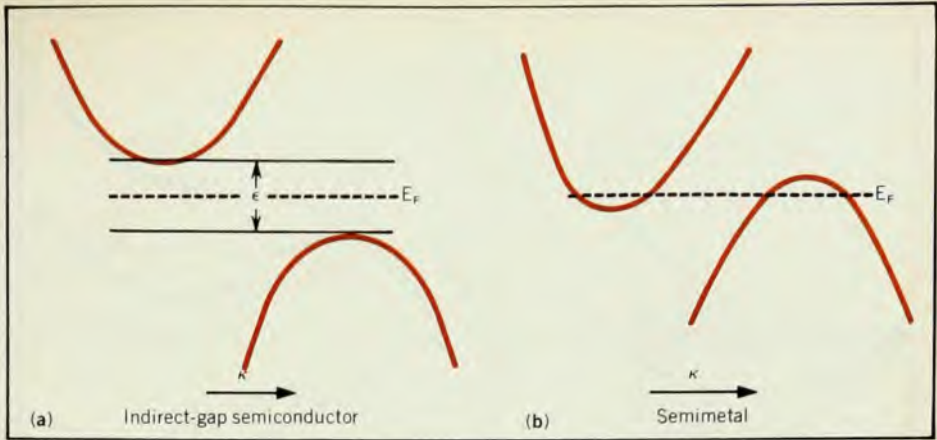
Band-crossing transitions

We turn now to metal-insulator transitions in crystalline systems. Perhaps the simplest occurs when an indirect-gap intrinsic semiconductor with a band form is illustrated in figure 4a is transformed into a metal as in figure 4b, the conduction and valence bands now overlapping. Examples are the divalent metals Yb, Ba and Sr where the bands separate under pressure, or the semiconductor Ti_2O_3 where a gap between the two branches of the d-band is small, but overlap occurs either on raising the temperature or on alloying with about ten percent of V_2O_3 . A transition of this kind can be described by the model of noninteracting electrons, which predicts that the gap would decrease continuously to zero and then the number of electrons and holes would increase from zero upwards.

From 1949 onwards, however, I have maintained that the existence of even a very small number of free electrons and holes at zero temperature is impossible, because these holes would combine together to form pairs. This pairing occurs because a Coulomb field with potential $-e^2/\kappa r$ always yields a bound state. A concentration n of carriers, given by

$$n^{1/3}a_H \simeq 0.25 \tag{10}$$

where a_H is $\hbar^2\kappa/me^2$, the hydrogen radius, would be necessary to give enough screening to ensure that the screened field $-(e^2/\kappa r) \exp(-qr)$ leads to no bound



Band-crossing transitions. The conduction and valence bands of an indirect gap non-metal (a); bands overlap to form a semimetal (b). Figure 4

state. It was therefore predicted that as some parameter (for example, volume) was changed, there would be a sudden change (at zero temperature) in the number of carriers from zero to a value given by equation 10. This argument suggested that there might be some "excitonic" phase formed of electron-hole pairs before truly metallic behavior sets in. Several theorists have developed this concept, and the idea of an insulating superfluid consisting of a Bose condensation of electron-hole pairs is fascinating. But, to the best of my knowledge, such a superfluid has not been observed, as might have been expected, for instance, when an electron-hole gas is formed by optical excitation of a semiconductor. What forms in all cases investigated is an electron-hole gas with metallic properties. If this is what normally occurs, an argument by W. F. Brinkman and T. M. Rice in 1973 gives a better way of considering the transition than that given originally by the present author, which is as follows. If in a semiconductor, as illustrated in figure 4a, the conduction and valence bands contain n electrons and holes respectively, their energy is

$$\frac{6}{5} \frac{\hbar^2 n^{2/3}}{m_{\text{eff}}} - c \frac{e^2 n^{1/3}}{\kappa} \tag{11}$$

The first term represents the kinetic energy of the electron gas and the hole gases (neglecting any many-valley structure of the bands), the second term, the Coulomb attraction between the two, and the constant c depends strongly on band form and the way correlation is taken into account.

This expression has a minimum as shown in figure 5, and the energy there is given by

$$E_{\text{crit}} = \text{const. } me^4/\hbar^2\kappa^2 \tag{12}$$

and n by an equation of the same type as equation 10 with an appropriate constant. The argument is that, as the band gap ΔE decreases, n will jump discontinuously from zero to the value given by equation 10, at which

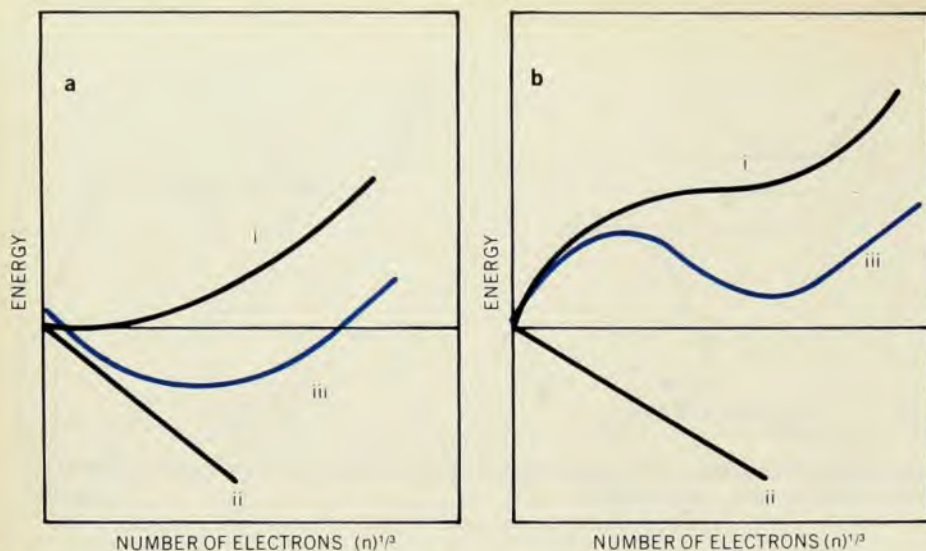
$$\Delta E = E_{\text{crit}} \tag{13}$$

It would be of considerable interest to observe a discontinuous change of this kind. For an experiment to be significant, it must be carried out at a low enough temperature for the number of electrons excited thermally across the gap to be negligible; one would then hope, if some parameter could be varied, to observe a discontinuous change from a finite value to zero in the activation energy ϵ for conduction.

But it is no use hoping to do this under pressure. If the free energy is plotted against volume, as in figure 6, there will be a change of slope at the transition, with the result that as the pressure is increased there will be a discontinuous change of volume from A to B. Moreover it is entirely possible that the crystal structure differs in the two phases. If the free energy of an alloy is plotted against composition, denoted by x , for ranges of x between A and B the alloy will separate into two phases. A single phase may perhaps be obtained by quenching; but until now this has not been done successfully. Moreover, in a disordered alloy the field in which the electrons move has a random element, and we shall see further on how this randomness affects our conclusions.

The Mott transition

Rather similar considerations apply to an array of one-electron centers. If they are far enough apart, they should form an antiferromagnetic lattice, as for instance in $TiBr_3$. I first suggested in 1949 that, if the distance between centers decreased, there should be transition to a metallic state with a density of electrons given by equation 10; the argument based on screening was used, and this kind of metal-insulator transition is sometimes called the Mott transition. More recently, in 1961, John Hubbard introduced the Hubbard Hamiltonian and the Hubbard intra-atomic energy U , defined as the average of e^2/r_{12} when two electrons are on one of the centers. If the centers are far apart, the energy necessary to take an electron from one center and put it on



Energy of an electron-hole gas (a) in a crystal and (b) for a noncrystalline system. Curve (i) denotes the kinetic energy, (ii) the potential energy, and (iii) their sum. The variation of energy with number of electrons n , shown in curve iii, has a minimum in both cases. Figure 5

another is just this quantity U ; it is the difference between the ionization energy and the electron affinity.

An extra electron placed on one of the centers can move to the next and so on through the lattice, with a Bloch wave function of the type in equation 2 and bandwidth B_1 ; we call this band of energies the upper Hubbard band. Similarly the "hole" from which an electron has been removed can move, the width of its "lower Hubbard band" being denoted by B_2 . Apart from certain refinements, this treatment predicts a metal-insulator transition when the two Hubbard bands overlap; that is, when

$$U - \frac{1}{2}(B_1 + B_2) = 0 \quad (14)$$

It is a curious coincidence that, for hydrogen-like centers, this leads to just the condition of equation 10, with a value of the constant about 0.2. The treatment as given by Hubbard does not lead to a discontinuous change in n , but the band gap described here by equation 13 is immediately applicable, and a discontinuous change is predicted, from an antiferromagnetic insulator state to one that is metallic and may be antiferromagnetic too.

Discontinuous and continuous

We may ask, has this behavior been observed? The behavior of doped *uncompensated* semiconductors was for a long time regarded as an example of this type of transition, and indeed the concentration of centers at which metallic behavior sets in does obey equation 10 for a very wide range of materials, the best value of the constant being 0.2. On the other hand the activation energy for conduction (normally denoted by ϵ_2) goes continuously to zero as n is increased. This may be because the centers are distributed at random in space. However, another random array of one-electron

centers, that provided by the solvated electrons in solutions of alkali metals in ammonia, shows a solubility gap with a consolute point at -42°C ; only above this temperature can a continuous metal-insulator transition be observed as the concentration of alkali is changed. According to figure 6, this gap suggests that there is a discontinuous change in ϵ_2 , and frozen solutions of ammonia in a solvent such as hexamethylphosphoramide may prove to be the best way of observing it.

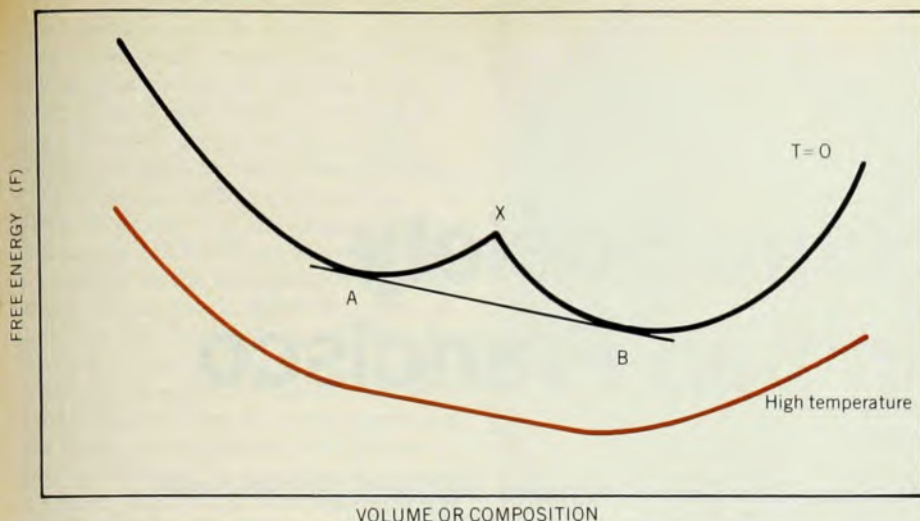
If so, we need a criterion to determine how much disorder in a system is compatible with a discontinuity in ϵ_2 . I have suggested⁴ such a criterion, though its detailed consequences have yet to be worked out. The idea is illustrated in figure 5. At the bottom of a band (Hubbard or otherwise), the density of states in a crystalline material behaves like $E^{1/2}$, but in a noncrystalline system an exponential tail is expected. Thus in equation 11, the first term, behaving like $n^{2/3}$, must be replaced by a term that varies as a low power of n for small n , and only for values of n that bring the Fermi energy out of the tail will a behavior as $n^{2/3}$ be approached. The energy term that must replace that in 11 is shown by the dotted line in figure 5; only if a minimum results at a negative value of E is a discontinuity to be expected in n .

We surmise that this criterion, if worked out in detail, might show that a discontinuity is to be expected for metal in ammonia but not for metal in silicon-phosphorus. In the former the range Δa in the distances a between centers will be much smaller than in Si:P, in comparison with the hydrogen radius. For silicon-phosphorus, in particular, doubt now arises⁵ on whether the concentration at which the transition occurs is determined by equation 14 at all, or whether it is an Anderson transition in which the Hubbard U plays little role. Certainly the

transition shows all the properties illustrated in figure 2, and the metallic state has few of the properties (such as greatly enhanced electronic specific heat and positive thermopower) expected just on the metallic side of a metal-insulator transition. The condition for Anderson localization in an impurity band has been worked out by B. T. Debnay⁶ and is of the form in approximation 10 with the constant equal to 0.4, although E. N. Economou and P. D. Antiniou,⁷ using a less realistic model, find that if disorder is "off-diagonal," that is to say with no random fields such as those present in a compensated semiconductor, localization cannot occur in mid-band. At present, then, the role of the Hubbard U remains uncertain.

Many non-metals show transition to the metallic state with increasing temperature, and there is certainly no uniform model that can describe them all. Thus Ti_2O_3 at low temperatures is a nonmetal with a small band gap (about 0.2 eV) separating a filled and empty band both derived from the titanium 3d orbitals. The material has the corundum structure and the gap depends on the ratio c/a . As the temperature is raised, electrons are excited into the conduction band, so c/a changes in the sense needed to diminish the gap, thus allowing more electrons to be excited with consequent increase in entropy. At about 400 K the bands begin to overlap giving metallic behavior. Overlap can also occur on alloying with about 10% V_2O_3 . One might hope to see the predicted discontinuous change in the density of current carriers in this system, but unfortunately the vanadium ions produce an impurity band, and metallic behavior begins here at much lower concentrations than are needed for band crossing.

The compounds V_2O_3 and VO_2 both show sharp transitions to metallic behavior as the temperature is raised; in the former metallic behavior can also be produced by alloying with Ti_2O_3 , the metal being an antiferromagnet, and also by applying pressure. The low-temperature phase in V_2O_3 is an antiferromagnetic insulator, and so the transition can be described as a kind of Mott transition. The change to the metallic state is due to a change in c/a that increases the bandwidth B relative to U and the driving force when the transition occurs as a function of temperature is the high entropy of a highly correlated "metal." VO_2 with, in the high-temperature phase, the corundum structure, appears to be a metal in which a broad and narrow d-band overlap,⁸ giving a high electronic specific heat and entropy. In the low temperature phase a structural distortion has the result that the bands do not overlap, and the narrow filled band should, if pairing were not allowed, give an antiferromagnetic insulator. However, the rutile structure does allow pairing, so



Free energy at a metal-insulator transition. The variation here is shown as either a function of composition (x) or volume. Note discontinuity in both cases at $T = 0$. Figure 6

vanadium atoms move towards each other, forming diamagnetic pairs. This in our view is not the cause of the non-metallic behavior, because even without pairing the Hubbard gap would exist; it is an example of the tendency of one-electron centers to form pairs if the structure allows, of which we shall give another example below (Ti_4O_7).

A quite different situation is presented by Fe_3O_4 , in which Fe^{2+} and Fe^{3+} order on available sites at low temperatures, giving nonmetallic behavior, and show a sharp transition at 110 K to a situation where they are random, the conductivity then being of order $10^2 \text{ ohms}^{-1} \text{ cm}^{-1}$. This "Verwey" transition is named after its discoverer E. J. Verwey. There is some controversy as to whether the problem should be treated as a charge-density wave that disappears as the temperature is raised, or as a Bragg-Williams disordering of heavy current carriers, perhaps polarons. The entropy of the transition is about $\frac{1}{2}Nk \log 2$, about half what would be suggested by the latter hypothesis, but other phenomena such as a Mössbauer splitting suggest a charge density wave. Doubtless a correct treatment will contain elements of both representations. A similar behavior is shown by Ti_4O_7 where the number of electrons is also half the number of sites; at low temperatures they are ordered, at high temperatures disordered with near metallic behavior, but an intermediate range of temperature appears in which the material conducts fairly well. In this temperature range σ shows a small activation energy but at the same time the material is *diamagnetic*. It is surmised⁹ that the electrons form diamagnetic pairs, the titanium atoms in each pair being displaced towards each other, but that these pairs are mobile with an activation energy of polaron type. We call them "bipolarons." Bipolarons may also exist in some other transition-metal compounds, but Ti_4O_7

is certainly the clearest current example.

There is still much that is obscure about these fascinating materials.¹⁰ Some, perhaps V_2O_3 may be so complicated—involving correlation, lattice distortion, polarons and a complicated band structure—that a simple theory may forever elude us. In materials showing a Verwey transition, such as Fe_3O_4 and Ti_4O_7 , the same may be true, but some features invite a qualitative explanation. One is that quite weak alloying, about 1% of V_4O_7 in Ti_4O_7 for instance, suppresses the ordered low-temperature phase. This is hardly to be understood on a Bragg-Williams model and suggests that the concept of a charge-density wave of bipolarons may have to be adopted.

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