

Metal–Insulator Transitions

Second Edition

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Preface to the Second Edition

The first edition of this book, published in 1974, described metal-insulator transitions due to disorder (the Anderson transition), the Mott-Hubbard transition resulting from intra-atomic interaction (the Hubbard U), as well as certain other types such as band-crossing transitions and those of Verwey as in Fe_3O_4 . Since then, our understanding of the Anderson transition has been completely transformed by the scaling theory of Abrahams *et al.* (1979), experiments that supported its conclusions and by the theory of interactions between electrons initiated by Altshuler and Aronov (1979) for the metallic state and by Efros and Shklovskii (1975) for hopping conduction. The part of the book that deals with these transitions, both in impurity bands and other systems, has been completely rewritten. Our description of the Mott-Hubbard transition has changed less, except that the transition in doped silicon and germanium is now believed to be of Anderson type, even though the formula for the critical concentration derived by the present author in 1949 for a Mott transition is in satisfactory agreement with experiment for almost all doped semiconductors. Our motive for a new edition dealing with the transition in crystalline conductors, particularly the transitional-metal oxides, is primarily the renewed interest in these materials generated by the discovery of the high-temperature superconductors. Some of these, it is widely believed, are antiferromagnetic insulators doped sufficiently heavily to become metallic. We include a chapter putting this point of view, in the context of what we know in general about metal-insulator transitions. We also describe our new understanding of the phenomenon in liquids.

In the first edition we expressed our debt to many colleagues with whom we have discussed the subject matter of this book, particularly J. V. Acrivos, P. W. Anderson, K. F. Berggren, M. H. Cohen, B. R. Coles, M. Cyrot, E. A. Davis, J. Friedel, L. Friedman, J. Goodenough, G. Gruner, P. Hagenmuller, S. Maekawa, T. M. Rice, M. Pollak, J. C. Thompson, and R. M. White. We now add the names of C. Cyrot-Lackmann, J. P. Doumerc, P. P. Edwards, M. Kaveh, J. Morgan, M. Pepper, G. A. Thomas and many others. We are also particularly grateful to

many authors who contributed articles on aspects of the subject to a *Festschrift* presented to the author on the occasion of his eightieth birthday, without which parts of the book could hardly have been written.

We depart from the usual notation in two ways. The transfer integral, usually denoted by t , is written I , following our first edition and Mott and Davis (1979). Also, instead of “transition metal”, we write “transitional metal” (following J. Friedel), in order to avoid confusion with the former word as used in the title of this book.

Nevill Mott

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Wigner and Verwey Transitions

1 Wigner crystallization

The metal-insulator transitions described in the preceding chapters occur for electrons in a narrow band, for which the orbitals can be described by the tight-binding method as made up of atomic wave functions. More than a decade before this kind of transition was considered, Wigner (1938) proposed that for low densities a gas of free electrons at zero temperature would crystallize into a non-conducting state, even in the absence of any lattice field. The argument is as follows. We suppose that there are n electrons per unit volume neutralized by a background of uniform positive charge ("jellium"). Then, if we localize each of them in a volume n^{-1} , this costs an amount of energy, per electron,

$$\sim \hbar^2 n^{2/3} / m. \quad (1)$$

On the other hand, there is a gain of interaction energy between electrons

$$\sim e^2 n^{1/3}. \quad (2)$$

At low densities, then, energy is always to be gained by such localization, which should take place when

$$n^{1/3} a_H = \text{constant} \quad (a_H = \hbar^2 / me^2)$$

The evaluation of the constant, since it must compare the energies of a low-density electron gas with the crystallized state, is difficult. An estimate (Mott 1961, Kugler 1969) is 0.05.

The concept of a degenerate electron gas superimposed on a uniform background of positive charge is of course difficult to realize. The present author (Mott 1967) suggested that a good approximation to this situation would be a highly doped and very strongly compensated semiconductor, for which the Bohr radius $\kappa \hbar^2 / me^2$ was large compared with the distance between centres. This idea was developed by Durkan *et al.* (1968); and Somerford (1971) used strong magnetic fields to induce such a transition in n-type InSb; Care and March (1971) and Rousseau *et al.* (1970) gave evidence to show that conduction in such a lattice is due to thermally generated vacancies.

Our conjecture as to the properties of a Wigner crystal are as follows.

- (i) It will be non-metallic and antiferromagnetic (or perhaps ferromagnetic, cf. Edwards and Hillel 1968).
- (ii) There will be two conduction mechanisms:
 - (a) excitation of electrons to the "conduction band"—a process similar to that described earlier in this chapter for "Mott-Hubbard" insulators; the material should be an intrinsic semiconductor and both electrons and holes can carry current;
 - (b) formation of vacancies, which are mobile, as described by Care and March (1971); the vacancies are identical with the holes mentioned in (a), but, if they are formed, the gas (if held at constant volume) undergoes compression.
- (iii) As the temperature is raised, the lattice can "melt"—by which we mean that long-range order will disappear.

Wigner crystallization is a subject on which there is little theoretical work, and it is not certain whether the phenomenon has been observed.

Experimental data on $\text{Hg}_{1-x}\text{Cd}_x\text{Te}$, a material in which for appropriate values of x the gap disappears, have been claimed as showing evidence for such a phenomenon (Nimtz *et al.* 1979, Rosenbaum *et al.* 1985).

Whether or not the available sites for electrons lie on the crystal lattice, the electrons may crystallize under the influence of their mutual repulsion. At low temperatures the system will then be non-conducting. There will be an energy of order $e^2/\kappa a$ needed to transfer an electron to a "wrong" site, so the material should behave like an intrinsic semiconductor. However, this energy should decrease as more and more electrons are transferred, so that as the temperature is raised we expect a first- or second-order transition to a state in which the sites are occupied in a random way. The analysis of such a transition could be analogous to that given originally by Bragg and Williams (1934) (for references to early work see Mott and Jones 1936, p. 29) for the order-disorder transition in alloys of the types AB, AB_3 , etc. We call such transitions "Verwey transitions", since they were first proposed by Verwey (1935) for Fe_3O_4 .

Such transitions are expected particularly for transitional-metal oxides of mixed valence, where polaron formation increases the effective mass of the carriers, so that the kinetic energy is small compared with the potential energy. The Hubbard intra-atomic energy U (Chapter 2, Section 9), of order $\langle e^2/r_{12} \rangle$ and equal to several electron volts, must be distinguished from the much smaller energy $\sim e^2/\kappa a$ needed to move an electron (polaron) onto a "wrong" site. This chapter is devoted to the discussion of materials in which the number of sites, N , is greater than n and in which, because of polaron formation or for other reasons, $\hbar^2/m_p a^2$ is much smaller than U , so that an essentially localized model is a useful first approximation, though $\hbar^2/m_p a^2$ and $e^2/\kappa a$ may be comparable (here m_p is the polaron effective mass).

We suppose that we have a gas of n electrons moving between N equivalent sites, and $N > n$. Then we have to ask whether or not such a material will conduct at low temperatures. There are several possibilities.

- (i) It will form a degenerate electron gas and conduction will be of the kind usual in metals.
- (ii) If the N sites are distributed at random or if there is some other kind of disorder then Anderson localization may occur at the Fermi energy, so that conduction at low temperatures will be by hopping (Chapter 1, Section 15). This can be the case under certain circumstances for doped and compensated semiconductors for low concentrations and for $\text{La}_{1-x}\text{Sr}_x\text{VO}_3$ (Chapter 7) and the oxide superconductors (Chapter 9).

The materials to be discussed in this chapter are Fe_3O_4 (magnetite) and Ti_4O_7 . In both, the number of carriers is half the number of available sites. At low temperatures they take up ordered positions, under the influence of their mutual repulsion; in both, there is a first-order transition as the temperature is raised to a state in which the carriers are at random sites. In Ti_4O_7 the carriers are bipolarons, as has already been pointed out in Chapter 2.

2 Fe_3O_4 (magnetite)

A recent review of the experimental situation has been given by Honig (1985). It is pointed out that the electrical properties, particularly near to the transition, are very sensitive to purity and specimen preparation, and that much of the extensive experimental work is therefore open to doubt. None the less, the broad features of the behaviour of this material are clear. The history of the so-called Verwey transition in this material goes back to 1926, when Parks and Kelly (1926) detected an anomalous peak near 120 K in the heat capacity of a natural crystal of magnetite. The first detailed investigations were those of Verwey and co-workers (Verwey 1939, Verwey and Haayman 1941, Verwey *et al.* 1947), who showed that there was a near discontinuity in the conductivity at about 160 K. The conductivity as measured by Miles *et al.* (1957) is shown in Fig. 8.1.

In this spinel, to which we may give the formula $\text{Fe}^{3+}(\text{Fe}^{2+} + \text{Fe}^{3+})\text{O}_4$, below 119 K the Fe^{2+} and Fe^{3+} ions are ordered onto alternate (011) B-site layers, distorting the crystal into orthorhombic symmetry (Goodenough 1963, p. 185). It will be seen from Fig. 8.1 that there is a first-order transition at 119 K (the Verwey temperature T_V) and that above this the conductivity increases by a further factor ~ 10 and eventually decreases slightly. Its largest value ($\sim 10^2 \Omega^{-1} \text{cm}^{-1}$) is rather low for a normal metal, however short the mean free path. In the ordered phase at the lowest temperatures the observed activation energies are small ($\sim 0.03 \text{eV}$), and conduction is probably by a hopping mechanism, allowed on account of vacancies or other defects. At temperatures below 850 K the ions on A- and B-sites are ordered antiferromagnetically, so the current carriers (on the B-sites) have moments ordered ferromagnetically above and below T_V .

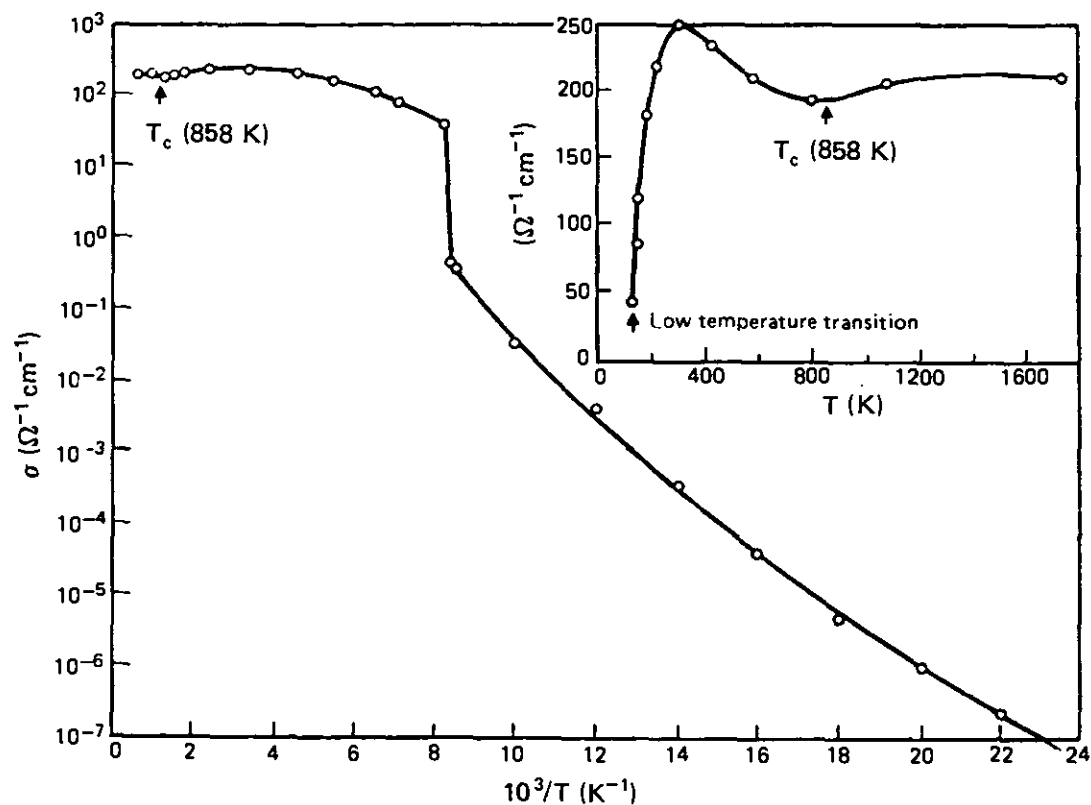


Fig. 8.1 Conductivity of Fe_3O_4 as a function of reciprocal temperature. After Miles *et al.* (1957).

As explained in Section 1, we believe in common with most other investigators that the Hubbard U can be considered large compared with the bandwidth,[†] so that the conductivity is due to the fact that the number of carriers (Fe^{2+}) is smaller (by a factor 2) than the number of the B-sites, on which there is a mixture of Fe^{2+} and Fe^{3+} . Thus the quantity that determines the electrical properties must be the ratio between the interaction between electrons (Fe^{2+} and Fe^{3+}), which we write as $V = e^2/\kappa R$, and the bandwidth B . We write $V_0 = e^2/\kappa a$ for the interactions between carriers on nearest-neighbour sites. On general grounds we should guess that if V_0/B is small then the material will be a metal, and if it is large then a Wigner crystallization will occur. An analysis of the condition for crystallization was given by Lorenz and Ihle (1972) and Cullen and Callen (1971, 1973). The latter authors found that an ordered insulator will occur when $V_0/B \gtrsim 3$. They also predicted multiple ordering for somewhat lower values of V_0/B ; such ordering is in fact observed in the insulating phase, there being much evidence from Mössbauer and other measurements discussed by these authors. It therefore seems right to conclude that V_0/B does not greatly exceed the critical value.

[†] It is likely that the band is formed from non-degenerate orbitals, in which case, since all spins are parallel, two electrons could not be on the same Fe atom even if U were not large.

We now ask what values of V_0 and B are likely. Bandwidths in transitional-metal oxides are 1–2 eV; the high magnetic-ordering temperature would not lead us to expect a small value of B , if it is given by a term of the type B^2/U (Chapter 3, equation (7)). We think that a value of V_0 of order 5 eV is very unlikely, in view of the small activation energies involved. It seems much more plausible that the carriers are small (or intermediate) polarons, so that

$$V_0 = \frac{e^2}{\kappa a},$$

with κ the background *static* dielectric constant, and B the polaron bandwidth, perhaps ~ 0.1 eV.

It is clear, however, that the activation energy for conduction at low temperatures, and the ordering energy, cannot be wholly electrostatic, since V_0/B lies not too far from the critical value, and the kinetic energy B must be taken into account. This follows particularly from the considerations of Anderson (1956), who pointed out that, if nearest-neighbour order is maintained, the electrostatic energy needed to produce long-range disorder is less than 10% of that to produce complete disorder; he also showed that half the entropy ($nk_B \ln 2$) for complete disorder is produced by disorder in which short-range order is maintained. The σ – $1/T$ curve shown in Fig. 8.1 suggests that it is necessary only to double the temperature after the transition to obtain a maximum value of the conductivity, and thus presumably complete disorder. Therefore no simple electrostatic model will be sufficient (cf. Fazekas 1972, Sokoloff 1972).

The assumption that the carriers are small or intermediate polarons in no way militates against discussions of the band structure of the ground state (see e.g. Camphausen *et al.* 1972, Cullen and Callen 1971, 1973). The absence of Jahn–Teller distortion (Goodenough 1971) also, in our view, indicates *not* the absence of a polaron mass-enhancement but rather a value of V_0/B not too far from the critical value. These conclusions seem to be in agreement with the considerations of Sokoloff (1972), who used a description in terms of a degenerate band of small polarons. Samara (1968) showed that pressure lowers the temperature of the Verwey transition. If this depended only on $e^2/\kappa a$ then the opposite should be the case. But pressure will increase B , and push the substance nearer to the critical value for the metal–insulator transition.

Miyahara (1972) showed that most impurities, including a deficit of metal, lower T for the metal–insulator transition. More recently, Whall and co-workers (see Graener *et al.* 1979) showed that 5% of fluorine in $\text{Fe}_3\text{O}_{4-x}\text{F}_x$ destroys the sharpness of the transition;† their results are shown in Fig. 8.2, where $\log \sigma$ is plotted versus $1/T$. The straight line in the insulating range does not necessarily mean that conduction is by hopping; it is suggested that, near the transition, conduction is by excitation of electrons from the Fe^{2+} sites into the Fe^{3+} sites, from which they can move by band motion, whether or not they form polarons.

† Later work by Barlow *et al.* (1987) shows that traces of the sharp transitions, on doping with F or Ge, remain at higher concentrations.

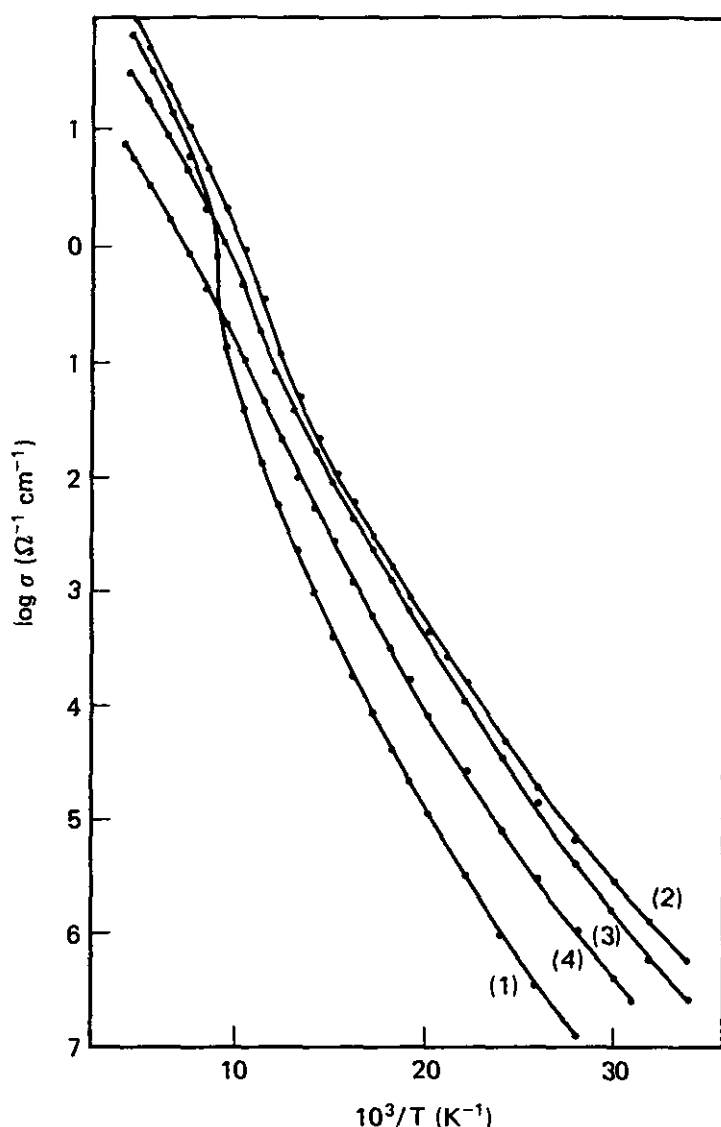


Fig. 8.2 Conductivity of $\text{Fe}_3\text{O}_{4-x}\text{F}_x$ for various values of x as a function of reciprocal temperature (Whall *et al.* 1978): (1) $x=0.025$; (2) 0.05; (3) 0.1; (4) 0.15. It will be seen that the Verwey transition disappears somewhere between $x=0.025$ and 0.05.

The disappearance of the sharp Verwey transition was discussed by Mott (1979), who suggested that at low temperatures the material is a “Wigner glass”, the electrons (Fe^{2+} ions) being frozen into random sites and the whole system stabilized by the fluorine. Discussion of the thermopower measurements show, according to Mott (1979), that a hopping mechanism is operative at low T . Ihle and Lorenz (1985), however, consider that the electrons in the “wrong” sites move by a small polaron band mechanism.

The mechanism for conduction at temperatures above the transition has long been a matter of controversy. It is very low for a metal, less than $300 \Omega^{-1} \text{cm}^{-1}$, of the order of σ_{min} . The temperature dependence is small (see Fig. 8.1). Ihle and Lorenz (1980, 1985) pointed out that traditional one-particle concepts break down because there is strong short-range order due to Coulomb repulsion between heavy charge carriers (perhaps intermediate polarons, see Chapter 2, Section 2). Mott (1979) proposed that the high-temperature phase in the pure

material can be considered as a Wigner glass (like the impure material below T_c), stabilized by the entropy of disorder. The conductivity would be expected to be low, but to calculate it is difficult. Ihle and Lorenz (1985 and earlier papers) attempt this by combining hopping and band conduction.

3 Ti_4O_7

In this material also the sharp Verwey transition disappears with traces of impurity—in $(\text{Ti}_{1-x}\text{V}_x)_4\text{O}_7$ when $x > 0.01$ (Schlenker *et al.* 1976, Gourmala *et al.* 1978, Ahmed *et al.* 1978). Below 90 K the conductivity then behaves like $\exp[-(T_0/T)^{1/4}]$. The above authors believe that the conduction process giving these low activation energies is the hopping of an electron from one V-Ti^{3+} pair to a V-Ti^{4+} pair, there being some compensation to make this possible.

Mott (1979, p. 357) discusses the amount of impurity needed to make the sharp Verwey transition disappear. The small value, $x \sim 0.01$, for Ti_4O_7 is difficult to explain.

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