Electronic Properties of Amorphous Materials

Changes are considered which occur when the long-range order typical for crystals disappears.

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Modern electronic industry makes more and more use of devices based on the dynamics of electrons in solids rather than in a vacuum. Their design was made possible by the enormous progress achieved in the last few decades in our understanding of the electronic processes in crystals. The development of the theory was greatly facilitated by a systematic use of the essential property of crystals: translational symmetry—a term meaning that the arrangement of atoms is considered to be the same throughout the whole body. One must take, of course, a proper account of surfaces, but experience has shown that many properties of crystals with linear dimensions as small as a few thousand angstroms are the same as those of crystals with infinite dimensions.

Now there is a large category of solids in which the atomic arrangement lacks translational symmetry (or, as I shall also call it, long-range order). Typical representatives are various kinds of glasses (some metal oxides, sulfides, selenides, and tellurides and their alloys), amorphous selenium, some other elemental semiconductors (germanium, silicon, boron, tellurium) condensed as thin films on a substrate kept at sufficiently low temperatures, and many organic solids. Then, of course, also lacking translational symmetry are all liquids, whose properties, contrary to what was thought earlier, have been shown to be much more like those of a solid than like those of a gas. It is therefore a natural step in the physics of the electronic properties of solids to try to understand what happens when the translational symmetry disappears and to construct a theory which would be applicable to amorphous bodies. The task appears to be a difficult one. No direct approach has been suggested so far, and the first attempts are based on the idea that the atomic structure of an amorphous body can be considered to be a relatively small perturbation of the corresponding crystalline structure.

Recently new electronic devices have been designed in which chalcogenide semiconducting glasses are used. These devices will probably soon find important application in the electronics industry. This will undoubtedly give impetus to new, more intensive theoretical and experimental work in this area.

In this article I first explain the differences in the atomic structure of a crystal and of an amorphous body and show the consequences of these differences for the theory of electronic states. I then discuss the results of a few optical and transport measurements which give some evidence about the changes which occur through loss of the long-range order.

Atomic Structure

In an amorphous body or in a liquid the atomic arrangement is not a totally chaotic one. The chemical forces between the atoms tend to bind the atoms in the same way that they are bound in the crystal. The consequence of these forces is that the arrangement of the nearest and next-nearest neighbors of a given atom is not very different from that in a crystal. Diffraction of x-rays by a crystal gives sharp spots or lines from which the exact positions of atoms forming the lattice can be determined. Diffraction of x-rays by an amorphous body gives more-or-less broad bands from which the distributions of the probabilities of finding an atom at a distance r from the chosen atom at $r = 0$ can be deduced. Figure 1 shows such a distribution for amorphous germanium. We see that even in germanium in the amorphous state there is a certain correlation of the atomic positions, which, contrary to the situation in the crystal, extends only over small distances (several lattice constants). We speak about the short-range order, and in the case of germanium, as seen in Fig. 1, the short-range order is similar in amorphous germanium and in the crystal.

A simple characteristic of the short-range order is the number of nearest neighbors of an atom, the so-called coordination number. If this number is the same in the disordered form as in the crystalline form, the disordered atomic arrangement can be considered a perturbation of the crystalline lattice, and many properties of the disordered form can be deduced from the crystalline properties by means of an appropriate perturbation procedure. Both amorphous germanium and crystalline germanium have a coordination number of 4, and indeed both are semiconductors. Molten germanium, however, has properties of a metal, very different from crystalline germanium; in fact, the coordination number increases from 4 to 8 during melting. This is an example of a general statement stressed by Ioffe and his school (*1*): the basic electronic properties are determined by the short-range order.

To understand the difference between an amorphous body and a crystal with nearly the same short-range order we must realize that random fluctuations in distances and angles which may be practically negligible over one or a few lattice cells lead to large mean-square deviations over long distances. As the statistics show, these latter quantities increase as $(N)^{\frac{1}{2}}$ (where N = number of cells), so that the order over large N (the long-range order) disappears.

Electronic Properties of Crystals

Quantum theory immediately draws most important conclusions concerning the electronic states in a crystal from

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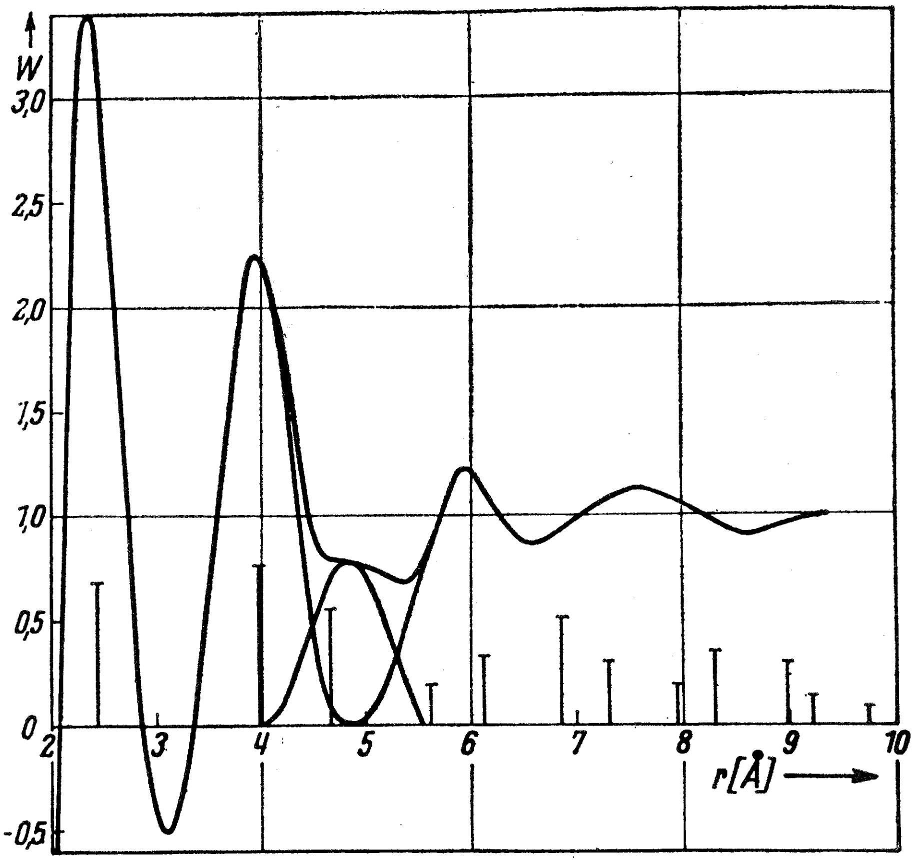


Fig. 1. Distribution of atoms in amorphous germanium. W is the average number of atomic positions at the distance r from a chosen atom at $r = 0$. The vertical lines show the position of atoms in crystalline germanium. [After Richter (*16*)]

the mere existence of the translational symmetry.

Wave functions of an electron in a crystal are plane waves similar to those in a vacuum, except that their amplitude is a periodic function with the same period as that of the atomic arrangement. These waves (the so-called Bloch waves) propagate freely through the whole lattice; they have a wavelength λ, and can be characterized by a wave vector $\mathbf{k}$ whose direction shows the direction of propagation of the wave and whose magnitude is $|\mathbf{k}| = 2\pi/\lambda$. The product $\hbar\mathbf{k}$ (where $\hbar = h/2\pi$ and h is the Planck constant) has the dimension of a momentum, and indeed it has many properties analogous to the momentum of an electron in free space; one therefore calls $\hbar\mathbf{k}$ the quasi-momentum of the electron.

The energies of an electron in the crystal can be grouped into bands of allowed and forbidden energies; the difference between two adjacent levels in a band of allowed energies is, for all practical purposes, negligible. Such a system of allowed and forbidden bands is completely analogous to the bands of allowed and forbidden frequencies of an electromagnetic wave propagating along a line with a periodic structure. According to the Fermi principle, the allowed energy levels, starting from the lowest, are filled with electrons in such a way that each state can contain only two electrons, with opposite spins. A completely filled or a completely empty band cannot carry electric current. Of the bands that contain electrons at zero temperature, the one of highest energy is called the valence band. In metals, it is incompletely filled with electrons; this is the cause of the high electrical conductivity of metals. In insulators and semiconductors, the valence band is completely filled at zero temperature. Electrical conductivity can take place if electrons are excited from the valence band into the energetically next-higher band—the conduction band. Electrical current is then carried both by the electrons in the conduction band (negatively charged carriers) and the vacant electronic states near the top of the valence band—the so-called holes (positively charged carriers).

The energy of an electron in each allowed state is a function of $\mathbf{k}$, and of n, which denotes the band; in the case of the valence band and the conduction band we use the designations v and c in place of n. The energy difference between a minimum for the conduction band E_c ($\mathbf{k}_c$) and a maximum for the valence band E_v ($\mathbf{k}_v$) is called the energy gap E_g, and equals $E_c(\mathbf{k}_c) - E_v(\mathbf{k}_v)$. It is a direct gap if $\mathbf{k}_c = \mathbf{k}_v$ or an indirect one if $\mathbf{k}_c \neq \mathbf{k}_v$.

Excitation of an electron from the valence band into the conduction band can occur either by thermal excitation or by irradiation with electromagnetic waves. In the former case the smallest of the gaps determines the activation energy of the electric conduction. In the regions of infrared, visible, and ultraviolet radiation a photon can cause a transition from the valence band into the conduction band which must satisfy two conditions: during the transition both the energy and the quasi-momentum $\hbar\mathbf{k}$ must be conserved. If the energy of the photon is $\hbar\omega$, we have $E_c(\mathbf{k}_c) - E_v(\mathbf{k}_v) = \hbar\omega$ (conservation of energy) and $\hbar\mathbf{k}_c - \hbar\mathbf{k}_v =$ photon momentum (conservation of momentum). The photon momentum is $h/\lambda_{\mathrm{elmag}}$, where λ_{elmag} is the wavelength of light ($\lambda_{\mathrm{elmag}} = 2\pi c/\omega$, c is the velocity of light). The photon momentum is much smaller than electron quasi-momentum $\hbar|\mathbf{k}| = h/\lambda$, because λ is of the order 10^0 to 10^1 angstroms and λ_{elmag} is $\sim 10^3$ to 10^5 angstroms. Therefore, in the spectral region considered, $\mathbf{k}_c = \mathbf{k}_v$ ($\mathbf{k}$ vector is conserved during the transition). Transitions satisfying this rule are called direct transitions.

Let us recall once more that all these statements are direct consequences of the translational symmetry. Of course, in a real crystal the translational symmetry is not perfect. There are various lattice imperfections, such as thermal vibrations of the atomic lattice, foreign atoms at various places in the lattice, and atomic vacancies. The important assumption is that these defects can be treated as small perturbations of the perfect lattice; this assumption is well justified even at high temperatures and for defect concentrations below 10^{19} per cubic centimeter (the concentration of atoms forming the lattice is of the order 10^{22} per cubic centimeter). This means that we consider the movement of an electron to be free (its state is well described by a Bloch wave function) until the electron "collides" with some imperfection (another way to express this is to say that the electron wave is "scattered" by the imperfection). Were it not for these collisions, the ideal lattice would have infinite electrical conductivity. Such a description evidently makes sense if the free path of an electron is considerably longer than the lattice constant a.

There are two types of defects that are very important in semiconductors: donors and acceptors. A donor is neutral in the ground state; in the ionized state it frees an electron and is positively charged. An acceptor is neutral in the ground state; in the ionized state it binds an electron. These defects give rise to energy states in the energy gap which are bound states, as distinguished from the free Bloch states. Donors with energies near those of the conduction band give, at non-zero temperature, electrons to the conduction band and cause n-type conductivity. Acceptors with energies near those of the conduction band capture electrons from the valence band; free holes are formed, and we have p-type conduction. The two last-mentioned kinds of conduction are referred to as impurity conduction, as distinguished from the intrinsic conduction arising from the excitation of electrons from the valence band to the conduction band.

If the concentrations of imperfections are high enough, electric current can be carried by electrons jumping from one defect to the next (the so-called hopping mechanism of electric conductivity activated by the atomic thermal motion).

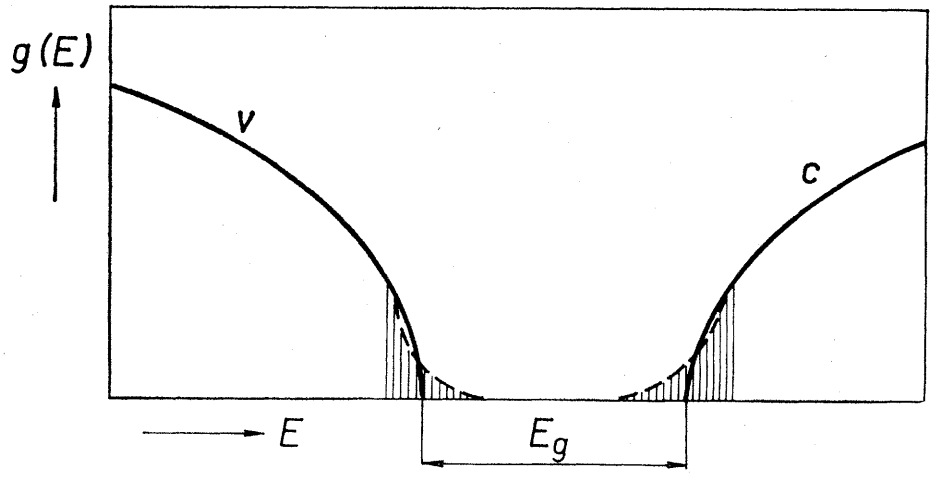


Fig. 2. Dependence of the density of electron states $g(E)$ on energy E. Solid lines correspond to the crystal, dashed lines to the disordered structure, according to theoretical considerations. (Shaded areas) Regions in which the states are localized; (c) conduction band; (v) valence band; (E_g) energy gap in the crystal.

Electronic States in an Amorphous Body

When we consider an amorphous solid, most of the laws mentioned in the last section do not make sense: if there is no long-range order, Bloch functions are not ground states of an electron and the quasi-momentum $\hbar\mathbf{k}$ is no longer a quantum number.

Gubanov (*2*) has suggested a procedure for extending these extremely useful notions to disordered structures. He assumes that the difference between the short-range-order atomic arrangement in an amorphous body and in the corresponding crystal is small. The necessarily large differences over long distances are taken care of by a transformation of coordinates. In place of the usual Cartesian coordinates, Gubanov introduces certain curvilinear coordinates and shows that the ground-state wave functions are linear combinations (*3*) of wave functions which are Bloch functions in these curvilinear coordinates. Their usefulness is limited (they seem not to be appropriate for the description of optical and transport effects), but they make it possible to calculate roughly the electron energy states. Gubanov has shown that they form bands not too different, in the approximation considered, from those of the corresponding crystal (there are only changes in band width and in gaps). It is difficult to apply Gubanov's theory to quantitative calculations concerning actual amorphous materials, and, as far as I am aware, no such attempt has been made.

It is possible to obtain detailed information on electronic states in the one-dimensional case [see the review paper by Mott (*4*)]. One considers a chain of atoms with the interatomic distance a in the crystalline state; in the disordered state the distance is $a\,(1 + \epsilon\gamma)$, where ϵ is a constant characterizing the degree of disorder and γ is a random variable with a convenient distribution (average values, $\bar{\gamma} = 0$, $\bar{\gamma}^2 = 1$). An important result is that some band states represented by wave functions extending through the whole crystal (Bloch functions) become localized in the disordered structure (the wave function has an appreciable value only in a small volume). An example is shown in Fig. 2, in which the density of states (the number of allowed states per cubic centimeter of the solid per unit energy) is plotted against the energy of the states. We see that some states near the band extrema become localized, and they even extend into the gap. There are more localized states if ϵ is larger, and at high enough values of ϵ the gap disappears, being completely filled with these states.

It is not easy to extend these results to three dimensions. Here the tendency

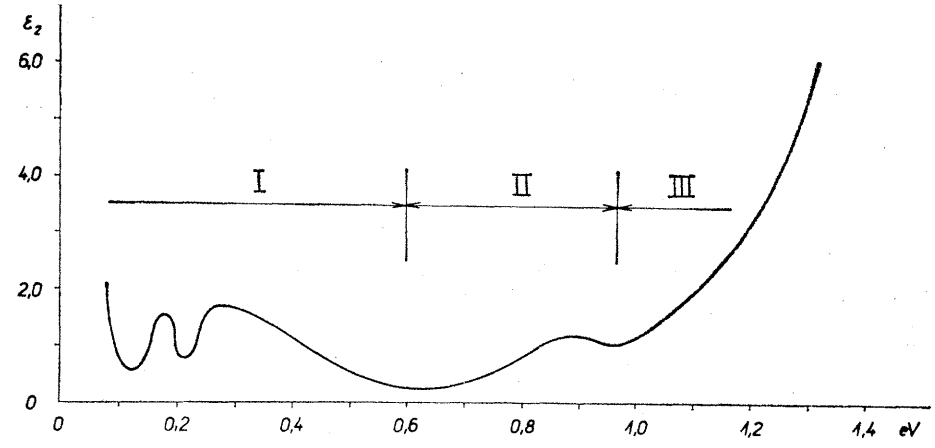


Fig. 3. Absorption in amorphous germanium. [After Tauc *et al.* (*8*)]

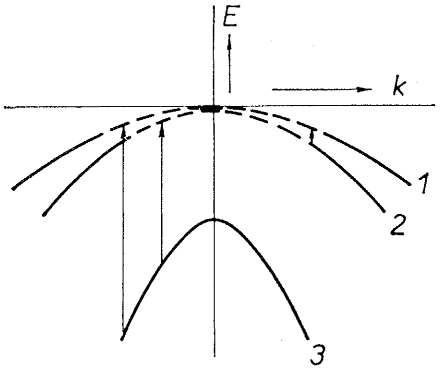


Fig. 4. Detailed band structure at the top of the valence band of germanium. (Dashed lines) Vacant upper states; (arrows) direct electron transitions into these states.

to form localized states is certainly smaller, because in the one-dimensional case any perturbation gives rise to a localized state, whereas in the three-dimensional case it does so only if the perturbation is deep enough and sufficiently extended spacially. Intuitively, we can imagine that in the one-dimensional case every wave is reflected by an obstacle, whereas in the three-dimensional case the wave can bend and go around. Mott (*4*) considers it very probable that, in general, localized states caused by disorder may exist even in three-dimensional cases.

Optical Properties

We may expect that with amorphous materials, as has been true with crystals, investigation of the optical properties will give us information on the electronic states. It is known that metals show little change of optical properties on melting (*5*). The optical properties in molten metal are determined by conduction electrons and are, in many cases, very well described by the simple classical formulas deduced by Drude—in fact, better described than the optical properties in solid metals, in which the anisotropy of the band structure complicates the situation. In glasses, little optical work has been done on amorphous and crystalline phases of the same material, to see the difference. This difference has been studied in elemental semiconductors such as selenium, tellurium, germanium, and silicon. As a simple example I discuss germanium, and the recent work of Grigorovici, myself, and our collaborators (*6–8*).

Figure 3 shows the dependence of the imaginary part of the dielectric constant ϵ_2 on photon energy in the infrared part of the spectrum. For the comparison with theory, it is advantageous to consider ϵ_2 (rather than say the absorption constant) because the absorption of electromagnetic energy is proportional to ϵ_2.

In range I of Fig. 3 the spectrum has a structure which is very similar to that found in crystalline *p*-type germanium, except that the peaks are shifted about 0.1 electron volt toward lower energies. This spectrum in crystalline germanium has been interpreted as being due to direct optical transitions between the various branches of the valence band (Fig. 4). At room temperature the upper states in bands 1 and 2 of Fig. 4 have their electrons excited to the acceptors; electrons from band 3 can be excited to the free states in bands 1 and 2, giving rise to the two observed peaks in the absorption spectrum. The low-energy edge corresponds to the transitions from band 2 into band 1. It is natural to use the same interpretation for amorphous germanium, which has been known (*7*) for a long time to be *p*-type semiconductor with a high concentration of acceptors (of the order 10^{18} per cubic centimeter). These acceptors are probably atomic vacancies in disordered lattice; in fact, amorphous germanium is less dense than crystalline germanium. These vacancies may be filled by some foreign atoms, probably oxygen. The vacancies are known to act as acceptors in crystalline germanium.

This interpretation accords well with the observed spectra. It is important to realize that it is essentially based on the conservation of the **k** vector and that therefore the **k** vector has a meaning for the electron wave functions in the valence band. This means that the ground-state wave functions are practically Bloch functions extending over a large volume.

Let us now consider region III of Fig. 3. The absorption is here due to transitions from the top of the valence band to the bottom of the conduction band; it is the so-called absorption edge. It has been shown (*6, 7*) that, in the photon-energy range 1 to 1.6 electron volts, the quantity $\hbar\omega(\epsilon_2)^{\frac{1}{2}}$ is a linear function of the photon-energy $\hbar\omega$. Such a dependence is typical for indirect transitions—that is, transitions for which the **k** vector is not conserved. This observation can be interpreted as indicating that the wave functions of the conduction-band states into which the transitions take place are linear combinations of the conduction-band Bloch wave function with **k** in a certain range. The observed absorption is then given as a sum of transitions for each of which the **k** vector is conserved. Such considerations give an estimate concerning the range of the **k** vectors from which the wave functions at the bottom of the conduction band are formed. Under certain assumptions (*7*) it can be shown that this range is of the order 10^{21} cm^{-3}. According to the uncertainty relation of the quantum theory, this result can be expressed in a more pictorial way: the conduction-band wave functions extend over a volume $(2\pi)^3/10^{21}$, or $\sim 10^{-19}$ cubic centimeter, which contains only several thousands of atoms.

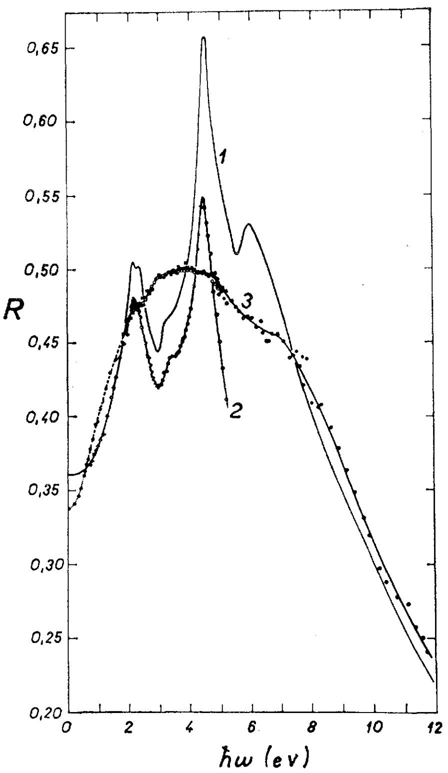
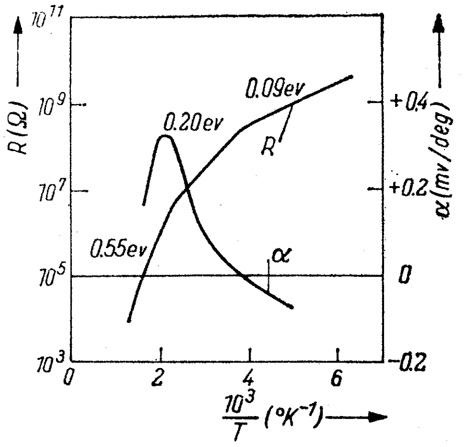


Fig. 5. Reflectivity *R* of amorphous germanium (curve 3) compared with the reflectivity measured on a single crystal (curve 1) and on a polycrystalline layer obtained by evaporation on a heated substrate (curve 2). [After Tauc *et al.* (*6*)]

Figure 5 shows the reflection spectrum of crystalline germanium compared with that of amorphous germanium in the region of fundamental absorption (valence-band → conduction-band transitions). We note that the spectrum of crystalline germanium shows a sharp structure which is due to the exact conservation of the **k** vector during the transition. Such a structure is absent in the spectrum of amorphous



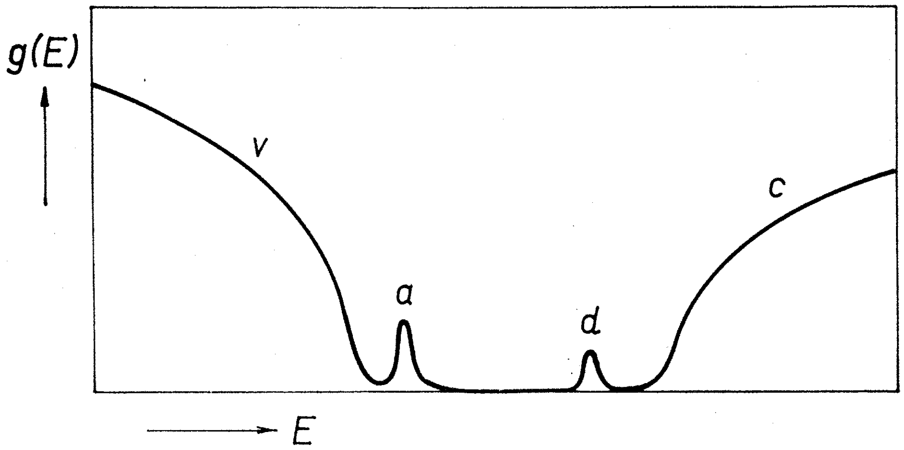


Fig. 6 (left). Temperature dependence of the resistance, R (in ohms), the thermoelectric power, α, in an amorphous germanium film. [After Grigorovici *et al.* (*10*)] Fig. 7 (right). The density of states $g(E)$ in amorphous germanium as deduced from the experiments discussed. (*v*) Valence band states; (*a*) acceptor states; (*d*) donor states; (*c*) conduction band states.

germanium, but in general appearance the two spectra are similar. Again, this can be understood if we assume that the wave functions in amorphous germanium are formed as linear combinations of the crystalline wave functions of the same band and that therefore the **k** vector is not conserved during the transitions.

The tail observed in region II of Fig. 3 is believed to be due to the absorption on lattice defects—the atomic vacancies acting as acceptors, as mentioned above.

Transport Properties

Many amorphous materials (for example, oxide glasses) are insulators, but many of them conduct electricity. It has been known for a long time that the electrical conductivity of metals decreases somewhat on melting, and the decrease can be in most cases satisfactorily explained in terms of the changes in atomic vibrations (*4, 5*). Amorphous selenium, tellurium, germanium, silicon, and the many chalcogenide glasses behave like semiconductors (*9*). Let us again consider amorphous germanium. It always shows *p*-type conductivity, due very probably to atomic vacancies acting as acceptors. Its conductivity appears to be little influenced by admixture of impurities in low concentrations, but no systematic work on the role of impurities has yet been done.

Figure 6 shows the temperature dependence of the electrical resistance of a film of amorphous germanium. Also, Fig. 6 shows a plot of the thermoelectric power; its sign gives the sign of the charge of current carriers. We see that at low temperatures the current is carried by negative charges. Grigorovici (*10*) suggested that in this region this is due to the hopping of electrons between acceptors, discussed above. The electrons originate from donors whose existence in amorphous germanium is assumed; these may be interstitial atoms, which are known to act as donors in crystalline germanium.

At higher temperatures the thermoelectric power becomes positive because the conductivity of holes at the top of the valence band, when the valence electrons are thermally excited, predominates over the impurity conductivity. At room temperature the conductivity is about 10^{-2} (ohm-cm)$^{-1}$, a value much smaller than that we would expect if we accept the conclusion reached from the optical measurements that the wave functions at the top of the valence band are practically Bloch functions. Indeed, Gubanov estimates the free mean path of an electron (or hole) in a disordered structure to be of the order a/ϵ^2, which, for a typical value $\epsilon \sim 0.1$, gives a conductivity of the order 10^2 (ohm-cm)$^{-1}$.

To explain the observed facts, Banyai (*11*) suggested that the states at the top of the valence band and the bottom of the conduction band are localized up to energies of about 0.2 electron volt in each band. However, later observations of the *p*-bands have shown that the wave functions at the top of the valence band are nonlocalized. Therefore another mechanism for lowering the conductivity must be looked for. Stuke (*4*) has suggested that, as in the case of selenium [even crystalline selenium (*12*)], the conductivity is probably lowered by the presence of potential barriers which the carriers pass over by means of thermal excitation. Indeed, a barrier of height 0.2 electron volt would introduce, at room temperature, a factor $\exp(-E/kT) = \exp(-0.2/0.025) \simeq 3.10^{-4}$, which would explain the difference between the measured and the expected conductivities. The origin of these barriers may be fluctuations of concentrations of vacancies (that is, acceptors), fluctuations of internal stresses which change the energy gap, or the presence of macroscopic regions formed during the growth of the layer.

The last part (the high-temperature part) of the conductivity curve of Fig. 6 has an activation energy of 0.55 electron volt and is explained as being the intrinsic conduction, with an energy gap of 1.1 electron volt [higher than the optical gap (0.9 electron volt), the difference being due perhaps to the presence of barriers].

In this way we have obtained a picture explaining the properties of amorphous germanium considered here. The corresponding density of the band states is shown in Fig. 7. The suggested picture is probably too simple and should be looked upon as a first hypothesis. There are effects which are difficult to understand on the basis of this picture. The sign of the Hall constant is negative (*13*) at the temperature at which the sign of the thermoelectric power is positive, although in crystals the two signs are the same; this difference seems to be a general property of amorphous semiconductors. In amorphous germanium there exists a photoelectric effect in the spectral regions where absorption takes place (*13*), but it is very weak, and this weakness seems difficult to reconcile with the idea of the presence of barriers. It appears that much more experimental and theoretical work must be done before the properties of even the simplest amorphous nonmetallic materials are completely understood.

Ovshinski (*14*) found a particularly useful transport effect in chalcogenide glasses and applied it to the design of a new device called the Ovonic switch. The device consists of an amorphous film between two electrodes. It has a high resistance at voltages below a certain threshold voltage. If the threshold voltage is surpassed, the resistance drops by several orders of magnitude. The transition time is exceedingly rapid (less than 150 picoseconds), and the effect is completely reversible. It was found empirically that amorphous materials are far better for this purpose than crystalline materials. Owing to the simplicity of production, the insensitivity to radiations, and characteristics useful for many applications (*15*), the Ovonic switch may find an important position among modern semiconducting devices. Ovshinski observed other unexpected effects in chalcogenide glasses, which may be used in the design of devices; for example, with some materials a device constructed along the lines of the Ovonic switch can remain in the blocking state or the conductivity state for a very long time and can be switched from one state to the other by an electric pulse. The nature of these effects is only partially understood.

Summary

A possible approach to the understanding of electronic properties of amorphous materials is to compare them with the corresponding crystalline materials, whose properties have been well explained. This approach has been exploited in the simple case of amorphous germanium, and I have indicated how the observed optical properties can be used to obtain information on the changes of electronic states, and what complications arise when we try to understand the transport properties.

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NEWS AND COMMENT

The Synthesis of DNA: How They Spread the Good News

1. Public Relations Aspects

Several months ago a research group, headed by Arthur Kornberg of Stanford University, concluded that it had succeeded in a long-standing quest—the in vitro synthesis of biologically active deoxyribonucleic acid (DNA). Kornberg and his colleagues, Robert L. Sinsheimer of California Institute of Technology, and Mehran Goulian, a Stanford postdoctoral fellow now on the University of Chicago faculty, prepared a report of their findings. In September the report was submitted to the *Proceedings of the National Academy of Sciences* (PNAS) by Kornberg, who is a Nobel laureate and a member of the Academy. Publication was scheduled for the December issue, which would normally have come out around the third week of this month. But because of production delays connected with PNAS's end-of-the-year index, the issue is not expected to go to press until early January.

That the work of the Kornberg group would eventually attract great attention within the scientific community was assured by the significance of the findings, Kornberg's reputation, and the prestigious place of publication. How it might fare in the outside world was an altogether separate question; for, despite the belief that the public ought to care about basic research and, therefore, does, it can scarcely be said that the man in the street has his transistor tuned for the latest word from the workshops of DNA research. Nevertheless, before news of the Kornberg report had an opportunity to become visible through the normal channels of scientific communication it had become front-page news, on 15 December, throughout the nation. In accompaniment to this news, two traditionally reticent government agencies, the National Institutes of Health and the National Science Foundation, publicly announced that they had shared the costs of Kornberg's work. And, on the eve of the appearance of the newspaper stories, no less a figure than the President of the United States had a last-minute insertion made in a speech to laud the Kornberg group for having "unlocked a fundamental secret of life." Speaking at the Smithsonian Institution on the occasion of the 200th anniversary of the *Encyclopaedia Britannica*, President Johnson advised his audience to look to the newspapers the next day for "one of the most important stories you ever read." On 17 December the New York *Times*, having had a few days to ruminate on the implications of the Kornberg report, informed its read-