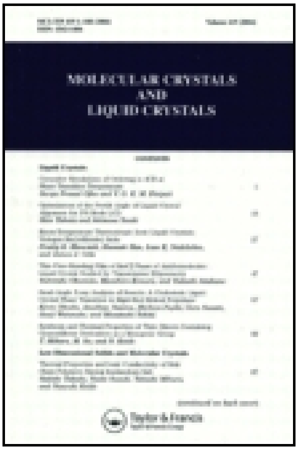

Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

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Studies of Organic Semiconductors for 40 Years—III

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Studies of Organic Semiconductors for 40 Years—III

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In this article the author takes upon himself the task of unrolling a local history of organic semiconductors in GERMANY. In doing so, he is well aware of the fact that describing a national development can only highlight isolated spots which are detached from the universal context. A composition of these mosaic stones to reflect the international interrelations of scientific thinking and searching, consisting always of giving and taking at the same time, can only be achieved by a synopsis of all the contributions within this series. In this connection the critical review by Pope and Swenberg[1] is worth mentioning which has solved this task for the years until 1980 in an outstanding way; a compressed update of the same authors covers the time until 1983.[2] A data compilation of selected experimental results may be found in Reference [85K1].

In the list of references of the present contribution only such work has been incorporated that had its physical or mental origin in Germany. In view of the great number of joint collaborations among authors of different nations, however, the borderlines will appear rather artificial and arbitrary in some cases. —The author has tried to completely cover the early literature from the very first beginning until 1969. The very rapid increase in the subsequent years, however, forced selectivity.

PREHISTORY

Photoconduction in organic crystals was discovered long time ago at the beginning of this century (1906) by the Italian scientist Pochettino,[3] who did experiments with anthracene, a material which much later (~1960) became the prototype of an organic photoconductor for over 25 years or so. Two review articles have been devoted solely to this substance (Kepler,[4] Karl [75K1]).

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[1] Pope, M., and Swenberg, Ch.E., *Electronic Processes in Organic Crystals* (Clarendon Press, Oxford 1982).

[2] Pope, M., and Swenberg, Ch.E., *Ann. Rev. Phys. Chem.*, **35**, 613–55 (1984).

[3] Pochettino, A., *Accad. Lincei Rend.*, **15** (1), 356 (1906) and **15** (2), 171 (1906).

In Germany a problem of interest at that early time was whether such phenomena as photopolymerization and fluorescence of organic substances had to do with a preceding photoionization of the molecules. In order to clarify these questions, Stark and Steubing [08S1] (see also [10S1]) at the University of Greifswald, and Byk and Borck [10B1] at the University of Berlin investigated the external photoelectric effect of solid anthracene and of a number of other fluorescing organic materials. At the same time (1910) Königsberger and Schilling at the University of Freiburg/Brg. performed experiments on the electrical dark-conductivity of several organic compounds [10K1]. These authors could not yet foresee that silicon, whose conductivity they measured between −190 and 850°C at the same time, should become much more famous than anthracene, which they studied between room temperature and several degrees above its melting point. Whereas the dark resistivity of the latter was "infinite" at room temperature for the experimental facilities available at that time, conductivity begun to become measurable at 200°C on the *quadrant electrometer* used, which had a deflection of "0.7 mm per 0.001V" (!). The conductivity increased by about one order of magnitude until the melting point at 213°C and slightly more above. One remarkable conclusion was that of the *electronic* nature of conductivity, derived from the absence of electrolytic polarization and hence ionic conduction. In 1913 W. E. Pauli[5] in Göttingen studied the external photoeffect in anthracene as a function of the exciting wavelength [13P1]; in course of his experiments he concluded from the charging and discharging behaviour of his samples (observed on an electrometer) that there was a room temperature dark conductivity as well. M. Volmer, writing his Habilitation thesis at the University of Leipzig, reported at the same time [13V1] that photoemission from solid anthracene into vacuum required a wavelength shorter than 225 nm (5.5eV) and was therefore obviously not involved in the normal process of fluorescence emission. Simultaneously he had thus determined for the first time a (solid state) ionization threshold of a highest occupied molecular orbital (HOMO), for which the best present day's value is 5.75(10)eV, cf. [85K1]. It is also remarkable that he made an organic solid state photodetector by melting thin anthracene layers between two platinum wires on a fused silica support. In contrast to the external photoeffect, this photoconductivity set in already at longer wavelengths, parallelling the anthracene VIS/UV absorption spectrum which begins at 400nm. In view of potential molecular electronic devices, one minor detail still deserves mentioning: protection against the attacks of oxygen and humidity could be achieved by a thin cover layer of paraffine, which did not degrade the photoelectric properties of this photocell.

From then on anthracene seems to have been forgotten for nearly 40 years, until its scintillator properties, its potential use as (electrical) crystal counter for high energy particle radiation and its model character for organic dyes used for sensitization of alkali-vacuumphotocells and silver halide photographic emulsions were recognized.

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[4] Kepler, R. G., *Organic Molecular Crystals: Anthracene*, in: ed. Hannay, N. B., *Treatise on Solid State Chemistry*, 3 (Plenum Publ. Corp. New York 1976), 615–678.

[5] He is not the inventor of the "Pauli Principle."

THE ANTHRACENE AND NAPHTHALENE ERA

Dark and photoconductivity in presence of trapping and space charge

H. Pick of the colour center group of R. W. Pohl at Göttingen seems to have been among the first in this country who extended their attention to organic semiconductors at that time. The need for careful material purification and *single* crystal growth was clearly recognized by him. With these prerequisites the anisotropy of dark conductivity in anthracene was determined as $\sigma_a : \sigma_b : \sigma_{c^*} = 1 : 1 : 0.19$ (at 148°C) [53M1], a ratio which is surprisingly close to the (28°C, sign-averaged) mobility anisotropy, measured with much greater effort much later (cf. [80K2]).

Dark and photoconductivity studies of anthracene were continued by several authors [57R1, 60B1, 61B1, 62H1, 65B1, 65B3, 67B1, 69B3, 70B1] and extended to other aromatic crystals, such as naphthalene [54P1, 62B1], stilbene [56D1], and pyrene [64B1, 65B2]. DC photoconductivity measurements were soon complemented by pulsed time-of-flight experiments, first introduced into the organic field by Kepler[6] and by LeBlanc.[7] With well zone-refined (cf. [80K1]) and highly perfect single crystals, allowing charge carriers to drift across macroscopic distances (~1mm) without being captured by chemical or structural traps, the time-of-flight method proved to be a very versatile technique not only for the determination of anisotropic electron and hole mobilities [62H2, 64B1, 65B2, 67B3, 67B4, 70H1], but also for obtaining additional kinetic information on generation, recombination and trapping of charge carriers [66B2, 67B3, 67B4, 68K1, 70K1, 72H2].

There is no problem in the field of organic photoconductors on which more time and paper has been spent over all the years than on that of trapping. Trapping leads to space charge and thus distorts the external electric field. Injection of space charge [66R1], space charge-limited currents [62M1, 62M2, 67H3, 69B2], transport with multiple shallow trapping (in conjunction with thermally activated detrapping) [69S1, 75P1], thermally [64H3] and optically [75R1, 76K1, 78K1] stimulated currents, and the question of the chemical and physical causes [68B4, 75P1] and the energetic distribution function of traps [63H2, 66B1] have worldwide kept busy generations of researchers (and still do so). This is especially true for materials which cannot (or could not yet) be brought to a sufficiently high purity level (an overall purity of better than 99.9999% is often required for the study of the intrinsic transport mechanisms in organic photoconductors, cf. [69S1, 75P1]).

ESTABLISHING THE BASIC PRINCIPLES OF PHOTOCONDUCTION

Light, excitons, electrodes and dye layers

The energy levels. First of all, ultraviolet photoelectron spectroscopy, UPS, is a powerful method to determine the valence levels. E. E. Koch seems to be the only one in this country who has applied this method on organic photoconductors [79G1]. Photoinjection from metal electrodes with different Fermi levels has been

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[6] Kepler, R. G., *Phys. Rev.*, **119**, 1226 (1960).

[7] LeBlanc, O. H., Jr., *J. Chem. Phys.*, **33**, 626 (1960).

used to determine the relative energy of the valence band edge and of the conduction band edge, and to derive from these data a band gap value for anthracene [68B1, 68B2, 68B3, 68V1, 68V2, 69B1]. Dark injection from electrolytic contacts with suitable redox potential can also give estimates of band energies [67B2, 69M1, 69M2]. Trials to learn about bands from hot carrier injection have been made [72B1, 77S1]. From an evaluation of autoionization data of electronically excited molecules a band gap of anthracene of 4.0eV and of tetracene of 3.1eV has been obtained, in fair agreement with several other approaches. Transitions to homo charge transfer states, detected by electromodulation spectroscopy under the strong intramolecular S_0–S_1 absorption, have been used to extrapolate to the energy needed for full charge separation [81S3, 83S2]; distinction has to be made between optically created unrelaxed and adiabatically reached states, cf. [87S6].

Intrinsic charge carrier generation and recombination. There is a general agreement since the ingenious investigations by Ch. Braun, USA, that intrinsic charge carrier photogeneration in substances like anthracene is a stepwise process, where first an initial "geminate" electron-hole pair, still bound by its mutual Coulombic interaction, is created, which in turn is subject to the competing processes of thermally activated and electric field-assisted full separation, and geminate recombination. The details, however, namely the question of how and where the initial pair is formed, and how the final diffusive process is correctly described (dimensionality, dielectric anisotropy, cf. [71K2], scattering length, hopping site energy distribution in disordered materials, etc.) have called for additional activities [83R1, 87R1, 87S6]. Moreover, charge carrier pair generation at metallic, electrolytic and dielectric electrodes needed separate investigation [73H1, 74M2, 75H3, 75G1, 78C1]. Magnetic field modulation has been used as one special tool [82P1], high time resolution as an other [82E1]. Recombination of non-geminate electrons and holes constitutes an additional loss channel, which is important e.g. whenever charge carriers are created at high density by strongly absorbed light; using the initial recombination losses before the charge clouds of opposite sign are separated by a field, allows one to extract the bimolecular recombination constant [71K1]. A nice alternative is to let electron and hole clouds drift through each other and to observe the losses time-resolved [67B3], cf. [74K1].

The role of excitons and sensitizers. The energy of a single exciton, albeit not sufficient for carrier *pair* separation, can be used to create a *single* free charge carrier, whenever an extrinsic level is available which lies energetically sufficiently deep in the band gap to compensate for the lacking energy. Intrinsically, singlet excitons can create mobile charge carriers only via bimolecular (in the light intensity quadratic) collisional processes [69B3, 71B1]. At electrodes with suitable levels (Fermi level of a metal [71K3, 72K1, 72K2, 74S1, 75G2], or molecular level of an adsorbed dye molecule [68G1, 81M1, 81W1]) excitons can dissociate and leave behind one free charge carrier in the organic crystal; hot holes created optically in a metal electrode have also been made responsible for charge carrier injection by light [75G1]. Excitation of an adsorbed dye molecule as a first step is an alternative possibility [76G1]. Hole injection from an excited dye aggregate through an alkane Langmuir-Blodgett barrier to anthracene end groups, adsorbed on an anthracene crystal, has been studied with 100 ps time resolution [87W1]. Interaction of triplet excitons with trapped charge carriers can not only lead to free carriers, which can be sensitively (10^{-14} A) detected as a current thus allowing an elegant measurement of triplet absorption spectra [75R1], the interaction also involves a nonstationary (quartet/doublet) spin pair state, which gives rise to a modulation of the photocurrent by a magnetic field vector. The full triplet fine structure tensor of the anthracene molecule in the Z-1 unit cell of an anthracene: pyromelliticdianhydride single crystal has thus been determined [79Z1].

Charge carrier transport. Charge carrier transport requires characterization by the full, temperature dependent electron and hole mobility tensors, if necessary (non-Ohmic case) at specified field strengths. The measurement of intrinsic, non-trapping-influenced mobilities over extended temperature ranges and down to low temperatures was only possible as extremely high purity single crystals of naphthalene and perylene became available [85K2, 85K3, 85W1, 85W2, 85W3], cf. [84K1, 85K1]. The result, the first observation of non-Ohmic band transport with comparatively high mobilities (several hundred cm^2/Vs) and saturation of the drift velocity at high electric fields at about $2 \cdot 10^6$ cm/s justified the extreme purification efforts (up to 500 zone passages in several zone refining steps performed after careful prepurification), cf. [85W1]. A saturation of drift velocities was also observed for the room temperature drift velocity of holes in anthracene at ultrahigh fields, thereafter [87B3].

ORGANIC DYESTUFFS AND PIGMENTS

Search for stable big molecules

Basic work. Photoconduction in (solid) organic dyestuffs and pigments is an old topic (H. Akamatu, Japan, D. D. Eley, UK, A. T. Vartanyan, USSR). In this country it is intimately connected with the name of H. Meier [58M1, 58M2, 59M1, 59N1, 65M1, 69M3], (see also his book review [74M1]). In spite of lack of high purity single crystalline material, the general trends discovered with pressed powder pellets of different classes of materials are remarkable. Doping with traces of electron acceptors, such as chloranil or iodine, was frequently found to increase photoconductivity by orders of magnitude [63M1].

The class of phthalocyanines, closely connected with the name of C. Hamann [63H1, 64H2, 66H1, 67H1, 67H2, 67S1, 71H1], deserves special mentioning because of the very high stability of most of its representatives. Thin layers can easily be made by vacuum evaporation [68H1]; single crystal growth is possible from the gas phase [67H1, 71H1]. Rectifying contacts [64H1], gas sensors [65K1] and on/off switching of high conductivity [78H2, 84F1] have been reported for lead phthalocyanine. Great activities with chemically modifying phthalocyanines and porphyrines to create new properties, are presently under way in the group of M. Hanack [85M3, 85M4, 86H1, 86H2, 87H3], see also [87H2, 87H4].

Routes towards high quantum yield solar cells. Thin dye films on inorganic semiconductors have been considered as promising candidates for photovoltaic and photoconductive devices. Thus copper-phthalocyanine on CdS and rhodamine B on AgI gave −220 mV and +350 mV (on the inorganic side), respectively [64M1, 65M2, 65M3], see also [76H1, 82V1]. In order to push up quantum yields, the primary geminate electron-hole pairs have to be hindered from recombining. A gradient of the electrochemical potential and a sufficiently large initial separation have been recognized as being essential. Along these lines F. Willig and coworkers performed ingenious experiments with molecular assemblies, consisting of 3 different chromophors in successive molecular layers on a perylene single crystal. These gave a current yield of 0.3 holes per absorbed photon for red light. The chromophors were arranged in such sequence that a stepwise change of the standard redox potential (with a total change of 0.5 eV) was obtained [86W1]. Very detailed microscopic understanding achieved recently for the initial charge separation process in bacterial photosynthetic reaction centers by Fischer and Scherer [87F1] will certainly allow to learn from nature how to further optimize these assemblies.

THE RADICAL ION SALT AGE

Materials which unite semiconductivity, metallic conduction and superconductivity

Excited state versus ground state charge transfer. The combination of donor-like molecules, i.e. molecules with *high* lying filled HOMO levels (in a downward electron binding energy scale with zero at vacuum level), and acceptor-like molecules, i.e. molecules with comparatively low lying empty LUMO levels, opens up the possibility of *inter*molecular transitions of an electron from the HOMO level of one molecule to the LUMO level of an adjacent molecule, cf. [61B2]. In the so called donor:acceptor complexes this transition needs much more than kT thermal energy and thus occurs only on optical excitation. Depending on the HOMO and LUMO energies [59B1], these optical charge transfer transitions, however, need less, or even much less photon energy than the intramolecular transitions and can therefore lie in the visible or even near IR. Nevertheless, an electron is transferred over one intermolecular distance. Final charge carrier separation (from the CT-geminate pair) and transport in this class of materials has only recently been adequately treated, namely in well purified material (anthracene: pyromelliticdianhydride) [75K1, 87M1, 88M1]. Charge transfer transitions also occur in one-component crystals (but are usually hidden under the strong intramolecular electronic transitions). Their detection by electromodulation spectroscopy has been the merit of G. Weiser and his colleagues [81S3, 81S4, 83S2].

Highly conductive materials. When donor and acceptor type molecules with extreme positions of the HOMO and LUMO levels (i.e. with extreme reducing and oxidizing potential) are combined (these molecules are at the borderline of chemical stability), partial or full charge transfer can require less energy than the Madelung energy (and other minor contributions) gained by stabilization of the system through formation of an ionic (or partially ionic) lattice. An "open shell" radical-ionic ground state is therefore formed and the partially filled bands give rise to considerable dark conductivity. Frequently (but not necessarily) the donor and acceptor molecules are arranged in separate linear stacks; the counter ions can also be inorganic ions, such as e.g. PF_6^-; the organic stacks can then be separated to such extent that conduction is strongly one-dimensional (or in some cases two-dimensional, see below). Several semiconducting TCNQ complexes were investigated in the early days in the group of H. Hänsel [69M6, 69Z1, 70Z1, 70Z2, 72M1, 72M2, 72Z1], methyl-phenothiazine:iodine and methyl-triphenylarsonium:TCNQ (and other TCNQ complexes) in the group of H. W. Helberg [74D1, 75K3, 76R1, 85P1], who have specialized in studying the microwave complex dielectric tensor components in order to extract the dielectric and the conductivity tensors, see also [87K2].

The first electrochemical synthesis of an organic$^+$/inorganic$^-$ radical ion salt in Germany was performed by H. P. Fritz et al. in 1978 [78F1]. These authors determined the X-ray crystal structure and reported a pressed pellet room temperature conductivity of $(\text{naphthalene})_2^+ \, PF_6^-$ of $\sigma = 0.12$ S/cm and a narrow ESR line of 0.2 Gauss width, which decreased down to 0.0025 Gauss in perfect single crystals in later measurements in the group of J. von Schütz [83M1] and thus can be used for very accurate spin resonance measurements of magnetic fields, cf. [83D1, 86D1]. The $2 \cdot 10^{21}$ spins per cm^3 display a metallic-like Pauli susceptibility above 240K, where a metal to semiconductor phase transition occurs on further cooling. A narrow ESR linewidth, correlated with metallic conductivity, was also observed by M. Schwoerer and coworkers in fluoranthenyl radical ion salts [81E1] (made in the group of G. Wegner [80K4]; a number of conductive perylene radical ion salts were synthesized in the group of H. J. Keller and studied by D. Schweitzer and colleagues [80K3]. These findings, together with the discovery of organic superconductivity by the Danish-French cooperation Bechgaard/Jérome (1980), initiated a true rush of synthetic activities, crystal structure determinations [82E2, 82E3, 87E1], ESR and NMR investigations [82S1, 83D2, 83S4, 84D1, 85D1, 85M1, 85M2, 85S1, 86M2, 86S3, 87D1, 87D2, 87D4, 87H6, 87M2, 88H1], conductivity measurements [83S4, 85S3, 87S1] and other characterization work [82H1, 83G1]. These results can be summarized as follows: From Overhauser shift measurements in ESR and (more precisely) from Knight shift measurements in NMR of electron and nuclear spin resonance frequencies the existence of metallic conductivity is clearly established in the high temperature phases of many of these radical ion salts; ESR pulse–echo experiments in a magnetic field gradient, performed in the group of M. Mehring, have yielded the electron diffusion tensor (1.6 and 0.001 cm^2/s for directions parallel and perpendicular to the stack axis, respectively, in $(\text{fluoranthene})_2^+ \, X^-$, where $X = AsF_6$, SbF_6, PF_6, etc.; [87M2], and ^{13}C and ^1H Knight shift measurements the atomic orbital contributions to the conduction-band molecular orbitals [87M2]. These measurements also give valuable information on the lower temperature semiconductor properties which are smoothly approached. Shubnikov-de Haas quantum oscillations, recently observed with $(TMTSF)_2ReO_4$ [86S1], but not yet well understood, will probably allow the extraction of still more detailed information on band structure and transport properties in these highly conductive materials, and thus contribute to an understanding of their semiconductor properties, too.

The synthesis of N,N′-dicyanoquinodiimines, "DCNQIs", (molecules similar to the TCNQs), by S. Hünig and his coworkers opened up a completely new field of organic semiconductors and metals [86A1, 86S2, 87A1, 87S2, 88A1, 88E1, 88K1, 88S2, 88T1, 88W1]. The copper radical *anion* salt of a DCNQI-derivative, (2,5-dimethyl-N,N'-DCNQI)$_2$ Cu, remains metallic down to 0.45 K (whereas most others transform to semiconductors between 130 and 70 K on cooling). It exhibits the highest conductivity ever measured in the organic field, 500 000 S/cm at 3.5 K [86A1].

Electrochemical synthesis of another very exciting radical ion salt, di[bis(ethylene-dithiolo)tetrathiafulvalene] triiodide, $(\text{BEDT-TTF})_2^+\ I_3^-$, exhibiting a crystal structure with two-dimensional interactions among the organic molecules and hence a nearly isotropic conductivity (60–250 S/cm) in the crystallographic *ab* plane, was described 1984 by H. J. Keller, D. Schweitzer and coworkers [84B2]. If pressure (12 kbar) is applied, its α-modification remains metallic down to 0.1 K; without pressure a phase transition to a semiconducting state occurs at 135K [85S4], cf. [87S7]. A β-modification, on the other hand, was found to exhibit ambient pressure *superconductivity* below 1.05 K [85S4, 85S5], in accordance with the initial observations by Yagubskii et al., USSR. Meissner effect measurements confirmed true volume superconductivity but indicated higher transition temperatures under pressure [85V1]. After a special temperature/pressure cycling an obviously stable ("α_t") modification could be obtained with a sharp transition at 8.1K to an ambient-pressure homogeneous superconducting state [85C1, 87S3, 87S4, 87S5]. A recent report in literature (Urayama *et al.*, Japan) on a 10K organic superconductor, (BEDT-TTF)$_2$Cu(SCN)$_2$, was confirmed and the system further characterized [88G1]. —Notice that the transition temperature of organic superconductivity has risen with a slope of 1.2K per year since its first discovery in 1980!

THE POLYMERIC WORLD

Order in disorder

Normal polymers combine ordered structures with disorder in that more or less extended sections of the polymer chain possess well defined translational periodic order, on the one hand, whereas, on the other, they carry kinks then and when which interrupt the order. In addition there is usually a certain spread of chain lengths. Several neighbouring chains can be parallel over a certain length, thus forming crystalline islands, which are embedded in a fully amorphous or a partially oriented matrix. These features give polymers their attractive mechanical and processing properties, but render them extremely difficult objects for electrical transport investigations. Disorder (and omnipresent impurities) lead to broad distribution functions of the energy levels, with concomitant dispersive transport (and trapping). Polymers with no conjugated double bonds belong to the best insulators, yet slow motion of injected electronic space charge is possible (besides residual ionic conduction). Temperatures and field dependences have been studied by space charge limited ("SCL") current and by thermally stimulated current ("TSC") techniques [83S1, 84S1, 85S2], yielding information on the energy level (trap) distribution function and on transport [80H1, 80H2, 87B2]. Charge carrier mobilities are usually *very* low and strongly increase with increasing crystallinity [85H1]. A very detailed consideration of the structural, dynamic, and long time characteristics of the respective polymer sample is necessary, before models developed for the fully crystalline or fully amorphous materials can be applied [87H1], cf. [82S2].

Polymeric photoconductors. Photoconductive polymers usually contain π-electron systems. Conjugated polyenes with small but uniform chain length, such as e.g. the carotenes, have been used as model systems [65S1, 77B1] for the longer nonuniform polyacetylene (see e.g. the review [84R1]). An important question with the latter is, if the polymeric chains conduct as "molecular wires" and what the appropriate transport model would be. A "fast" component (μ = 2 cm^2/Vs), obtained from sub-nanosecond pulsed photoconductivity measurements in the group of S. Roth [83Y1] (cf. [88B1]) has very recently been attributed by these authors to polaronic transport [88B1] rather than to solitons (widely discussed in literature).

Polydiacetylenes (discovered in this country by G. Wegner) are distinguished by the unique feature that several representatives can be obtained as rather perfect single crystals by solid state polymerization of the monomer crystals. This fact has attracted H. Bässler's interest to use them as ordered one-dimensional model systems [76L1, 78L1] for studying generation, transport [78R1], recombination [76R2] and trapping [83S3] of charge carriers. In contradiction to other results reported in literature, no exceedingly high (time-of-flight) mobilities were obtained [78R1]. These findings could later be explained on the basis of a careful consideration of the one-dimensional physics on the polymer chain and the dispersiveness of the macroscopically observed transport [88B2].

Photoconduction of poly(arylenevinylene)s [78O1, 80O1], polymeric metalorganic compounds [86M1] and of poly(vinylcarbazole) [87K1, 87M3, 87S8] have been other topics of interest. Transport in the latter material, which has widely been used in electrophotography, has been especially carefully studied as a function of time over many orders of magnitude (10 decades of time) by D. Haarer and coworkers [87M3]. The "flat" (rather than exponential) trap distribution found, gives rise to the novel feature of a gradual transition from dispersive to nondispersive transport during a single transit. —Percolation aspects have been found important in polymers blended with a low molecular weight component [87D3].

Dark conductive polymers. Polyacetylene and many other polymers with double bonds can be made conductive by "doping," i.e. partial oxidation or reduction (cf. 84W1). These redox reactions can frequently also be performed electrochemically (cf. [87H5, 87W3]), in a similar way as in the preparation of small molecular radical ion salts, and studied by ESR, cf. [87W3]. Moreover, well defined single crystals of oligomeric representatives of the latter class, e.g. *p*-quaterphenyl—PF_6, can serve as ideal model systems for studying packing, molecular interactions and charge transport in the corresponding conductive polymers [85E1], cf. [87E1]. Poly(pyrrole)s [87B1], conjugated bridged macrocyclic metal complexes [86H3] and poly(phthalocyanine)s [87W2] have been other topics of interest. Two outstanding recent results deserve special mention: Stretch-aligned iodine-doped Narmann polyacetylene has reached the surprisingly high conductivity of 80 000 S/cm at room temperature with an anisotropy of 23:1 [88S1]. The measurements are perfectly described by the Sheng model. —Poly(pyrrole)s, surrounded with insulating cycloalkyl "spacers" of different size, have been synthesized in the group of G. Wegner and have demonstrated the overwhelming importance of *interchain* charge carrier hopping processes for the macroscopic conduction behaviour of polymers.

Transport and disorder. In the limit of a perfectly ordered crystal lattice, absence of impurities, and $T \rightarrow 0$K a coherent band transport should occur even in weakly bonded molecular solids, whereas disorder, impurities and more efficient charge carrier scattering at elevated temperatures would destroy coherency and lead to (much slower) hopping transport. Coupled coherent and incoherent transport, expected for intermediate parameters, has been treated in the classical papers by H. Haken, P. Reineker and G. Strobl [72H1, 73H2]. Charge carrier dispersive hopping transport in fully disordered media has been the subject of several publications of H. Bässler and coworkers [79S1, 81S1, 81S2, 82B1, 82L1, 84B1, 86R1]. The question of whether electronic transport by (charged) bond alternation defect ("soliton") mobility occurs in conjugated polymers with a right–left degenerate ground state, has led to a lively discussion. No soliton-like transport could be established in polyacetylene doped at different levels; a Mott type hopping was inferred, instead [86E1].

FUTURE TRENDS

Miniaturization down to the molecular level

After 40 and more years of basic research on organic semiconductors and conductors, which have lifted the veil of strangeness from this class of materials to a great extent so that they revealed many specific and sometimes unique fundamental properties, it is felt by many people working in the field, that the time is mature for seeking applications of what has been learned academically. In view of the additional very interesting nonlinear optical properties of a number of organic crystals and thin films, together with the unique photochromic and pyroelectric properties of several representatives, "molecular devices" have a real chance of coming into the realm of reality. There is general agreement that these will necessarily have to be *micro*circuits with an integration as high as possible. Thin organic films on inorganic semiconductors are one possible approach. But much activity is even directed to the molecular level, where molecular wires, molecular switches and molecular memory devices are expected. In this connection quantum well problems in molecular chain heterostructures are one of several interesting theoretical aspects [86R2, 87R2, 88R1]. Some of these ideas are presently being followed in a Stuttgart cooperation (see below).

RESEARCH PROGRAMS AND CONFERENCES

Research on organic semiconductors and conductors in Germany has been supported by the Deutsche Forschungsgemeinschaft and by the Max Planck Gesellschaft in individual contracts. From 1978 till 1988 the Stiftung Volkswagenwerk has generously financed an interdisciplinary research program "Physics and Chemistry of Unconventional Materials." 1986 a local cooperative program (Sonderforschungsbereich) at the Universities of Stuttgart and Tübingen, "Physical and Chemical Fundaments of Molecular Electronics," has been founded which is being funded by the Deutsche Forschungsgemeinschaft. The German activities on the organic materials field are reflected by several international conferences organized in this country (Molecular Crystal Symposium VI and IX, 1973 at Schloβ Elmau and 1980 at Mittelberg in the Northern Alps, respectively; Sixth International Conference on the Chemistry of the Organic Solid State, Freiburg/Brg. 1982; Emil Warburg Symposium on Unconventional Photoactive Solids, 1982 at Schloβ Mitwitz and 1987 at Schloβ Elmau), and by a number of symposia, seminars and workshops.

A considerable number of *review papers and books* [61B2, 65H1, 67B1, 74K1, 74M1, 75K1, 78H1, 80H3, 80K1, 81H1, 81W1, 83S1, 84B1, 84K1, 84W1, 85K1, 87E1, 87H2, 87H4, 87S3, 88B1, 88B2] document the activities in the two parts of Germany on the field of *organic* photoconductors, semiconductors, metals and superconductors.

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