10. C. Benoit à la Guillaume, D. Scalbert, T. Dietl, *Phys. Rev. B* **46**, 9853 (1992).
11. J. Okabayashi *et al.*, *Phys. Rev. B* **58**, R4211 (1998).
12. C. Zener, *Phys. Rev.* **81**, 440 (1950); *Phys. Rev.* **83**, 299 (1950). A similar model for the nuclear ferromagnetism was developed by H. Fröhlich and F. R. N. Nabarro [*Proc. R. Soc. London Ser. A* **175**, 382 (1940)].
13. T. Dietl, A. Haury, Y. Merle d'Aubigné, *Phys. Rev. B* **55**, R3347 (1997).
14. See, for example, G. L. Bir and G. E. Pikus, *Symmetry and Strain-Induced Effects in Semiconductors* (Wiley, New York, 1974).
15. See, for example, J. K. Furdyna, *J. Appl. Phys.* **64**, R29 (1988).
16. D. Bimberg *et al.*, *in Landolt-Börstein, New Series*, vol. 17a, O. Madelung, M. Schulz, W. Weiss, Eds. (Springer-Verlag, Berlin, 1982); I. Broser *et al.*, in *Landolt-Börstein, New Series*, vol. 17b, O. Madelung, M. Schulz, W. Weiss, Eds. (Springer-Verlag, Berlin, 1982); O. Madelung, W. von der Osten, U. Rössler, in *Landolt-Börstein, New Series*, vol. 22a, O. Madelung, Ed. (Springer-Verlag, Berlin, 1987); R. Blachnik *et al.*, in *Landolt-Börstein, New Series*, vol. 41B, U. Rössler, Ed. (Springer-Verlag, Berlin, 1999); I. Stolpe *et al.*, *Physica B* **256–258**, 659 (1998).
17. A. Twardowski, P. Świderski, M. von Ortenberg, R. Pauthenet, *Solid State Commun.* **50**, 509 (1984).
18. T. Jungwirth, W. A. Atkinson, B. H. Lee, A. H. MacDonald, *Phys. Rev. B* **59**, 9818 (1999).
19. Y. Shapira *et al.*, *Phys. Rev. B* **33**, 356 (1986).
20. M. Sawicki *et al.*, *Phys. Rev. Lett.* **56**, 508 (1986); P. Głód, T. Dietl, M. Sawicki, I. Miotkowski, *Physica B* **194-196**, 995 (1994).
21. H. Ohno, F. Matsukura, T. Omiya, N. Akiba, *J. Appl. Phys.* **85**, 4277 (1999).
22. D. Ferrand *et al.*, *J. Crystal Growth*, in press (e-print available at xxx.lanl.gov/abs/cond-mat/9910131).
23. See, for example, M. A. Paalanen and R. N. Bhatt, *Physica B* **169**, 223 (1991).
24. B. Beshoten *et al.*, *Phys. Rev. Lett.* **83**, 3073 (1999).
25. J. Blinowski, P. Kacman, J. A. Majewski, in *High Magnetic Fields in Semiconductor Physics II*, G. Landwehr and W. Ossau, Eds. (World Scientific, Singapore, 1997), pp. 861–864.
26. Parameters of ZnO were obtained by fitting the kp model (*14*) to energies calculated by D. Vogel, P. Krüger, and J. Pollmann [*Phys. Rev. B* **54**, 5495 (1996)] and by taking β(ZnMnO) = β(ZnMnSe).
27. T. Dietl, H. Ohno, F. Matsukura, J. Cibert, D. Ferrand, data not shown.
28. K. Kim, W. L. R. Lambrecht, B. Segall, M. van Schilfgaarde, *Phys. Rev. B* **56**, 7363 (1997).

29. The work at Tohoku University was supported by the Japan Society for the Promotion of Science and by the Ministry of Education, Japan.

19 October 1999; accepted 10 December 1999

Ambipolar Pentacene Field-Effect Transistors and Inverters

J. H. Schön,* S. Berg, Ch. Kloc, B. Batlogg

Organic field-effect transistors based on pentacene single crystals, prepared with an amorphous aluminum oxide gate insulator, are capable of ambipolar operation and can be used for the preparation of complementary inverter circuits. The field-effect mobilities of carriers in these transistors increase from 2.7 and 1.7 square centimeters per volt per second at room temperature up to 1200 and 320 square centimeters per volt per second at low temperatures for hole and electron transport, respectively, following a power-law dependence. The possible simplification of the fabrication process of complementary logic circuits with these transistors, together with the high carrier mobilities, may be seen as another step toward applications of plastic electronics.

Organic thin-film field-effect transistors (FETs) have been studied extensively throughout the last decade, and tremendous progress in performance of these devices has been achieved (*1–4*). Among these organic materials, pentacene has been found to have the highest mobilities for hole transport (p channel) (*5*, *6*). State-of-the-art organic thin-film transistors reach performances similar to those of devices prepared from hydrogenated amorphous silicon (a-Si:H), with mobilities around 1 $cm^2\ V^{-1}\ s^{-1}$ and on/off ratios surpassing 10^6, and with the use of high–dielectric constant gate insulators, operating voltages as low as 5 V can be achieved (*7*). These accomplishments demonstrate that the use of organic electronic devices may become feasible and desirable in areas in which large area coverage, mechanical flexibility, low-temperature processing, and overall low cost are required. Potential applications include low-end data storage, such as identification tags or smart cards (*8*), and even switching devices for active displays (*9*), especially because the integration of organic FETs and organic light-emitting diodes into smart pixels has been demonstrated (*9–11*). However, organic FETs have worked only as unipolar devices in accumulation or depletion, never in inversion. To exploit advantages of complementary logic, such as low-power dissipation, good noise margins, robust operation, and ease of circuit design, two different organic materials have to be used. The different semiconductors can be embedded into one heterostructure device (*12*, *13*) or into many separate devices (*14–17*), leading to all-organic digital circuits. The limitation of charge transport by only one carrier type is generally ascribed to effective trapping of the other carrier in the material itself as well as at the interface to the gate dielectric (*12*, *13*). Therefore, the use of ultrapure, high-quality materials seems to be a prerequisite to overcome this limitation.

Here, we report on organic FETs based on pentacene single crystals working as ambipolar devices both in accumulation (p type) and inversion (n type). High-purity pentacene single crystals were grown by physical vapor transport in a stream of hydrogen (*18*). Space-charge–limited current measurements (*19*) revealed trap concentrations (for holes) and acceptor densities as low as 10^{13} and 10^{11} cm^{-3}, respectively. Gold source and drain contacts (thickness of 50 nm) were evaporated through a shadow mask, defining a channel length between 25 and 50 μm and a width of 500 to 1500 μm. Al_2O_3 was deposited as gate dielectric layer by radio frequency–magnetron sputtering (capacitance $C_i \approx$ 30 nF cm^{-2}; thickness of 250 nm). Finally, the gate electrode (thickness of 100 nm) was prepared by thermal evaporation of gold (Fig. 1).

Typical device characteristics at room temperature of a pentacene single-crystal FET (Fig. 2) show the device working in both accumulation and inversion modes. The device operation of organic transistors is well described by standard FET equations (*20*), as previously shown (*7*). For accumulation (hole transport), the mobility is 2.7 $cm^2\ V^{-1}\ s^{-1}$, and the on/off ratio at 10 V is 10^9. Typical threshold voltages are in the range of −1 V. In combination with the steep subthreshold slope of 200 meV per decade (Fig. 3), this low threshold voltage indicates the high quality of the pentacene single crystal as well as the pentacene-Al_2O_3 interface. An electron mobility of 1.7 $cm^2\ V^{-1}\ s^{-1}$ and an on/off ratio of 10^8 are measured for operation in inversion. The higher threshold voltage of about 5 V reveals a higher density of traps for electrons than for holes. Nevertheless, n-channel transport can be obtained in pentacene devices. The observed field-effect mobility is similar to previous time-of-flight mobilities measured on related compounds such as naphtalene or anthracene (*21*).

Because no organic material has to be patterned, the use of ambipolar devices can substantially simplify the fabrication of complementary metal oxide semiconductor (CMOS)–

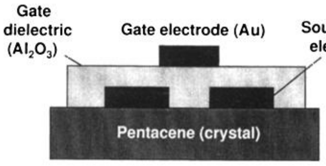

Fig. 1. Schematic structure of the FETs based on single crystalline pentacene. Gold and Al_2O_3 were used as electrode and gate insulator materials, respectively.

Bell Laboratories, Lucent Technologies, Mountain Avenue, Murray Hill, NJ 07974, USA.

*To whom correspondence should be addressed. E-mail: hendrik@lucent.com

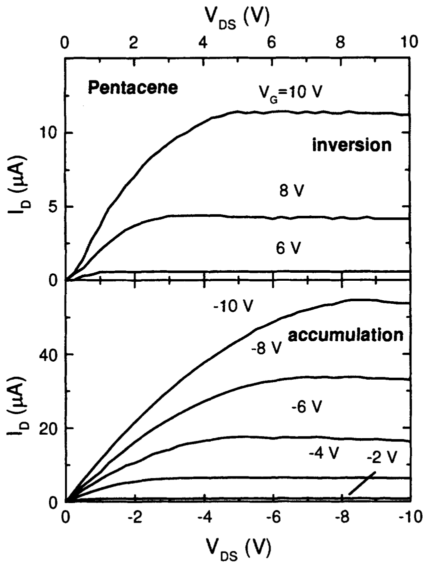

Fig. 2. Drain current (I_D) versus drain-source voltage (V_{DS}) characteristics at room temperature of a pentacene single-crystal transistor in inversion mode (n type, top) and accumulation mode (p type, bottom). V_G, gate voltage.

like circuits. Complementary circuits in which single transistors operate either as n- or p-channel device have been proposed and analyzed for a-Si:H–based FETs (*22*, *23*). From the transfer characteristic of a CMOS-like inverter circuit based on ambipolar pentacene FETs (Fig. 4), a gain as high as 10 has been measured. Moreover, inverters with a gain as high as 23 have been prepared with different metals as source and drain electrodes for n- and p-channel operation. Because of the high mobilities observed in the present devices, especially for n-type transport, a substantial improvement of switching speed (*15*, *17*) and performance of organic complementary circuits can be expected from the use of ambipolar transistors in addition to the simplification of processing.

The high quality of the pentacene single crystals and of the pentacene-Al_2O_3 interface opens up possibilities for studying the physics of charge transport in these organic semiconductors. Measurements on thin-film devices demonstrated a wide variation of the temperature-dependent mobility even for nominally similar preparation conditions, which is ascribed to trap levels, contact effects, and grain boundaries (*4*, *24*). However, it is worth mentioning that these extrinsic defects mainly influence the charge transport in thin films at low temperature. At room temperature, however, similar mobilities as in single crystals can be achieved in pentacene thin films. We prepared pentacene thin films by organic vapor phase deposition on flexible plastic substrates and used them as a basis for FETs, which also revealed ambipolar activity. Growth of large grain (>15 μm) films at elevated temperatures with low grain boundary trap densities seems to be the key parameter for such devices.

The field-effect mobility on single-crystal devices in which the influence of grain boundaries, traps, and residual disorder is minimized (Fig. 5) increases, following a power law from 2.7 or 1.7 $cm^2 V^{-1} s^{-1}$ at room temperature up to 1200 or 300 $cm^2 V^{-1} s^{-1}$ at low temperatures for holes or electrons, respectively. This temperature dependence and the very high values of the mobility at low temperature suggest that the charge transport is governed by band-like motion rather than by hopping processes. These results are in line with our temperature-dependent space-charge–limited current measurements on pentacene single crystals and also with time-of-flight measurements on naphtalene single crystals (*20*, *25*), where mobilities as high as 400 $cm^2 V^{-1} s^{-1}$ were measured at low temperatures. Therefore, it appears that on a phenomenological level, classical inorganic semiconductor physics may provide an adequate description of charge transport and device performance in polyacene materials.

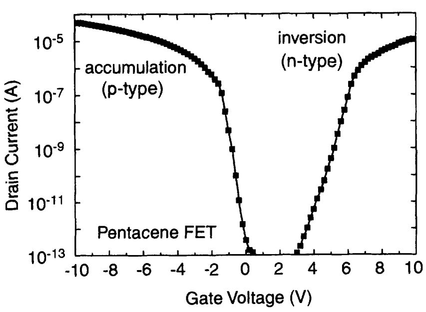

Fig. 3. Drain current versus gate voltage characteristics at room temperature of a pentacene single-crystal transistor. The on/off current ratio ($|V_{DS}|$ = 10 V) is 10^8 and 10^9 for n- and p-channel operation, respectively.

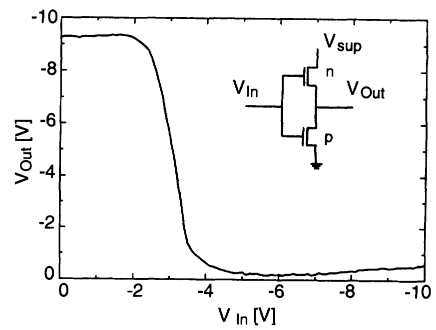

Fig. 4. Transfer characteristic of a CMOS-like ambipolar pentacene inverter circuit (see inset) for a supply voltage V_{sup} of −10 V. The p-channel transistor was used as driver for the n-channel load. A gain of 10 was obtained. V_{In}, input voltage; V_{Out}, output voltage.

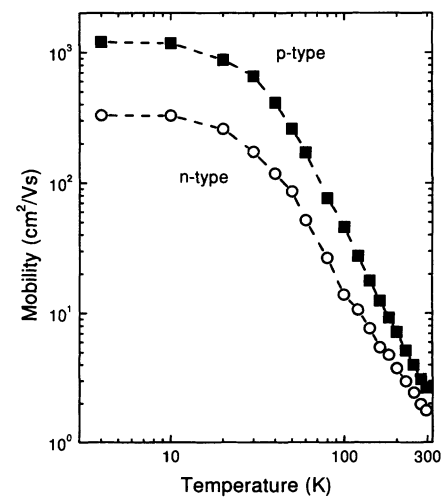

Fig. 5. Temperature dependence of the field-effect mobility of a pentacene single-crystal FET showing a power-law dependence for p-channel as well as for n-channel operation.

References and Notes

1. F. Garnier, G. Horowitz, D. Fichou, A. Yassar, *Synth. Met.* **81**, 163 (1996).
2. C. D. Dimitrakopoulos, B. K. Furman, T. Graham, S. Hedge, S. Purushothaman, *Synth. Met.* **92**, 47 (1998).
3. G. Horowitz, *Adv. Mater.* **10**, 365 (1998).
4. H. E. Katz, Z. Bao, A. Dodabalapur, in *Handbook of Oligo- and Polythiophenes*, D. Fichou, Ed. (Wiley-VCH, Weinheim, Germany, 1998), pp. 459–489.
5. S. F. Nelson, Y.-Y. Lin, D. J. Gundlach, T. N. Jackson, *Appl. Phys. Lett.* **72**, 1854 (1998).
6. Y. Y. Lin, D. J. Gundlach, S. F. Nelson, T. N. Jackson, *IEEE Trans. Electron Devices* **44**, 1325 (1997).
7. C. D. Dimitrakopoulos, S. Purushothaman, J. Kymissis, A. Callegari, J. M. Shaw, *Science* **283**, 822 (1999).
8. D. de Leeuw, *Phys. World* (March 1999), p. 31.
9. H. Klauk, D. J. Gundlach, J. A. Nichols, T. N. Jackson, *IEEE Trans. Electron Devices* **46**, 1258 (1999).
10. H. Sirringhaus, N. Tessler, R. H. Friend, *Science* **280**, 1741 (1998).
11. A. Dodabalapur *et al.*, *Appl. Phys. Lett.* **73**, 142 (1998).
12. A. Dodabalapur, H. E. Katz, L. Torsi, R. C. Haddon, *Science* **269**, 1560 (1995).
13. ______, *Appl. Phys. Lett.* **68**, 1108 (1996).
14. A. R. Brown, A. Pomp, C. M. Hart, D. M. de Leeuw, *Science* **270**, 972 (1995).
15. H. Klauk, D. J. Gundlach, T. N. Jackson, *IEEE Electron Devices Lett.* **20**, 289 (1999).
16. C. J. Drury, C. M. J. Mutsaers, C. M. Hart, M. Matters, D. M. de Leeuw, *Appl. Phys. Lett.* **73**, 108 (1998).
17. Y. Y. Lin *et al.*, *Appl. Phys. Lett.* **74**, 2714 (1999).
18. Ch. Kloc, P. G. Simpkins, T. Siegrist, R. A. Laudise, *J. Cryst. Growth* **182**, 416 (1997).
19. J. H. Schön, Ch. Kloc, R. A. Laudise, B. Batlogg, *Phys. Rev. B* **58**, 12952 (1998).
20. S. M. Sze, *Physics of Semiconductor Devices* (Wiley, New York, 1981), p. 442.
21. N. Karl, *Organic Semiconductors*, vol. 17 of *Landolt-Börnstein, New Series, Semiconductors*, O. Madelung, M. Schulz, H. Weiss, Eds. (Springer-Verlag, Berlin 1985), subvolume 17i, pp. 106–218.
22. G. W. Neudeck, H. F. Bare, K. Y. Chung, *IEEE Trans. Electron Devices* **34**, 344 (1987).
23. M. Matsumura and Y. Nara, *J. Appl. Phys.* **51**, 6443 (1980).
24. J. H. Schön and B. Batlogg, *Appl. Phys. Lett.* **74**, 260 (1999).
25. W. Warta and N. Karl, *Phys. Rev. B* **32**, 1172 (1985).
26. One of the authors (J.H.S.) gratefully acknowledges financial support by the Deutsche Forschungsgemeinschaft. Furthermore, we thank E. Bucher for use of equipment and Z. Bao, A. Dodabalapur, H. Katz, and G. A. Thomas for helpful discussions.

27 September 1999; accepted 23 December 1999