



Fig. 4. Disappearing images of more vortices in a single chain ($T = 57$ K, $B_{\perp} = 0.8$ mT, $\theta = 80^{\circ}$). We attribute the disappearance of the vortex images to the synchronous oscillation of vortices between the node vortices a and b along the chain direction, just like a coupled oscillation.

where a much larger number of chain vortices disappeared (Fig. 4).

If thermal fluctuations excite the oscillations of chain vortices, the temperature at which the oscillation begins and the vortex image disappears (T_d) must differ depending on the vortex density, because the vortex density changes the spring constant of the chain vortices and also the periodic potential distribution for chain vortices. When we measured T_d as a function of vortex density or B_{\perp} , we found that the T_d values greatly depended on B_{\perp} (Fig. 5). For example, T_d was as low as 20 K when $B_{\perp} = 1.7$ mT, whereas T_d was as high as 70 K when $B_{\perp} = 0.4$ mT.

The tendency for T_d to decrease when B_{\perp} increases can be qualitatively understood as follows: When B_{\perp} increases and the spacing a between vortices becomes narrower, the amplitude of the potential oscillation shown in Fig. 3C decreases as a result of the overlap of each vortex magnetic field. Lowering the potential barrier will naturally make chain vortices start oscillating at lower T . The spring constant k between closer chain vortices increases, thus making it easier for a more rigid incommensurate chain of vortices to slide freely as a whole. This explains why our experimental results show that T_d tends to decrease as B_{\perp} increases (Fig. 5).

We expected to be able to detect the direct indication of the vortex movement at the very beginning of the oscillation under the assumption that vortex lines move as straight rods; hence, we attempted to observe the vortex motion while T gradually increased and crossed T_d . However, the individual vortex images were gradually blurred, and no sign of vortex movement was detected. As a result, our experiments cannot rule out the possibility that a chain vortex line was split into pancake vortices (4) that oscillated in each layer independently for different layers, thus blurring the averaged magnetic field of the vortex line. Even in this case, the partial disappearance of vortex images can be explained because the oscillation of pancake chain vortices in each layer can have the nodes fixed by the surrounding straight vortex lines belonging to the triangular lattices.

The disappearance of the vortex images

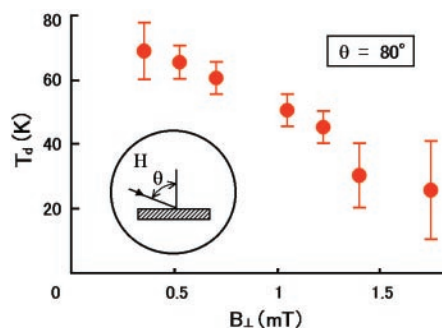


Fig. 5. Disappearance temperature T_d versus magnetic flux density perpendicular to the film B_{\perp} . T_d decreased greatly as B_{\perp} increased. This tendency can be explained by assuming the movement of chain vortices as being that of a coupled oscillation of chain vortices situated on the periodic potential distribution (see Fig. 3C). When vortices are dense (large B_{\perp} and small a), the periodic potential barrier becomes lower as a result of the overlap of the magnetic fields of surrounding triangular vortices, and chain vortices can more easily start oscillating at lower T , thus leading to lower T_d .

may be explained by assuming that the vortex lines are greatly tilted (7). Indeed, the image contrast was shown to decrease when vortex lines were trapped along columnar defects tilted by 70° (13). However, in the present experiment we were able to prove that the observed vortex image disappearance was not attributable to the vortex-line tilting, because the vortex images remained invisible even at a large defocusing distance Δf . The image contrast of tilted vortex lines, which is weak at a relatively short Δf , must become as vis-

ible as that of nontilted vortex lines at greater Δf s (13). Our experiments cannot rule out the possibility that the vortex lines do not simply tilt, but that their magnetic fluxes are somehow blurred in the chain direction (14).

The above discussion may be too simplified and may overlook several features of this phenomenon. For example, the adjacent lines of vortices on both sides of the disappearing vortex chains in Figs. 3A and 4 are not straight; they are slightly bent in such a way that the regions of the disappeared chains are wider at the vortex oscillation and narrower at the oscillation nodes. This may be due to the effect of the locally violent oscillation of vortices.

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14. More detailed simulations and experiments to pinpoint the cause of the disappearance of chain-vortex images are now in progress (A. Tonomura *et al.*, in preparation).
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Field-Effect Modulation of the Conductance of Single Molecules

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Field-effect transistors based on two-component self-assembled monolayers of conjugated and insulating molecules were prepared; the conductance through them can be varied by more than three orders of magnitude by changing the applied gate bias. With very small ratios of conjugated to insulating molecules in the two-component monolayer, devices with only a few "electrically active" molecules can be achieved. At low temperatures, the peak channel conductance is quantized in units of $2e^2/h$ (where e is the electron charge and h is Planck's constant). This behavior is indicative of transistor action in single molecules. On the basis of such single-molecule transistors, inverter circuits with gain are demonstrated.

Modern microelectronics and computation are advancing at an extremely fast rate. At the heart of this field has been the technology of complementary logic metal-oxide semiconductor (CMOS)-based integrated circuits. However, fundamental physical

limitations as well as increasingly prohibitive cost associated with fabrication facilities might slow down further miniaturization of devices to increase computation power within the next couple of decades (1, 2). To continue the trend to ever faster and

cheaper computers, new schemes will be required. One such approach is toward molecular devices (3, 4), where the idea is that only a few or even just one molecule could be used to perform basic electronic functions (5). Previous work has tended to focus on two-terminal devices: for example, negative differential resistance (6), rectifiers (7), and reconfigurable switching (8). Three-terminal devices, i.e., transistors, could offer several advantages for logic operations compared with two-terminal switches, the most important of which is "gain." As one example of that, an electro-mechanical amplifier that uses a single C_{60} molecule has been reported (9). In addition, field-effect devices based on single nanotubes have been demonstrated (10–14). However, for these devices, the active channel length was defined by the resolution of lithography rather than the molecular scale (15). Here, we report on field-effect transistor (FET) action, i.e., conductance modulation through a third electrode, in devices consisting of only several "electrically active" molecules. The channel length of 10 to 20 Å is defined by the thickness of a self-assembled monolayer (SAM) rather than lithography. Moreover, the peak conductance at low temperatures is quantized in units of $2e^2/h$, indicating that the conductance of single molecules is modulated.

We prepared vertical self-assembled monolayer field-effect transistors (SAMFETs) (15)

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using monolayers of alkanedithiols (e.g., 1,5-pentanedithiol) mixed with 4,4'-biphenyldithiol or 5,5'-terthiophenedithiol (Fig. 1A). The alkanedithiol was chosen to have a length similar to that of the conjugated molecule. A highly doped Si substrate, which is structured with conventional lithography and anisotropic etching to define vertical steps, was used as the gate electrode. A thin layer of thermally grown SiO_2 was used as a gate insulator. To fabricate a source electrode, we deposited a gold layer by thermal evaporation. A thiol-terminated monolayer was then self-assembled on the gold surface. Ellipsometry was used to confirm that only a monolayer has been deposited. Finally, the drain electrode was prepared by shallow angle shadow evaporation of gold onto a cooled substrate (~ 100 K) to avoid damage to the SAM and form an active contact area of about $0.8 \times 0.1 \mu m^2$. As the gated conduction in organic FETs takes place only within 50 Å from the semiconductor-insulator interface (16), only several thousand molecules will be probed in such SAMFETs, where the channel length is defined by the thickness of the monolayer. No signs of macroscopic phase separation were observed experimentally in similar two-component SAMs with data reported in the literature (17–19). We therefore assume that the conjugated molecules, i.e., 4,4'-biphenyldithiol or 5,5'-terthiophenedithiol, are homogeneously and statistically distributed within the alkanedithiol matrix. However, the possibility of bundles of conjugated molecules within the SAM cannot be ruled out completely. Nevertheless, the use of two-component SAMs allows us to study transistor action in single molecules or small bundles of molecules.

Control experiments reveal that samples consisting of pure alkanedithiol SAMs do not show any conductance modulation because of gate bias variation. All the data shown here are results from 4,4'-biphenyldithiol, even though similar characteristics have been observed for 5,5'-terthiophenedithiol.

The transfer characteristics of the SAMFETs scale with the ratio of conjugated to insulating molecules in the two-component SAM (Fig. 1B), suggesting that the properties of individual molecules are responsible for the conductance modulation rather than interactions of molecules with their nearest neighbors. However, it is difficult to explain the strong conductance modulation solely on the basis of quantum-mechanical treatment of simple coherent transport through a monolayer as first-principle calculations indicate a modulation of more than one order of magnitude for p-phenylenedithiol (20). However, it is particularly noteworthy that resonances and strong modulation have been observed and studied for single-molecule junctions (21). Additional effects could include a gate modulation of the potential distribution along the channel length, a suppression of the barrier at the contacts (22–24), or a conformational change of the molecule (25). At low temperatures (below 200 K), the transfer characteristics show a well-pronounced peak of the conductance as a function of gate voltage (Fig. 2A). Although quite broad, this peak might be related to resonant electron transfer. However, more detailed investigations will be needed to develop a better understanding of

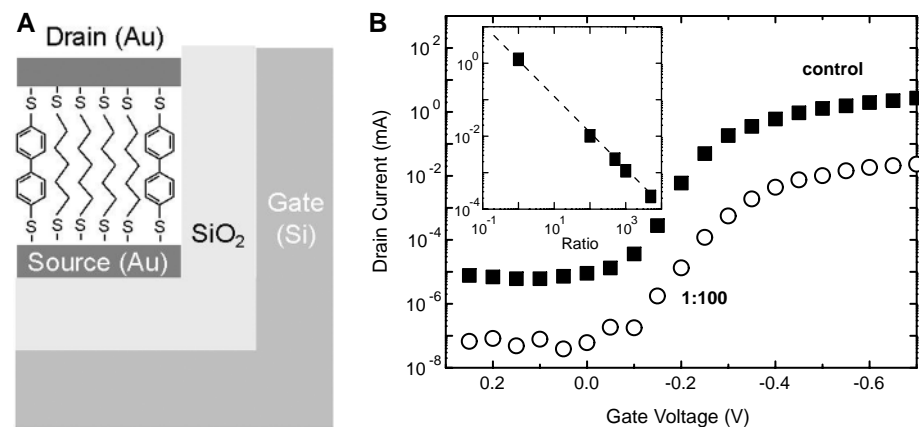


Fig. 1. (A) SAMFET structure. A highly doped Si substrate is used as the gate electrode, a thermally grown SiO_2 layer acts as gate insulator, the gold source electrode is deposited by thermal evaporation, and the active semiconducting material is a two-component SAM of alkanedithiols mixed with 4,4'-biphenyldithiol or 5,5'-terthiophenedithiol. The drain contact is defined by shallow angle shadow evaporation of gold. The active region of the device is magnified. (B) Transfer characteristics of two SAMFETs (drain-source voltage of -0.5 V). The control corresponds to a "pure" 4,4'-biphenyldithiol SAM, whereas the second one is based on a two-component SAM (4,4'-biphenyldithiol to alkanedithiol ratio is 1:100). The inset shows the current at a gate and drain-source voltage of -0.5 V as a function of the mixture ratio.

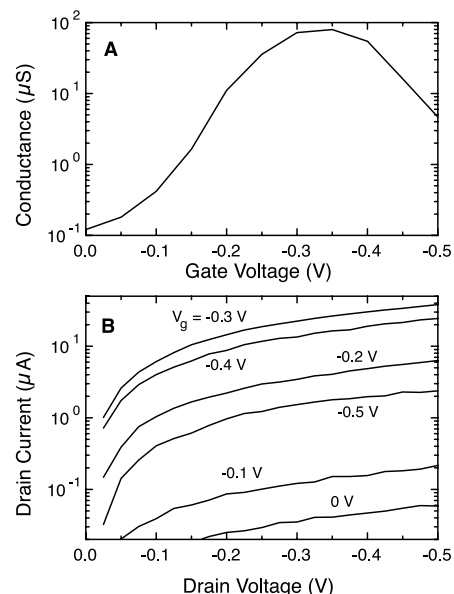


Fig. 2. (A) Transfer characteristics at 4 K of a two-component SAMFETs (ratio 1:5000). (B) Transistor characteristics at 4 K of a two-component SAMFETs (ratio 1:5000).

the microscopic processes underlying the transistor action (Fig. 2B) in these devices. For very "dilute" (ratio 1:5000) two-component SAMs, the peak conductance of the different investigated devices is quantized in units of $2e^2/h$ (Fig. 3A). Assuming a homogeneous distribution of the conjugated molecules in the SAM, we should find about two molecules in the active area of the device. However, in some devices, one or also more than two molecules might assemble in this region. This is in accordance with the results depicted in Fig. 3A, assuming that the maximum conductance through a single molecule is given by $2e^2/h$. We found that the value of the gate voltage leading to the conductance peak can differ

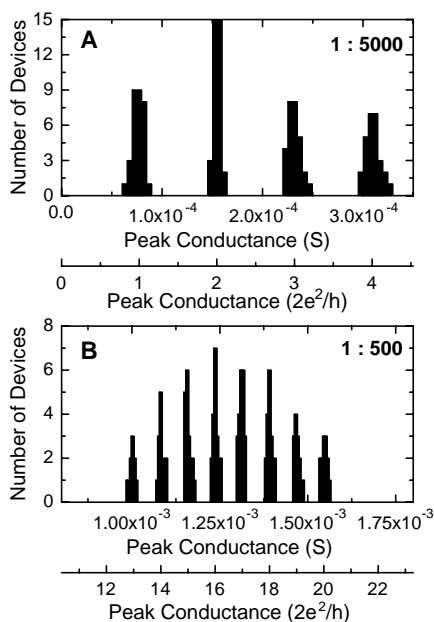


Fig. 3. Histogram of the peak conductance at 4 K of various two-component SAMFETs for a ratio 1:5000 (A) and 1:500 (B) revealing quantization in units of $2e^2/h$.

from device to device and is ascribed to different positions of the active molecule with respect to the gate insulator. Consequently, the effective field experienced by the molecule varies for different SAMFETs depending on its position relative to the gate insulator (vertical step). The peak conductance quantization is also observed for two-component SAMs with lower ratios of alkanedithiols compared with the conjugated species (Fig. 3B).

These results indicate that the conductance through a single molecule can be modulated by the field effect, leading to transistor action in a single molecule or small cluster. As the off-current of the SAMFET is limited by the leakage through the alkanedithiol matrix, the actual conductance modulation should be many orders of magnitude larger than 10^3 , which is observed for the devices reported here. From the transfer characteristics, a transconductance (differential change of the drain current with the gate voltage for a constant drain voltage) in the range of $150 \mu\text{A/V}$ is estimated for a single molecular SAMFET at low temperature (4 K), decreasing to about $10 \mu\text{A/V}$ at room temperature. One obvious way of increasing this value would be to decrease the gate insulator thickness. The control of placement of single molecules though more complexed self-assembly techniques (26) may lead to further improvements in device performance as well as simpler device design. Nevertheless, a small signal voltage gain, necessary for amplification, was observed for inverters consisting of two SAMFETs, which exhibited switching characteristics of one single molecule in the active channel. Figure 4 shows the static switching characteristics of such an inverter circuit, which is a basic building block for more complex logic circuits. Here, the output switches from logic "1" (-2 V) to logic "0" (0 V) when

the input switches from logic 0 to logic 1. A gain up to 10 is obtained (see inset in Fig. 4), demonstrating the possibility of switching and amplification based on single molecular devices.

Although many challenges remain, the demonstration of FET action with a single electroactive molecule can be regarded as an important step toward "molecular electronics." Moreover, the use of two-component SAMs will allow the study of gating effects on various molecules. With molecular heterostructures and molecular wires, it might be possible to combine the insulating and semiconducting properties as well as the contacts within a single molecule or supramolecular architecture.

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Fig. 4. Output characteristics at 4 K of an inverter with two two-component SAMFETs (ratio of 1:5000). The small signal gain is plotted in the inset revealing the possibility of amplification and switching of SAMFETs based on single molecular operation. V_s is the supply voltage of -2 V .

