$H \propto r$, as predicted by Eq. (4). The nonlinear behavior of I_{709} and H versus P can be understood: When the drop diameter reaches a significant fraction of the laser-spot size (defined by the focusing condition), the single drop can more efficiently collect the e-h pairs excited by the laser.

For the maximum possible drop radius in this sample, $r_{\rm max} = 0.68$ mm = half the sample thickness, we calculate from Eq. (5) the resonant field H = 36 kOe, comparable to the field for the *C* line at maximum pump power [Fig. 3(b)]. A thinner sample yielded a correspondingly lower maximum resonant field. At the very highest laser pump powers the relative spacing of the lines appears to change, possibly indicating a change in the shape of the drop as its size approaches the sample dimensions. Although earlier experiments¹ reported drop sizes of ~ 10 μ m, recent experiments here¹² report large drops.

Finally, we have used MDR to study decay kinetics of large drops, to be reported separately.¹³

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Negative Magnetoresistance in *n*-Channel (100) Silicon Inversion Layers

I. Eisele and G. Dorda Forschungslaboratorien, Siemens AG, München, West Germany (Received 8 April 1974)

The transverse magnetoresistance effect of (100) *n*-channel silicon inversion layers was measured at low temperatures. Negative and/or positive effects have been obtained depending on surface-state density.

Transverse magnetoresistance measurements have been carried out on *n*-channel metal-oxidesemiconductor field-effect transistors in a temperature range between 2 and 10 K. The oxide thickness was 120 nm and the geometry of the channel (channel length 400 μ m, channel width 40 μ m) allowed additional information about the Hall mobility. Transistors on a (100) silicon surface were selected so that a wide range in threshold voltage, as well as the corresponding maximum mobility, was represented.

The charge-carrier concentration responsible

for the transport in metal-oxide-semiconductor transistors is caused by a sufficiently strong electric field perpendicular to the surface and is confined to the semiconductor region near the Si-SiO₂ interface. As a consequence of the electrical surface field, the energy of the charge carriers perpendicular to the surface is quantized and the carriers behave almost as a two-dimensional electron gas.¹ At sufficiently low temperatures the electrons in an *n*-channel (100) surface are restricted to the lowest sub-band corresponding to the isotropic effective mass parallel



FIG. 1. Transverse magnetoresistance effect $\Delta \rho / \rho_0$ versus magnetic field *H* at 4.2 K. Parameter is the threshold voltage V_T . The surface-state density increases with decreasing V_T .

to the surface $(m_T = 0.21m_0)$. From statistics we know further that the Fermi level in the temperature range considered lies within the conduction sub-band. Considering both of these facts we are led to the assumption that the transverse magnetoresistance should be zero if we apply the commonly used theories. Despite the expected behavior at low temperatures we observed pronounced magnetoresistance effects at 4.2 K (Fig. 1) that depend on the magnetic field and, surprisingly, change sign gradually with sample quality. The behavior is clearly positive for high-quality samples, i.e., low concentrations of interface states, as is documented by a large threshold voltage (V_T = 3.3 V) and a high effective-mobility maximum (μ_{max} =1.75 m² V⁻¹ sec⁻¹). For lower quality samples the relative resistance change at low gate voltages becomes more and more negative as can be seen from the other profiles with $V_T = 1.9 \text{ V} (\mu_{\text{max}} = 0.65 \text{ m}^2 \text{ V}^{-1} \text{ sec}^{-1}) \text{ and } V_T = 0.4 \text{ V} (\mu_{\text{max}} = 0.38 \text{ m}^2 \text{ V}^{-1} \text{ sec}^{-1}).$ In Fig. 2 the transverse magnetoresistance is plotted versus V_{c} $-V_T$, where V_G is the applied gate voltage normal to the surface and V_T the threshold voltage at which the channel current flow begins. V_{c} $-V_T$ is proportional to the induced carrier density in the channel. For high-quality samples the effect is positive in the measured range and rises to a maximum value of about 5% at $V_G - V_T$ = 4.2 V. For low-quality samples the negative effect is only observed at low electrical surface



FIG. 2. Magnetoresistance $\Delta \rho / \rho_0$ versus effective gate voltage $V_G - V_T$, where V_G is the applied gate voltage and V_T the threshold voltage; the oxide thickness was 120 nm. Parameter is the surface-state density that corresponds to V_T .

fields and decreases rapidly with increasing $V_G - V_T$, becoming positive for $V_G - V_T \ge 10.6$ V.

Similar positive and negative profiles have been observed for highly doped semiconductors.²⁻⁶ The positive effect was described on the basis of a reducing overlap of neighboring impurity states whereas, for the explanation of the negative behavior, a model was proposed in which the scattering produced by localized spins is reduced by the magnetic field. It was demonstrated that the negative effect increased with increasing doping (impurity) concentration.

The obvious connection between the magnetoresistance effect and the concentration of the interface states seems also to be the key for the analysis of the present results. At low temperatures the existence of a hopping process between localized states was recently proposed for low chargecarrier concentrations in the inversion channel.⁷ This idea seems to be valid because at these concentrations the mobility is much smaller than μ_{max} . The application of the spin scattering theory to the inversion layer also seems to be appropriate for our results because the interfacestate densities N_{ss} for low-quality samples (N_{ss} $\geq 10^{16}$ m⁻²) are comparable to the bulk impurity concentrations in doped semiconductors where the negative magnetoresistance effect was found. For doped semiconductors Boon⁸ derived equations describing a negative magnetoresistance by the interaction between localized spin centers and conduction electrons. We modified this theory by introducing Fermi statistics instead of

Boltzmann statistics. In this case the electron concentrations with spin up (n_{\pm}) and spin down (n_{\pm}) in the quantum limit are given by

$$n_{\dagger}(H) = \frac{m_T}{2\pi\hbar^2} \left(\boldsymbol{E}_{\rm F} - \boldsymbol{E}_{\rm 0} + \frac{\Delta \boldsymbol{E}(H)}{2} \right), \tag{1}$$

$$n_{\downarrow}(H) = \frac{m_T}{2\pi\hbar^2} \left(E_{\rm F} - E_0 - \frac{\Delta E(H)}{2} \right), \tag{2}$$

where m_T is the transverse effective mass of electrons, E_F the energy of the Fermi level, E_0 the minimum energy of the lowest sub-band, and $\Delta E(H)$ the energy difference between spin-up and spin-down carriers. This holds only for $E_F - E_0$ $\gg kT$. Introducing these equations we obtain the final expression for the conductivity change $\Delta \sigma / \sigma_0$:

$$\frac{\Delta\sigma}{\sigma_0} = \frac{\hbar^2}{16m_T a^2 (E_F - E_0)} \left(\frac{g\mu_B H}{kT}\right)^2,$$
(3)

provided that $g \mu_{\rm B} H \ll kT$, where *a* is the spacing between localized spins, g the Landé factor, $\mu_{\rm B}$ the Bohr magneton, and H the magnetic field. For magnetic fields such that $g\mu_{\rm B}H \gg kT$, the conductivity change saturates at $\Delta\sigma/\sigma_0 = \lambda_0/2a$ with a mean free path λ_0 due to all scattering processes excluding the spin scattering. According to Eq. (3) the negative magnetoresistance should be proportional to H^2 and T^{-2} , and therefore we studied the temperature dependence of sample c of Fig. 1. The results are presented in Fig. 3 for various temperatures. It should be noted that with increasing temperature the negative effect rapidly decreases, thus revealing more and more the positive effect. Its existence is based on the fact that the magnetic field reduces the mobility of the hopping probability by reducing orbital overlap of surface centers. Discussing the influence of the magnetic field on the bound wave function yields an effect proportional to H^2 and independent of temperature.⁶ The temperature dependence of sample a of Fig. 1 showed indeed that the positive magnetoresistance effect is almost independent of temperature up to 18 K.

The data of Fig. 3 have been fitted by combining both effects according to

$$\frac{\Delta\sigma}{\sigma_0}(T,H) = \alpha \frac{H^2}{T^2} - (\beta H)^n.$$
(4)

The solid lines in Fig. 3 represent this semiempirical equation with $\alpha = 2.0$, $\beta = 0.016$, and, in agreement with sample *a*, $1 \le n(H) \le 2$. Equation (4) satisfactorily describes the experimental results for low magnetic fields. At higher fields a



FIG. 3. Temperature dependence of the magnetoresistance effect of sample c of Fig. 1. The solid lines correspond to the theoretical profiles.

deviation occurs that is caused either by a beginning saturation of the spin reorientation or by a positive effect with n(H) > 2. The value of α yields a/g = 1.4 nm. Several authors found that with decreasing electron concentration the g factor in the inversion layer exceeds by far the usual value of 2 for free electrons.⁹⁻¹¹ The theory in the case of a half-filled Landau level with all spins in the same direction accounts for a maximum of g = 8. At low magnetic fields more than one Landau level is occupied and a smoothing of the g-factor oscillation occurs. In agreement with the literature we thus suppose that g = 4seems to be a reasonable value for our low electron concentrations. For the distance between the localized surface centers we then calculate a = 5.6 nm, corresponding to a surface density $N_{ss} = 3.2 \times 10^{16}$ m⁻². In a different way the surface-state density can be estimated by measuring the mobility ratio μ_{Hall}/μ_{eff} which in our case yielded $N_{ss} = 2.1 \times 10^{16} \text{ m}^{-2.12}$ This excellent agreement is an additional support of the presented model.

In conclusion, we would like to point out that this is the first observation of a negative magnetoresistance in connection with surface states. This transverse effect at the surface differs substantially from the negative tangential magnetoresistance effect. If the phenomenological deVOLUME 32, NUMBER 24

scription with spin scattering is modified taking a two-dimensional sub-band structure into consideration, the theory appears to be acceptable and quite successful in describing the experimental results.

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Magnetic Studies of Tetrathiofulvalinium Tetracyanoquinodimethanide (TTF-TCNQ)

Y. Tomkiewicz, B. A. Scott, L. J. Tao, and R. S. Title IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598 (Received 28 January 1974)

EPR and magnetic-susceptibility studies of TTF-TCNQ indicate that in the temperature region $T \gtrsim 60^{\circ}$ K this compound consists of a metallic (TTF)⁺ chain and a (TCNQ)⁻ chain with localized spins. The metal-insulator transition at ~ 60°K is accompanied by a strong interchain interaction.

The magnetic properties of a large number of TCNQ salts have previously been studied.¹ Most of the low- and intermediate-conductivity TCNQ complexes [conductivities below 10 $(\Omega \text{ cm})^{-1}$] exhibit a temperature-activated susceptibility, whereas the susceptibility of the highly conductive TCNQ complexes shows a very weak temperature dependence. In TTF-TCNQ, however, despite the unusually high conductivity^{2,3} the magnetic susceptibility behavior² resembles that of low-conductivity salts.

In order to understand this unusual behavior, we have made an extensive EPR investigation of TTF-TCNQ single crystals as well as static magnetic-susceptibility studies of polycrystalline samples. The EPR measurements were performed at X and K bands. (In the following, whenever the frequency is not specified the related experiment was performed at X band.) Two different types of single crystals were used: Solutiongrown⁴ crystals, in which the b axis is the needle axis [conductivity along the b axis ~ (3.3-10) $\times 10^2/\Omega$ cm],⁵ were used for the $\vec{H}_1 \parallel \vec{b}$ orientation, while vapor-grown⁶ crystals, in which the b axis is almost perpendicular to the needle axis [conductivity along the *b* axis ~ $(2-6.7) \times 10^2/\Omega$ cm],⁵ were used for the $\vec{H}_1 \perp \vec{b}$ orientation. Crystals of highest conductivity exhibited Dyson-Bloembergen line shapes at the $\vec{H}_1 \perp \vec{b}$ orientation. No significant variations of line widths or *g* values, for a common orientation, were found between samples grown by these two different methods. The static susceptibility of polycrystalline TTF-TCNQ crystals was measured by the Faraday method in a helium gas atmosphere in the temperature range 2-400°K.

In solutions the reported g values of $(TTF)^+$ and $(TCNQ)^-$ are, respectively, 2.008 38⁷ and 2.0025.⁸ In view of this large difference between the g values one might expect to observe two distinct absorptions in the solid TTF-TCNQ (actually four because there are two molecules per unit cell⁹). However, over the entire temperature range 20-300°K,¹⁰ in which the susceptibility was found to be temperature activated, only a single line was observed. The principal values of g at room temperature are $g_{zz} = 2.0026 \pm 0.0003$, g_{xx} = 2.0065 ± 0.0003, and $g_{yy} = 2.0086 \pm 0.0003$. The z