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Molecular Crystals and Liquid Crystals Incorporating Nonlinear Optics

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Electrical Switching Effect in Organic Charge Transfer Compounds

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ELECTRICAL SWITCHING EFFECT IN ORGANIC CHARGE TRANSFER COMPOUNDS

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Abstract A current switching effect in charge transfer compounds has been investigated by probing photoconductivity. It has been found that the life time of photocurrent is suddenly elongated when the crystal is switched to the on-state of current.

INTRODUCTION

Since Potember et al. discovered an electrical switching effect in Cu-TCNQ films[1], the current switching phenomenon in organic materials has attracted much attention in connection with interest in molecular electronics. Recently, we have found a switching effect in various types of organic charge transfer (CT) compounds which have one-dimensional columns of alternately stacked donor(D) and acceptor(A) molecules(...DADADA...).[2,3]

Experiments on single crystals have indicated that the J-E characteristics observed in bulk crystals exhibit a negative resistance region as shown in Fig.1. When a load resistor is connected in series with the crystal, a switching effect takes place with a large hysteresis (see the inset of Fig.1). We have also demonstrated that a pronounced negative resistance effect is observed only in the low temperature phase of the ionic CT crystals[3]. In the ionic crystal $TTeC_1TTF$-TCNQ, whose J-E curves are shown in Fig.1, a phase transition takes place at about T_c=240K. Above T_c, the lattice is quasi-uniform, and below T_c, the stack is dimerized as schematically represented by ...$\underline{D^+A^-}$ $\underline{D^+A^-}$ $\underline{D^+A^-}$ $\underline{D^+A^-}$... This phase transition is a kind of spin-Peierls transition, and we refer the low temperature state as the bond ordered wave (BOW) state. As shown in Fig.1, the negative resistance effect is enhanced in the BOW phase.

The negative resistance effect has been observed in various BOW

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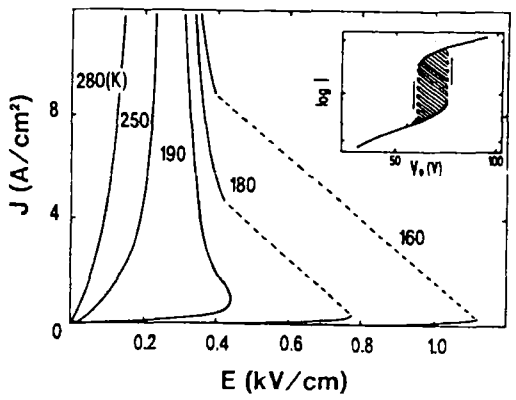

FIGURE 1
J-E characteristics for $TTeC_1TTF$-TCNQ single crystal at various temperatures. The inset shows the relation between the current and the source voltage.

semiconductors, but the underlying mechanism remains still unsolved. We suggest that the switching of Cu-TCNQ films is also likely related to a similar mechanism. This mechanism is thought to be different from those proposed for the switching phenomena in inorganic semiconductors or chalcogenide glass films[4], such as a Zener tunneling from the valence to the conduction band, an avalanche breakdown, or a heating effect.

The experimental results suggest that the negative resistance effect is inherent to the BOW state. To tackle this problem, it is crucial to get information on the nature of high current (negative resistance) state. In this work, we have measured the photoconductivity to probe the change of lattice dimerization before and after switching. It is shown that the high current state has a nature similar to the high temperature phase. Based on these results, we suggest that, in the high current state, the three-dimensional order of dimerization is appreciably disturbed.

EXPERIMENTAL RESULTS

The electrical circuit in the measurement of photocurrent was identical to that used in the measurement of dc I-V characteristics.[3] The light source was an excimer-dye laser tuned at wavelength 581nm, with the pulse width of 15ns, the repetition rate of 0.5Hz. The laser power was made sufficiently weak to avoid the heating effect by laser irradiation. The decay curves of photocurrent were recorded by a digital oscilloscope.

The photocurrent decays at a low electric field (E=150V/cm) are shown in Fig.2(a). The current I(t) is normalized by the initial

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photocurrent I(t=0). The decay at high temperature quasi-uniform phase is very slow. As the temperature is lowered, the decay time becomes shorter, and in the low temperature dimerized phase, it is appreciably shortened. The decay curves follow a power dependence on time, rather than an exponential form. Although the microscopic mechanism for the photoconductivity is not well understood as yet, the characteristic decay rate of photocurrent seems to reveal some important aspect of the degree of lattice dimerization.

In Fig.2(b), we plotted the decay curves of photocurrent at 160K in the low and high current states. The decay curves are essentially independent of field so long as the sample remains in the low current state. However, after the crystal is switched to the high current state, the decay time of photocurrent is suddenly elongated. Note that such an elongation effect of photocurrent decay after switching (at 160K) is similar to the change of decay curve when the crystal undergoes a transition to the high temperature quasi-uniform phase.

The observed change of the photocurrent decay is not attributable to the heating effect. If the change of decay is totally due to the heating effect, the temperature in the high current state must be about 100K higher than the bath temperature of 160K, when estimated from the experimental temperature dependence of the photocurrent decay curve. A rough estimate of heating gives a temperature rise in sample less than 20K, since the power loss in the high current state is only about five times larger than that in the low current state.

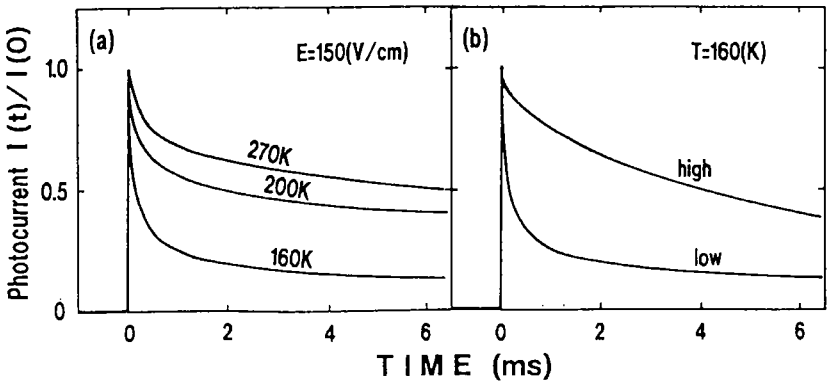

FIGURE 2
Decay characteristics of photocurrent (a) at different temperatures at low field(E=150V/cm), (b) for high and low current state at 160K (BOW phase).

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DISCUSSION

In the BOW organic semiconductors including dimerized CT crystals and conjugated polymers, the current is considered to be supported by charge carriers associated with low-energy excitations due to the strong electron-lattice interaction. In trans-polyacetylene, the existence of charge solitons or polarons has been theoretically predicated and experimentally confirmed[5]. In CT compounds, the carriers are also thought to have a nature similar to those in conjugated polymers.

In the dimerized phase (BOW state), a defect of dimerization is supposed to contribute to the conductivity. It is expected, then, that if the density of such carriers is increased beyond a certain limit, the BOW state may be significantly disturbed, leading to a kind of transition to a non-BOW state. The experimental results in Fig.2 clearly indicate that the photoconductive response is very similar between the high temperature phase and the high current state in BOW. We speculate that the negative resistance state corresponds to a state where the BOW state is effectively broken by an excessive current flow. The negative resistance state may be regarded as a current-induced quasi-uniform state analogous to that in the high temperature phase.

The negative resistance effect in these crystals has been empirically explained assuming a self-increase of current.[3] Such a cooperative nature of nonlinear conductivity can be attributed to the effect of three-dimensional interaction between the one-dimensional DA stacks. A definite model based on this picture will be presented elsewhere together with more quantitative analysis of experimental results.

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