Materials Chemistry and Physics 148 (2014) 431–434

Contents lists available at ScienceDirect

Materials Chemistry and Physics

journal homepage: www.elsevier.com/locate/matchemphys

# Effects of a metallic front gate on the temperature-dependent electronic property of pentacene films

Yow-Jon Lin [a, *], Hou-Yen Tsao [a], Day-Shan Liu [b]

[a] Institute of Photonics, National Changhua University of Education, Changhua 500, Taiwan
[b] Graduate Institute of Electro-Optical and Materials Science, National Formosa University, Huwei 632, Taiwan

## HIGHLIGHTS

- For the metal-capped and uncapped pentacene films, the mobility was researched.
- The mobility was dramatically increased by capping an In (Au) layer.
- The induced strain by capping a metal layer is found.
- The strain may lead to the electron–phonon coupling variation.
- The enhanced mobility is attributed to the weakened electron–phonon coupling.

## ARTICLE INFO

*Article history:*
Received 17 September 2013
Received in revised form
13 April 2014
Accepted 7 August 2014
Available online 29 August 2014

*Keywords:*
Organic compounds
Thin films
Electrical properties
Transport properties
Hall effect

## ABSTRACT

The effect of a metallic front gate on the temperature-dependent electronic property of pentacene films was investigated in this study. The carrier mobility exhibits strong temperature dependence, implying the dominance of tunneling (hopping) at low (high) temperatures. The room-temperature mobility was drastically increased by capping an In (Au) layer on the pentacene front surface. However, the carrier concentration is not affected. An increase in the phonon energy occurs for In-capped or Au-capped pentacene samples, which corresponds to the abrupt transition to the nonlocal electron–phonon coupling. The enhanced mobility by capping a metal layer is attributed to a change in the electron–phonon coupling.



## 1. Introduction

Organic semiconductors play an increasing role in many fields. Nowadays, organic thin film transistors (OTFTs) have been intensively investigated in order to apply their remarkable attributes to more electronic devices, such as organic light-emitting diode panels, displays and flexible logic circuits [1]. Among the organic semiconductors considered for use as active materials in OTFTs, pentacene is the most widely used and studied because of its outstanding mobility. Charge transport in organic semiconductors has been extensively studied [2–10]. These new experimental results have renewed the interest in developing theoretical models to better understand the charge-transport mechanisms in organic molecular crystals [2]. The key quantity that characterizes charge transport is the carrier mobility. The temperature dependence of carrier mobilities exhibits a universal power-law behavior at low temperatures and a temperature independent or slightly thermally activated behavior at high temperatures [11]. The crossover from bandlike to hopping transport in the carrier mobilities occurs around room temperature [8,11–14]. Here, we investigated the effect of a metallic front gate on the Hall-effect carrier mobility ($\mu$) of pentacene films. The room-temperature mobility was drastically increased from 11.5 to 24.1 (29.6) $cm^2\ V^{-1}\text{-}s^{-1}$ by capping an In (Au) layer on the pentacene front surface. Our results indicate that the enhanced mobility is due to a change in the electron–phonon coupling. The raised $\mu$ by metallic capping layers has never been reported until now.

## 2. Experiment procedure

A 70 nm thick pentacene (Luminescence Technology Corp.) layer was deposited on the glass substrates by vacuum thermal evaporation and the evaporation rate was 1.8 nm $min^{-1}$ (as monitored by a

* Corresponding author. Tel.: +886 4 7232105x3379; fax: +886 4 7211153.
*E-mail address:* rzr2390@yahoo.com.tw (Y.-J. Lin).

http://dx.doi.org/10.1016/j.matchemphys.2014.08.010
0254-0584/

quartz crystal microbalance after thickness calibration with atomic force microscopy measurements). The substrate temperature was fixed at 90 °C [8]. The pentacene films were deposited on glass substrates for the conductivity measurements. The glass samples were cleaned in chemical cleaning solutions of acetone and methanol, rinsed with de-ionized water, and blow-dried with $N_2$. For all samples, the carrier concentration and mobility were obtained from the Hall-effect measurements (Ecopia HMS-5000) based on van der Pauw method. Four thick gold top contacts were deposited by sputtering using a mechanical mask. The HMS-5000 includes software with current–voltage curve capability for checking the ohmic integrity of the user made sample contacts. In addition, Au and In were used as the capping layer located on the pentacene front surface, respectively. The capping layer was fabricated by depositing Au (In) metal on the pentacene layer through a shadow mask. The thickness of these capping layers is fixed as 60 nm. The inset of Fig. 1 shows a schematic plane view of the Au-capped (In-capped) sample. To confirm the influence of the capping layer on the device performance, the bottom-contact OTFTs were fabricated. A $SiO_2$ layer was grown on the n-type Si wafer with resistivity of 0.001 Ω cm using a dry oxidation process as a gate oxide layer. The thickness of the $SiO_2$ film is 265 nm according to ellipsometric measurements. The insulator capacitance per unit area ($C_i$) of 12 nF $cm^{-2}$ was obtained from capacitance–voltage measurement. The source/drain ($S/D$) electrodes were fabricated by depositing Au metal on the thermally grown $SiO_2$ layer through a shadow mask. The thickness of $S/D$ electrodes is 60 nm. Then, a 140-nm-thick pentacene layer was deposited on the thermally grown $SiO_2$/n-type Si substrates by vacuum thermal evaporation. The substrate temperature was fixed at 90 °C. The small-area metallic front gate was fabricated by depositing In (Au) metal on the pentacene layer through a shadow mask. The devices have the channel width ($W$) of 700 μm and the channel length ($L$) of 100 μm. The transfer characteristics of devices were measured using a Keithley Model-4200 semiconductor characterization system. The drain current ($I_D$) versus gate-source voltage ($V_{GS}$) curve was scanned from 10 to −40 V, with a fixed drain-source voltage ($V_{DS}$) at −40 V. In the saturation region, $V_{GS}$-dependent $I_D$ measurements at a constant $V_{DS}$ provide a method to examine the relationship between the threshold voltage ($V_{TH}$) and the field-effect carrier mobility ($\mu_{FE}$).

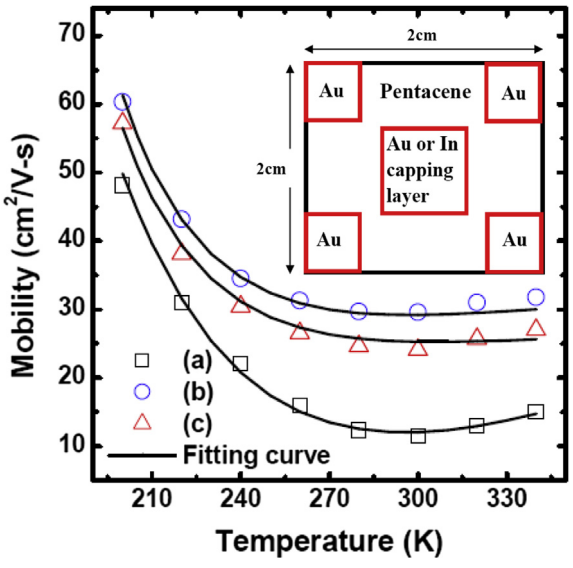


**Fig. 1.** The carrier mobility as a function of temperature [(a) uncapped, (b) Au-capped and (c) In-capped pentacene samples]. Inset: a schematic plane view of the Au-capped (In-capped) sample.

## 3. Experimental results and discussion

To obtain the carrier mobility as a function of temperature ($T$), the Van der Pauw–Hall measurements were performed in the temperature range of 200–340 K. The uncapped and capped pentacene samples show p-type behavior. Fig. 1 shows the carrier mobility as a function of $T$. It is found that the carrier mobilities are strongly temperature-dependent from 200 to 340 K for uncapped, Au-capped, or In-capped pentacene samples. The carrier mobility decreases with increasing $T$ reaching a minimum at $T = 300$ K, and then increases with further increasing of $T$ beyond that point. Early models suggested that thermally activated small polaron hopping describes the motion of carriers through an organic crystal [15]. Seo et al. suggested that charge transport in pentacene occurs by an activated-transport process in the temperature range of 40–300 K [16]. Our measurements showed a significant different from that reported, which cannot be explained with activation hopping. It cannot explain the sign reversal of the bulk mobility versus temperature derivative ($\partial\mu/\partial T < 0$).

Coropceanu et al. presented an overview of polaron models that have been reported to describe the charge transport mechanisms in organic materials [2]. An important feature of the small polaron theory provides the origin of the bandlike to hopping crossover: the electron–phonon interaction is seen to reduce the transfer integral (bandwidth). The band narrowing is temperature dependent with the polaron band narrower at higher temperatures. Thus, the polaron mass is larger at higher temperatures, thereby reducing the bandlike mobility. Since the hopping contribution increases with temperature, a crossover from one regime to the other is predicted. For strong local electron–phonon couplings ($g^2 >> 1$), two distinct temperature regimes occur; see Fig. 18 of Ref. [2]: (i) at low temperatures, the mobility is bandlike; (ii) as temperature increases, the hopping term starts to dominate, and the mobility exhibits a crossover from coherent transport to an incoherent, temperature-activated transport. According to polaron models, the total mobility can be expressed to a good approximation as a sum of two contributions [2]:

$$\mu = \mu_{tun} + \mu_{hop} \tag{1}$$

$$\mu_{tun} = \frac{ea^2\omega_o}{k_B T}\left[\frac{g^2}{\pi\sinh\left(\frac{\hbar\omega_o}{2k_B T}\right)}\right]^{0.5} \exp\left[\frac{-2g^2}{\sinh\left(\frac{\hbar\omega_o}{2k_B T}\right)}\right] \tag{2}$$

$$\mu_{hop} = \frac{ea^2t^2}{k_B T\hbar^2\omega_o}\left[\frac{\pi\sinh\left(\frac{\hbar\omega_o}{2k_B T}\right)}{g^2}\right]^{0.5} \exp\left[-2g^2\tanh\left(\frac{\hbar\omega_o}{4k_B T}\right)\right] \tag{3}$$

where $\mu_{tun}$ is the tunneling mobility, $\mu_{hop}$ is he hopping mobility, $e$ is the electron charge, $g$ is a coupling constant, ħ equals to $h/2\pi$ ($h$ is the Planck constant), $\hbar\omega_o$ is the phonon energy, $a$ denotes the spacing between molecules, $t$ is the transfer integral, and $k_B$ is Boltzmann's constant. Here, the first term of Eq. (1) is due to electron tunneling (coherent electron transfer) and dominates transport at low temperatures; the second term of Eq. (1) is related to hopping motion (incoherent electron transfer) and becomes dominant at higher temperatures. To extract the charge-transport parameters, the relationship between $\mu$ and $t$ ($\hbar\omega_o$, $a$ or $g$) was examined based on the equations [Eqs. (1)–(3)]. We found that the reduction of $\hbar\omega_o$ may lead to suppressed (enhanced) tunneling (hopping) conduction. Upon $t$, tunneling conduction is not affected. However, hopping conduction is sensitive to the $t$ variation. In