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THE EFFECT OF OXYGEN ON THE PHOTOCONDUCTIVITY OF C_{60} FILM: ACTION SPECTRUM AND TEMPERATURE DEPENDENCE

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Abstract We report the effect of oxygen on the action spectrum of the steady-state photoconductivity (PC) of C_{60} film and its temperature dependence. The steady-state PC decreases more than 4 orders of magnitude upon exposure to oxygen; the PC decreases faster below $\hbar\omega \approx 2.3$ eV. The low-temperature PC spectrum of the oxygen-free C_{60} is similar to the high-temperature PC spectrum of C_{60} exposed to oxygen. The steady-state PC exhibits a peak at $T \approx 240$ K in the oxygen-free C_{60}, which disappears in C_{60} exposed to oxygen. The results indicate that the oxygen in C_{60} film acts as an efficient deep trap for photoexcited carriers and reduces the structural order.

INTRODUCTION

Since the discovery[1] of C_{60} many experimental and theoretical studies have been carried out to understand its electronic and optical properties. However, many important issues have remained unsettled. One of these issues is the magnitude of band gap in pristine solid C_{60}.[2] Since measurement of the PC action spectrum can determine the energy gap for photogeneration of charge carriers, we address this issue through comprehensive measurements of the steady-state PC of C_{60}.

Various different PC action spectra have been reported by a number of groups.[3-8] Therefore, it is difficult to unambiguously determine the energy gap. Since oxygen has been known to affect the optical and electronic properties of C_{60},[8-11] we found it necessary to examine systematically its effect on the PC action spectrum. Furthermore, the PC spectrum has not been studied as a function of temperature. As is well known, solid C_{60} undergoes a first-order structural phase transition from a high-temperature face-centered-cubic to a low-temperature simple-cubic structure near $T_C \approx 249$ K.[12] Thus, it is also important to study the effect of the structural phase transition on the PC action spectrum.

In this paper, we focus on the effect of oxygen on the steady-state photoconductivity (action spectrum and its dependence on temperature and modulation frequency of light) of C_{60} film.

EXPERIMENTAL

The preparation of C_{60} film has been described in previous publications.[3,13] The steady-state PC measurements of C_{60} film in the surface-cell configuration were carried out using standard modulation technique in the temperature ranges between 50 - 300 K. The excitation light from a 150 W xenon lamp was modulated at a frequency of 39 Hz, and the PC response was detected by a lock-in amplifier. More details regarding the steady-state PC measurements can be found in early publications.[3,13]

RESULTS AND DISCUSSION

Figure 1 presents the evolution of the steady-state PC action spectra taken at 300 K upon exposure to oxygen. The results reveal a dramatic decrease of the steady-state PC upon exposure to oxygen, by more than 4 orders of magnitude from its value in pristine

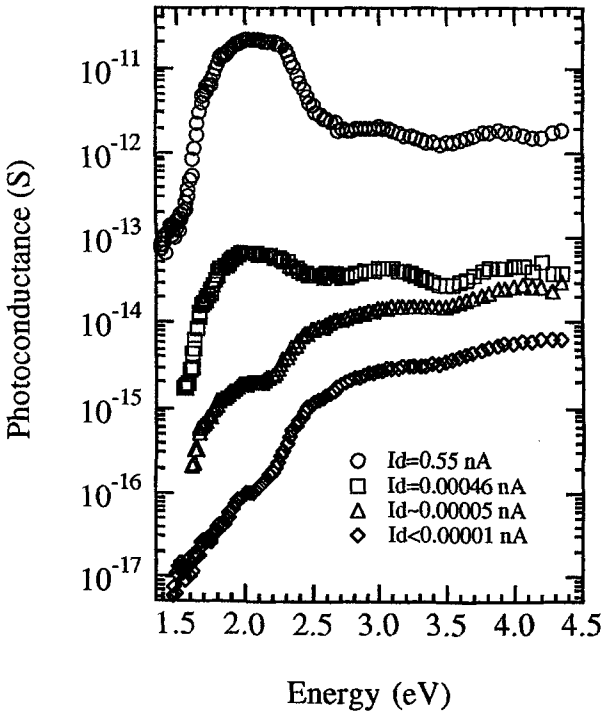

Figure 1 The room temperature steady-state PC action spectrum of pristine C_{60} film and its evolution with increasing oxygen content. The dark current (I_d) of each curve is indicated for each stage of oxygen exposure.

C_{60}. While exposure of C_{60} film to oxygen does not change significantly the optical absorption spectrum (it only slightly broadens the absorption peaks[11]), it modifies significantly the PC excitation spectrum.

The steady-state PC of the pristine C_{60} shows a sharp increase at $\hbar\omega \approx 1.6$ eV, coincident with the onset of optical-absorption.[14] However, as the film is exposed to air, the PC decreases particularly faster at $\hbar\omega < 2.3$ eV. Thus, oxygen within C_{60} films generates a (spatial and potential) disorder that localizes the electronic states at the band edges. This is due to the smaller contribution to the PC from carriers photoexcited at localized states than that from carriers occupying extended band states. In C_{60} fully contaminated by oxygen, it appears that the mobility gap that separates extended and localized states is at $\hbar\omega \approx 2.3$ eV.

The temperature dependence of the steady-state PC action spectra is depicted in Figure 2 for oxygen-free C_{60} (a) and C_{60} fully exposed to oxygen (b).

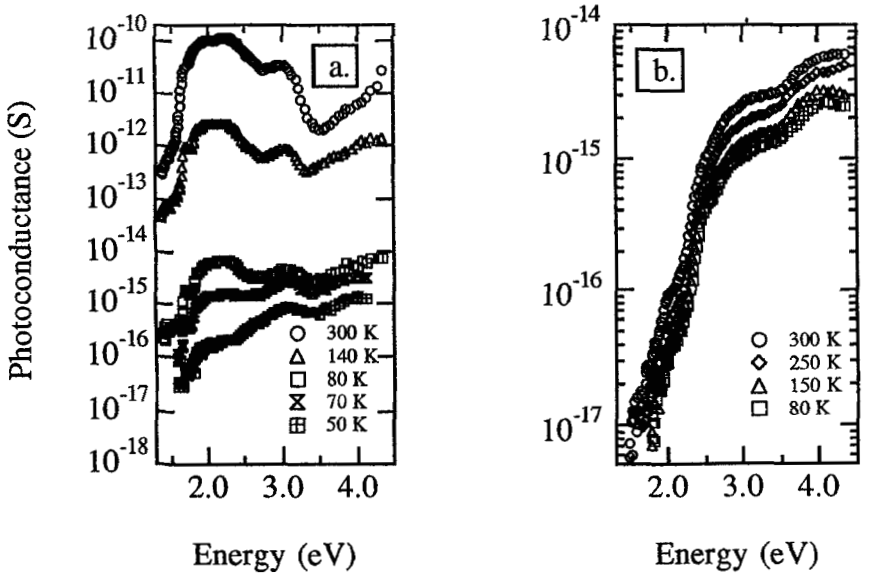

Figure 2 The temperature dependence of the steady-state PC action spectra for oxygen-free C_{60} (a) and C_{60} fully exposed to oxygen (b).

In oxygen-free C_{60}, the steady-state PC decreases dramatically as the temperature decreases, by more than 5 orders of magnitude at T=50 K. In contrast, in C_{60} fully exposed to oxygen, the PC shows very weak temperature dependence. The latter behavior indicates the effective quenching of the multiple trapping transport mechanism by the deep traps that are generated by oxygen in the bulk of C_{60} film.[13]

The spectra shown in Fig. 1 and Fig. 2 (a) are very similar, indicating that reducing of the ambient temperature affects the PC spectral dependence of C_{60} film in a similar qualitative way as exposing the film to oxygen. The well-resolved structure in the PC action spectra at the low energy region has been associated with vibronic transitions.[14]

The chopping frequency dependence of the steady-state PC shown in Figure 3 also reveals the similarity between reducing of the temperature and exposing of C_{60} to oxygen: The lifetime estimated from the frequency dependence of the steady-state PC is affected similarly by decreasing the temperature and exposing C_{60} to oxygen.

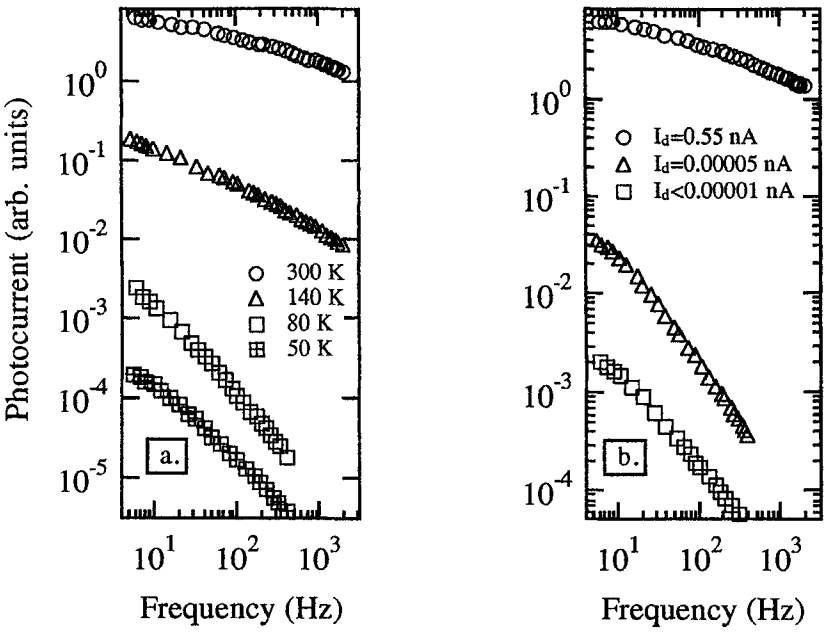

Figure 3 The chopping frequency dependence of the steady-state PC measured at $\hbar\omega$=2.64 eV for oxygen-free C_{60} at various temperature (a) and for various stages of oxygen exposure at 300 K (b).

Figure 4 displays the temperature dependence of the steady-state PC at $\hbar\omega$=2.64 eV of oxygen-free C_{60}, C_{60} partially-contaminated by oxygen, and C_{60} fully-contaminated by oxygen: Similar qualitative temperature dependences are observed at other photon energies as well. Fig. 4 also includes the dark conductance, which is characterized by an activation energy of about 0.5 eV.

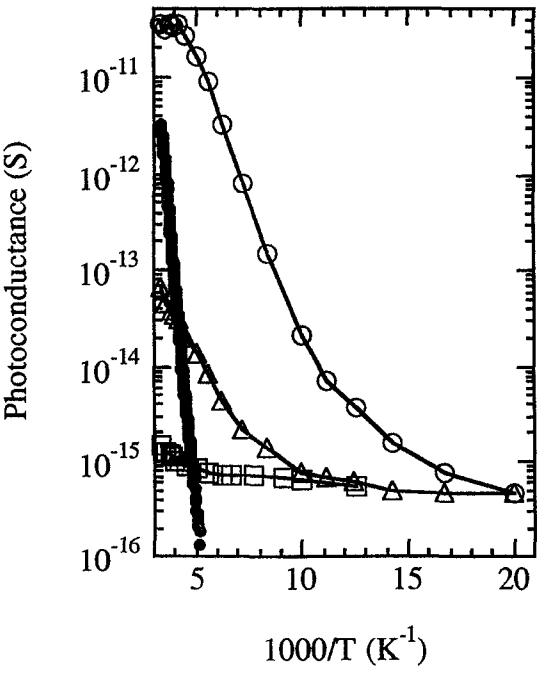

FIGURE 4 The temperature dependence of the steady-state PC at $\hbar\omega$=2.64 eV for oxygen-free C_{60} (○), C_{60} partially-contaminated by oxygen (△), and C_{60} fully-contaminated by oxygen (□), as well as the dark conductance of oxygen-free C_{60} (●). The lines are plotted as a guide to the eye.

The steady-state PC of oxygen-free C_{60} exhibits a peak at T≈240 K. This PC peak disappears as the C_{60} film is exposed to oxygen. In addition, exposure to oxygen decreases the characteristic activation energy of the steady-state PC, reduces the temperature region at which the transport is activated, and eventually in film fully exposed to oxygen causes the steady-state PC to become almost temperature independent. The results indicate that the thermally activated transport mechanism (multiple trapping transport) in oxygen-free C_{60} is effectively quenched in C_{60} contaminated by oxygen.

CONCLUSION

We have investigated the dynamics of photoexcited carriers in pristine C_{60} film and its evolution as the film is progressively exposed to oxygen by the steady-state PC measurement. The effect of oxygen contamination on the steady-state PC action

spectrum of C_{60} is qualitatively similar to the effect of reducing the ambient temperature of pristine C_{60}: in both cases the magnitude of the steady-state PC is severely reduced, where the change is faster at $\hbar\omega<2.3$ eV. In addition, exposure of C_{60} film to oxygen modifies the crystalline structural order to the extent that the maximum in the PC near 240 K at which pristine C_{60} undergoes a structural phase transition disappears. These observations indicate that oxygen in C_{60} film creates deep traps that effectively localize the photocarriers and thereby modifies the carrier dynamics and recombination kinetics. In particular, the thermally-activated transport mechanism (multiple trapping transport) in oxygen-free C_{60} is effectively quenched in C_{60} contaminated by oxygen, and consequently the steady-state PC becomes almost temperature independent.

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