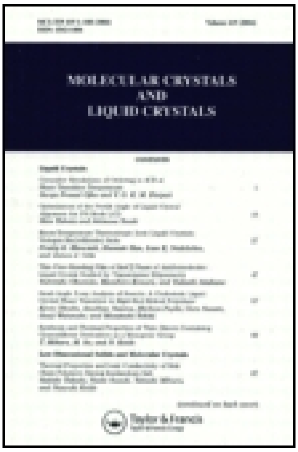

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Time-Resolved Photoinduced Absorption and Optical Kerr Effect in Fullerene C_{60}

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TIME-RESOLVED PHOTOINDUCED ABSORPTION AND OPTICAL KERR EFFECT IN FULLERENE C_{60}

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Abstract Time-resolved photoinduced absorption in fullerene C_{60} solutions is reported. The nonlinear index of refraction n_2 and third-order susceptibility $\chi^{(3)}$ in C_{60} film, measured by the optical Kerr gate technique, are also reported.

INTRODUCTION

To study kinetics of the excited state of fullerene C_{60}, time-resolved photoexcitation properties of C_{60} have been measured [1-4]. The optical properties of fullerene C_{60} and C_{70} have shown a reverse saturable absorption behavior [5]. Here we use the photoinduced absorption (PA) pump-probe technique, with femtosecond pulses, to investigate the photoexitation dynamics of fullerene C_{60}.

Optical nonlinearities in C_{60} have also been investigated [6-8]. However, almost all measurements of $\chi^{(3)}$ in fullerenes have been performed by means of third harmonic generation and four-wave-mixing experiments [6-8]. The optical Kerr effect (OKE) in C_{60} has not been tested. In an all-optical switching system, Kerr gates perform differently from four-wave-mixing gates. For four-wave mixing gates, phase matching leads to parametric gain so that a small signal can control a large signal. Kerr gates are a three-terminal device; there is no gain since a large pulse is used to control a smaller pulse.

The OKE is a transient optical-field-induced birefringence. A sufficiently intense pump wave propagating through a nonlinear media induces an intensity-dependent birefringence which can alter the polarization of a weaker probe beam. At steady-state, the intensity of a linearly polarized probe transmitted through a crossed polarizer can be expressed as

$$I = I_0 \sin^2(\Delta\Phi/2) \qquad (1)$$

$$\Delta\Phi = 2\ \pi\ n_2\ I_{in}\ L/\lambda$$

where I_0 is the probe beam intensity, I_{in} is the pump intensity, n_2 is the nonlinear index of refraction of the sample, $n_2 = 16\ \pi^2\ \chi^{(3)}/cn^2$, c is light velocity, n is the refractive index, and L is the thickness. Generally, $\Delta\Phi$ is sufficiently small so I can be approximated by

$$I \sim I_0(\pi\ n_2\ I_{in}\ L/\lambda)^2 \qquad (2)$$

The OKE signal I which is reported here originates from the real part of $\chi^{(3)}$, where $\chi^{(3)} = \chi^{(3)}{}_{1212} + \chi^{(3)}{}_{1221}$.

EXPERIMENTAL

Pulses with 250 fs duration at 5 Hz, tunable 575 nm to 610 nm were obtained from a Lambda Physik 500 FS. Peak power of the pump pulse was about 10^6 W. Photon density could reach 10^{17} cm^{-2}. The probe intensity was about 5% of the pump. For the transient photoexcitation measurement, a conventional one-color pump-probe technique was used. For the OKE experiment, the sample was placed between two crossed polarizers. The probe beam was initially polarized 45^0 with respect to the pump. The transient transmission intensity of the probe was measured as a function of delay time of the pump.

In order to estimate the nonlinear index of refraction in C_{60}, the OKE for CS_2 at STP ($n_2 = 2 \times 10^{-11}$ esu) [9] was measured as a standard. In addition, the OKE signal produced by the substrate (glass) was taken into account. We measured the OKE signal of the glass and then subtracted it from total the OKE signal to get a net signal of the sample

$$\sqrt{I_{net}} = \sqrt{I_{total}} - \sqrt{I_{glass}}$$

RESULTS AND DISCUSSION

C_{60} with a purity > 99.9% was dissolved in spectroscopic-grade dichlorobenzene and carbon disulfide (CS_2). Solution concentration was ~ 10^{-2} M. The absorption spectrum of C_{60} in dichlorobenzene in the region of 400 nm - 700 nm is plotted in Fig. 1. The weak absorption bands in the 410-620 nm region are assigned to the singlet dipole-forbidden transition in C_{60} molecules. Fig. 2 shows the time-dependent transmissions of C_{60} in dichlorobenzene and CS_2. At least 100 shots were taken and averaged for each experimental point. Both figures show a step-like change in the PA signal after o.6 ps. From o.6 ps to our experimental limit of 1.2 ns, no change in the PA signals with delay time was exhibited. Immediately after the pump excitation, the S_1 state is populated yielding the PA signal. Deexcitation of S_1 has two primary channels, either direct return to the ground state or inter-system crossing to the triplet manifold. A rate constant for intersystem crossing of 1.5 x 10^9 s; with the assumption that the quantum yield for triplet formation is near unity, has been reported [2]. Our results provide confirmation. In fact, when the time delay is in the nanosecond range, the PA signal results from absorption of both the S_1 and the S_3 states. The lifetime of the S_3 state has been reported to be on the order of μs [3,12].

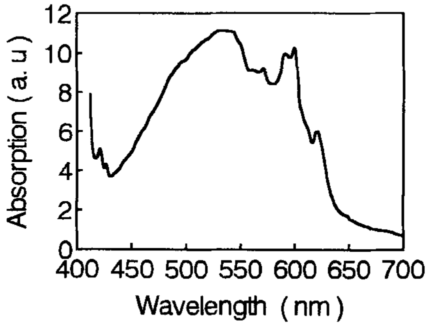

Fig. 1 Optical absorption spectrum of C_{60} in dichlorobenzene.

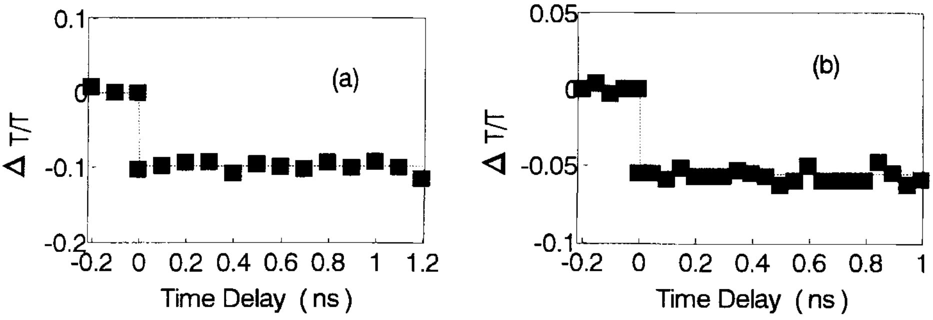

Fig. 2 Time -resovled PA of C_{60} in (a) dichlorobenzene at 585 nm and (b) CS_2 at 596 nm, the dashed line is for eye guide.

The OKE measurement in fullerene C_{60} film was performed at 585 nm. Here a C_{60} saturated solution was spin cast onto a glass substrate to form the film. A peak transmission of 0.3% was observed (Fig. 3). The time-dependent OKE signal presented in Fig. 3 was observed to have symmetric shape about t = o. The signal had a third-order correlation, $I \sim I_0 \times I^2_{in}$ (see Eq (2)). This implies that the Kerr gate in a C_{60} film immediately responds to the gate pulse. In other words, an instantaneous (10^{-15} s) electronic nonlinearity in the C_{60} film is responsible for the observed OKE. C_{60} molecules in the film are more or less frozen, so the contributions of molecular reorientation and redistribution to n_2 are insignificant. Surprisingly, the OKE temporal response in the C_{60} film is not affected by its PA relaxation (Fig. 2). Possibly the ground state contributions to the $\chi^{(3)}{}_{1221}$ tensor component dominate because excited-state contributions to the $\chi^{(3)}{}_{1221}$ tensor component are associated with the orientation correlation function of the transition dipoles [10,11]. Although absorption losses exist for the tested wavelengths, resonance enhancement is expected.

The results at 585 nm in C_{60} film showed $n_2 \sim 8.0 \times 10^{-10}$ esu and $\chi^{(3)} \sim 1.5 \times 10^{-11}$ esu. Generally, for an isotropic sample, the electronic $\chi^{(3)}(e)$ has the following relationship: $\chi^{(3)}_{1221}(e) = \chi^{(3)}_{1212}(e)$. Our measured value of $\chi^{(3)}_{1221}$ would be 7.5×10^{-12} esu which is close to 4.3×10^{-12} esu, as reported in Ref [6]. From a device point of view, we note that C_{60} film can be of potential use in a Kerr-effect light gate, because of possible resonance enhancement of $\chi^{(3)}$ and a femtosecond time response.

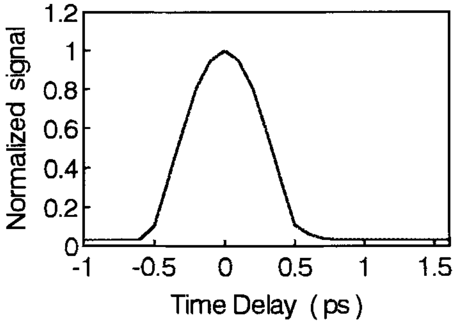

Fig. 3 Transmitted Signal vs various delay time of C_{60} film.

In summary, the PA showed no decay within a nanosecond range in solution fullerene C_{60}. The OKE in C_{60} film showed a femtosecond gate response and its nonlinear index of refraction ($n_2 \sim 8.0 \times 10^{-10}$ esu) and third-order susceptibility ($\chi^{(3)} \sim 1.5 \times 10^{-11}$ esu) were measured.

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