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Study of Nonlinear Optical Properties of C_{60} Benzene Solution

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The nonlinear optical properties of fullerene C_{60} in benzene solution were studied. The large third-order nonlinear susceptibility $\chi^{(3)}$ 4×10^{-9}esu has been measured by degenerate four wave mixing and the picosecond nonlinear response time 17ps originating from the electronic processes were obtained by using incoherent photon echo technique.

In 1985, a molecule consisting of sixty carbon atoms was identified in a cluster beam produced by laser ablating graphite[1]. The stability of this novel species was ascribed to the geodestic properties inherent in a truncated icosahedral hollow cage. From mass spectrometer, X-ray and electron diffraction studies, that is consists of hexagorally packed spheroidal molecule with 1nm array spacing, a result beautifully consistent with the proposed structure[2]. This special structure of fullerene C_{60} may be used as novel nonlinear optical material.

In our experiment, the fullerene C_{60} was obtained by operation of carbon arc in atmosphere of helium and soluble products were extracted from the soot by the Soxhle method using benzene. The resulting solution contained C_{60} and C_{70} in a 5:1 as estimated by FTMS apparatus and Nicoled 5DX FT- IR spectraphotometer analysis, the mass spectrum of the fullerene shown in Fig 1.

The nonlinear optical properties of C_{60} in benzene solution were studied. The nonlinear susceptibility $\chi^{(3)}$ was measured by the degenerate four wave mixing (DFWM). The dye laser at 585nm pumped by SHG of pulsed YAG laser was used as a source, which is split into three equal beams, the angle between probe beam and forward pump beam was about 2°, according to the theory of DFWM, the $\chi^{(3)}$ can be obtained from

$$\chi^{(3)}_{s} = \left|\chi^{(3)}_{r}\right| (C_s/C_r)^{1/2}(n_s/n_r)^2 \times \{L\alpha/[(1-e^{-L\alpha})e^{-L\alpha/2}]\} \quad ①$$

where $\chi^{(3)}_{r}$ is the nonlinear susceptibility of reference sample CS_2, it is known to be 6.8×10^{-13} esu, C_s and C_r are conjugate reflectivity of sample and reference, respectively . The hyperpolarizability of single molecule γ_{1111} can also be calculated from $\chi^{(3)}$ in formula ②

$$\gamma_{1111} = \chi^{(3)}\varepsilon_0/\{N/[(n^2+1)/3]^4\} \quad ②$$

N is the molecular concentration, $(n^2+1)/3$ is the Lorentz local field factor. From our experiments, the $\chi^{(3)}$ of C_{60} was determined as about 4×10^{-9} esu in the concentration of 10^{-3}mol/l, the γ_{1111} of C_{60} was calculated as about 3×10^{-28}esu.

The photon echo signal which is measured as a function of the delay time between the pulses is displayed in Fig 2. Experimental apparatus consist of a broadband R6G dye laser pumped by SHG of YAG laser producting pulse of 10 ns duration at repetition rate of

5HZ, we employ laser pulse with the bandwidth $\Delta\lambda$ of about 1.3nm, at 565nm, associated with $\tau_c \cong \lambda^2/\pi C \Delta\lambda$, of 0.5ps. Output dye laser is split into two beams (wavevector $\boldsymbol{K_1}$, $\boldsymbol{K_2}$), the second pulse is delayed in time, both pulses are focussed onto the C_{60} benzene solution, the angle is about 2 °, photon echo emitted in the direction $2\boldsymbol{K_2} - \boldsymbol{K_1}$ are detected and accumulated by a Boxcar intergrator. From Fig2, the nonlinear response time 17ps originating from the electronic process was obtained.

1) H.W.Kroto, J.R.Heath, S.C.O'Brien, R.F.Curl and R.E.Smalley Nature(London) 318 (1985) 162

2) W.Kratschmer, L.D.Lamb K.Fostiropoulos and D.R.Huffman Nature(London) 374 (1990) 354

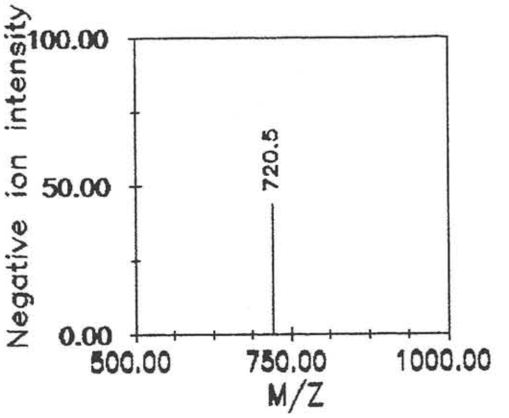

Fig.1 Mass spectrum of fullerene C_{60} prepared by extraction in benzene

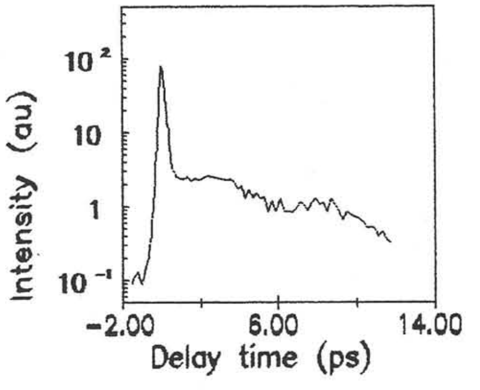

Fig. 2 Photon echo via delay time between pump and probe beams