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Negative group velocity in C_{60} due to RSA effect

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ABSTRACT We experimentally observed negative group velocity in C_{60} toluene solution at the wavelength of 532 nm for the first time. The observed time advancement of the modulated signal appears to be due to the RSA (reverse saturable absorption) effect of the C_{60} molecule. The largest time advancement of 71.65 ms was obtained with a C_{60} sample of 1.5-mm length with the concentration of 1.39×10^{-3} mol/l; the corresponding group velocity was −0.021 m/s. We could control the group velocity by adjusting the modulation frequency and the input intensity.

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Controllable slow and fast light in nonlinear media are of fundamental interest in physics. Their attractive features are not only helpful for deepening our understanding of physical laws but also in their potential applications in telecommunications and signal processing. Using the electromagnetically induced transparency (EIT) technique, the group velocity of light was slowed down significantly in atomic vapor due to a quantum interference effect [1–4]. The technique of coherent population oscillation (CPO) was also used to realize sub- and superluminal propagation at room temperature [5–15]. And, many other methods have been used to achieve slow and fast light, such as two-wave mixing [16], stimulated Brillouin scattering [17], stimulated Raman scattering [18], etc. Recently, slow and fast light were also realized in many new types of matter, such as optical fibers [17, 19, 20], quantum dots [21], quantum wells [11, 22], biological thin films [15], photonic crystals [23] and semiconductors [24]. Slow and fast light in new materials would be of undoubted significance in developing these researches to broad fields.

In a previous paper, we examined pulse propagation in C_{60} toluene solution at the wavelength of 514.5 nm experimentally [25]. It was shown that the reverse saturable absorption (RSA) effect was achieved in C_{60} at 514.5 nm and superluminal propagation was observed in it. We could control the group velocity by adjusting the modulation frequency. In this paper, we discuss the possibility of the generation of superluminal propagation due to the RSA effect at the wavelength of 532 nm and obtain negative group velocity in C_{60} for the first time. We found that we could control the time advancement by changing not only the modulation frequency but also the input intensity. One of the striking effects predicted in our experiment was that superluminal propagation could be realized in a broad wavelength range due to the wide RSA effect region of C_{60}. These methods could also be used in other materials with similar properties.

Reverse saturable absorption is a phenomenon which occurs due to higher absorption from a photo-excited state compared with that from the ground state. Figure 1 gives the ground-state absorption spectrum of C_{60} solution. From this figure we can see that C_{60} has relatively weak and even absorption from 420 nm to 1200 nm compared with that from 300 nm to 420 nm and the RSA effect can be realized in this region easily [26]. At the condition of RSA, the absorption of the incident photons results in excitation of the C_{60} molecules from the ground state S_0 to the singlet state S_1 and these molecules quickly transfer to the long-lived lowest triplet state T_1 through fast intersystem transitions. RSA results because the absorption from the triplet state T_1 is stronger than that from the ground state S_1. By virtue of the RSA effect, fast light can be observed in C_{60} and it is valid from the analysis of the density matrix model [25].

From the above analysis, we chose a Nd:YAG laser at 532 nm as light source and realized the superluminal propagation with negative group velocity in C_{60} under the RSA condition. The experimental setup is shown in [25]. Briefly, the modulated sinusoidal signal from an electro-optic modulator was divided into signal light and reference light. The signal light was focused by a lens with the focal length of 11 cm and injected into a quartz cell of 1.5 mm that is full of C_{60} toluene solution with the concentration of 1.39×10^{-3} mol/l. The transmitted signal and the reference signal were received by detectors and fed into a two-channel oscilloscope for comparison. We can obtain the waveforms of the signal light and the reference light and the time advancement by data processing.

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Figure 2 gives the typical waveforms of the input signal and the output signal at the modulation frequency of 50 Hz when the input power was 180 mW. The output signal arrived at the detector 1.07 ms earlier than the reference signal with the group velocity of −1.5 m/s and kept its original waveform nearly unchanged. By adjusting the modulation frequency, we also obtained the relation between the time advancement and the modulation frequency and calculated the corresponding group velocity. As shown in Fig. 3, the largest time advancement of 71.65 ms at 4 Hz was observed with the corresponding group velocity of −0.021 m/s. And, this time advancement was the largest one observed in C_{60}, up to now, to our knowledge. The time advancement decreased with the increase of modulation frequency and disappeared when the modulation frequency exceeded several kilohertz. This is due to the intrinsic property of fixed lifetime and transition rate of electronic states of C_{60}.

It is well known that the RSA effect is intensity related, so we observed the time advancement at different input powers. Figure 4 demonstrates the time advancement of the output pulse relative to the input pulse a function of input power when the modulation frequency was 20 Hz. From this figure we can see that, initially, the time advancement increased with the input power, for the RSA effect became stronger and stronger with the increasing of the input intensity. But, the time advancement nearly ceased to further increase with the input power when the input intensity was larger than about 300 mW. This saturation phenomenon of the time advancement may be attributed to the local heating effect. When the input power was strong enough, the convection of the liquid could not counteract the heating effect effectively. So, the lifetime of the absorbing states of the C_{60} molecule decreased due to the local heating and led to the saturation of the time advancement.

From Fig. 4, we can also see that larger time advancement and group velocity could be obtained at higher input power and changing the input power was an alternative way to change the group velocity, as well as adjusting the modulation frequency. And,

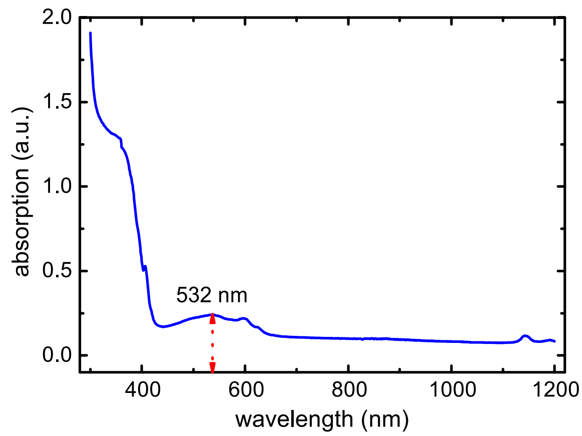

FIGURE 1 Ground-state absorption spectrum of C_{60} molecule

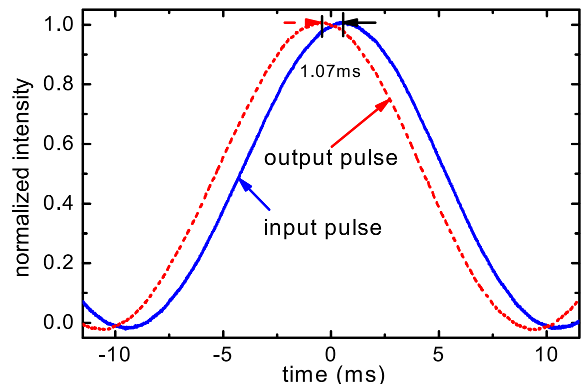

FIGURE 2 Normalized waveforms of input (*blue*) and output (*red*) pulses. The modulation frequency is 50 Hz. Input power is 180 mW

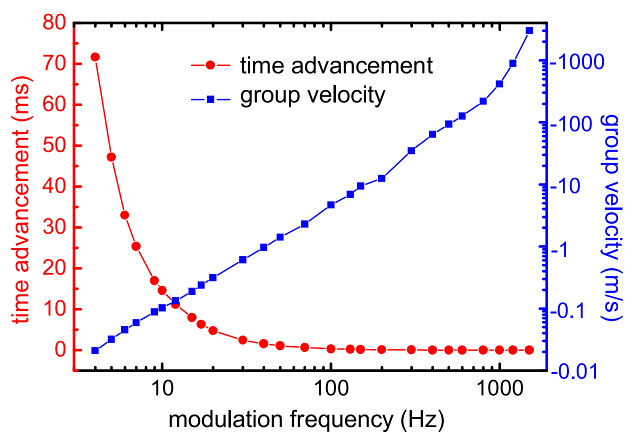

FIGURE 3 Relation between time advancement (*red*) and modulation frequency. And, the corresponding group velocity (*blue*). The input power is 180 mW

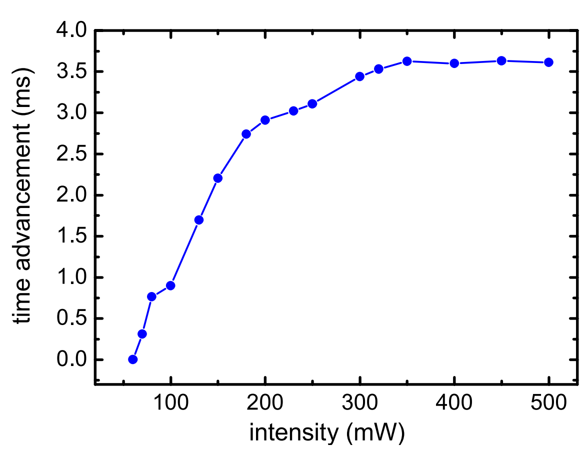

FIGURE 4 Relation between time advancement and input intensity. The modulation frequency is 20 Hz

even larger advancement was possible under higher input intensity with proper considerations.

In conclusion, we analyzed the possibility of superluminal propagation due to the RSA effect at 532 nm in C_{60} and experimentally obtained the negative group velocity in it for the first time. We could control the time advancement, i.e. the group velocity, not only by changing the modulation frequency but also by adjusting the input laser intensity. The largest time advancement of 71.65 ms was observed with the corresponding group velocity of -0.021 m/s. These analyses would be reasonable in other nonlinear media with similar properties.

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