Dynamics of Rapid Melting and Recrystallization

Lu Jie

National Synchrotron Radiation Lab (NSRL) & School of Nuclear Science and Technology

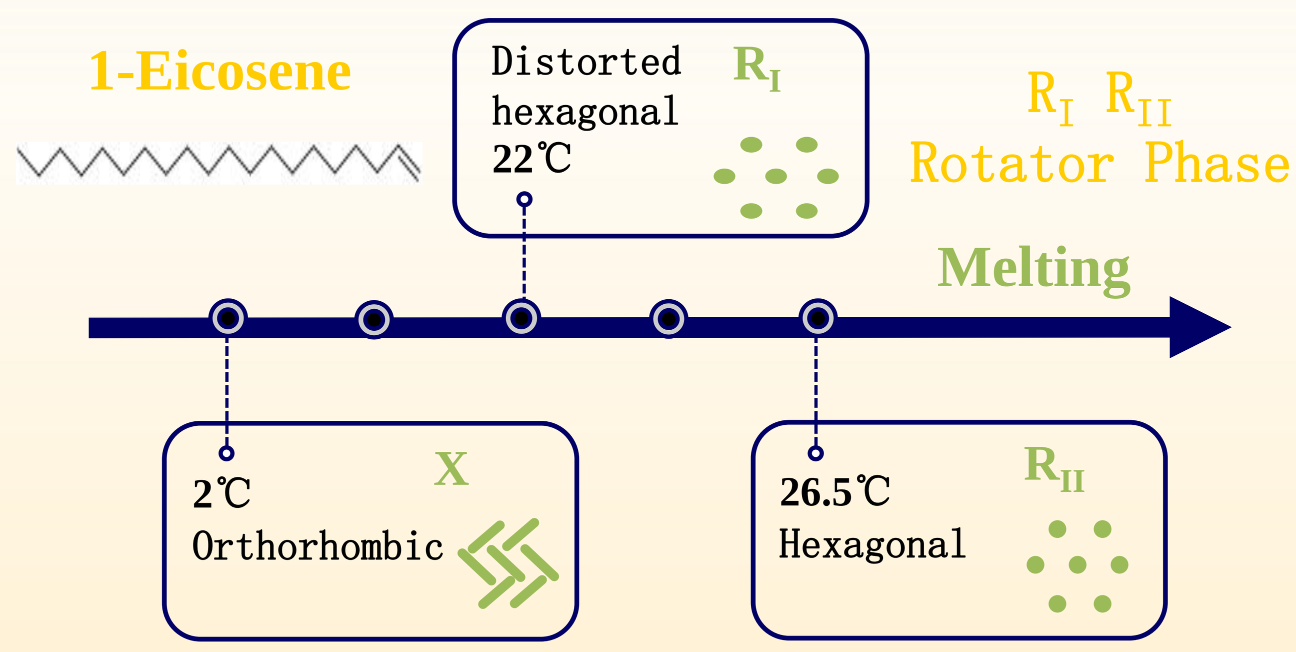
University of Science and Technology of China (USTC), Hefei, China

Introduction

Since crystals are widely spread in nature, phenomena of melting and recrystallization have been intensively studied by multiple methods such as X-ray scattering, DSC and molecular dynamic simulation. At the same time, very few reports are devoted to the study of melting induced by resonant absorption of mid-infrared lasers. It is already known that resonant absorption induces much greater thermal effect than that caused by non-resonant absorption with friction movements among molecules. Thus it is very important to understand the intrinsic mechanism of resonant melting. What is the difference of melting process between resonant and non-resonant absorption? Does it have any relationship with the vibrational mode or the position of excited groups on molecular chains?

In the current work we exploit pump-probe method to investigate the dynamics of rapid melting and recrystallization based on mid-infrared QCLs.

Experiments and Data



1-eicosene is a rodlike molecule with simple structure, which has three different crystal phases. It is very convenient to locally heat according to the position of different groups.

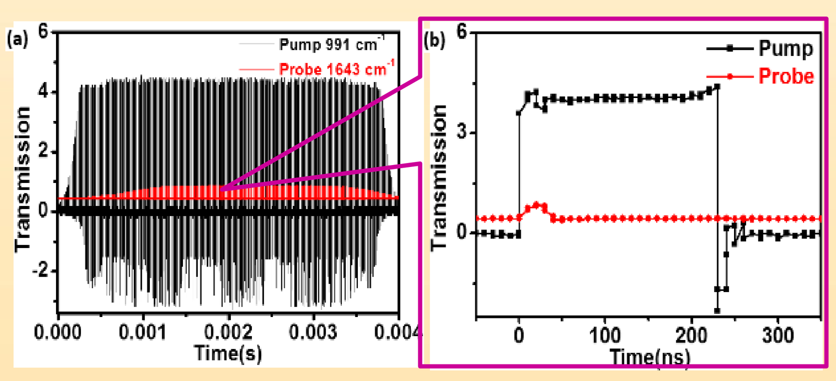


Fig.3 (a) Transmission change of probe beam at 1643 cm^{-1} versus time and (b) partially enlarged view of a single pulse.

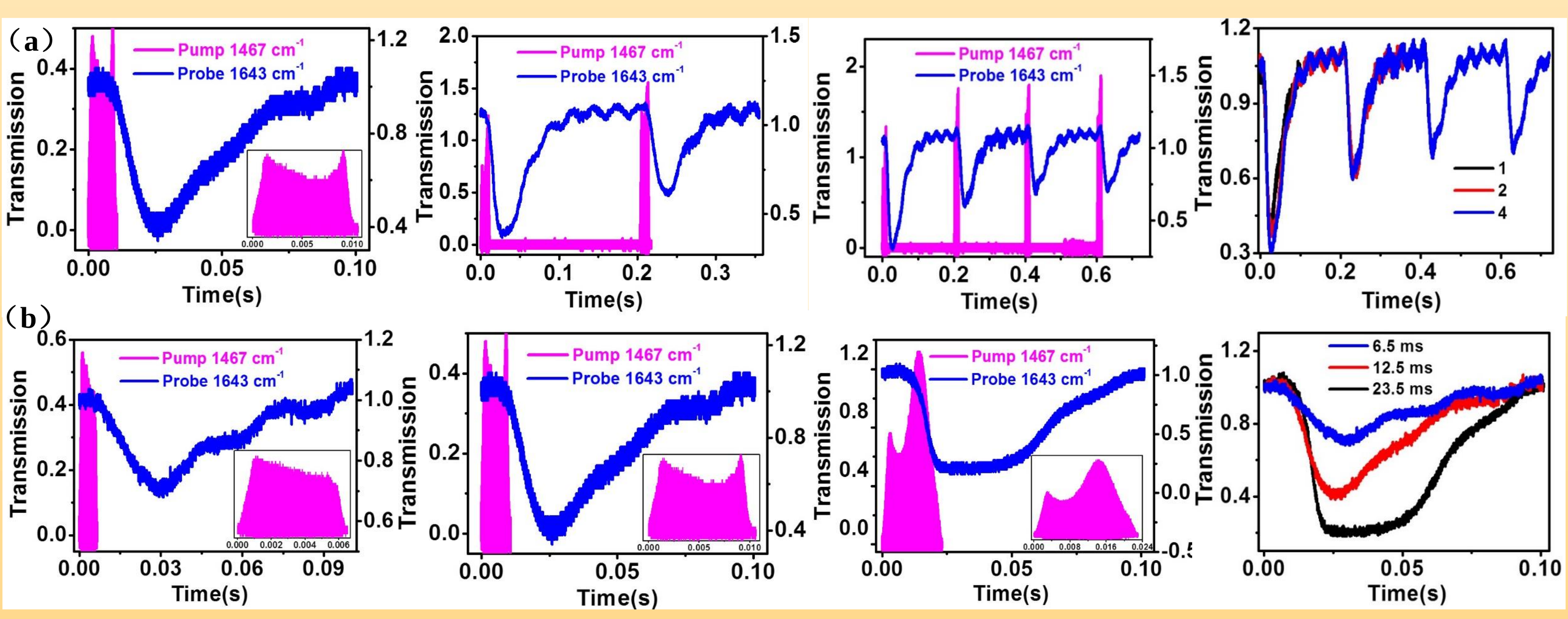


Fig.4 Changes in the transmission intensity of probe beam versus time for different (a) pump frequency and (b) pump time. The wavenumber of pump beam was tuned to 1467 cm^{-1} corresponding CH_2 bending vibration while that of probe beam was tuned to 1643 cm^{-1} corresponding to C=C stretching vibration.

Pump-Probe Method

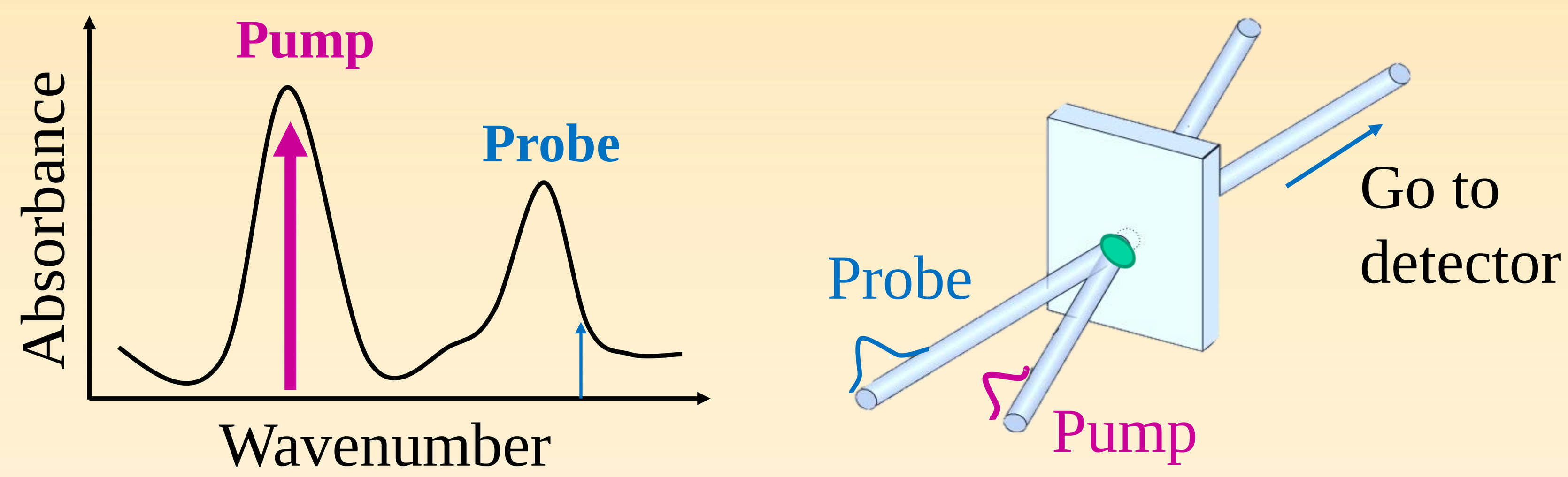


Fig.1 Schematic of pump-probe method.

Intense infrared pulses were regarded as pump beam to excite the particular groups on molecular chains while weak infrared pulses comprised probe beam to monitor molecular movements. The pump and probe beams were focused on the same location of sample. The wavelength of the resonant IR radiation was adjusted in such a way that it coincided exactly with the absorbance peak in the spectrum of 1-eicosene corresponding to the selected groups.

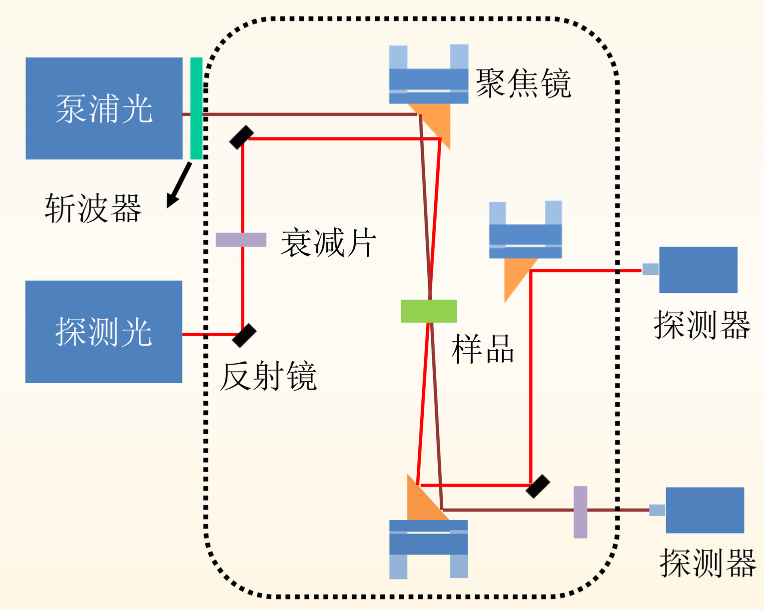
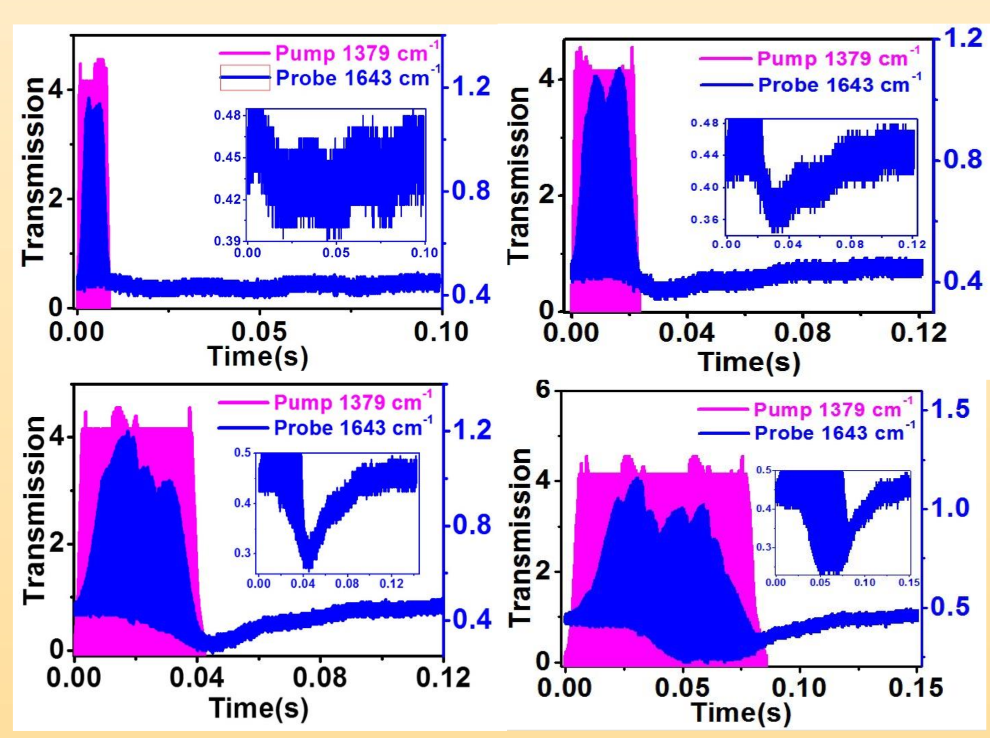


Fig.2 Diagram for light path

The vibration amplitude of excited molecules increases under irradiation of resonant pump beam. Thermal effect intensifies and phase transition occurs as temperature reaches the melting point. The intensity of probe beam changes as excited molecules move. Thus relationship between the intensity of probe beam and molecular motions can be established.

Plans

Fig.5 Transmission change of probe beam versus time for different pump time. The wavenumber of pump beam was tuned to 1379 cm^{-1} corresponding CH_2 bending vibration while that of probe beam was tuned to 1643 cm^{-1} corresponding to C=C stretching vibration.



The pump beam at different wavenumber excited distinct groups on the molecular chain inducing various transmission change of probe beam. It is necessary to further investigate if this phenomenon is only the properties of 1-eicosene or more general. Does the effect of resonant groups relate to the overall molecular arrangement? Thus more and more efforts need to be devoted to instinct mechanism.

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