Nuclear Instruments and Methods in Physics Research A304 (1991) 797
North-Holland

Section XII. FEL applications

Picosecond FEL experiments on condensed matter systems

Michael D. Fayer
Department of Chemistry, Stanford University, Stanford, CA 94305, USA

Invited paper

Applications of picosecond pulses from a free electron laser (FEL) are discussed. A specific example, photon echo experiments on a near-infrared dye molecule, is described.

A FEL which is capable of producing picosecond-timescale pulses provides unique capabilities for conducting condensed matter research [1]. The FEL can be used to obtain information of fundamental importance on a wide variety of materials such as high-temperature superconductors, semiconductors, including amorphous materials and multiquantum wells; inorganic and organic crystals and glasses; biological materials, for example hemoglobin, proteins, DNA, and membranes; surfaces and the nature of molecules on surfaces; polymeric solids, and liquids. In all cases it is the unique operating characteristics of a picosecond FEL, providing single, high-powered, picosecond pulses tunable across the IR spectrum, which makes it possible to probe matter with methods that have previously been impossible.

The IR spectrum, while not readily accessible with conventional sources of ps pulses, is extremely important for the understanding of the properties of condensed matter systems. Visible and ultraviolet light correspond to energies which are resonant with high-energy states of systems, e.g. electronic states of molecules. The IR, however, corresponds to the relatively low-energy states of molecular and atomic systems, e.g. the vibrational states of small and large molecules, polymers, and proteins, as well as the band gaps in novel semiconductors and multiquantum well structures, and the pair energy gap in high-temperature superconductors. Thus the FEL can open vast new areas of materials research for study which cannot be readily explored with conventional laser systems.

The requirements of modern ultrafast experiments place extreme demands on the laser source. This is particularly true for optical nonlinear experiments which are becoming widely used with conventional lasers. Biophysical and materials science applications of FELs are important. A variety of techniques are possible, including infrared pump/probe and infrared pump/Raman-probe experiments of vibrational relaxation in solids, proteins, and surfaces; two-color experiments using one infrared color and a visible or UV wavelength (from a synchronized conventional laser); and two infrared colors simultaneously generated by a single FEL. As a detailed example, photon echo experiments were performed. These are the first time-resolved picosecond experiments and the first nonlinear experiments using a FEL as the laser source.

Photon echo experiments were performed on a near-infrared dye molecule in a polymeric solid at liquid-helium temperatures using the Stanford SCA/FEL [2]. The photon echo is a nonlinear experiment which involves three interactions of the sample with the radiation field. The incoming pulses generate a polarization in the sample which gives rise to an outgoing coherent pulse of light which propagates in a unique direction. This outgoing pulse is the signal, and is referred to as the echo. As the time between the incoming pulses is delayed, the echo signal becomes smaller. This is called the echo decay. It contains detailed information on the dynamics and the intermolecular interactions in the medium. By measuring the echo decay as a function of temperature, the extent of the fluctuations of the medium and the nature of the motions and interactions were determined [2,3].

References

[1] D.D. Dlott and M.D. Fayer, J. Opt. Soc. Am. B6 (1989) 977.
[2] S.R. Greenfield, Y.S. Bai and M.D. Fayer, Chem. Phys. Lett. 170 (1990) 133.
[3] L.R Narasimhan, K.A. Littau, D.W. Pack, Y.S. Bai, A. Elschner and M.D. Fayer, Chem. Rev. 90 (1990) 439.