3P076

Ultrafast Scanning Near-Field Optical Microscope Based on Nonlinear Group Velocity Dispersion Compensation

(Institute for Molecular Science and The Graduate University for Advanced Studies,[a] Waseda University [b]) ○Hui Jun Wu [a], Kohei Imura [b], and Hiromi Okamoto [a]

[Introduction] Surface plasmons (SP) of metal nanostructures have attracted great interests recently for its fundamental importance not only in the field of basics nano-optics but also for their application to wide research areas in physics, chemistry and biology. However, details of spatial and temporal characteristics of plasmons are still not fully understood, partly because the spatial scale of plasmons is much smaller than the diffraction limit of light, and thus conventional optical microscope is of little use. Near-field method can overcome this problem, and can explore plasmons in real space. The time scale of plasmon dynamics is of order of 10-20 fs or even shorter, and ultrafast technique that can resolve 20 fs (or less) is essential to directly observe the dynamics. By combining the near-field method with the ultrafast time-resolved technique, we can construct an ultrafast near-field optical microscope which enables studies of spatial and temporal behaviors of plasmons at one time. We previously reported time- and space-resolved studies on nanomaterials based on the ultrafast scanning near-field optical microscope (SNOM). In those works the time resolution was limited to ~100 fs by the light source. This time resolution is not enough to further clarify the dynamic properties of surface plasmons. In the present study, we construct an ultrafast SNOM with much higher time resolution, using a further shorter pulsed light source. Since group velocity dispersion (GVD) arising from optical components (especially the optical fiber of the near-field probe) seriously broadens the pulse width, higher time resolution cannot be obtained by simply replacing the laser source to a shorter pulse one. To solve this problem, a pulse-shaping device is combined with the light source before introducing the light to the near-field microscope. We have succeeded in developing a basic technology to achieve the time-resolution as high as lifetime of plasmons at the probe tip of the near field microscope.

[Experimental] Figure 1 shows a schematic diagram of the experimental setup. In this setup, the near-field microscope was replaced with far-field second-harmonic (SH) generation optics to examine the performance of the pulse shaping system. The laser beam from a Ti:sapphire laser with ca. 15-fs pulse duration was dispersed by a grating and collimated by a concave mirror, and incident onto a deformable mirror. We can adjust phase shifts (optical path lengths) of the dispersed frequency components of the incident light, by controlling the surface shape of the deformable mirror. A prism pair (SF14) was adopted to pre-compensate linear GVD effects arising from optical components. These devices act as a pulse shaping system. The laser beam was then coupled into a 150-mm long optical fiber. Output beam from the fiber was collimated by an objective lens and focused onto a BBO crystal to generate SH photons. SH photons were detected by a photomultiplier tube and were fed to the computer. A genetic algorithm was used to give the highest SH signal intensity, by optimizing the surface shape of the deformable mirror in an adaptive way.

[Results and Discussion] In the previous SNOM system in our lab, the GVD effect was compensated by a grating pair, with which we could recover the original pulse width (~100 fs) at the near-field tip. In the present study, we first tried to shorten the pulse width only with the prism-pair GVD compensator. Figure 2a shows a typical auto-correlation trace of the pulse obtained. The autocorrelation width obtained (~135 fs) in this setup is found to be much broader than that of the original pulse (~23fs, autocorrelation width). This is due to the nonlinear GVD occurred in the fiber. Such a nonlinear GVD contribution becomes significant, as the pulse width is extremely short.

In order to get rid of the nonlinear GVD contribution, the deformable mirror of the pulse shaping system was operated. The deformable mirror finely tunes optical path length of each wavelength component of the light. Figure 2b shows an autocorrelation trace of the pulses obtained using the pulse shaper. The autocorrelation width obtained (33fs) is much shorter than that obtained without the pulse shaper, and is close to the correlation width of the original Ti: sapphire laser output. The obtained pulse width after the fiber might be short enough to clarify the plasmon dynamics. We are planning to explore spatial and temporal characteristics of metal nanostructures after combining this device with a near-field microscope.

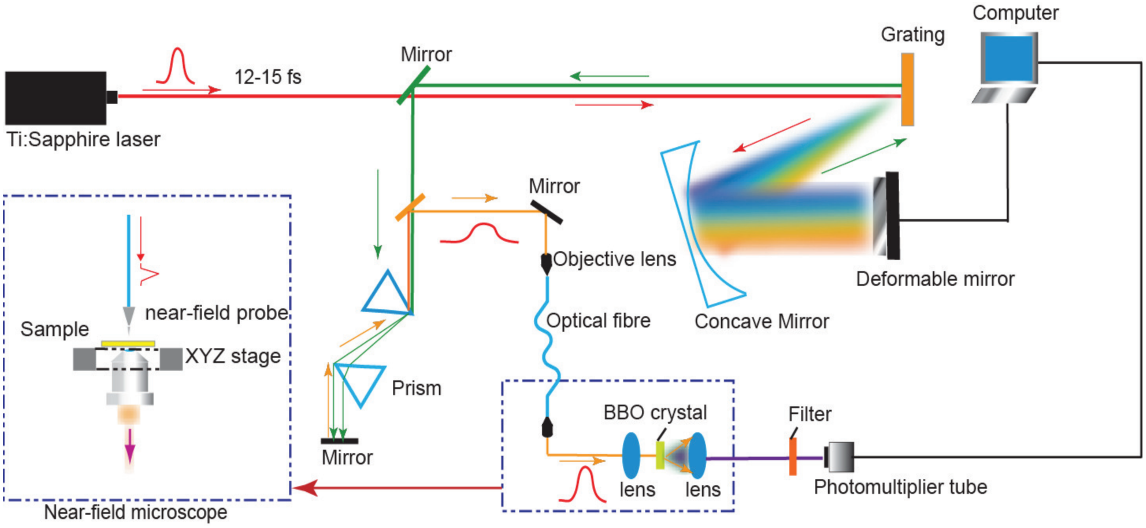

Fig 1. Schematic diagram of the experimental setup.

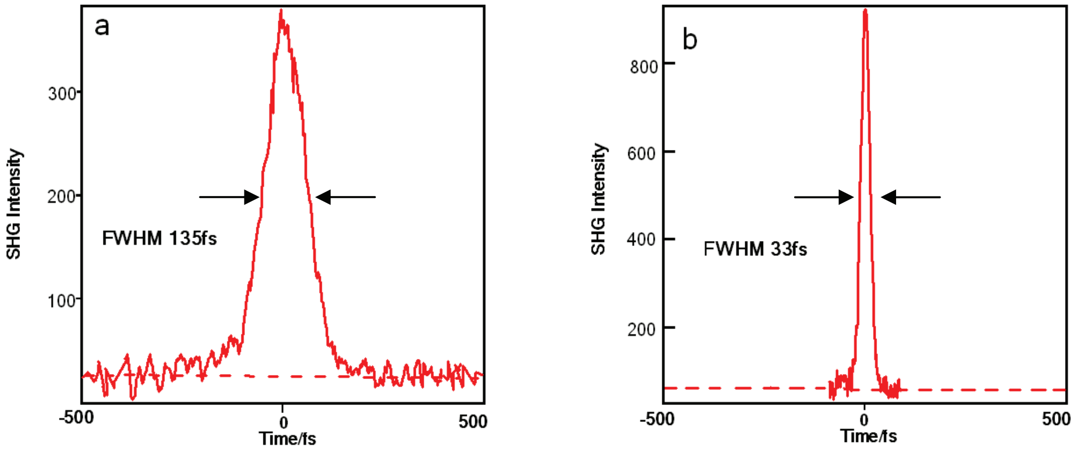

Fig 2. Autocorrelation trace before (a) and after (b) optimization. The dotted lines indicate base line of the SHG signal.