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Periodic structures on germanium induced by high repetition rate femtosecond laser

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ABSTRACT

Laser-induced periodic surface structures (LIPSS) are studied on germanium surface in air by the femtosecond pulsed laser with repetition frequency of 76 MHz and wavelength λ of 800 nm. Three types of LIPSS were found and they are low-spatial-frequency LIPSS (LSFL), high-spatial-frequency LIPSS (HSFL), and LSFL superimposed with HSFL. The period Λ_{LSFL} of LSFL shrinks quickly from approximately 650 nm to 400 nm ($\sim\lambda/2$) when lowering the scanning speed. Comparatively, the period Λ_{HSFL} of HSFL keeps almost constant between 90 and 100 nm ($\sim\lambda/8$) when the scanning speed and the laser pulse energy vary. LSFL and HSFL coexist when the laser pulse energy is around 3.3 nJ/pulse and the scanning speed ranges between 3 and 8 mm/s. The surface plasmon polariton waves make a contribution to the formation of LIPSS and the fourth harmonic generation (FHG) might be involved in the formation of HSFL.

1. Introduction

Laser-induced periodic surface structures (LIPSS, ripples) are attracting more attention due to new features imposed by femtosecond (fs) lasers, which can produce surface patterns with spatial period Λ much smaller than the irradiated laser wavelength λ and this nanostructuring beyond the diffraction limit is of importance in science and technology due to its simplicity and controllability [1–3]. In general, two distinct types of LIPSS induced by fs lasers have been observed: low-spatial-frequency LIPSS (LSFL, coarse ripples, near sub-wavelength ripples: $0.5 \leq \Lambda/\lambda$) and high-spatial-frequency LIPSS (HSFL, fine ripples, deep sub-wavelength ripples: $\Lambda/\lambda < 0.5$). LSFL can be formed under a relatively wide laser parameters and are easily observed, in contrast, HSFL are not so common as LSFL, difficult to be produced and observed, and may come from diverse origins such as self-organization [4], second and higher harmonic generation [5,6], excitation of surface plasmon polaritons (SPP) [7–9] and redistribution of the electric field [10]. Although both of these LIPSS have been reported on different materials, including semiconductors [11–15], metals [16–19] and dielectrics [20–22], the formation conditions and mechanisms of these surface periodic structures, especially of HSFL, are not particularly clear. Under fs laser irradiation, the permittivity of materials, especially semiconductors and dielectrics, is transiently adjusted and the materials show a tendency of metallization, which is important to the formation of LIPSS and has to be considered when exploring the formation mechanism of LIPSS. As an important semiconductor, germanium has the similar structure as silicon. Both of them are the elements of the carbon group and possess the same number of outermost electrons. Besides, they are all diamond-type structure. Furthermore, germanium possesses special optical and electrical properties between metals and semiconductors, such as high refractive index, high absorption coefficient and narrow band gap. Therefor germanium is a good choice for understanding the difference in LIPSS between metals and semiconductors.

In this work, the LIPSS on single crystal germanium are studied by irradiation of a high-repetition-rate femtosecond pulsed laser in air. The femtosecond laser with high repetition rate (MHz) has been proved to be a good tool to induce HSFL due to its low pulse energy (nJ) and low etching rate per pulse, and HSFL are usually formed in a narrow range of laser fluence and scanning speed (or pulse number) under femtosecond laser irradiation [23,24]. The results reveal that the surface plasmon polaritons play a crucial role in LSFL ($\Lambda_{LSFL} \approx \lambda/2$) and HSFL ($\Lambda_{HSFL} \approx \lambda/8$) on Ge surface might be related to fourth harmonic generation.

2. Experimental

A compact Ti: sapphire tunable laser system (Millennia Pro 6s, Spectra Physics in USA) was used for providing femtosecond laser

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pulses with central wavelength λ of 800 nm, repetition rate of 76 MHz, duration of 50 fs and maximum single pulse energy of 40 nJ. The specific experimental setup can be found in Ref. [23]. Due to the low pulse energy of the fs laser, a 25× focusing objective with numerical aperture (NA) of 0.40 was employed to reach the fluence threshold of LIPSS formation on germanium. The focused spot is the Gaussian beam and its diameter D was estimated with the relation $D = 1.22\lambda/NA = 2.5$ μm. The peak fluence $F = 2E_p/(\pi D^2/4)$ on the target was estimated with the measured laser pulse energy E_p and spot diameter D. In our experiment, the incident laser pulse energies ranged from 2.2 to 3.7 nJ/pulse (the corresponding fluences from 0.089 to 0.151 J/cm^2) and laser scanning speeds were 0.5–10 mm/s. The ⟨1 0 0⟩ monocrystalline single-sized polished germanium samples with a thickness of 525 μm were fixed on the stage of a high precision computer-controlled three-dimensional translation stage (Newport in USA) and scanned by tightly focused femtosecond laser in air environment. The surface morphology was characterized by field emission scanning electron microscope (SEM, Ultra 55, Zeiss Instrument Company in Germany).

3. Results

Fig. 1 shows SEM images of LIPSS observed on germanium surfaces after femtosecond laser pulses irradiation with different laser scanning speeds in air. The femtosecond laser pulse energy is fixed at 3.3 nJ/pulse, and the laser scanning speeds are 10, 8, 5, 3, 1 and 0.5 mm/s, respectively. The numbers of laser pulses per spot are described by the relation $N = D \cdot f/v$ [25]. Here, $D = 2.5$ μm is the focused Gaussian spot diameter, $f = 76$ MHz is the pulse repetition frequency and v is the laser scanning speed. The corresponding numbers of irradiated laser pulses are 19,000, 23,750, 38,000, 63,000, 190,000 and 380,000, respectively. The insets of Fig. 1 (a) and (b) are the magnified SEM images. The scanning direction S and laser polarization E are indicated in Fig. 1(a). With decreasing the laser scanning speed (increasing the number of laser pulses), a clear evolution process of LIPSS is revealed on the germanium surface. When the laser scanning speed is as high as 10 mm/s in Fig. 1 (a), the distinct, regular and sharp LSFL with orientation perpendicular to the laser polarization are observed and the spatial period of LSFL is 610 ± 38 nm. The ridges and grooves of these LSFL are very smooth, and some grooves of LSFL have a tendency to generate HSFL with smaller period of around 95 nm, as shown in the inset of Fig. 1(a). With decreasing the laser scanning speed, the LSFL become blurred, and the HSFL emerge for the first time at the grooves of LSFL for the scanning speed of 8 mm/s in Fig. 1(b) and then spread out on all the LSFL when the scanning speed is lower to 5 mm/s. Further slowing the scanning speed, the LIPSS were fragmented in Fig. 1(d) and (e), then both LSFL and HSFL were destroyed in Fig. 1(f). Admittedly, although the laser beam used

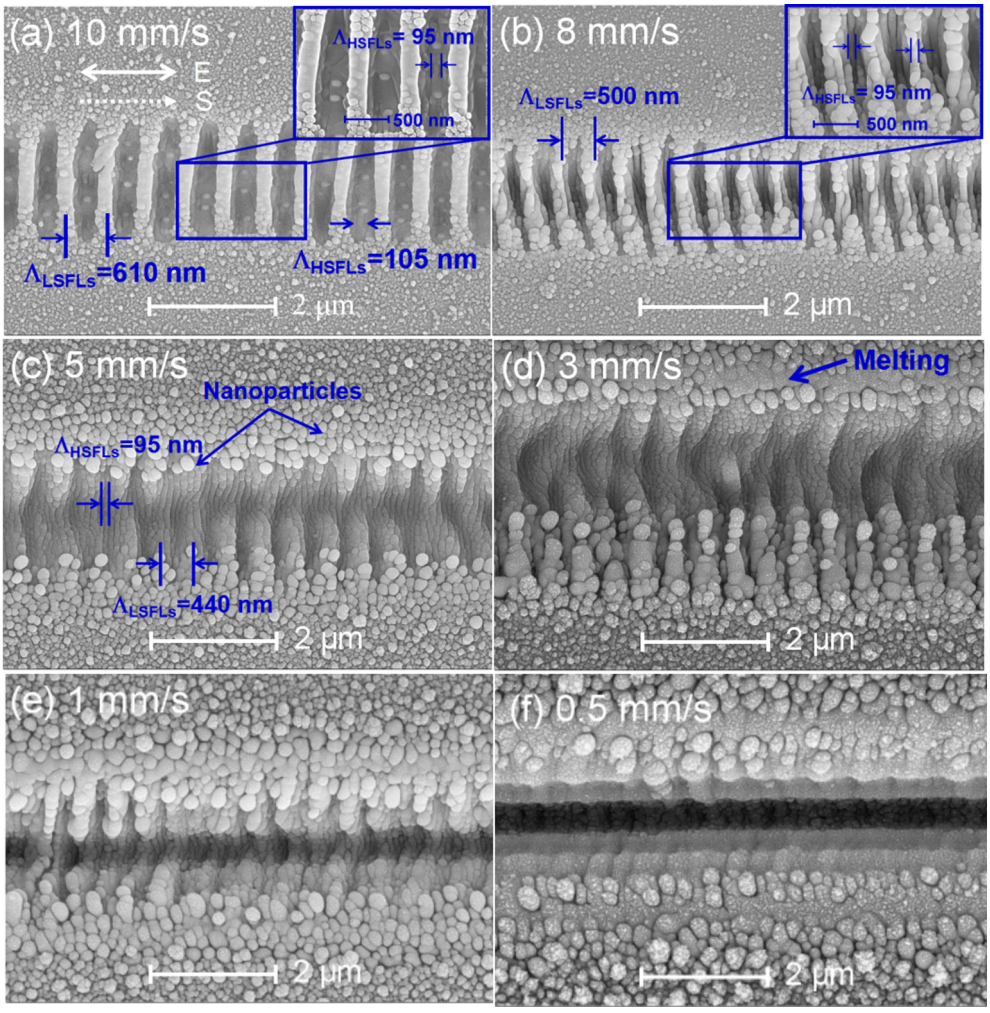

Fig. 1. Formation of LIPSS on germanium surface investigated by different laser scanning speeds and the same pulse energy of 3.3 nJ/pulse. The insets are magnified SEM images. The scanning direction S and laser polarization E are depicted in (a).