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Near-IR resonant response of Ge microparticles fabricated by femtosecond laser printing

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Abstract. Recently introduced femtosecond laser printing approach was applied for the controlled fabrication of single germanium microparticles (MPs). The characterization of MPs was performed by scanning electron microscopy and near-IR optical spectroscopy. The size of obtained MPs is about 1-1.5 μm while their shape is close to spherical. Theoretical calculations of MP scattering cross section were performed on the basis of Mie theory. It is shown that extinction spectra of Ge MPs possess sharp resonances in near IR spectral region, which are identified by means of multipole decomposition analysis.

1. Introduction

Semiconductor and dielectric micro- and nanostructures attract increasing attention of researchers nowadays due to their high potential as a substitution for plasmonic metal-based metamaterials. The latter suffer from intrinsic Ohmic energy losses at optical frequencies, which limit considerably the device efficiency. In contrast, so-called all-dielectric metamaterials are characterized by much lower absorption losses, and, moreover, can exhibit strong Mie-type resonances in their optical response, which is favorable for the realization of the fundamental task of nanophotonics – light manipulation at the nanoscale [1]. Different types of metamaterials have been shown by employing silicon – most widely used material to date [2]. One of the progressive techniques for the fabrication of nearly ideally spherical metal and semiconductor micro- and nanoparticles is known as femtosecond (fs) laser printing [3, 4]. Si nanoparticles with sizes of a few hundred nanometers, produced by this method, demonstrated outstanding optical properties in visible spectral range due to excitation of electric and magnetic dipole resonances [5]. Going beyond silicon, in our most recent paper we further developed the fs laser printing technique and fabricated single spherical Ge and SiGe nanoparticles [6]. Such novel objects are found to possess a crystalline structure as revealed by Raman scattering measurements. Moreover, both Ge and SiGe nanoparticles also exhibit Mie-type resonances in the visible light scattering spectra which make them the promising candidates for the needs of nanophotonics [6].



In present paper we demonstrate the fabrication of single Ge microparticles (MPs) by the fs laser printing method. Optical characterization of obtained MPs in near-IR spectral region is also provided. Germanium is known as a material used in photodetectors for near-IR range due to its appropriate band gap. On the other hand, relatively low absorption losses of Ge in near-IR comparing to visible region allow one to consider Ge MPs as elementary building blocks of all-dielectric metamaterials for use in near-IR range.

2. Experimental details

Thin Ge film with the thickness of 100 nm was used as a donor for MP fabrication by means of fs laser printing approach [6]. The film was deposited on 150 μm -thick standard borosilicate wafer (Borofloat 33, Plan Optik AG) by the e-beam evaporation at room temperature using ultra high vacuum growth system [7]. A commercial femtosecond laser system (Spectra Physics) with 800 nm central wavelength, 50 fs pulse duration, 3 mJ maximum pulse energy and 1 kHz repetition rate was used for fs laser printing of MPs. Laser beam was focused on a transparent substrate with donor Ge thin film by means of 20 mm focal length lens. MPs were transferred on a receiver glass substrate located directly underneath the thin film. The distance between the donor and receiver substrates was equal to about 10 μm . Scattering spectra were collected in forward-scattering geometry using a microscopic setup equipped with CCD camera. Visualization of generated MPs was performed by means of dark-field microscopy. Theoretical calculations of scattering cross sections for ideally spherical Ge MPs were performed using Mie theory, while dispersion relations of real and imaginary parts of complex dielectric permittivity were taken from the respective database [8].

3. Results and discussion

Figure 1 shows dark-field microscopic image of Ge MP arrays printed by 800 nm single fs laser pulses. Bright spots correspond to 3x3 MP subarrays obtained at fixed laser pulse energy, while the latter gradually increased from left to right side of the image. The inset of Figure 1 demonstrates SEM image of a chosen Ge MP, which size is about 1.43 μm and shape is close to spherical. Overall MP size is varied from about 1 to 1.5 μm depending on the energy of employed laser pulses. The crystalline structure of MPs is evident from Raman spectroscopy measurements (not shown here), which revealed a sharp peak at 300 cm^{-1} characteristic for bulk crystalline Ge. The modification of the donor Ge film surface under fs laser treatment is shown in Figure 2. The initial formation and eventual ejection of Ge MP upon high enough pulse energies can be clearly seen. An increase of laser pulse energy was also accompanied by the rise of molten film area from about 3 to 4.5 μm in diameter. The reproducibility of fabricated MPs can be estimated to amount to 90 %. At the same time it is worth noting that the change of the laser wavelength to 400 nm (laser second harmonic) led to the formation of invariably multiple nanoparticles instead of single ones. The latter emphasize the necessity of proper adjustment of experimental parameters to achieve the control over the laser printing approach.

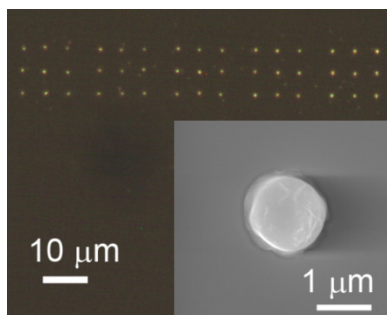


Figure 1. Dark-field microscopic image of Ge MP arrays printed by 800 nm single fs laser pulses. The inset shows SEM image of a chosen Ge MP.

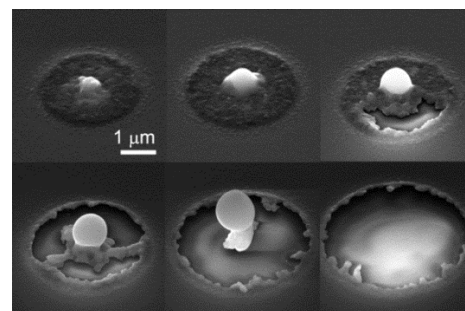


Figure 2. SEM images of the donor Ge film surface irradiated by single 800 nm fs laser pulses with gradually increasing energy. Scale bar is common for all images.

An optical response of fabricated MPs was studied in the size-related near-IR spectral range. Figure 3 demonstrates for comparison the experimental extinction spectrum of Ge MP, which is shown in the inset of Figure 1, and both theoretical scattering and extinction cross sections for equal-sized ideally spherical Ge MP (1430 nm in diameter). While the experimental spectrum is quite noisy due to the lack of the signal, it reproduces well in general the corresponding theoretical dependence. An absence of total agreement may be also explained by the deviation of MP shape from the ideal sphere. Multipole decomposition analysis of the scattering cross section revealed the dominance of magnetic-type contributions (dipole and quadrupole) responsible for the appearance of sharp peaks in the spectrum. The demonstrated resonant behavior of fabricated Ge MP optical response may be used both in photonics and IR sensorics.

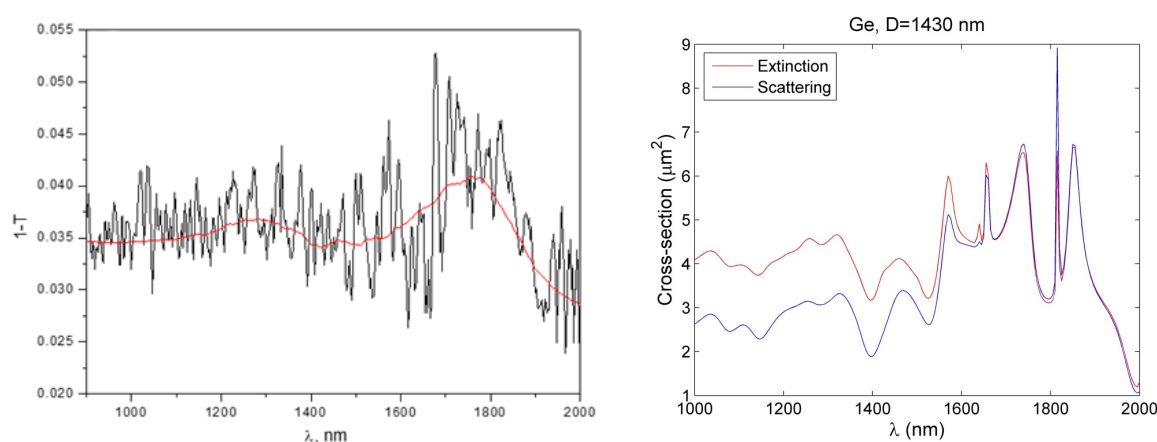


Figure 3. (Left) Experimental extinction spectrum of fabricated Ge MP (shown in the inset of Figure 1) and (right) theoretical scattering and extinction cross sections for equal-sized ideally spherical Ge MP.

4. Conclusions

In conclusion, we realized the reproducible fs laser printing of single Ge microparticles, which were characterized by SEM and visualized by dark-field microscopy. Near-IR optical response of fabricated Ge MPs was studied both experimentally and theoretically with a satisfactory agreement showing the dominance of magnetic-type multipole contributions.

Acknowledgments

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