Investigation of plasmonic response of metal nanoparticles to ultrashort laser pulses

Submitted by

Dmitry Konstantinovich Polyushkin

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I certify that all material in this thesis which is not my own work has been identified and that no material has previously been submitted and approved for the award of a degree by this or any other university.

I lovingly dedicate this thesis to all my family, especially to my wonderful wife Yulia, who fills my life with joy and happiness; to my father Konstantin, mother Irina and sister Daria who have always provided me with their unconditional love and support through the years; to my grandparents Gennady and Nina and my aunt Ekaterina who with my parents have brought me up in love and care; and in memory of my dear grandparents Ivan and Nadezhda.

Abstract

In this thesis the interaction of ultrashort laser pulses with metal nanostructures is investigated via two different phenomena: coherent acoustic oscillations of nanoparticles and generation of THz pulses on metal surfaces.

Both of these effects rely on the collective oscillations of free conduction electrons in metal surfaces, plasmons. The field of plasmonics gained a great interest in the last twenty years due to the unique properties of these surface modes. It is the effects of the resonant response of plasmonic structures to incident electromagnetic wave, in particular, in visible and infrared bands and the concentration of the electromagnetic field in small subwavelength regions with significant enhancement of the incident field that make plasmonics so attractive for various applications, such as biochemical sensing, enhanced fluorescence, surface-enhanced Raman scattering, and second harmonic generation, amongst others.

Investigation of the coherent particle vibrations is performed using the pump-probe technique which allows measurement of the transient transmission signals. The expansion and subsequent contraction of the nanoparticle following the ultrashort laser pulse excitation lead to a shift of the plasmon band which can be traced by transient spectroscopy. We have investigated the effect of the particle thickness on the frequency of the fundamental vibrational mode. In addition, we measured the vibrational particle response during the particle shape deformation, both symmetrical and asymmetrical.

Exploration of the THz generation phenomena on plasmonic structures was performed using THz time-domain spectroscopy, the method which allows tracing of the generated THz field in the time-domain. We were able for the first time to measure the THz pulses generated from arrays of metal nanoparticles. Our observations verify the role of the particle plasmon mode in the generation of THz pulses. In addition, by exploring the dependence of the THz emission on the femtosecond pulse intensity we showed a high nonlinearity in the THz generation mechanism. The experimental results were assessed in the context of a recently proposed model where the THz radiation is generated via the acceleration of the ejected electrons by ponderomotive forces.

To reveal another proposed mechanism of the THz generation from plasmonic structures, namely optical rectification, we investigated the THz generation and electron emission from the arrays of nanoparticles and nanoholes. Our results suggest that both mechanisms may contribute to generation of THz pulses from the same sample under different illumination conditions.

In addition to periodic arrays of nanoparticles and nanoholes, THz generation from random metal-dielectric films was investigated. The microstructuring of such films allowed selective THz frequency generation which was explained by a model of dipole THz emitters. In addition, the effects of low temperature and pressure on the THz generation efficiency were investigated.

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