Spaser Spectroscopy with Subwavelength Spatial Resolution

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Abstrasct

The advances of nanotechnology open an opportunity of development of existent spectroscopy methods. We suggest a novel method that combines the ideas of tip-enhanced optical spectroscopy and of laser spectroscopy. The latter, offering no spatial resolution, provides an extremely high sensitivity and reveals even prohibited (non-dipolar) transitions. The combination is enabled by invention of plasmonic laser (spaser) [1], which generates plasmons due to nonradiative energy transfer from the gain medium (quantum dot) to SPP, localized at a nanoparticle. Recently we have suggested a spaser radiating 1D plasmons [2]. The device suggested here is based on 1D spaser generating plasmons on a needle with narrow tip. We show that the sensitivity allows detection of some dozens of atoms with high (sub-wavelength) spatial resolution.

1. Introduction

Recent development of electrodynamics is largely governed by a tendency to expansion of the application range of optical devices to the fields where the electronic, magnetic, X-ray and other devices were of conventional use. The examples are future optical computers, optical memory, optical lithography of high (subwavelength) spatial resolution, as long as scanning near-field optical microscopy (SNOM). It is the use of near field that makes possible exceeding the limit of $\lambda/2$, restricting the lower bound for the size of optical devices and for the optical microscope resolving power. Most promising are the systems including metal and thus supporting the near-field excitations – surface plasmon-polaritons (SPPs). SPPs are used in SNOM, where the SPP is excited by incident laser beam. The scattering of SPP by an analyzed sample creates a far field, intensity (and in some schemes phase) are measured and used for the image retrieval.

Even greater achievement is Tip-Enhanced Optical Spectroscopy (TEOS), exploiting the principles of SNOM but analyzing the spectral response of the sample. Thus, both image and the composition of the sample are obtained. The most popular is the method based on the Raman spectroscopy – Tip-Enhanced Raman Spectroscopy (TERS), taking advance of large field intensity in SPP for even larger (square) enhancement of the nonlinear effect.

ISBN 978-952-67611-2-1

In the current paper we suggest a novel method that combines the ideas of TEOS and of laser spectroscopy. The latter, offering no spatial resolution, provides an extremely high sensitivity and reveals even prohibited (non-dipolar) transitions. The combination is enabled by invention of plasmonic laser (spaser), which generates plasmons due to nonradiative energy transfer from the gain medium (quantum dot) to SPP, localized at a nanoparticle. The high field intensity favors high sensitivity even for linear methods, nothing to say of TERS. The advantage of a new method over the standard laser spectroscopy, besides the spatial resolution, is the weakness of nano-sized spaser compared to the macroscopic laser, which permits easier suppression of lasing by the analyzed sample and, thus, increases the sensitivity.

2. Principal Scheme

A principal scheme of the spaser spectroscope is shown in Fig. 1. The needle geometry typical of the near-field devices supports plasmon, which is closed between the needle tip and a Bragg reflector, formed at the needle surface. The resonant plasmonic mode of the obtained cavity is amplified by a set of quantum dots deposited onto the needle or by any other gain medium. Thereby, a 1D spaser is formed. The field concentrated near the tip interacts effectively with analyzed sample. The field localization assures spatial resolution of the order of the tip curvature radius. The radiation tunneling through the Bragg reflector on its way to a spectral analyzer may be additionally amplified by quantum dots. The spectrum of the sample is displayed as a set of relatively narrow dips in the spectral line of spaser.

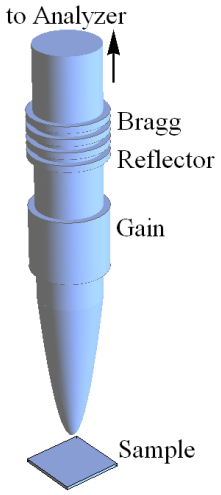

Fig. 1. Geometry of Spaser Spectroscope.

3. Spaser analysis

The laser dynamics is described by Maxwell-Bloch equations, which follow from the Maxwell equations and from the equations for the density matrix of the gain medium atoms. To avoid the solution of the system of PDEs, one conventionally uses the single-mode approximation, representing the electric field as a product of a time amplitude and a cavity mode $\mathbf{E}(\mathbf{r})$. In our case the latter is the field distribution in the plasmon of needle, restricted by the tip and the Bragg reflector. Further simplification of the equations is attained by the slow amplitudes approximation, measuring all the frequencies from that of the cavity mode and neglecting the second derivatives of the amplitudes. This gives the system of ODEs for the field amplitude a, polarization of the gain medium σ and population inversion D:

$$\begin{aligned}
\dot{a} &= -a/\tau_a - i\Omega\sigma - i\Omega_S\sigma_S + F(t),\\
\dot{\sigma} &= (i\delta - 1/T_2)\sigma + i\Omega aD,\\
\dot{D} &= -(D - D_0)/T_1 + 2i\Omega(a^*\sigma - \sigma^* a),\\
\dot{\sigma}_S &= (i\delta_S - 1/T_{2S})\sigma_S + i\Omega_S aD_S,\\
\dot{D}_S &= -(D_S - D_{0S})/T_{1S} + 2i\Omega_S(a^*\sigma_S - \sigma_S^* a).
\end{aligned} \qquad (1)$$

ISBN 978-952-67611-2-1

4. Conclusions

A convenient way of the lasing spectrum calculation is the use of the Wiener-Khinchin theorem $S(\omega) = (2\pi)^{-1} \int_{-\infty}^{\infty} C(t) \exp(i\omega t)\, dt$. The result is shown by red line in Fig. 2. The sample changes generation spectrum. At weak interaction (the needle is far from the sample, i.e., at small values of Ω_S) the dip is also weak (green line in Fig. 2). The growth of interaction increases the dip (blue line in Fig. 2). But finally the dip broadens (black line in Fig. 2), which leads to the decrease of its depth and of the spectral resolution. Thus, an optimal position of needle exists, which may be found from maximum of the dip depth

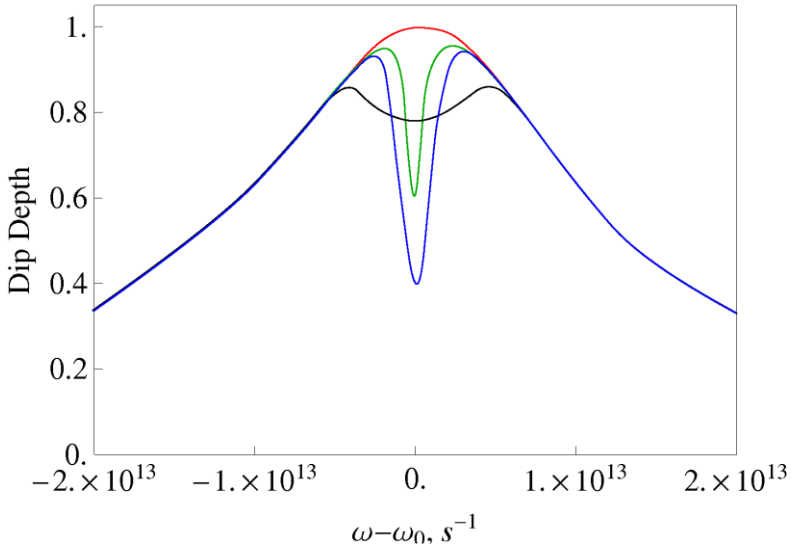

Fig. 2. The lasing spectrum of spaser (black line) and its change by interaction with sample, so that the red, green and blue lines correspond to the values of interaction constant Ω_S of $0.5 \cdot 10^{12}$, $1.7 \cdot 10^{12}$ and $5 \cdot 10^{12}$ s^{-1}, respectively.

The calculations show that the device easily detects some dozens of atoms.

References

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