Non-exponential decay of dark localized surface plasmons

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Abstract: It is shown that the decay of the weakly coupled to radiation (dark) modes of subwavelength plasmonic nanostructures is strongly nonexponential. Their lifetime is overestimated by conventional exponential relaxation time obtained in the standard Markovian approximation. These effects are manifestations of the strong dispersion and near-field feedback. The developed theoretical framework introduces an ensemble of local relaxation degrees of freedom coupled to plasmonic mode in order to describe its decay due to material losses. The macroscopic description of the decay process leads to the specific memory function of the system, evaluated from the modal and material dispersions of the plasmonic nanostructure. Proper knowledge of the relaxation behavior is vital for various applications relying on light-matter interactions of emitters with nanoscale objects, such as fluorescence manipulation, bio-imaging, sensing, spasers, sub-diffraction optics, Raman scattering, and quantum optics.

OCIS codes: (240.6680) Surface plasmons; (270.5580) Quantum electrodynamics.

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1. Introduction

Electromagnetic cavities are extensively used for manipulation of light-matter interactions since the original work of Purcell [1], where the environment was shown to provide significant modification of spontaneous emission rates at radio frequencies. In the optical range, various systems, such as photonic crystals [2], pillars [3], toroidal microcavities [4], metal nano-particles [5], and metamaterials [6–9] are used for different applications, where manipulation of light emission is required. The recent progress in both fabrication and modeling enabled design of metallic nanometric cavities with precisely controlled optical properties, opening new prospects for interesting practical applications, including sensing [10, 11], optical trapping [12], cancer imaging and therapy [13], plasmonic lasers [14–16], spasers [17] and other active plasmonic components [18], enhanced nonlinearities [19–21], control of emission properties [22], transformation optics [23], and many others.

Metal nanoparticles, having negative permittivity at certain frequencies, can support excitations, called localized surface plasmon resonances, even if structures are of subwavelength dimensions [24]. The eigenmodes of nanostructures may be estimated using different tools, such as hybridization method [25], surface integrals [26] or other numerical techniques. The eigen frequencies (resonances) may be tuned using particle-particle coupling [27], particle elongation [28], concavity tuning [29], and ultimately by evolutionary methods [30], providing possibility to achieve on-demand spectrum of resonance modes. Here we distinguish between bright modes of plasmonic nanoparticles with strong dipolar moment, hence efficiently radiating, and higher-order dark modes, weakly or not at all radiating into the far-field. While bright plasmonic modes may be used for the enhancement of radiation efficiency of emitters, using the so-called optical antenna configurations, dark modes have been proposed for trapping of light signals and for guiding along chains of particles with suppressed far-field losses [31]. Dark modes, not being coupled to the far-field radiation, can be excited only in the near-field by closely situated emitters [32] or directly using electron beam impact [33].

The knowledge of the relaxation behavior of plasmonic dark modes is of potential interest for both fundamental understanding of the process as well as for various above discussed applications, where lifetime estimations may limit or improve predictions on possible performances. To the best of our knowledge, the decay law of the dark mode decay is generally assumed to be exponential [34] and, in theory, determined by material losses if the far-filed radiation is neglected. However, any physical system with an energy spectrum, bounded from below, could not decay to its ground state obeying exponential law [35], and localized surface plasmons, having discrete and bound spectrum, are not uncommon. The exponential decay law of a physical system relies on the assumption (also called the Markov approximation) that the system has no memory and its next state is only depends on the present and not on the past. The deviations from exponential laws were studied mostly for decays of quantum systems, such as hydrogen and more generally two-level atoms [36, 37].

Here we develop a theoretical framework to account for memory effects in plasmonic dark mode decay. We show that the steep dispersion of the nano-cavity eigenmodes together with strong near-field feedback lead to considerable deviations from generally assumed conventional exponential decay law with up to 40% changes in the relaxation time.

2. Exponential versus non-exponential decay laws

The relaxation time of an open cavity is determined by a competition between the far-field radiative losses and the material losses in its components. While dipolar (‘bright’) plasmonic resonances predominantly decay by coupling to the far-field radiation (apart from very small particles), the lifetime of higher-order dark modes, e.g., quadrupole mode with the field distribution for a spherical nanoparticle shown in Fig. 1, is determined by material losses. The decay rate of a dark cavity mode can be written in general case as [38–41]

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$$\Gamma_C = 2\omega_0 \frac{\int dV \varepsilon_0 \varepsilon_{im} \left|\vec{E}\left(\vec{r},\omega_0\right)\right|^2}{U_{tot}}, \tag{1}$$

where ω_0 is the resonant frequency, $\vec{E}\left(\vec{r},\omega_0\right)$ is the associated electric field, U_{tot} is the total energy in the cavity, ε_0 is the vacuum permittivity, and $\varepsilon = \varepsilon_r + i\varepsilon_{im}$ is the complex, frequency dependent relative permittivity. The intensity decay coefficient Γ_C for the quadrupole resonance of a plasmonic sphere can be analytically calculated in the quasistatic approximation to be

$$\Gamma_C = \frac{41\omega_p^2 \gamma \omega_0^2}{\left(\omega_0^2+\gamma^2\right)^2 \cdot \left[\frac{41}{2}\left(1 - \frac{\omega_p^2}{\left(\omega_0^2+\gamma^2\right)} + \frac{2\omega_p^2\omega_0^2}{\left(\omega_0^2+\gamma^2\right)^2}\right) + \frac{189}{8}\varepsilon_d\right]}, \tag{2}$$

where ω_p is the plasma frequency of conduction band electrons, γ is the electron collision rate determining the material (Ohmic) losses, ε_d is the permittivity of the embedding medium, and the Drude model for the metal permittivity was used with $\varepsilon = 1 - \frac{\omega_p^2}{\omega\left(\omega + i\gamma\right)}$. These analytical expressions for the both decay rate and the material dispersion are used here for convenience, while any other numerically or experimentally evaluated data may be employed as well. The conventional decay rate (Eqs. (1, 2 corresponds to the exponential time dependence of the intensity: $I(t) = I(0)\exp\left(-\Gamma_C t\right)$, if the mode is not overdamped. However, this description neglects the memory effects of the system, originating from the steep modal and material dispersion near the resonant frequency and, as will be shown below, significantly overestimates the lifetime of plasmonic dark modes.

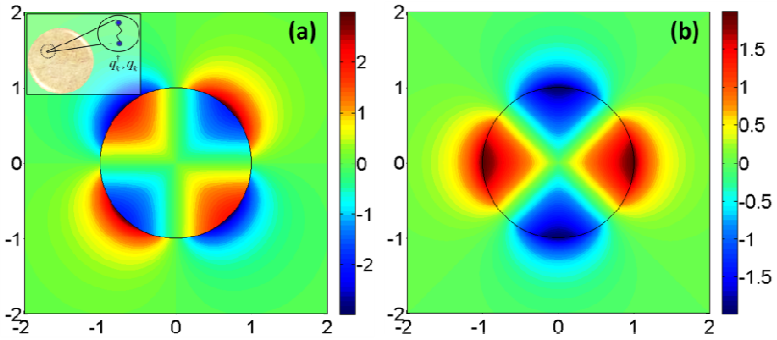

Fig. 1. The electric field distribution associated with a quadrupolar mode of a spherical nanoparticle in the quasistatic approximation. Color maps of (a) radial and (b) azimuthal electric field components. The quadrupole resonance condition is $\varepsilon(\omega) = -1.5\varepsilon_d$. All distances are normalized to the sphere radius. Color scale represents the electric field amplitude in arbitrary units. Inset in (a) illustrates the assembly of local oscillators acting as the damping reservoir; $q_k^\dagger, q_k$ are the creation and annihilation operators corresponding to k^{th} oscillator.

3. Theoretical model of the non-exponential decay law

In the presented model, the memory function is introduced by the cavity mode coupling to the ensemble of local oscillators (inset in Fig. 1(a)), accounting for the material losses. In this treatment, the spatial shape of the cavity mode is evaluated by solving classical Maxwell's equations for lossless case, and the overall stored energy is calculated by the Brillouin formula [40, 41], taking into account the dispersion of the real part of a material permittivity.

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This approach is known in quantum mechanical quantization schemes as 'modes-of-the-universe' or 'mode decomposition' approach and, in principle, may simultaneously include losses and dispersion by introducing an ensemble of local oscillators ([42] and references therein). Here, we separate the loss channel from the dispersion behavior of the real part of the permittivity, maintaining the Kramers–Kronig relation between them. This approach provides direct access to the loss channels without extra need to treat an additional coupling with the real part of the dispersion and solve complex equations for polariton modes of the system. In our model, the particle is divided into a set of infinitesimal volumes, each one contains the absorption degree of freedom represented by a local oscillator. The infinite ensemble of such oscillators will act as the reservoir, which is treated within the Heisnberg-Langevin approach for quantum theory of decay [43]. The Hamiltonian of the system, separated into the energy of the free field and the reservoir modes, and the field-reservoir interaction part, is given by

$$\begin{aligned} &\hat{\mathrm{H}} = \hat{\mathrm{H}}_0 + \hat{\mathrm{H}}_I \\ &\hat{\mathrm{H}}_0 = \hbar\omega_0 a^\dagger a + \sum_q \hbar\omega_k q_k^\dagger q_k ; \hat{\mathrm{H}}_I = \hbar\sum_k g_k \left(q_k^\dagger a + a^\dagger q_k\right), \end{aligned} \tag{3}$$

where $a^\dagger(a)$ and $q_k^\dagger(q_k)$ are the creation (annihilation) operators of the cavity and reservoir modes, respectively, and g_k is the constant describing the coupling between the field and the reservoir. The formal solution of the equation of motion for the operators, averaged over the reservoir modes ($\langle ... \rangle_R$), is given by the following integral equation:

$$\begin{aligned} &\tilde{a}(t) = \langle a(t) \rangle_R e^{i\omega_0 t} \\ &\dot{\tilde{a}} = -\sum_k \left(g_k^2 \cdot \int_0^t dt' \tilde{a}(t') e^{i(\omega_0 - \omega_k)(t-t')} \right). \end{aligned} \tag{4}$$

The conventional decay rate corresponds to the solution of Eq. (4) under the Markovian and Weisskopf-Wigner [43] approximation (memory-less exponential process). It may be shown also, that $g_k^{\,2} \sim \varepsilon_{im}$. Thus, the final equation for the time evolution of the field amplitude which takes into account the memory effects is given by

$$\dot{\tilde{a}} = -\int_0^t F(t-\tau)\tilde{a}(\tau)d\tau, \tag{5}$$

where

$$F(t) = \frac{\Gamma_C}{2\pi}\int_0^\infty \left(\frac{\omega_0^2 + \gamma^2}{\omega^2 + \gamma^2} \right) \cdot \frac{\omega_0}{\omega} e^{i(\omega_0 - \omega)t} d\omega \ . \tag{6}$$

The memory function $F(t)$ is responsible for the time evolution of the field operators. It should be noted, that neither derivations above nor the subsequent analytical solution are relying on the slow variation in time approximation of $\tilde{a}(t)$ in Eq. (4), employed in the Weisskopf-Wigner approach.

The solution of Eqs. (5) and 6 may be written in the close integral form using the Laplace transformation and the convolution property:

$$a(t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \frac{a(0)}{is + \frac{\Gamma_C}{2}\left(\frac{\omega_0^2 + \gamma^2}{(\omega_0 - s)^2 + \gamma^2} \right) \cdot \frac{\omega_0}{(\omega_0 - s)}} e^{ist} ds \ . \tag{7}$$

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The asymptotic behavior of the solution can be found by approximating the zero-order of the integrand by Lorenzian function. This will result in an exponential decay law. However, the higher-order contributions of the denominator's Taylor expansion are adding the oscillatory factor to the exponential decay.

4. Discussion

Assuming macroscopic number of cavity photons, quantum operators may be replaced by classical amplitudes. Figure 2 (a) shows the time evolution of the mode power for two different material loss parameters. In both cases $\varepsilon_r = -1.5$ was used, corresponding to the quadrupole resonance of the sphere (~350 nm wavelength for Ag) and material losses were varied with $\varepsilon_{im} = 0.5$ and $\varepsilon_{im} = 0.1$. As was mentioned above, plasmonic resonances may be tuned by various techniques, and the desired dark modes may be shifted into the infrared part of the spectrum, where relative material losses are smaller. The numerical results (Eq. (7) deviate from the conventional theory. In both cases, nonexponetinal decay has been observed, as was qualitatively predicted in the discussion after Eq. (7). The deviations are more pronounced with the increasing Ohmic losses. For large losses, the lifetime of the dark mode is overestimated by the conventional model by about 40%. To emphasize the deviation from the exponential law, we plotted the log-scale ratio of the numerical result and the exponential law: on this graph the horizontal line y = 0 represents correspondence with the conventional theory (Fig. 2 (b)). As can be seen, pronounced oscillatory behavior is present, manifesting that the decay is not purely exponential.

Bright plasmonic modes may be treated in the similar fashion, distinguishing between radiative and Ohmic losses. Absorption, scattering and extinction cross-sections of silver spheres (material parameters from [44]) were evaluated using the Mie theory and are depicted in Fig. 3 (a). The extinction of the 50 nm radius sphere is dominated by scattering, while for the 20 nm radius the absorption is the main loss channel. For silver spherical nanoparticles with the radius bigger than 30 nm radiation losses can be considered predominant [39].

The decay of bright modes still may be described by Eq. (3) with the local oscillators accounting for material losses are replaced by the free-space radiation modes. The coupling constant, resulting from the interaction Hamiltonian $\hat{\mathrm{H}}_I = -\vec{d}\cdot\vec{E}$, is given by $g_k{}^2 = \frac{\omega_0}{2\hbar\varepsilon_0 V}|d|^2\cos^2\theta$ [43], here V is the quantization volume and $d = \frac{12\pi\varepsilon_0\varepsilon_d\hbar R^3}{\partial\,\mathrm{Re}(\varepsilon)/\partial\omega_{\omega=\omega_0}}$ is the effective plasmonic dipole moment [39] with R being the sphere radius and θ inclination angle in the spherical coordinate system. The equation of motion for the dipolar plasmonic mode annihilation operator (a_d), similar to Eq. (5), is then $\dot{a}_d(t) = -\frac{\sqrt{\varepsilon_d^3}R^3\left(\omega_0^2+\gamma^2\right)^2}{\pi c^3\omega_p^2\omega_0}\int_0^t d\tau\, a_d(\tau)\int_0^\infty d\omega\cdot\omega^3 e^{i(\omega_0-\omega)(t-\tau)}$. In this equation, however, the last integral is undefined, meaning that more complicated integration contours for the Laplace transformation [36] or step-by-step solution, approximating the derivative by finite differences [37] should be used. We employed the latter method for the treatment of the dipolar mode decay behavior. The results are depicted on Fig. 3(b) showing that the numerical solution (blue line) is very close to the conventional Weisskopf-Wigner approach (red-dashed line) and the memory effects are not significant in radiative decay of bright plasmonic modes.

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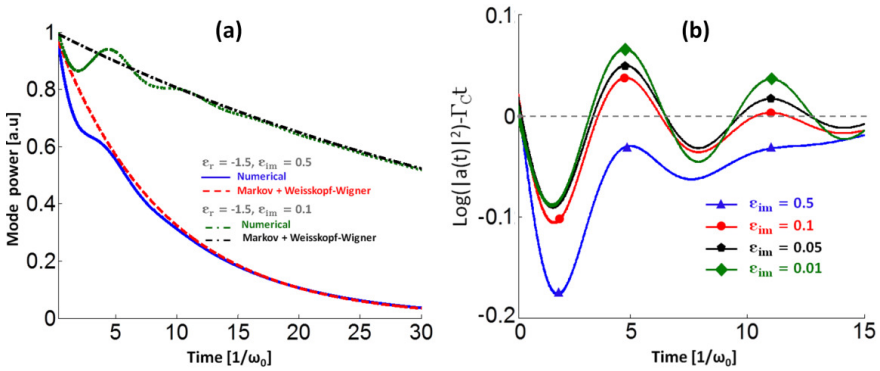

Fig. 2. (a) The time evolution of the plasmonic quadrupole mode's power for different loss parameters in the quasistatic regime. Blue (red) and green (black) lines correspond to the memory-effect-based model (conventional exponential decay) for $\varepsilon = -1.5 + 0.5i$ and $\varepsilon = -1.5 + 0.1i$, respectively. (b) Deviation of the decay process from the exponential law $[\log(|a(t)|^2) - \Gamma_c \cdot t]$: 4 curves correspond to different material loss parameters $\varepsilon_{im} = [0.5; 0.1; 0.05; 0.01]$. Horizontal dashed line y = 0 corresponds to the exponential decay.

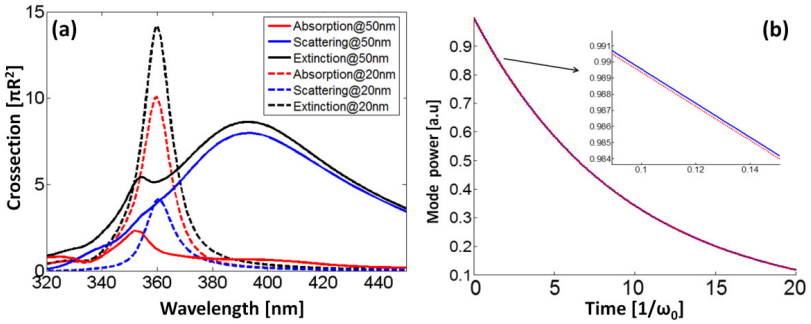

Fig. 3. (a) Absorption (red), scattering (blue) and extinction (black) cross-sections for the silver particles with the radii of 50 nm (solid lines) and 20 nm (dashed lines) and permittivity from [44]. (b) The time evolution of the dipolar plasmonic mode's power: red line corresponds to the standard Weisskopf-Wigner theory; blue line is the full numerical solution using the memory equation. Insert shows the zoomed area of the same curve.

5. Conclusion

We have developed a theoretical framework to describe the decay of dark plasmonic modes. The introduction of the local degrees of freedom coupled to the plasmonic mode results in the macroscopic description of the process in terms of the memory function, evaluated from the modal and material dispersions and strong near-field feedback. The dark mode decay is strongly nonexponential if the Ohmic losses are strong. The standard exponential decay law has been shown to overestimate the lifetime by up to 40%, suggesting that the proposed model should be considered to obtain realistic estimation of performances of plasmonic devices based on dark modes. The developed formalism may be strictly applied for any open cavity (not necessarily plasmonic) where radiative leakage may be treated in the same way as material losses. In particular, dipolar plasmonic resonances, decaying both radiatively and nonradiatively may be described in the same fashion paying additional attention to the competition between different decay channels. Interestingly, other quantum effects in plasmonics, associated with peculiarities of material responses, have recently been shown to provide unusual metastable states [45] and to correct overestimated by standard theory predictions on the near-field enhancements [46].

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The memory effects may also substantially influence the spasing phenomenon. The spasing into the dark modes, yet experimentally demonstrated, is very promising, since it may directly generate strongly confined evanescent modes with almost no leakage in the far-field radiation. The rate equations, conventionally describing the spaser action [17] depends only on the actual time, neglecting the memory effects. The latter effects may lead to interesting and important for spasing action phenomena, similar to the effects of the memory of atoms in laser systems which was shown to reduce the laser linewidth below the usual Schawlow-Townes limit [47].

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