Nano-stratification of local field and atomic bistability in low-dimensional structures

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Abstract: Nano-scale strata due to broken uniformity of local field in strongly self-interacting low-dimensional structures are predicted. They result in giant field resonances and atomic optical bistability, including ultimate case of two coupled atoms. ©2008 Optical Society of America

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Optical properties of sufficiently dense materials are greatly affected by the near-field interactions between neighboring atoms, the best known manifestation of which is the local vs incident field phenomenon and related Lorentz–Lorenz or Clausius–Mossotti relations. In most theories of such interactions a standard (and often implicit) assumption is that the local field and polarization are uniform in near neighborhood of each participating atom at least at the distances shorter than the light wavelength, $\lambda$.

We discovered that if the local uniformity is not presumed, the system of interacting atoms may exhibit spatial variations of the local field resulting in sub-$\lambda$ strata with a nano-scale period. Because of angular averaging, the 3D interaction significantly inhibits the effect of angular anisotropy of near-field dipole interaction, so the best candidates to explore the new phenomenon are low-dimensional systems. The most revealing is the case of a 1D string of particles (such as, e.g., a string of quantum dots, long molecules, carbon nano-tubes, etc). In these systems the nano-strata are strongly anisotropic with respect to the laser polarization and sensitive to the sign of the interaction, especially in near vicinity of an atomic resonance. In a string of particles, if the laser is linearly polarized, one can distinguish between two drastically different cases, that can be described as co-interaction and counter-interaction. The nearby particles co-interact if they have positive sign of interaction and the field is normal to the string (or negative sign of interaction and the field is parallel to the string), whereas counter-interaction happens at reverse combination of these parameters. In the co-interaction mode, the nano-strata are of a much longer spatial period than in the counter-interaction mode. However, the most striking manifestation of the anisotropy is huge resonant enhancement of the strata in the co-interaction case, that is due to the eigen-modes of the string of finite length (Fig. 1). These resonances are absent in the case of counter-interaction. Nano-strata emerge if a certain critical condition is met on the strength of interaction and particle density. These nano-strata are expected to be most pronounced near atomic resonances with enhanced dipole momenta, e.g. in quantum dots, alkali vapors at certain densities, etc. Because of huge local field enhancement, the nonlinearity due to atomic resonance may result in strongly pronounced giant hysteresises and optical bistability in the string of even a few atoms (including the ultimate case of 2-atoms pair); the incident intensity required for that could be much lower than the saturation intensity.

The local field $E_L(\vec{r})$ acting upon a 2-level atom at the point $\vec{r}$ in the presence of (a) an external incident field with the amplitude $E_{\text{in}}$ and frequency $\omega$, and (b) the near-field of surrounding atoms regarded as dipoles is

$$E_L(\vec{r}) = E_{\text{in}}(\vec{r}) - \frac{\mu_0}{\varepsilon_0 \Delta \omega_{\text{res}}} \sum_{\vec{r}'} \frac{1}{|\vec{r} - \vec{r}'|^3} \frac{3\langle \vec{\mu} \cdot \vec{E}_L(\vec{r}') \rangle}{1 + \frac{3}{\delta^2} + \left(\frac{\langle \vec{E}_L(\vec{r}') \rangle^2}{\Delta \omega_{\text{sat}}}\right)},$$

where $\vec{\mu}$ is a unit vector in the direction of $\vec{r} - \vec{r}'$, $\varepsilon$ is a background dielectric constant, $\vec{d}_0$ is an atomic dipole moment; $\Delta \omega_{\text{res}}$ is an atomic resonance linewidth; $\delta = (\omega - \omega_0)/\Delta \omega_{\text{res}}$ is laser detuning from the atomic frequency $\omega_0$, $\vec{r}$ is the position of a nearby atom; $\Delta \omega_{\text{sat}} = h^2 \Delta \omega_{\text{res}}/\mu_0^2 \tau$ is the saturation intensity, and $\tau$ is the life-time of excited atom; the summation in (1) is done over all the atoms. Eqn. (1) is very general and also reflects nonlinear atomic saturation.

We introduce the strength of interaction between neighboring atoms as

$$Q = -A(\rho) \frac{2 \alpha \lambda_0 (\mu_0/\varepsilon)^2}{\pi l_0^2 (\delta + i) \Delta \omega_{\text{res}}},$$

where $\alpha = 1/137$ is the fine-structure constant, $\lambda_0$ is the laser wavelength, $l_0$ is the spacing between atoms, and $A(\rho)$ is an anisotropy factor due to the dipole angular diagram. In 1D case $A(\rho) = 1$ for the laser field parallel to the string, and $A(\rho) = -1/2$ for the field normal to the string. We found that once $Q$ exceeds critical value, $|Q| > Q_c = O(1)$, the uniformity of the local field [i.e. $E_L(\vec{r}) = \text{const}$ if $E_{\text{in}}(\vec{r}) = \text{const}$ within distances $\sim \lambda$] can be broken by boundaries,
impurities, vacancies in the crystal structure, etc. Many physical systems can satisfy this condition; even gases or vapors such as Na can satisfy it at a pressure below 1 atm. For a 1D-string, $Q_{cr} \approx 1$ in the near-neighbor approximation and $Q_{cr} \approx 1.2$ for the full sum (1).

The co-interaction mode corresponds to $\text{Re}(Q) > 0$, whereby the system exhibits long-period and large-amplitude strata with the wave-numbers $k_{long} \ll l_a^{-1}$, while $\text{Re}(Q) < 0$ — to counter-interaction, $k_{short} = O(l_a^{-1})$. These wave-numbers are determined by the relationship $QG(k_p l_a) = 1$, where $G(\xi)$ is a periodic function of $\xi$ with the period of $2\pi$ [$G(\xi) = \cos(\xi)$ in the near-neighbor approximation]. Fig. 1 depicts the local field variations from atom to atom in the finite string of the size $N = 512$ subjected to an incident field with a semi-sinusoid profile (having a peak at a boundary). The co-interaction mode exhibits huge enhancement of the local field vs the incident field; the intensity enhancement can be as large as $(\omega/\Delta \omega_{res})^2$. The enhancement peaks at the string-size resonances at $k_{long}l_a \approx \pi/(N+1)$; they are clearly seen in the plot of strata amplitude vs the strength $Q$ for fixed $N = 512$ in Fig. 2. Under sufficiently strong driving (but still much below saturation), the system can exhibit large “tsunami”-like hysteresis and optical bistability due to nonlinearity, see Fig. 3 depicting the intensity of the local field vs detuning $\delta$ for various intensities of the incident field at the main resonance.

Most of the above features are preserved down to the ultimate and most basic case of a two-atom system, exhibiting giant enhancement at the main resonance, hysteresises and bistability both for co- and counter-interaction. In the latter case it is a split-fork bistability (the polarization and local field are suppressed in one atom and enhanced in the other one), see Fig. 4. These effects indicate great potential of the new phenomenon for molecular and atomic computers. The detailed consideration of 2D nano-strata are presented in our other paper at this conference [1].

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