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Radiation of Dynamic Toroidal Moments

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**Figure S1.** Evolution of the simulated electric field \( E_z \) and magnetic field \( H_z \) of the T2 mode at 2.5 eV from 0 to \( \pi \) at \( \pi/4 \) intervals. Grey circles denote the nanoholes. The impact locations of the electron probe are indicated by the white dots. Red arrows show the directions of magnetic dipoles. Scale bars are 200 nm.

Figure S1 gives a glance of the field evolution of the T2 mode over half a harmonic period of \( \pi \), where the full harmonic period of the T2 mode takes 2\( \pi \). In general, the magnetic field \( H_z \) (red curve) has a time shift of \( \pi/2 \) with respect to the electric field \( E_z \) (black curve) due to the dynamic nature of electromagnetism. At the beginning of the period (i.e. 0), a loop of magnetic dipoles is only observed in the lower 4 nanoholes (\( H_z \) field at the bottom in Figure S1), and the corresponding electric field is mostly concentrated in the silver bridge between the lower 4 nanoholes. Hence, only one toroidal dipole is shown. After a time of \( \pi/4 \), an additional loop of magnetic dipoles appears in the upper 4 nanoholes, which assembles another toroidal dipole. Together with the toroidal dipole excited in the lower 4 holes, they now form an antiparallel pair of toroidal dipoles as already presented in Figure 2a at 2.5 eV). At the time of \( \pi/2 \), only the toroidal dipole in the upper 4 nanoholes is dominant (\( H_z \) field in Figure S1). Therefore, these two constituent toroidal dipoles appear one after another in the time domain with a shift of \( \pi/2 \).

Indeed, electric and magnetic fields are synchronous in free space. This can be understood by simply applying the following Maxwell equation:
\[ \nabla \times \vec{H} = \frac{\partial D}{\partial t} \Rightarrow -i\vec{k} \times \vec{H} = i\omega \varepsilon_0 \vec{E} \Rightarrow \vec{E} = -\frac{1}{\omega \varepsilon_0} \vec{k} \times \vec{H} \] (1)

where \( \vec{k} \) is the wave-vector and \( \varepsilon_0 \) is the free-space permittivity. Eq. (1), tells us that the transversal components of the electric and magnetic fields are synchronous.

However, in this paper we discuss the phase relation between electric and magnetic field components in matter. Assuming an electromagnetic field oscillating inside a bulk material, after some straight-forward algebra we obtain the following relation between \( E_z \) and \( H_z \), as

\[ H_z = -\frac{\omega \varepsilon_0 \varepsilon_r k_z}{k_x k_z} E_z \] (2)

This is already telling us that the phase relation between the electric and magnetic fields is given by the phase of the permittivity as \( \angle \varepsilon_r = \tan^{-1}(\varepsilon'' \varepsilon'') \) where \( \varepsilon_r \) and \( \varepsilon'' \) are the real- and imaginary parts of the complex-valued permittivity.

Next, we discuss resonant conditions. The shape of the anti-symmetric toroidal moment can be nicely fitted with a Lorentzian function of the sort which is obtained normally for harmonic oscillators. The equation of motion for a harmonic oscillator, excited by a time-dependent harmonic force, is given by

\[ \ddot{p} + \gamma \dot{p} + \omega_0^2 p = F_0 \cos(\omega t) \] (3)

where \( p \) is the induced polarization, \( \gamma \) is the damping ratio, \( \omega_0 \) is the resonant frequency, and \( F_0 \) is the magnitude of the applied force. The stationary response \( p \) is written as \( p = p_0 \cos(\omega t + \varphi) \), with the amplitude and phase given by

\[ p_0 = \frac{F_0}{\sqrt{(\omega_0^2 - \omega^2)^2 + \omega^2 \gamma^2}} \quad \text{and} \quad \varphi = \tan^{-1} \left( \frac{\omega \gamma}{\omega_0^2 - \omega^2} \right), \] (4)

respectively. For \( \omega = \omega_0 \), we have obtained \( \varphi = \pi/2 \). This means that the deriving phase at highly resonant conditions is given by \( \varphi = \pi/2 \), regardless of the damping of the oscillator.

To apply this analogy to our case, we briefly mention here, that the toroidal moment is a hybrid solution and is not obtainable with quasi-static analysis. This means that the retardation plays an important role here. In this case, the force is given by the magnetic-vortex circulating around the relativistic current distribution of the electron, and this force will excite the polarization and hence the \( z \)-component of the toroidal moment excitation in our structure. In this regard, the exact shift of \( \varphi = \pi/2 \) will be obtained.
**Figure S2.** Illustration of cathodoluminescence detection in the experiment (a) and simulation (b). Blue arrows depict the radiation of the excited toroidal moments in the plasmonic heptamer structure.

Figure S2 shows different cathodoluminescence collection schemes used in the experiment and simulations. In the experiment, the radiation is collected by the parabolic mirror (light grey area in Figure S2a); it collects in principle the light emission in $+z$, $-y$ and $\pm x$ directions. In contrast, only the $+z$ component of the far-field radiation is calculated in the simulation, as if putting a detection plane above the specimen (black line in Figure S2b). The reason for this treatment is that the silver thin film in the simulation is constructed as an infinite plane. Any detection planes truncating the film infinity in $x$-$y$ plane will collect not only the radiation of excited modes, but also the propagating surface plasmons. Therefore, the emitted light in directions other than $+z$ is neglected in the simulated CL spectra.
**Figure S3.** Radial electric dipole mode sustained by the plasmonic heptamer cavity. The simulated magnetic field $H_z$ (a) and electric field $E_z$ (b) of the mode at 3.3 eV (376 nm). Grey circles denote the nanoholes. The impact location of the electron probe is indicated by the white dot. Red and black arrows show the directions of magnetic and electric dipoles, respectively. Scale bar is 200 nm.