S1 The effective excitation field for a dipole nanoparticle above a reflective substrate

With the Fourier transformation, the focal field $\boldsymbol{E}_f$ and its reflected part $\boldsymbol{E}_f^r$ can be, respectively, expressed as

$$\boldsymbol{E}_f(\boldsymbol{r}) = \iint \hat{\mathbf{E}}(k_x, k_y; 0)\mathrm{e}^{\mathrm{i}[k_x x+k_y y+k_z z]}\, dk_x dk_y \tag{S1.1-a}$$

$$\boldsymbol{E}_f^r(\boldsymbol{r}) = \iint \hat{\mathbf{E}}^{\mathrm{r}}(k_x, k_y; 0)\mathrm{e}^{\mathrm{i}[k_x x+k_y y-k_z z]}\, dk_x dk_y \tag{S1.1-b}$$

$\hat{\mathbf{E}}(k_x, k_y; 0)$ and $\hat{\mathbf{E}}^{\mathrm{r}}(k_x, k_y; 0)$, respectively, represent the Fourier spectrum for the incident focusing field and its reflected one. In angular spectrum theory, the free-space far field for a light source is entirely defined by its spatial frequency spectrum. Vice versa, according to the reversibility principle of beam path, the spatial frequency spectrum for the focal field can be obtained from its far field at the exit pupil of the objective lens by the expression

$$\hat{\mathbf{E}}(k_x, k_y; 0) = \frac{ife^{-ikf}}{2\pi k_z}\mathbf{E}_\infty(k_x, k_x). \tag{S1.2}$$

Here, f is the focal length of the objective lens. The far-field $\mathbf{E}_\infty$of the focal field can be step-by-step derived by the following process.

By mathematically expressing the linearly polarized beam before the beam splitter in the collimating optical path as $\mathbf{E}_{\mathrm{in}}=\mathrm{E}_{\mathrm{in}}\hat{\mathrm{n}}_{\mathrm{x}}$, it will be firstly transformed into a cylindrical radially polarized beam $\mathrm{E}_{\mathrm{in}}\hat{\mathrm{n}}_{\rho}$ by a polarization convertor, and will subsequently experience wavefront shaping by the specific mask to be denoted as $\mathrm{E}_{\mathrm{in}}\mathrm{P}(\rho)\ \hat{\mathrm{n}}_{\rho}$. Here, $\mathrm{P}(\rho)$ represents the apodization function of the illumination light field which is determined both by the shaping MASK and the numerical aperture of the objective lens. According to the energy conservation law between the two sides of the objective lens and the polarization transformation principle (from $\hat{\mathrm{n}}_{\rho}$ to $\hat{\mathrm{n}}_{\theta}$), the transmitted angular spectrum $\widehat{\mathrm{E}}_\infty(\theta, \phi)$ immediately after the objective lens is derived as $\mathrm{E}_{\mathrm{in}}\mathrm{P}(\theta)\sqrt{\frac{cos\theta}{n}}\hat{\mathrm{n}}_{\theta}$. In terms of the spatial frequencies $\mathbf{k} = (k_x, k_y, k_z)$ with the transformation formulas $\hat{\mathrm{n}}_{\theta} = \begin{bmatrix} cos\theta cos\phi \\ cos\theta sin\phi \\ -sin\theta \end{bmatrix}$ and $\mathbf{k} = \begin{bmatrix} k_x \\ k_y \\ k_z \end{bmatrix} = k\begin{bmatrix} sin\theta cos\phi \\ sin\theta sin\phi \\ -cos\theta \end{bmatrix}$, the far field for the focal field can be derived as

$$\mathbf{E}_{\infty}(k_x, k_x) = \frac{\mathrm{E_{in}P}(k_x,k_y)}{k\sqrt{(k_x^2+k_y^2)}}\sqrt{\frac{k_z}{nk}}\begin{bmatrix} k_x k_z \\ k_y k_z \\ -(k_x^2 + k_y^2) \end{bmatrix} \tag{S1.3}$$

where $k = 2\pi n/\lambda$ denotes the wave number for wavelength λ in a medium with a refractive index of n.

And the angular spectrum of the reflected focusing field can be obtained by multiplying the Fresnel reflection coefficient and an additional propagating phase factor, which is written as

$$\hat{\mathbf{E}}_{\infty}^{\mathrm{r}}(k_x, k_x) = r^p \, \mathrm{e}^{2ik_z z_0}\mathbf{E}_{\infty}(k_x, k_x) \tag{S1.4}$$

where r^p is the Fresnel reflection coefficient of the substrate for the p-polarized beam that only exists in the incident angular spectrum, and z_0 denotes the axial position of the upper surface of the substrate relative to the focal plane.

By inserting Equations (s1.1-s1.4) into the effective excitation expression (Equation (1)) in the main thesis, the effective excitation field above the substrate for a dipole source is derived, giving the following expression:

$$\boldsymbol{E}_{exc} = \frac{if e^{ikf}}{2\pi k}\mathcal{F}^{-1}\left\{\frac{\mathrm{E_{in}P}(k_x,k_y)}{k_z\sqrt{(k_x^2+k_y^2)}}\sqrt{\frac{k_z}{nk}}\begin{bmatrix} k_x k_z \\ k_y k_z \\ -(k_x^2 + k_y^2) \end{bmatrix}(\mathrm{e}^{ik_z z} + r^p \, \mathrm{e}^{2ik_z z_0}\mathrm{e}^{-ik_z z})\right\} \tag{S1.5}$$

$\mathcal{F}^{-1}$ represents the inverse Fourier transformation (iFT) operator.

It is clear that in addition to the vector LFM in the illumination system, the modulation of a reflective substrate to the excitation light field is also included in the expression (s1.6). Thus, the effective excitation for a dipole nanoparticle can be further modulated by tailoring the Fresnel reflection coefficient $r^p$and altering the gap z_0 between the focal plane and the upper surface of the substrate.

S2 The detection Green function for a dipole above a reflective substrate

For a dipole source above the reflective substrate with a gap of z_d, its radiating field in the upper half of space and the reflected part of its radiating field in the lower half of space can both be collected by the objective lens. The dipole radiation field in a homogeneous space is defined as the dynamic green function (DGF), denoted as $\overleftrightarrow{\boldsymbol{G}}_0(\boldsymbol{r}, \boldsymbol{r_0})$. The responding field in the upper space of the dipole in a reflective substrate can be written as

$$\boldsymbol{E}_1(\boldsymbol{r}) = \frac{\omega^2}{\varepsilon_0 c^2}[\overleftrightarrow{\boldsymbol{G}}_0(\boldsymbol{r}, \boldsymbol{r_0}) + \overleftrightarrow{\boldsymbol{G}}_0^r(\boldsymbol{r}, \boldsymbol{r_0})] \cdot \boldsymbol{P}(\boldsymbol{r_0}) \tag{S2.1}$$

by involving the reflection part of the free space DGF, denoted as $\overleftrightarrow{\boldsymbol{G}}_0^r(\boldsymbol{r},\boldsymbol{r_0})$. $\boldsymbol{r}$ is the arbitrary position vector in object space, and $\boldsymbol{r_0}$ denotes the position vector of the induced dipole. To simplify the derivation, the dipole used for deviating the DGF of the detection system is set as the focus of the objective lens, which coincides with the original point of the Cartesian coordinate system. Thus, $\boldsymbol{r_0} = (\boldsymbol{0},\boldsymbol{0},\boldsymbol{0})$. For any other induced dipoles in the vicinity of the focus, its responding field on the image focal plane of the detection system can be acquired with the linear transformation invariant principle. For a general excitation case, the dipole moment vector $\boldsymbol{P}(\boldsymbol{r_0})$ can be divided into three fundamental components as $(p_x, p_y, p_z)^T$, where T denotes the transpose operator to the row vector.

In terms of angular spectrum theory, the free space DGF $\overleftrightarrow{\boldsymbol{G}}_0$ of the dipole can be expressed as follows:

$$\overleftrightarrow{\boldsymbol{G}}_0 = \frac{i}{8\pi^2}\iint\left(\overleftrightarrow{M}^s + \overleftrightarrow{M}^p\right)\mathrm{e}^{\mathrm{i}[k_x x+k_y y+k_z z]}dk_x dk_y \tag{S2.2}$$

where the angular spectrum for an s-polarized and p-polarized light beam for a vector dipole is, respectively, a 3×3 tensor expressed as

$$\overleftrightarrow{M}^s = \frac{1}{k_z(k_x^2+k_y^2)}\begin{bmatrix} k_y^2 & -k_x k_y & 0 \\ -k_x k_y & k_x^2 & 0 \\ 0 & 0 & 0 \end{bmatrix},$$

$$\overleftrightarrow{M}^p = \frac{1}{k^2(k_x^2+k_y^2)}\begin{bmatrix} k_x^2 k_z & -k_x k_y & k_x(k_x^2+k_y^2) \\ -k_x k_y k_z & k_x^2 & k_y(k_x^2+k_y^2) \\ k_x(k_x^2+k_y^2) & k_y(k_x^2+k_y^2) & (k_x^2+k_y^2)^2/k_z \end{bmatrix}.$$

For the reflected DGF, it can be denoted as

$$\overleftrightarrow{\boldsymbol{G}}_0 = \frac{i}{8\pi^2}\iint\left(\overleftrightarrow{M}_r^s + \overleftrightarrow{M}_r^p\right)\mathrm{e}^{\mathrm{i}[k_x x+k_y y+k_z z]}dk_x dk_y \tag{S2.3}$$

and the angular spectrum for the reflected DGF can be obtained from that of the free space DGF by multiplying the Fresnel reflection coefficient of the substrate and the dipole–substrate distance related phase delay factor, written as $\overleftrightarrow{\mathrm{M}}_\mathrm{r}^\mathrm{s} = \mathrm{r}^\mathrm{s}\mathrm{e}^{2\mathrm{i}k_z z_d}\overleftrightarrow{\mathrm{M}}^\mathrm{s}$ and $\overleftrightarrow{\mathrm{M}}_\mathrm{r}^\mathrm{p} = -\mathrm{r}^\mathrm{p}\mathrm{e}^{2\mathrm{i}k_z z_d}\overleftrightarrow{\mathrm{M}}^\mathrm{p}$.

Combining the expressions from (S2.1) to (S2.3), the total radiation field in the upper half of space of a dipole source above a reflective substrate can be rewritten as

$$\boldsymbol{E}_1(\boldsymbol{r}) = \frac{\omega^2}{\varepsilon_0 c^2}\iint \hat{\mathbf{E}}_0(k_x, k_y; 0)\mathrm{e}^{i[k_x x+k_y y+k_z z]}\, dk_x dk_y \cdot \mathbf{P}(\boldsymbol{r_0}). \tag{S2.4}$$

Here, $\hat{\mathbf{E}}_0(\mathrm{k_x}, \mathrm{k_y}; 0)$ denotes the total angular spectrum, expressed as

$$\hat{\mathbf{E}}_0(k_x, k_y; 0) = \frac{i}{8\pi^2}\{[\overleftrightarrow{M}^s + \overleftrightarrow{M}^p] + [\overleftrightarrow{M}_r^s + \overleftrightarrow{M}_r^p]\}. \quad \text{(S2.5)}$$

Now, the field at the collection aperture of the objective lens can be directly obtained from the far-field angular spectrum of the dipole by

$$\mathbf{E}_\infty(k_x, k_y) = -2\pi i k_z \hat{\mathbf{E}}_0(k_x, k_y; 0)\frac{e^{ikf}}{f} \quad \text{(S2.6)}$$

In the following, the propagation and modulation processes in the detection system will be step-by-step described mathematically.

The field collected by the objective lens can be expressed as

$$\mathbf{E}_1^\infty(k_x, k_y) = \{[\mathbf{E}_\infty(k_x, k_y)\cdot \hat{n}_\phi]\hat{n}_\phi + [\mathbf{E}_\infty(k_x, k_y)\cdot \hat{n}_\theta]\hat{n}_\rho\}\sqrt{\frac{n}{cos\theta}} \quad \text{(S2.7)}$$

where the energy conservation law before and after collection by the objective lens is satisfied, and the vector component $\hat{n}_\phi$ remains unaffected, but the vector component $\hat{n}_\theta$ is transformed into $\hat{n}_\rho$. And the field after transmitting the annular aperture can be denoted by the following expression by multiplying its apodization function:

$$\mathbf{E}_2^\infty(k_x, k_y) = \mathbf{E}_1^\infty(k_x, k_y)\cdot \mathrm{P}(k_x, k_y). \quad \text{(S2.8)}$$

In the detection optical path, while passing through the polarization convertor, the radially polarized and azimuthally polarized beam is transformed into a set of orthogonal polarized light, which can be expressed as

$$\mathbf{E}_{2'}^\infty(k_x, k_y) = \{\ \mathbf{E}_2^\infty(k_x, k_y)\cdot \hat{n}_\rho]\hat{n}_x + [\ \mathbf{E}_2^\infty(k_x, k_y)\cdot \hat{n}_\phi]\hat{n}_y\} \quad \text{(S2.9)}$$

By expressing the field before the tube lens in cylindrical coordinates with the institution of $\hat{n}_x = \cos\phi\hat{n}_\rho - \sin\phi\hat{n}_\rho\hat{n}_\phi$ and $\hat{n}_y = \sin\phi\hat{n}_\rho + \cos\phi\hat{n}_\phi$, and by applying the energy conservation and polarization transformation of the tube lens, the angular spectrum immediately after the tube lens can be written as

$$\mathbf{E}_3^\infty(k_x, k_y) = \{\ \mathbf{E}_{2'}^\infty(k_x, k_y)\cdot \hat{n}_\rho]\hat{n}_{\theta'} + [\ \mathbf{E}_2^\infty(k_x, k_y)\cdot \hat{n}_\phi]\hat{n}_\phi\}\sqrt{cos\theta'}. \quad \text{(S2.10)}$$

Thus, the focusing field on the image space can be obtained by the following:

$$\boldsymbol{E}_{im}(\boldsymbol{r}') = \frac{if' e^{ik'f'}}{2\pi}\iint \mathbf{E}_3^\infty(\mathrm{k_x}, \mathrm{k_y})\frac{e^{ik'_z z'}}{k'_z k'_z} e^{i[k_x x + k_y y]}\, d\mathrm{k_x} d\mathrm{k_y} \quad \text{(S2.11)}$$

where f' is the focal length for the tube lens. With the sine condition at the objective lens and tube lens $\rho = fsin\theta = f'sin\theta'$, and the magnification factor M, the physical quantity in the object space and the image space can be interconnected as $x' = Mx$; $y' = My'$; $z' = zM^2/n$; $k_{x'} = Mk_x$; $k_{y'} =$

$\mathrm{Mk_y}$; $k_{z\prime} = k_0 cos\theta'$. As the focal length of the tube lens is much larger that that of the objective lens, $f' \gg f$, $sin\theta' = (f/f')sin\theta \approx 0$ and $\cos\theta' = \sqrt{1-(f/f')^2 sin^2\theta} \approx 1$.

Combining the equations from (S2.4) to (S2.11) and the related variable relationship between the object space and image space, as well as between the real space and the k space, the effective detection green function for a vector dipole above a reflective substrate can be expressed as the iFT of the transfer function $\overleftrightarrow{\mathbf{T}}_{\mathrm{det}}$ of the super resolution detection microscopy system as follows:

$$\overleftrightarrow{\mathbf{G}}_{\mathrm{det}}(\boldsymbol{r}', \boldsymbol{r_0}) = \frac{f\prime e^{i(kf-k_0 f\prime)}}{8\pi^2 k_0 f M^2} \mathcal{F}^{-1}\{\overleftrightarrow{\mathbf{T}}_{\mathrm{det}}\} \tag{S2.12}$$

The transfer function tensor includes three column vectors $\overleftrightarrow{\mathbf{T}}_{\mathrm{det}} = [\mathbf{T}_{\mathrm{x}}, \mathbf{T}_{\mathrm{y}}, \mathbf{T}_{\mathrm{z}}]$. For the dipole component oriented along the x-, y-, and z- axis, it, respectively, has the following expressions:

$$\mathrm{T_x} = \mathcal{A}_{\mathrm{det}} \begin{bmatrix} \frac{\mathcal{C}(k_x^3 k_z^2 + k_x k_y^2 k_z^2 + k_x (k_x^2+k_y^2)^2)}{k^3} \\ \frac{\mathcal{D}(-k_y^3 - k_x^2 k_y)}{k_z} \\ 0 \end{bmatrix},$$

$$\mathrm{T_y} = \mathcal{A}_{\mathrm{det}} \begin{bmatrix} \frac{\mathcal{C}(k_y^3 k_z^2 + k_x^2 k_y k_z^2 + k_y (k_x^2+k_y^2)^2)}{k^3} \\ \frac{\mathcal{D}(k_x^3 + k_x k_y^2)}{k_z} \\ 0 \end{bmatrix},$$

$$\mathrm{T_z} = \mathcal{A}_{\mathrm{det}} \begin{bmatrix} \frac{\mathcal{C}\prime[-k_x^2 k_z (k_x^2+k_y^2) - k_y^2 k_z (k_x^2+k_y^2) - (k_x^2+k_y^2)^3/k_z]}{k^3} \\ 0 \\ 0 \end{bmatrix},$$

where $\mathcal{A}_{\mathrm{det}} = \frac{P(k_x,k_y)k_z}{(k_x^2+k_y^2)^{3/2}} \sqrt{\frac{k_z}{nk}} \mathrm{e}^{ikz\left[\left(\frac{f\prime}{f}\right)^2 - \frac{1}{2}\frac{k_x^2+k_y^2}{k^2}\right]}$, $\mathcal{C} = \mathrm{e}^{-ik_z z_d} - r^p \mathrm{e}^{ik_z z_d}$, $\mathcal{D} = \mathrm{e}^{-ik_z z_d} + r^s \mathrm{e}^{ik_z z_d}$, and $\mathcal{C}' = \mathrm{e}^{-ik_z z_d} + r^p \mathrm{e}^{ik_z z_d}$.

According to all of the above derivations, the reflection response of the substrate to the radiation responding of an induced dipole above a substrate is included in the detection green function tensor. The final derived results indicate that there only exists a lateral field component in the confocal image plane for the induced dipole above the substrate, which is same as that without the substrate. However, each image field component for a dipole oscillating along any directions is enhanced or weakened by the constructive or destructive interference with the reflected parts.

S3 The reflected imaging field of the incident focusing field by the substrate

If a sample has a feature size larger than the sub-wavelength excitation spot, such as a nanotriangle with a smooth surface, and a side length larger than a wavelength, the light–matter interaction in the planar area of the nanotriangle can be dealt with by the Fresnel equations. In this case, the image field is the reflected illumination field. However, at the boundary and vertex area, the theory of nano-optics should be applied, including the multiple–multiple method, the volume integral method, total green function, and so on. Furthermore, if microscopy is applied in imaging a nanoparticle with a feature size much smaller than its spatial resolution limit evaluated by the free space PSF, the reflected or transmitted illumination field at the substrate will also reach the image plane as a background field unless a special detection system is applied, such as in dark-field microscopy in which only the scattered field is detected. Thus, the theoretical derivation of the image field for the reflected illumination field has a very critical role, while an extended planar nanostructure or a very small nanoparticle is considered. For the later imaging case, the absorption of the small nanoparticle in the illumination field is very weak, so it can be ignored.

By replacing the angular spectrum expression for dipole radiation above the substrate in Equation (S2.5) with that for the incident focusing field at the focal plane, as described in Equation (S1.2), the image field for the reflected focusing field is derived to have the following expression:

$$\boldsymbol{E}_B^{im}(\boldsymbol{r}') = \frac{if' e^{-ik_0 f'}}{2\pi k_0 M^2} \mathcal{F}^{-1}\left\{\mathrm{P}(\mathrm{k_x}, \mathrm{k_y}) \mathrm{e}^{2\mathrm{i}\mathrm{k_z}\mathrm{z_d}} \mathrm{r^p}(\mathrm{k_x}, \mathrm{k_y}) \mathrm{e}^{\mathrm{ikz}\left[\left(\frac{f'}{f}\right)^2 - \frac{1}{2}\frac{\mathrm{k_x^2}+\mathrm{k_y^2}}{\mathrm{k^2}}\right]}\right\}. \qquad \text{(S3.1)}$$

We can see that there only exists the x-polarized component on the image plane for the reflected background field.