Supplemental Material: Multiple Scattering Enhanced Single Particle Sensing

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We are interested in the distribution, in the case of a scalar field, of $\gamma_1 = 1 + \sum_{i=1}^{N} A_i$ as defined in the main text, where $A_i = (k_0^2/\varepsilon_0)\exp[-i\boldsymbol{k}_{\rm out}\cdot(\boldsymbol{r}_i - \boldsymbol{r}_{N+1})]\sum_{j=1}^{N} M_{ij}^{-1}\alpha G_{j,N+1}$. We assume that the phase of A_i is uniformly distributed over the full 2π range and independent of the amplitudes $|A_i|$. Making the further assumption that N is large, such that the central limit theorem can be applied, it then follows that the phasor sum $\sum_{i=1}^{N} A_i$ is a zero mean complex circular Gaussian random variable with variance $\sigma_1^2 = N\langle|A_i|^2\rangle/2$ [1]. Consequently, the probability density function of γ_1 is a rotationally symmetric Gaussian with mean of 1 and the same standard deviation. The amplitude $|\gamma_1|$ in turn follows a Rician distribution. In order to calculate σ_1^2, we expand M_{ij}^{-1} as a Neumann series to give

$$A_i = e^{-i\boldsymbol{k}_{\rm out}\cdot(\boldsymbol{r}_i - \boldsymbol{r}_{N+1})}\sum_{k=0}^{\infty}\left(\frac{k_0^2}{\varepsilon_0}\right)^{k+1}\sum_{l_1,\ldots,l_{k-1},j}\alpha G_{il_1}\alpha G_{l_1l_2}\alpha G_{l_2l_3}\ldots\alpha G_{l_{k-2}l_{k-1}}\alpha G_{l_{k-1}j}\alpha G_{j,N+1}. \tag{1}$$

Since $G_{l_il_j}$ describes free propagation from $\boldsymbol{r}_{l_j}$ to $\boldsymbol{r}_{l_i}$ and α describes a scattering event, A_i corresponds to a sum over all scattering paths visiting scatterers $N+1 \to j \to l_{k-1} \to l_{k-2} \to \ldots \to l_2 \to l_1 \to i$ and the sum implicitly excludes terms where $l_p = l_{p+1}$ (i.e. the same scatterer is not visited consecutively). It is useful to define the sum over k, describing propagation from $\boldsymbol{r}_i$ to $\boldsymbol{r}$ via all scattering paths, more generally for any start and end points $\boldsymbol{r}'$ and $\boldsymbol{r}$ respectively. We denote this quantity $G_{sc}(\boldsymbol{r},\boldsymbol{r}')$ as it is (ignoring some prefactors) the Green's function for the disordered system including the N initial scatterers, and it is given by

$$G_{sc}(\boldsymbol{r},\boldsymbol{r}') = \sum_{k=0}^{\infty}\left(\frac{k_0^2}{\varepsilon_0}\right)^{k+1}\sum_{l_1,\ldots,l_k}\alpha G(\boldsymbol{r},\boldsymbol{r}_{l_1})\alpha G_{l_1l_2}\alpha G_{l_2l_3}\ldots\alpha G_{l_{k-2}l_{k-1}}\alpha G_{l_{k-1}l_k}\alpha G(\boldsymbol{r}_{l_k},\boldsymbol{r}'). \tag{2}$$

Following the approach in Ref. [2], replacing each Green's function by its Fourier decomposition and ignoring paths with repeated scatterers (i.e. assuming each $\boldsymbol{r}_{l_i}$ is distinct), one can average over the intermediate scatterer positions to give

$$\begin{aligned}\langle G_{sc}(\boldsymbol{r},\boldsymbol{r}')\rangle &= \int\left[1 - n\alpha\frac{k_0^2}{\varepsilon_0}\widetilde{G}(\boldsymbol{q})\right]^{-1}\alpha\frac{k_0^2}{\varepsilon_0}\widetilde{G}(\boldsymbol{q})e^{i\boldsymbol{q}\cdot(\boldsymbol{\rho}-\boldsymbol{\rho}')}\frac{d^2\boldsymbol{q}}{(2\pi)^2}\\ &= \int\frac{-4\mu}{k_{\rm SPP}^2 - q^2 + 4n\mu}e^{i\boldsymbol{q}\cdot(\boldsymbol{\rho}-\boldsymbol{\rho}')}\frac{d^2\boldsymbol{q}}{(2\pi)^2},\end{aligned} \tag{3}$$

where $\widetilde{G}(\boldsymbol{q}) = -4A_0\exp(-2ak_{\rm SPP}z_s)/[k_{\rm SPP}^2 - q^2]$ is the transverse Fourier transform of $G_{\rm SPP}$ defined in Eq. (9) of the main text and μ is also defined in the main text. Noting that averaging restores translational invariance, we can define the Fourier space representation $\langle\widetilde{G}_{sc}(\boldsymbol{q})\rangle = \int\langle G_{sc}(\boldsymbol{r},\boldsymbol{r}')\rangle\exp[-i\boldsymbol{q}\cdot(\boldsymbol{\rho}-\boldsymbol{\rho}')]d^2\boldsymbol{\rho}$, which is given by

$$\langle\widetilde{G}_{sc}(\boldsymbol{q})\rangle = \frac{-4\mu}{k_{\rm SPP}^2 - q^2 + 4n\mu}. \tag{4}$$

Eq. (4) shows that the average effect of scattering is to shift the SPP wavevector [3]. In order to calculate σ_1^2, we must evaluate $\langle A_iA_i^*\rangle = \langle G_{sc}(\boldsymbol{r}_i,\boldsymbol{r}_{N+1})G_{sc}^*(\boldsymbol{r}_i,\boldsymbol{r}_{N+1})\rangle$. As G_{sc} is a sum over all multiple scattering paths, $|A_i|^2$ contains the interference of all paths. Within the ladder approximation, we assume that the phase difference between different scattering paths mean they will tend to average out over realisations unless they are identical (i.e. visit the same scatterers in the same order) [4]. This allows $\langle|A_i|^2\rangle$ to be expressed as

$$\begin{aligned}\langle|A_i|^2\rangle = &\int\langle G_{sc}(\boldsymbol{r}_i,\boldsymbol{r}_{N+1})\rangle\langle G_{sc}^*(\boldsymbol{r}_i,\boldsymbol{r}_{N+1})\rangle\frac{d^2\boldsymbol{\rho}_i}{L^2}\\ &+\sum_{k=1}^{\infty}\sum_{s_1,\ldots,s_k=1}^{N}\int|\langle G_{sc}(\boldsymbol{r}_i,\boldsymbol{r}_{s_1})\rangle\langle G_{sc}(\boldsymbol{r}_{s_1},\boldsymbol{r}_{s_2})\rangle\langle G_{sc}(\boldsymbol{r}_{s_2},\boldsymbol{r}_{s_3})\rangle\ldots\langle G_{sc}(\boldsymbol{r}_{s_k},\boldsymbol{r}_{N+1})\rangle|^2\frac{d^2\boldsymbol{\rho}_i}{L^2}\frac{d^2\boldsymbol{\rho}_{s_1}}{L^2}\frac{d^2\boldsymbol{\rho}_{s_2}}{L^2}\ldots\frac{d^2\boldsymbol{\rho}_{s_k}}{L^2},\end{aligned} \tag{5}$$

where the first term is the contribution from the interference of paths sharing no scatterers and each term in the sum over k is the contribution from the interference between one path from A_i visiting k scatterers $s_1, s_2, \ldots, s_k$ and a conjugated path (from A_i^*) visiting the same k scatterers in the same order. In using $\langle G_{sc}\rangle$ instead of $G_{\rm SPP}$ to propagate between each of these k shared scatterers, we account for any scattering during this propagation from other scatterers outside this set of k scatterers. The integration $\int \ldots d^2\boldsymbol{\rho}_{s_j}/L^2$ arises from averaging over the scatterer positions. Inserting the Fourier decomposition $\langle G_{sc}(\boldsymbol{r},\boldsymbol{r}')\rangle = \int \langle \widetilde{G}_{sc}(\boldsymbol{q})\rangle \exp[i\boldsymbol{q}\cdot(\boldsymbol{\rho}-\boldsymbol{\rho}')]d^2\boldsymbol{q}/(2\pi)^2$ for each factor of $\langle G_{sc}\rangle$ results in

$$\begin{aligned}\langle |A_i|^2\rangle = &\frac{1}{L^2}\int \left|\langle \widetilde{G}_{sc}(\boldsymbol{q})\rangle\right|^2 d^2\boldsymbol{q}/(2\pi)^2 \\ &+\sum_{k=1}^{\infty}\sum_{s_1,\ldots,s_k=1}^{N}\int \langle \widetilde{G}_{sc}(\boldsymbol{q}_1)\rangle\langle \widetilde{G}_{sc}(\boldsymbol{q}_2)\rangle\ldots\langle \widetilde{G}_{sc}(\boldsymbol{q}_k)\rangle\langle \widetilde{G}^*_{sc}(\boldsymbol{q}'_1)\rangle\langle \widetilde{G}^*_{sc}(\boldsymbol{q}'_2)\rangle\ldots\langle \widetilde{G}^*_{sc}(\boldsymbol{q}'_k)\rangle \\ &\times e^{i(\boldsymbol{q}_1-\boldsymbol{q}'_1)\cdot(\boldsymbol{\rho}_i-\boldsymbol{\rho}_{s_1})}e^{i(\boldsymbol{q}_2-\boldsymbol{q}'_2)\cdot(\boldsymbol{\rho}_{s_1}-\boldsymbol{\rho}_{s_2})}\ldots e^{i(\boldsymbol{q}_k-\boldsymbol{q}'_k)\cdot(\boldsymbol{\rho}_{s_k}-\boldsymbol{\rho}_{N+1})}\frac{d^2\boldsymbol{\rho}_i}{L^2}\frac{d^2\boldsymbol{\rho}_{s_1}}{L^2}\frac{d^2\boldsymbol{\rho}_{s_2}}{L^2}\ldots\frac{d^2\boldsymbol{\rho}_{s_k}}{L^2} \\ &\times\frac{d^2\boldsymbol{q}_1}{(2\pi)^2}\frac{d^2\boldsymbol{q}'_1}{(2\pi)^2}\frac{d^2\boldsymbol{q}_2}{(2\pi)^2}\frac{d^2\boldsymbol{q}'_2}{(2\pi)^2}\ldots\frac{d^2\boldsymbol{q}_k}{(2\pi)^2}\frac{d^2\boldsymbol{q}'_k}{(2\pi)^2}.\end{aligned} \tag{6}$$

Integrating over each position gives a factor $(2\pi)^2\delta(\boldsymbol{q}_i-\boldsymbol{q}'_i)/L^2$, while the sum over scatterers $\sum_{s_1,\ldots,s_k}$ reduces to a factor of N^k. Therefore, Eq. (6) becomes

$$\langle |A_i|^2\rangle = \frac{1}{L^2}\int \left|\langle \widetilde{G}_{sc}(\boldsymbol{q})\rangle\right|^2 d^2\boldsymbol{q}/(2\pi)^2 + \sum_{k=1}^{\infty} N^k\left(\frac{1}{L^2}\int\left|\langle \widetilde{G}_{sc}(\boldsymbol{q})\rangle\right|^2\frac{d^2\boldsymbol{q}}{(2\pi)^2}\right)^{k+1}. \tag{7}$$

The integral can be evaluated, resulting in

$$\begin{aligned}\int\left|\langle \widetilde{G}_{sc}(\boldsymbol{q})\rangle\right|^2\frac{d^2\boldsymbol{q}}{(2\pi)^2} &= \int\frac{16|\mu|^2}{(k_{\rm SPP}^2-q^2+4n\mu)(k_{\rm SPP}^{*2}-q^2+4n\mu^*)}\frac{d^2\boldsymbol{q}}{(2\pi)^2} \\ &= \frac{4|\mu|^2}{\mathrm{Im}[k_{\rm SPP}^2+4n\mu]} \\ &= \frac{\sigma_{\rm SPP}}{l_{\rm abs}^{-1}+4n\,\mathrm{Im}[\mu]/\,\mathrm{Re}[k_{\rm SPP}]}.\end{aligned} \tag{8}$$

Therefore, Eq. (7) reduces to

$$\langle |A_i|^2\rangle = \frac{1}{L^2}\frac{\sigma_{\rm SPP}}{l_{\rm abs}^{-1}+4n\,\mathrm{Im}[\mu]/\,\mathrm{Re}[k_{\rm SPP}]} + \frac{1}{L^2}\sum_{k=1}^{\infty}\left(\frac{N}{L^2}\right)^k\left(\frac{\sigma_{\rm SPP}}{l_{\rm abs}^{-1}+4n\,\mathrm{Im}[\mu]/\,\mathrm{Re}[k_{\rm SPP}]}\right)^{k+1}. \tag{9}$$

Using $\sigma_1^2 = N\langle |A_i|^2\rangle/2$, recalling $n=N/L^2$ and $n\sigma_{\rm SPP}=l_s^{-1}$ gives

$$\begin{aligned}\sigma_1^2 &= \frac{1}{2}\sum_{k=1}^{\infty}\left(\frac{l_s^{-1}}{l_{\rm abs}^{-1}+4n\,\mathrm{Im}[\mu]/\,\mathrm{Re}[k_{\rm SPP}]}\right)^k \\ &= \frac{1}{2}\frac{l_s^{-1}}{l_{\rm abs}^{-1}+4n\,\mathrm{Im}[\mu]/\,\mathrm{Re}[k_{\rm SPP}]-l_s^{-1}}\end{aligned} \tag{10}$$

which is the result in Eq. (10) in the main text.

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