PNAS
www.pnas.org

Supplementary Information for

Quantum phase-sensitive diffraction and imaging using entangled photons

Shahaf Asban, Konstantin E. Dorfman and Shaul Mukamel

Shahaf Asban, Konstantin E. Dorfman and Shaul Mukamel
sasban@uci.edu,dorfmank@lps.ecnu.edu.cn ,smukamel@uci.edu

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www.pnas.org/cgi/doi/10.1073/pnas.1904839116

Supporting Information Text

1. The reduced density matrix

Below we focus on the two-photon subspace of the density matrix. The density matrix in the interaction picture takes the form,

$$\rho\left(t\right)=\mathcal{T}e^{-i\int d\tau\mathcal{H}_{I,-}\left(\tau\right)}\rho_{\mu}\otimes\rho_{\phi},$$

where $\rho\left(t=0\right)=\rho_0=\rho_{\mu}\otimes\rho_{\phi}$. To first order in the interaction $\mathcal{H}_I=\int d\boldsymbol{r}\sigma\left(\boldsymbol{r},t\right)\boldsymbol{A}^2\left(\boldsymbol{r},t\right)$ we get,

$$\rho^{(1)}\left(t\right)=\rho_{\mu}\otimes\rho_{\phi}-i\int d\tau\left[\mathcal{H}_I\left(\tau\right),\rho_0\right],$$
$$\rho^{(1)}_{int}\left(t\right)=-i\int dtd\boldsymbol{r}\sigma\left(\boldsymbol{r},t\right)\boldsymbol{A}^2\left(\boldsymbol{r},t\right)\rho_0+i\rho_0\int dtd\boldsymbol{r}\sigma\left(\boldsymbol{r},t\right)\boldsymbol{A}^2\left(\boldsymbol{r},t\right),$$

for diffraction we take $\boldsymbol{A}^2=A_pA_d^{\dagger}+h.c.$ where the p=pump and d=diffracted modes. By tracing over the matter degrees of freedom we get,

$$\rho^{(1)}_{int,\phi}\left(t\right)=-i\int dtd\boldsymbol{r}\left\langle\sigma\left(\boldsymbol{r},t\right)\right\rangle\boldsymbol{A}^2\left(\boldsymbol{r},t\right)\sum_{s,i,s'i'}\Phi_{si}\Phi^*_{s'i'}|\mathbf{1}_s\mathbf{1}_i\rangle\langle\mathbf{1}_{s'}\mathbf{1}_{i'}|$$
$$+i\sum_{s,i,s'i'}\Phi_{si}\Phi^*_{s'i'}|\mathbf{1}_s\mathbf{1}_i\rangle\langle\mathbf{1}_{s'}\mathbf{1}_{i'}|\int dtd\boldsymbol{r}\left\langle\sigma\left(\boldsymbol{r},t\right)\right\rangle^*\boldsymbol{A}^2\left(\boldsymbol{r},t\right).$$

The vector-potential is given by,

$$\boldsymbol{A}\left(\boldsymbol{r},t\right)=i\sum_k\hat{\epsilon}_ka_ke^{i\left(\boldsymbol{k}\cdot\boldsymbol{r}-\omega_kt\right)}+h.c.$$
$$\boldsymbol{A}^2\left(\boldsymbol{r},t\right)\rightarrow A_pA_d^{\dagger}+h.c.$$
$$=\sum_{d,p}\left(\hat{\epsilon}_pa_pe^{i\left(\boldsymbol{k}_p\cdot\boldsymbol{r}-\omega_pt\right)}-\hat{\epsilon}^*_pa_p^{\dagger}e^{-i\left(\boldsymbol{k}_p\cdot\boldsymbol{r}-\omega_pt\right)}\right)\left(\hat{\epsilon}_da_de^{i\left(\boldsymbol{k}_d\cdot\boldsymbol{r}-\omega_dt\right)}-\hat{\epsilon}^*_da_d^{\dagger}e^{-i\left(\boldsymbol{k}_d\cdot\boldsymbol{r}-\omega_dt\right)}\right),$$

We will look at the two photon subspace that corresponds to the signal,

$$\rho^{(1,2)}_{int,\phi}\left(t\right)=-i\sum_{s,i,s'i'}\sum_{d,p}\Phi_{si}\Phi^*_{s'i'}\int dtd\boldsymbol{r}\left\langle\sigma\left(\boldsymbol{r},t\right)\right\rangle e^{i\left(\boldsymbol{k}_{dp}\cdot\boldsymbol{r}-\omega_{dp}t\right)}\hat{\epsilon}_p\hat{\epsilon}^*_da_d^{\dagger}a_p|\mathbf{1}_s\mathbf{1}_i\rangle\langle\mathbf{1}_{s'}\mathbf{1}_{i'}|+h.c.,$$

and since only the signal beam interacts with the p,d modes we use,

$$a_pa_d^{\dagger}|\mathbf{1}_s\mathbf{1}_i\rangle=\delta_{ps}|\mathbf{1}_d\mathbf{1}_i\rangle.$$

Finally,

$$\rho^{(1,2)}_{int,\phi}\left(t\right)=-i\sum_{d,s,i,s'i'}\hat{\epsilon}_s\cdot\hat{\epsilon}^*_d\Phi\left(\boldsymbol{k}_s,\boldsymbol{k}_i\right)\Phi^*\left(\boldsymbol{k}'_s,\boldsymbol{k}'_i\right)\left\langle\sigma\left(\boldsymbol{k}_{ds},\omega_{ds}\right)\right\rangle\left[|\mathbf{1}_d\mathbf{1}_i\rangle\langle\mathbf{1}_{s'}\mathbf{1}_{i'}|\right.$$
$$\left.+i\hat{\epsilon}^*_s\cdot\hat{\epsilon}_d\left\langle\sigma\left(\boldsymbol{k}_{ds'},\omega_{ds'}\right)\right\rangle^*|\mathbf{1}_s\mathbf{1}_i\rangle\langle\mathbf{1}_d\mathbf{1}_{i'}|\right].$$

Using the Schmidt decomposition we arrive at,

$$\rho^{(1,2)}_{int,\phi}\left(t\right)=-i\sum_{d,s,i,s'i'}\hat{\epsilon}_s\cdot\hat{\epsilon}^*_d\sum_{nm}\sqrt{\lambda_n\lambda_m}u_n\left(\boldsymbol{k}_s\right)v^*_n\left(\boldsymbol{k}_i\right)u^*_m\left(\boldsymbol{k}'_s\right)v_m\left(\boldsymbol{k}'_i\right)\left\langle\sigma\left(\boldsymbol{k}_{ds},\omega_{ds}\right)\right\rangle\left[|\mathbf{1}_d\mathbf{1}_i\rangle\langle\mathbf{1}_{s'}\mathbf{1}_{i'}|\right.$$
$$\left.+i\hat{\epsilon}^*_s\cdot\hat{\epsilon}_d\left\langle\sigma\left(\boldsymbol{k}_{ds'},\omega_{ds'}\right)\right\rangle^*|\mathbf{1}_s\mathbf{1}_i\rangle\langle\mathbf{1}_d\mathbf{1}_{i'}|\right].$$

Expanding the complex exponent of the Fourier transform using the summation of mixed space basis functions (one in plane waves and the other in real-space), taking the trace with respect to the signal beam yields the reduced density matrix of the idler is finally given by,

$$\rho_{Idler}=\sum_{n,m,i,i'}\mathcal{P}_{nm}v^*_n\left(\boldsymbol{k}_i\right)v_m\left(\boldsymbol{k}_i\right)|\mathbf{1}_i\rangle\langle\mathbf{1}_{i'}|+h.c \qquad [1]$$

where we have defined $\mathcal{P}_{nm}=i\beta_{nm}\sqrt{\lambda_n\lambda_m}$. The expectation value of the intensity of the idler beam results in Eq.(1) in the main text.

2. The coincidence measurement

The setup for the coincidence measurement is depicted in Fig.1 of the main text. An entangled photon pair created by parametric down conversion is separated by a beam splitter BS into *signal* (s) and *idler* (i) beams with wave-vector, frequency and polarization $(\boldsymbol{k}_m, \omega_m, \epsilon_m)$ where $m \in \{s, i\}$. The signal beam undergoes a diffraction by the material sample prepared by an actinic pulse. The image is generated by the coincidence measurement of the signal and idler beams by two detectors, which provides an intensity-intensity correlation function ($g^{(2)}$ type). It is recorded Vs. the frequency of the signal photon $\bar{\omega}_s$ and position in the idler (transverse) detection plane $\bar{\boldsymbol{\rho}}_i$. The image is defined by the intensity correlation function of the detected photon-pair,

$$S\left[\bar{\boldsymbol{\rho}}_i\right] = \int d\boldsymbol{X}_s d\boldsymbol{X}_i G_s\left(\boldsymbol{X}_s, \bar{\boldsymbol{X}}_s\right) G_i\left(\boldsymbol{X}_i, \bar{\boldsymbol{X}}_i\right) \times \left\langle \mathcal{T} \hat{I}_s\left(\boldsymbol{r}_s, t_s\right) \hat{I}_i\left(\boldsymbol{r}_i, t_i\right) \mathcal{U}_I(t) \right\rangle, \quad [2]$$

The gating functions G_m represent the details of the measurement process (1, 2). $\hat{I}_m\left(\boldsymbol{r}_m, t_m\right) \equiv \hat{\boldsymbol{E}}^{(-)}_{m,R}\left(\boldsymbol{r}_m, t_m\right) \cdot \hat{\boldsymbol{E}}^{(+)}_{m,L}\left(\boldsymbol{r}_m, t_m\right)$ is the field intensity. $\hat{\boldsymbol{E}}^{(\pm)}$ are the negative and positive frequency components of the electric field operator. The electric field is given by the $\boldsymbol{E}\left(\boldsymbol{r}, t\right) = \sum_k \boldsymbol{E}^{(+)}_k\left(\boldsymbol{r}, t\right) + \boldsymbol{E}^{(-)}_k\left(\boldsymbol{r}, t\right)$ such that,

$$\boldsymbol{E}^{(+)}_k\left(\boldsymbol{r}, t\right) = \left(\boldsymbol{E}^{(-)}_k\left(\boldsymbol{r}, t\right)\right)^{\dagger} = \sqrt{\frac{2\pi\hbar\omega_{\boldsymbol{k}}}{V_{\boldsymbol{k}}}} \sum_{\nu} \epsilon^{(\nu)}_{\boldsymbol{k}} a_{\boldsymbol{k},\nu} e^{i\boldsymbol{k}\cdot\boldsymbol{r} - i\omega_{\boldsymbol{k}} t}, \quad [3]$$

with polarization $\epsilon^{(\nu)}_{\boldsymbol{k}}$ and the field annihilation (creation) operator $a_{\boldsymbol{k},\nu}\left(a^{\dagger}_{\boldsymbol{k},\nu}\right)$. The photon coordinates $\boldsymbol{X}_m \equiv \left(\boldsymbol{r}_m, t_m, \boldsymbol{k}_m, \omega_m\right)$ are mapped by the gating to the detected domain $\bar{\boldsymbol{X}}_m \equiv \left(\bar{\boldsymbol{r}}_m, \bar{t}_m, \bar{\boldsymbol{k}}_m, \bar{\omega}_m\right)$. The subscripts L/R stand for left and right super-operators which specify from which side they act on an ordinary operator (3), i.e. $\mathcal{O}_R \varrho \equiv \varrho \mathcal{O}$ and $\mathcal{O}_L \varrho \equiv \mathcal{O}\varrho$. $\mathcal{T}$ represents super-operators time ordering and $\mathcal{U}_I(t) \equiv \exp\left[-\frac{i}{\hbar}\int_{t_0}^{t} d\tau \mathcal{H}_{I,-}(\tau)\right]$ is the interaction picture propagator. The *off-resonance* radiation/matter coupling is $\mathcal{H}_I = \int d\boldsymbol{r} \sigma\left(\boldsymbol{r}, t\right) \boldsymbol{A}^2\left(\boldsymbol{r}, t\right)$ with the vector field $\boldsymbol{A}\left(\boldsymbol{r}, t\right) = -\frac{1}{c}\dot{\boldsymbol{E}}\left(\boldsymbol{r}, t\right)$. The subscript $(-)$ on a Hilbert space operators represents the commutator $\mathcal{O}_- \equiv \mathcal{O}_L - \mathcal{O}_R$. $\langle \cdots \rangle$ denotes the average with respect to the initial density matrix of the light and matter.

Expanding $\mathcal{U}_I(t)$ to first order in the interaction, and subtracting the noninteracting background, the image is finally given by a 6-point correlation function,

$$\mathcal{S}\left[\bar{\boldsymbol{\rho}}_i\right] = \frac{2A}{\hbar}\mathfrak{Re}\int d\boldsymbol{X}_s d\boldsymbol{X}_i G_s\left(\boldsymbol{X}_s, \bar{\boldsymbol{X}}_s\right) G_i\left(\boldsymbol{X}_i, \bar{\boldsymbol{X}}_i\right) \int_{-\infty}^{t_s} d\boldsymbol{r}' d\tau \left\langle \sigma\left(\boldsymbol{r}', \tau\right)\right\rangle_{\mu} \times \left\langle \mathcal{T} \hat{\boldsymbol{E}}^{(-)}_{s,R}\left(\boldsymbol{r}_s, t_s\right) \cdot \hat{\boldsymbol{E}}^{(+)}_{s,L}\left(\boldsymbol{r}_s, t_s\right) \hat{\boldsymbol{E}}^{(-)}_{i,R}\left(\boldsymbol{r}_i, t_i\right) \cdot \hat{\boldsymbol{E}}^{(+)}_{i,L}\left(\boldsymbol{r}_i, t_i\right) \boldsymbol{A}^{(+)}\left(\boldsymbol{r}', \tau\right) \boldsymbol{A}^{(-)}\left(\boldsymbol{r}', \tau\right) \right\rangle_{\phi}. \quad [4]$$

The subscripts ϕ, μ represent field and the matter degrees of freedom, respectively. Explicitly by the 10 field operator correlation function,

$$\mathcal{S}\left[\bar{\boldsymbol{\rho}}_i\right] = \frac{2A}{\hbar}\mathfrak{Re}\int d\boldsymbol{X}_s d\boldsymbol{X}_i G_s\left(\boldsymbol{X}_s, \bar{\boldsymbol{X}}_s\right) G_i\left(\boldsymbol{X}_i, \bar{\boldsymbol{X}}_i\right) \int_{-\infty}^{t_s} d\boldsymbol{r}' d\tau \left\langle \sigma\left(\boldsymbol{r}', \tau\right)\right\rangle_{\mu} \quad [5]$$

$$\times \sum_{\boldsymbol{k}_s, \boldsymbol{k}_i} \sum_{\boldsymbol{k}'_s, \boldsymbol{k}'_i} \Phi\left(\boldsymbol{k}_s, \boldsymbol{k}_i\right) \Phi^*\left(\boldsymbol{k}'_s, \boldsymbol{k}'_i\right)$$

$$\times \left\langle \boldsymbol{0}_{s'}, \boldsymbol{0}_{i'} \right| a^{\dagger}_{\boldsymbol{k}_s,\mu_s} a^{\dagger}_{\boldsymbol{k}_i,\mu_i} a_{\boldsymbol{k}'_s,\mu_s} a_{\boldsymbol{k}'_i,\mu_i} \mathcal{T} \hat{\boldsymbol{E}}^{(-)}_{s,R}\left(\boldsymbol{r}_s, t_s\right) \cdot \hat{\boldsymbol{E}}^{(+)}_{s,L}\left(\boldsymbol{r}_s, t_s\right) \hat{\boldsymbol{E}}^{(-)}_{i,R}\left(\boldsymbol{r}_i, t_i\right) \cdot \hat{\boldsymbol{E}}^{(+)}_{i,L}\left(\boldsymbol{r}_i, t_i\right) \boldsymbol{A}^{(+)}\left(\boldsymbol{r}', \tau\right) \boldsymbol{A}^{(-)}\left(\boldsymbol{r}', \tau\right) \left| \boldsymbol{0}_s, \boldsymbol{0}_i \right\rangle, \quad [6]$$

Contracting the field operators defined in Eq. 3 of the intensity-intensity expectation value Eq. 2, with the vector potential of the scattered modes (initially in the vacuum), We find a nonvanishing linear contribution. Assuming spatial gating for the idler tracing over the signal yields, operators results in,

$$\mathcal{S}\left[\bar{\rho}_i\right] = C\mathfrak{Re}\int d\boldsymbol{r}_s dt_s dt_i \sum_{s,s',i,i',d} \Phi\left(\boldsymbol{k}_s, \boldsymbol{k}_i\right) \Phi^*\left(\boldsymbol{k}'_s, \boldsymbol{k}'_i\right) \int d\boldsymbol{r}'\, dt\, \left\langle \sigma\left(\boldsymbol{r}, t\right)\right\rangle_{\mu} \times e^{i\boldsymbol{k}_{ds'}\cdot\boldsymbol{r}_s - i\omega_{ds'} t_s} e^{i\boldsymbol{k}_{ii'}\cdot\boldsymbol{r}_i - i\omega_{ii'} t_i} e^{i\boldsymbol{k}_{ds}\cdot\boldsymbol{r} - i\omega_{ds} t},$$

by integration over the signal and matter coordinates and expanding the transverse two-photon amplitudes in Schmidt modes we obtain,

$$\mathcal{S}\left[\bar{\boldsymbol{\rho}}_i\right]=\mathcal{A}\mathfrak{Re}\sum_{nm}\sqrt{\lambda_n\lambda_m}\beta_{nm}v_n^*\left(\boldsymbol{\rho}_i\right)v_m\left(\boldsymbol{\rho}_i\right), \tag{7}$$

where $\mathcal{A}=C\int d\omega_d d\omega_s d\omega_i G\left(\omega_s\right)G\left(\omega_d\right)\left|G\left(\omega_i\right)\right|^2 A\left(\omega_s+\omega_i\right)A^*\left(\omega_d+\omega_i\right)$ and,

$$\begin{aligned}\beta_{nm}&=\sum_{s,d}u_n\left(\boldsymbol{q}_s\right)\left\langle\sigma\left(\boldsymbol{q}_d-\boldsymbol{q}_s,k_{ds}^z,\omega_{ds}\right)\right\rangle_\mu u_m^*\left(\boldsymbol{q}_d\right)\\&=\int d\boldsymbol{r}\,u_n\left(\boldsymbol{\rho}\right)\left\langle\bar{\sigma}\left(\boldsymbol{\rho}\right)\right\rangle_\mu u_m^*\left(\boldsymbol{\rho}\right),\end{aligned} \tag{8}$$

and $\bar{\sigma}\left(\boldsymbol{\rho}\right)=\sum_{ds}\sigma\left(\boldsymbol{\rho}\right)$. This expresses the role of the charge density in diffraction in an intuitive manner, generating weighted rotations.

We next derive an expression for the far field diffraction after rotational averaging. This coincidence image in the far-field yields a similar expression to the one calculated from the reduced density matrix in Eq.(1) with additional spatial phase factor characteristic to far-field diffraction. Estimation of Eq.(6), using initial entangled state of the field for the setup depicted in Fig.1 of the main text, followed by rotational averaging and far-field approximation we obtain,

$$\begin{aligned}\mathcal{S}\left[\bar{\boldsymbol{\rho}}_i\right]=C\mathfrak{Re}\int d\omega_s\mathscr{E}\left[\omega_s\right]\int d\boldsymbol{\rho}_s\Phi\left(\boldsymbol{\rho}_s,\bar{\boldsymbol{\rho}}_i\right)\times\\\int d\boldsymbol{\rho}'\,\Phi\left(\boldsymbol{\rho}',\bar{\boldsymbol{\rho}}_i\right)\boldsymbol{\sigma}\left(\boldsymbol{\rho}'\right)e^{-i\boldsymbol{Q}_s\cdot\boldsymbol{\rho}'}.\end{aligned} \tag{9}$$

Here $\boldsymbol{Q}_s=\frac{\omega_s}{c}\hat{\rho}_s$, $\mathscr{E}\left[\omega_s\right]=\int d\omega_i G\left(\omega_s\right)G\left(\omega_i\right)\left|A\left(\omega_s+\omega_i\right)\right|^2$is a functional of the frequency, $\mathcal{S}=-\left(S-S_0\right)$ is the image with the noninteracting-uniform background (S_0) subtracted, and $\bar{\boldsymbol{\rho}}_i$ is the mapping onto the detector plane with the corresponding sign. $\boldsymbol{\sigma}\left(\boldsymbol{\rho}\right)\equiv\sum_{\alpha;a,b}\langle a|\hat{\sigma}\left(\boldsymbol{\rho}-\boldsymbol{\rho}_\alpha\right)|b\rangle$ denotes a matrix element of the charge-density operator with respect to the eigenstates $\{a,b\}$ and α specify the location of particles initially.

The matter is initially in a superposition state, created by a preparation process. $\sigma\left(\boldsymbol{\rho}\right)$ denotes summation over the longitudinal direction and $\boldsymbol{\rho}_\alpha$ are positions of particles in the sample. Substituting the Schmidt decomposition (Eq.(**??**)) in Eq.(9) gives,

$$\begin{aligned}\mathcal{S}\left[\bar{\boldsymbol{\rho}}_i\right]=C\mathfrak{Re}\int d\omega_s\mathscr{E}\left[\omega_s\right]d\boldsymbol{\rho}_s\sum_{nm}^{\infty}\sqrt{\lambda_n\lambda_m}u_n\left(\boldsymbol{\rho}_s\right)v_n^*\left(\bar{\boldsymbol{\rho}}_i\right)\times\\v_m\left(\bar{\boldsymbol{\rho}}_i\right)\int d\boldsymbol{\rho}'u_m^*\left(\boldsymbol{\rho}'\right)\boldsymbol{\sigma}\left(\boldsymbol{\rho}'\right)e^{-i\frac{\omega_s}{c}\hat{\rho}_s\cdot\boldsymbol{\rho}'}.\end{aligned} \tag{10}$$

This shows a smooth transition from momentum to real space imaging. For low Schmidt modes that do not vary a lot along the charge density scale, the last term yields $\boldsymbol{\sigma}\left(\boldsymbol{Q}_s,\cdot\right)\propto\int d\boldsymbol{\rho}'u_m^*\left(\boldsymbol{\rho}'\right)\boldsymbol{\sigma}\left(\boldsymbol{\rho}'\right)e^{-i\frac{\omega_s}{c}\hat{\rho}_s\cdot\boldsymbol{\rho}'}$. Then this quantity is projected on u_n and reweights the corresponding idler modes. When many of these projections are measured, the resulting image is the real-space image of the charge density. Expressing the complex exponent as superposition of Schmidt modes such that,

$$\mathcal{S}\left[\bar{\boldsymbol{\rho}}_i\right]\propto\mathfrak{Re}\sum_{nm}^{\infty}\gamma_{nm}\sqrt{\lambda_n\lambda_m}v_n^*\left(\bar{\boldsymbol{\rho}}_i\right)v_m\left(\bar{\boldsymbol{\rho}}_i\right) \tag{11}$$

where,

$$\gamma_{nm}=\sum_k\beta_{km}\int d\boldsymbol{\rho}_s d\omega_s\mathscr{E}\left[\omega_s\right]u_n\left(\boldsymbol{\rho}_s\right)u_k^*\left(\boldsymbol{Q}_s\right), \tag{12}$$

introduced by Eqs.(15, 16) in the main text. Here β_{nm} is the same overlap defined for the density matrix. From the definition of $\boldsymbol{Q}_s$ it is evident that its angular component of u_k is identical the corresponding in u_n and therefore γ_{nm} is composed of summation over modes with the same angular momentum if one considers LG basis set.

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