Supplemental Document

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Quantum-randomized polarization of laser pulses derived from zero-point diamond motion: supplement

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Quantum-randomized polarization of laser pulses derived from zero-point motion of diamond: supplemental document

This supplementary material outlines the derivation of the Stokes field arising from the excitation of the F_{2g} Raman modes where the wave-vector and linear polarization of the pump field is oriented along the [110] and [$1\bar{1}0$] cubic crystal axes respectively and presents the results of the NIST-800-90B test suite for statistical independence [1], applied to Stokes polarization measurements.

1. DERIVATION OF THE STOKES FIELD

The quantum mechanical equations of motion for stimulated Raman scattering (SRS) under the canonical quantization formalism, in the case of multiple excited modes generalize to [2],

$$\frac{\partial}{\partial\tau}\hat{Q}_n^\dagger(z,\tau) = -\Gamma\hat{Q}_n^\dagger(z,\tau) + \hat{F}_n^\dagger(z,\tau) + i\boldsymbol{\kappa}_n\mathbf{E}_p^*(\tau)\hat{\mathbf{E}}_S(z,\tau) \tag{S1a}$$

$$\frac{\partial}{\partial z}\hat{\mathbf{E}}_S(z,\tau) = -iC\sum_n \boldsymbol{\kappa}_n^*\mathbf{E}_p(\tau)\hat{Q}_n^\dagger(z,\tau). \tag{S1b}$$

where the symbols are defined as follows;

- $\tau = t - z/v$ is the time coordinate in the pump-pulse reference frame, where v is the velocity of the pump and Stokes fields. This implicitly assumes no material dispersion.
- $\hat{Q}_n^\dagger$ are the collective atomic operators for the n-th vibrational mode and $\hat{F}_n^\dagger$ are the corresponding Langevin operators that represent the zero-point motion of the crystal lattice [2, 3].
- $\hat{\mathbf{E}}_S$ is the (total) Stokes field operator.
- $\mathbf{E}_p = \hat{\mathbf{e}}_p E_p$ is the (classical) pump field that is assumed to be undepleted, thus exhibiting no z-dependence.
- $\boldsymbol{\kappa}_n$ are the polarizabilities associated with F_{2g} Raman modes, and are represented as tensors in this formulation to account for polarization effects.
- Γ is the phonon damping rate.
- $C = 2\pi N\hbar\omega_S v/c^2$, where N is the atomic density of the crystal, ω_s is the Stokes frequency and c is the speed of light in a vacuum.

The first step to solving Eq. S1 is to eliminate the partial derivative in z by taking the spatial Laplace transform. Assuming no initial Stokes field in z, Eq. S1 becomes

$$\frac{\partial}{\partial\tau}\hat{q}_n^\dagger(s,\tau) = -\Gamma\hat{q}_n^\dagger(s,\tau) + i\boldsymbol{\kappa}_n\mathbf{E}_p^*(\tau)\hat{\boldsymbol{\mathcal{E}}}_S(s,\tau) + \hat{f}_n^\dagger(s,\tau), \tag{S2a}$$

$$\hat{\boldsymbol{\mathcal{E}}}_S(s,\tau) = -iC\sum_n \frac{1}{s}\boldsymbol{\kappa}_n^*\mathbf{E}_p(\tau)\hat{q}_n^\dagger(s,\tau), \tag{S2b}$$

where $\hat{\boldsymbol{\mathcal{E}}}_S$, $\hat{q}_n^\dagger$ and $\hat{f}_n^\dagger$ represent the Laplace transforms of $\hat{\mathbf{E}}_S$, $\hat{Q}_n^\dagger$ and $\hat{F}_n^\dagger$ respectively.

Substitution of Eq. S2b into Eq. S2a yields *coupled* ordinary differential equations (ODEs), rather than the single ODE as in [2]. In tensor form, this coupled ODE system is

$$\frac{\partial}{\partial\tau}\hat{\mathbf{q}}^\dagger(s,\tau) = (\mathbf{M} - \mathbf{I}\Gamma)\,\hat{\mathbf{q}}^\dagger(s,\tau) + \hat{\mathbf{f}}^\dagger(s,\tau), \tag{S3}$$

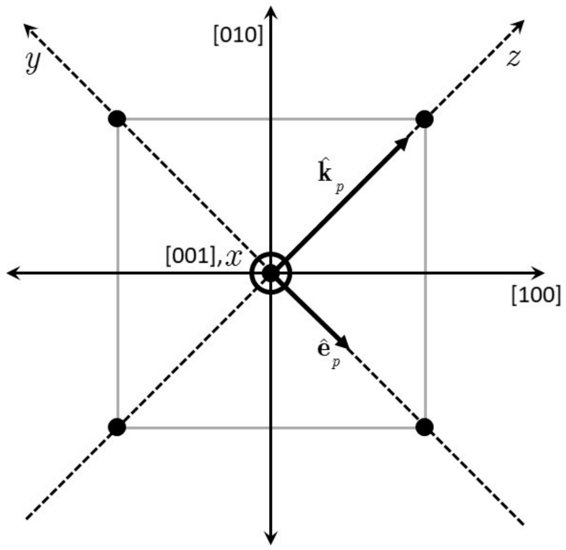

Fig. S1. Diagram illustrating the propagation direction $\hat{\mathbf{k}}_p$, and polarization $\hat{\mathbf{e}}_p$ with respect to the face-centered cubic axes of the diamond, and the rotated basis $\{x, y, z\}$. Positions of in-plane carbon atoms (filled circles) are depicted for reference.

where $\hat{\mathbf{q}}^\dagger = \{\hat{q}_1^\dagger, \hat{q}_2^\dagger, \hat{q}_3^\dagger\}$, $\hat{\mathbf{f}}^\dagger = \{\hat{f}_1^\dagger, \hat{f}_2^\dagger, \hat{f}_3^\dagger\}$, $\mathbf{I}$ is the identity matrix and

$$M_{ij} = \frac{1}{s} C \left(\mathbf{E}_p^* \boldsymbol{\kappa}_i\right) \left(\mathbf{E}_p^* \boldsymbol{\kappa}_j\right)^\dagger = \frac{1}{s} C \sum_k \sum_l \sum_m \kappa_{kli} \kappa_{kmj}^* E_{p,l}^* E_{p,m}, \tag{S4}$$

and where $\kappa_{ijk} = \kappa_{ij}$ for the k-th mode.

Eq. S3 has no analytic solution in the general case, however for certain high-symmetry crystal and pump configurations it reduces to a more tractable form. The polarizability tensors of the F_{2g} Raman modes with respect to the original coordinate basis of Eq. S1 are [4];

$$\begin{pmatrix} 0 & 0 & 0 \\ 0 & -d & 0 \\ 0 & 0 & d \end{pmatrix}, \begin{pmatrix} 0 & \frac{d}{\sqrt{2}} & \frac{-d}{\sqrt{2}} \\ \frac{d}{\sqrt{2}} & 0 & 0 \\ \frac{-d}{\sqrt{2}} & 0 & 0 \end{pmatrix}, \begin{pmatrix} 0 & \frac{-d}{\sqrt{2}} & \frac{-d}{\sqrt{2}} \\ \frac{-d}{\sqrt{2}} & 0 & 0 \\ \frac{-d}{\sqrt{2}} & 0 & 0 \end{pmatrix}. \tag{S5}$$

For a $[1\bar{1}0]$-polarized pump propagating in the $[110]$-direction, the tensor $\mathbf{M}$ becomes

$$\mathbf{M} = \frac{1}{s} C d^2 \left|E_p(\tau)\right|^2 \begin{pmatrix} 1 & 0 & 0 \\ 0 & \frac{1}{2} & -\frac{1}{2} \\ 0 & -\frac{1}{2} & \frac{1}{2} \end{pmatrix}, \tag{S6}$$

indicating that the equation for $\hat{q}_1$ becomes uncoupled from the other two ODEs, which remain coupled. The solutions for $\hat{q}_n$ are

$$\hat{q}_1^\dagger(s,\tau) = \int_0^\tau \tfrac{1}{2} e^{-\Gamma(\tau-\tau')} e^{a(\tau,\tau')/s} \hat{f}_1^\dagger(s,\tau') d\tau', \tag{S7a}$$

$$\begin{aligned} \hat{q}_{2,3}^\dagger(s,\tau) = \int_0^\tau & \tfrac{1}{2} e^{-\Gamma(\tau-\tau')} \left(1 + e^{a(\tau,\tau')/s}\right) \hat{f}_{2,3}^\dagger(s,\tau') \\ & + \tfrac{1}{2} e^{-\Gamma(\tau-\tau')} \left(1 - e^{a(\tau,\tau')/s}\right) \hat{f}_{3,2}^\dagger(s,\tau') d\tau', \end{aligned} \tag{S7b}$$

where

$$a(\tau,\tau') = C d^2 \int_{\tau'}^\tau \left|E_p(\tau'')\right|^2 d\tau''. \tag{S7c}$$

Inserting these solutions into Eq. S2b yields

$$\hat{\mathcal{E}}_S(s,\tau) = i\frac{Cd}{s}E_p(\tau)\int_0^{\tau} e^{-\Gamma(\tau-\tau')}e^{a(\tau,\tau')/s}\left[\hat{\mathbf{x}}\frac{1}{\sqrt{2}}\left(\hat{f}_2^{\dagger}(s,\tau')+\hat{f}_3^{\dagger}(s,\tau')\right)+\hat{\mathbf{y}}\hat{f}_1^{\dagger}(s,\tau')\right]d\tau', \quad \text{(S8)}$$

where $\hat{\mathbf{x}} = \{1,0,0\}$ and $\hat{\mathbf{y}} = \{0,1,0\}$ denote unit vectors in the x and y directions respectively. Finally, the inverse Laplace transform is taken to determine the Stokes field

$$\begin{aligned}\hat{\mathbf{E}}_S(z,\tau) = iCdE_p(\tau)\int_0^{\tau}\int_0^{z} H(z,z',\tau,\tau')\times \\ \left[\hat{\mathbf{x}}\frac{1}{\sqrt{2}}\left(\hat{F}_2^{\dagger}(z',\tau')+\hat{F}_3^{\dagger}(z',\tau')\right)+\hat{\mathbf{y}}\hat{F}_1^{\dagger}(z',\tau')\right]dz'd\tau',\end{aligned} \quad \text{(S9a)}$$

where

$$H(z,z',\tau,\tau') = e^{-\Gamma(\tau-\tau')}I_0\left(\sqrt{4(z-z')a(\tau,\tau')}\right), \quad \text{(S9b)}$$

and I_0 denotes the 0-th order modified Bessel function of the first kind [2]. Note that $\hat{\mathbf{E}}_S$ is perpendicular to the propagation (z) direction as required.

2. NIST-800-90B TEST SUITE OUTPUT

Prior to processing, measured Stokes polarization orientations were mapped to integers between 0-255. No other data pre-processing was performed before applying the test suite.

A. Test output

Read in file 130mW2.bin, 98695 bytes long.
Dataset: 98695 8-bit symbols.
Output symbol values: min = 18, max = 238

Table S1. Test Summary

Test	Status	Reference
Compression Test	Pass	S2
Over/Under Test	Pass	S3
Excursion Test	Pass	S4
Directional Runs Test	Pass	S5
Covariance Test	Pass	S6
Collision Test	Pass	S7
iid Shuffle Tests	Pass	
Chi-square Independence	Pass	
Chi-square Stability	Pass	
Compression Sanity Check	Pass	S8
Collision Sanity Check	Pass	

** Passed iid shuffle tests

Chi square independence
score = 5200.75, degrees of freedom = 5103, cut-off = 5420.88
** Passed chi-square independence test

Chi square stability
score = 1815.91, degrees of freedom = 1782 cut-off = 1972.19
** Passed chi-square stability test

IID = True

min-entropy = 6.67081

Collision sanity check...
Dividing dataset into 4-tuples
Check rule 1 - do three or more 4-tuples have the same value?...Pass
Check rule 2 - probability of number of collisions below cutoff; number of collisions = 0, cutoff = 2.82464...Pass

sanity check = PASS
time: (326.563 sec)

Table S2. Compression Test (Passed)

Scores	Ranks
10019	921
9994	788
9997	355
9988	501
9990	210
10005	321
10008	425
10009	526
9995	307
9999	517
	0

Table S3. Over/under Test (Passed)

Scores		Ranks	
12	4813	326	64
18	4907	964*	620
13	4894	501	535
14	4915	552	629
13	4948	501	830
12	4865	311	345
14	4828	530	102
15	4905	743	634
13	4924	501	775
14	4907	547	584
		1	0

Table S4. Excursion Test (Passed)

Scores	Ranks
4824.09	837
5313.39	927
4777.79	848
6278.13	981*
3924.94	604
1976.11	22*
4036.51	664
3242.06	380
4132.48	670
2793.2	244
	2

Table S5. Directional Runs Test (Passed)

Scores			Ranks		
6558	7	4916	498	606	342
6532	6	4931	298	501	665
6515	6	4909	160	501	161
6596	6	4904	817	501	73
6600	7	4904	847	621	53
6549	7	4926	435	617	558
6573	6	4908	638	501	170
6548	6	4924	450	501	521
6572	6	4908	648	501	166
6535	6	4930	328	501	660
			0	0	0

Table S6. Covariance Test (Passed)

Scores	Ranks
47.579	994*
8.15688	696
13.8632	783
-9.3806	304
-14.4233	228
13.1026	743
12.6675	775
15.029	780
-21.3057	116
-4.36311	413
	1

Table S7. Collision Test (Passed)

Scores			Ranks		
1	15.601	44	501	517	483
1	15.8024	48	501	805	766
1	15.2772	45	501	147	501
1	15.4467	50	501	408	875
1	15.3615	39	501	173	27*
1	15.8853	44	501	811	457
1	16.0104	48	501	901	746
1	15.2525	41	501	105	145
1	15.8902	45	501	811	501
1	15.4317	45	501	373	501
			0	0	1

Table S8. Compression Sanity Check

Dataset	Compressed Length	Cutoff	Status
1	80152	65834.2	Pass
2	79952	65834.2	Pass
3	79976	65834.2	Pass
4	79904	65834.2	Pass
5	79920	65834.2	Pass
6	80040	65834.2	Pass
7	80064	65834.2	Pass
8	80072	65834.2	Pass
9	79960	65834.2	Pass
10	79992	65834.2	Pass
	1		

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