Supporting Information for
Selective Phonon Stimulation Mechanism to Tune Thermal Transport

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S1. IR Active Phonon Modes in RDX

Numerous experimental studies have investigated and reported the absorption of IR radiation by the molecules in RDX \(^1\text{-}^8\). The fraction of radiation energy absorbed is often reported as transmittance or absorbance, or % transmission or % absorption. In this work, we assume that the percent of energy absorbed % Absorption = (100 - % transmission), or absorbance = (1-transmittance) while neglecting any reflectance (transmittance = 1 is the same as % transmission = 100). Based on the IR spectroscopy data in the literature, fifteen IR active modes in RDX spanning the complete phonon spectrum are identified for stimulation as shown in Table S1. The optical energy input \((E_{in,\phi_z})\), the energy absorbed \((E_{abs,\phi_z})\) and the corresponding increasing in
phonon population ($\Delta n_{\Phi_s}$) are shown in Table S2. An example of spectral profile of the optical pulse used for stimulation assumed in this work is shown in Figure S1.

Table S1. Fifteen IR active modes in RDX selected for stimulation, corresponding mode assignment and % absorption obtained from literature$^{1-8}$. Subscript $s$ denotes that these modes have been selected for stimulation. Superscripts: $w =$ wag, ro = rotation, b = bending, u = umbrella, st = stretching, t = twist, sc = scissoring, rc = rocking, ax = axial, eq = equatorial, fo = folding, as = asymmetric.

| IR active Mode | Frequency $\omega_{IR}$ (cm$^{-1}$) | Mode Assignment | Mode Energy, $E_{\Phi_s}$ (meV) | Equilibrium Occupation $n_{\Phi_s}$ | % Absorption $n_{\Phi_s}$ |
|----------------|-----------------------------------|-----------------|-------------------------------|----------------------------------|--------------------------|
| 22.74          | Translation                        | 2.82            | 57.1140                       | 19.54                           |
| 43.99          | Translation                        | 5.46            | 29.2848                       | 4.21                            |
| 71.25          | $w$(all)NO$_2$                     | 8.83            | 17.8920                       | 17.14                           |
| 83.38          | $ro$(all)NO$_2$                    | 10.34           | 15.2178                       | 33.08                           |
| 190.87         | NC$_2$                            | 23.67           | 6.3760                        | 23.81                           |
| 405.03         | b+60/Ring 9–11                    | 50.22           | 2.7603                        | 28.32                           |
| 503.51         | b+75/Ring 9–11                    | 62.43           | 2.1339                        | 27.94                           |
| 582.36         | b+75/Ring 9–11                    | 72.21           | 1.7866                        | 64.80                           |
| 786.90         | $w$C-N+ $sc$(eq)NO$_2$            | 97.57           | 1.2147                        | 50.67                           |
| 914.84         | $st$(eq)N-N+ $ro$CH$_2$           | 113.43          | 0.9898                        | 30.18                           |
| 1140.67        | $st$(eq)CH$_2$                    | 141.44          | 0.7202                        | 51.33                           |
| 1299.72        | $st$(eq,ax)N-N+ $ro$CH$_2$        | 161.16          | 0.5893                        | 36.99                           |
| 1501.31        | $st$(ax)NO$_2$                    | 186.15          | 0.4661                        | 20.69                           |
| 1739.89        |                                | 215.74          | 0.3605                        | 70.97                           |
| 2823.76        | $st$(ax)CH$_2$                    | 350.13          | 0.1310                        | 23.51                           |
Table S2. Optical energy input ($E_{in,\Phi_s}$), energy absorbed ($E_{abs,\Phi_s}$) and corresponding increase in phonon population ($\Delta n_{\Phi_s}$) of the 15 IR active modes in RDX selected for stimulation.

| IR active Mode Frequency $\omega_{IR}$ (cm$^{-1}$) | $E_{in,\Phi_s}$ (eV) | $E_{abs,\Phi_s}$ (eV) | $\Delta n_{\Phi_s}$ |
|---|---|---|---|
| 22.74 | 1.00 | 0.20 | 69.33 |
| 43.99 | 1.94 | 0.08 | 14.94 |
| 71.25 | 3.13 | 0.54 | 60.81 |
| 83.38 | 3.67 | 1.21 | 117.36 |
| 190.87 | 8.40 | 2.00 | 84.46 |
| 405.03 | 17.82 | 5.05 | 100.46 |
| 503.51 | 22.15 | 6.19 | 99.10 |
| 582.36 | 25.62 | 16.60 | 229.87 |
| 786.90 | 34.61 | 17.54 | 179.76 |
| 914.84 | 40.24 | 12.14 | 107.06 |
| 1140.67 | 50.17 | 25.76 | 182.10 |
| 1299.72 | 57.17 | 21.15 | 131.22 |
| 1501.31 | 66.03 | 13.66 | 73.40 |
| 1739.89 | 76.53 | 54.31 | 251.75 |
| 2823.76 | 124.20 | 29.20 | 83.38 |

Figure S1. Example of spectral profile of the optical pulse used for stimulation assumed in this work (centered at IR active mode frequency with a linewidth of 1 cm$^{-1}$). 1 eV optical energy input is used for stimulating the band at 22.74 cm$^{-1}$ and the energy input is assumed constant across the frequency band.
S2. Fermi’s Golden Rule based 3-phonon Scattering Rate

The crystal Hamiltonian can be written as

\[ H = H_0 + H_3 + H_4 \ldots \]  \hspace{1cm} \text{(S1)}

where \( H_0 \) is the harmonic term, \( H_3 \) and \( H_4 \) are anharmonic terms also referred to as first and second order perturbation terms, respectively. In this work, we only consider up to the first order perturbation term of the Hamiltonian. The first order perturbation term \( H_3 \) is defined as\(^{14,15}\)

\[ H_3 = H_{\phi_1,\phi_2,\phi_3}^{(3)}(a_{\phi_1}^\dagger + a_{\phi_1})(a_{\phi_2}^\dagger + a_{\phi_2})(a_{\phi_3}^\dagger + a_{\phi_3}) \]  \hspace{1cm} \text{(S2)}

where \( \phi \) is the phonon mode index (\( \phi_1,\phi_2,\phi_3 \) are the mode indices of the three phonons involved in scattering, \( \phi \) refers to a mode corresponding to a negative wavevector), \( H_{\phi_1,\phi_2,\phi_3}^{(3)} \) are Fourier transforms of the third order Interatomic Force Constants (IFCs), \( a_{\phi}^\dagger \) and \( a_{\phi} \) are creation and annihilation operators respectively with \( a_{\phi}^\dagger \left| n_{\phi} \right> = \sqrt{n_{\phi} + 1} \left| n_{\phi} + 1 \right> \) and \( a_{\phi} \left| n_{\phi} \right> = \sqrt{n_{\phi}} \left| n_{\phi} - 1 \right> \), and \( n_{\phi} \) is the phonon mode population given by Bose-Einstein (BE) statistics. The coefficients \( H_{\phi_1,\phi_2,\phi_3}^{(3)} \) are related to the analogous coefficients \( V_{\phi_1,\phi_2,\phi_3}^{(3)} \) defined by Born and Huang\(^{16}\). They are related by
\[ H_{\phi_1, \phi_2, \phi_3}^{(3)} = \frac{\hbar^2}{6N^2} \sum_{\alpha_1, \alpha_2, \alpha_3} \sum_{b_1, b_2, b_3} \sum_{l, l_2, l_3} \Phi_{\alpha_1, \alpha_2, \alpha_3}^{b_1, b_2, b_3} e^{i k_{l_2} r_{l_2}} e^{i k_{l_3} r_{l_3}} \left( m_b, m_{b_2}, m_{b_3} \right) \]  

where \( N \) is the total number of k points, \( \omega_{\phi} \) is the angular frequency of the phonon mode, Kronecker delta \( \Delta_{k_1 + k_2 + k_3} \) enforces momentum conservation, and \( V_{\phi_1, \phi_2, \phi_3}^{(3)} \) is the cubic anharmonic matrix of third order IFCs defined as

\[ V_{\phi_1, \phi_2, \phi_3}^{(3)} = \sum_{\alpha_1, \alpha_2, \alpha_3} \sum_{b_1, b_2, b_3} \sum_{l, l_2, l_3} \Phi_{\alpha_1, \alpha_2, \alpha_3}^{b_1, b_2, b_3} e^{i k_{l_2} r_{l_2}} e^{i k_{l_3} r_{l_3}} \left( m_b, m_{b_2}, m_{b_3} \right) \]  

where \( b \) is the index of atoms in the unitcell, \( l \) is the index of cells in a supercell, \( \alpha \) represents the three Cartesian directions, \( m_b \) is the mass of the \( b^{th} \) atom, \( \Phi_{\alpha_1, \alpha_2, \alpha_3}^{b_1, b_2, b_3} \) are the third order force constants, \( e^{\phi_{\alpha_1}} \) are the phonon mode eigenvectors, and \( \mathbf{k} \) represents the wavevector.

With the above expression for the anharmonic Hamiltonian, Maradudin and Fein formulated a method to calculate the intrinsic phonon scattering rates using the perturbation theory (Fermi’s Golden Rule or FGR)\(^{14} \). Based on FGR, the probability of transition from an initial state \( |i\rangle \) to a final state \( |f\rangle \) for the 3-phonon emission process \( \phi_1 \rightarrow \phi_2 + \phi_3 \) is given by

\[ \frac{2\pi}{\hbar} |\langle f | H_3 | i \rangle|^2 \delta(E_i - E_f) \sim n_{\phi_1}(1 + n_{\phi_2})(1 + n_{\phi_3}) |H_{\phi_1, \phi_2, \phi_3}^{(3)}|^2 \]  

(S5)
Similarly, the transition probability for the process $\phi_1 \leftrightarrow \phi_2 + \phi_3$ is given by

$$\frac{2\pi}{\hbar} |\langle i | H_3 | f \rangle|^2 \delta(E_i - E_f) \sim (1 + n_{\phi_1})n_{\phi_2}n_{\phi_3}|H_{\phi_1,\phi_2,\phi_3}^{(3)}|^2$$

(S6)

where $E_i$ and $E_f$ are energy of the initial and the final state respectively. The transition probabilities for the absorption processes $\phi_1 + \phi_2 \rightarrow \phi_3$ and $\phi_1 + \phi_2 \leftarrow \phi_3$ can be expressed in a similar manner.

The rate of change of occupation of the mode $\phi_1$ can be calculated based on the transition probabilities for the 3-phonon processes as

$$\frac{\partial n_{\phi_1}}{\partial t} = - \sum_{\phi_2,\phi_3} \left\{ \frac{1}{2} n_{\phi_1}(1 + n_{\phi_2})(1 + n_{\phi_3}) - (1 + n_{\phi_1})n_{\phi_2}n_{\phi_3} \right\} L -
+ \left[ (1 + n_{\phi_3})n_{\phi_1}n_{\phi_2} - n_{\phi_3}(1 + n_{\phi_1})(1 + n_{\phi_2}) \right] L^+$$

(S7)

The first two terms of the summation in Eq. (S7) account for the emission process (the difference between $\phi_1 \rightarrow \phi_2 + \phi_3$ and $\phi_1 \leftarrow \phi_2 + \phi_3$), and the last two terms account for the absorption process (the difference between $\phi_1 + \phi_2 \rightarrow \phi_3$ and $\phi_1 + \phi_2 \leftarrow \phi_3$). Using the single mode relaxation time approximation (SMRTA), which assumes that the non-equilibrium population of any mode is calculated independently of other phonon modes i.e., $n_{\phi_1} = n_{\phi_1}^0 + n'_{\phi_1}$ and $n_{\phi_2} = n_{\phi_2}^0$, $n_{\phi_3} = n_{\phi_3}^0$,

Eq. (S7) can be reduced to
\[ \frac{\partial n_{\phi_1}}{\partial t} = - n'_\phi \sum_{\phi_2, \phi_3} \left\{ \frac{1}{2} \left( 1 + n_{\phi_2}^0 + n_{\phi_3}^0 \right) L_- + \left( n_{\phi_2}^0 - n_{\phi_3}^0 \right) L_+ \right\} \]  

(S8)

where \( n'_\phi \) is the perturbation in population of the mode \( \phi_1 \), \( n_\phi^0 \) is the equilibrium phonon population of the mode \( \phi \) given by BE statistics, and the summation on the right side is the intrinsic 3-phonon scattering rate \( \Gamma_{\phi_i} \) for mode \( \phi_1 \).

\[ \Gamma_{\phi_1} = \sum_{\phi_2, \phi_3} \left\{ \frac{1}{2} \left( 1 + n_{\phi_2}^0 + n_{\phi_3}^0 \right) L_- + \left( n_{\phi_2}^0 - n_{\phi_3}^0 \right) L_+ \right\} \]  

(S9)

where \( L_\pm \) accounts for the conservation of crystal momentum and energy and the probability of transition from an initial state to a final state for absorption (\( - \)) and emission (\( + \)) processes.

The expression for \( L_\pm \) is given by FGR as\(^{17} \)

\[ L_\pm = \frac{\pi \hbar}{4N} \left| V^{(3)}_\pm \right|^2 \Delta \pm \frac{\delta(\omega_{\phi_1} \pm \omega_{\phi_2} - \omega_{\phi_3})}{\omega_{\phi_1} \omega_{\phi_2} \omega_{\phi_3}} \]  

(S10)

where \( V^{(3)}_\pm \) is cubic anharmonic matrix of the third order IFCs,

\[ V^{(3)}_\pm = \sum_{\alpha_1, \alpha_2, \alpha_3} \sum_{b_1, b_2, b_3} \frac{\phi_{\alpha_1, \alpha_2, \alpha_3} e^{i k_1 b_1 \alpha_1} e^{i k_2 b_2 \alpha_2} e^{i k_3 b_3 \alpha_3}}{m_{\phi_1} m_{\phi_2} m_{\phi_3}} e^{\pm ik_{\pm} r_{\pm}} e^{-i k_{\perp} r_{\perp}} \]  

(S11)

The scattering rate \( \Gamma_{\phi_i} \) can be considered to be a product of two terms, \( \left| V^{(3)}_\pm \right|^2 \) which indicates the anharmonicity of the modes and a 3-phonon phase space volume \( P_{3,\phi_1} \) calculated as
\[ P_{3, \Phi_1} = \sum_{\phi_2, \phi_3} \left\{ \frac{1}{2} \left( 1 + n_{\phi_2}^0 + n_{\phi_3}^0 \right) \Delta - \frac{\delta(\omega_{\phi_1} - \omega_{\phi_2} - \omega_{\phi_3})}{\omega_{\phi_1} \omega_{\phi_2} \omega_{\phi_3}} + (n_{\phi_2}^0 - n_{\phi_3}^0) \Delta + \frac{\delta(\omega_{\phi_1}^S - \omega_{\phi_2} - \omega_{\phi_3})}{\omega_{\phi_1} \omega_{\phi_2} \omega_{\phi_3}} \right\} \] (S12)

which indicates the number of allowed three phonon scattering events that follow conservation of crystal momentum and energy.

When phonons in band \( \Phi_S \) are stimulated, the population of stimulated phonons is driven out of equilibrium and the scattering rate in Eq. (S9) can be re-written by replacing \( n_\phi^0 \) with \( n_\phi^S \) for the stimulated modes

\[ \Gamma_{\phi_1}^{\Phi_S} = \sum_{\phi_2 \notin \Phi_S, \phi_3 \notin \Phi_S} \left\{ \frac{1}{2} \left( 1 + n_{\phi_2}^0 + n_{\phi_3}^0 \right) L_- + (n_{\phi_2}^0 - n_{\phi_3}^0) L_+ \right\} + \sum_{\phi_2 \notin \Phi_S, \phi_3 \notin \Phi_S} \left\{ \frac{1}{2} \left( 1 + n_{\phi_2}^0 + n_{\phi_3}^S \right) L_- + (n_{\phi_2}^0 - n_{\phi_3}^S) L_+ \right\} + \sum_{\phi_2 \notin \Phi_S, \phi_3 \notin \Phi_S} \left\{ \frac{1}{2} \left( 1 + n_{\phi_2}^S + n_{\phi_3}^0 \right) L_- + (n_{\phi_2}^S - n_{\phi_3}^0) L_+ \right\} + \sum_{\phi_2 \notin \Phi_S, \phi_3 \notin \Phi_S} \left\{ \frac{1}{2} \left( 1 + n_{\phi_2}^S + n_{\phi_3}^S \right) L_- + (n_{\phi_2}^S - n_{\phi_3}^S) L_+ \right\} \right\} \] (S13)

where \( n_\phi^S = n_\phi^0 + \Delta n_{\Phi_S} \). The resulting change in scattering rate can be calculated as Eq. (S13) - Eq. (S9)
\[ \Delta \tau_{\phi_1}^{\phi_1} = \tau_{\phi_1}^{\phi_1} - \tau_{\phi_1} = \sum_{\phi_2 \in \Phi, \phi_3 \in \Phi_3} \left\{ \left( \frac{L_2 - L_1}{2} - L_1 \right) (n_{\phi_3}^e - n_{\phi_3}^0) \right\} + \sum_{\phi_2 \in \Phi_3, \phi_3 \in \Phi_3} \left\{ \left( \frac{L_2 - L_1}{2} - L_1 \right) (n_{\phi_3}^e - n_{\phi_3}^0) \right\} - n_{\phi_3}^0 + \left( \frac{L_2 - L_1}{2} + L_1 \right) (n_{\phi_2}^e - n_{\phi_2}^0), \quad (S14) \]

Next, the phonon lifetimes are calculated as

\[ \tau_{\phi_1}^{\phi_1} = \left| \frac{1}{\Delta \tau_{\phi_1}^{\phi_1}} \right|, \quad (S15) \]

and the phonon mean free paths are calculated as

\[ \Lambda_{\phi_1}^{\phi_1} = \left| v_{g,\phi_1} \right| \tau_{\phi_1}, \quad (S16) \]

where \( v_{g,\phi_1} \) is the phonon mode group velocity. Subsequently, the modewise diffusivity is calculated as

\[ D_{\phi_1}^{\phi_1} = \left| v_{g,\phi_1} \right| \Lambda_{\phi_1}^{\phi_1}, \quad (S17) \]

the scalar thermal conductivity is calculated as

\[ \kappa_s^{\phi_1} = \frac{1}{3} \sum_{\phi_1} C_{v,\phi_1} \left| v_{g,\phi_1} \right| \Lambda_{\phi_1}^{\phi_1}, \quad (S18) \]

where \( C_{v,\phi_1} \) is the phonon mode specific heat.

**S3. Measures for Quantifying Effects of Phonon Band Stimulation**
We quantify the effects of band stimulation by calculating average values of percent change in modewise phonon properties. Specifically, the average percent change in scattering rates (average over all modes) when phonons in band \( \Phi_s \) are stimulated is calculated as

\[
\% \Delta \Gamma^{\text{avg}, \Phi_s} = \frac{1}{\text{no. of modes}} \sum_{\phi_1} 100 \frac{\Gamma_{\phi_1}^{\Phi_s} - \Gamma_{\phi_1}}{\Gamma_{\phi_1}}, \tag{S19}
\]

Similarly, the average percent change in modewise diffusivity is calculated as

\[
\% \Delta D^{\text{avg}, \Phi_s} = \frac{1}{\text{no. of modes}} \sum_{\phi_1} 100 \frac{D_{\phi_1}^{\Phi_s} - D_{\phi_1}}{D_{\phi_1}}, \tag{S20}
\]

and the percent change in scalar thermal conductivity is calculated as

\[
\% \Delta \kappa_s^{\Phi_s} = 100 \frac{\kappa_{\phi_1}^{\Phi_s} - \kappa_s}{\kappa_s}, \tag{S21}
\]

**S4. Modewise Percent Contribution to Thermal Conductivity**

For each mode \( \phi_1 \), the modewise percent contribution to thermal conductivity is defined as

\[
\kappa_{\phi_1} = 100 \frac{\kappa_{\phi_1}}{\kappa_s}
\]

where \( \kappa_{\phi_1} \) is the modewise thermal conductivity defined as

\[
\kappa_{\phi_1}^{\text{PGM}} = \frac{1}{3} (C_{\phi_1} | \mathbf{v}_{g, \phi_1} | \Lambda_{\phi_1})
\]

for the Phonon Gas Model and

\[
\kappa_{\phi_1}^{\text{AF}} = (C_{\phi_1} D_{\phi_1})
\]

for the Allen-Feldman model. Here \( C_{\phi_1}, \mathbf{v}_{g, \phi_1}, \Lambda_{\phi_1} \),
and $D_{\phi_i}$ are the modewise specific heat, group velocity, phonon mean free path and diffusivity respectively.

![Figure S2](image_url)

**Figure S2.** Modewise percent contribution to thermal conductivity for PGM and AF models. Although, PGM treats all phonons as propagating carriers while neglecting the diffusive nature of transport, the modewise percent contribution of the phonons to thermal conductivity is similar for both PGM (propagating carriers) and Allen-Feldman model (diffusive carriers).

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