Supplementary Materials

Sodium Diffusion and Dynamics in $Na_2Ti_3O_7$: Neutron Scattering and Ab-initio Simulations

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Table S1 The fitted lattice parameters and atomic positions as obtained from the Rietveld refinement of the structural model for $Na_2Ti_3O_7$. The available experimental data [1] from literature are also given.

	Our data a = 3.8008(1) Å b = 8.5604(2) Å c = 9.1252(2) Å α = 101.598(2) Å V = 290.84(1) Å Z = 2 Space Group: $P2_1/m$			From Ref. [1] a = 3.806 Å b= 8.566 Å c = 9.133 Å α = 101.57° V = 291.71 Å³ Z = 2 Space Group: $P2_1/m$		
	x	y	z	x	y	z
Ti1	0.25	0.1470(4)	0.9844(4)	0.25	0.1461	0.9850
Ti2	0.25	0.2533(4)	0.6755(4)	0.25	0.2489	0.6759
Ti3	0.25	0.0288(4)	0.2808(4)	0.25	0.0286	0.2803
Na1	0.25	0.6730(9)	0.5944(9)	0.25	0.683	0.5929
Na2	0.25	0.4959(10)	0.1565(10)	0.25	0.4999	0.1553
O1	0.25	0.2333(8)	0.1853(7)	0.25	0.2188	0.1865
O2	0.25	0.1594(12)	0.4573(7)	0.25	0.1465	0.4612
O3	0.25	0.4476(9)	0.6485(14)	0.25	0.4413	0.6510
O4	0.25	0.3365(8)	0.9072(7)	0.25	0.3246	0.9081
O5	0.25	0.0177(8)	0.7483(7)	0.25	0.0161	0.7532
O6	0.25	0.7822(9)	0.3237(13)	0.25	0.8035	0.3182
O7	0.25	0.9070(8)	0.0381(8)	0.25	0.9152	0.0440

Rp: 15.4 %, Rwp: 20.3 %, χ^2: 2.09
Bragg R-factor: 11.3 %, Rf-factor= 6.30 %

[1] O. V. Yakubovich and V. V. Kireev, Refinement of the crystal structure of $Na_2Ti_3O_7$, Crystallography Reports 48 (2003) 24-28

FIG. S1 Powder XRD data recorded for $Na_2Ti_3O_7$ using Cu Kα radiation. Rietveld refinement of the structural model (Space Group: $P2_1/m$). The R-factors and χ^2 values of the fit are given in Table S1.
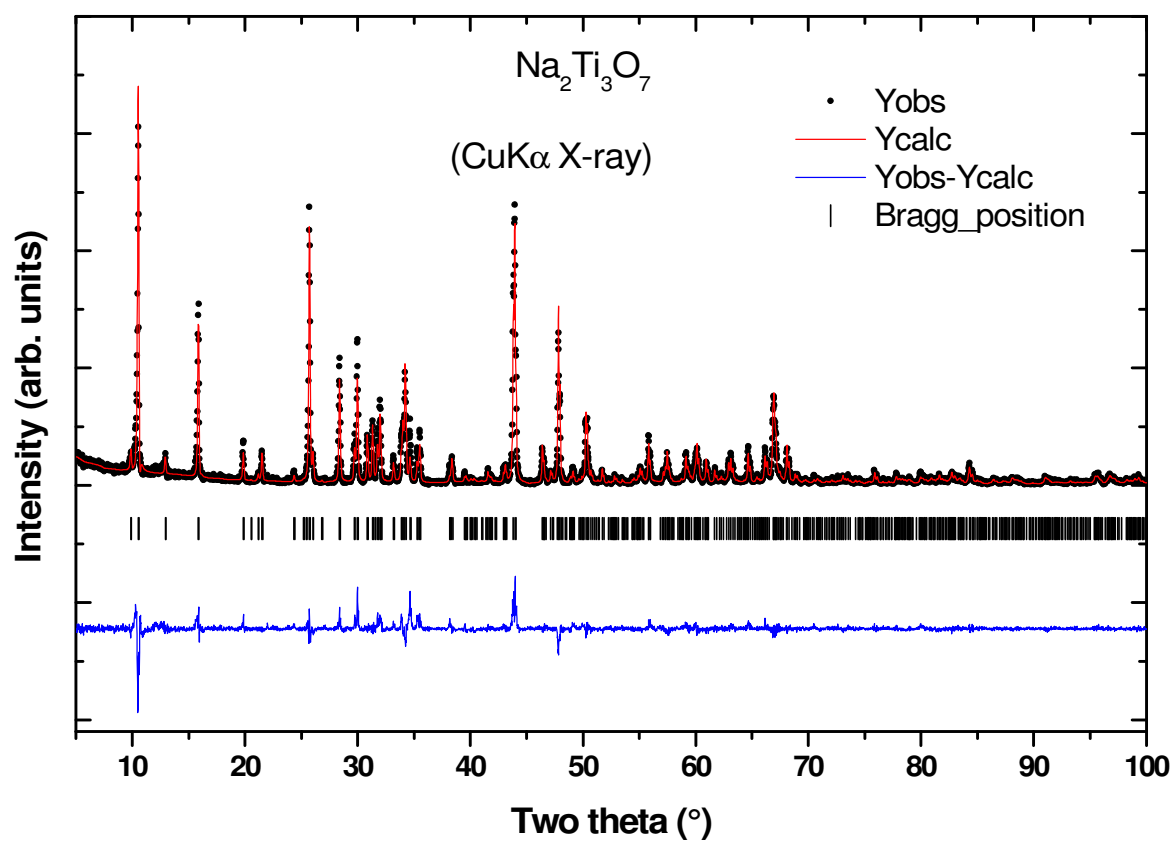
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FIG S2 (Color online) The measured QENS spectra of Na$_2$Ti$_3$O$_7$ at 1173 K along with the instrumental resolution (vanadium at 300 K).
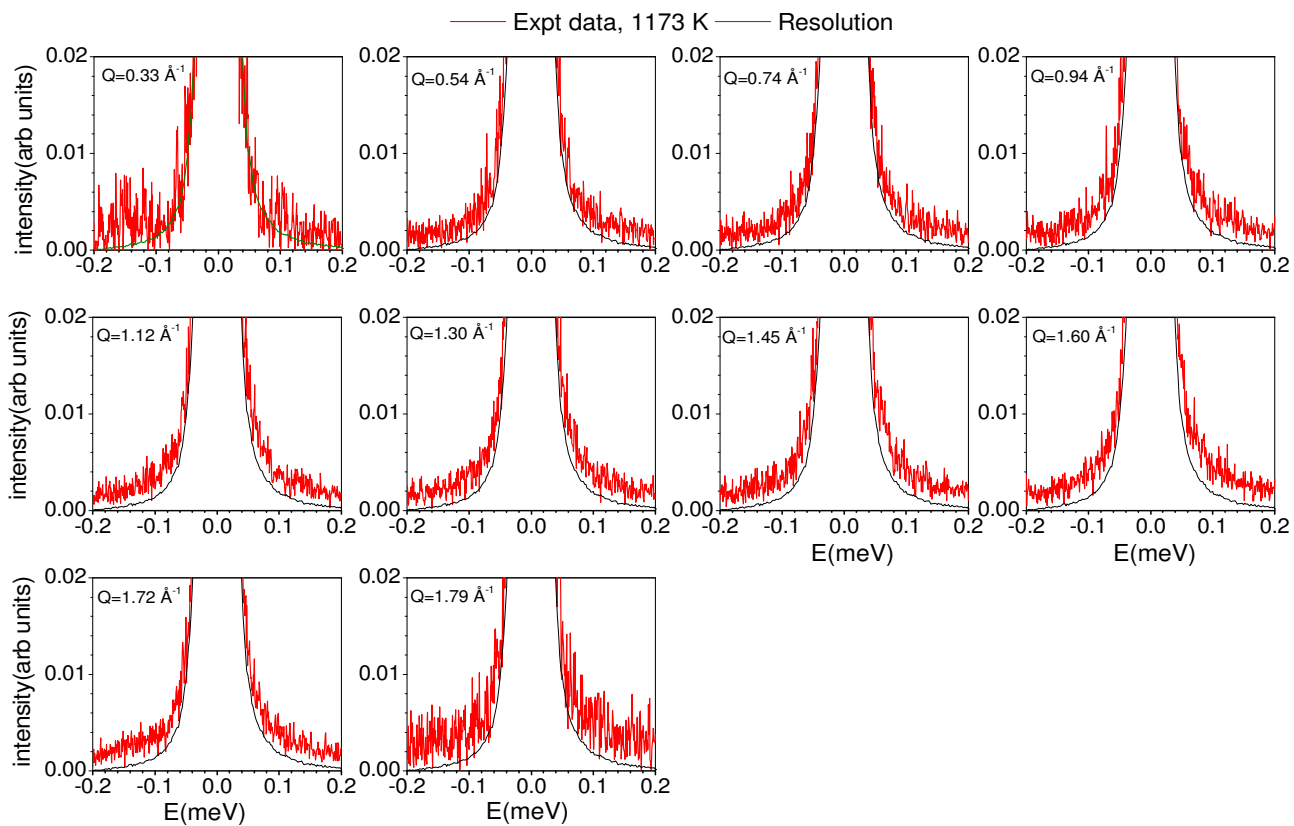
FIG S3 (Color online) Observed dynamical neutron scattering function S(Q, E) of $Na_2Ti_3O_7$ integrated over all wave-vector transfers (Q) at various temperatures. The experimental S(Q, E) data at various temperatures have not been normalized to unity at E=0.
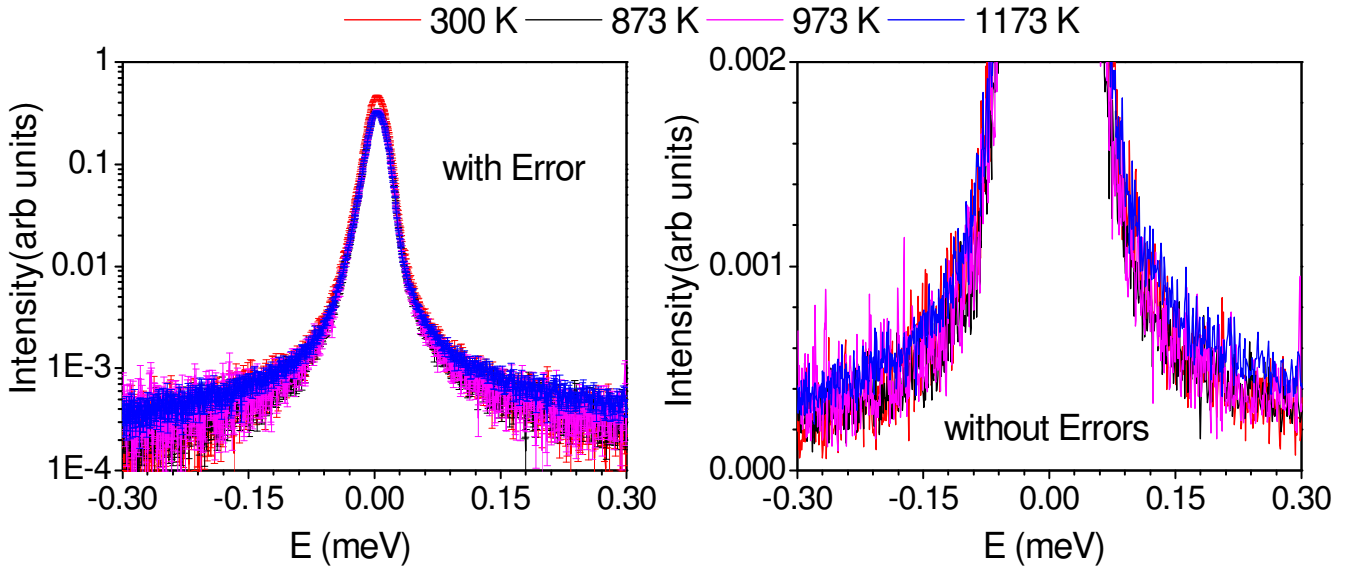
FIG S4 (Color online) The Q integrated S(Q,E) spectra at 973 K and 1173 K. Please note that at 873 K, the statistics are still very poor and do not allow us to estimate the FWHM. We found the estimated FWHM at 973 K and 1173 K is ~5 and ~121 μeV, respectively. However, the QENS broadening at 973 K is very small and may not be reliable due to the poor statistics of the data. Hence we feel that significant QENS broadening is seen only at 1173 K.
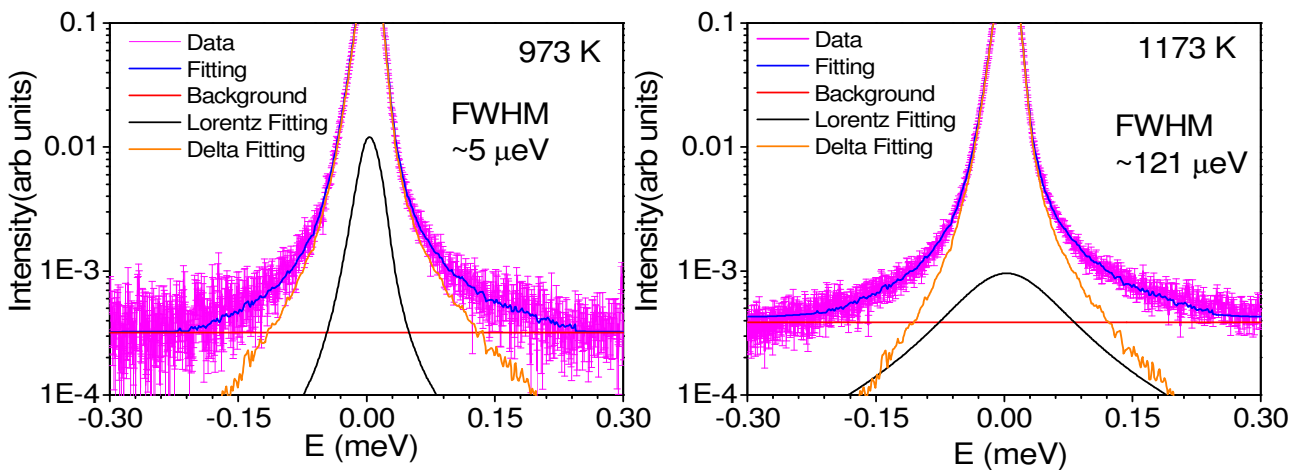
FIG S5 (Color online) The calculated incoherent dynamical structure factor of Na (black) and total coherent (red) dynamical structure factor using AIMD trajectories with one Na vacancy in a (3×2×2) supercell at 1800 K. The incoherent contribution of Na dynamics dominates over the total-coherent scattering. The incoherent neutron scattering cross-section of the Ti and O atoms is negligible compared to that of the Na atom.
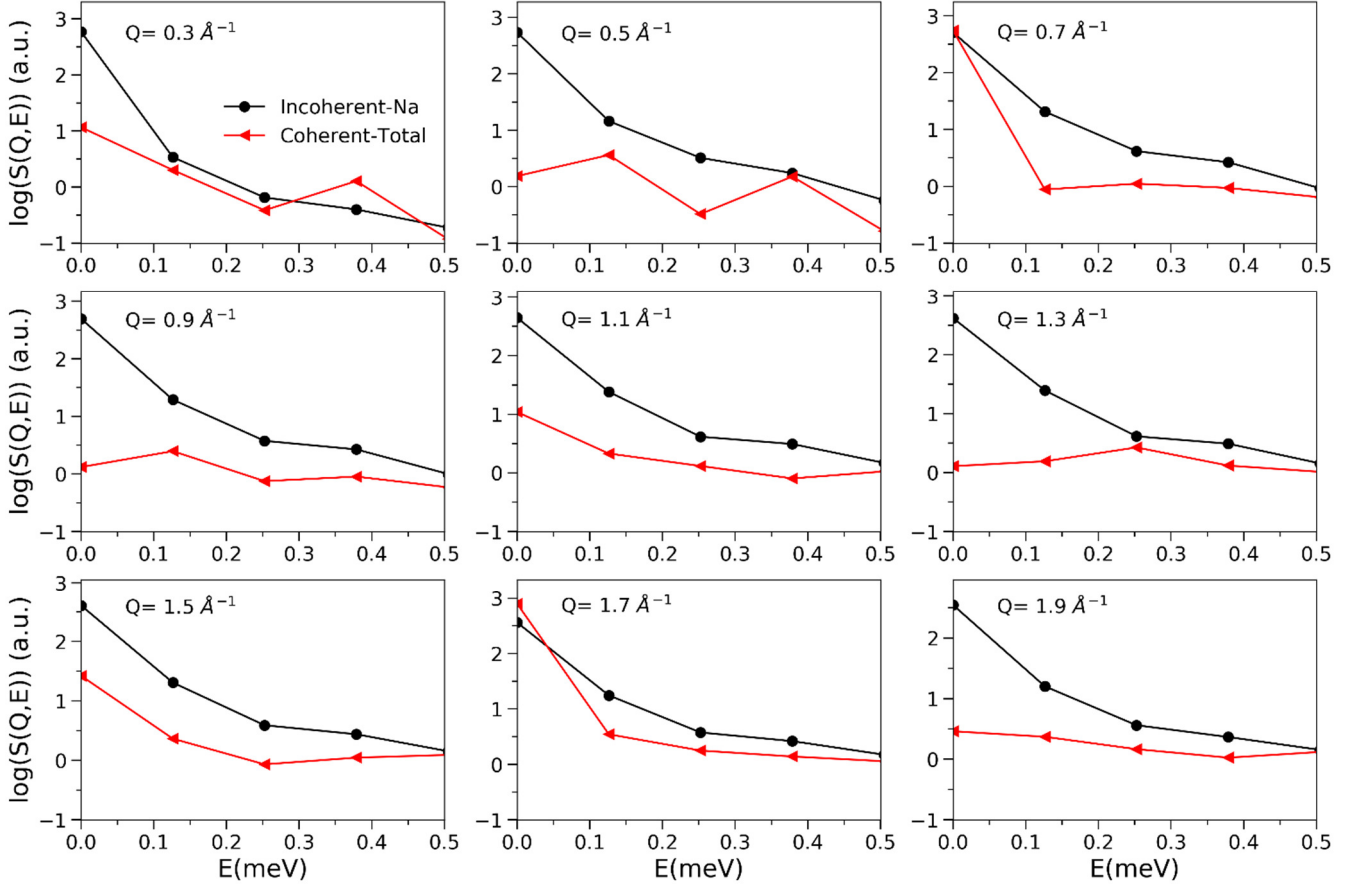
FIG S6 (Color online) The calculated neutron-weighted coherent, incoherent, and total S(Q) in $Na_2Ti_3O_7$ with one Na vacancy in a (3×2×2) supercell at 1800 K (solid-lines), compared with the measured intensity from QENS measurements at 1173 K. The incoherent scattering is mainly from the Na atom since the incoherent cross-section of Ti and O is much smaller. The weak Bragg reflections in the calculated coherent scattering are not visible in the experiments.
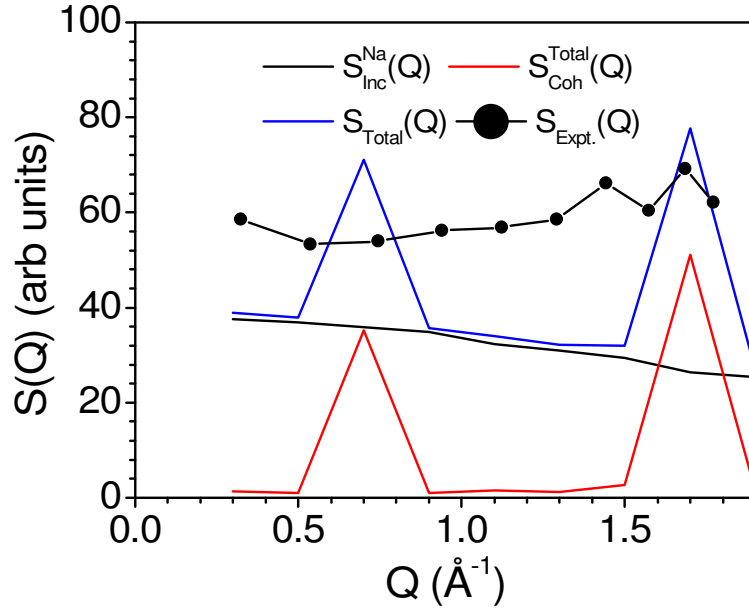

FIG S7 (Color online) The calculated MSD components for Na atoms in $Na_2Ti_3O_7$ with one Na vacancy in a (3×2×2) supercell along the Cartesian x, y, and z-axis. The x- and y-axis are along the a-and b-axis, respectively, of the monoclinic unit cell.
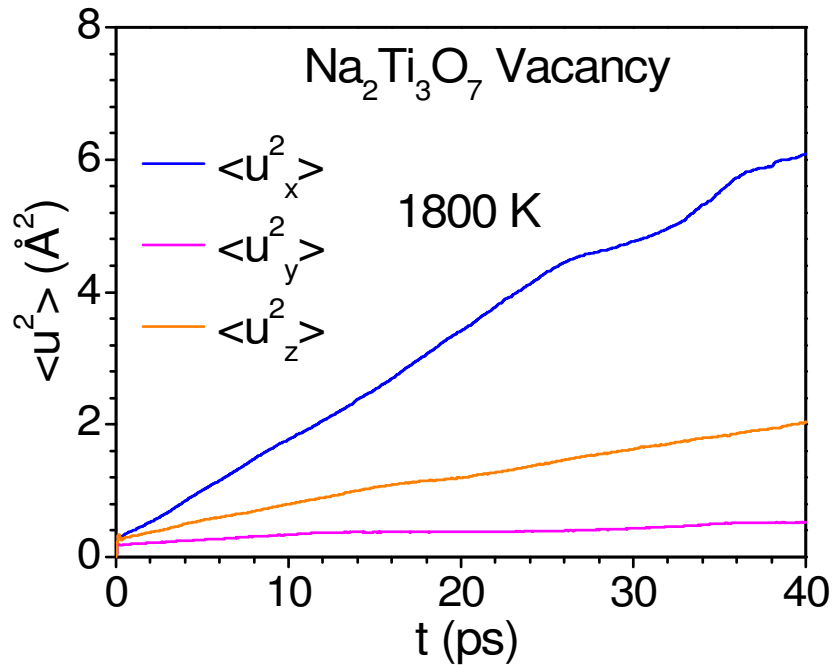
FIG S8 (Color online) The calculated incoherent S(Q, E) of Na from AIMD trajectory with one Na vacancy in a (3×2×2) supercell at 1800 K (solid black circles) and estimated HWHM (Γ) obtained from single Lorentzian fits (dotted lines).
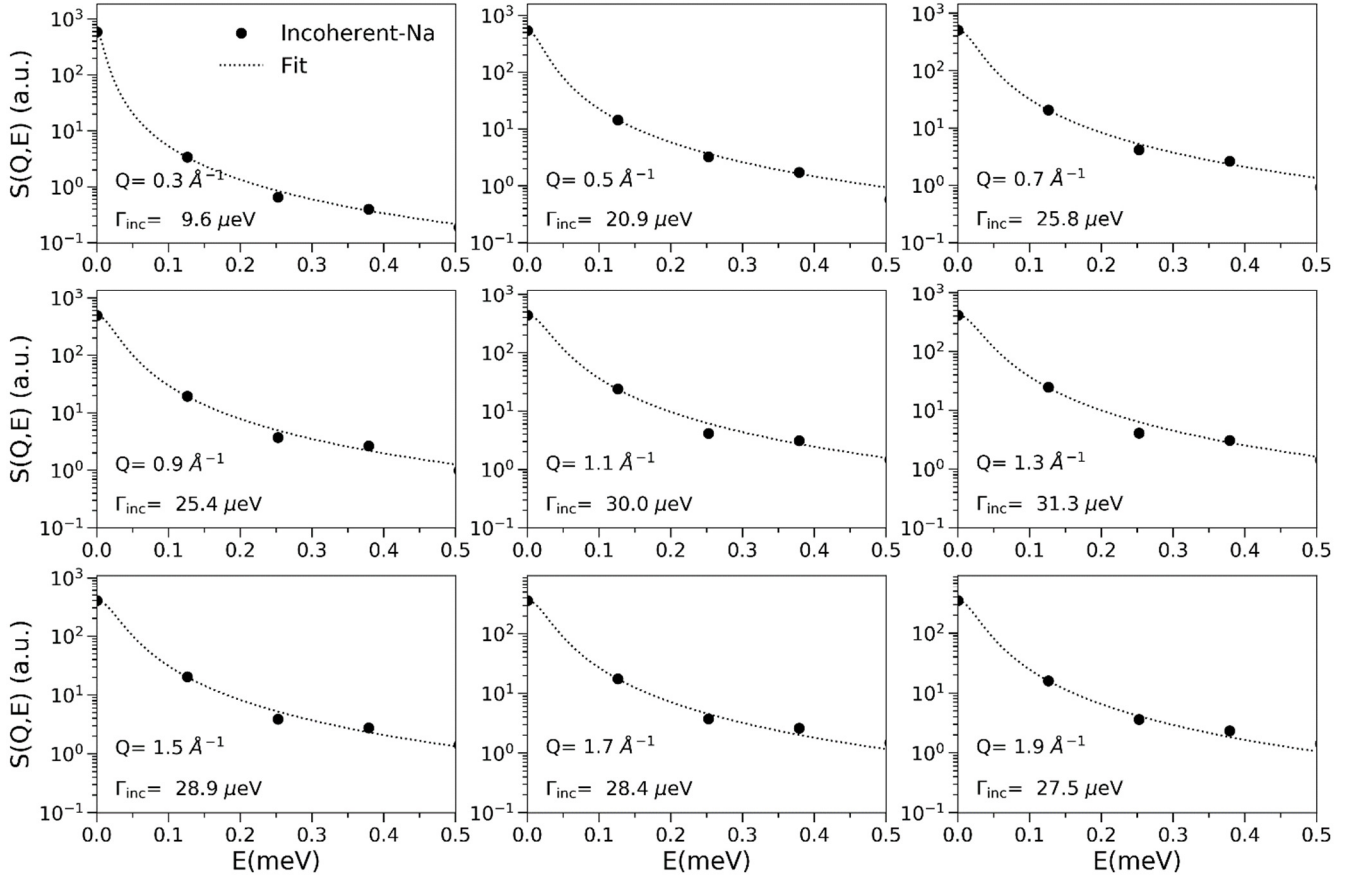
FIG S9 (Color online) The calculated Q-dependence of half-width at half maximum (HWHM) of Lorentzian obtained from simulated Na incoherent $S(Q, E)$ for $Na_2Ti_3O_7$ with one Na vacancy in a $(3\times2\times2)$ supercell at 1800 K.