MOLECULAR DYNAMICS SIMULATIONS OF EXAFS IN GERMANIUM

J. Timoshenko, A. Kuzmin, J. Purans

Institute of Solid State Physics, University of Latvia, Riga, Latvia

E-mail: timoshenkojanis@inbox.lv

Abstract

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$$f_A(r) = -B\exp(-\lambda_2 r)$$

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$$f_C(r) = \begin{cases} 1, & r < R - D \\ \frac{1}{2} - \frac{1}{2}\sin\left(\frac{\pi}{2}\frac{(r-R)}{D}\right), & R - D < r < R + D \\ 0, r > R + D \end{cases}$$

$$b_{ij} = \left(1 + \beta^n \zeta_{ij}^n\right)^{-1/2n}$$

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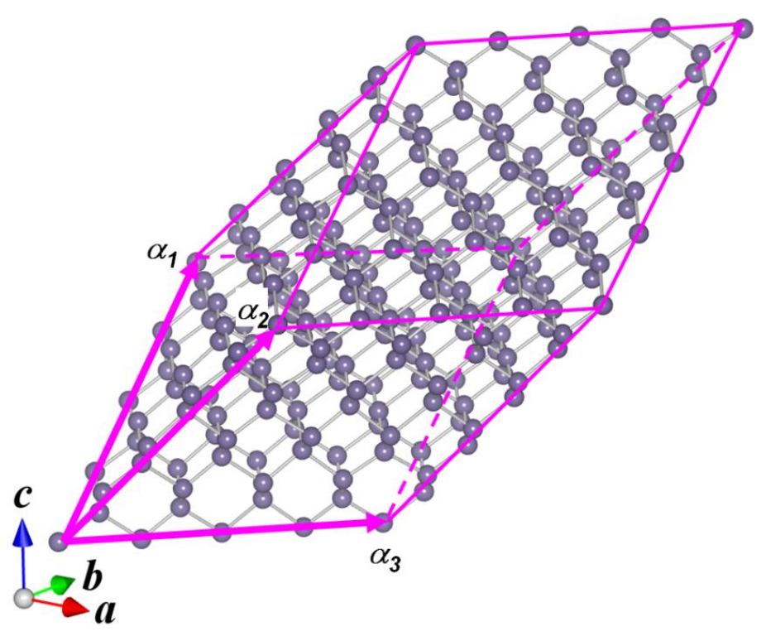

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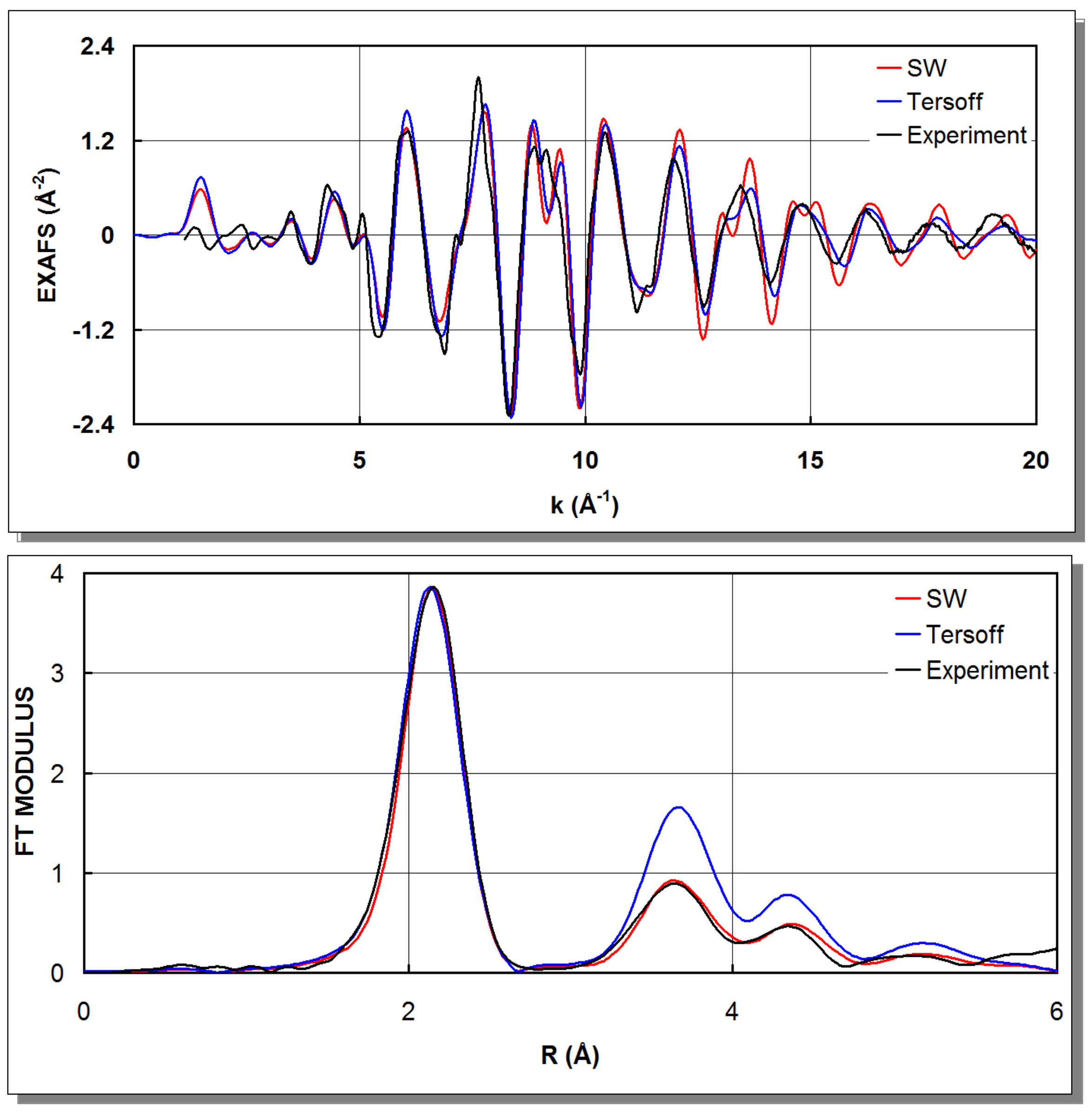

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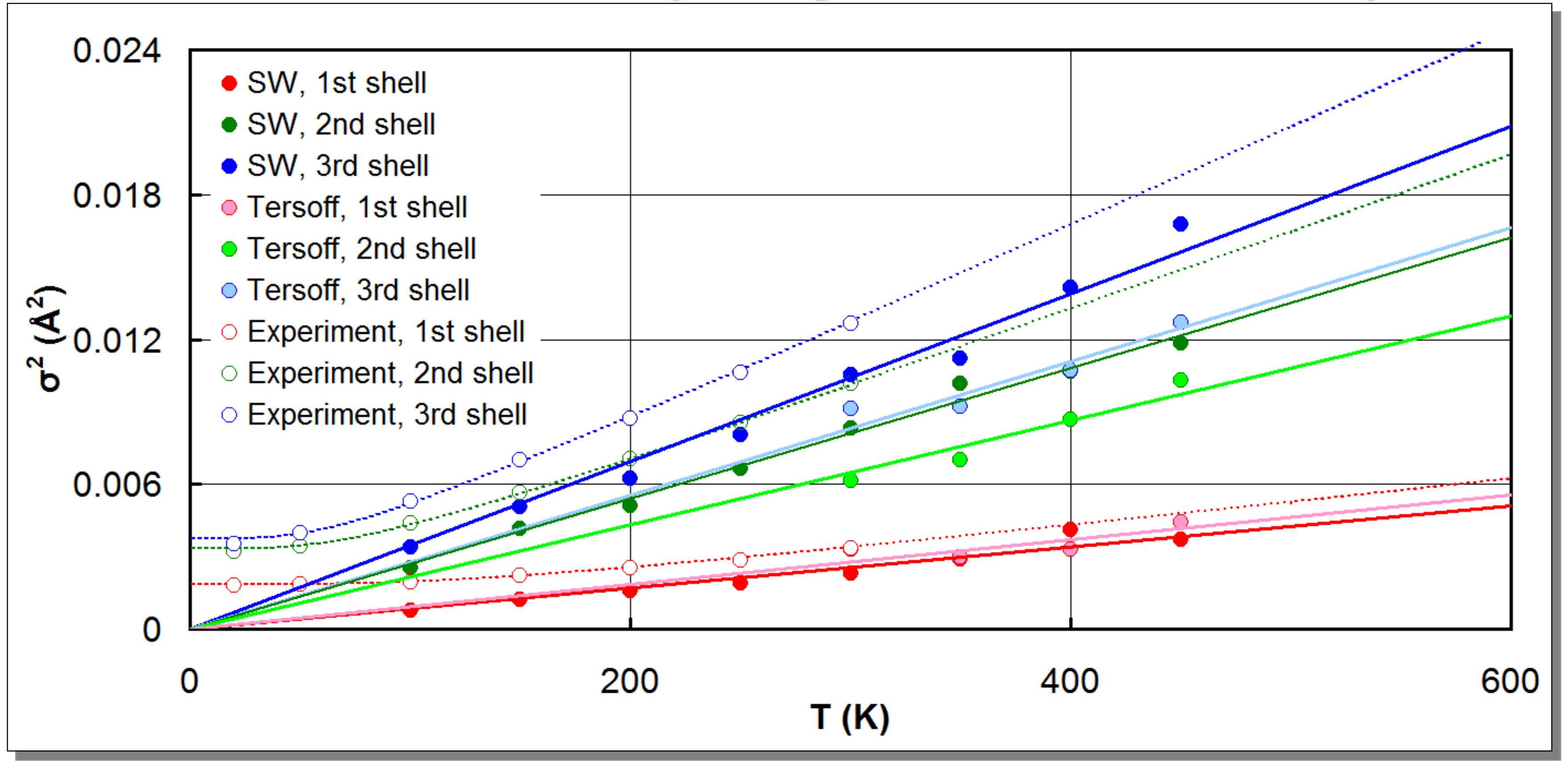

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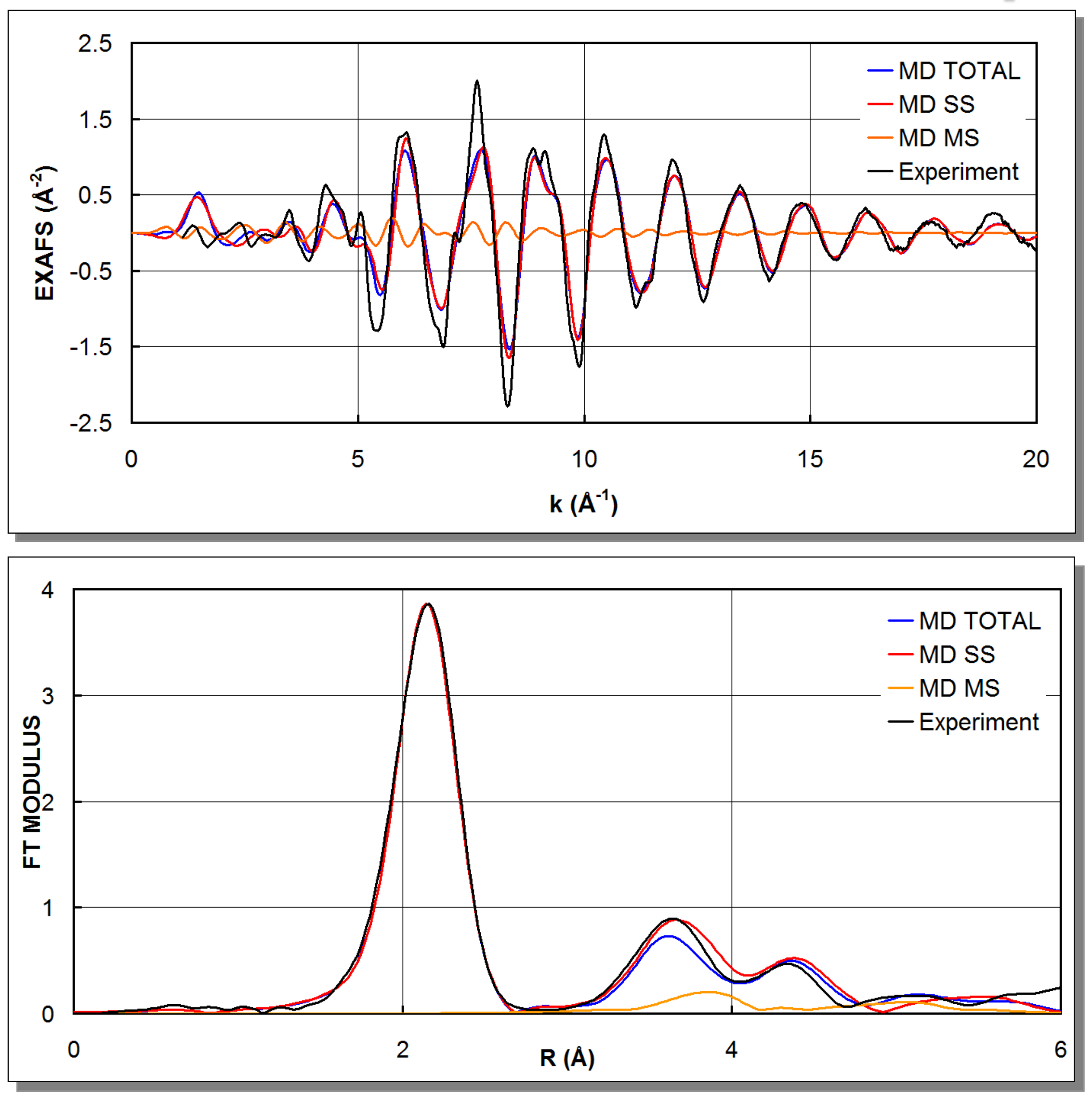

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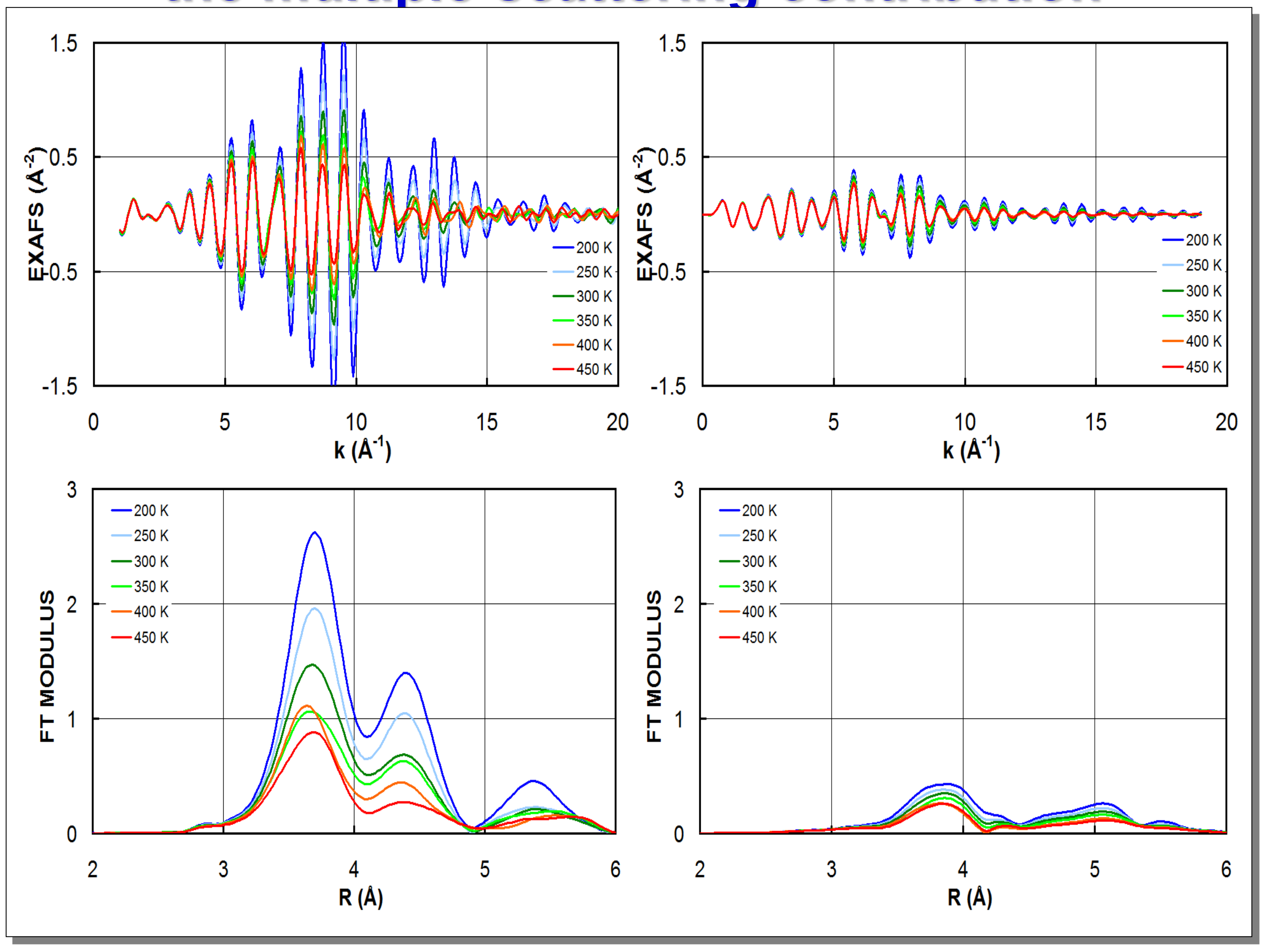

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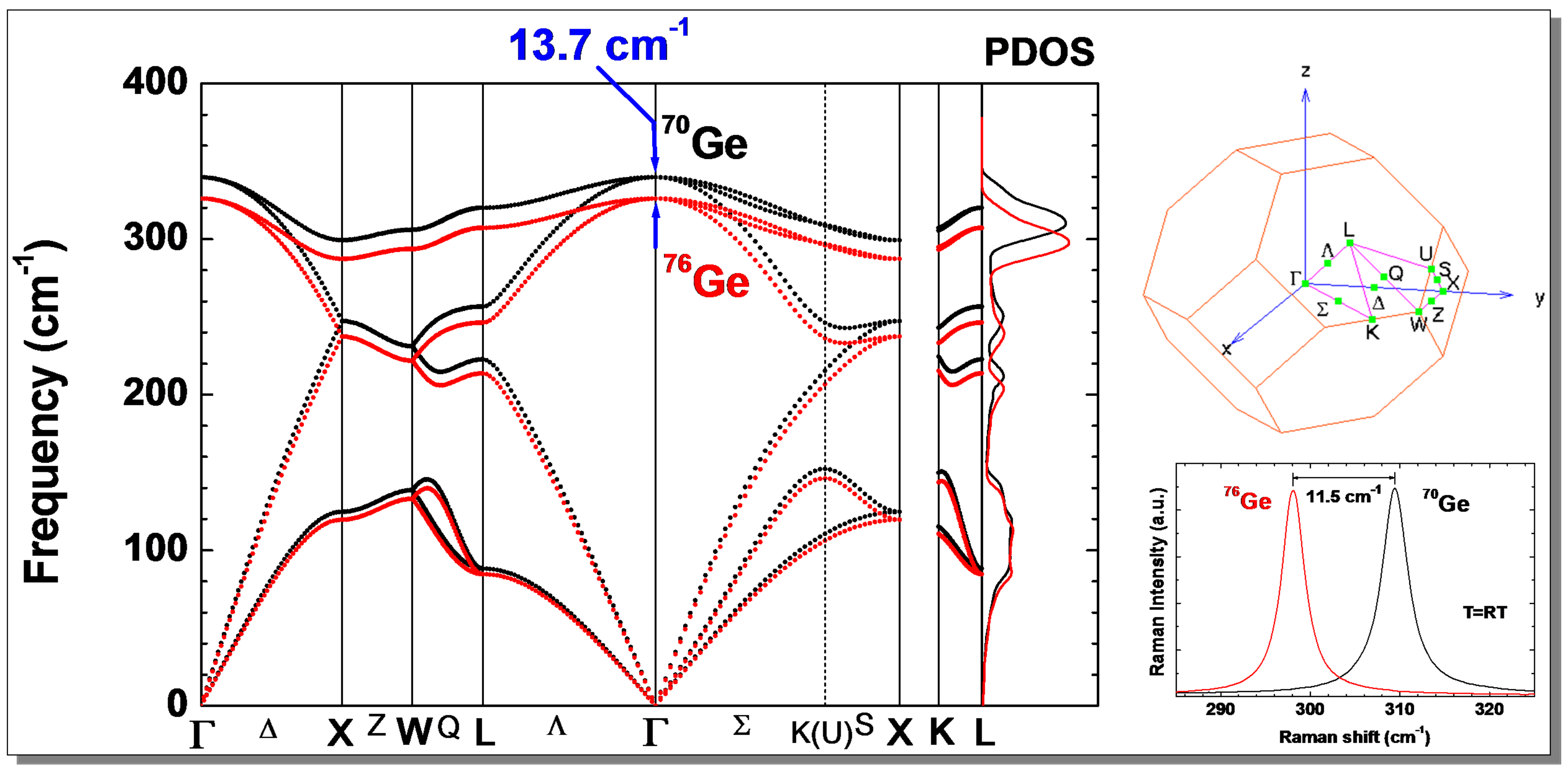

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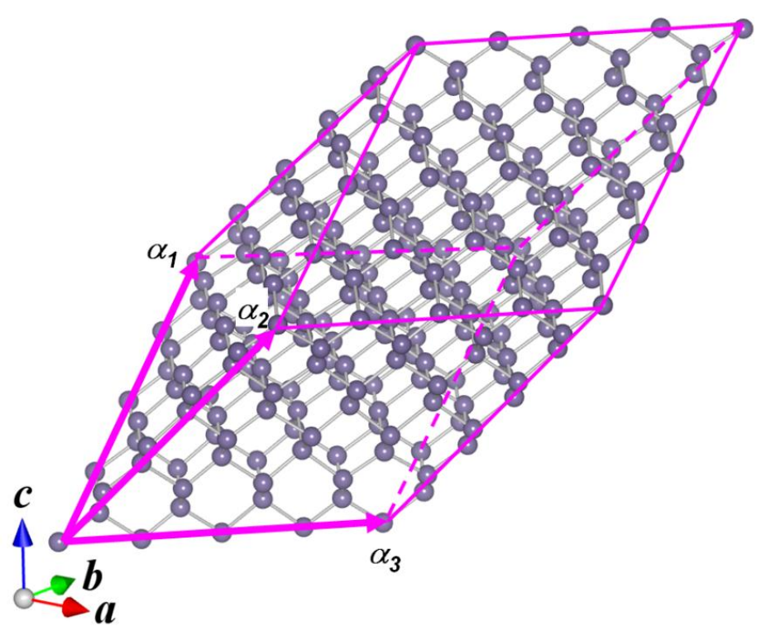

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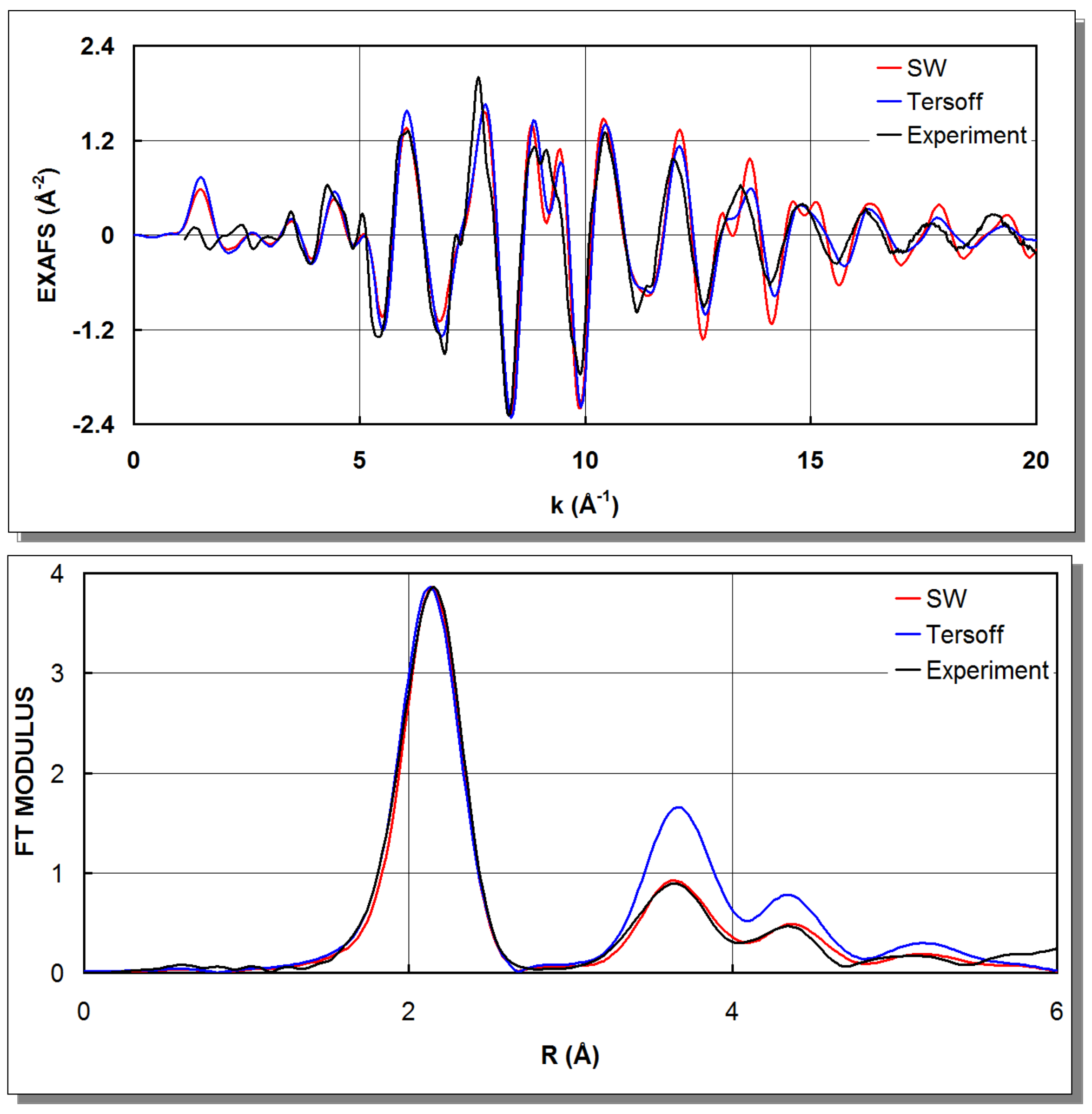

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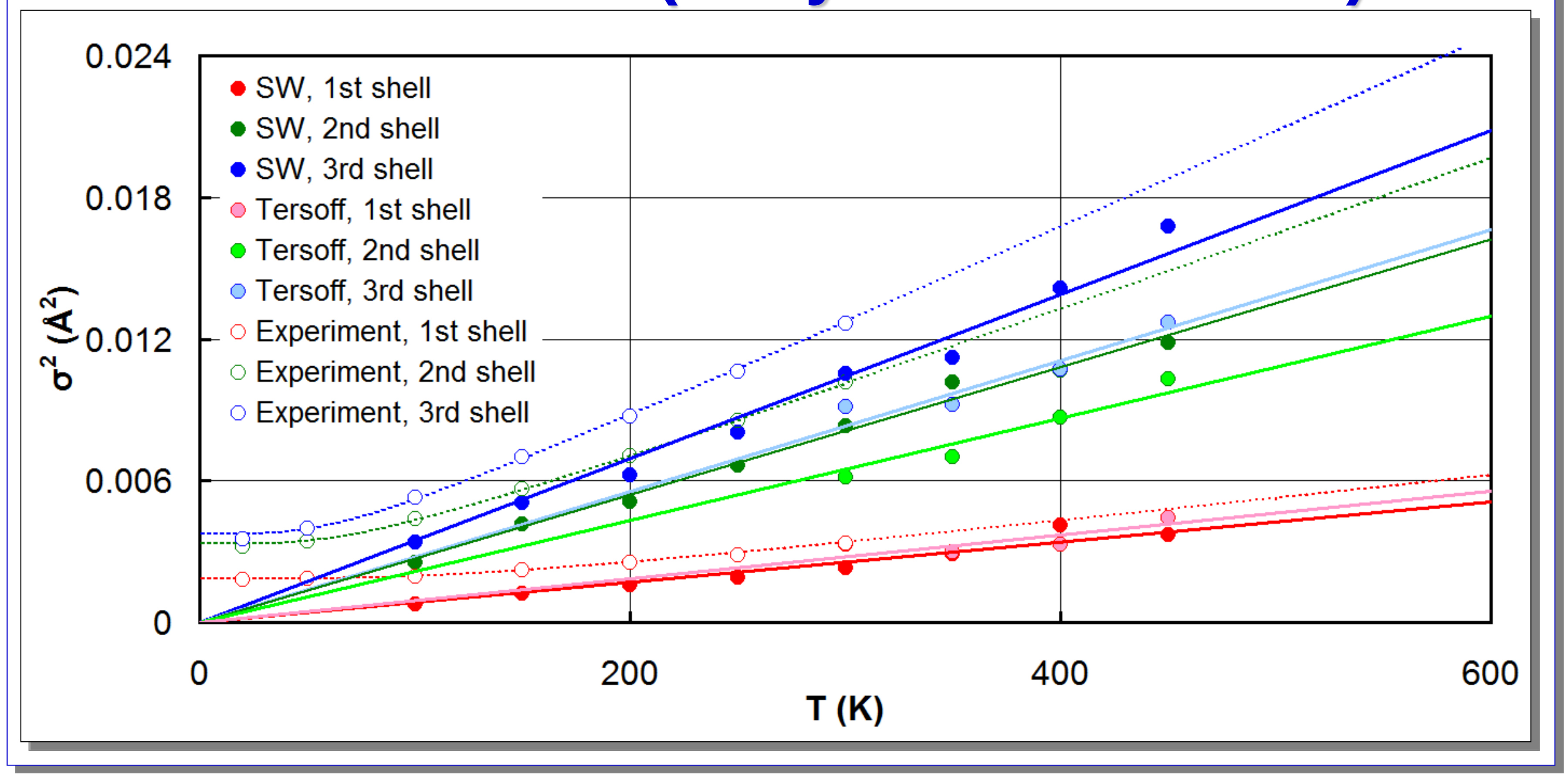

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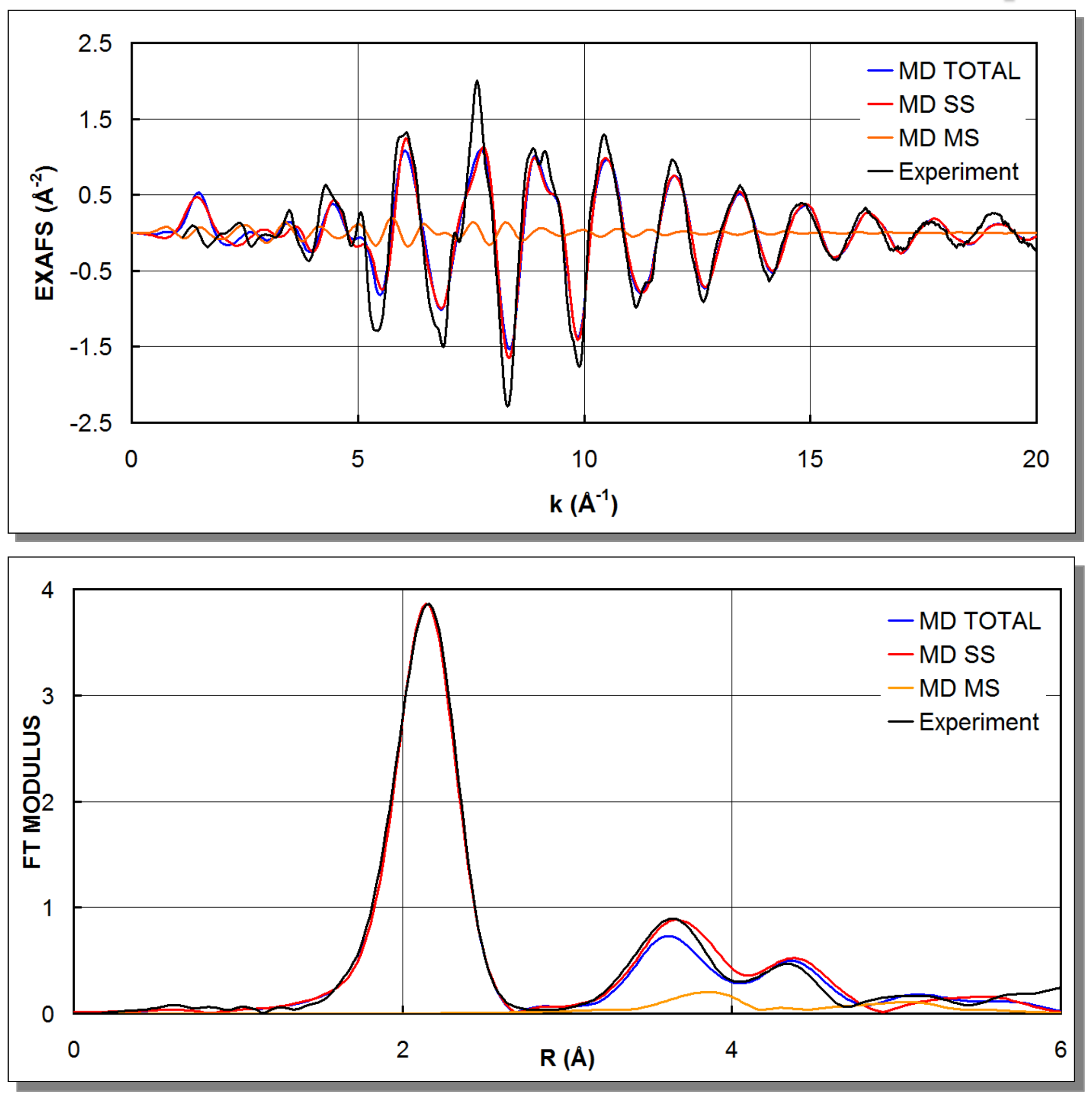

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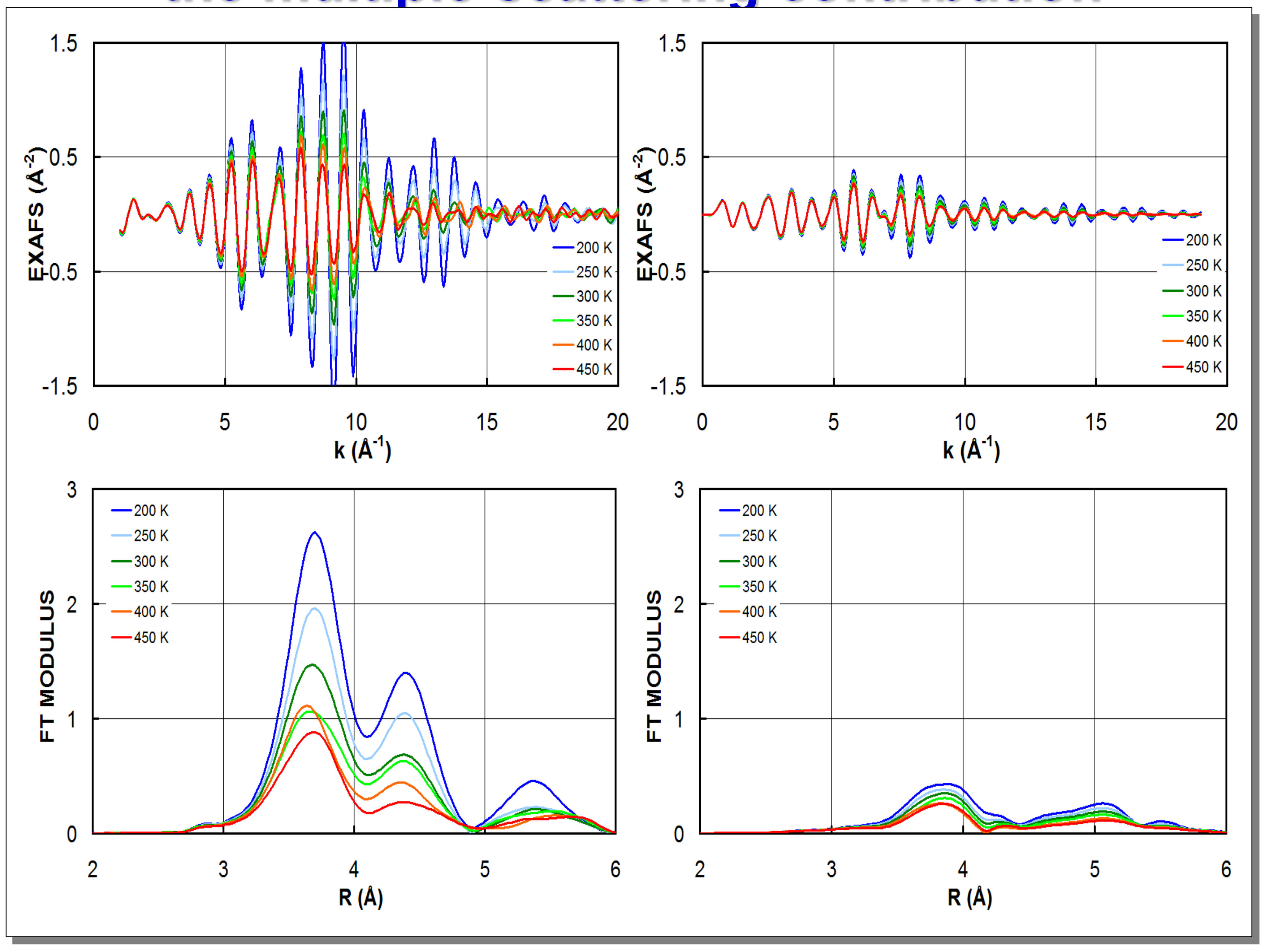

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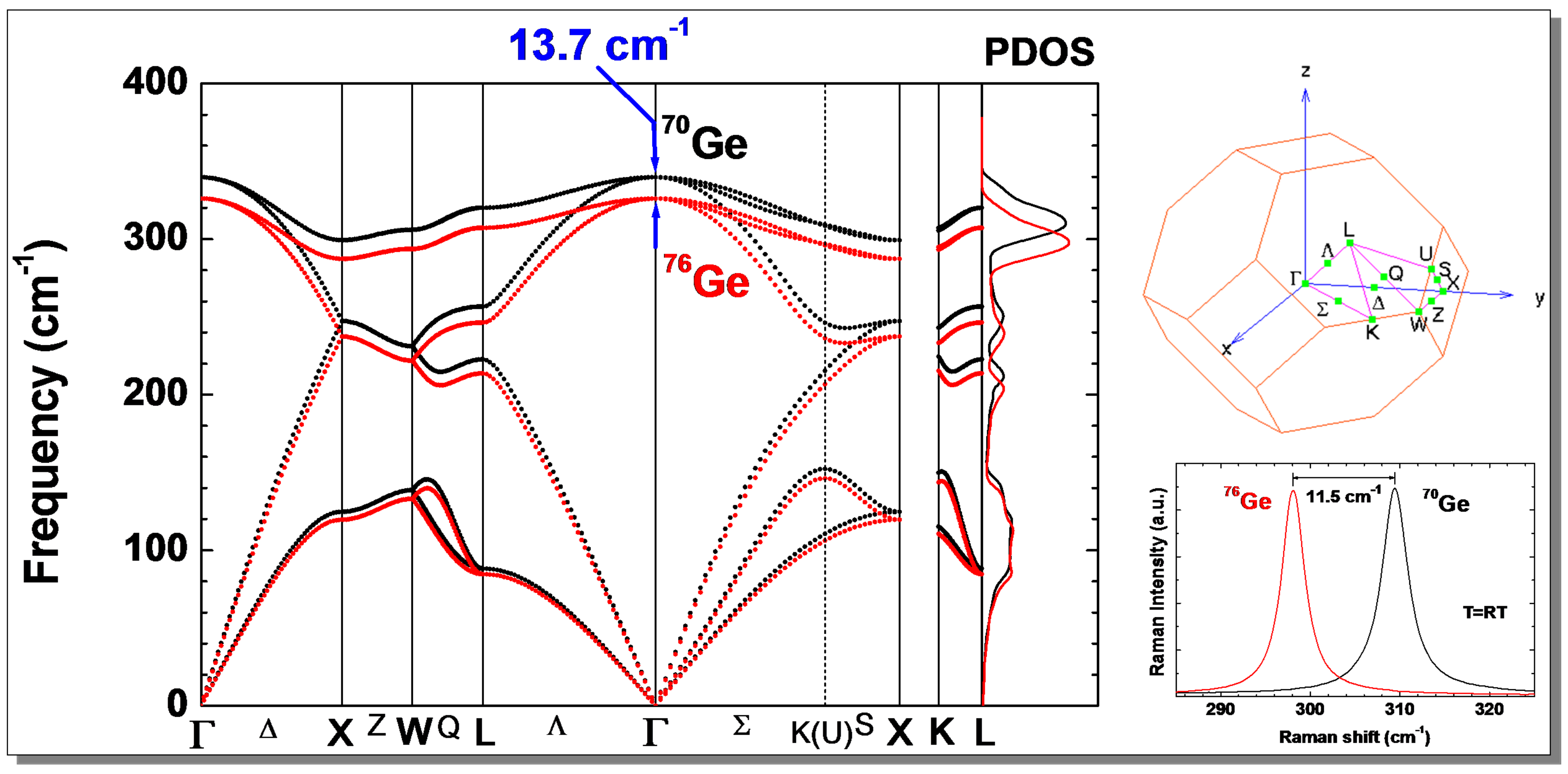

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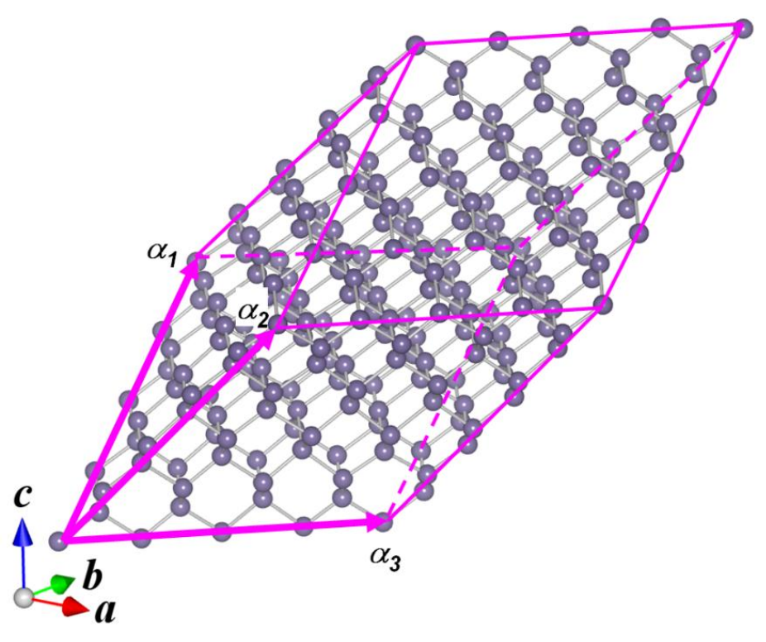

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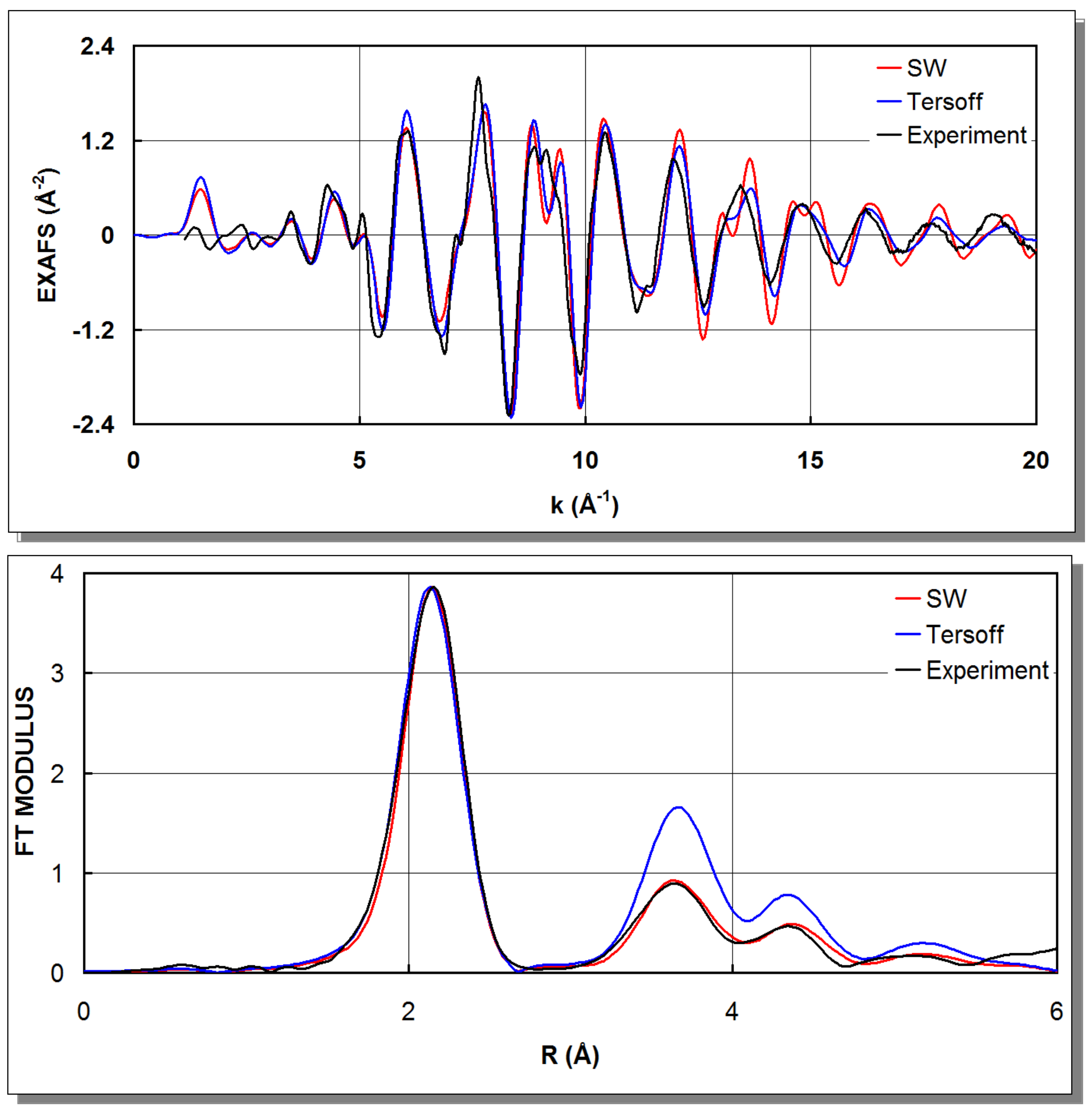

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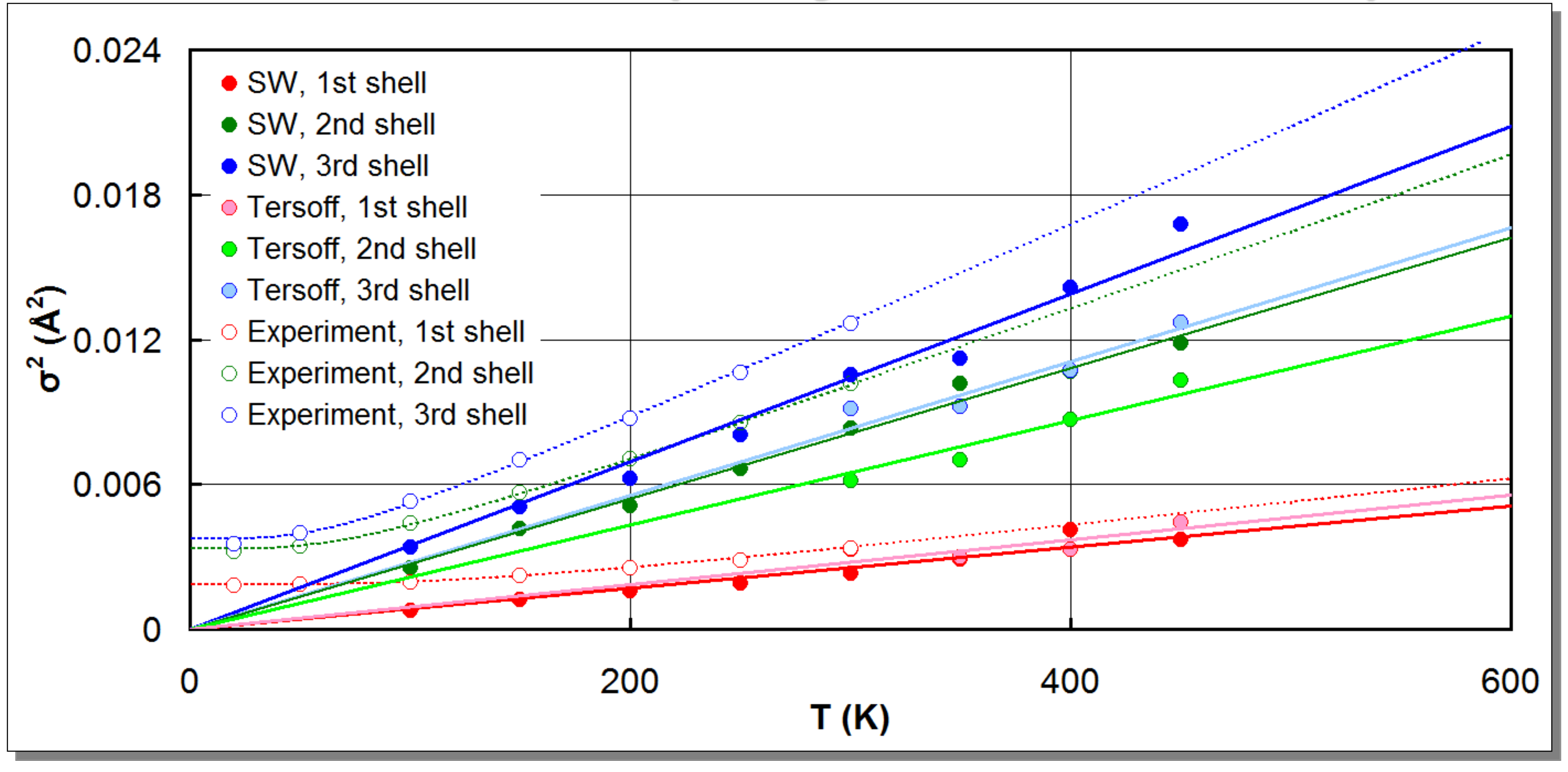

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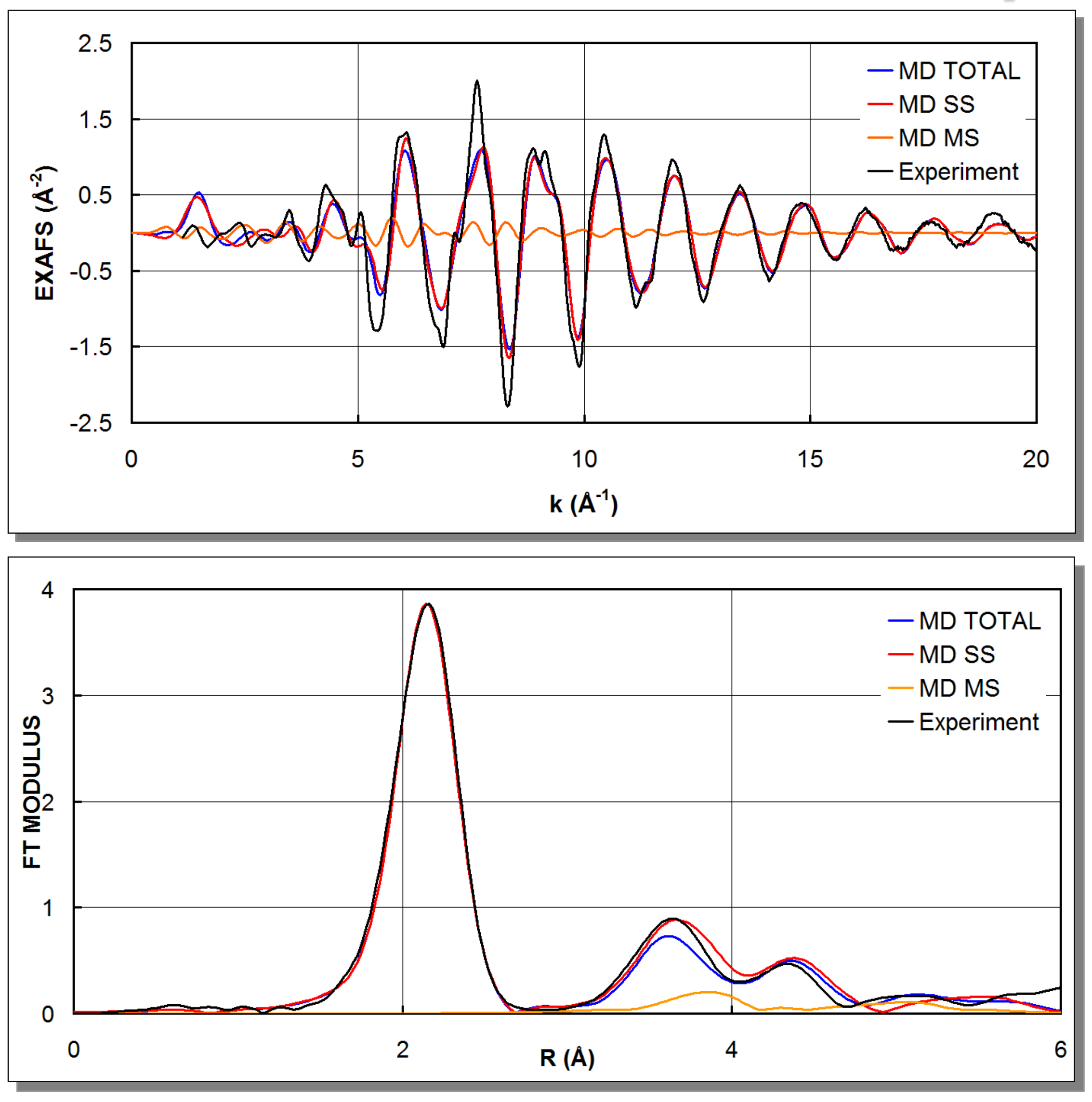

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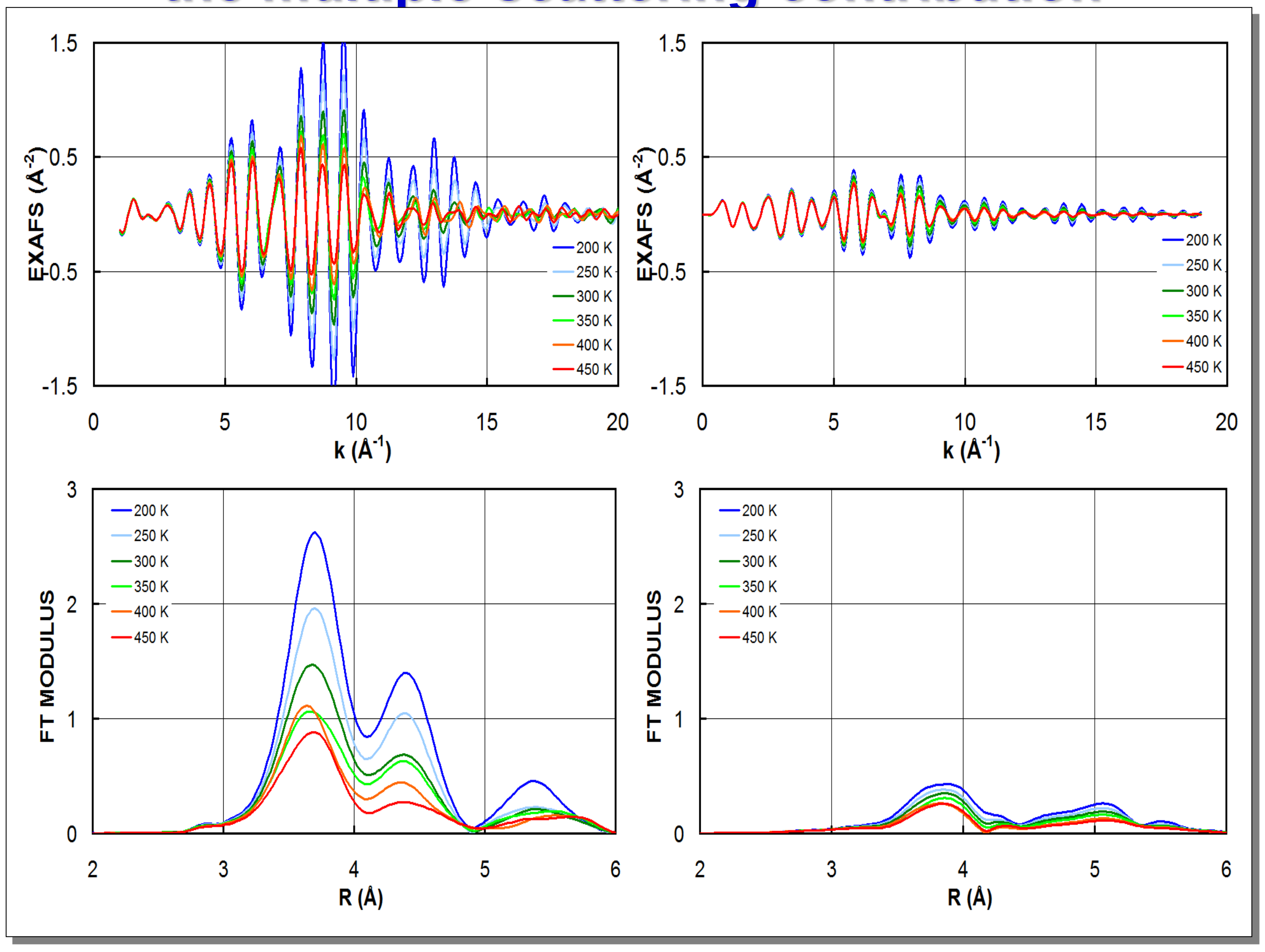

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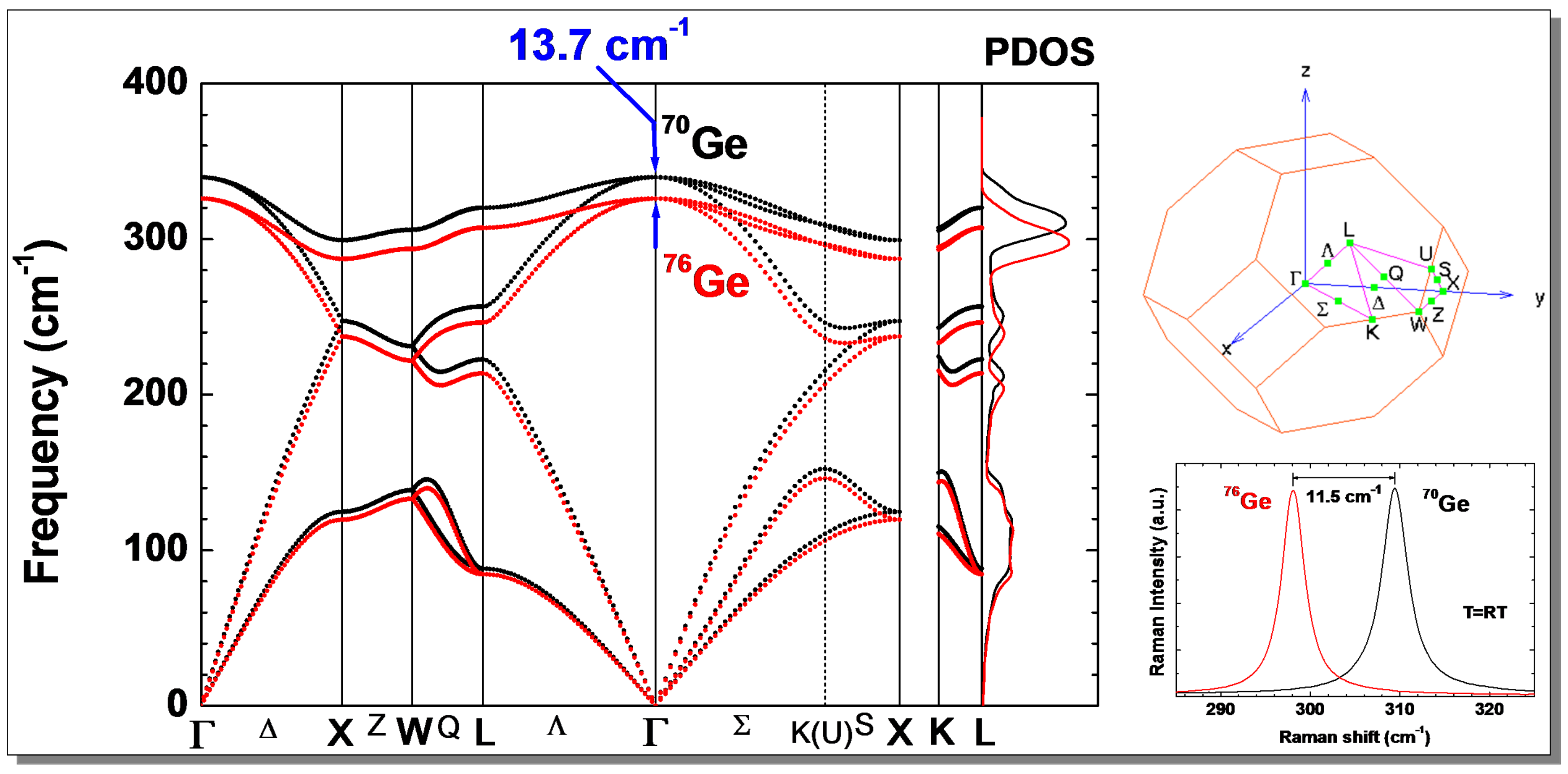

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MOLECULAR DYNAMICS SIMULATIONS OF EXAFS IN GERMANIUM

J. Timoshenko, A. Kuzmin, J. Purans

Institute of Solid State Physics, University of Latvia, Riga, Latvia

E-mail: timoshenkojanis@inbox.lv

Abstract

Classical molecular dynamics (MD) simulations of the Ge K-edge EXAFS have been performed with the aim to estimate the thermal effects within the first three coordination shells and their influence on the single-scattering and multiple-scattering contributions. The effect of the isotopic mass has been also evaluated.

Introduction

The accurate analysis of the Ge K-edge EXAFS in germanium is a long standing problem due to the presence of multiple-scattering (MS) contributions, which strongly influence the "classical" EXAFS analysis, based on the single-scattering (SS) approach [1]. Our previous analysis [2] of thermal effects in two isotopes of ^{70}Ge and ^{76}Ge within the first three coordination shells has been performed using both SS and MS models. We found that while the ratio of the Einstein frequencies for the second and third shells agrees well for the two models, the absolute values of Einstein frequencies are slightly overestimated in the SS model [2]. Unfortunately, the MS EXAFS analysis is limited by two factors: the simplified description of thermal effects within the MS model and a large number of correlated model parameters required.

In this work we present for the first time the classical molecular dynamics (MD) simulation of the Ge K-edge EXAFS using recently developed approach [3].

Molecular Dynamics (MD) Simulations

Interatomic forces: $F_i = -\nabla_i V(r_1, r_2, ..., r_n, \Theta_1, \Theta_2, ..., \Theta_m)$

Tersoff potential [4]:

$$V(r_1, r_2, ...r_n, \Theta_1, \Theta_2, ..., \Theta_m) = \sum_i V_i = \frac{1}{2}\sum_{i \neq j} V_{ij}$$

$$V_{ij} = f_C(r_{ij})[a_{ij} f_R(r_{ij}) + b_{ij} f_A(r_{ij})]$$

$$f_A(r) = -B\exp(-\lambda_2 r)$$

$$f_R(r) = A\exp(-\lambda_1 r)$$

$$f_C(r) = \begin{cases} 1, & r < R-D \\ \frac{1}{2} - \frac{1}{2}\sin\left(\frac{\pi}{2}\frac{(r-R)}{D}\right), & R-D < r < R+D \\ 0, r > R+D \end{cases}$$

$$b_{ij} = (1 + \beta^n \zeta_{ij}^n)^{-1/2n}$$

$$\zeta_{ij} = \sum_{k \neq i,j} f_C(r_{ik}) g(\Theta_{ijk}) \exp[\lambda_3^3 (r_{ij} - r_{jk})^3]$$

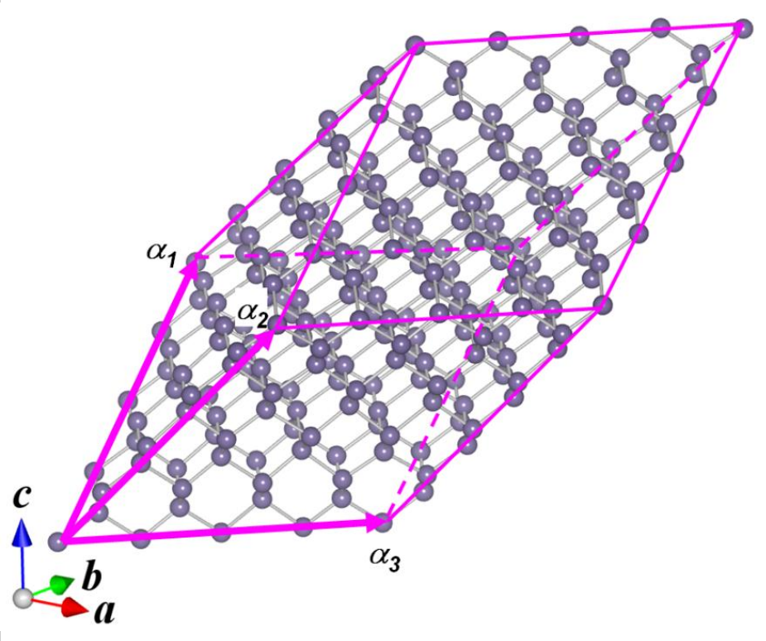
$$g(\Theta) = 1 + \frac{c^2}{d^2} - \frac{c^2}{d^2 + (h - \cos\Theta)^2}$$

$$a_{ij} = (1 + \alpha^n \eta_{ij}^n)^{-1/2n}$$

$$\eta_{ij} = \sum_{k \neq i,j} f_C(r_{ij}) \exp[\lambda_3^3 (r_{ij} - r_{jk})^3]$$

A, keV	1.849	*β ·10⁷*	4.357
B, keV	0.487	*n*	0.436
λ₁, Å	2.480	*λ₃, Å*	1.732
λ₂, Å	1.736	*c ·10⁻⁵*	1.015
R	2.7	*d*	17.51
D	0.3	*h*	-0.601
α	0		

Supercell 5 x 5 x 5



Stillinger-Weber (SW) potential [5]:

$$V(r_1, r_2, ...r_n, \Theta_1, \Theta_2, ..., \Theta_m) = \frac{1}{2}\sum_{i,j} V_{ij} + \frac{1}{6}\sum_{i,j,k} V_{ijk}$$

$$V_{ijk} = \varepsilon f_3\left(\frac{\vec{r}_i}{\sigma}, \frac{\vec{r}_j}{\sigma}, \frac{\vec{r}_k}{\sigma}\right)$$

$$V_{ij} = \varepsilon f_2\left(\frac{r_{ij}}{\sigma}\right)$$

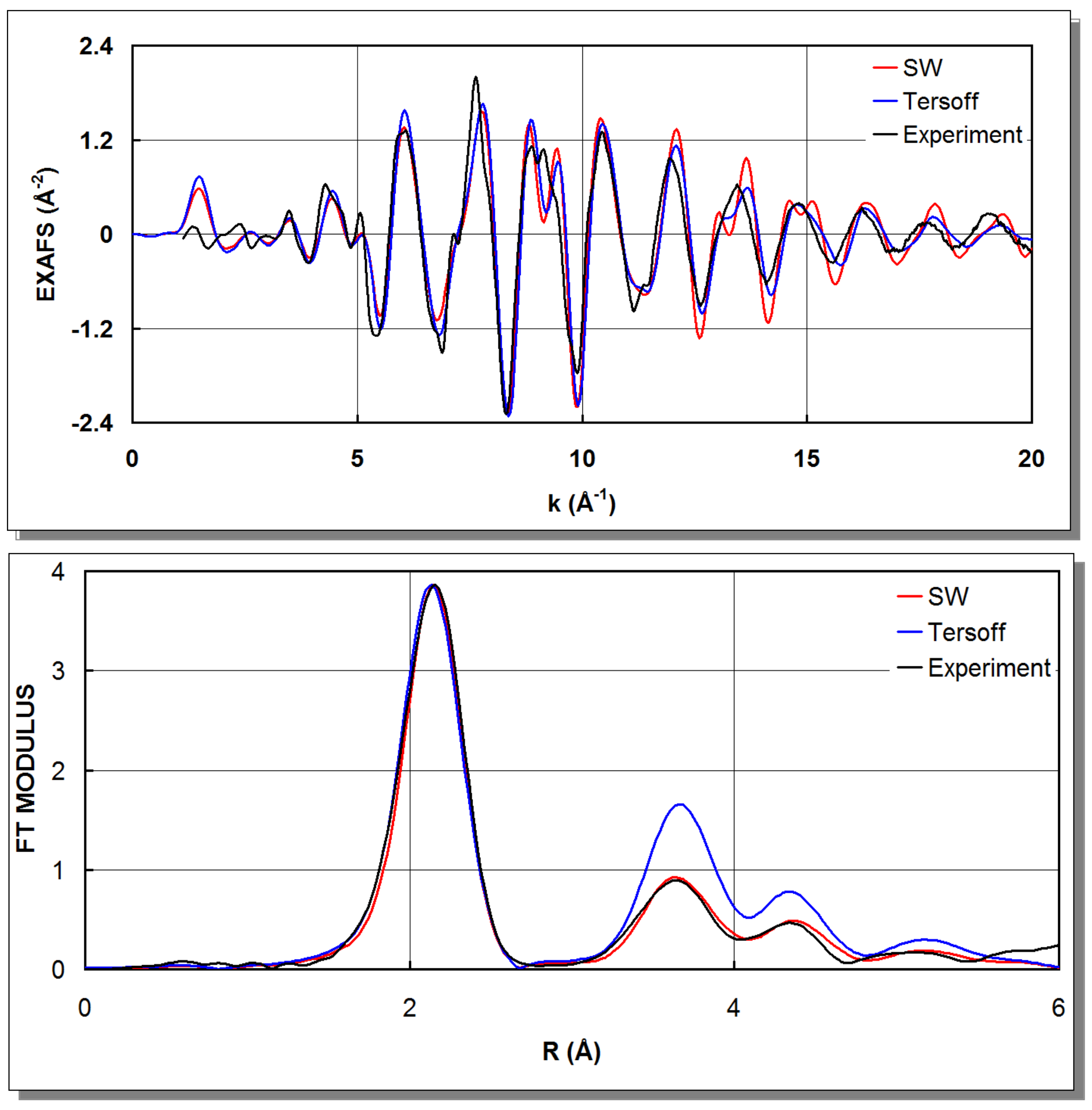
$$f_3(\vec{r}_i, \vec{r}_j, \vec{r}_k) = h(r_{ij}, r_{ik}, \Theta_{jik}) + h(r_{ji}, r_{jk}, \Theta_{ijk}) + h(r_{ki}, r_{kj}, \Theta_{ikj})$$

$$f_2(r) = \begin{cases} A(Br^{-p} - r^{-q})\exp(1/(r-a)), & r < a \\ 0, & r \geq a \end{cases}$$

$$h(r_{ij}, r_{ij}, \Theta_{jik}) = \lambda \exp\left(\frac{\gamma}{r_{ij} - a} + \frac{\gamma}{r_{ik} - a}\right)(\cos\Theta_{jik} - \cos\Theta_0)$$

A	7.050	*λ*	31
B	0.602	*γ*	1.2
p	4	*ε, eV*	1.93
q	0	*σ, Å*	2.181
a	1.8	θ₀, ⁰	109.5

Force-field models: SW vs. Tersoff



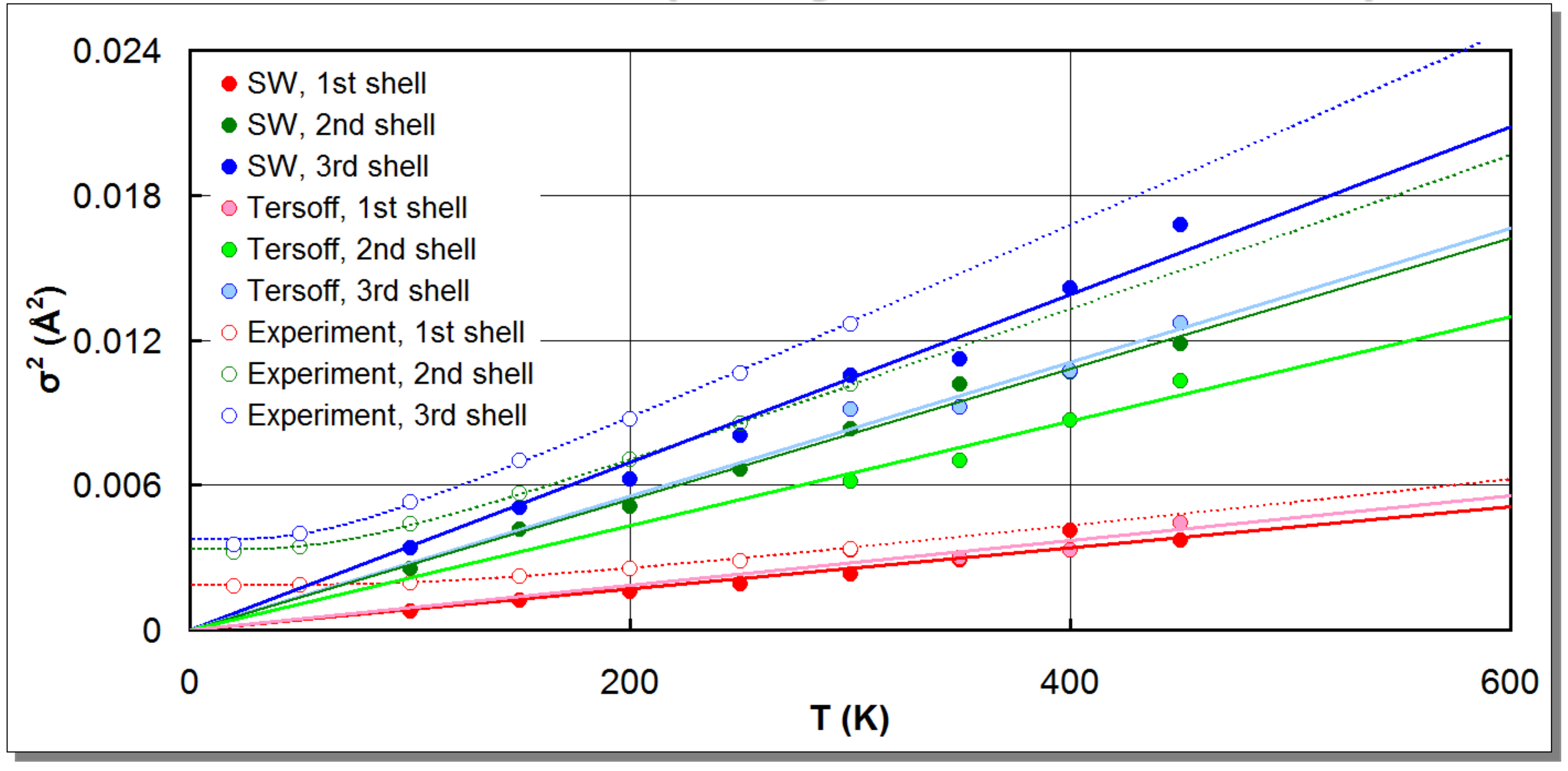
Experimental (T = 300 K) and configuration-averaged (T = 300 K, up to 6.5 Å) EXAFS spectra $\chi(k)k^2$ and their Fourier transforms for two different force-field models:

1) Stillinger-Weber (SW)
2) Tersoff.

The contribution from the 2nd and 3rd shells (peaks at ~3.7 and ~4.4 Å) are overestimated in the case of the Tersoff potential model.

The SW potential gives EXAFS signal being in better agreement with the experiment and will be used further.

Parallel MSRD (Debye-Waller factors)



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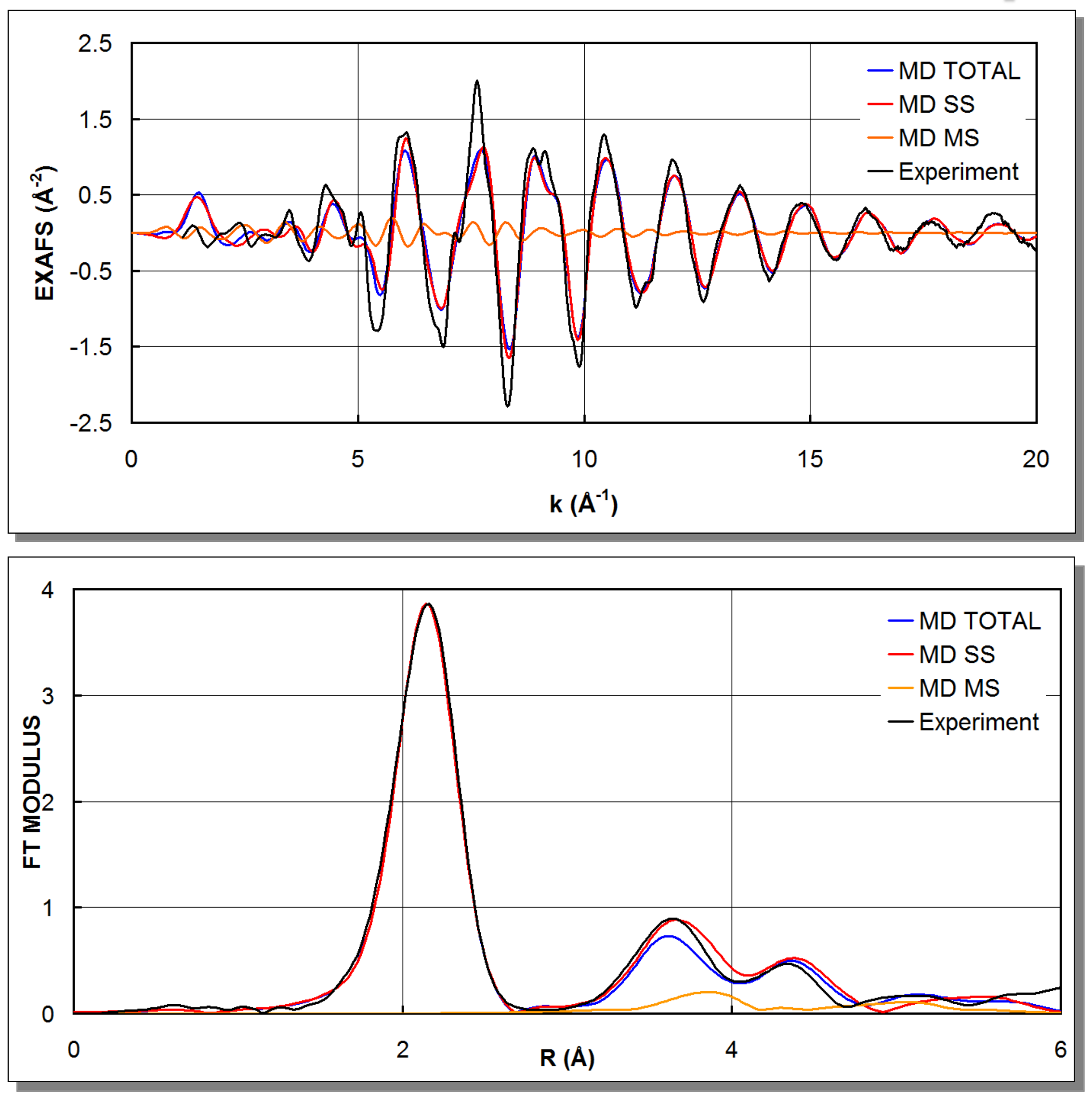
MD-EXAFS vs. Experiment

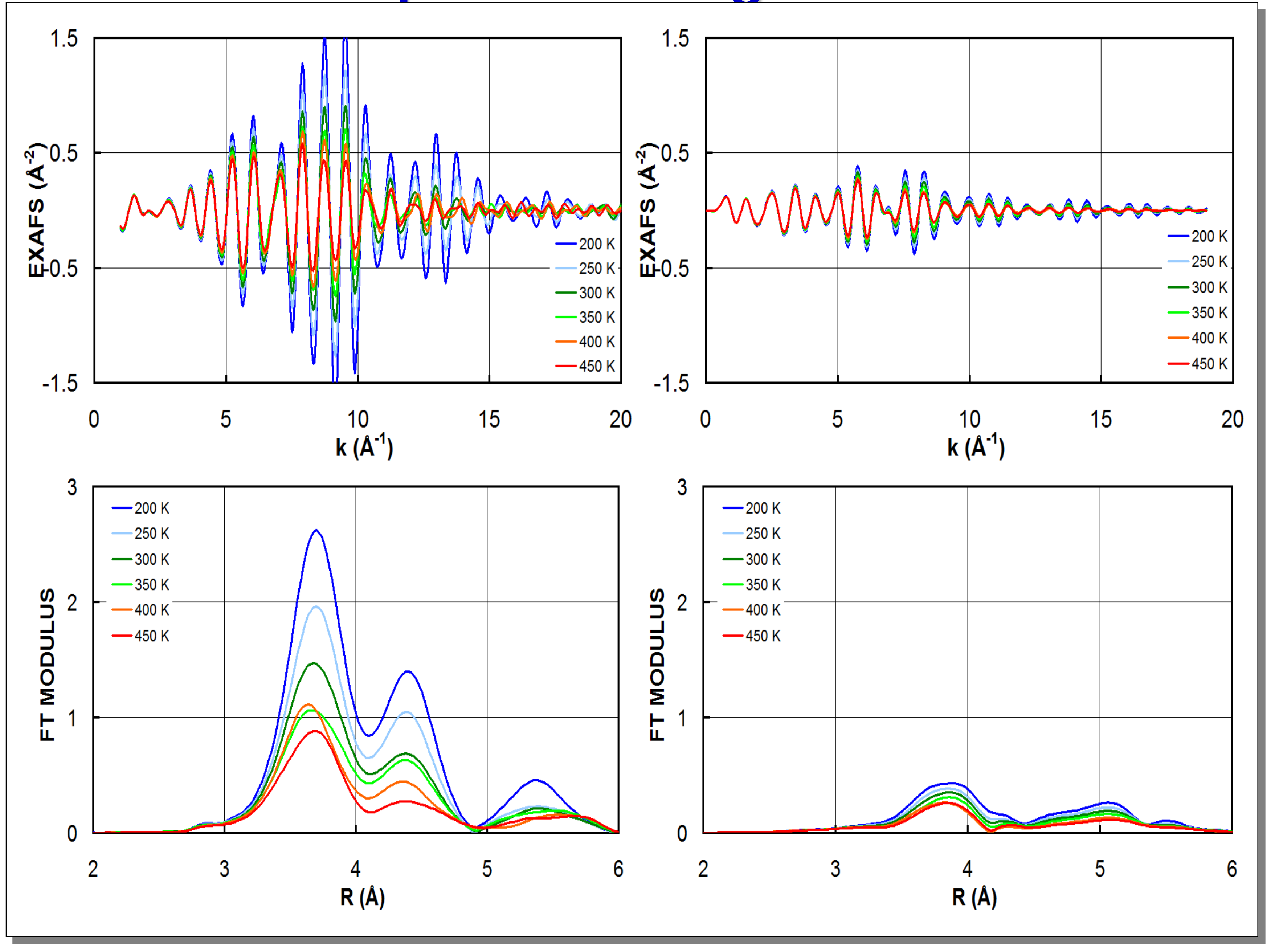


Experimental (T = 300 K) and configuration-averaged (T = 350 K, up to 6.5 Å) EXAFS spectra $\chi(k)k^2$ and their Fourier transforms.

The single-scattering (SS) and multiple-scattering (MS) contributions are also shown.

The MS effects contribute mainly under the second peak at 3.8 Å.

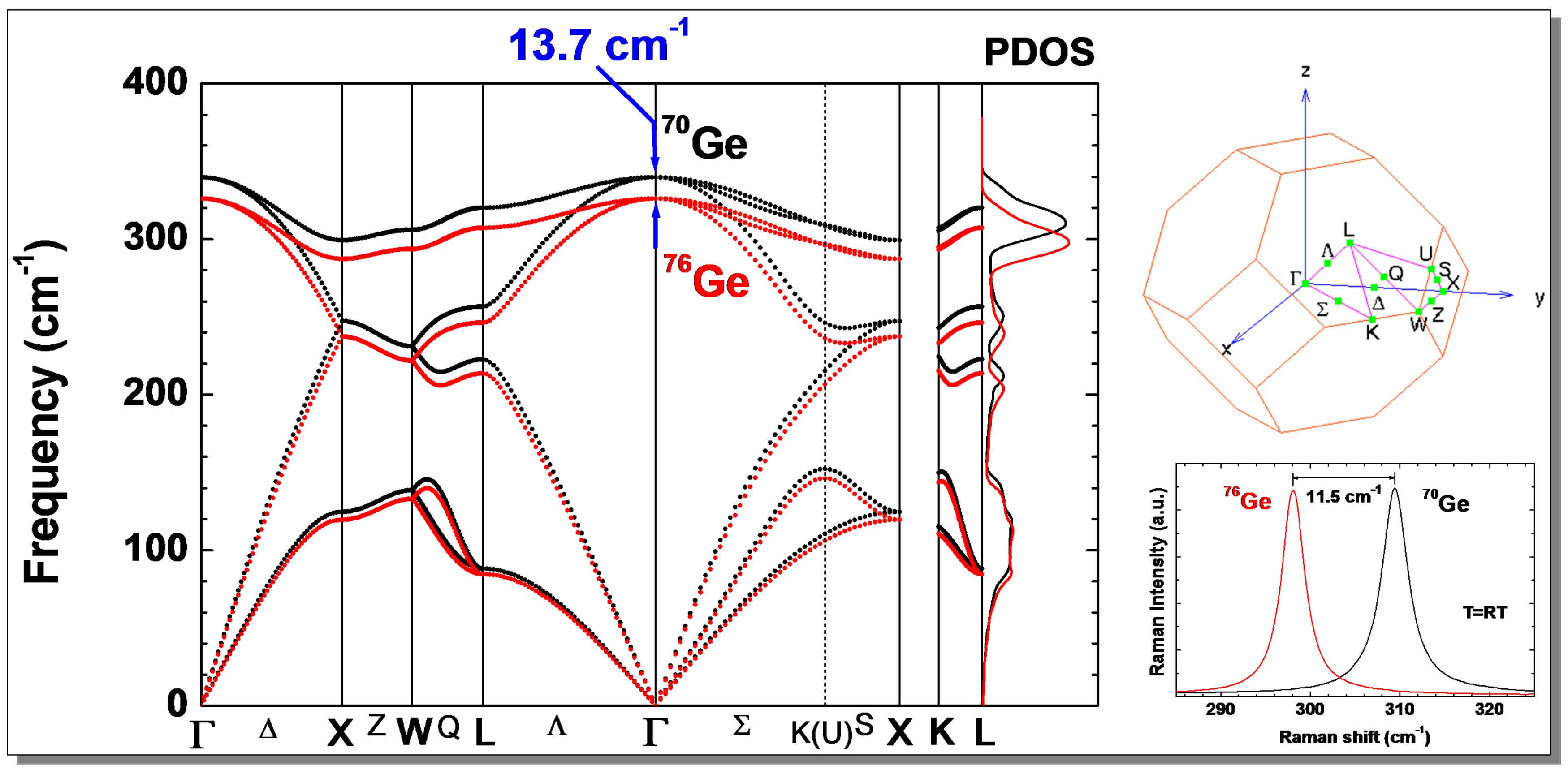
MD-EXAFS: Temperature dependence of the multiple-scattering contribution



Configuration-averaged EXAFS spectra $\chi(k)k^2$ (upper left panel) and their Fourier transforms (FTs) (lower left panel), calculated in the temperature range from 200 K to 450 K. Multiple-scattering contributions to EXAFS spectra (upper right panel) and their FTs (lower right panel).

The MS effects are less sensitive to the thermal disorder.

Isotopic effect



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