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Observations of different thermal scattering models in view of graphite based materials

R. Dagan

Karlsruhe Institute of Technology, Institute for Nuclear Waste Disposal, P.O. Box 3640, 76021 Karlsruhe, Germany

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The full free gas vs. the full solid state models (both energy dependent)

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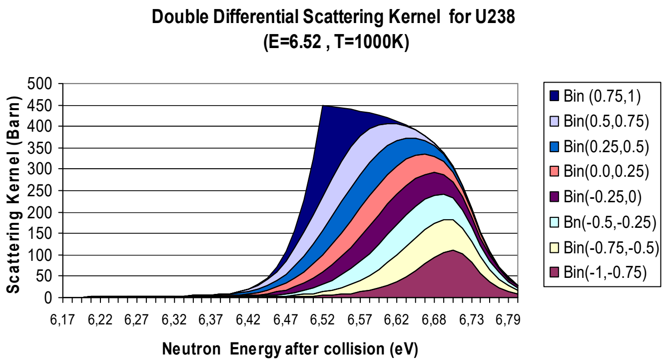


IDEAL GAS KERNEL: ENERGY DEPENDENT $\sigma_s(E_r)$

$$\sigma_s^T(E \to E', \vec{\Omega} \to \vec{\Omega'}) =$$

$$\frac{1}{4\pi E}\sqrt{\frac{A+1}{A\pi}}\int_{\epsilon_{max}}^{\infty} d\xi \int_{\tau_0(\xi)}^{\tau_1(\xi)} d\tau$$

$$\left[\frac{(\xi+\tau)}{2}\right]\left(\frac{A+1}{A}\right)^2 \sigma_s\left[\left(\frac{A+1}{A^2}\right)\frac{[\xi+\tau]^2}{4}k_B T\right]$$

$$\exp\left\{v^2 - \left[\frac{(\xi+\tau)^2}{4A} + \frac{(\xi-\tau)^2}{4}\right]\right\}\left[\frac{\epsilon_{max}\epsilon_{min}(\xi-\tau)^2}{B_0 sin\hat{\varphi}}\right]$$

Solid state based expression of Word & Trammel (1980)

$$\frac{d^2\sigma_R}{d\Omega dE} = \frac{v}{2\pi}\left(\frac{\Gamma_n}{2k_i}\right)^2\left(\frac{E_f}{E_i}\right)^{1/2}\int_{-\infty}^{\infty} dT \exp(+iET)\int_0^{\infty} dt \exp[a(t)]\int_0^{\infty} dt'$$
$$\times \exp[a^*(t')] \times W(T, t, t'),$$

$$W(T, t, t') = \left\langle \exp\left[-i\vec{k}_i\vec{r}(T-t')\right]\exp\left[i\vec{k}_f\vec{r}(T)\right]\right.$$
$$\left.\exp\left[-i\vec{k}_f\vec{r}\right]\exp\left[i\vec{k}_i\vec{r}(-t)\right]\right\rangle.$$

$$a(t) = -i(E_r - E_i - i\Gamma/2)t.$$

Solid state based phonon expansion scattering

$$Phonon\ Expansion:\ S(\alpha,\beta) = e^{-\alpha\lambda} \sum_{n=0}^{\infty} \frac{1}{n!} [\alpha\lambda]^n \mathcal{T}_n(\beta)$$

$$\#\ \mathcal{T}_0(\beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\beta t}\, dt = \delta(\beta);$$

$$\#\ \mathcal{T}_1(\beta) = \frac{P(\beta) e^{-\frac{\beta}{2}}}{\lambda};$$

$$\#\ \mathcal{T}_n(\beta) = \int_{-\infty}^{\infty} \mathcal{T}_1(\beta')\mathcal{T}_{n-1}(\beta - \beta')\, d\beta';$$

$$\mathcal{T}_n(\beta) = e^{-\beta} \mathcal{T}_n(-\beta);$$

$$\#\ \beta = \frac{E - E'}{kT};\ \alpha = \frac{E' + E - 2(EE'\mu)^{\frac{1}{2}}}{AkT};$$

$$\#\ P(\beta) = \frac{\rho(\beta)}{2\beta sinh\left(\frac{\beta}{2}\right)};\ \lambda = \int_{-\infty}^{\infty} P(\beta) e^{-\frac{\beta}{2}}\, d\beta;$$

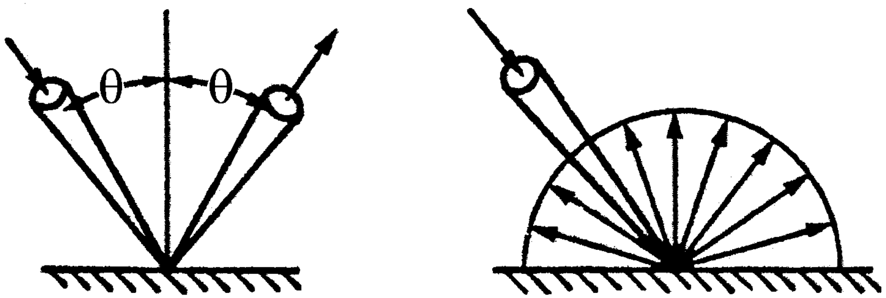
$$\#\ \rho(k,k',+t) = \frac{k \cdot k'}{2M} \int_0^{\infty} \frac{\rho(\omega)}{\omega} \left[\coth\left(\frac{\omega}{2kT}\right)\cos(\omega t) - i sin(\omega t)\right] d\omega$$

- *How important is the resonance treatment in low energies?*

- *Should Optical Model based scattering replace this method?*
  case in which the elastic coherent is dominant, like Graphite.

Other scattering models ???

Duderstadt (1976): "Although one introduces several horrifyingly brutal approximations, at least to the solid state physicist, for nuclear engineer they are acceptable".. This will mean that neutrons behave like light , or X-ray



Ideal mirroring reflection

Ideal diffuse reflection

Real surface reflection

How "horrifying" is the solid state physicists' approximations?

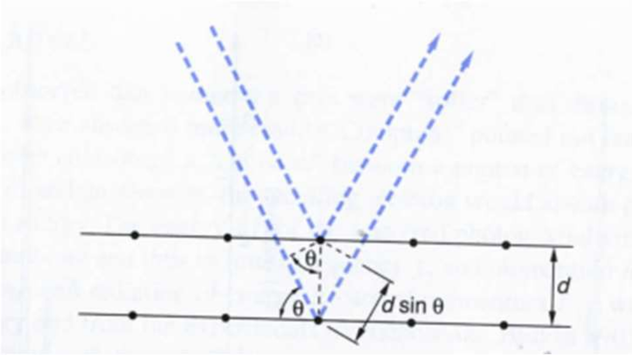
Two options for X ray based scattering

Bragg scattering:

$$2d \sin\theta = n\lambda$$

λ Wavelength

n an integer (depth of the layer)

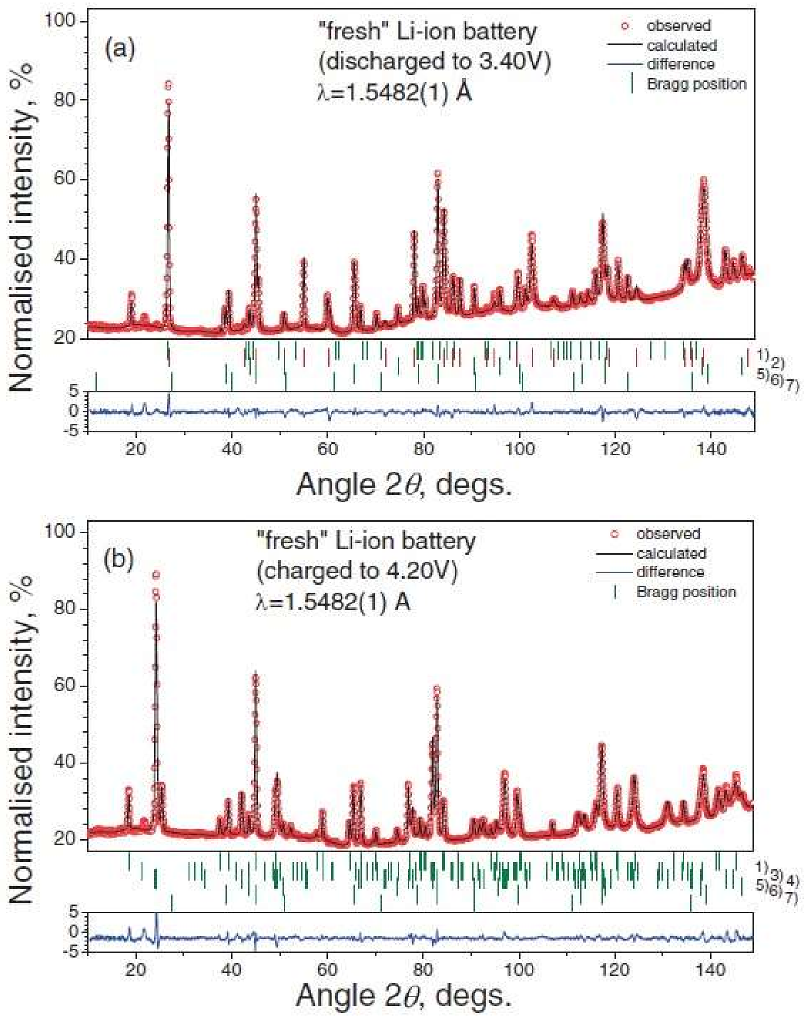


source : R. Tipler , R. Llewellyn "Modern Physics"

- Small angle Diffusive scattering kernel based on x-ray rough surface scattering
- "**assuming ….molecular structure can be neglected…**"

Source: P. Müller-Buschbaum, Polymer Journal (2013) 45, 34–42

Scattering kernels for investigation Li-Ion Battery based on Bragg scattering (coherent- elastic)



- Calculated positions of Bragg peaks in charged and discharged state of Li-Ion Battery, when the anode is Graphite
- Inelastic-incoherent completely ignored

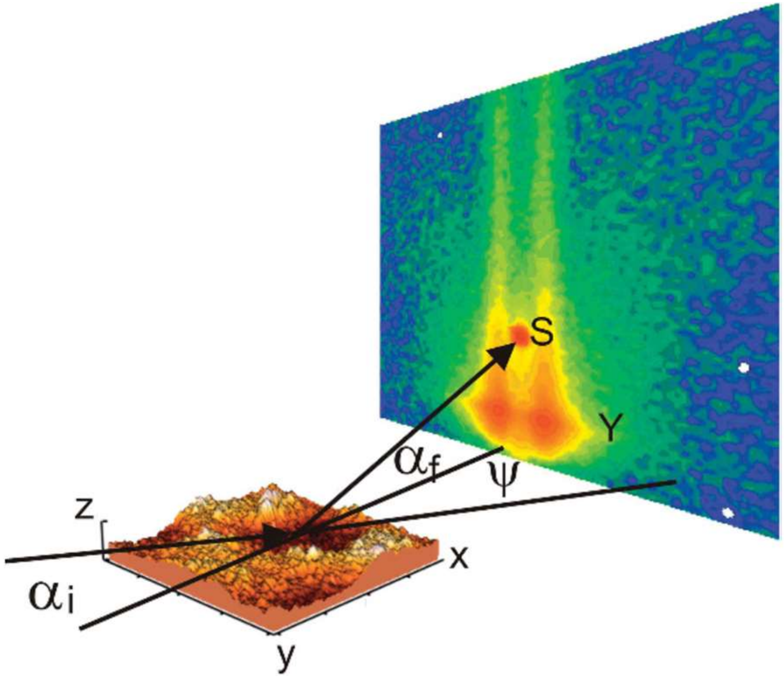
- Reference: Fatigue Process in Li-Ion Cells: An In Situ Combined Neutron Diffraction and Electrochemical Study, O. Dolotko et al., *Journal of The Electrochemical Society*, 159 (12) A2082-A2088 (2012)

$$2d \sin \theta = n\lambda$$

λ Wavelength

n an integer (depth of the layer)

Schematic of the scattering geometry used in GISANS (Grazing incidence small Angle neutron scattering)



Scattering vector $q = (q_x, q_y, q_z)$

q_x=2π(cosΨcosα$_f$ –cosα$_i$) / λ

q_y=2π(sinΨcosα$_f$) / λ

q_z=2π(sinα$_i$ +sinα$_f$) / λ

- The sample is placed in the (x, y) plane, and the incident neutron beam is along the x axis.
- The resulting scattering pattern is anisotropic and typically exhibits a Yoneda peak (marked with Y) and a specular peak (marked with S)
- The scattering kernel approach is based upon Fresnel transmission and reflection coefficients
- Reference: Grazing incidence small-angle neutron scattering: challenges and possibilities,

Source: P. Müller-Buschbaum, Polymer Journal (2013) 45, 34–42

Coupling chemical binding and free gas model via the effective temperature: validity of the SCT approximation

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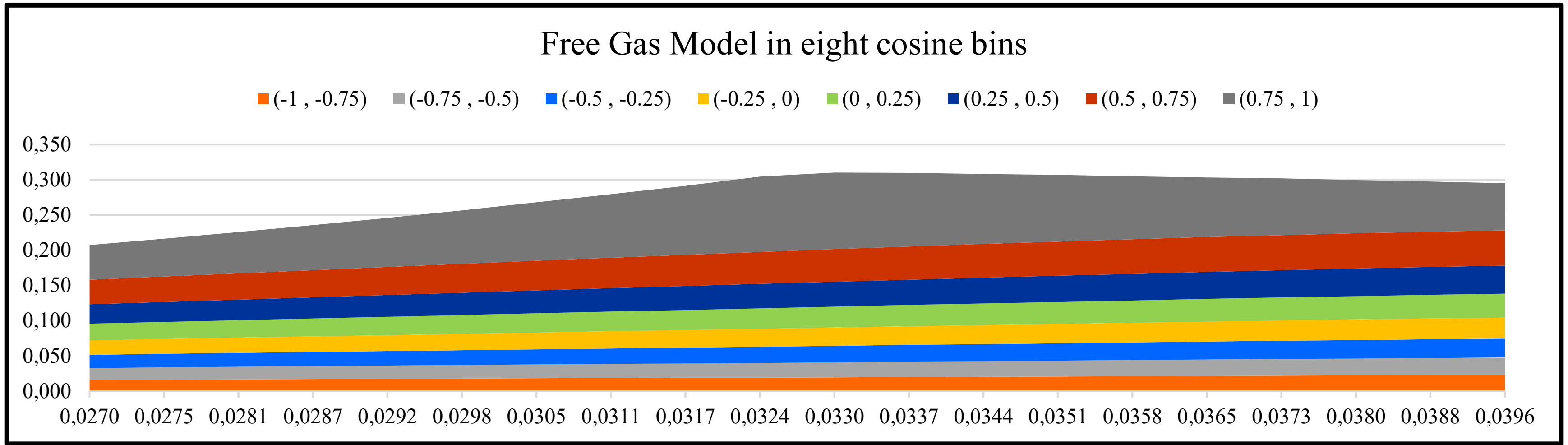
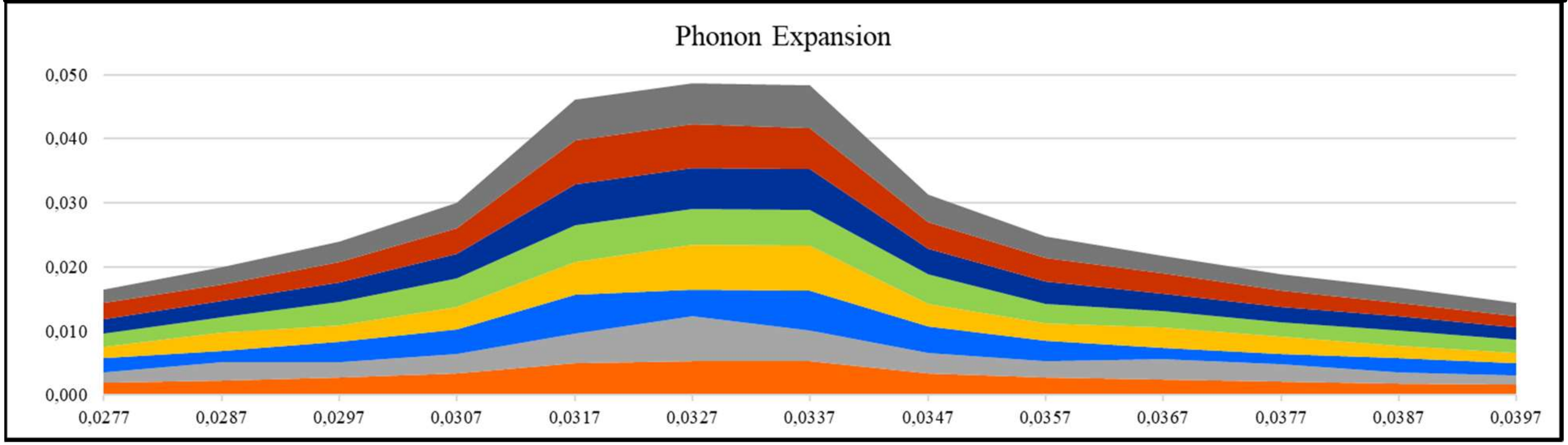
Graphite: Comparison between SCT app. and phonon expansion at E=0.0327 ev (n=12) (via MATLAB)





Deriving the equation for agitating target: connection to the azimuth angle

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$$\sigma_s^T\left(E\rightarrow E',\vec{\Omega}\rightarrow\vec{\Omega}'\right)=\frac{1}{2\pi}\sigma_s^T\left(E\rightarrow E',\mu_0^{lab}\right)=\frac{1}{2\pi v}\left(\frac{A+1}{A}\right)^4\left(\frac{A}{\pi}\right)^{3/2}$$

$$\int 2\pi u^2 du\int d\mu_u\int c^2dc\int (u')^2du'\int d\mu_{u'}\int\frac{2}{\sin\varphi}\frac{\delta(u'-u)}{(u')^2}\exp\left[v^2-(A+1)\left(\frac{u^2}{A}+c^2\right)\right]$$

$$\frac{4vv'c^2}{B_0'}\delta(\cos\varphi-\cos\hat{\varphi})u\sigma_s(E_r)\frac{P(u,\mu_0^{cm})}{2\pi}d\cos\varphi$$

$$\frac{1}{uvc}\delta\left[\mu_u-\frac{(v^2-c^2-u^2)}{2uc}\right]\frac{1}{2u'ck_BT}\delta\left[\mu_{u'}-\frac{(v')^2-(u')^2-c^2}{2u'c}\right]$$

- The derivation of the equation is based on the fulfillment of all constraints marked by δ
- Note the azimuth angle is connected to the polar angle

$$(\vec{\Omega}\cdot\vec{\Omega}')=\mu_0^{lab}\ ;\ \varphi=\varphi_{u'}-\varphi_u$$

$$\cos\hat{\varphi}=R=\left[\left(4vv'c^2\mu_0^{lab}-C_0'\right)/B_0'\right]$$

- not incorporated in DBRC or $S(\alpha,\beta)$

Summary

- Thermal scattering analysis based on suggestions by J. Rowlands could help to better understanding of the different approaches
- More experiments in the thermal range for graphite based materials are under consideration.
- For thermal scattering: OMP can't comply with temperature, chemical binding or energy dependency, yet the idea of "optical model" is being used for Batteries and Photovoltaic cells.
- The azimuth angle, elastic coherent data should be considered for MC codes. In those cases the inelastic incoherent part could be presented by SCT approximation.
- **Response to Duderstadt (1976): The solid state physicists' approximations are " quite horrifying" as well.**