# Understanding Aging of TlBr

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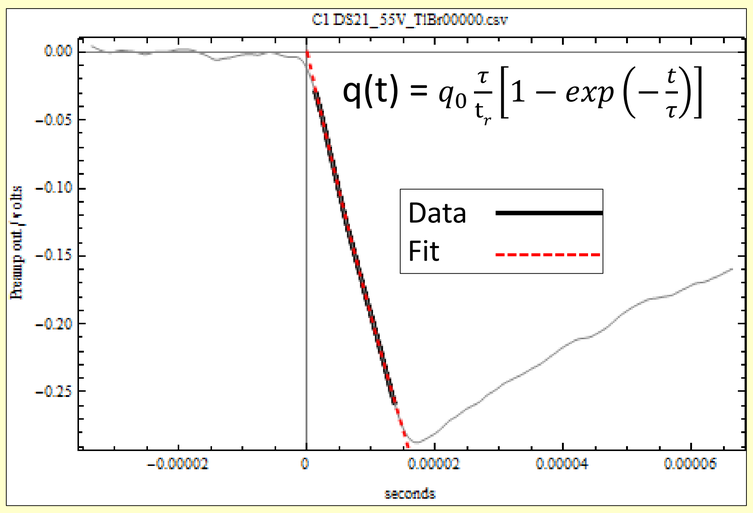
## SUMMARY

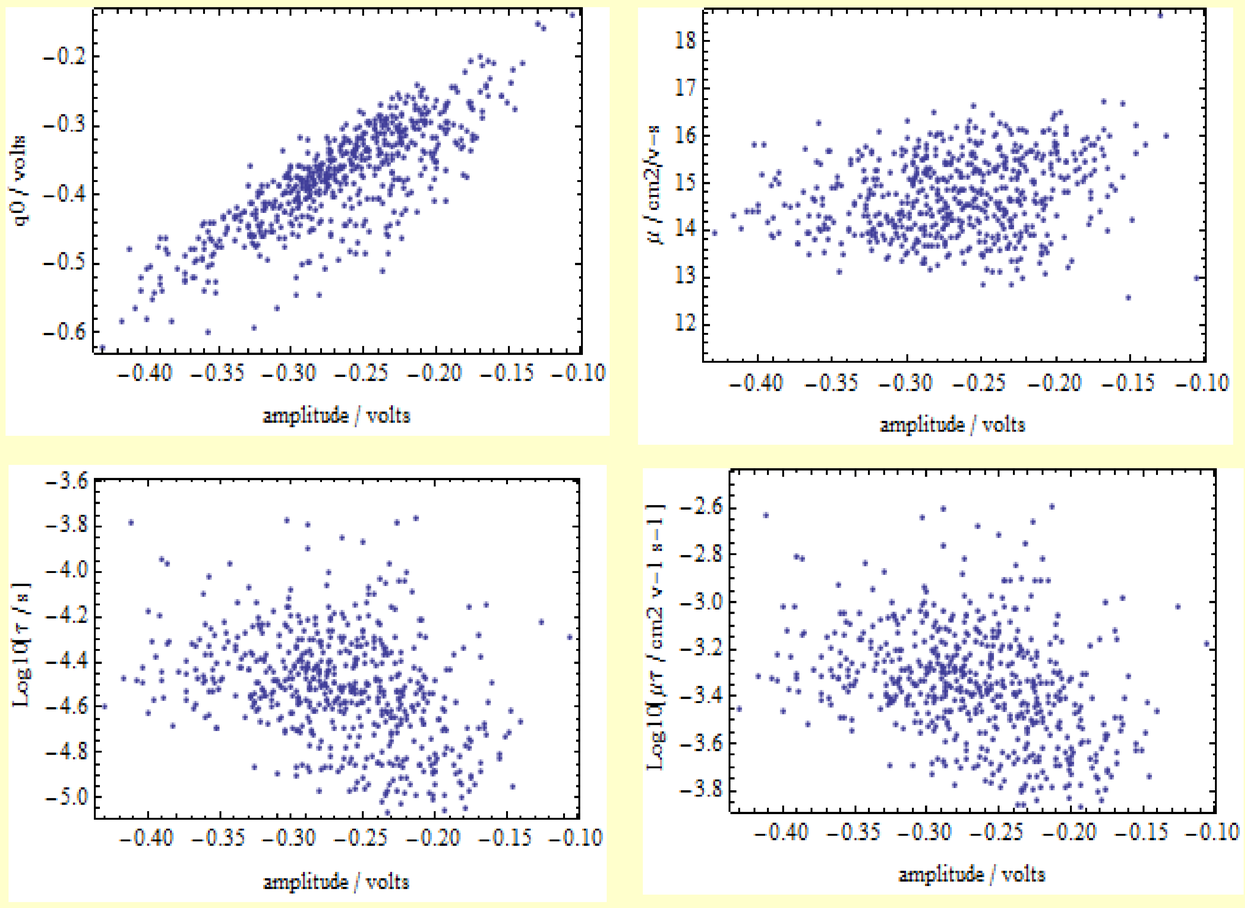
**TlBr has promising γ– and X- radiation detection properties but the performance changes with time under bias. Our charge transient experiments indicate that this may come from structural changes near the contacts, but the experiments alone cannot identify the nature of the structural changes. To enable molecular dynamics simulations of structural evolution of TlBr under external electric fields, we have developed a robust TlBr interatomic potential. Challenging simulations of ionic conduction under external fields are demonstrated, thereby enabling future work to study structural evolution under external fields.**

## Charge Transient Experiments (E = 425 Volts/cm)

1. **α particle transients induced in vacuum on the negative gold electrode were fit to the time dependent Hecht equation. This enables independent determination of charge equivalent, $q_0$, electron mobility, μ and trapping time, τ, on a pulse-by-pulse basis.**
2. **Correlation plots show pulse amplitudes are uncorrelated with μ or τ, but strongly correlated with $q_0$. We conclude that α attenuation near the negative contact caused large variance of charge generation in the active semiconductor, consistent with recent observations on second phase material growth under accelerated aging [1].**




(a) Pulse-by-pulse determination of charge $q_0$, trapping time τ, and $t_r$ = thickness/μE.




***(b)*** Correlation plots showing that variance of amplitudes is caused by variation of charge generated, not transport.

[1] "Visualization of TlBr Ionic Transport Mechanisms by the Accelerated Device Degradation Technique", Amlan Datta, Piotr Becla, and Shariar Motakef, NIMA Proceedings-D-14-00185, Submitted.

## Towards Molecular Dynamics Simulations of Ionic Conduction Induced Structure Evolution

1. **Modified the analytical functions of the Stillinger-Weber (SW) potential so that it can be used for many crystal structures as opposed to the conventional SW potential which can only be used for tetrahedral diamond-cubic, zinc-blende, or wurtzite crystals;**
2. **Implemented an equation-free (eqfree) potential model in the public molecular dynamics (MD) codes LAMMPS. The eqfree works for any modified functions for SW, Tersoff, and embedded-atom method (EAM) potentials. Hence, future modifications of such potentials no longer require changing MD codes;**
3. **Parameterized the modified SW potential for TlBr. This TlBr potential has the capability of predicting the crystalline growth of TlBr (CsCl – lattice) during MD simulations, which not only validates the lowest energy for the TlBr-CsCl, but also validates that the TlBr-CsCl is robust enough for simulations of ionic conduction under very high external electric fields.**

***(a)*** Model reproduces experimental lattice constants a, c (Å), cohesive energy $E_c$ (eV), and elastic constants $C_{11}$, $C_{12}$, $C_{13}$, $C_{33}$, and $C_{44}$ (eV/Å$^3$)

| material | structure | method | a | c | $E_c$ | $C_{11}$ | $C_{12}$ | $C_{13}$ | $C_{33}$ | $C_{44}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| **Tl** | hcp | MSW | 3.408 | 5.582 | -1.850 | 0.240 | 0.140 | 0.137 | 0.242 | 0.053 |
| | | Exp [2-4] | 3.450 | 5.520 | -1.850 | 0.277 | 0.235 | 0.187 | 0.376 | 0.055 |
| **TlBr** | CsCl | MSW | 3.985 | ----- | -2.389 | 0.275 | 0.104 | 0.104 | 0.275 | 0.067 |
| | | Exp [2-4] | 3.985 | ----- | -2.389 | 0.275 | 0.104 | 0.104 | 0.275 | 0.067 |

[2] J. D. H. Donnay, and H. M. Ondik, "crystal data, determinative tables", 3rd ed., Vol. 2 (inorganic compounds), U. S. Department of Commerce, National Bureau of Standards, and Joint Committee on Power Diffraction Standards, U. S. A., 1973; [3] I. Barin, "thermochemical data of pure substances", VCH, Weinheim, 1993; [4] G. Simmons, "single crystal elastic constants and calculated aggregate properties", Southern Methodist University Press, 1965; [5] G. E. Morse, and A. W. Lawson, J. Phys. Chem. Solids, 28, 939 (1967).

***(b)*** Model captures crystalline growth of TlBr (CsCl crystal)




***(c)*** Demonstration of simulations of interstitial ionic conduction under external fields




## Conclusions

**Transient experiments suggest that the aging of TlBr may come from structural changes near the contacts. Molecular dynamics model has been developed for TlBr. Growth simulations verifies that this model allows structural evolution to be simulated under large external electric fields. It enables future studies to identify the nature of the structural evolution near the contacts.**

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