**APPENDIX A: Nonlinear creep resistance observed for lithium indentation and the impact on linear viscoelastic analysis**

The indentation of lithium considered in the main text shows hardness values well above the yield stress (approximately 46 to 350 times higher) but still well below the elastic limit. These observations occur at indentation depths up to ~350 nm and prior to a distinctive strain burst signifying the initiation or the onset of dislocation glide plasticity. These paradoxical observations have been rationalized in the main text by application of diffusional creep mechanisms where it is considered that the indentation strain rate is the same order as the creep strain rate, a constraint that is also consistent with the analysis proposed by both Bower *et al*.1 and Ginder *et al*.2 Under these assumptions, a measure of the resistance to indentation deformation is of the form

where has units of viscosity, though not measured in pure shear. The experimental measurements of the depth dependent hardness and the strain rate for the slow strain rate experiment can be substituted into Eq. (A1) to give

and for the high strain rate experiment (constant loading rate),

where and are constants. Equations (A2) and (A3) indicate an indentation size effect in the Li results, as that there is an increasing resistance to creep deformation as increases. For a viscoelastic solid, such an increase in viscosity with strain or time would be described in the rheological nomenclature as rheopectic behavior. In considering a time (strain) dependent viscosity, Pandey *et al*.3 note ‘the reciprocity principle, which links the Laplace transforms of the creep compliance and the relaxation modulus, is not satisfied here. This is because the time-varying Maxwell model is not a time-invariant linear system, as assumed by the application of the Laplace transforms.’ The review process for this paper suggests that experts in the field are not aware that rheopectic behavior may invalidate the Lee *et al*.4 and Ting5 Laplace inversion approach to indentation creep analysis. That is, Lee *et al*.4 and Ting5 both rely on Laplace transform inversion of the Boltzmann integral for development of their indentation creep analysis. It is well known that the Boltzmann integral linkage between the creep compliance and relaxation modulus is a consequence of the applicability of the convolution theorem for the integral transform, leading to

where is the relaxation modulus (not to be confused with the reduced indentation modulus) and is the creep compliance. If Eq. (A4) is not true, then the Lee *et al*.4 inversion approach is not rigorously valid.

The purpose of this appendix is to examine if Eq. (A4) is satisfied for the Nabarro-Herring creep model proposed here and in similar form as proposed by Li *et al*.6 We will first examine if reciprocity (Eq. (A4)) holds for the Ting5 indentation analysis when the viscosity is constant with depth. This will be followed with a consideration of reciprocity when the viscosity is a function of indentation depth. Ting5 considered determining the relaxation modulus by examining the load, , required to maintain a constant *elastic* indentation depth, . For the creep compliance, Ting5 separately considered the implementation of a constant load where the indentation depth ‘creeps’ with time. For this analysis to be specific to geometrically self-similar indentation, the material response is related to hardness, , through the relationship with the load, , and indentation depth, as follows

From Eq. (A5), the time dependence of the load required to maintain a constant indentation depth, , as the initial elastic displacement in a linear viscoelastic solid is then related to the creep strain rate through the differential equation

where is taken to be a constant based on the indenter tip geometry. The solution for the time dependence of the load is

Following Ting,5 Eq. (A7) then defines the effective indentation relaxation modulus as

The indentation creep compliance is obtained through a constant load, , experiment relating the indentation strain rate from Eq. (A5) to the linear viscoelastic strain rate,

Giving the time dependence for the indentation depth,

or using the Sneddon7 solution for the elastic contact area,

Following Ting,5 this produces an expression for the effective indentation creep compliance,

Eqs. (A8) and (A12) can now substituted into Eq. (A4) to give

Equation (A13) is the expected relationship between compliance and relaxation modulus for a linear viscoelastic solid when the superposition principle, the Boltzmann integral and the deconvolution theorem are valid.8

Eqs. (A8) and (A12) are now compared with the results of Eqs. (A1) – (A13) when an indentation size dependence of the effective viscosity for the Nabarro-Herring creep mechanism is included. Again considering the constant indentation depth experiment for the relaxation modulus,

with as the initial elastic contact depth. Following the same procedure leading to Eq. (A8),

Although Eq. (A15) has the same form as Eq. (A8), it should be noted that is dependent on the initial load. That is, the relaxation modulus depends on the initial load.

For the relaxation modulus related to the proposed Nabarro-Herring creep mechanism, the variation in the indentation depth under a constant load is determined from

leading to the time dependence of the indentation depth,

Then the indentation creep compliance is

Equation (A18) is analogous to Eq. (A15) in that the creep compliance is seen to be a function of the initial load through . According to Lee *et al*.4 (p. 433), this is in an indication of nonlinearity of the viscoelastic operators. Substitution of Eqs. (A15) and (A18) into Eq. (A4) produces an integral which cannot be evaluated analytically, although clearly the result cannot be the same as in Eq. (A13). However, for small values of time in Eq. (A15), the first order Taylor series for the integrand leads to the formulation (to second order in time)

where the initial effective viscosity is

Other forms of the Taylor series expansion return similar results for the integrand consistent with loss off reciprocity. The failure of Eq. (A19) to satisfy Eq. (A4) invalidates the superposition principle used to derive the Boltzmann convolution integral, and thus application of the convolution theorem for inversion. It is on this basis that we have pursued an alternative approach to that implemented by Lee *et al*.4 and Ting.5