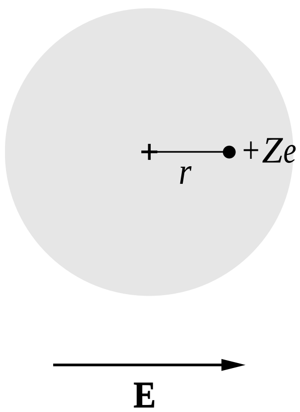
A MODEL FOR $\chi^{(3)}$, PART 1

- Assumptions:
  - ▷ Model atomic nucleus as a point charge, $+Ze$
  - ▷ Model electrons as a continuous negative charge distribution $\rho(\mathbf{r})$
    - ◦ Assume spherical symmetry:

$$\rho(\mathbf{r}) = \rho(r)$$

- Apply a uniform external electric field:



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A MODEL FOR $\chi^{(3)}$, PART 2

- Restoring force on nucleus:

$$F = \frac{Ze}{r^2} q(r) \quad \text{where} \quad q(r) = 4\pi \int_0^r \rho(r')\, r'^2 \, dr'$$

- At equilibrium, $F = ZeE$

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A MODEL FOR $\chi^{(3)}$, PART 3

- Response to applied field:
  - ▷ Atomic dipole moment:

$$\mu = Zer$$

  - ▷ Macroscopic polarization (dipole moment per unit volume):

$$P \approx N\mu$$

$$N = \text{atomic number density}$$

- **Uniform-sphere model**:

$$\rho(r) = \frac{3Ze}{4\pi a^3} \qquad (a = \text{radius of sphere of negative charge})$$

  - ▷ Restoring force on nucleus:

$$F = \frac{Ze}{r^2} q(r) = \frac{Ze}{r^2}\left(\frac{4\pi}{3} r^3 \rho\right) = \frac{(Ze)^2}{a^3} r$$

  - ▷ Displacement at equilibrium is linear in E:

$$r = \frac{a^3}{Ze} E$$

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A MODEL FOR $\chi^{(3)}$, PART 4

- Response to applied field in uniform-sphere model:
  - ▷ Atomic dipole moment:

$$\mu = a^3 E = \alpha E$$

$$\alpha = \text{atomic polarizability} \sim 10^{-23}\ \text{cm}^3$$

  - ▷ Macroscopic polarization (dipole moment per unit volume):

$$P \approx N\mu$$

$$N \sim 10^{22}\ \text{cm}^{-3}$$

  - ▷ Dielectric constant:

$$\epsilon = 1 + 4\pi\chi^{(1)} \approx 1 + 4\pi N\alpha \sim 2$$

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A MODEL FOR $\chi^{(3)}$, PART 5

- **Nonuniform-sphere model**:

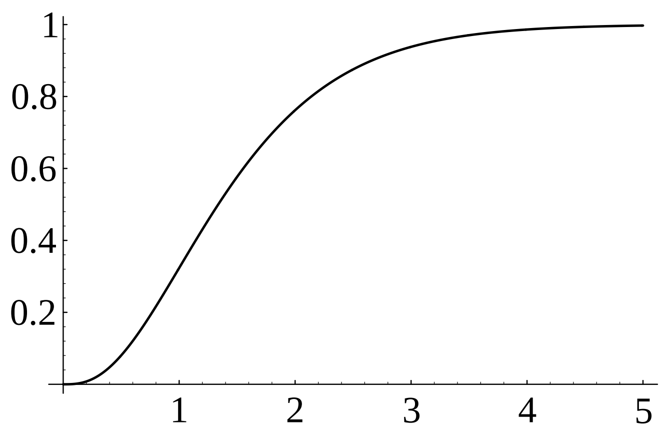
$$\rho(r) = \frac{Ze}{\pi a_0^3}\, e^{-2r/a_0}$$

$$a_0 = \text{Bohr radius of outer electrons}$$

▷ Charge inside sphere of radius r:

$$q(r) = Ze\left(1 - e^{-2r/a_0}\right) - \frac{Ze}{2} e^{-2r/a_0}\left(\frac{2r}{a_0}\right)\left[2 + \frac{2r}{a_0}\right]$$

Charge vs. radius



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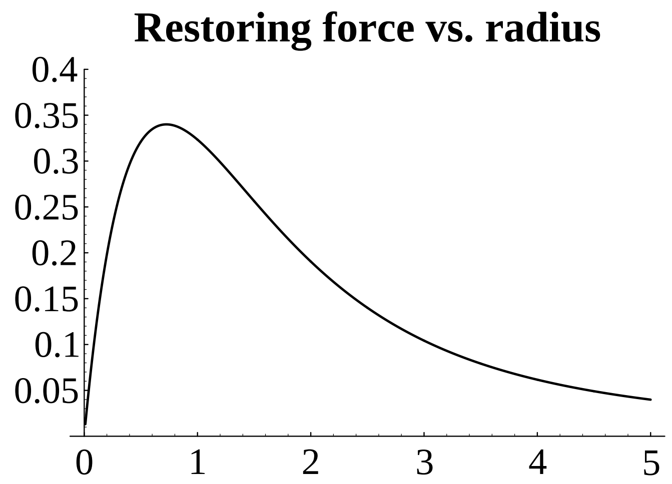
A MODEL FOR $\chi^{(3)}$, PART 6

- Response to applied field for nonuniform sphere:
  - ▷ Restoring force on nucleus at equilibrium:

$$F = \frac{Ze}{r^2} q(r)$$



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A MODEL FOR $\chi^{(3)}$, PART 7

- Taylor series for atomic dipole moment:

$$\mu = Zer(E) = Ze\left[\left.\frac{dr}{dE}\right|_{E=0} E + \frac{1}{2}\left.\frac{d^2r}{dE^2}\right|_{E=0} E^2 + \frac{1}{6}\left.\frac{d^3r}{dE^3}\right|_{E=0} E^3 + \cdots\right]$$

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A MODEL FOR $\chi^{(3)}$, PART 8

- Linear regime:
  - ▷ Charge inside radius r when $r \ll a_0$:

$$q(r) = \frac{4Ze}{3}\left(\frac{r}{a_0}\right)^3$$

  - ▷ At equilibrium,

$$E = \frac{q(r)}{r^2} = \frac{4Zer}{3a_0^3}$$

  - ▷ Linear polarizability:

$$\alpha = \frac{Zer}{E} = \tfrac{3}{4}a_0^3$$

  - ▷ Numerical estimate of $\epsilon \approx 1 + 4\pi N\alpha$:
    - ∘ Take $a_0 \approx 2 \times 10^{-8}$ cm, $N \approx 10^{22}$ cm^{-3}
    - ∘ Find $\epsilon - 1 \approx .5$ (right order of magnitude)

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A MODEL FOR $\chi^{(3)}$, PART 9

- Nonlinear regime:
  - ▷ Charge inside radius r when $r \ll a_0$:

$$q(r) \approx Ze\left[\frac{4}{3}\left(\frac{r}{a_0}\right)^3 - 2\left(\frac{r}{a_0}\right)^4 + \frac{8}{5}\left(\frac{r}{a_0}\right)^5\right]$$

  - ▷ At equilibrium,

$$E = \frac{q(r)}{r^2} \approx Ze\left[\frac{4r}{3a_0^3} - \frac{2r^2}{a_0^4} + \frac{8r^3}{5a_0^5}\right]$$

    but we need $r(E)$, not $E(r)$
  - ▷ Contribution to $\chi^{(3)}$:

$$\left.\frac{d^3r}{dE^3}\right|_{E=0} = \left.\frac{3(d^2E/dr^2) - (dE/dr)(d^3E/dr^3)}{(dE/dr)^5}\right|_{r=0} = \frac{2673}{320}\frac{a_0^7}{(Ze)^3}$$

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A MODEL FOR $\chi^{(3)}$, PART 10

- Nonlinear regime:
  - ▷ Numerical estimate of

$$\chi^{(3)} \approx \frac{Ne}{6}\frac{d^3r}{dE^3}$$

    - ◦ Take $a_0 \approx 10^{-8}$ cm, $N \approx 10^{22}$ cm^{-3}
    - ◦ Find $\chi^{(3)} \approx 0.6 \times 10^{-15}$ esu (correct order of magnitude for SiO_2)
    - ◦ Note extreme sensitivity to value of a_0

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