PHYSICAL REVIEW B VOLUME 54, NUMBER 5 1 AUGUST 1996-I

Giant enhancement of cubic nonlinearity in a polycrystalline quasi-one-dimensional conductor

David Stroud
Department of Physics, Ohio State University, Columbus, Ohio 43210

(Received 20 March 1996)

We show that the cubic nonlinear response of a polycrystalline quasi-one-dimensional conductor, such as a conducting polymer, can be expressed exactly in terms of the single-crystal cubic susceptibility and the electric fields in the analogous linear polymer. We also propose a simple nonlinear decoupling approximation which allows the polycrystalline nonlinear susceptibility to be simply estimated. Using this method, we show that local field effects may hugely enhance the nonlinear susceptibility of the polycrystal above its single-crystal value. A comparable enhancement is shown to exist in the conductivity noise. [S0163-1829(96)06229-7]

I. INTRODUCTION

Quasi-one-dimensional (1D) organic polymers may have a huge cubic nonlinear response at optical frequencies.[1] Since such a large nonlinearity is equivalent to a highly intensity-dependent dielectric function, these materials may be useful as intensity-dependent filters, self-focusing optical media, etc. Because of these possibilities, many papers have discussed possible microscopic mechanisms for this observed nonlinearity.[1]

In this work, we study, not the mechanism, but rather, how the nonlinearity of quasi-1D material may be affected if the polymer is a polycrystal. Polycrystalline structures are certainly of practical interest, because it can be difficult to prepare quasi-1D materials as single crystals. Unless special preparation techniques are used, these materials will tend to have their principal axes randomly oriented in space—in other words, to be polycrystalline.

Our main result is that this random structure may hugely enhance the effective cubic nonlinearity of the organic polymer. This enhancement is caused by a local field effect: the cubic nonlinearity depends on the cube of the *local* electric field, and this can be greatly increased above the applied field by local fluctuations in the dielectric properties.

Local field effects have already been discussed extensively for suspensions of *isotropic* nonlinear material in a linear host.[2–8] In this case, the cubic response is greatly enhanced near the frequency of the surface plasmon resonance of the suspended particles. More recently, an experiment has shown that a layered microstructure of alternating nonlinear dielectrics can have an effective cubic nonlinearity which is larger than that of of either pure component.[9] On the theoretical side, it has been shown that the enhancement factor is proportional to the averaged fourth power of the electric field in a related *linear* medium.[9–12]

In this paper, we will show that a similar result also applies to polycrystals. Using this formalism, we will show that a large enhancement can occur in a polycrystal of quasi-1D material, if the components of the dielectric tensor have suitable values.

The remainder of this paper is organized as follows. The next section describes a method for treating cubic nonlinearities in a polycrystalline material. Following this, we give some simple numerical examples, based on model calculations, and conclude with a brief discussion.

II. FORMALISM

We consider an anisotropic material, in which **D** and **E** have a nonlinear relation of the form

$$D_i = \epsilon_{ij} E_j + \chi_{ijkl} E_j E_k E_l . \tag{1}$$

Here D_i and E_i are the ith Cartesian components of **D** and **E**, ϵ_{ij} and χ_{ijkl} are second-rank and fourth-rank Cartesian tensors, and we use the Einstein convention that repeated indices are summed over. Initially, we assume that we are working at zero frequency, and that all of the quantities in Eq. (1) are real. We also assume that ϵ_{ij} is symmetric, which insures that it can be diagonalized with real eigenvalues.

We are concerned with a polycrystal of this material. The principal axes of the crystallites are assumed to be randomly oriented—that is, they point with equal probability in any direction. Equation (1) is then understood to express the constitutive relation in a coordinate system fixed in the crystallite (body coordinates). It can be converted to laboratory coordinates by a transformation using appropriate rotation matrices.

We define the effective dielectric function ϵ_e and cubic nonlinear susceptibility χ_e of the polycrystal by generalizing the method of Ref. 10. The sample is assumed to have volume V, bounded by surface S, on which the potential Φ is specified as $\Phi(\mathbf{x}) = -\mathbf{E}_0 \cdot \mathbf{x}$. This choice insures that the volume averaged electric field within V is $\mathbf{E}_0$.[13] If the polycrystal is macroscopically isotropic, ϵ_e and χ_e may be defined by the relation[10,12,14–17]

$$\langle \mathbf{D} \rangle = \epsilon_e \mathbf{E}_0 + \chi_e |\mathbf{E}_0|^2 \mathbf{E}_0 , \tag{2}$$

where $\langle\,\rangle$ denotes a volume average. It can be shown that this definition is equivalent to

$$W \equiv V \langle \mathbf{D} \rangle \cdot \mathbf{E}_0 \equiv V \{ \epsilon_e E_0^2 + \chi_e E_0^4 \} , \tag{3}$$

where we have introduced an energylike function W from which ϵ_e and χ_e may be derived.[10]

Note that even though the constitutive relationship (1) is nonlinear and anisotropic, the local fields **D** and **E** still satisfy the usual electrostatic equations

$$\nabla \cdot \mathbf{D} = 0; \tag{4}$$

$$\nabla \times \mathbf{E} = 0. \tag{5}$$

From the second of these, **E** can be expressed as the negative gradient of a scalar potential, $\mathbf{E} = -\nabla \Phi$. However, Φ no longer obeys Laplace's equation, because of the complicated (and inhomogeneous) constitutive relation.

The definition (3) can be written in another equivalent form, which is more convenient for approximations, namely

$$W = \int \{\epsilon_{ij} E_i E_j + \chi_{ijkl} E_i E_j E_k E_l\} d^3x \equiv W_2 + W_4 . \tag{6}$$

It is easy to prove, from the electrostatic equations (1), (4), and (5), that expressions (3) and (6) are equivalent. The proof follows closely the analogous proof for isotropic two-component composites.[10] Thus, to determine ϵ_e and χ_e, we need only evaluate W for the actual microstructure of the polycrystalline sample, using Eq. (6).

We now show that to *first* order in χ_{ijkl}, just as in the isotropic case, χ_e involves the fourth moment of the electric field in a related *linear* problem. We first write

$$\mathbf{E} = \mathbf{E}_{\mathrm{lin}} + \delta \mathbf{E}, \tag{7}$$

where $\mathbf{E}_{\mathrm{lin}}$ is the electric field in a linear medium with the same $\epsilon_{ij}(\mathbf{x})$ but with $\chi_{ijkl}(\mathbf{x}) = 0$; and $\delta\mathbf{E}$ is the additional electric field due to the nonlinearity. By definition, $\delta\mathbf{E}$ is at least first order in χ_{ijkl}. Since χ_{ijkl} is assumed to be a small perturbation on the linear medium, it is sufficient to calculate **W** only to first order in χ. To this order, we may neglect the contribution of $\delta\mathbf{E}$ to the fourth order term, since this will only have an effect on W of *second* order in the χ_{ijkl}'s.

The term involving ϵ_{ij} may be written

$$W_2 = \int [\epsilon_{ij}(E_{\mathrm{lin};i} + \delta E_i)(E_{\mathrm{lin};j} + \delta E_j)] d^3x. \tag{8}$$

That portion of W_2 which is *first* order in $\delta\mathbf{E}$ can be written

$$\delta W_2^{(1)} = \int [2\epsilon_{ij} E_{\mathrm{lin};i} \delta E_j] d^3x, \tag{9}$$

where we have used the symmetry of ϵ_{ij}. Writing $\delta E_j = -\nabla_j \delta\Phi$ and integrating by parts, we obtain

$$\delta W_2^{(1)} = 2\left\{ \int [\nabla_j(\epsilon_{ij} E_{\mathrm{lin};i}) \delta\Phi] d^3x - \int \nabla_j(\epsilon_{ij} E_{\mathrm{lin};i} \delta\Phi) d^3x \right\}. \tag{10}$$

But

$$\nabla_j(\epsilon_{ij} E_{\mathrm{lin};i}) = \nabla \cdot \mathbf{D}_{\mathrm{lin}} = 0, \tag{11}$$

where $\mathbf{D}_{\mathrm{lin}}$ is the displacement vector in the related linear medium, and is, like the total displacement **D**, divergence free. The integrand of the second term is the divergence of a vector whose jth component is $\epsilon_{ij} E_{\mathrm{lin};i} \delta\Phi$. Using the divergence theorem, we can convert this integral into a surface integral, which vanishes because $\delta\Phi = 0$ on S. Thus $\delta W_2^{(1)}$ also vanishes.

The effective coefficients ϵ_e and χ_e are given, according to Eq. (3), by the coefficients of VE_0^2 and VE_0^4 in W. Using the results just proven, we have

$$\epsilon_e = \frac{\int \epsilon_{ij} E_{\mathrm{lin};i} E_{\mathrm{lin};j} d^3x}{VE_0^2} \tag{12}$$

and

$$\chi_e = \frac{\int \chi_{ijkl} E_{\mathrm{lin};i} E_{\mathrm{lin};j} E_{\mathrm{lin};k} E_{lin;l} d^3x}{VE_0^4}. \tag{13}$$

Thus, just as in an isotropic composite, both ϵ_e and χ_e can be expressed (to lowest order in the nonlinearity) in moments of the electric field in the related *linear* medium.

Next, we write down a simple approximation for χ_e analogous to the ''nonlinear decoupling approximation'' (NDA) of isotropic nonlinear composites.[14] Hereafter, we drop the subscript ''lin;'' unless otherwise stated, only linear fields will be discussed. First, we prove a simple lemma which allows us to state this approximation more clearly. Denoting the eigenvalues of ϵ_{ij} by ϵ_i, we write

$$\epsilon_e = \frac{\epsilon_i \int E_i(\mathbf{x})^2 d^3x}{VE_0^2}, \tag{14}$$

where $E_i(\mathbf{x})$ is the field component parallel to the ith principal axis at $\mathbf{x}$. Equation (14) implies that

$$\frac{\langle E_i^2 \rangle}{E_0^2} = \frac{\partial \epsilon_e}{\partial \epsilon_i}, \tag{15}$$

where $\langle\ \rangle$ denotes a spatial average. This is the analog of a result in isotropic composites,[13]

$$\langle E^2 \rangle_\alpha = \frac{1}{p_\alpha} \frac{\partial \epsilon_e}{\partial \epsilon_\alpha}, \tag{16}$$

where $\langle\ \rangle_\alpha$ denotes the average of a quantity in the αth component, p_α the volume fraction of that component, ϵ_α its dielectric constant, and ϵ_e the effective composite dielectric constant.

We now consider the nonlinear susceptibility explicitly. For convenience, we will assume that, in the body coordinate system, the elements of χ all vanish except χ_{iijj}, with indices equal in pairs. Then from Eq. (13),

$$\chi_e = \chi_{iijj} \frac{\langle E_i^2 E_j^2 \rangle}{E_0^4}. \tag{17}$$

The NDA is specified by the assumption that

$$\langle E_i^2 E_j^2 \rangle \approx \langle E_i^2 \rangle \langle E_j^2 \rangle, \tag{18}$$

or, upon using Eq. (15),

$$\chi_e = \chi_{iijj} \left(\frac{\partial \epsilon_e}{\partial \epsilon_i} \right) \left(\frac{\partial \epsilon_e}{\partial \epsilon_j} \right). \tag{19}$$

This is closely analogous to the equation

$$\chi_e = \frac{1}{p_\alpha} \left(\frac{\partial \epsilon_e}{\partial \epsilon_\alpha} \right)^2 \tag{20}$$

which specifies the NDA in an isotropic composite.[14]

Equation (19) is particularly useful given an approximation for $\epsilon_e(\epsilon_1,\epsilon_2,\epsilon_3)$. The simplest of these is the effective-medium approximation (EMA),[18] which gives

$$\sum_{i=1}^{3} \frac{\epsilon_i-\epsilon_e}{\epsilon_i+2\epsilon_e}=0. \tag{21}$$

For a uniaxial material with principal elements ϵ_1, ϵ_2, and ϵ_2, Eq. (21) simplifies to

$$\frac{\epsilon_1-\epsilon_3}{\epsilon_1+2\epsilon_e}+2\,\frac{\epsilon_2-\epsilon_e}{\epsilon_2+2\epsilon_e}=0. \tag{22}$$

The physical solution to this quadratic equation must give $\epsilon_e(\epsilon_1,\epsilon_1,\epsilon_1)=\epsilon_1$:

$$\epsilon_e=\frac{\epsilon_2+\sqrt{\epsilon_2^2+8\epsilon_1\epsilon_2}}{4}. \tag{23}$$

From Eqs. (19) and (23), χ_e is greatly enhanced if $\epsilon_1 \gg \epsilon_2$, which gives

$$\epsilon_e \approx \sqrt{\epsilon_1\epsilon_2/2}. \tag{24}$$

In this regime, if only χ_{2222} is nonzero, and if $\epsilon_1 \gg \epsilon_2$, then

$$\chi_e \approx \chi_{2222}\,\frac{\epsilon_1}{2\epsilon_2}, \tag{25}$$

which gives an *arbitrarily large enhancement* if $\epsilon_1/\epsilon_2 \to \infty$.

The physical origin of this enhancement is a large increase in local electric fields. In each individual crystallite, the uniaxial (high-dielectric-constant) direction predominates in carrying displacement current. But because the crystallites are randomly oriented, there is an impedance mismatch at every grain boundary. The EMA predicts that, because of this mismatch, the polycrystal is exactly at the percolation threshold for carrying displacement current. This leads to a large local field enhancement in the low-conductivity crystal directions. Therefore, χ_e, which depends on the fourth power of the local electric field, is greatly increased over χ_{2222}.

We can also introduce a *crossover field* E_x, defined as the electric field at which the linear and nonlinear contributions to the displacement current become comparable:

$$\epsilon_e E_X=\chi_e|E_X|^2E_X. \tag{26}$$

Using the EMA

$$E_X=\sqrt{\epsilon_e/\chi_e}\approx\left(\frac{\epsilon_2}{\chi_{2222}}\right)\sqrt{\epsilon_2/\epsilon_1}. \tag{27}$$

Thus the crossover field is *reduced* relative to the corresponding field in the single crystal by a factor of $\sqrt{\epsilon_2/\epsilon_1}$—that is, nonlinear effects set in at a much lower field in the polycrystal than in the single crystal.

Next, we prove a connection between χ_e and the conductivity fluctuations in a related noise problem.[19–23] A similar theorem has also been proved for isotropic composites.[10,14] We assume that our polycrystal is a member of an ensemble of polycrystals. Each member of the ensemble is assumed to have the same geometry, but a different conductivity tensor. Let the conductivity tensor of one such member be denoted $\sigma_{ij}(\mathbf{x})+\delta\sigma_{ij}(\mathbf{x})$, where $\delta\sigma_{ij}(\mathbf{x})$ represents the fluctuating part, while $\sigma_{ij}(\mathbf{x})$ is the same for each member of the ensemble (but is position dependent). The fluctuating part is assumed to have zero mean, i.e.,

$$\langle \delta\sigma_{ij}(\mathbf{x})\rangle_{\rm av}=0, \tag{28}$$

where $\langle\,\rangle_{\rm au}$ is to be understood as an ensemble average. We assume also that

$$\langle \delta\sigma_{ij}(\mathbf{x})\,\delta\sigma_{kl}(\mathbf{x}')\rangle_{\rm av}=a_{ijkl}(\mathbf{x})\,\delta(\mathbf{x}-\mathbf{x}'). \tag{29}$$

The fluctuations in conductivity might arise, for example, from a special noise process, which causes the conductivity of the sample to fluctuate in time, such that there are no correlations between fluctuations measured at the same time but at different spatial points within the sample. The fourth-rank tensor $a_{ijkl}(\mathbf{x})$ describes the strength of these conductivity fluctuations.

Within this model, from Eq. (12), the mean-square conductivity fluctuations of the *polycrystal* are determined by

$$\begin{aligned}V^2\langle(\sigma_e+\delta\sigma_e)^2\rangle_{\rm av}E_0^4=\Big\langle \int d^3x\int d^3x'\,E_i(\mathbf{x})(\sigma_{ij}(\mathbf{x})\\ +\delta\sigma_{ij}(\mathbf{x}))E_j(\mathbf{x})E_k(\mathbf{x}')(\sigma_{kl}(\mathbf{x}')\\ +\delta\sigma_{kl}(\mathbf{x}')E_l(\mathbf{x}'))\Big\rangle_{\rm av}.\end{aligned} \tag{30}$$

We simplify this using Eqs. (28) and (29) for the ensemble averages of $\delta\sigma_{ij}$ and its square. The term on the right-hand side which involves $\sigma_{ij}(\mathbf{x})\sigma_{kl}(\mathbf{x}')$ gives $V^2\sigma_e^2E_0^4$. The cross terms involving products of σ and $\delta\sigma$ vanish. Finally, the term quadratic in $\delta\sigma$ simplifies to a single integral over $a_{ijkl}(\mathbf{x})$. When this is evaluated, we obtain

$$V\langle(\delta\sigma_e)^2\rangle_{\rm av}=a_e, \tag{31}$$

where a_e is the effective cubic nonlinear susceptibility of a medium whose linear conductivity is $\sigma_{ij}(\mathbf{x})$ and whose cubic nonlinear conductivity is $a_{ijkl}(\mathbf{x}')$. Thus, just as in an isotropic composite, there is a connection between the mean-square conductivity fluctuations in a linear polycrystal with conductivity noise, and the effective nonlinearity of a related nonlinear polycrystal.[24]

III. NUMERICAL EXAMPLE

To illustrate the predicted enhancement of nonlinear susceptibility, we consider a highly oversimplified model of a quasi-1D conductor. In the high-conductivity direction, we assume a Drude metal with dielectric function

$$\epsilon_1(\omega)=1-\omega_p^2/[\omega(\omega+i/\tau)]. \tag{32}$$

In the perpendicular direction, we postulate an insulator with a constant dielectric function,

$$\epsilon_2(\omega)=1. \tag{33}$$

Within this model, the EMA predicts that the polycrystal is precisely at the percolation threshold, so that the enhancement of χ_e will be especially large. In a possibly more realistic situation with finite conductivity in both parallel and perpendicular directions, the enhancement may be smaller.

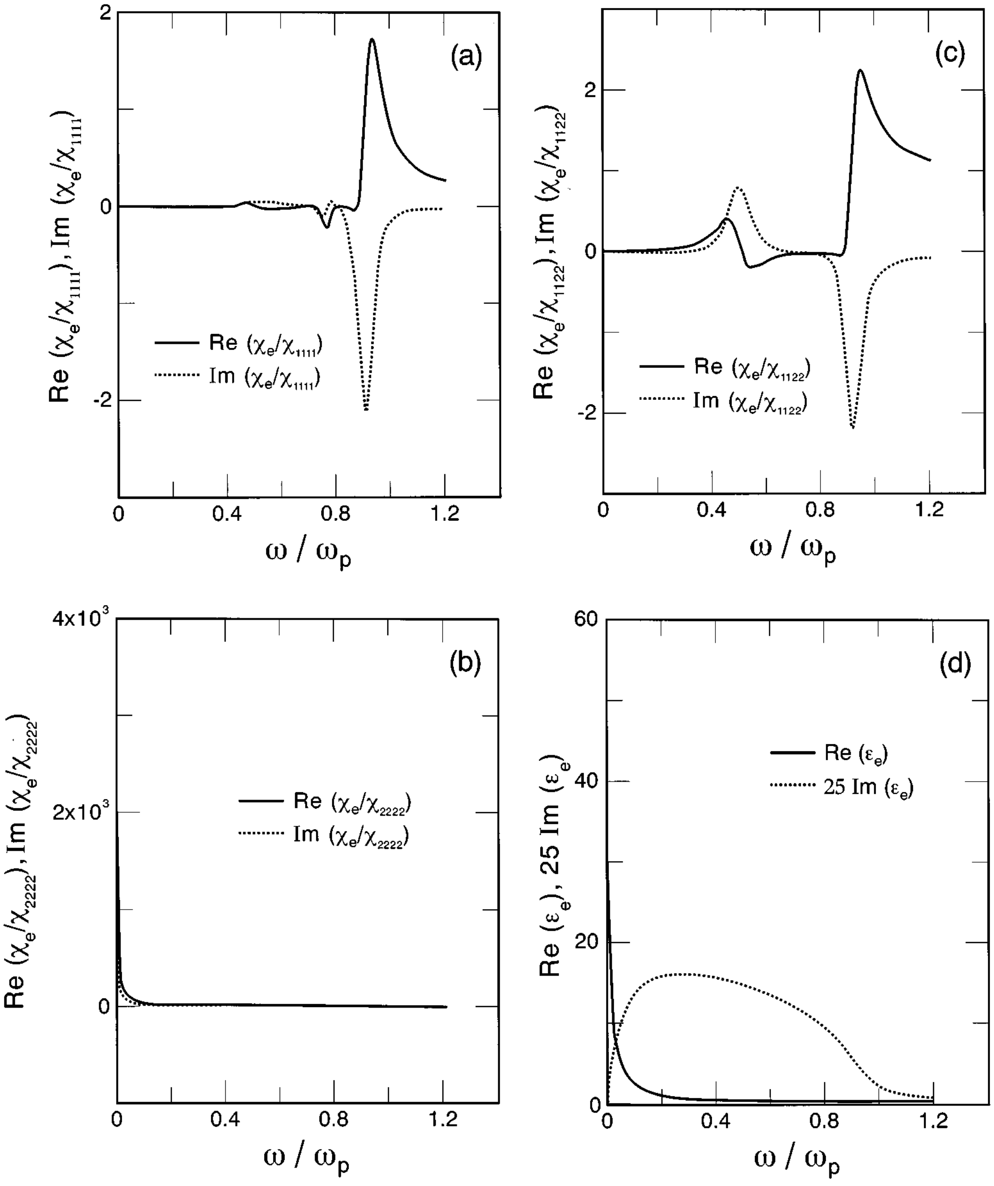

FIG. 1. (a) This is a plot of χ_e/χ_{1111} in a polycrystalline material, assuming that the only nonzero component of the single-crystal nonlinear susceptibility tensor is χ_{1111} (parallel to the high-conductivity axis). Calculation carried out using the nonlinear decoupling approximation [Eq. (19)] within the linear effective medium approximation [Eq. (22)], with single-crystal dielectric functions given by Eqs. (32) and (33) with $\omega_p\tau=10$. (b) Same as (a) except we plot χ_e/χ_{2222} assuming only component χ_{2222} is nonzero. (c) Same as (a) except χ_e/χ_{1122} is plotted, assuming only χ_{1122} is nonzero. (d) Plot of the linear dielectric function $\epsilon_e(\omega)$ in the EMA.

Because the principal components ϵ_1 and ϵ_2 are, in general, complex at finite frequencies (as are the components of the nonlinear susceptibility tensor), Eq. (17) must be modified slightly. The appropriate generalization is

$$\chi_e = \chi_{iijj} \frac{\langle E_i^2 |E_j|^2 \rangle}{E_0^4}, \tag{34}$$

where E_i and E_j are in general complex. This form takes correct account of the phase relationships between $\mathbf{E}$ and $\mathbf{E}_0$ at finite frequencies. The generalized NDA is now

$$\chi_e = \chi_{iijj} \left(\frac{\partial \epsilon_e}{\partial \epsilon_i} \left| \frac{\partial \epsilon_e}{\partial \epsilon_j} \right| \right). \tag{35}$$

To apply this approximation to the present example we solve Eq. (22) for ϵ_e and compute the specified derivatives, assuming $\omega_p\tau=10$. The results are shown in Fig. 1. Clearly, there is an enormous enhancement at low frequencies provided χ_{2222}, the transverse component, is nonzero. This large enhancement occurs because the ratio $|\epsilon_1/\epsilon_2|$ becomes arbitrarily large at low frequencies. If χ_{2222} is nonzero, there is only a modest enhancement due to the parallel components of the tensor at higher frequencies. Note that the predicted enhancement may be somewhat magnified by the use of the EMA and the NDA, which predicts that the polycrystal is precisely at the percolation threshold. In addition, if our model dielectric function had a lesser contrast between the high and low conductivity directions, the enhancement would be smaller, with or without the NDA.

IV. CONCLUSIONS

We have derived an exact formal expression for the cubic nonlinear susceptibility χ_e of a polycrystal. As previously shown for composites with isotropic components, we find that χ_e in a polycrystal depends entirely on the fourth moments of the electric field in the *linear* limit. To evaluate these moments, we have also described a simple nonlinear decoupling approximation for estimating χ_e from the corresponding linear dielectric function. This approximation predicts that χ_e can be enormously enhanced in a polycrystal at appropriate frequencies, provided that the constitutive parameters of the single crystal are appropriately chosen. In particular, if the component χ_{2222} of the nonlinear susceptibility in the direction perpendicular to the high conductivity axis is nonzero, there may be a large enhancement in χ_e. A similar enhancement is demonstrated for the corresponding conductivity noise in a polycrystalline material.

The present results may be of practical importance. As noted in the Introduction, many quasi-one-dimensional conductors have large cubic nonlinear susceptibilities, whose origins have been attributed to a wide variety of microscopic mechanisms. But whatever the microscopic origin, these materials may be even more nonlinear as polycrystals than as single crystals. Since polycrystals are often more easily prepared than single crystals, this result may lead to a method for preparing even more strongly nonlinear conducting polymers. It would be of great interest, therefore, to test these predictions experimentally.

ACKNOWLEDGMENTS

It is a pleasure to thank Professor A. J. Epstein, Professor P. M. Hui, and Dr. Ohad Levy for many useful conversations. I am also most grateful to Professor S. Doniach and the Department of Applied Physics at Stanford University for their warm hospitality while this work was being completed. This work was supported by NSF Grant No. DMR94-02131.

---

[1] For recent reviews of nonlinear optical properties of organic polymers, see, for example, the articles in *Nonlinear Optical Properties of Organic Polymers*, edited by David J. Williams, ACS Symposium Series No. 233 (American Chemical Society, Washington, D.C., 1983), or in *Organic Materials for Nonlinear Optics*, edited by R. Hann and D. Bloor (Royal Society of Chemistry, London, 1989), Vols. I and II.

[2] For recent reviews, see, e.g., the articles in J. Opt. Soc. Am. B **6**, 531 (1989); or C. Flytzanis, Prog. Opt. **29**, 2539 (1992).

[3] R. Jain and R. C. Lind, J. Opt. Soc. Am. **73**, 647 (1983).

[4] D. Ricard *et al.*, Opt. Lett. **10**, 511 (1985); F. Hache *et al.*, J. Opt. Soc. Am. B **3**, 1647 (1986); F. Hache *et al.*, Appl. Phys. A **47**, 347 (1988).

[5] P. Roussignol *et al.*, J. Opt. Soc. Am. B **4**, 5 (1987).

[6] See, for example, D. Stroud and Van E. Wood, J. Opt. Soc. Am. B **6**, 778 (1989); A. E. Neeves and M. H. Birnboim, *ibid.* **6**, 787 (1989); Y. Q. Li *et al.*, *ibid.* **6**, 814 (1989).

[7] J. W. Haus *et al.*, J. Opt. Soc. Am. B **6**, 797 (1989); J. W. Haus *et al.*, Phys. Rev. A **40**, 5729 (1990); M. J. Bloemer *et al.*, IEEE J. Quantum Electron. **26**, 1075 (1990).

[8] V. Shalaev and collaborators have carried out extensive calculations of the nonlinear response of fractals. See, e.g., A. V. Butenko *et al.*, Sov. Phys. JETP **67**, 60 (1988); A. V. Butenko *et al.*, Z. Phys. D **10**, 81 (1988); S. G. Rautian *et al.*, JETP Lett. **47**, 243 (1988); V. M. Shalaev *et al.*, Physica A **185**, 181 (1992).

[9] R. W. Boyd *et al.*, Phys. Rev. Lett. **74**, 1871 (1995).

[10] D. Stroud and P. M. Hui, Phys. Rev. B **37**, 8719 (1988).

[11] A. Aharony, Phys. Rev. Lett. **58**, 2726 (1987).

[12] D. J. Bergman, Phys. Rev. B **39**, 4598 (1989).

[13] D. J. Bergman, Phys. Rep. **43**, 378 (1978).

[14] X. C. Zeng *et al.*, Phys. Rev. B **38**, 10 970 (1988); X. C. Zeng *et al.*, Physica A **157**, 192 (1989).

[15] N. Kalyaniwalla *et al.*, Phys. Rev. A **42**, 5613 (1990).

[16] K. W. Yu *et al.*, Phys. Rev. B **47**, 14 150 (1993).

[17] C. S. Yang and P. M. Hui, Phys. Rev. B **44**, 12 559 (1991).

[18] D. A. G. Bruggeman, Ann. Phys. (Leipzig) **24**, 636 (1935); R. Landauer, J. Appl. Phys. **23**, 779 (1952).

[19] R. Rammal *et al.*, Phys. Rev. A **31**, 2662 (1985).

[20] R. Rammal *et al.*, Phys. Rev. Lett. **54**, 1718 (1985).

[21] L. de Arcangelis *et al.*, Phys. Rev. B **31**, 4725 (1985); **34**, 4656 (1986).

[22] M. A. Dubson *et al.*, Phys. Rev. B **39**, 6807 (1989).

[23] S. W. Kenkel and J. P. Straley, Phys. Rev. B **29**, 6299 (1984).

[24] In our discussion of noise, we discuss conductivity fluctuations rather than fluctuations in dielectric constant, since most noise measurements deal with conductivity noise. Since at zero frequency **J**, like **D**, is divergenceless, all the relevant equations and derivations are identical in form for both types of noise.