Nonlinear Dielectric Materials*

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Summary—**The following is a brief description of nonlinear dielectrics from the standpoint of the fundamental physics involved. Specific available materials and technical applications are considered only by way of illustration of general properties. It is shown that, although dielectric nonlinearity and ferroelectricity are quite different phenomena, the fact that ferroelectrics have high dielectric constants makes them the most likely materials to exhibit a high degree of nonlinearity at electric field strengths safely below the breakdown values.**

Introduction

THE MACROSCOPIC electromagnetic properties of matter are commonly described by specifying the current density J, electric displacement D, and magnetic induction B as functions of the electric and magnetic "intensities" E and H. In the majority of substances these relations are found to be linear, leading to the definitions of the conductivity, dielectric constant, and permeability tensors. The meaning of the word "linear" must be made more precise as soon as we consider time-varying fields; in particular, we must be careful to distinguish between nonlinearity and dispersion. The simple statement that $D(t)$ is proportional to $E(t)$ will not do; by common usage it is understood that linearity is concerned with such a proportionality in the frequency domain rather than in the time domain. Thus, consider a Fourier integral representation of the fields:

$$E(t) = \int_{-\infty}^{\infty} E(\omega)e^{i\omega t}d\omega$$
$$D(t) = \int_{-\infty}^{\infty} D(\omega)e^{i\omega t}d\omega \tag{1}$$

By linearity we mean that the material is characterized by some unique function $\epsilon(\omega)$ such that

$$D(\omega) = \epsilon(\omega)E(\omega). \tag{2}$$

This corresponds in general to no simple relation between $D(t)$ and $E(t)$. Thus, a linear dielectric material is characterized intuitively by the following conditions:

1. No frequencies are present in $D(t)$ which not are present in $E(t)$.
2. If $E_1(t)$ produces $D_1(t)$, which we write compactly as $E_1 \rightarrow D_1$ and $E_2 \rightarrow D_2$, then $(a_1E_1+a_2E_2) \rightarrow (a_1D_1+a_2D_2)$, with similar definitions for linear conductors and magnetic media.

It is possible to have one but not both of these conditions satisfied: for example, consider the corresponding magnetic properties of water placed in a strong but inhomogenous magnetic field. For applied frequencies within the range of the proton Larmor frequencies, condition 1 is satisfied but not the superposition condition 2.[1]

Strictly speaking, all such linear laws may be regarded as approximations for several different reasons. In the first place, they would presumably fail in any material substance at sufficiently high field strengths; *i.e.*, breakdown or saturation effects would occur. Secondly, there really are no unique relations between the above vectors since in the work of the highest accuracy one would always expect to find that, for example, the electric displacement does not depend only on the electric field, but also on every other physical condition of the material such as temperature, state of stress and strain, degree of illumination, and even the entire past history of the specimen. Thus, even if it should be found that D is exactly proportional to E at constant temperature, the fact that the dielectric constant so defined is a function of temperature gives rise to electrocaloric effects, in which a sudden change in electric field produces a change in temperature. Thus, the linearity or nonlinearity of a substance could depend on its degree of thermal contact with its surroundings; *i.e.*, on whether it is operated under isothermal or adiabatic conditions. Similarly, if a dielectric is linear under conditions of constant stress, it might not be so under conditions of constant strain. In practice, however, these are usually extremely small effects.

Finally there are more esoteric examples provided by modern physical theories, according to which even a perfect vacuum should have nonlinear properties. In quantum electrodynamics one finds that the phenomenon of scattering of light by light (a violation of condition 2) should occur due to the formation and subsequent annihilation of electron-positron pairs;[2] the cross-section for this process is, however, so small that experimental confirmation is not to be thought of. Another effect is predicted by General Relativity; an electromagnetic field contains energy and therefore mass. This produces a gravitational field which can in turn deflect a light beam, again in violation of condition 2. Once again, we do not expect any experimental confirmation in the laboratory! In order to be extremely cautious about the experimental situation, however, we note that electrical measurements of the highest accuracy are never performed with intense fields; if appreciable deviations from Maxwell's equations did occur in free space at field strengths in excess

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[1] F. Bloch, *Phys. Rev.*, vol. 70, p. 460; 1946.
A. Bloom, *Phys. Rev.*, vol. 98, p. 1105; 1955.
[2] O. Halpern, *Phys. Rev.*, vol. 44, p. 855; 1934.
H. Euler and B. Kockel, *Naturwiss.*, vol. 23, p. 246; 1935.

of 10^5 v/cm or 10^5 oersted, they would almost certainly have escaped experimental detection thus far.[3]

In spite of the above considerations, the vast majority of substances is found to be linear to a high degree of accuracy at all field strengths commonly attained, hence we denote as "nonlinear" only those substances in which there are substantial and easily demonstrable effects arising from violation of conditions 1 or 2 above, occurring at easily produced field strengths. Each such substance is potentially capable of important technical applications, since by violation of condition 1 we generate harmonics and beat frequencies, while violation of condition 2 enables one to modulate one signal by the presence of another.

In the case of conductivity and permeability, substances which are nonlinear in the above sense have long been known; thyrite and, to a certain extent, electrolytic cells and various solid-state devices, such as rectifiers, may be regarded as media with nonlinear conductivity (more correctly as circuit elements with nonlinear conductance), while the hysteresis and saturation effects in ferromagnetic materials respresent great nonlinearity. By contrast, nonlinear dielectric materials, although not entirely unknown in the past, have not until recently been available in forms of interest to electrical engineers. Probably the earliest known example of dielectric nonlinearity was the discovery, over 80 years ago, of the Kerr electro-optical effect.[4] Glass and many liquids, in particular nitrobenzene, develop optical birefringence in fairly strong electric fields; thus the dielectric constant at optical frequencies varies with a low-frequency electric field in violation of the superposition condition. An example of a nonlinear capacitance at microwave frequencies is provided by crystal rectifiers, particularly the germanium welded-contact variety,[5] in which the barrier capacitance varies strongly with bias voltage. By far the most important nonlinear dielectrics, however, are the ferroelectric crystals or ceramics.

Ferroelectrics

From a phenomenological point of view, ferroelectricity may be defined as the electric analog of ferromagnetism, and the fundamental criterion of ferroelectricity is the existence, at certain temperatures, of hysteresis between D and E. The static electric displacement then depends not only on the applied electric field but also on the past history, in such a way that with sufficiently slow periodic variation of E we obtain a D-E hysteresis loop exactly like the familiar B-H curves of ferromagnetic materials. Above a certain temperature T_c, called the Curie point, this hysteresis disappears, but the relation between D and E may remain appreciably nonlinear up to temperatures far above T_c. Since oscillograms illustrating these effects have been published recently in this journal,[6] they will not be repeated here. In the neighborhood of the Curie point more complicated phenomena are sometimes found.[7]

Several distinct classes of ferroelectrics, with widely different chemical composition and crystal structure, are now known. The first to be discovered was Rochelle salt (sodium potassium tartrate tetrahydrate), widely used for its piezoelectric properties.[8] This substance appears to be unique in that it possesses two Curie temperatures (-18°C and $+23$°C) and is ferroelectric between them. Other ferroelectric tartrates[9,10] show only a single Curie point. Mueller[11] has shown that the electrical, mechanical, and thermal properties of Rochelle salt can be correlated very satisfactorily by a single thermodynamic free-energy function valid on both sides of the Curie points. This is important not only from the standpoint of economy of description, but it indicates that the ferroelectric-"paralectric" phase transition at the Curie points is probably not a very drastic rearrangement from a molecular point of view as is the case in many phase transitions, for example, that between diamond and graphite.[12] This conclusion seems well established also for the other classes[13] of ferroelectrics.

Another class of ferroelectrics is represented by the salt potassium dihydrogen phosphate, KH_2PO_4 and other substances of similar chemical composition and crystal structure (*i.e.*, the alkali and ammonium phosphates and arsenates).[14] Although they have found applications based on their electro-optical properties, the fact that their Curie points are at liquid-air temperatures limits their usefulness as nonlinear dielectrics in the purely electrical sense. KH_2PO_4 is at present unique in that the molecular mechanism of its properties (different arrangements of hydrogen bonds) is probably

[3] There is, of course, indirect evidence associated with atomic theory suggesting that the laws of electrostatics hold at field strengths far beyond this limit; for example, the fact that the wavelengths of the spectral lines of hydrogen can be calculated to great accuracy on the assumption that the electric field of the nucleus is a coulomb field. However, this could hardly be called an electrical measurement.

[4] M. Born, "Optik," p. 365, J. Springer, Berlin; 1933.

[5] H. C. Torrey and C. A. Whitmer, "Crystal Rectifiers," Chap. 13, M.I.T. Radiation Laboratory series No. 15; McGraw-Hill Book Co., Inc., New York; 1948.

[6] W. P. Mason and R. F. Wick, "Ferroelectrics and the dielectric amplifier," Proc. IRE, vol. 42, pp. 1606–1620; November, 1954.

[7] W. J. Merz, *Phys. Rev.*, vol. 91, p. 513; 1953.

[8] W. G. Cady, "Piezoelectricity," McGraw-Hill Book Co., Inc., New York, 1946.

[9] W. J. Merz, *Phys. Rev.*, vol. 82, p. 562; 1950, vol. 83, pp. 226, 656; 1951.

[10] B. T. Matthias and J. K. Hulm, *Phys. Rev.*, vol. 82, pp. 108; 1951.

[11] H. Mueller, *Phys. Rev.*, vol. 47, p. 175; 1935, vol. 57, pp. 829–842; 1940, vol. 58, pp. 565–805; 1941, *Zeit. Krist.*, vol. 99, p. 122, 1938, *Ann. N. Y. Acad. Sci.*, vol. 40, p. 321; 1940.

[12] R. Smoluchowski, et al., "Phase Transformations in Solids," John Wiley & Sons, New York; 1951.

[13] H. R. Danner and R. Pepinsky, *Phys. Rev.*, vol. 99, p. 1215; 1955.

[14] G. Busch and P. Scherrer, *Naturwiss.*, vol. 23, p. 737; 1935.
G. Busch, *Helv. Phys. Acta.*, vol. 11, p. 269, 1938.
C. C. Stephenson and J. G. Hooley, *Phys. Rev.*, vol. 56, p. 121; 1939.
W. Bantle and P. Scherrer, *Nature*, vol. 143, p. 980; 1939.
J. and K. Mendelssohn, *Nature*, vol. 144, p. 595, 1939.

understood with greater certainty than for any other ferroelectric.[15,16]

The third class of ferroelectrics,[17] represented by the cubic form of barium titanate, $BaTiO_3$ and several similar substances, is at the present time the most interesting from a theoretical standpoint and the most useful in terms of applications. The Curie point of $BaTiO_3$ is at 120°C, and at room temperature the most nearly perfect crystals grown to date[7] exhibit hysteresis loops with coercive force as low as 600 v/cm and a spontaneous polarization of about 26 microcoulombs /cm^2, very much higher than in the first two classes of ferroelectrics. The dielectric properties of $BaTiO_3$ single crystals above the Curie point have been measured by Drougard, Landauer, and Young,[18] with results that may be summarized as follows. For crystals that are free to distort in accordance with their electrostrictive properties (*i.e.*, under conditions of zero stress), the behavior is given within experimental error by the free-energy function

$$F(P, T) = F_0(T) + A(T)P^2 + B(T)P^4, \tag{3}$$

where P is the dielectric polarization, F_0 the free energy at zero polarization (which is irrelevant for dielectric properties, although it largely determines the specific heat of the material), and A, B are linear functions of temperature, given by the empirical equations

$$\begin{aligned} A &= 3.8 \times 10^{-5}\,(T - 105) \\ B &= 4.5 \times 10^{-15}\,(T - 175), \end{aligned} \tag{4}$$

which are in cgs units, with the temperature in degrees centigrade. The electric field is, from thermodynamics,

$$E = \partial F/\partial P = 2AP + 4BP^3. \tag{5}$$

Therefore, the incremental (small signal) dielectric constant is

$$\begin{aligned} \epsilon &= 1 + 4\pi(\partial P/\partial E) = 1 + 4\pi/(2A + 12BP)^2 \\ &\simeq 4\pi A^2/(2A^3 + 3BE^2), \end{aligned} \tag{6}$$

the approximation being valid at field strengths for which the cubic term in (5) is small compared to the linear one. Since B is negative in the temperature range where these experiments were performed (119°C to 150°C), we have the rather surprising result that application of a biasing field increases the dielectric constant of the crystal. This phenomenon is seen clearly in the oscillograms of Merz,[7] and may be shown by thermodynamic arguments[19] to be connected with the fact that the crystals exhibit a first-order transition at the Curie point; *i.e.*, as the temperature is lowered, the spontaneous polarization jumps discontinuously from zero to a finite value (about 18 microcoulombs /cm^2). Some of the first crystals grown, which were less perfect, exhibited a second-order transition and, as required by thermodynamics, a positive sign of B.[20] It must be remembered, however, that this increase due to bias occurs only under conditions of zero stress, and therefore can be seen only at sufficiently low frequencies, below all of the mechanical vibration modes of the crystal. Measurements made at Stanford University by Mr. V. Varenhorst at frequencies of 20, 40, and 120 mcs showed in all cases a decrease in dielectric constant with bias voltage. The results were complicated by temperature hysteresis effects which persist above the Curie point and are not understood, but in a typical case at 40 mcs and 130°C, application of a biasing field of 1700 v/cm lowered the dielectric constant from 6,700 to 6,000.

Growth of good single crystals of $BaTiO_3$ is still a difficult and costly art, and most of its applications to date have involved the ceramic material, often with various added impurities. The ceramic also exhibits dielectric nonlinearity, a typical result[6] being a decrease of dielectric constant with biasing field such that a field of 10 kv/cm lowers ϵ from 1,400 to 1,100, while a field of 30 kv/cm lowers it to 700. Similar results have been found at Stanford University, with an interesting additional qualitative observation that the loss tangent of a ceramic at radio frequencies may be lowered substantially by application of a biasing field of a few kv/cm. Further data on properties of ceramics have been given by von Hippel.[21]

Many details concerning the physical properties of $BaTiO_3$ have been omitted here; in particular the phenomena of domain formation and motion which are essential to an understanding of the properties of single crystals below the Curie point. These have been described by Forsbergh,[22] Merz,[23] and Little.[24] A recent discussion of the theory of ferroelectrics has been given by Devonshire.[25]

Theory of Dielectrics

It might be supposed that with modern knowledge of the properties of atoms and molecules, it would be a straightforward matter to calculate the dielectric constant of any material of known composition from first principles. Unfortunately, this turns out to be an extremely complicated problem on which little progress has been made; only in the case of gases where the dielectric constant is very close to unity and the similar case of dilute solutions of polar molecules in nonpolar liquids can one claim quantitative success. Although it

[15] J. C. Slater, *Jour. Chem. Phys.*, vol. 9, p. 16; 1941.
[16] C. C. Stephenson and J. G. Hooley, *Jour. Am. Chem. Soc.*, vol. 66, p. 1937; 1944.
[17] von Hippel, Breckenridge, Chesley, and Tisza, *Jour. Ind. Eng. Chem.*, vol. 38, p. 1097; 1946.
[18] M. E. Drougard, R. Landauer, and D. R. Young, *Phys. Rev.*, vol. 98, p. 1010; 1955.
[19] E. T. Jaynes, "Ferroelectricity," Princeton University Press, Princeton, N. J., Chap. 3; 1953.
[20] W. J. Merz, *Phys. Rev.*, vol. 76, p. 1221; 1949.
[21] A. von Hippel, "Dielectric Materials and Applications," John Wiley & Sons, New York; 1954.
[22] P. W. Forsbergh, Jr., *Phys. Rev.*, vol. 76, p. 1187; 1949.
[23] W. J. Merz, *Phys. Rev.*, vol. 88, p. 421, 1952; vol. 95, p. 690; 1954.
[24] E. A. Little, *Phys. Rev.*, vol. 98, p. 978; 1955.
[25] A. F. Devonshire, *Phil. Mag. Suppl.*, vol. 3, p. 85; 1954.

is easy to describe in words the mathematical procedure that would give a rigorous treatment, in practice one must make from the start drastic simplifications which make it difficult to interpret whatever agreement or disagreement with experiment is found. The basic reason for this is that the properties of a crystal are not merely the properties of a single molecule, multiplied by the number of molecules present, but the interactions between them are an essential part of the picture, and these are never taken into account in a correct way. From the point of view of quantum mechanics, we would have to say that it is not meaningful in any precise way to speak of the states and behavior of individual molecules or atoms, but the only really correct attitude is the global one in which we enumerate the possible quantum states (wave functions) of the crystal as a whole. Almost without exception, however, theoretical treatments of dielectric properties of solids have been based on the concepts developed by Clausius and Mosotti about 100 years ago. Here one regards a solid as composed of a large number of polarizable objects (in various cases the mechanism of polarization may be thought of as distortion of electronic distributions of atoms or ions, motion of ions, or rotation of molecular aggregates having a permanent dipole moment) each with polarizability α, so that each object, in an electric field F, develops a dipole moment

$$M = \alpha F. \tag{7}$$

The field F is not, however, the same as the macroscopic applied field E; because of the interaction of the polarizable objects with each other, there is an additional term commonly taken as proportional to the net polarization, with a proportionality constant β, known as the Lorentz factor.

$$F = E + \beta P. \tag{8}$$

Lorentz showed that if the polarizable objects are arranged in a cubic or random array, and each maintains the same constant dipole moment (no thermal agitation effects), β would have the value $4\pi/3$. If there are N of these polarizable objects per unit volume, the polarization is

$$P = N\alpha F = N\alpha(E + \beta P),$$

so that the dielectric susceptiblity becomes

$$\chi = P/E = N\alpha/(1 - N\alpha\beta). \tag{9}$$

Introducing the dielectric constant

$$\epsilon = 1 + 4\pi\chi, \tag{10}$$

and assuming the Lorentz value $\beta = 4\pi/3$, we arrive at the well-known Clausius-Mosotti formula

$$\frac{\epsilon - 1}{\epsilon + 2} = \frac{4\pi N\alpha}{3}, \tag{11}$$

which is presented in some textbooks as if it were a rigorous relation.

Many refinements of this treatment have been made, and a very complete account of them may be found in the recent book of Böttcher.[26] They have led to some improvement in agreement with experiment but not to any appreciably deeper understanding, because the basic concepts remain the local polarizability and local field F, which in modern theory no longer have a precise meaning. Nevertheless, this classical treatment contains enough of the truth to be very useful in giving a qualitative understanding of dielectrics, provided certain precautions are observed. In the first place, (11) cannot be used when the polarizability is due to freely rotating permanent dipoles (*i.e.*, polar molecules) except in the case of high dilution when it reduces to

$$\epsilon - 1 = 4\pi N\alpha \ll 1.$$

From statistical mechanics, one can calculate the polarizability of a rotating dipole of moment M, with the result $\alpha = M^2/3kT$, with k Boltzmann's constant and T the temperature in degrees Kelvin. Eq. (11) predicts an infinite dielectric constant, *i.e.*, ferroelectricity, when $N\alpha\beta > 1$, so that this should occur at sufficiently low temperatures for any substance with rotating dipoles. However, if we insert the numerical values, we find that many polar substances should be ferroelectric at temperatures far above their boiling points! This is the famous "$4\pi/3$ catastrophe," which was resolved by Onsager[27] with the observation that strong correlations between the motions of nearby dipoles reduce the effective Lorentz factor; the results of his approximate treatment are obtained if we formally replace β in the above equations by $4\pi/(2\epsilon+1)$; the opposite conclusion is then obtained that ferroelectricity does not occur unless the polarizability becomes infinite.

The fact that ferroelectricity is so easy to "explain" when one uses poor mathematical approximations has long plagued theoreticians and has delayed any reliable understanding of the true cause of ferroeelctricity. For example, Rochelle salt was for many years treated as an assembly of rotating dipoles exhibiting the catastrophe of (11). Another example of a model which predicts ferroelectricity as a result of poor approximation is an array of harmonic oscillators interacting with each other. Such an oscillator with a particle of charge e, mass m, and resonant frequency ω has a temperature-independent polarizability of $e^2/m\omega^2$, and therefore according to the above equations one can always produce a ferroelectric array by making the resonant frequency sufficiently small. However, this model is so simple that it can be treated rigorously; an orthogonal transformation of coordinates enables one to find the states of the array as a whole, and it is found when the problem is treated correctly that ferroelectricity cannot occur unless the polarizability of a single oscillator becomes infinite. We

[26] C. J. F. Böttcher, "Theory of Electric Polarisation," Elsevier Press, Amsterdam; 1952.
[27] L. Onsager, *Jour. Am. Chem. Soc.*, vol. 58, p. 1486; 1936.

also note that Slater's theory of KH_2PO_4, already mentioned as probably the best available theory of a ferroelectric, does not make use of electrostatic interactions but rather ones of a more direct mechanical nature. In spite of these and other considerations,[28] many people believe that in barium titanate we have the realization of the classical polarization catastrophe; in the present writer's opinion it is doubtful whether any existing theory is free of the effects of poor approximations of the above type, and interactions other than electrostatic may well be the essential ones.

If, disregarding these precautions, we assume that a classical type of theory should have at least a qualitative usefulness, what can be said about the expected occurrence of dielectric nonlinearity? According to the above equations, this would require that either the polarizability or the Lorentz factor be field-dependent. We consider separately the three cases that the polarization is due to: (a) rotating permanent dipoles; (b) translational motion of ions; or (c) electronic distortion of atoms or ions.

(a) *Rotating Dipoles.* The polarizability $\alpha = M^2/3kT$ given above is an approximation valid only at field strengths F such that $MF \ll kT$. The exact expression, first calculated by Langevin in 1905, is

$$\alpha = (M/F)L(a) = M^2/3kT - M^4F^2/45k^3T^3 + \cdots,$$

where $L(a) = \coth a - a^{-1}$ is the Langevin function and $a = MF/kT$. From this we find that at $a = 1$ the polarizability is lowered by about 6 per cent, and for $a > 5$, $L(a)$ is essentially equal to unity, so that α varies inversely as the internal field F. Since molecular dipole moments are of the order of 10^{-18} esu, we find that at room temperature an appreciable nonlinearity could be expected only for F greater than about 4×10^4 esu, or 1.2×10^7 v/cm. Since we must remember to use the Onsager field for F, the applied field E would have to be of the same order of magnitude; thus a measurable nonlinearity due to saturation of rotating dipoles could be expected only at very low temperatures and intense field strengths.

(b) *Translational Motion of Ions.* Here the prospects are considerably brighter. In many types of crystals the size of the lattice is determined by the larger ions that have to fit into it, and if small ions are also present, they may be free to move in the interstices, through distances of an appreciable fraction of an Angstrom, but cannot move further due to contact with the larger ions. It is seen without any calculation that this results in a contribution to the polarization of the crystal which saturates rather abruptly at a certain value. KH_2PO_4 is undoubtedly of this type, with movable hydrogens; other crystals of similar structure, even though not ferroelectric, might be expected to show nonlinearity. However, materials with high dielectric constants should provide the most favorable possibilities, since according to the above equations we then obtain an internal field F which is considerably "amplified" above the applied field E. If appreciable motion occurs, the Lorentz factors might also vary.

(c) *Electronic Distortion.* As a simple example, consider an atom which has a ground state ψ_0 with energy E_0, and an excited state ψ_1 with energy E_1, such that the matrix element of the dipole moment operator between them,

$$M_{01} = e \int_{\psi_0} {}^*z\psi_1 dV$$

does not vanish.[29] Using quantum mechanics and statistical mechanics,[30] the following formula for polarizability may be obtained:

$$\alpha = \frac{|M_{01}|^2}{kT} \cdot \frac{\tanh a}{a}$$

where

$$a = \frac{[4(E_1 - E_0)^2 + |M_{01}|^2F^2]^{1/2}}{kT}$$

It is seen that appreciable nonlinearity requires that a be of order unity or greater and that the term in F^2 must contribute substantially to a. Therefore, since M_{01} will typically be of the order of magnitude 10^{-18} esu, if the two energy levels are sufficiently close together the situation is about the same as in the case of rotating dipoles. If a high dielectric constant leads to great internal field strengths, these conditions might be met, although it appears that the case of movable ions remains the most favorable to development of strong nonlinearity.

[28] J. M. Luttinger and L. Tisza, *Phys. Rev.*, vol. 70, p. 954, 1946; vol. 72, p. 257; 1947.

[29] L. I. Schiff, "Quantum Mechanics," McGraw-Hill Book Co., Inc., New York, sec 25; 1949.

[30] See ref. 19 (pp. 58–60) for a similar calculation.

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