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Formation of polarization-echo and polarization-avalanche signals in nonpiezoelectric dielectrics

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We develop a quantum-mechanical theory of coherent polarization effects that can arise not only in piezoelectrics but also in all physically inhomogeneous nonpiezoelectric dielectrics. An anharmonic mechanical oscillator coupled with an electric dipole moment is used as the model. It is shown that this model can describe simultaneously both the polarization echo and the polarization avalanche. The polarization avalanche can be illustratively represented as the relaxation of the oscillations of an initially compressed or bent body after the external driving force has been removed. The theory is constructed using the formalism of cyclotron and ferrimagnetic echo, thus demonstrating that all these effects have a common physical character, being concrete representations of one and the same dynamic property of nonlinear physical systems.

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INTRODUCTION

An arbitrary system of weakly interacting particles, excited independently of one another by an external coherent field, can generate under certain conditions superradiant signals of the Bloch-induction,[1] Hahn echo,[2] or boson avalanche[3] type. All these signals are generating by an oscillating macroscopic polarization formed by a set of coherently excited multipoles. This common property of all the induction, echo, and avalanche phenomena can point the way to the search and prediction of new analogous physical effects.

The search for these coherent effects can follow two directions: a) Search for new types of "carriers" that radiate multipoles unconnected with spins but having individual physical properties and definite oscillation eigenfrequencies. Examples of such carriers are powder particles, domain wals, fluxoids, rotons, dislocations, and others, b) Search for multipoles of new physical nature and connected with the spins. The most interesting example of this kind is the discovery of elastic multipoles connected with the electron and nuclear spins and generating spin-phonon echo signals.[4] Of course, greatest interest attaches to observation of new types of radiating multipoles connected with carriers of a new type.

By now, echo-type signals have been observed in such physical objects as ferrimagnetic powders[5] and ferroelectric and piezoelectric powders (see, e.g., Ref. 6–8). The echo signals in powders are due to oscillating polarization of the electric dipoles that are either spontaneously present in the sample (ferroelectrics), or result from piezoelectric coupling (piezoelectrics). These phenomena were named "polarization echo" (PE).

We show in this paper that PE can be observed in a large new class of powdered materials—powdered centrosymmetric dielectrics. The piezoelectric properties of centrosymmetric dielectrics are the result of the inhomogeneous distribution of the charge in a layer near the surface. These dielectric will be called hereafter "local piezoelectrics."

It has become obvious, following the publication of Ref. 9, that echo-type signals can be observed in two principally different quantum systems: of the spin type, wherein the energy spectrum of each particle is bounded, and of the oscillator type, where the spectrum of an individual particle is equidistant and not bounded from above. This result found a general theoretical explanation in terms of abstract Lie algebras. It has turned out that echo signals arise whenever an algebra that described the unified properties of an external generator and of a quantum system becomes unsolvable. As to echo of the spin type (STE) or of the oscillator type (OTE), it can be expressed in terms of concrete representations of abstract unresolvable Lie algebra. The OTE has a number of important and interesting features compared with STE: under certain conditions the signal increases without limit with increasing excitation; ac-

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cumulation of the signal intensity with increasing number of exciting pulses is possible. These features explain the large group of practical applications that came into being after the observation of the OTE. In only one respect did the oscillator systems turn out to be less promising than the spin systems: because of the unbounded energy spectrum, it is impossible to obtain in them population inversion and effects of the boson avalanche (BA) type.

It is shown in the present paper that under sufficiently strong anharmonicity of the oscillators, when their spectrum becomes nonequidistant, BA becomes possible. Thus, various oscillator systems with ultralong phase-memory times can also be used to generate powerful electromagnetic and acoustic field pulses in the BA regime.

In Sec. 1 we obtain the Hamiltonian of a locally piezoelectric particle having nonlinear elastic properties. The PE is generated by a sample consisting of a set of such particles with different eigenfrequencies. In Sec. 2 we derive a formula for the intensity of the PE signal. We show that a system of locally piezoelectric "powder granules" produces echo signals whose intensity increases initially with increasing time interval between the pulses; this is confirmed by experimental data.[10] The identity of the theoretical result of Ref. 10 with the previously known result of Ref. 11 is established. It follows hence that the term "electroacoustic echo" introduced[12] after the "polarization echo" concept[13] describes a phenomenon that is just as quantum-mechanical as STE, a premise rejected by a number of investigators. In Sec. 3 we consider BA generation in a system of anharmonic oscillators. Only a photon avalanche (PTA) can develop in a powder, since the "powder granules" are not coupled acoustically with one another. The onset of a phonon avalanche (PNA) is possible in those cases when the oscillators are "packed" in a single crystal or in glass.

1. HAMILTONIAN OF LOCALLY PIEZOELECTRIC PARTICLE

The fact that dielectric crystals have surfaces causes a space charge to be produced in them, and one of the consequences of this charge is an inhomogeneous distribution of the polarization over the crystal.[14] The polarization in the surface layer produces a piezoelectric coupling between the external electric field and the mechanical stress, irrespective of whether the material is piezoelectric or not.

All the calculations pertain to a piezoelectric plate with the following dimensions and parameters: a, b, and d are respectively the length, width, and thickness; c, q, ε, and ρ are respectively the elastic modulus, the electrostriction constant, the permittivity, and the density. According to Ref. 15, the axes and dimensions can be chosen such that all the electric and mechanical quantities are scalars, viz., the polarization $P=(0,0,P_z)$, the electric field intensity $E=(0,0,E_z)$, the strain $S=(S_1,0,0,0,0,0)$ and the mechanical stress $T=(T_1,0,0,0,0,0)$. In the nonferroelectric phase the average polarization of such a plate is $P_0=0$, therefore the charge distribution inside the plate is characterized by a polarization moment

$$M_p=\int_{-z_0}^{z_0} P_0(z)\,z\,dz, \tag{1}$$

where z is the coordinate along the plate thickness.

The stresses produced in the two surface layers of the sample have opposite signs, so that the constant electric field leads to a bending of the plate, and the alternating field leads to flexural oscillations. Taking the nonlinear elastic effects into account, we can write down the electromechanical equations in the form

$$T=c_2S+c_3^{\text{eff}}S^2+c_4^{\text{eff}}S^3-q(P_0+P)^2, \tag{2}$$

$$E=(\varepsilon_0\varepsilon)^{-1}(P_0+P)-2q(P_0+P)S, \tag{3}$$

where

$$S=z\,\partial^2v/\partial x^2=zr,\quad c_3^{\text{eff}}={}^3/_2c_2+3c_4,\quad c_4^{\text{eff}}={}^1/_2c_2+6c_3+4c_4.$$

Here P_0 is the static internal polarization, P is the harmonic polarization induced by the field $E\exp(i\omega t)$, v is the transverse displacement of the plate, and c_2, c_3, and c_4 are the second, third, and fourth order elastic moduli. Solving (2) and (3) we can obtain the potential energy of the sample in an external field:

$$W_p=\frac{c_2abd^3}{24}(1-l^2)r^2+\frac{c_4^{\text{eff}}abd^5}{320}r^4-pE, \tag{4}$$

where $l^2=48q^2M_p^2\varepsilon\varepsilon_0/d^4c_2$, and the electric dipole moment of the plate is given by

$$p=\varepsilon\varepsilon_0ab\cdot 2qM_pr. \tag{5}$$

The curvature of the plate in the equilibrium state can be obtained from Eq. (3):

$$r_{eq}=\frac{24qM_pE\varepsilon\varepsilon_0}{d^3c_2(1-l^2)}, \tag{6}$$

using the equilibrium value of the bending moment

$$M=b\int_{-z_0}^{z_0}T(z)\,z\,dz=0.$$

Solving the equation of motion for the bending plate with the boundary conditions ($M=0$, $\partial M/\partial x=0$, $x=\pm x_0$, where x is the coordinate along the sample), we obtain the natural frequency of the oscillations

$$\omega_0=\frac{3^{1/2}\pi^2d}{32a^2}\left[\frac{c_2(1-l^2)}{\rho}\right]^{1/2}, \tag{7}$$

and the kinetic energy of the plate

$$W_k={}^1/_2m^{\text{eff}}\dot r^2, \tag{8}$$

where $m^{\text{eff}}=\rho abd(4a/3\pi)^4$. The total energy of the sample in an external field

$$W=\frac{m^{\text{eff}}\omega_0r^2}{2}+\frac{m^{\text{eff}}\dot r^2}{2}+\frac{c_4^{\text{eff}}abd^5}{320}r^4-pE \tag{9}$$

is the energy of an anharmonic oscillator executing flexural oscillations under the influence of an electric field.

In operator form, the interaction of such an anharmonic oscillator with an electric field is described by an unperturbed Hamiltonian

$$\mathcal{H}_0=\hbar\omega_0(a^+a+{}^1/_2)+\mu\hbar\omega_0(a^++a)^4 \tag{10}$$

and by an interaction Hamiltonian

$$\mathcal{H}_1=-p_0E_0(a^+e^{-i\omega t}+ae^{i\omega t}), \tag{11}$$

where $p_0=\varepsilon\varepsilon_0 ab\cdot 2qM_p(\hbar/2m^{\text{eff}}\omega_0)^{1/2}$ is the dipole-moment constant, ω is the field frequency, $\mu=c_{\text{eff}}^4 abd^5\hbar/4\times320(m^{\text{eff}})^2\omega_0^3$ is the interaction constant, and a^+ and a are creation and annihilation operators defined for our problem by the formula

$$a(a^+)=\frac{1}{2^{1/2}}\left[\left(\frac{m^{\text{eff}}\omega_0}{\hbar}\right)^{1/2}r\pm i\frac{m^{\text{eff}}}{(m^{\text{eff}}\omega_0\hbar)^{1/2}}\dot{r}\right]. \tag{12}$$

Using the technique proposed in Ref. 16, we can rewrite the unperturbed Hamiltonian $\mathcal{H}_0$, with accuracy to first order in $\mu\hbar\omega_0$, in diagonal form

$$\mathcal{H}_0'=\hbar\omega_0(a^+a+1/2)+6\mu\hbar\omega_0[(a^+a+1/2)^2+1/2], \tag{13}$$

and the interaction Hamiltonian in the form

$$\mathcal{H}_1'=\mathcal{H}_1+(\text{rapidly oscillating terms}). \tag{14}$$

2. POLARIZATION ECHO IN POWDERS OF LOCAL PIEZOELECTRICS

The total Hamiltonian of a powdered sample consisting of a large number N of the anharmonic oscillators considered in the preceding section can be written in the form

$$\mathcal{H}=\sum_{j=1}^{N}\left\{\hbar\omega_0^j\left(a_j^+a_j+\frac{1}{2}\right)+\mu_j\hbar\omega_0^j\left[\left(a_j^+a_j+\frac{1}{2}\right)^2+\frac{1}{2}\right]-p_0^jE_0^j(a_j^+e^{-i\omega t}+a_je^{i\omega t})+\hbar\Delta\omega_j a_j^+a_j\right\}, \tag{15}$$

where the last term describes effects of inhomogeneity of the local field, with $\Delta\omega$ the parameter of the scatter of the local field.

An electric-field pulse E_1 of duration Δt_1 applied at the instant $t=0$ induces in the system a coherent electric dipole

$$p=\sum_{j=1}^{N}p_j$$

and causes the moment to rotate around the field direction at a frequency ω. After the pulse is turned off, this macroscopic coherent moment becomes dephased within a characteristic relaxation time T_2^* because of the presence of local inhomogeneities in the system, but the dipole moments of the individual particles are preserved. A second pulse E_2 of duration Δt_2 applied at the instant of time $t=\tau$ restores the phases of the individual dipole moments and produces an echo at the instant of time $t=2\tau$. In our case the echo formation and the restoration of the phases are due to the nonlinear anharmonic coupling of the particle oscillations after the first pulses with the oscillations after the second pulse.

A quantum-mechanical calculation of the PE signal by the technique proposed in Ref. 11 yields for the maximum echo-signal intensity at the instant $t=2\tau$ the formula

$$I_{\max}=I_0N^2\Theta_1^2\Theta_2^4e^{-4\tau/T_2}(\mu+2\omega_0\mu\tau)^2, \tag{16}$$

where the factor $\exp(-4\tau/T_2)$ describes the damping of the oscillations in the system, I_0 is the radiation intensity of an individual dipole moment, $\Theta_1=\hbar^{-1}E_1\Delta t_1 p$, $\Theta_2=\hbar^{-1}E_2\Delta t_2 p$. Let us compare the result with that of Ref. 10, where PE signals were considered in piezoelectric powders and all the calculations were made in the framework of classical physics. In that paper they calculated the maximum electric voltage signal at the instant of time $t=2\tau$ following the action of two electric-field pulses; this voltage was generated in a circuit containing the investigated piezoelectric powder:

$$V_{\max}(t)=\frac{3}{160}G_R(0)c_4\frac{K^4}{(1+K^2)^3}\frac{\omega T_2}{2}\frac{\varepsilon_0}{\varepsilon}\frac{\varepsilon_0^2}{\rho^2}\frac{m^4(d)}{d^7}\times abN\Delta t_1\Delta t_2^2E_1E_2^2e^{-2\tau/T_2}(1-e^{-2\tau/T_2}). \tag{17}$$

Here $G_R(0)$ is the rf response of the circuit and of the receiver, $K^2=e^2(\varepsilon c_2)^{-1}$ is the electromechanical coefficient, e is the piezoelectric coefficient, $m(d)=d\sin kz$, k is the wave vector of the fundamental mode of the oscillations of the piezoelectric plate,

$$N=G_\Omega(0)\int_{-\infty}^{\infty}G_1(\xi)G_2^2(\xi)\,d\Omega \tag{18}$$

is the number of all the modes of all the particles [$G_\Omega(0)$ describes the mode distribution], $\Omega\equiv\omega-\omega_0$ is the difference frequency,

$$G(\xi)=\frac{\sin\xi}{\xi}e^{-i\xi},\quad \xi=\left(\omega+i\frac{1}{T_2}\right)\frac{\Delta t}{2}. \tag{19}$$

The remaining quantities in (17) have the same meaning as in this paper. Expression (17) can be shown to be identical with

$$V_{\max}(t)=G_R(0)\frac{3}{160}\frac{abdc_4}{\varepsilon\varepsilon_0\hbar}p_1\left(\frac{1}{1+K^2}\right)^3\frac{\omega T_2}{2}\times(\hbar^{-1}E_1\Delta t_1p_1)(\hbar^{-1}E_2\Delta t_2p_1)^2Ne^{-2\tau/T_2}(1-e^{-2\tau/T_2}). \tag{20}$$

Comparing the quantum-mechanical maximum intensity $I_{\max}$ (16) with the maximum power determined from (20) we can see that they coincide if we put

$$I_0=C\left[G_R(0)\frac{3}{160}\frac{abdc_4}{\varepsilon\varepsilon_0\hbar}p_1\left(\frac{1}{1+K^2}\right)^3\frac{\omega T_2}{2}\right]^2\frac{\omega}{2\pi}, \tag{21}$$

where C is the capacitance of the capacitor that contains the powder sample, and

$$p_1\equiv\hbar\varepsilon_0Km(d)d^{-2}\rho^{-1/2}.$$

3. PHOTON AND PHONON AVALANCHES IN POWDERS OF LOCAL PIEZOELECTRICS

We consider now the evolution of a system of local piezoelectrics with nonequidistant spectrum in a negative absolute temperature state produced by a short electric-field pulse. The relaxation process is accompanied by generation of a boson field, and the bosons of this field have different wave vectors k. Under real conditions, however, bosons are produced with wave vectors $\mathbf{k}_0$ and with minimum damping. When these appear, all the particles begin to interact via this boson field and the system goes over into a superradiant state described by some parameter φ. If the real system is described by a two-level model, then $\sin^2(\varphi/2)$ turns out to be proportional to the probability of the boson-induced transition of the particle from the ground state to an excited state. According to Ref. 3, the intensity of the coherent spontaneous radiation is determined by the formula

$$I(t)=I_0(\mathbf{k}_0)\frac{N^2\lambda^2\operatorname{ctg}^2(\varphi_1/2)}{A(e^{\eta t}+\operatorname{ctg}^2(\varphi_1/2)e^{-\eta t})^2}, \tag{22}$$

where $\eta \equiv I_0(\mathbf{k}_0)N\lambda^2/\hbar\omega_0 A$, $\varphi_1 = \varphi(t_0)$, A is the cross-section area of the radiating end face, $I_0(\mathbf{k}_0)$ is the intensity of the coherent radiation of an isolated particle into a unit solid angle in the k direction, and λ is the radiation wavelength. From (22) we can obtain the temporal characteristics of the avalanche, namely the formation time t_0 and the duration t_R

$$t_0 = \frac{1}{2}\frac{(\ln N)A\tau^{(0)}}{N\lambda^2}, \quad t_R = \frac{A\tau^{(0)}}{N\lambda^2}, \tag{23}$$

where $\tau^{(0)}$ is the spontaneous relaxation time of an isolated particle.

Since the powder particles are not coupled acoustically with one another, no phonon avalanche (PNA) can develop in such a sample. The main characteristic of the photon avalanche (PTA) in a local-piezoelectric powder is the spontaneous pseudospin–photon relaxation time, defined by

$$\tau_{phot}^{(0)} = \hbar V_0/4\pi Q_{el} p^2, \tag{24}$$

where Q_{el} is the electric quality factor and V_0 is the volume of the sample. Since the electric oscillations are coupled with the elastic oscillations of the powder particles, an important role is played in the PTA formation by the acoustic quality factor Q_{ac}. The elastic-oscillation losses characterized by Q_{ac} decrease the number of active particles and shorten the relaxation times $T_1 > t_0$ and $T_2 > t_0$, and this may suppress the PTA signal.

The formalism presented above can be used to describe avalanche-like processes in individual dielectric crystals. These crystals can have point defects and dislocations that violate the local symmetry in a region $l_0 \ll \lambda_s$, where λ_s the sound wavelength. In this case ω_0 is no longer the natural frequency of an eleastic resonator as in the case of a powder particle, but the natural frequency of an electromagnetic or of an electromagnetic-elastic system. These locally perturbed regions generate phonons if they can be inverted by electric excitation. To find the spontaneous lifetime of such a perturbation in a crystal, we resort to the analogy between electromagnetic and elastic oscillations. We shall speak of the phonon radiated by an object produced in a locally piezoelectric region just as we speak of photons radiated by an electric dipole. In the case of a two-level model, the interaction between the elastic dipole of the jth object with the phonon field in a crystal is described in the form

$$U_{elast} = G\varkappa, \tag{25}$$

where E_{elast} is the elastic energy of the individual object and is given in (4), G is the pseudospin-phonon interaction constant, and $\varkappa = ra^2/d$ is the relative deformation. If the deformation is produced by zero-point elastic oscillations of the jth object

$$\varkappa_0^j = \frac{a^2}{2d}\left(\frac{12\hbar\omega_0}{d^3abc_2}\right)^{1/2}, \tag{26}$$

then the time of the pseudospin-phonon relaxation is[4]

$$\tau_{phon}^{(0)} = \frac{2\hbar\rho V_0 v_{s_j}^2}{9G^2 Q_{ac}}. \tag{27}$$

We now estimate the PTA and PNA relaxation time for a $BaTiO_3$ crystal in the nonferroelectric phase with the following dimensions and parameters: $a = 10^{-3}$ m, $b \sim d = 10^{-4}$ m, $q = 3\times10^3$ m·kg^{3}C^{-1} sec, $M_p = 3\times10^{-11}$ C, $\varepsilon = 10^3$, $c_2 = 14\times10^{10}$N·m^{-2}, $\rho = 6\times10^3$ kg·m^{-3}, $v_s = 5\times10^3$ m·sec^{-1}, $\omega_0 = 10^6$ rad·sec^{-1} [from expression (7)], $\varkappa_0 = 10^{-12}$, and $G = 5\times10^{-17}$ J·(def.un.)$^{-1}$. For a voltage 1 V the electric dipole moment of the sample is $p = 4\times10^{-18}$ C·m. The relaxation times are then $\tau_{phon}^{(0)} = 10^{-2}Q_{ac}^{-1}$ sec and $\tau_{phot}^{(0)} = 10^{-8}Q_{el}^{-1}$ sec.

We see thus that such locally perturbed regions as dielectric powder particles or domainlike formations in dielectric high-symmetry single crystals can possess simultaneously both electric and elastic multipoles and can furthermore generate signals of the BA type. The type of the boson field generated in the BA process will depend on the sample preparation and on the excitation conditions. Intense polarization avalanches (PA) will be observed at sufficiently long polarization-memory times. These times are substantially longer in samples with layered structure: the polarization excitation formed in one of the layers is transmitted by the electric field pulse to another layer with long memory time. For example, a ZnO layer in a monolithic or powdered sample with layers of the type $(n-Si)+(SiO_2)+(ZnO)$ +electrodes can increase the memory time from 10^{-3} – 10 sec to 10 hours at an excitation frequency 130 MHz, a pulse duration 10^{-6} sec, and an electric field amplitude 50 V·cm^{-1}.[17]

CONCLUSION

Our results add greatly to the number of substances in which PE and PA signals can be sought. Since practically all real bodies contain various imperfections and defects, it follows that at sufficiently low temperatures, sufficiently short pulses, and sufficient sensitivity of the receiving apparatus it is possible to observe in them echo responses. On the other hand, modern microelectronics is capable of producing artificial systems of electromagnetic and elastic multipoles in which echo signals of the type discussed can be observed. It seems that these signals are promising means of estimating the laws governing the distributions of the physical parameters of these systems. A circumstance of greater interest is that artificial construction of a system of microscopic oscillators is automatically accompanied by the appearance of "memory" properties that can be used to control these systems and to produce internal-memory elements. The physical cause of this phenomenon is that the microscopic semiconductor elements begin to exchange virtual photons and phonons, and this leads to formation of collective properties in these elements.

The possibility of BA generation by a system of anharmonic oscillators leads to the conclusion that all intensive actions on substances should be accompanied by avalanche-like responses, inasmuch as the nonlinear properties of matter begin to manifest themselves as a rule in the case of strong actions. Since the avalanches start out from a position of unstable equilibrium, the type of action that produces this unstable state is quite immaterial, i.e., avalanche processes are an effective method of generating certain coherent physical fields at the expense of the energy of other fields, irrespective of the relation between their wave vectors. For example,

an electromagnetically compressed substance can generate spontaneously hypersound of very short wavelength.

Note added in proof (6 February 1979). Let us describe more accurately the principle of finding the number N of the anharmonic oscillators, which determines the PA intensity, and the role of the piezoelectric plates containing these oscillators. It follows from our paper that the macroscopic parameters of the plate determine the resonant frequencies of the microscopic oscillators, i.e., the plates form acoustic resonators for the active centers. When the plate oscillates, alternating molecular electroelastic fields are produced in its volume and on its surface. These fields can polarize the centers both via orientation mechanism and by displacements of the free surface and volume charges. The most interesting properties of the electric polarization occur when the electroacoustic oscillations of the sample deflect considerably the active centers from their equilibrium positions in the solid matrix and drive them into the excited potential minima (traps). This property of electric and magnetic crystals has been in use for a long time to record information contained in acoustic and electromagnetic waves, and various physical mechanisms of this internal memory have been investigated. The observation of PE in these same substances shows that the formation of a quantum superposition of the states of the center in the principal minimum and in the trap makes it possible to generate spontaneous acoustic and electromagnetic radiation within the framework of free and induced inductions, and also of ordinary and stimulated echo. It is remarkable that the time of capture of the center by the trap plays the role of the time of longitudinal pseudospin relaxation T_1, which determines the time limit of the appearance of the stimulated PE. Since the powder granules can have high Q's, a case of nonresonant PE excitation is possible, described by a generalization of formula (16):

$$I_{\max}=I_0N^2\left[\frac{\omega_1\omega_2^2}{\nu_1\nu_2^2}\left(\frac{\omega_0-\omega_{01}}{\nu_1}\Theta_1+1\right)\Theta_1\Theta_2^2\right]^2\mu^2e^{-4\tau/T_2}(1+2\omega_0\tau)^2,$$

$$\Theta_\alpha=\nu_\alpha\Delta t_\alpha,\quad \nu_\alpha=[(\omega_0-\omega_{0\alpha})^2+\omega\alpha^2]^{1/2},\quad \omega_\alpha=\hbar^{-1}E_\alpha p,\quad \alpha=1,2,\quad \Theta_\alpha<1.$$

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