Acoustic spectroscopy of DNA in the gigahertz range

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We find a parametric resonance in the gigahertz range of DNA dynamics, generated by pumping hypersound. The resonance may be accompanied by the formation of localized phonon modes due to the random structure of elastic modulii of DNA.

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I. INTRODUCTION

We aim at finding *possible* effects of resonance attenuation of hypersound propagating in a sample of molecules of DNA. Our actual goal is to urge experimentalists to start looking for this. To that end we suggest that one should employ the interaction between solvents and molecules of DNA to generate phonon modes of DNA by pumping GHz excitations in the liquid. The strong viscous interactions of molecules and solvent may result in a dragging that could promote torsional phonon modes generating the interstrand ones and result in the additional absorption of hypersound. Thus, the study of solutions of DNA could provide important information as to the nature of the liquid state, precisely through taking into account effects of dissipation in solvents in the GHz range. It is worth noting that hypersound acoustics has made considerable progress in recent years and has become a powerful method in experimental research [1–4]. In the context of DNA it may turn out to be a valuable means for studying hydrodynamic phenomena.

The DNA molecule has unusual elastic properties owing to its helicoidal symmetry and the sequence of base pairs. From the physical viewpoint, i.e., neglecting its genetic information, the latter looks random. Therefore, elastic excitations of the DNA take place in random media. According to general theory (see Ref. [5]), they can be localized, that is, confined to a limited set of base pairs. It is generally accepted that the vibrational dynamics of DNA has a bearing upon biological phenomena in cells ([6]; see also Ref. [7]). One may expect that the phenomenon of localization could be of importance as well.

A specific feature of the elastic dynamics of the DNA is elastic phonon modes in the duplex. Their existence has been confirmed by the experimental research using Raman scattering [8–12], far-infrared absorption [13,14], and Brillouin scattering [15]. In Ref. [16] the vibrational modes have been studied using submillimeter-wave absorption spectroscopy in the range ∼0.01–10 THz. Thus, Woolard *et al.* [16] have found multiple dielectric resonances in the long-wavelength portion of the submillimeter-wave regime, i.e., ∼1–30 cm^{-1}, which they ascribe to phonon modes of DNA. These results should provide valuable methods of biodiagnostics [17]. But, to our knowledge, so far there have been no experimental work that investigated the generation of the phonon modes by external means.

Theoretical study of phonon modes of DNA has revealed elastic excitations of the duplex that may correspond to the approximate helicoidal symmetry of a molecule of DNA [18,19]. It is important that phonon modes of DNA are believed to be strongly attenuated owing to an interaction with ambient medium. The effect can be mitigated, to some extent, by preparing samples of appropriate character. Thus, films formed by molecules of DNA appear to be less prone to the attenuation in regard to phonon modes. In contrast, it is especially strong in liquid solutions of DNA. It should be noted that the estimates are based on the Navier-Stokes hydrodynamics in the *sub*-GHz range and, to an even larger extent, the classical Stokes formula for the viscous drag at small Reynolds numbers [20]. This argumentation fails in the GHz range. Specifically, Van Zandt [21] (see also Ref. [22]) demonstrated that underdamped phonon modes can exist within the framework of Maxwell hydrodynamics; see also Ref. [23]. Thus, it remains to be explored whether some phonon modes are damped.

In this paper we follow the analysis performed by Chia C. Shih and S. Georghiou who have put forward powerful arguments in favor of the existence of underdamped vibrational modes of a molecule of the DNA, besides the overdamped ones [24]. The authors visualize a molecule of DNA as a duplex of two strands formed by sugar-phosphate backbones framed by base pairs located inside the strands and assume that the bases are shielded by sugar phosphates from the bombardment by molecules of solvent, the base pairs being influenced by the ambient medium indirectly, through their interaction with the backbone. The motion of the bases has the shape of librations inside the cages formed by the sugars of the backbone. Consequently, the dynamics of the backbone is damped owing to the strong attenuation caused by the medium, whereas that of the base pairs turns out to be underdamped. Their angular frequencies are insensitive to the viscosity and lie in the low range of the Raman spectrum. Thus, the backbone modulates the motion of base pairs in accord with the environmental medium.

II. LIBRATIONAL DYNAMICS OF BASE PAIRS

In this paper we are concerned with the phonon modes of DNA. To that end we employ the theoretical model worked out in our earlier paper [25], in which we follow the guidelines cast by H. Kappellmann and W. Biem [26]. For the convenience of

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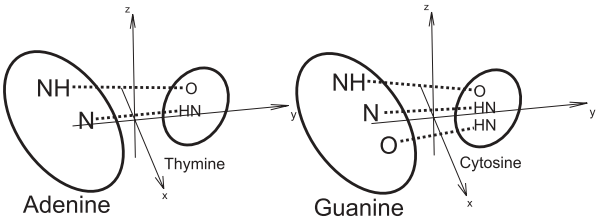

FIG. 1. Base pairs: adenine-thymine and guanine-cytosine. Dashed lines designate hydrogen bonds.

the reader we recall the main points of the duplex structure of DNA. One has to take into account:

(1) The DNA's having two strands
(2) The base pairs being linked by hydrogen bonds
(3) The helical symmetry.

We shall utilize a one-dimensional lattice model of DNA that accommodates these requirements, in agreement with Ref. [24]. The details and the relevant formulas of our model are contained in the Appendix.

According to the model worked out by Crick and Watson [27], one can visualize a molecule of the DNA as a structure that has a backbone formed by two chains composed of alternate sugar and phosphate groups. The chains are wound around each other in a spiral and linked together by four special chemicals, or bases, that are attached to the chains and arranged in pairs. There are four types of the bases: adenine (A) and guanine (G), called purines, and cytosine (C) and thymine (T), called pyramidines. Each type of base on one strand couples with just one type of base on the other strand by means of hydrogen bonds, so that purines are attached to pyramidines, only pairs (A-T) and (G-C) being allowed (see Fig. 1). It is important that the formation of pairs (A-T) and (G-C) involve two and three hydrogen bonds, respectively, so that their coupling energies are different.

The Crick-Watson model of DNA allows a random sequence of bases along one strand. In contrast, the backbone of the double helix formed by sugar and phosphate groups is regular. This circumstance provides a basis for our work.

Elastic properties of DNA depend on more refined details of its structure. It is important that each base is attached to a sugar of the respective strand. Together, the base and the sugar form a complex called a nucleoside. The complex of a sugar, a base, and an adjacent phosphate group is called nucleotide. Thus, we may consider the duplex of the DNA as comprising two chains of nucleotides attached to each other by hydrogen bonds (see Fig. 2). The essential point is that a nucleoside has internal degrees of freedom due to the orientation of the base relative to the sugar as was shown by Donohue and Trueblood [28].

There is no general agreement as to the size of displacements of bases inside the nucleoside so that one has to be content with only a qualitative picture of the motion. It is necessary to allow as well for the relative motions of bases that make up a base pair. According to the existing nomenclature ([29]; see also Ref. [30]), they are described by the angles of tilt, roll, and twist. One should also take into account that the distance between the bases of a base pair can vary so that the hydrogen bonds inside the nucleoside may be broken if amplitudes are large enough. Thus, there are internal degrees of freedom due to the relative motion of bases inside base pairs.

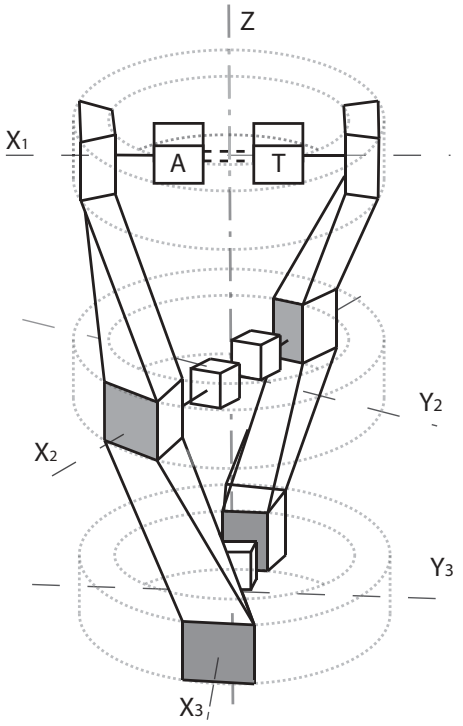

FIG. 2. Base pairs: adenine-thymine and guanine-cytosine. Dashed lines designate hydrogen bonds.

On the other hand the strands can also change their relative position or conformation by twisting around the axis of the double helix. It is important that relative equilibrium positions of alternate base pairs are related through an angle Ω, which is equal to $\pi/5$ for the B form of the DNA.

Summarizing, we may state that (1) there are the internal degrees of freedom owing to the flexible orientation of bases with respect to sugars and relative motion of bases, which take place *inside the double helix*, and (2) *the external ones* are generated by winding the strands around the axis of the double helix.

To put this picture into a more quantitative form we have to employ some simplifications. To that end we turn the rather realistic Fig. 2 into the more abstract Figs. 3 and 4 by considering a molecule of DNA as a one-dimensional lattice, with sites and straight segments corresponding to its sugar

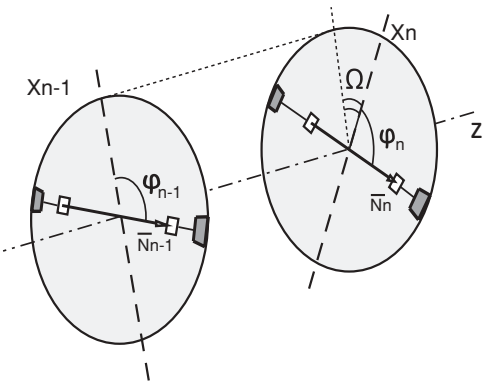

FIG. 3. Angles φ_n indicate deviation from equilibrium. Angle $\Omega = \pi/5$ denotes the pitch of the double helix.

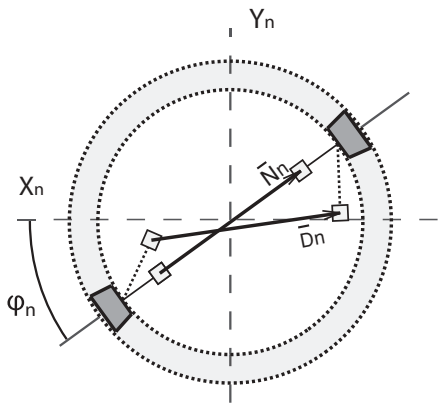

FIG. 4. Angles φ_n indicate twist deviation from equilibrium. Vectors $\vec{N}_n$ and $\vec{D}_n$ designate the unperturbed and perturbed states of the bases in a base pair. Vector $\vec{Y}_n$ equals the difference between $\vec{D}_n$ and $\vec{N}_n$, i.e., $\vec{Y}_n = \vec{D}_n - \vec{N}_n$.

and phosphate groups. We are neglecting the bending degrees of freedom by considering segments of DNA that are short enough. Taking into account its long persistence length of 500 Å we are still in a position to visualize a molecule of DNA as a straight double helix and at the same time to consider samples that are sufficiently large for studying torsional and interstrand vibrational modes due to the dynamics of nucleotides.

We take into account the internal motion of nucleosides by considering vectors $\vec{Y}_n$ at sites of the lattice. Vector $\vec{Y}_n$ equals zero if the base pair related to the pair of nucleosides at the nth site is at equilibrium. Otherwise $\vec{Y}_n$ describes a relative displacement of the basis, as illustrated in Fig. 3 and 4. Thus we cast the physical picture illustrated in Fig. 2 into a picture of one-dimensional lattice framed by vectors $\vec{Y}_n$ at its sites, as shown in Fig. 5. To simplify the problem we shall assume that $\vec{Y}_n$ are all perpendicular to the axis of the helix. We accommodate the possible twist of the strands through a scalar field, which we denote by φ_n, that describes the winding of the double helix.

Thus, we have obtained a model that provide a *qualitative description* for the dynamics of a molecule of DNA. It may be visualized as $(1 + \epsilon)$-dimensional one, in the sense that it is one dimensional, from the formal point of view, and at the same time *accommodates small librational motions of base pairs* into "two transversal dimensions" outside the axis of the duplex. This is due to $\vec{Y}_n$ being directed outside the axis of a molecule, while scalar angles φ_n describe the twist of the sugar-phosphate backbone.

The material constants determining the dynamics of our model (see the Appendix) read as follows:

(1) c is the sound velocity of elastic excitations

(2) I_n is the moment of inertia of the nth pair of nucleosides

FIG. 5. Lattice of vectors $\vec{Y}_n$ of relative positions of bases in a base pair.

(3) k_n is the torsional elastic constant of the nth base pair

(4) τ_n is the torsional elastic constant of the sugar phosphate backbone at the nth site

(5) ϵ_n is the elastic constants for the displacement of bases inside the nth base pair

(6) μ is the viscousity coefficient of surrounding liquid.

It is worth noting that, in real life, a molecule of DNA is not a perfect duplex so that elastic constants that determine the dynamics of the duplex (see the Appendix) are, generally, random sequences. Indeed, owing to different sets of base pairs, the conformational parameters of a molecule of DNA, such as angles determining positions of base pairs, suffer considerable, up to tens of percent, deviations from the regular positions [29]. Further, their being the two types, adenine-thymine and guanine-cytosine, results in these constants having different values for different pairs. Thus, in accord with the general theory of excitations in random media, the phonon modes of DNA may be localized. There are various theoretical approaches to the phenomena (see Ref. [5]).

In this paper we follow the theory worked out years ago by I. M. Lifshitz and his colleagues [31–36] (see also the excellent review [37]). According to the Lifshitz theory the dynamics of elastic excitations in random media, for example, lattices of real crystals or amorphous systems, has two important properties. First, there exists forbidden bands in the spectrum of elastic excitations, such as phonons. Second, there are excitations localized in bounded regions of the medium, their characteristic frequencies being located at the edges of the forbidden zones. The key observation due to Lifshitz was that defects of media could serve as centers for the localization of elastic excitations (see Refs. [32–36]). In his first paper on the subject he considered defects caused by an atom of an isotope that has a mass different from the regular one, and he showed that the defect could generate localized vibrations at its side. Later the approach was further extended to defects of a more general kind and higher dimensions (see Ref. [37]), for example, plains of defects, and also boundaries of crystals.

The study of spectra for disordered, or random, lattices has followed mainly two paths. A number of papers employed analytical methods, which generally could provide only qualitative description of the spectra (see Ref. [37]). It was P. Dean [38] who developed a powerful computer technique for analysis situs of the frequency distribution for disordered lattices. The results by Dean comply with the Lifshitz theory and provide a powerful insight into a specific structure of spectra.

III. HYPERSOUND SPECTROSCOPY

Phonon modes may result in the excessive absorption of hypersound in films and solutions of DNA. In fact, the passage of an acoustic wave may promote the transfer of molecules from the equilibrium state to a state in which phonon modes of molecules of DNA are excited. The time delays in this process and its reversal should lead to a relaxational dissipation of acoustic energy and an absorption of hypersound at certain resonance frequencies corresponding to frequencies of the phonon modes. Equally important, hypersound irradiation of molecules of DNA could make for generating phonon modes. Both the experimental data [8–12,15] and the theoretical

arguments [6,18,19] indicate that the frequencies of phonons of DNA are in GHz range, and therefore one may expect resonance interaction between hypersound waves and phonons of DNA. The availability of hypersound transducers (see Refs. [1,2]) suggests that there are technical means so as to employ high-resolution acoustical spectroscopy for studying phonon modes of DNA in GHz range.

The alleged attenuation requires a careful choice of right samples of DNA for studying phonon modes. By now films of DNA are generally employed to that end. Perhaps this could make for diminishing the attenuation effects previously mentioned. In fact, solutions of DNA are not the best proposition owing to the attenuation effects being large in this case. But, anyway, it should be very interesting to use liquid crystallin phases of DNA, as was done for inelastic x-ray scattering (see Refs. [11,39] and references therein).

The attenuation of phonon modes is closely related to the problem of hydration of DNA. It is alleged that there are two relaxational processes in a hydrated molecule of DNA due to the primary and the second hydration shell with relaxational times $\tau_1 = 4 \times 10^{-11}$ and $\tau_2 = 2 \times 10^{-12}$ s. The residence time for a molecule of water at grooves of a molecule of DNA is estimated as $0.2 \times 10^{-9} - 0.4 \times 10^{-9}$ sec [40]. Phonon dynamics having characteristic times 10^{-9}–10^{-12} s, there is a need for using the generalized hydrodynamics in the GHz range [23] to estimate, for example, the size of dissipation forces acting on a vibrating molecule. Presently, this problem is hard to solve, and we have only the information on the dispersion and attenuation of sound waves in this range, required for the theoretical treatment of the Mandelstam-Brillouin light scattering.

But, curiously enough, one may expect that the strong attenuation of some phonon modes could bring about the generation of others because of the attenuation due to the hydration shell we have mentioned. The latter primarily involve only external regions of the duplex or, according to Ref. [24], does not directly affect the librations of base pairs. Hence, we may suggest that an acoustic wave could drag the molecule, promote its torsional motion, and generate librations of base pairs owing to the internal dynamics of the duplex. We may infer from Eqs. (A5)–(A7) in the Appendix that our model allows for the process. The problem is to write down a reasonable force of molecular-liquid interaction.

For the convenience of numerical simulation it is worthwhile to choose appropriate scales for length, mass, and time that agree with the conformational structure of DNA. We shall take the following quantities as DNA units

(1) $M = 10^{-22}$g as unit of mass, by the order of magnitude close to the mass of a base pair

(2) $L = 3 \times 10^{-8}$ cm as unit of length, close to the distance between neighboring base pairs

(3) $T = 10^{-13}$ s as unit of time corresponding to the upper edge of phonon frequencies of DNA.

We present the results of our calculation by employing the indicated units. Then the material constants of our model are estimated as follows. The sound velocities c of a molecule of DNA are of order 10^5cm/s, according to various experimental and theoretical estimates [10,11]. In the units indicated we have therefore $c \sim 0.3$. The moment of inertia I_n of a base pair is equal by orders of magnitude $m_n R^2$, where R is the radius of a molecule of DNA, that is, ~ 10 Å. Therefore, we have $I_n \sim 10$ in the DNA units we have introduced. To obtain numerical estimates for the constants k_n, τ_n, ϵ_n we shall require that the values of sound velocities c and frequencies ω of librations inside base pairs be of the orders of magnitude $c = 0.3$ and 1, respectively. Since $k_n/m_n \sim c^2$ and $\tau_n/I_n \sim c^2$, we have $k_n \sim 0.1$ and $\tau \sim 1$, respectively. For the libration frequency we have $\epsilon_n/m_n \sim 1$, if we assume that it be of order 1 GHz. For the viscosity coefficient of water we have, accordingly, $\mu \approx 0.3$. Summarizing, we have the following characteristic quantities expressed in the DNA units (see the notations):

$$c \sim 0.3, \quad I_n \sim 10, \quad k_n \sim 0.1, \quad \tau_n \sim 1,$$
$$\epsilon_n \sim 1, \quad \mu \sim 0.3.$$

These estimates provide the basis for our numerical simulation.

As was mentioned, hydrodynamical theory at present does not provide reliable theoretical instruments for studying the interaction between a molecule and solvent, and one should turn to a rule of thumb to find a solution. The interaction could be small enough, and therefore proportional to the velocity of a molecule. Confining us to the picture given by Ref. [24], we may suggest that the viscous force imposed on the nth site of the molecule should have the form

$$d_n = -\gamma \dot{\varphi}_n, \tag{1}$$

where γ is a dissipative constant. The size of γ is difficult to assess. Estimates based on the conventional picture of liquid motion, that is, the Navier-Stokes one, are apparently far from reality because of small characteristic ("nano") times, and the specific structure of water close to the molecule (see Ref. [15]), so that there is a problem as to whether it is reasonable to employ the usual hydrodynamic viscosity coefficient in this situation. If we assume that it is possible, the dimension analysis gives for the moment of viscousity at site n (see the Appendix)

$$\frac{\gamma}{a^2} \propto \mu R^3$$

or

$$\gamma \propto \mu a^2 R^3.$$

In the introduced DNA units, $\mu \approx 0.3, a = 1, R \approx 3$, so that we have $\gamma \approx 10$. Here a is the distance between adjacent base pairs.

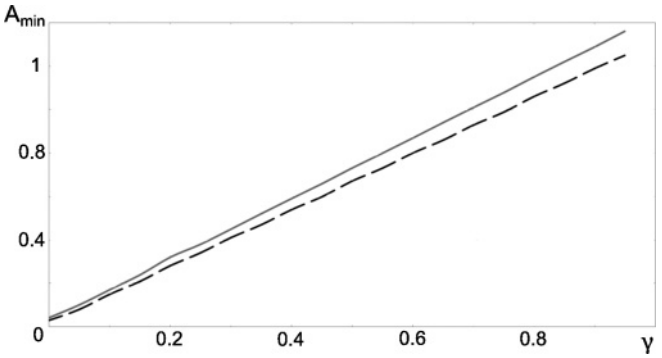

FIG. 6. Minimal amplitude $A_{\min}$ against dissipation γ. Resonance frequencies 1.3 (dashed) and 1.4 (solid). The analytical fit is linear approximations $1.296\gamma + 0{,}019$ and $1.395\gamma + 0{,}033$, respectively.

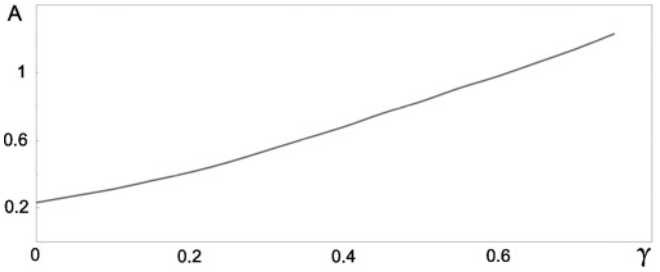

FIG. 7. Threshold $A_{\min}$ against the dissipation γ. The excitation frequency equals 1.55 and deviates from the resonance one. Number of sites $N = 300$, mass $m = 1$; elastic constants $\tau = 1$ or 1.5 (equal probabilities); $\epsilon = 1$; $k = 0.1$ or 0.15 (equal probabilities) $I = 10$. The value of $A_{\min}$ at $\gamma = 0$ is not equal to zero.

In choosing the drag due to the action of a sound wave on the molecule we have to take into account that the wavelength of a hypersound wave in GHz region is by several orders of magnitude larger than the molecular segment. The latter being of a few hundred Å, the force depends only on time. We assume also that the torsion of the molecule due to the force is small and may take it in the form

$$f = A\varphi_n \cos(\omega t). \tag{2}$$

The main point is to determine the nature of the force and to assess its size. If we confine ourselves to the *macroscopical* Navier-Stokes hydrodynamics, two different pictures emerge.

First, there could be an effect similar to that of the Rayleigh disk [41]. To see the point let us neglect the dissipation and attribute the force to the streamlines of flow around a molecule. There is the turning moment Q experienced by an oblate ellipsoid, or disk, in a vibrating fluid [42]:

$$Q = \frac{4}{3}\rho R^3 v^2 \sin 2\varphi.$$

The dimension analysis shows that there is the general formula for the turning moment

$$Q \propto \rho L^3 v^2 \varphi,$$

where L is the characteristic length of a body, v the velocity, and ρ the density of the fluid. In the case we are considering, we may suggest that the rotation angle is small so as to employ φ instead of its sinus. Since the coefficient at $\ddot{\varphi}$ is the product $a^2 I_n$, we obtain the expression

$$A \propto \rho R^3 a^2 v^2.$$

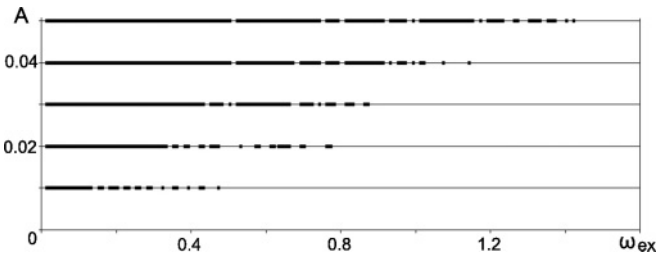

FIG. 8. Ranges of the resonance frequencies corresponding to the excitation amplitudes 0.01, 0.02, 0.03, 0.04, 0.05 (thick horizontal segments); $\epsilon = 1$, $k = 0.1$ or 0.15 (equal probabilities); $I = 10$, $\gamma = 0$.

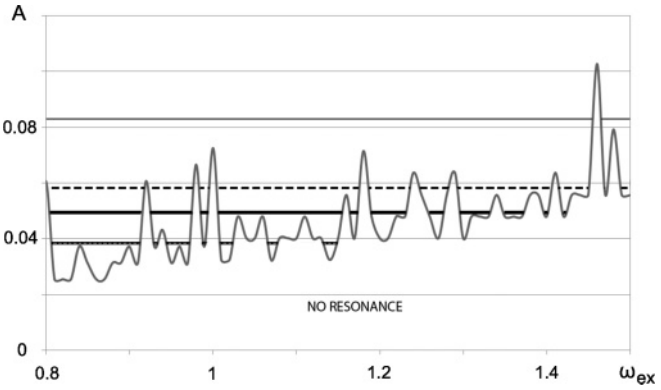

FIG. 9. Threshold curve $A_{\min}(\omega)$ against excitation frequency (thin line). Thick lines indicate segments of the frequencies ω corresponding to constant values $A \geqslant A_{\min}(\omega)$ for which there is resonance.

Now we are in a position to assess the value of A as regards the energy pumped in a sample. Let us recall that the density J_E of the energy current is given by the equation $J_E = \rho c v^2$, c being the velocity of sound in liquid. We are interested only in rough estimates. Therefore we assume $c \sim 10^5 \mathrm{cm/s}, \rho \sim 1g$. For the energy current $J_E = 1\mathrm{W/cm}^2$ we obtain $v = 10$ cm/s, or $v \sim 0.3, 10^{-4}$ in DNA units. In our equation R is the radius of the molecule, that is, $\sim 10^{-7}$cm, or $R \sim 3$ in DNA units. Consequently, we have $A \sim 10^{-7}$. Compared with the values of I_n, γ, it is too small to produce any appreciable effect. In fact, even for $I_E \sim 1\mathrm{kW/cm}^2$ we obtain only $A \sim 10^{-4}$, which is also too small. Therefore, the effect of the Rayleigh disk could not have any bearing on phonon modes of DNA. But it is worth noting that our estimate is due to the use of the laws of

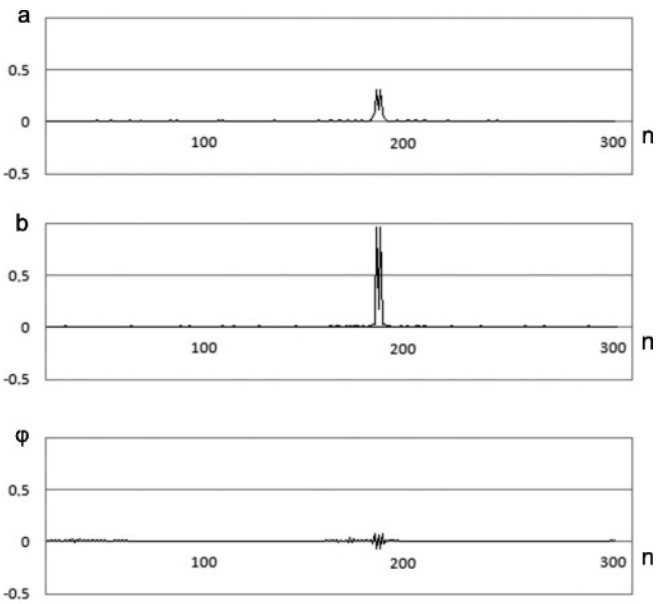

FIG. 10. Localized excitation on a random lattice. Dissipation $\gamma = 0$. The DNA units are employed. $N = 300$, $m = 1$; $\tau = 1$ or 1.5 (equal probabilities); $\epsilon = 1$; $k = 0.1$ or 0.15 (equal probabilities); $I = 10$; amplitude $A = 0.08$; excitation frequency $\omega = 1.5$. Time elapsed is 300 periods of the acoustic wave. The graphs correspond to the a, b, φ modes (view from the top).

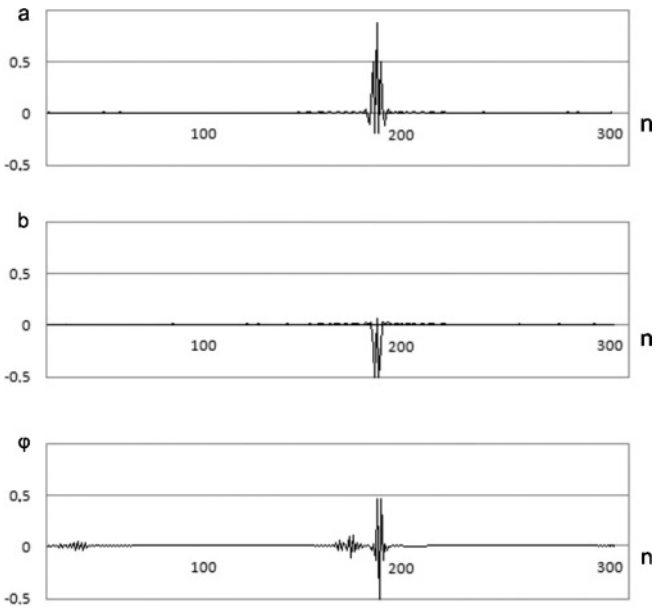

FIG. 11. Localized excitation on a random lattice. Dissipation $\gamma = 0.01$. The DNA units are employed. $N = 300, m = 1; \tau =$ 1 or 1.5 (equal probabilities); $\epsilon = 1; k = 0.1$ or 0.15 (equal probabilities); $I = 10$; amplitude $A = 0.1$; excitation frequency $\omega = 1.489$. Time elapsed is 1000 periods of the acoustic wave. The graphs correspond to the a,b,φ modes (view from the top).

macroscopical hydrodynamics, whereas there are no definite conclusions as to their validity on *microscopical scale*.

The second option is provided by the viscous interaction of the solvent with the molecule. We are looking for an analog

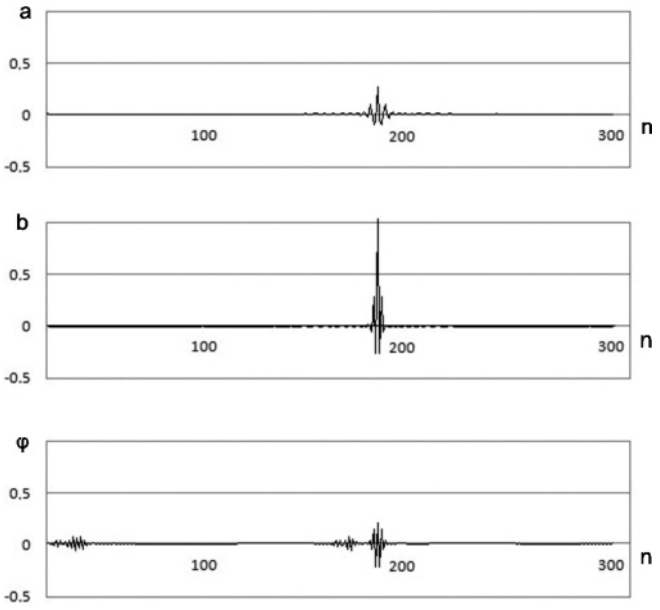

FIG. 12. Localized excitation on a random lattice. Dissipation $\gamma = 0.02$. The DNA units are employed. $N = 300$, $m = 1; \tau =$ 1 or 1.5 (equal probabilities); $\epsilon = 1; k = 0.1$ or 0.15 (equal probabilities); $I = 10$; amplitude $A = 0.1$; excitation frequency $\omega = 1.489$. Time elapsed is 1300 periods of the acoustic wave. The graphs correspond to the a,b,φ modes (view from the top).

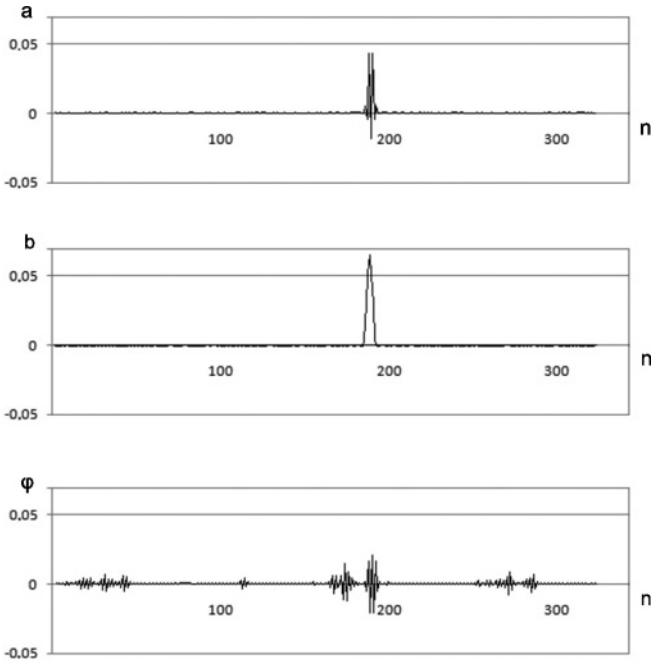

FIG. 13. Localized excitation on a random lattice. Dissipation $\gamma = 0.8$. The DNA units are employed. $N = 300, m = 1; \tau =$ 1 or 1.5 (equal probabilities); $\epsilon = 1; k = 0.1$ or 0.15 (equal probabilities); $I = 10$; amplitude $A = 1.3$; excitation frequency $\omega = 1.489$. Time elapsed if 760 periods of the acoustic wave. The graphs correspond to the a,b,φ modes (view from the top).

of the familiar Stokes force, $6\pi\mu R^2 v$. Using the dimensional analysis we obtain the expression

$$A \propto \mu R^2 a^2 v. \tag{3}$$

For $J_E = 1\text{W/cm}^2$ it provides the estimate $A \sim 10^{-4}$, or $A \sim 3 \times 10^{-3}$ for $J_E = 1\text{kW/cm}^2$.

Our estimates based on *conventional* hydrodynamics apparently preclude any opportunity for the observation of phonon modes of DNA by means of hypersound pumping. Similar arguments [20] are generally put forward in regard to phonon modes studied by the submillimeter absorption spectroscopy. But nonetheless *the sub-THz-phonon modes are observed* (see Ref. [16] and the references therein). We feel that this discrepancy between theory and experiment is likely to be due to the use of conventional hydrodynamics in the region where it does not work properly. In particular, it results in the values of the coefficients A and γ in Eqs. (A5)–(A7) in the Appendix, which precludes the existence of phonon modes. Therefore, in what follows we consider the consequences of A and γ being outside the range prescribed by the Navier-Stokes hydrodynamics and look for appropriate values that permit the existence of phonon modes.

IV. NUMERICAL SIMULATION OF PHONON MODES

The equations of motion are obtained by inserting the forces (1) and (2) in Eq. (A5). The equations are nonlinear and require numerical simulation for their studying. For the numerical simulation of our equations we have used the algorithms of Verlet, LeapFrog, and the explicit and implicit

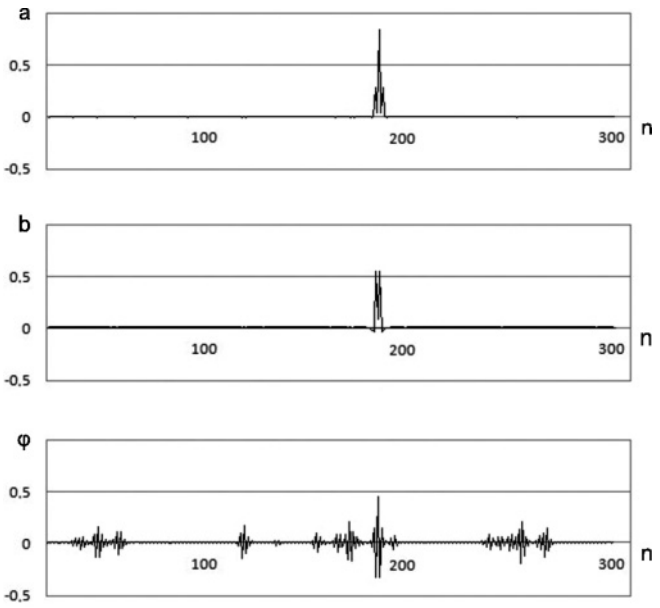

FIG. 14. Localized excitation on a random lattice. Dissipation $\gamma = 1$. The DNA units are employed. $N = 300, m = 1; \tau = 1$ or 1.5 (equal probabilities); $\epsilon = 1; k = 0.1$ or 0.15 (equal probabilities); $I = 10$; amplitude $A = 1.7$; excitation frequency $\omega = 1.489$. Time elapsed is 400 periods of the acoustic wave. The graphs correspond to the a, b, φ modes (view from the top).

Adams algorithms. We have systematically made comparisons between results provided by different algorithms so as to escape possible mistakes and numerical artifacts. We have taken the integration step 0.01, or 0.001 for verification, the unit of time being the period of external force generated by sound pumping.

We are going to simulate the generation of phonon modes by pumping hypersound. According to the equations of motion it is allowed owing to the term given by Eq. (2). Thus, we shall generate first a φ mode, and the latter shall promote the a_n and the b_n ones, according to Eqs. (A6) and (A7). As far as these modes are concerned, we have a parametric excitation through the interaction terms in Eqs. (A6) and (A7) in the Appendix, and, as we will show, there may emerge a parametric resonance at a certain frequency ω_{ex} of the excitation pulse. Here it should be noted that the usual theory of parametric resonance for the harmonic oscillator [41] cannot be directly employed because the Eqs. (A6) and (A7) of motion for the interacting φ_n, a_n, b_n chains are nonlinear, and we need to find the resonance frequencies by "trial and error," Lifshitz's theory providing at this point general recommendations.

It is important that there exists the threshold $A_{\min}$ that depends on the dissipative constant γ, the resonance taking place only for amplitudes of acoustic field that are large enough, $A \geqslant A_{\min}$, as illustrated in Figs. 6 and 7.

If the frequency of the acoustic wave is very close to the resonance one, $A_{\min}$ depends linearly on the dissipative constant γ (see Fig. 6), and for small γ, $A_{\min}$ is also small, so that there is no threshold value at $\gamma = 0$ and $A_{\min} = 0$. If the excitation frequency appreciably deviates from the resonance one, there is a threshold for $A_{\min}$ at $\gamma = 0$ (see Fig. 7).

The numerical simulation provides a description of the resonance frequencies against the excitation amplitude in accord with the general picture of parametric resonance (see Fig. 8). We have also considered the dependence of $A_{\min}$ on the excitation force (see Fig. 9), which has the band structure characteristic of parametries.

Thus, the numerical simulation of Eqs. (A5)–(A7) is in agreement with the general theory of parametric resonance.

Localization of a phonon mode excited by the resonant acoustic wave is illustrated in Fig. 10 for zero dissipative factor γ, and excitation amplitude $A = 0.08$; Fig. 11 for $\gamma = 0.01$, $A = 0.1$; Fig. 12 for $\gamma = 0.02$, $A = 0.1$; Fig. 13 for $\gamma = 0.8$, $A = 1.3$; and Fig. 14 for $\gamma = 1$, $A = 1.7$. As one can see, the same regions of a and b chains are excited due to the same $\omega = 1.489$ frequency.

Our results thus in agreement with Van Zandt and Saxena [22], who predicted the phonons in the submillimiter range corresponding to localized excitations spread over several base pairs.

V. CONCLUSION

We have demonstrated that the acoustic spectroscopy can be helpful in exploration of the conformational structure of DNA through the analysis of its phonon modes. The model we have constructed qualitatively agree with the experimental data by providing the existence of the phonon modes and the reasonable orders of magnitude for their frequencies. It implies a phonon localization due to the random structure of the duplex and shows that it would be worthwhile to study the action of hypersound on molecules of DNA. It is important that absorption peaks for hypersound could be expected at frequencies corresponding to the parametric resonance of phonon modes of DNA under hypersound pumping. In this respect the parametric resonance could be a valuable means for studying the vibrational modes of DNA.

There still remains a problem of accommodating the dissipative effects. The standard Navier-Stokes model promotes a size of dissipation that excludes the existence of phonon modes, but they are observed in real life [16]. The circumstance challenges theoreticians to work out an adequate framework for the hydrodynamics at the molecular scale. The part played by the dissipation, perhaps different from the Navier-Stokes one, could be crucial, and torsional modes of the backbone could make for understanding its nature on the nanoscale. Hypersound acoustic spectroscopy could be instrumental in this respect. To that end liquid samples may turn out to be more interesting solutions of DNA providing a probe into the physics of the liquid state. So far there has been remarkable progress in the experimental study of microhydrodynamics in the low-frequency range [43]. The observation of the indicated parametric resonance could serve a means for understanding the microhydrodynamics in the GHz range.

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APPENDIX

Using the notations introduced earlier we are going to formulate the dynamical equations for our system. The situation is similar to that of the Kirchhoff equations [45] for the equilibrium of a one-dimensional rod where the arc length plays the part of time. We follow the exposition given in Ref. [44]. We shall assume that bases of a base pair lie in the same plane. In fact, there are small deviations outside the planar position (see Ref. [29]) but aiming at a qualitative description of the dynamics of DNA we shall neglect it. For a given base pair we choose a specific, or local, coordinate system with the origin at the intersection of the axis of the double helix and the plane of bases, the latter being perpendicular to the axis; see Ref. [29] for details. The axes of the local coordinates are chosen in such a manner that the ζ axis is parallel to the axis of the double helix, the direction of the ξ axis indicates the equilibrium position of the base pair, and the θ axis is perpendicular to ξ and θ (see Fig. 2 of the present paper and Fig. 1 of Ref. [29]). Thus, we obtain a local system of coordinates, or frame, at every site of the lattice, the adjacent frames being rotated with respect to each other through an angle Φ. At equilibrium Φ is equal to

$$\Omega = 2\pi/10,$$

that is, the pitch of the double helix, whereas the deformation of the structure results in small deviations ϕ_n at sites, $n = 1,2,\ldots$, so that the angle of rotation of adjacent base pairs with respect to each other turns out to be equal to

$$\Phi = \Omega + \phi_{n+1} - \phi_n. \tag{A1}$$

Now we may express the strains and the stresses caused by the indicated deformations following the general prescription of the theory of elasticity (see Ref. [44], ch. 2, §§ [17–19]). The strain u_{nn+1} due to the rotational displacement of the base pairs corresponding to sites n,$n+1$ is given by

$$u_{nn+1} = \frac{1}{a}[R^{-1}(\Phi)\vec{Y}_{n+1} - \vec{Y}_n],$$

where $R(\Phi)$ is the matrix of a rotation round the axis of the double helix through the angle Φ, a being the distance between alternate base pairs. The strain due to the displacement of bases inside a base pair is $\vec{Y}$, the corresponding stress being

$$\epsilon_n \vec{Y}.$$

The strain $v_{n\,n+1}$ caused by the displacements of the sugar-phosphate backbones is determined by the twist angle ϕ_n and given by

$$v_{n\,n+1} = \frac{\tau_n}{a}[\phi_{n+1} - \phi_n].$$

The corresponding stresses read

$$\sigma_{n\,n+1} = \frac{k_n}{a}[R^{-1}(\Phi)\vec{Y}_{n+1} - \vec{Y}_n]$$

$$\text{and } \omega_{n\,n+1} = \frac{\tau_n}{a}(\phi_{n+1} - \phi_n),$$

respectively. Here k_n and τ_n are the torsional elastic constants. Hence, we have the following equation for the potential energy of the framed lattice representing the molecule:

$$U = \sum_n \left\{ \frac{\tau_n}{2a^2}[\phi_{n+1} - \phi_n]^2 + \frac{k_n}{2a^2}[R^{-1}(\Phi)\vec{Y}_{n+1} - \vec{Y}_n]^2 + \frac{\epsilon_n}{2}\vec{Y}^2 \right\}; \tag{A2}$$

here a is the distance between adjacent base pairs. Next, we shall write the kinetic energy. It comprises two terms: (1) the twist one corresponding to the sugar-phosphate chains

$$T_{\text{str}} = \sum_n \frac{I_n}{2}\dot{\phi}_n^2, \tag{A3}$$

where I_n is the moments of inertia of the nth pair of nucleotides, and (2) that due to the motion of base pairs

$$T_{\text{bases}} = \sum_n \frac{M_n}{2}\dot{\vec{Y}}_n^2, \tag{A4}$$

where M_n is the effective mass of the nth base pair. In our summations n is the number of a site corresponding to the nth base pair, and $n = 1,2,\ldots N$, N being the number of pairs in the segment of DNA under consideration.

The total energy of the system is the sum

$$H = T_{\text{str}} + T_{\text{bases}} + U.$$

From Eqs. (A2) and (A3) we derive the equations of motion by using the usual prescription [44].

Our next step is to split the $(1+\epsilon)$–dimensional lattice into three interacting linear chains. To that end we shall cast our variables in a complex form. Let us introduce complex quantities z_n in accord with

$$\vec{Y}_n = \left(Y_n^1; Y_n^2\right) \to z_n = Y_n^1 + iY_n^2.$$

Then we may cast the expression for the energy, $E = T + U$, in the form

$$T = \sum_{n=0}^{N} \frac{I_n\dot{\varphi}_n^2}{2} + \sum_{n=0}^{N} \frac{m_n\dot{z}_n\dot{z}_n^*}{2}$$

for the kinetic energy and

$$U = \sum_{n=0}^{N} \frac{\epsilon_n z_n z_n^*}{2} + \sum_{n=0}^{N-1} \frac{\tau_n}{2a^2}|\varphi_{n+1} - \varphi_n|^2 + \sum_{n=0}^{N-1} \frac{k_n}{2a^2}|z_{n+1} - e^{i(\Omega+\varphi_{n+1}-\varphi_n)}z_n|^2$$

for the potential one. On using the substitution

$$z_n = e^{in\cdot\Omega}\, y_n$$

we may cast these equations in the form

$$T = \sum_{n=0}^{N} \frac{I_n\dot{\varphi}_n^2}{2} + \sum_{n=0}^{N} \frac{m_n\dot{y}_n\dot{y}_n^*}{2}$$

and

$$U = \sum_{n=0}^{N} \frac{\epsilon_n y_n y_n^*}{2} + \sum_{n=0}^{N-1} \frac{\tau_n}{2a^2}|\varphi_{n+1} - \varphi_n|^2 + \sum_{n=0}^{N-1} \frac{k_n}{2a^2}|y_{n+1}e^{i(\varphi_{n+1}-\varphi_n)}y_n|^2.$$

We consider only small deviations from the equilibrium and assume that differences $(\varphi_{n+1} - \varphi_n)$ are small. Therefore, we have

$$e^{i(\varphi_{n+1}-\varphi_n)} = 1 + i(\varphi_{n+1} - \varphi_n) + O[(\varphi_{n+1} - \varphi_n)^2].$$

In what follows we shall neglect terms of the second order in φ.

Within this approximation the Lagrangian equations of motion have the form

$$a^2 I_n \ddot{\varphi}_n = \tau_{n-1}\varphi_{n-1} - (\tau_{n-1} + \tau_n)\varphi_n + \tau_n\varphi_{n+1} - k_n(a_n b_{n+1} - a_{n+1}b_n) + k_{n-1}(a_{n-1}b_n - a_n b_{n-1}), \tag{A5}$$

$$a^2 m_n \ddot{a}_n = -a^2\epsilon_n a_n + k_{n-1}a_{n-1} - (k_{n-1} + k_n)a_n + k_n a_{n+1} + k_n(\varphi_{n+1} - \varphi_n)b_{n+1} - k_{n-1}(\varphi_n - \varphi_{n-1})b_{n-1}, \tag{A6}$$

$$a^2 m_n \ddot{b}_n = -a^2\epsilon_n b_n + k_{n-1}b_{n-1} - (k_{n-1} + k_n)b_n + k_n b_{n+1} - k_n(\varphi_{n+1} - \varphi_n)a_{n+1} + k_{n-1}(\varphi_n - \varphi_{n-1})a_{n-1}, \tag{A7}$$

a_n and b_n being the real and imaginary parts of y_n. Thus, we split the equations of motion into three parts and cast the system in a form of three interacting one-dimensional lattices of variables a_n, b_n, φ_n. This mathematical device is crucial for the subsequent analysis of the dynamics.

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