Resonant Microwave Absorption of Selected DNA Molecules

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The resonant absorption of microwave energy by aqueous solutions containing DNA of known length is experimentally demonstrated. The resonances observed have relaxation times of hundreds of picoseconds. Absorption by linear and supercoiled circular DNA molecules is discussed in terms of a mechanism involving microwave excitation of acoustic modes of the double helix.

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We are investigating the absorption of microwave energy by aqueous solutions of DNA at room temperature. The absorption properties of solutions of biological macromolecules exposed to nonionizing radiation have generally been described by Debye relaxation theory.[1] This theory does not, however, explain the observed microwave absorption of DNA solutions. Recent biological experiments[2,3] have suggested frequency-specific effects from exposure to low-level (producing nonsignificant thermal induction) microwave or radio-frequency radiation. These biological effects also cannot be described by Debye relaxation processes and have frequently been viewed with disbelief by the physical community.

We have previously reported broadband microwave absorption of aqueous DNA solutions using both an optical heterodyne technique[4–6] and more standard dielectrometric methods.[7] These results suggested a direct interaction of the microwave field with the DNA molecule. Van Zandt and co-workers[8–10] have carried out extensive calculations using normal-mode analysis, lattice dynamics, and hydrodynamic theory. Their model describes vibrational modes[8] and defect resonances[10] for DNA polymers which could exhibit direct coupling to the microwave field. It has generally been thought, however, that the vibrational modes of isolated DNA molecules in aqueous solutions at room temperature would be overdamped by the solvent.[9] Hakim, Lindsay, and Powell[11] have measured the velocity of sound of an acoustic mode on oriented DNA fibers of varying humidity using Brillouin spectroscopy. Reduced solvent damping is attributed to the structure and orientation of the DNA molecules and water which constitute the fiber. Lindsay and Powell[12] have also recently observed a microwave resonance which is consistent with the description of Putnam *et al.*[10] of a defect resonance.

Our previous dielectrometric measurements[13] have also demonstrated a dramatic increase in the absorption of microwave energy for solutions of *E. coli* DNA that were being randomly nicked by low concentrations of the endonuclease DNase I (Fig. 1). We proposed that the nicking produced a

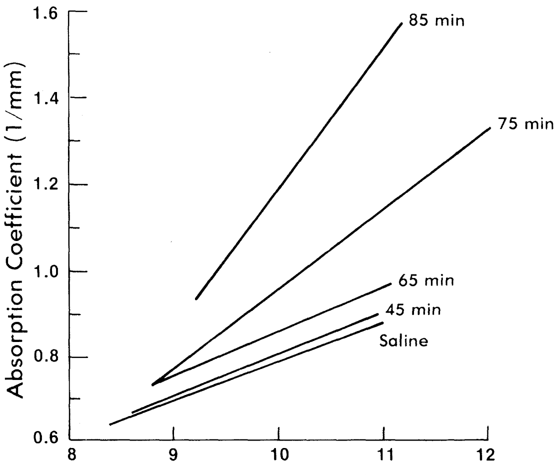

FIG. 1. Variation of the absorption coefficient (α) of *E. coli* DNA exposed to DNase I for 85 min. 1.75 g/l *E. coli* DNA (> 50 000 base pairs prior to DNase activity) dissolved in 9 g/l NaCl, 0.1 g/l DNase I, and 10 g/l $MgCl_2$ solution.

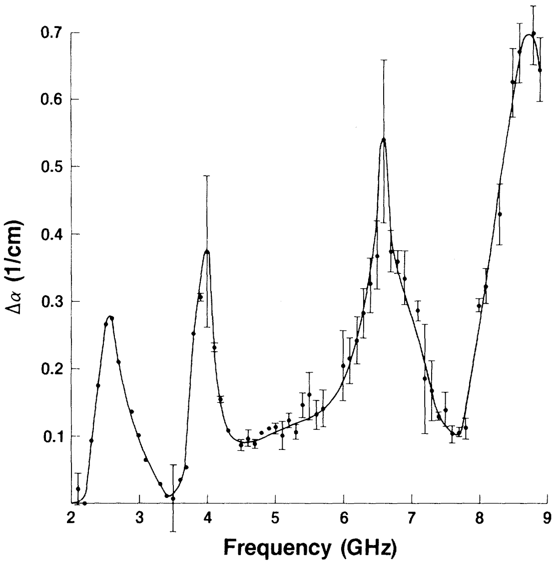

FIG. 2. Relative absorption coefficient of the fundamental and first three harmonics of 0.53 g/l 2734-bp supercoiled circular DNA. Relative measurements have background absorption subtracted. Error bars represent total spread in the data. DNA is dissolved in storage buffer [10 mM tris(hydroxymethyl)aminomethane, 10 mM NaCl, 1 mM ethylenediamine tetra-acetic acid, pH 7.5].

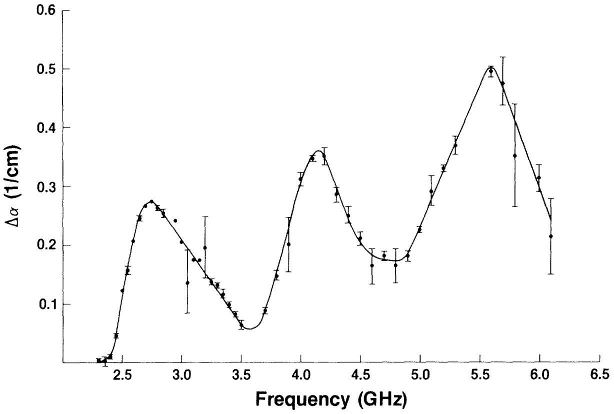

FIG. 3. Relative absorption coefficient of the first three harmonics of 0.50 g/l 2734-bp linear DNA dissolved in storage buffer. The corresponding fundamental expected near 900 MHz is not observed.

dynamic length distribution whose mean length decreased with time. Enhanced absorption occurred as the sample length distribution moved through the region giving significant absorption in the experimental frequency range (0.4 to 12 GHz).

To further investigate these phenomena we now have produced cloned DNA samples of uniform length and known sequence prepared by standard plasmid extraction procedures and restriction endonuclease digestion. To conserve sample material we have altered the exposure system limiting the higher end of the experimental frequency range. A solution of supercoiled circular DNA molecules of 2734 base pairs (bp) has resonant frequencies near 2.55, 4.00, 6.60, and 8.75 GHz (Fig. 2). A solution of 2734-bp linearized DNA has resonant frequencies near 2.75, 4.15, and 5.60 GHz (Fig. 3). A solution of equimolar 948-bp and 1786-bp linear DNA has resonant frequencies near 2.65 GHz (Fig. 4) and 4.10 GHz (Fig. 5). We believe that our success in demonstrating these specific resonances is due to the use of DNA of uniform length. It is important to recognize the necessity of stringent sample preparation and characterization in this research.

The observed relaxation times are surprisingly long (200 to 300 psec); it is not understood why there is apparently so little damping from the solvent. Van Zandt's hydrodynamic theory[9] models the solvent as a continuum fluid with "stick" boundary conditions at the surface of the molecule. Considerations of solvent structure and alternative boundary conditions[14] should lead to a theoretical model describing less severe solvent damping.

It is interesting to speculate about the possibility of this or higher excited states evolving into other energetic forms. Our earlier studies using the optical heterodyne technique[4-6] measured the thermal expansion of sample solutions that resulted from relaxation of excited polymer modes. We intend to investigate further whether these excited states can only relax into random thermal energy or if other energetic pathways are available.

The frequency-length dependence suggested by these experimental results (Figs. 2–5) can be

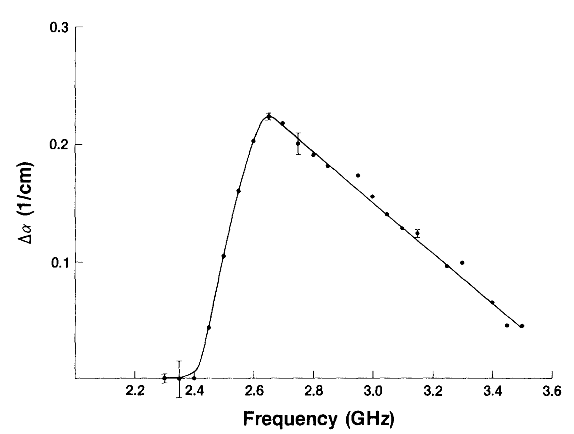

FIG. 4. Relative absorption coefficient assigned to the fundamental of 0.12 g/l 948-bp linear DNA dissolved in storage buffer.

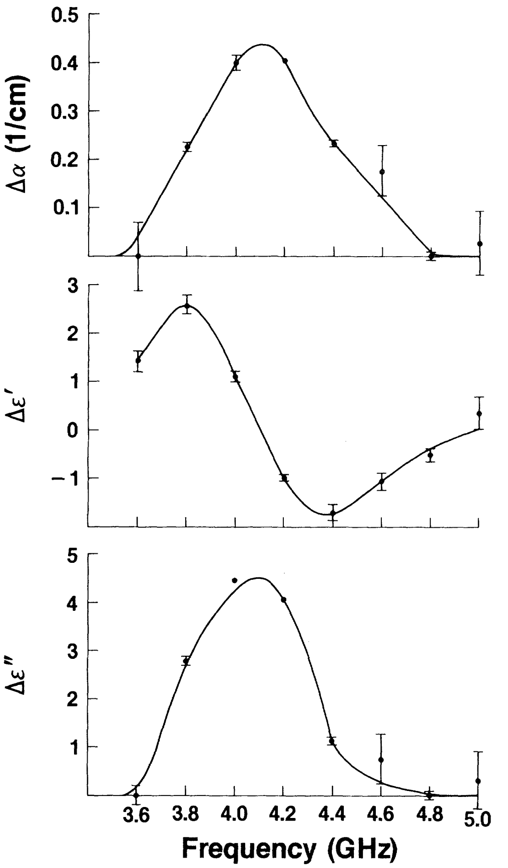

FIG. 5. Relative absorption coefficient ($\Delta\alpha$), and real ($\Delta\epsilon'$) and imaginary ($\Delta\epsilon''$) parts of the dielectric constant assigned to the first harmonic of 0.22 g/l 1786-pb linear DNA dissolved in storage buffer. These curves exhibit a classical absorption resonance. The corresponding fundamental expected near 1.4 GHz is not observed.

described by simple equations. The frequency ν of the observed resonances for DNA molecules and the length l of the molecules (determined by the number of base pairs) are related by

$$\nu_{\text{circular}} = (n+1)v\,l, \quad \nu_{\text{linear}} = 2(n+\tfrac{1}{2})v\,l,$$

where v is the acoustic velocity for circular or linear DNA molecules; n indicates the nth haromonic, the fundamental being described by n equal to zero. These relationships result from considerations of field-induced acoustic modes. The excitation must overlap (circular DNA) or reflect (linear DNA) in phase. In addition, net power flow from the field to the molecule must be possible; there is no net power flow for full wavelength (and multiples thereof) excitations on linear DNA. The measured acoustic velocities for linear DNA (1.67 km/s for 948 bp) compare favorably with that of Hakim, Lindsay, and Powell,[11] (1.69 km/s for the fully hydrated fiber). The measured acoustic velocity for supercoiled circular DNA molecules (2.32 km/s) is significantly faster. It is unclear to us why the velocities for the supercoiled circular and linear DNA molecules would be different. The model used to calculate the acoustic velocity,[8] however, depends on extensive electrostatic interactions between charged groups symmetrically distributed along the DNA molecule. Could these interactions be sufficiently sensitive to molecular configuration to account for this difference in acoustic velocity? Theoretical models have not yet been developed for circular DNA molecules.

The fundamental resonances described by these simple equations are not observed for 1786-bp and 2734-bp linear DNA but are observed for 948-bp linear and 2734-bp circular DNA. A plausible explanation for the missing fundamentals is suggested by the observed relaxation times. The observed resonant microwave absorption is a length-dependent event. In some sense the excitation must know the extent of the molecule to develop into a coherent mode; i.e., the excitation must constructively interfere to develop into an acoustic mode. The observed relaxation times (300 psec) and acoustic velocities suggest that an excitation can travel on the order of 1500 bp (linear DNA) and 2100 bp (supercoiled circular DNA) before dispersing. Linear DNA of 1786 bp and 2734 bp are too long for constructive interference to develop at the fundamental energy. The presence of the nth harmonics results from $(2n+1)$ times the fundamental energy being coupled into the DNA molecule. For circular DNA the excitation must travel half the molecular extent before constructive interference can develop. DNA solutions should fail to exhibit fundamentals and successive harmonics as the molecular length is increased. This model is additionally attractive because it provides a maximum length and minimum frequency of excitation as implied by our earlier results (Fig. 1).

Our experimental frequency range has a lower limit of 400 MHz. We have observed the falling edge of an absorption near 400 MHz for solutions of linear DNA. This absorption is independent of the length of linear DNA molecules and is not present for solutions of supercoiled circular DNA. This could correspond to the defect resonance described by Putnam *et al.*[10] and observed by Lindsay and Powell[12] which is associated with the premelting of the ends of DNA molecules.

Although there is no demonstrated relationship between these phenomena and reported genetic effects, these observations indicate that a mechanism exists for coherent frequency-specific deposition of microwave energy in DNA. This mechanism has surprising physical characteristics and intriguing implications for future polymer and biochemical research. It is unexpected that DNA solutions exhibit microwave resonances and fascinating that such sharp resonances occur in dense solutions at room temperature. It will be interesting to see if these phenomena can be used to alter biochemical processes and if similar phenomena exist in other symmetric macromolecules.

This work is from a dissertation to be submitted to the Graduate School, University of Maryland by G. S. Edwards in partial fulfillment of the requirements for the Ph.D. degree in Chemical Physics.

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