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FREE RADICALS IN PYRIMIDINES: ESR OF IRRADIATED SINGLE CRYSTALS OF OROTIC ACID[1]

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ABSTRACT

Hydrogen abstraction from $N_{(1)}$ occurs when single crystals of orotic acid are exposed to ionizing radiation at room temperature. The stable free radical has unpaired spin density on both $N_{(1)}$ and $C_{(5)}$ of the pyrimidine ring. Hyperfine coupling to the nitrogen nucleus shows axial symmetry, with $A_{\parallel}^{N} = 15.0$ gauss and $A_{\perp}^{N} = 0$. The hydrogen bonded to $C_{(5)}$ has principal hyperfine values, in gauss, of $A_x^H = 7.3$, $A_y^H = 22.2$, and $A_z^H = 15.1$. From the hyperfine couplings, the spin densities calculated for $N_{(1)}$ and $C_{(5)}$ respectively are 0.29 and 0.64. Principal values of the g-tensor are $g_u = 2.0059$, $g_v = 2.0052$, and $g_w = 2.0023$. The ESR analysis can be used to predict the molecular orientation in orotic acid crystals.

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KEY WORDS: Pyrimidines, orotic acid, electron spin resonance, free radicals.

INTRODUCTION

When organic crystalline solids are exposed to ionizing radiation at room temperature, the stable free radicals formed are usually neutral species resulting from the rupture of covalent bonds. In the majority of studies in which positive radical identification has been possible, the mechanism of radical formation involves either hydrogen abstraction from, or hydrogen addition to, the parent molecule. These reactions are usually quite selective, occurring at only one of several possible sites on the molecule. The basis for this selectivity is not fully understood, and no adequate method exists for predicting which stable radical will be formed by irradiating a particular compound.

It appears especially difficult to predict the site of free-radical formation in pyrimidines, in which both hydrogen abstraction and hydrogen addition reactions have been found to occur. It is not too surprising that compounds containing the thymine ring are subject to hydrogen addition reactions at the 5, 6 double bond (1-3), whereas dihydrothymine undergoes hydrogen abstraction at these same positions (4, 5). However, in cytosine, stable radicals formed both by hydrogen abstraction from $N_{(1)}$ and by hydrogen addition to $C_{(5)}$ of the pyrimidine ring have been identified (6). An interesting hydrogen addition reaction occurs in 5-nitro-6-methyluracil, where the hydrogen adds to the keto oxygen at the 4-position (7). The resulting radical has unpaired spin density at both $C_{(4)}$ and $C_{(6)}$, due to resonance with the π electrons in the 5, 6 double bond. Hydrogen addition to the oxygen atom bound to $C_{(4)}$ is also thought to occur in halogenated derivatives of uracil (8). Thus a variety of mechanisms for radical formation are seen, and it is

not clear what factors determine which mechanism prevails for a particular pyrimidine derivative.

Our work, aimed toward understanding the mechanisms of free-radical formation in irradiated pyrimidines, has now been extended to include a carboxylated derivative of uracil. We report here a detailed ESR study of free radicals in irradiated orotic acid (uracil-6-carboxylic acid). The primary free radical stable at room temperature has been identified and the parameters characterizing its ESR absorption have been determined. It was found that a simple first-order treatment of hyperfine interactions was inadequate to explain the observations, and an analysis following the theory of McConnell et al. (9) was used to verify the radical assignment and hyperfine coupling constants at all crystal orientations in the magnetic field.

EXPERIMENTAL PROCEDURE

Preparation of Crystals

Orotic acid was obtained commercially and used without further purification. This compound is only sparingly soluble in water, and our attempts to grow good single crystals from dilute aqueous solutions were unsuccessful. After trying several solvents, we found that a mixture of ethanol and NH_4OH was suitable for crystal growth. A solution of 50% ethanol and 50% 0.8 M aqueous NH_4OH (by volume) was saturated with orotic acid at 60° C. Slow cooling over a period of several days yielded large crystals with well-developed faces. These crystals showed no decomposition in air after harvesting.

In order to ascertain that the crystals obtained by this procedure

were orotic acid, a crystal was dissolved in water and analyzed by ultraviolet (UV) spectroscopy and thin-layer chromotography. The UV absorption spectra of the dissolved crystal and of a solution of the commercial preparation were identical, and the absorbancy ratios at 280/260 and 250/260 agreed with those reported in the literature for orotic acid (10). Ascending chromatography on Eastman thin layer 6065 (cellulose with fluorescence indicator) was carried out, using four solvents. With isopropanol: 30% HCl : H_2O (65: 10 : 25), the Rf values for the dissolved crystal and the commercial preparation were 0.68 and 0.66 respectively; with isopropanol : H_2O (50 : 50), the respective values were 0.18 and 0.18; with methanol: 30% HCl : H_2O (70 : 20 : 10), the values were 0.52 and 0.50; and with n-butanol : methanol : H_2O : 0.8 M NH_4OH (59 : 20: 20 : 1), the values were 0.22 and 0.25. All proportions given here are by volume.

To our knowledge, no report of the crystal structure of orotic acid appears in the literature. An orthogonal a'b'c' axis system, shown in relation to the crystal morphology in Fig. 1, was used to facilitate the ESR analysis. These axes were found to be axes of magnetic symmetry. Specifically, in the a'b' plane, the spectrum at any angle θ relative to a' (or b') is identical to that at an angle $-\theta$ with that axis. There are two magnetically distinct molecules in the a'b' plane. In the b'c' and c'a' planes, all molecules are magnetically equivalent. The orientation of the molecules relative to the a'b'c' axis system will be inferred later from the ESR analysis.

Irradiation

The crystals were irradiated at room temperature with 7-MeV

electrons from a linear accelerator. A total exposure of 10 megaroentgens was delivered at a rate of approximately 1 MR per minute. The radicals produced were quite stable, showing no appreciable decay or conversion at room temperature for several weeks.

ESR Spectroscopy

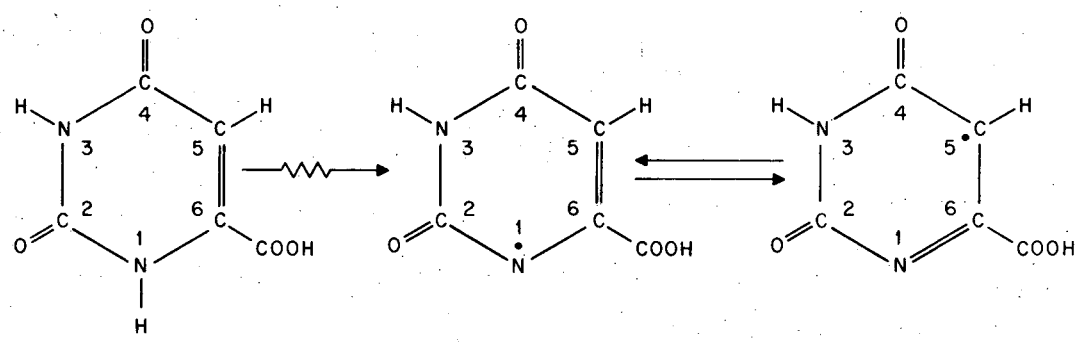
It was necessary to make ESR measurements at two microwave frequencies in order to analyze completely the hyperfine interactions observed. Data were taken at 9.4 GHz, using a Varian 4502 spectrometer, and at 16.2 GHz, using a microwave bridge constructed in our laboratory. In both cases, a cylindrical cavity was used to allow rotation of the magnet about the crystal. Hyperfine couplings were measured by comparison with a standard marker of Mn^{++} in MgO. DPPH was used as a standard for determining g-values.

All calculations were made using a CDC 6600 computer.

IDENTIFICATION OF THE RADICAL

The spectra observed for many crystal orientations consisted of a hyperfine doublet with each component further split into an equal-intensity triplet. Figure 2 shows this basic pattern at four orientations in the b'c' plane. It is apparent that the unpaired electron interacts with one proton ($I = 1/2$) and one N^{14} nucleus ($I = 1$) to give this hyperfine structure. Both the nitrogen and hydrogen hyperfine couplings showed considerable anisotropy. The nitrogen coupling had its maximum value with the magnetic field along the c' axis and was too small to be resolved everywhere in the a'b' plane. This suggests that the unpaired spin density producing the nitrogen coupling is in a nitrogen $2p\pi$ orbital whose

direction is along c'. The c' axis was found to be the direction of minimum g-value, further substantiating this assignment. The hydrogen coupling was typical of that expected for coupling of unpaired spin density in a carbon 2pπ orbital to an α proton. Furthermore, the intermediate principal value for the hydrogen coupling occurred with the field along c', as would be expected for an α proton attached to the pyrimidine ring. Thus, all the features of the ESR spectra can be accounted for by a radical with the resonance structures



formed by hydrogen abstraction from $N_{(1)}$. No other method of radical formation, aside from drastic molecular rearrangements, can explain the observations.

DETERMINATION OF ESR PARAMETERS

g-Value

With the magnetic field along c', the g-value was very near the free spin value for an unpaired electron. This was the smallest g-value observed at any orientation and must be the minimum principal value

for the g-tensor. The other two principal values should then be found in the a'b' plane. Although there are two magnetically distinct molecules in that plane, as evidenced by the hydrogen hyperfine coupling, the g-value variation for both molecules was nearly identical. Table I lists the principal values for the g-tensor and the direction cosines relating the principal axes to the a'b'c' coordinate system. In Fig. 3, the observed g-values are compared with those calculated from the parameters in Table I. It is likely that the direction of g_u (or g_v) is almost, but not quite, identical for the two magnetically distinct species.

Nitrogen Hyperfine Interaction

Within experimental error, the nitrogen hyperfine tensor is axially symmetric, with the two degenerate principal values being zero. The general expression for the hyperfine coupling at an arbitrary angle in the principal axis system is

$$A = (A_x^2 \cos^2\theta_x + A_y^2 \cos^2\theta_y + A_z^2 \cos^2\theta_z)^{1/2},$$

where A_x, A_y, A_z are the principal values and θ_x, θ_y, θ_z the corresponding direction cosines relating the direction of observation to the principal axes. Here, this expression reduces to

$$A = (A_{\parallel}^2 \cos^2\theta)^{1/2},$$

with θ being the angle between the field direction and the symmetry axis of the nitrogen hyperfine tensor. In Fig. 4, the observed values

for the nitrogen coupling are compared with the calculated values, based on parameters given in Table I.

Hydrogen Hyperfine Interaction

Ordinarily, the interaction of a single proton with the unpaired electron in a free radical splits the original absorption line(s) into a simple doublet. The two lines of the hyperfine doublet correspond to the two possible spin states ($M_I = \pm 1/2$) for the proton, the selection rule being that M_I does not change its value simultaneously with the change in the electron spin state. This situation prevails if the external magnetic field of the spectrometer is either much greater than or much less than the magnetic field produced by the electron at the site of the proton. In the former case, the proton is aligned either parallel or antiparallel to the external magnetic field, and its alignment is not expected to change during the electronic transition. In the latter case, the proton is aligned either parallel or antiparallel to the electronic magnetic field, and the electronic transition merely reverses the direction of this field. The proton spin state again is not expected to change, as it will still be quantized in the field.

A more complicated situation results when the external field is comparable in magnitude to the electronic field at the proton. In this case, the proton is oriented in the resultant of the external and electronic magnetic fields. When the electronic transition occurs, the field at the proton neither remains fixed nor reverses, but instead takes on some intermediate direction relative to the initial resultant field. The selection rule restricting a change in the proton spin state no longer applies, since the proton must now become oriented either parallel or antiparallel to

the new resultant field. Thus, four transitions become allowed rather than two. These four transitions comprise two pairs, referred to as the inner and outer doublets, each symmetrically disposed about the center of the original absorption line(s). The relative intensity of the two doublets depends on the angle between the resultant fields at the proton for the two electron spin states.

There are in general three unique orientations of a radical in the external field for which the proton hyperfine pattern is only a doublet even though the external field and the electronic field are comparable in size. These are called the canonical orientations, the directions in the radical being the principal hyperfine axes. For each of these orientations, the electronic field at the proton is parallel (or antiparallel) to the external field, so that the direction of the resultant field either remains the same or reverses during the electronic transition. Thus, the proton remains quantized, and the selection rule restricting a change in its spin state applies.

According to the theory of McConnel et al. (9), the resultant magnetic field at the proton, due to both the external and electronic fields, is in the direction of a unit vector $\vec{u}$ given by an equation of the form

$$A\vec{u} = \frac{g_I \beta_I}{g \beta} H\vec{u}_H - M_S(\vec{i}\, A_x \sin\theta \cos\phi + \vec{j}\, A_y \sin\theta \sin\phi + \vec{k}\, A_z \cos\theta).$$

In this equation, g and g_I are the electronic and nuclear g-factors respectively, β is the Bohr magneton, and β_I is the nuclear magneton; $\vec{u}_H$ is a unit vector in the direction of the external magnetic field, and $\vec{i}$, $\vec{j}$, $\vec{k}$ are unit vectors in the directions of the principal hyperfine axies, the corresponding principal hyperfine values being A_x, A_y, and A_z; θ and ϕ

are the polar and azimuthal angles relating the external field direction to the principal axis system. Thus, this equation may be evaluated for the two electron spin states $M_S = \pm 1/2$ to give two values, $\vec{u}'$ and $\vec{u}''$, for the direction of the resultant field at the proton. Similarly, two corresponding values of A are obtained, A' and A''. These parameters are then used to give both the splittings of the inner and outer doublets and their relative intensities. The inner doublet splitting equals A' - A'' and the outer doublet splitting equals A' + A''. The angle (ξ) between $\vec{u}'$ and $\vec{u}''$, given by $\cos \xi = \vec{u}' \cdot \vec{u}''$, determines the relative intensities of the inner and outer doublets. For the inner doublet, the normalized relative intensity is $\cos^2 (\xi/2)$, and for the outer doublet it is $\sin^2 (\xi/2)$.

The theory discussed above was used to analyze the proton hyperfine coupling for the free radical in irradiated orotic acid. It can be seen that both the inner and outer doublet splittings, as well as their relative intensities, depend on the magnitude of the external field. In Fig. 5, this behavior is seen for a crystal orientation with H along the a' axis, where all molecules in the unit cell are magnetically equivalent. At a microwave frequency of 9.4 GHz ($H \approx 3{,}330$ gauss), the inner and outer doublets are of comparable intensity, whereas at 16.2 GHz ($H \approx 5{,}770$ gauss), the inner doublet is much stronger than the outer doublet. At other crystal orientations, the outer doublet was more intense than the inner doublet. The spectra at both microwave frequencies are expected to be identical only when the crystal is in a canonical orientation, with H along a principal hyperfine axis.

In order to apply the theory of McConnell et al. (9) it is necessary to determine the principal hyperfine values A_x, A_y, A_z, and the directions of the

principal axes relative to the crystal axis system. In this case, the intermediate hyperfine value can be measured directly as the splitting when H is along c', since this direction is parallel to the p-orbital (see earlier discussion). The other two principal values are then found as the maximum and minimum splittings in the a'b' plane. Unfortunately, there are two magnetically distinct molecules in this plane, and a canonical orientation for one is not a canonical orientation for the other. Nevertheless, it was possible to obtain the tensor parameters for the hydrogen hyperfine interaction, and these are given in Table I. The principal hyperfine axes for the two molecules are approximately 40 deg apart.

With these parameters, it should be possible to explain both qualitatively and quantitatively the spectra observed at all crystal orientations. This was found to be the case, and is considered as justification for the radical assignment and the tensor parameters in Table I. The spectra in Fig. 5 agree well with the theoretical lines at both microwave frequencies. Figure 6 shows spectra at another orientation where there are two magnetically distinct molecules. Again, good agreement is seen between the calculated and observed spectra. Quantitatively, the splittings of the inner and outer doublets were predicted well by the theory. A comparison between the observed and calculated splittings for the two doublets, with the magnetic field in the a'b' plane, is shown in Figs. 7 and 8 for the two microwave frequencies. In this plane, the nitrogen hyperfine coupling was zero, so the proton coupling could be analyzed more carefully than in the other two planes. The theoretical lines are shown as dashed lines in regions where the normalized relative intensity

for a particular doublet falls below 0.30.

CALCULATION OF SPIN DENSITIES

The radical in irradiated orotic acid is formed by the same mechanism as one of the radicals in irradiated cytosine (6), and the spin densities at $N_{(1)}$ and $C_{(5)}$ can be compared for these two cases. Following the analysis used by Cook et al. (6) for cytosine, the spin density (ρ_N) in the $2p\pi$ orbital on $N_{(1)}$ may be calculated from the expression

$$A_d = B_0 (3 \cos^2\theta - 1) \rho_N,$$

where A_d is the dipolar component of the nitrogen coupling and B_0 is a constant equal to 17.1 gauss. The maximum value of A_d occurs when H is along the nitrogen $2p\pi$ orbital, so that $\theta = 0$ and $Ad = 2B_0 \rho_N$. In the present case, the nitrogen coupling is composed of both isotropic and anisotropic components, with the isotropic component of $1/3(A_x + A_y + A_z)$ being equal to $1/3\ A_{||}$, or 5 gauss. Thus, with $\theta = 0$, $A_d = 10$ gauss and $\rho_N = 0.29$. This value is exactly the same as that reported for the spin density on $N_{(1)}$ in cytosine (6).

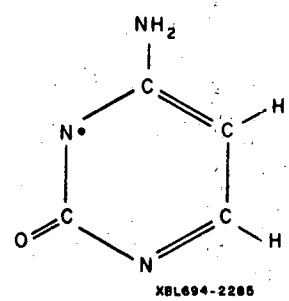
The spin density on $C_{(5)}$ (ρ_C) may be determined from the hydrogen hyperfine interaction. Here, the isotropic component of the coupling (A_f) is related to ρ_C by the formula of McConnel and Chestnut (11),

$$A_f = Q \rho_C,$$

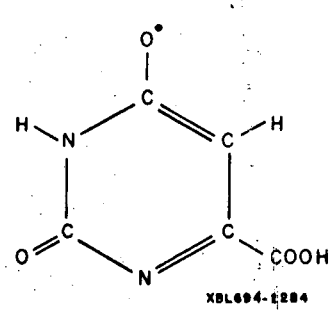
where Q is an empirical constant. With the value of Q used for cytosine, and with the principal values in Table I, ρ_C is found to be 0.64. In cytosine, the spin density on $C_{(5)}$ was found to be 0.72.

In comparing the radicals in these two compounds, it should be

noted that, in cytosine, an additional spin density of 0.15 resides on $N_{(3)}$, due to the resonance structure



This is not possible in orotic acid if this compound is in the diketo form, as is usually the case with uracil derivatives. It seems somewhat surprising that the spin density on $C_{(5)}$ is larger in cytosine than in orotic acid, since cytosine has the additional site for spin density on $N_{(3)}$. It may be that the difference is due to the different groups on $C_{(4)}$ in the two compounds. In orotic acid, some additional spin density may reside on $O_{(4)}$, due to the resonance structure



and this would not be possible in cytosine. There is, of course, no evidence for spin density on the oxygen atom, since the O^{16} nucleus has no magnetic moment.

MOLECULAR ORIENTATION IN THE CRYSTAL

Since the crystal structure of orotic acid is apparently not known, it is of value to predict the orientation of the molecules in the crystal from the ESR analysis. This can be done on the assumption that the radicals in the crystal do not reorient following hydrogen abstraction from $N_{(1)}$. First, from the direction of the minimum g-value and the maximum nitrogen hyperfine coupling, it can be concluded that the pyrimidine ring lies very nearly, and probably exactly, in the a'b' plane. This is the case for all molecules in the unit cell. In the a'b' plane, some additional information regarding the molecular orientation can be obtained from the directions of the hydrogen principal hyperfine values. Theoretically (12), the minimum principal value for an α proton should occur with the magnetic field along the C-H bond, and the maximum value in a direction perpendicular to both the C-H bond and the carbon p-orbital. Thus, these molecular directions can be related to the a' and b' axes. Due to the asymmetry of the orotic acid molecule, there are two possible orientations which will satisfy the conditions, these corresponding to a rotation of 180 deg about the C-H bond. Figures 9(a) and 9(b) show the two possible orientation schemes. In each case, two molecules are shown, corresponding to the two magnetically distinct species observed in this plane. It seems likely that there are four molecules in the unit cell of orotic acid, the two additional ones being related to the other two by centers of symmetry. These would not appear as distinct species in ESR data, however, since two molecules related by a center of symmetry are magnetically equivalent. It should be noted

that the ESR parameters only indicate the probable orientation of the molecules within the crystal, and give no information on the spatial arrangement of the molecules.

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Table I.

Principal values of the spectroscopic splitting tensor (g), the nitrogen hyperfine coupling tensor (A^N), and the hydrogen hyperfine coupling tensor (A^H), and the direction cosines relating the corresponding principal axes to the a'b'c' coordinate system. (Plus and minus signs for the direction cosines are for the two magnetically distinct molecules in the unit cell.)

	Principal value	Direction cosines		
		a'	b'	c'
g_u	2.0059	1	0	0
g_v	2.0052	0	1	0
g_w	2.0023	0	0	1
$A^N_{\parallel}$	15.0 gauss	0	0	1
$A^N_{\perp}$	0	$\perp$ to	(0,0, 1)	
A^H_x	7.3 gauss	±0.94	0.34	0
A^H_y	22.2	∓0.34	0.94	0
A^H_z	15.1	0	0	±1

FIGURE CAPTIONS

Fig. 1. Crystal habit of orotic acid, showing the orthogonal a'b'c' axis system employed for the ESR analysis.

Fig. 2. Second-derivative ESR spectra of an irradiated single crystal of orotic acid at four orientations of the magnetic field in the b'c' plane. The angle is relative to the b' axis. Solid bars beneath the spectra give the predicted line positions, based on tensor parameters from Table I. The microwave frequency was 9.4 GHz.

Fig. 3. Plot of the g-value in the three orthogonal planes of the a'b'c' axis system. Solid lines were calculated from tensor parameters in Table I; circles are experimental values. In the a'b' plane, solid and open circles are for the two magnetically distinct molecules in the unit cell.

Fig. 4. Plot of the nitrogen hyperfine coupling, as described in Fig. 3. No plot is shown for the a'b' plane, since the coupling was zero at all angles.

Fig. 5. Second-derivative ESR spectra taken at two microwave frequencies, with the magnetic field along the a' axis. The theoretical patterns beneath the spectra were calculated from the theory of McConnell et al. (9), using tensor parameters from Table I.

Fig. 6. Second-derivative ESR spectra taken with the magnetic field at an angle of 45 deg in the a'b' plane. Solid and dashed lines indicate theoretical patterns for the two magnetically distinct species in the unit cell.

Fig. 7. Comparison of the experimental value of the inner doublet (open circles) and the outer doublet (solid circles) with the theoretical values. The two magnetically distinct molecules are indicated as (I) and (II). Lines are broken in regions where the normalized relative intensity for the transition falls below 0.30. The microwave frequency was 9.4 GHz.

Fig. 8. Plots for the inner and outer doublets, as described in Fig. 7, but for a microwave frequency of 16.2 GHz.

Fig. 9. Expected orientations of the two magnetically distinct molecules of orotic acid in the a'b' plane, as predicted by the ESR analysis of hydrogen hyperfine coupling. The two schemes (a) and (b) cannot be distinguished on the basis of ESR results alone.

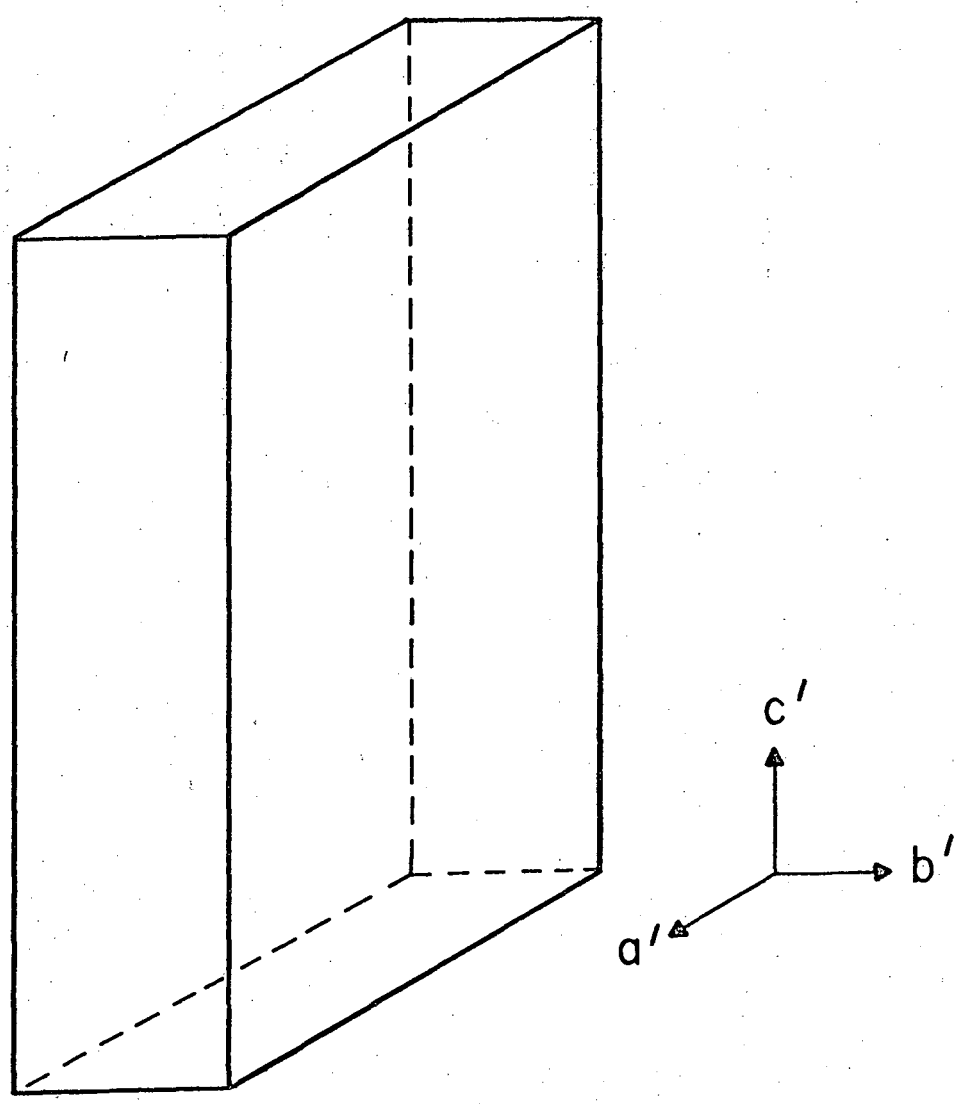

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Fig. 1

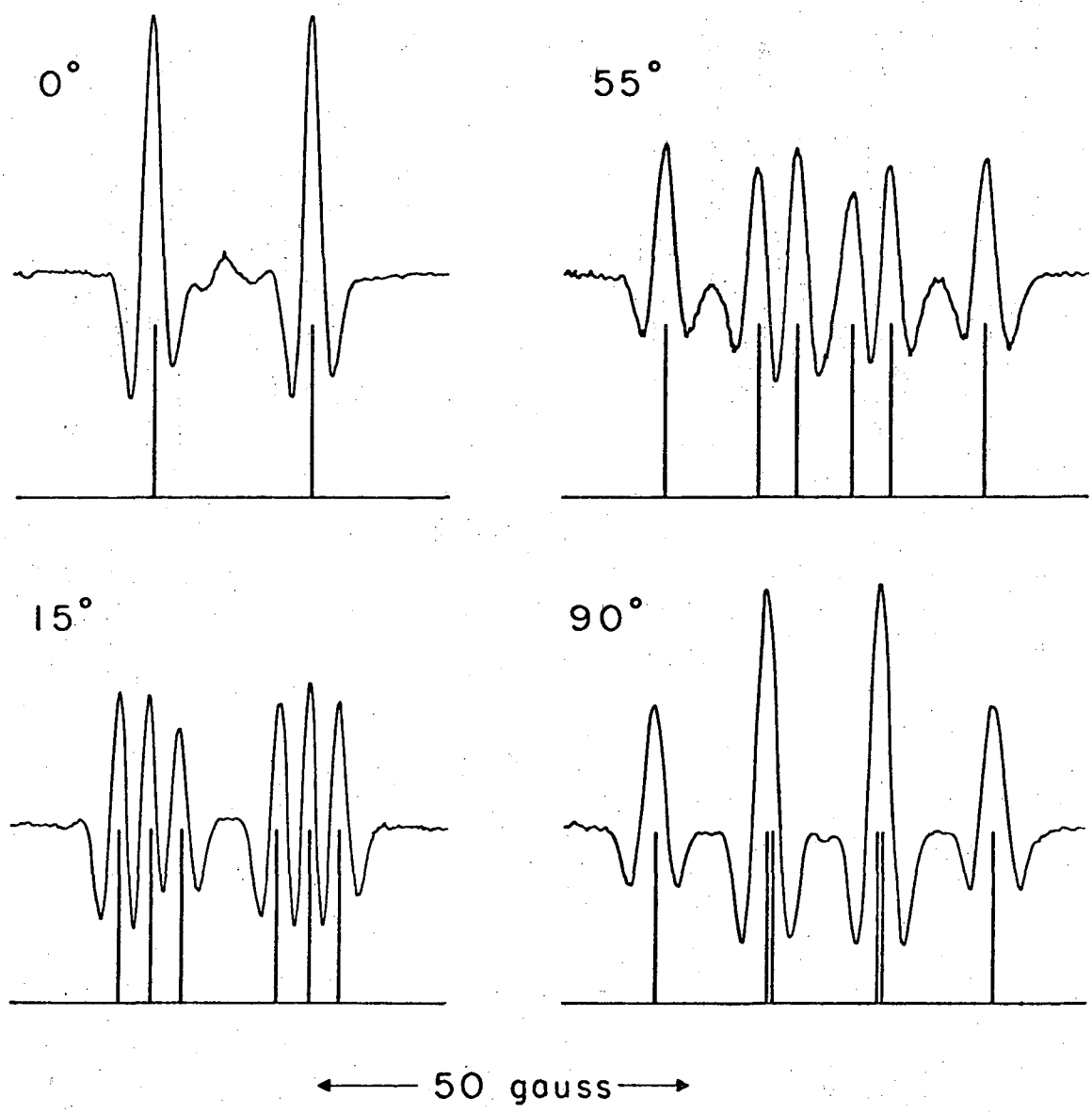

Fig. 2

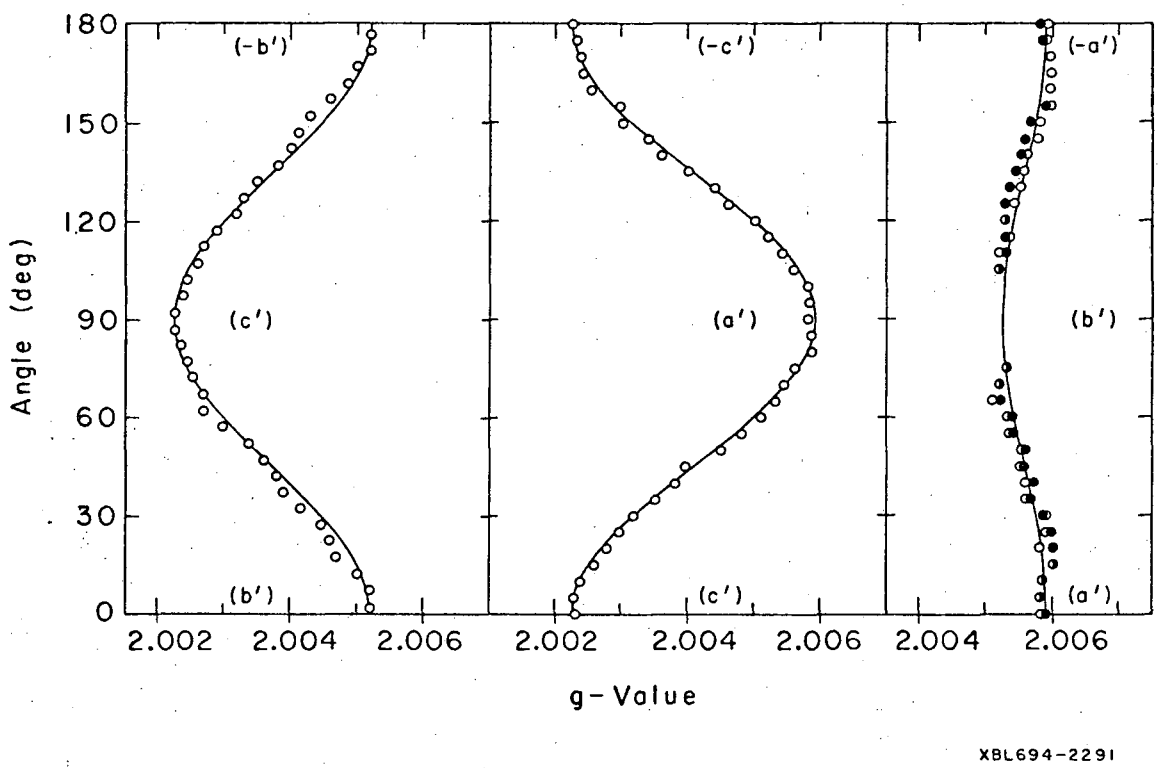
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Fig. 3

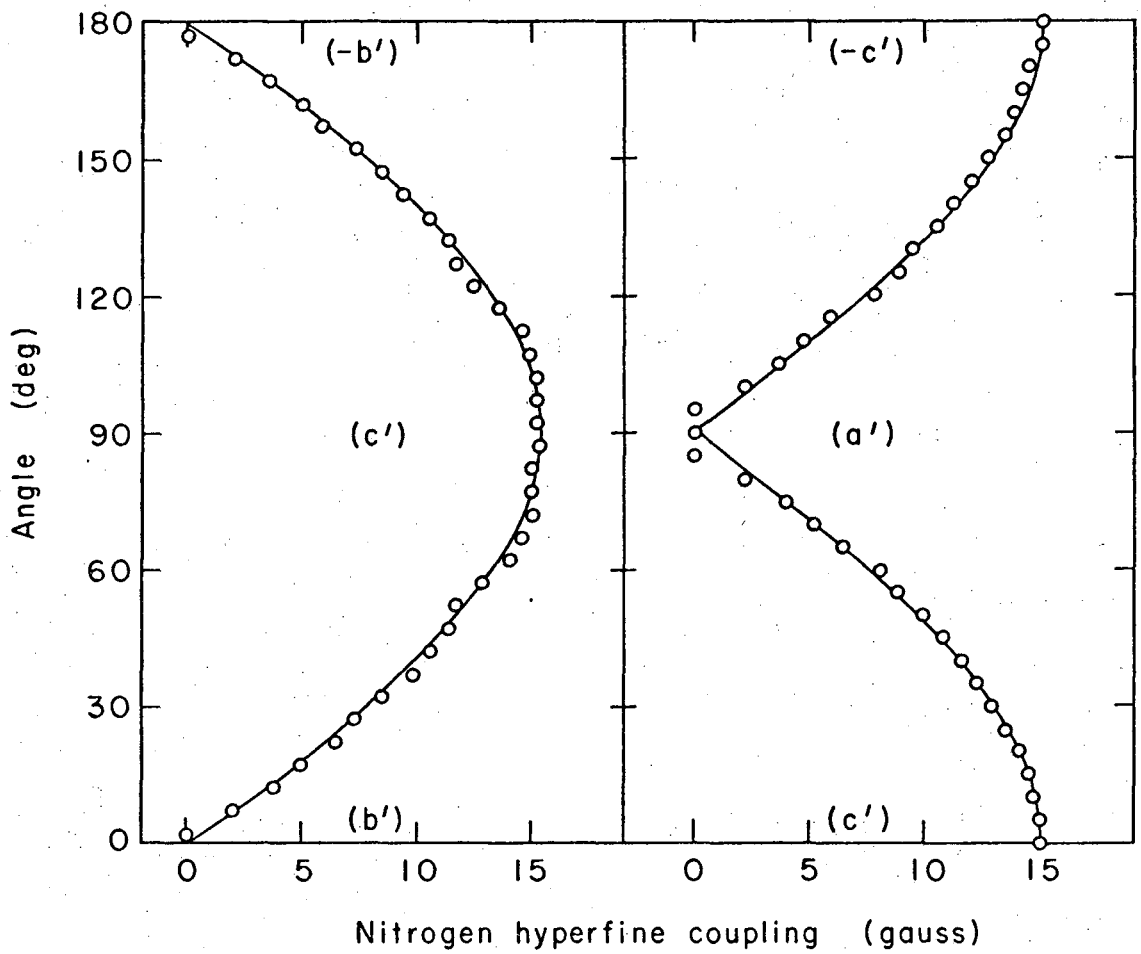

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Fig. 4

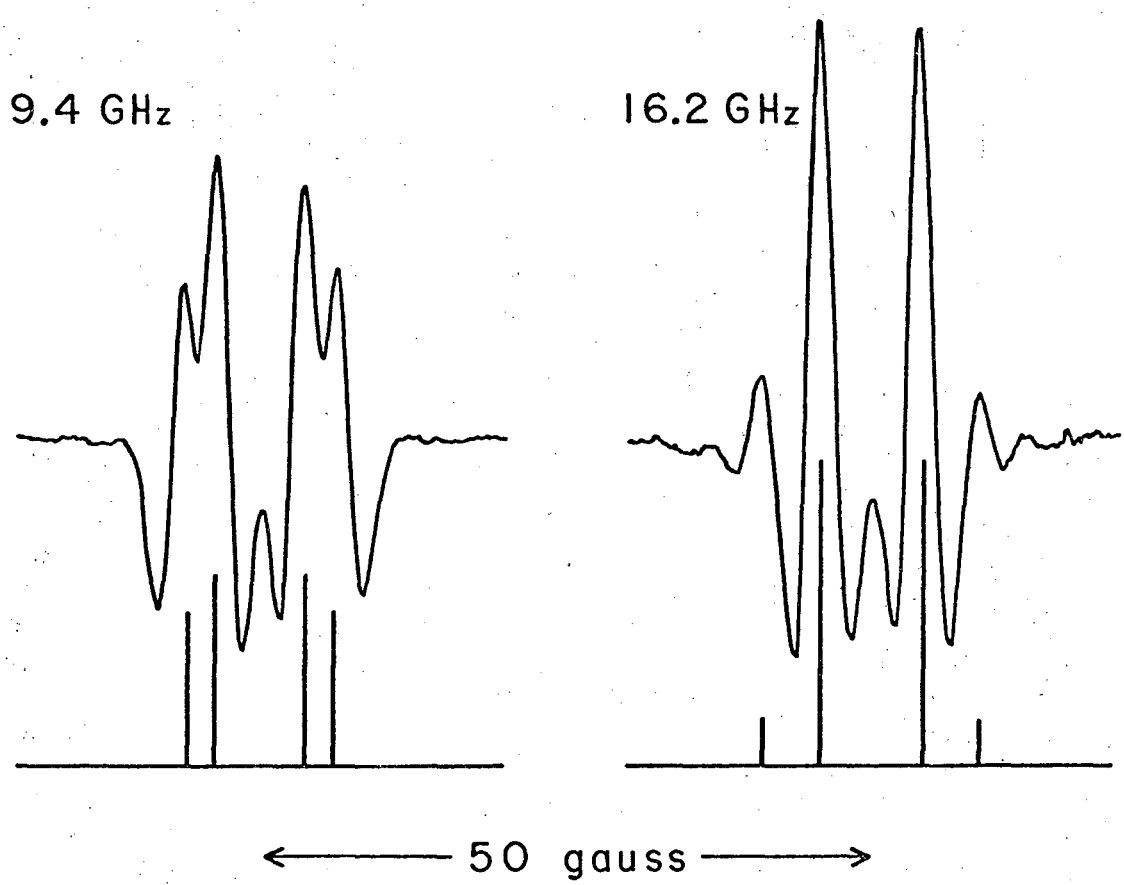

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Fig. 5

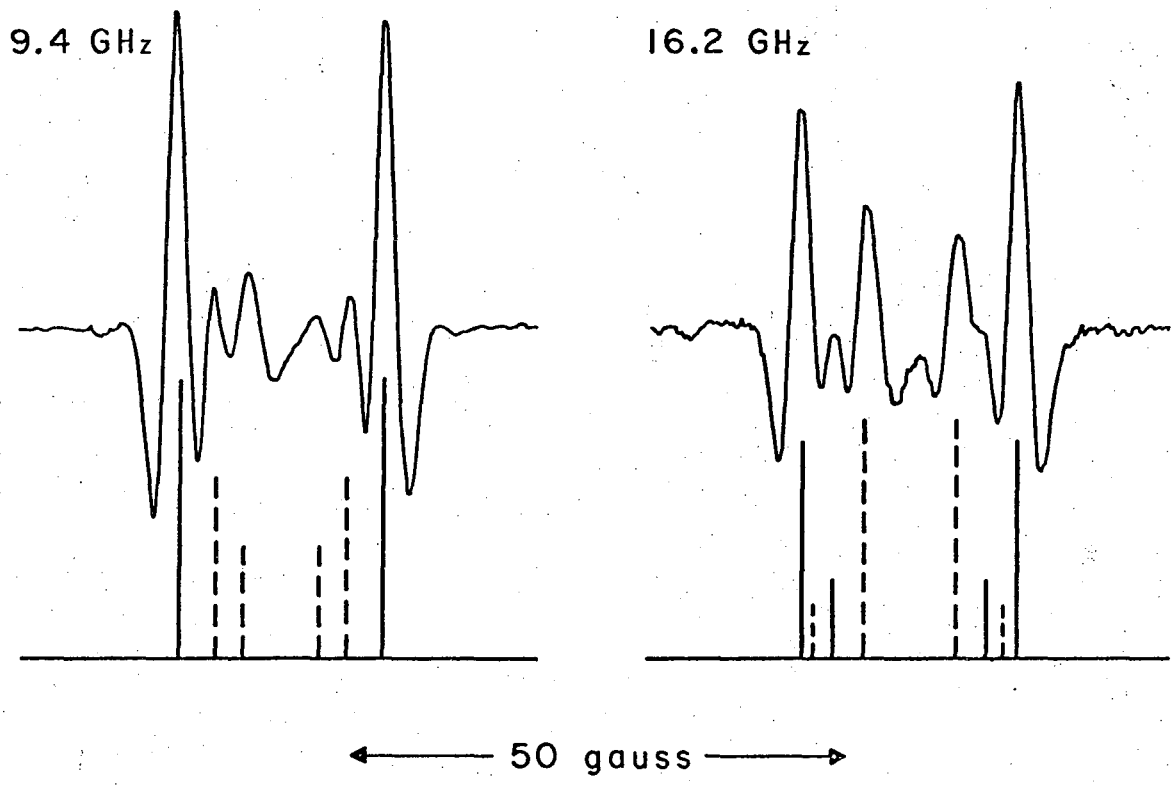

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Fig. 6

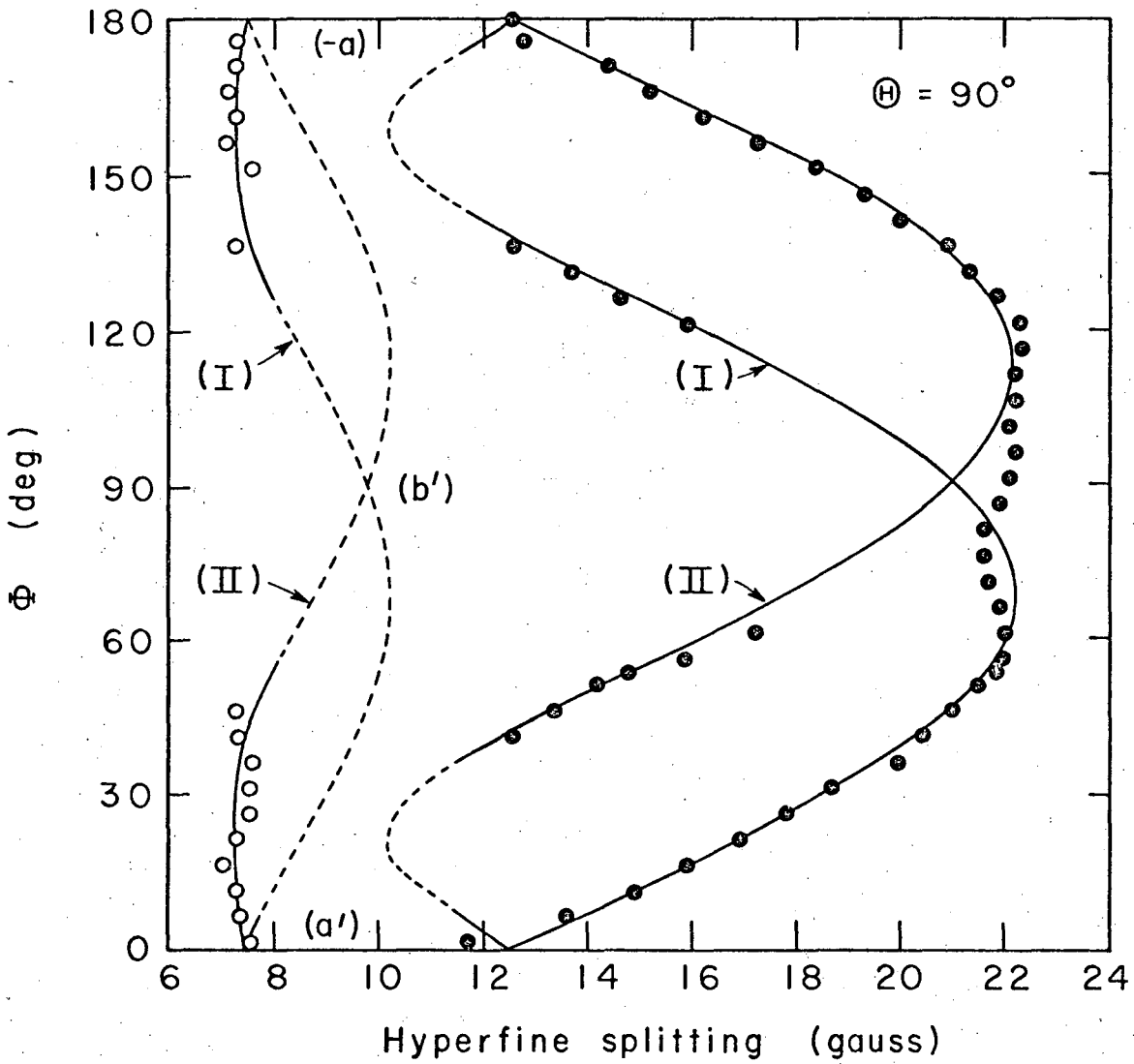

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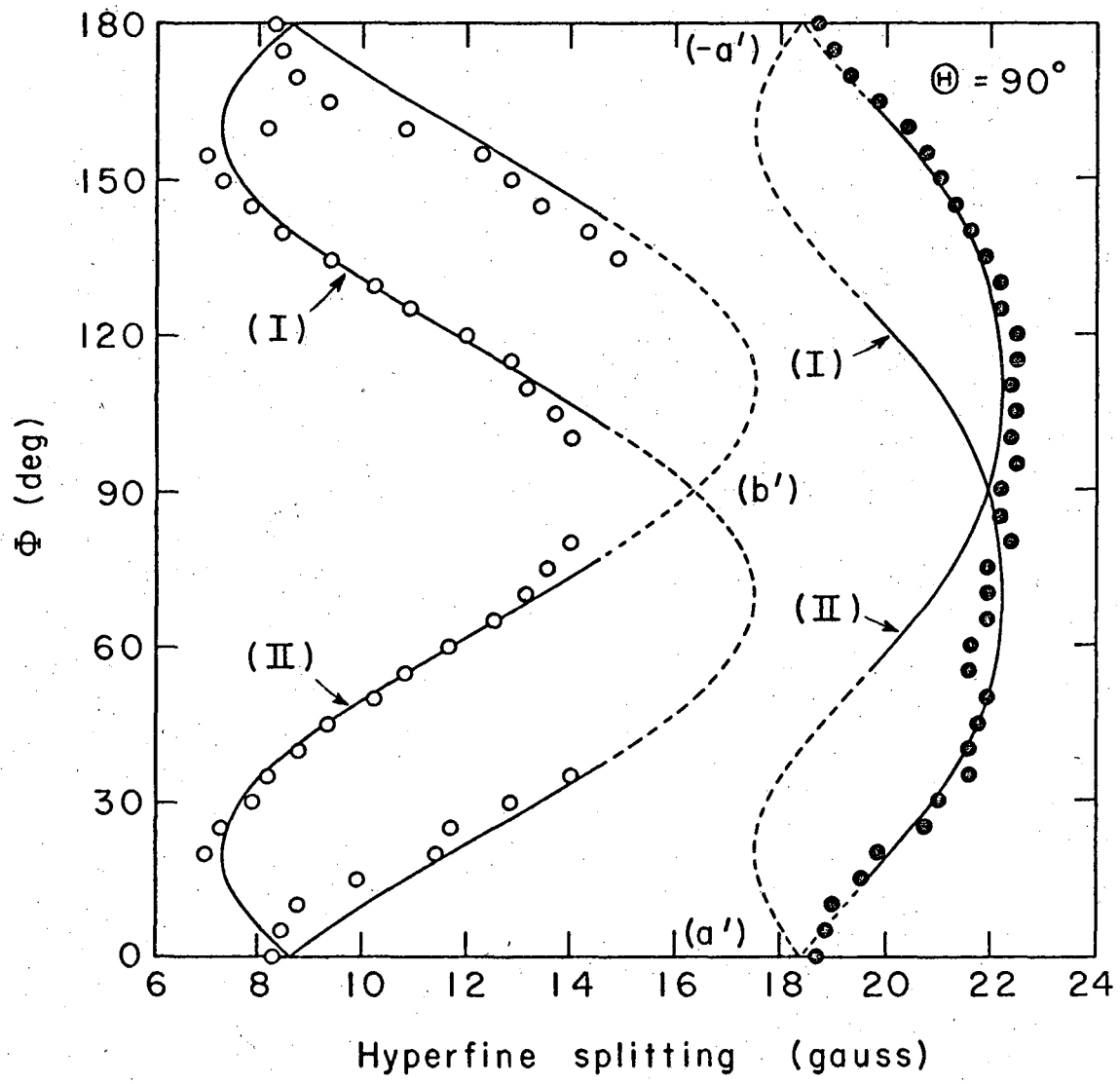
Fig. 7



XBL694-2288

Fig. 8

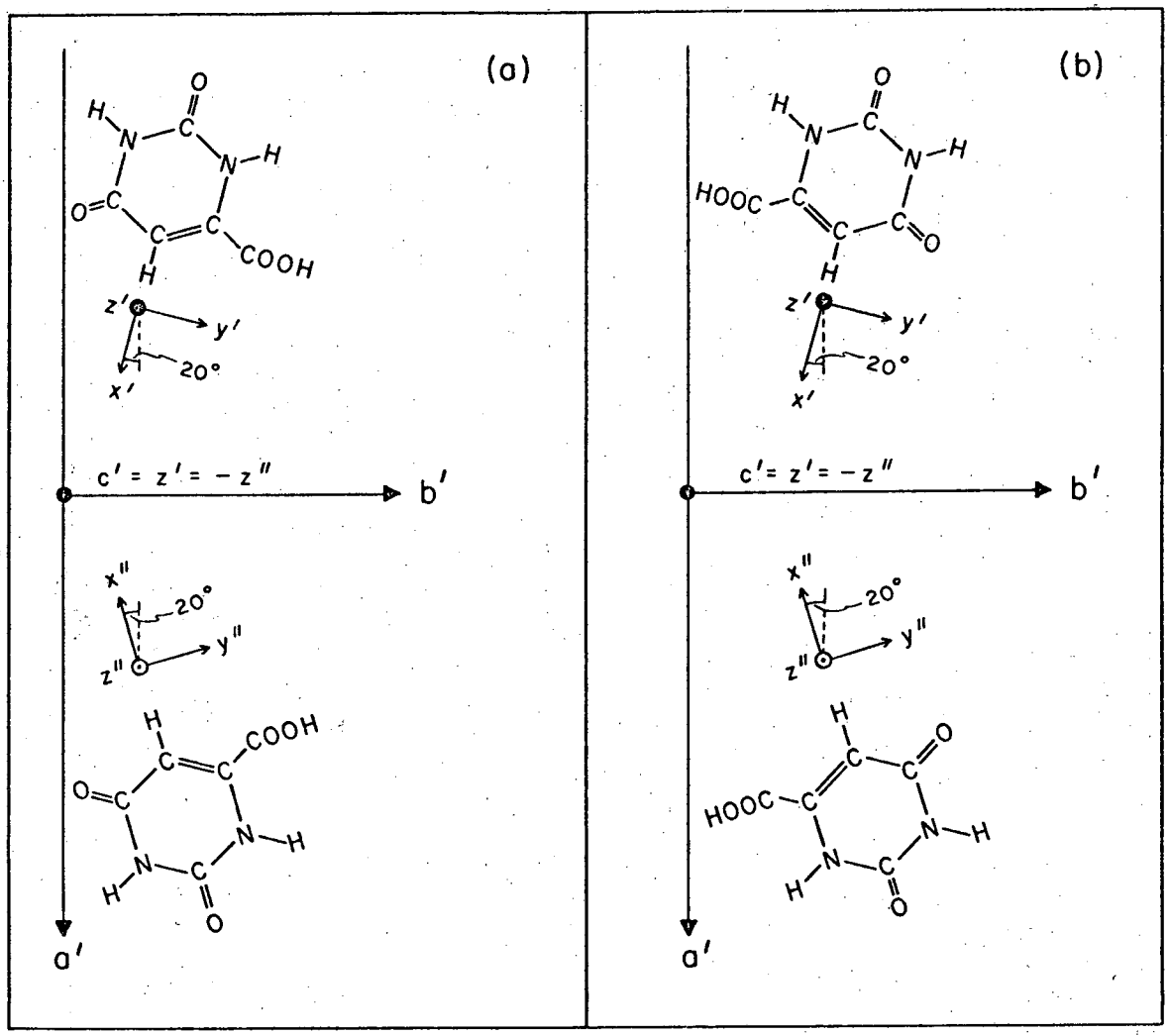


Fig. 9

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