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by

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BRILLOUIN SCATTERING STUDIES OF POLYVINYLIDENE
FLUORIDE FILMS SUBJECT TO UNIAXIAL STRETCH

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ABSTRACT

Brillouin scattering spectra of PVF₂ films were studied for the first time as a function of draw ratio. The two peaks observed in the VV spectrum are assigned as due to scattering from the crystalline and the amorphous regions. The orientation parameter of the polymer chains as induced by stretch is obtained as a function of draw ratio from the hypersonic frequency data. The temperature dependent study shows an anomaly at 50°C, corresponding to the dispersion temperature associated with the primary relaxation of the polymer chain.

INTRODUCTION

A great deal of interests have recently been paid to polyvinylidene fluoride (PVF₂) because of its strong piezoelectricity^{1,2,3}, pyroelectricity^{4,5} and nonlinear optical properties.⁴

PVF₂ crystallizes from the melt into spherulitic structures⁶ with the volume fraction of the crystalline region about 50%, depending on thermal history.⁷ The glass temperature from the amorphous region is about -50°C.⁸ In the crystalline region there are two forms (forms I and III) where the molecules are reported to have a planar zig-zag conformation⁹ with the dipole moment of the CH₂-CF₂ unit being parallel to the b-axis of the unit cell. Therefore, in these forms the PVF₂ crystal has a net polarization along the b-axis and is piezoelectric. A third form (form II), obtained from the melt, contains two molecular chains of tgtg' conformation and is thus intrinsically non-piezoelectric.

Strong piezoelectricity in uniaxially stretched PVF₂ films has been attributed to the molecular chain structure of form I. Imposing strain along the stretch axis diminishes the repulsion force between the CF₂ groups and induces a spontaneous polarization along the axis perpendicular to the film surface, as evidenced by the result of the piezoelectric resonance study.¹⁰ Since the polymer molecules are oriented

by stretching. The induced polarization due to stretching is closely related to the increase of orientation along the direction of stretch.

The X-ray diffraction and IR spectra have shown that the uniaxially oriented films are essentially composed of form I microcrystals with the molecular axis being highly oriented in the stretch axis; however, the biaxially oriented films are mainly composed of the microcrystals of form II.¹¹ While, the strong piezoelectric effect observed from a compressional stress applied perpendicular to the surface of the film¹⁰ has been interpreted as due to the change of form II to form I, NMR studies have failed to detect such a change.¹⁰ Raman studies of the intramolecular vibrational mode also have failed to detect any changes in the sample¹² after poling in a strong electric field.

The inability to detect the changes in the Raman spectral studies is from the sensitivity of the techniques, rather than the absence of preferred orientation. In this paper, we report the first Brillouin scattering measurements of the hypersonic frequency of PVF₂ films as a function of the stretching ratio and temperature. We have detected two pairs of Brillouin shifts on both sides of the Rayleigh peak and have interpreted the result as due to the longitudinal acoustic waves from both the crystalline and the amorphous regions. These observations clearly illustrate the sensitivity of Brillouin scattering technique as an effective method for the determination of the change of polymer orientation due to stretch.

We have related the change of the hypersonic frequency

as the PVF_2 film is stretched to the molecular orientation using the theory of Moseley.¹⁹

EXPERIMENTAL

Polyvinylidene fluoride (PVF₂) of MW~100,000 was obtained in powdered form polysciences Co. Films were melt cast between heated plates in an hydraulic press. The plate temperature was 200°C and the applied load was 2 metric tons. The hot films were kept in the condition for about one hour before quenched in ice water to improve the optical quality of the samples.

PVF₂ films obtained above were stretched in a manually turned film stretcher immersed in a silicone oil bath at 130°C. The PVF₂ film strips were mounted tautly in brass frames directly from the stretcher. The virgin film thickness was 0.260 mm. After stretch, the film thickness decreases to between 0.100 mm and 0.160mm, depending on the draw ratio. The draw ratio was determined by the linear displacement of marks made in the film. The films produced in this manner primarily composed of form II (or α) crystalline structure. The Brillouin scattering spectra of the films were taken with an optical system consisting of a single mode laser radiation at about 4880 Å as the excitation source. The scattering light at a specific polarization wave selected by a Glan Thompson prism polarizer. An interference filter which transmits only 4880±10Å blocked the Raman scattered light. A piezoelectrically scanned triple-pass Fabry-Perot interferometer provided the spectral analysis of the scattered light. The free spectral range was adjusted to be about 3 times of the highest Brillouin frequency shift and the overall instrumental finesse for all spectra was 35-40.

The Brillouin spectrum for each sample was recorded using several free spectral ranges to confirm the identity of the Brillouin shifts. The Brillouin spectra were obtained in various film orientations. The optimum spectra were obtained with the draw axis of the film oriented in the scattering plane but held at 45° to the laser beam while the scattered beam on the other side of the film was collected for further spectral analysis. In this scattering configuration, the scattering vector q lies directly along the draw axis of the film. Due to the presence of the intense surface flare, the scattered light was collected from the opposite side of the incident beam, with respect to the polymer film.

The PVF₂ films are birefringent. However, the average index of refraction of the films along the stretch direction can be determined by matching the refractive index of the films to mixture of organic liquids with known refraction index. The index for all films used in the present experiment did not change appreciably by stretch, with the average value found to be equal to 1.43.

Films having the form I crystalline phase were also prepared and studied. Form I samples were obtained by drawing films in a water bath at 60°C . Films with draw ratios about 4 were found to be of best optical quality and suitable for the angular dependent experiment, and for the temperature dependent measurements. The regulation of temperature was accomplished with a copper block with heating tapes wrapped around the outer surface.

A copper frame holding the film sample ($R_s = 4.1$) was inserted into a narrow channel in the block. Wide angle-bevelled openings of about 0.5 cm diameter on the sides of the block to allow the incident light to penetrate the sample and the scattered light to be observed with minimal reflections from the opening surfaces. Temperature was measured with a chromel-constantan thermocouple attached to the film surface close to the illumination region. Accuracy of the temperature was $\pm 2^\circ\text{C}$. For temperature greater than 150°C , the central Rayleigh component was very intense and covered up the Brillouin side bands.

RESULTS AND DISCUSSION

The Brillouin spectra were obtained in VV and VH configurations for PVF₂ prepared in various forms. The VV spectra display clear Brillouin peaks for the oriented films with draw ratio greater than 3. The Brillouin spectra for the two films of form I and II at the draw ratio equal to 4.2 are shown in Fig. 1. The VH spectra display very weak Brillouin peaks which occur at different frequency shifts from those appearing in the VV spectra. However, due to the poor signal to noise ratio reliable results are obtained only for $R = 4.2$. The peak frequency in the VH spectrum lies at a frequency between two peaks in the VV spectrum and is presumably associated with the transverse acoustic modes. However, further studies to provide a more detailed information of these peaks are needed.

There are two pronounced Brillouin peaks in the VV spectrum, with the peak at the lower frequency being considerably greater in intensity. Furthermore, the low frequency peak is fully polarized and its position depends strongly on the draw ratio of the film. The higher frequency peak is partially depolarized and its position changes only slightly with a change in draw ratio. The positions of the two Brillouin peaks in the VV spectrum are plotted as a function of R in Fig. 2. One notes that the position of the low frequency peak increases with increasing R and approaches an asymptotic value at $R > 6$; whereas the position of the high frequency peak decreases slightly with increasing R .

In the case of a uniform solid, one expects to observe three peaks in the Brillouin spectrum. The three peaks correspond to one longitudinal and two transverse acoustic modes. In the absence of mixing of modes the

longitudinal mode is polarized and the transverse modes are depolarized. The Brillouin spectrum would show a polarized band associated with the longitudinal mode at higher frequency and two depolarized bands at lower frequencies associated with the transverse modes. In the VV spectrum the longitudinal mode will have a greater intensity than those associated with the transverse modes. However, as is clearly shown in Fig. 1, the lower frequency peak has a much greater intensity than the high frequency peak. Further, only two peaks instead of the predicted three for a uniform solid are observed in the VV spectrum.

If one neglects the effect of birefringence the longitudinal and transverse mode frequencies are related by the Poisson ratio ν , as given by

$$\omega_T = \omega_L \left(\frac{1/2 - \nu}{1 - \nu} \right)^{1/2} \quad (1)$$

where ω_T and ω_L are the transverse and longitudinal frequencies, respectively. The Poisson ratio for a typical semicrystalline polymer is about 0.35¹³. The conventional assignment of the low frequency peak at the frequency equal to 5.5 GHz to the transverse wave can be rejected for one reason that Eq.(1) predicts that the longitudinal frequency to be 20 GHz, which we did not see.

In general scattering from the longitudinal wave is what one observes in the Brillouin spectra of semicrystalline or amorphous polymers above T_g , with the exception of samples in which longitudinal and transverse waves are mixed due to the presence of large scale spherulitic polymer crystals¹⁴. As a check of this general result, we have also carried out the Brillouin scattering experiment on a transparent poly(methyl methacrylate) block. A fully polarized doublet at 9.9 GHz from PMMA due to the

central Rayleigh peak corresponding to the scattering of the longitudinal wave appears clearly in the spectrum, yet without any trace of the scattered intensity from the transverse component either in the VV or VH spectrum. Furthermore, the small peak appearing on the high frequency side of the VV Brillouin spectrum of PVF₂ in Fig. 1 cannot be attributed to the inhomogeneous addition of the plasticizer as suggested in ref. 14 for the PVC film, for the reason that the materials used to prepare the PVF₂ films are rather pure.

It is thus reasonable to assume that the two peaks on both sides of the central Rayleigh component are scattering from the longitudinal hypersonic waves from the crystalline and the amorphous regions.

Support for this assignment is obtained from the Brillouin data of the films subject to various degrees of deformation due to stretch. Shown in Fig. 2 are the two peak frequencies as a function of draw ratio. The amorphous longitudinal frequency shown in the lower curve shows a steady increase with increasing R_S until a plateau value is reached at $R_S = 6.5$. This increase is taken as evidence of an increase of chain orientation. The upper curve which is associated with the crystalline longitudinal frequency shows a slight decrease with increasing R . This small decrease is entirely expected according to the theory of elasticity¹⁵ for a crystalline solid subject to a uniaxial stretch. It should be noted that the total decrease in F_B (or in sound velocity) in the crystalline region is small in comparison to the increase of F_B in the lower curve. The small decrease in F_B shows that little or no orientation occurs in the crystalline region when the PVF₂ film is stretched. This result is expected, considering the fact that in the crystalline

region the hypersonic frequency results mostly from the intramolecular interaction, whereas in the amorphous region the mechanism for the hypersonic transmission is largely intermolecular.

Having obtained the hypersonic frequency, we can determine the Hermans' orientation parameter α as a function of R by using the method proposed by Moseley.¹³ The propagation velocity v , of the acoustic wave in partially oriented polymer molecules is related to the orientation function $\langle \cos^2 \theta \rangle$ as¹⁸

$$\frac{1}{v^2} = \frac{1 - \langle \cos^2 \theta \rangle}{v_{\perp}^2} + \frac{\langle \cos^2 \theta \rangle}{v_{\parallel}^2} \quad (2)$$

where θ is the angle between the direction of sound propagation (i.e., q) and the chain axis. v_{\perp} and v_{\parallel} are the limiting velocities of the acoustic wave for a hypothetical sample in which the chain axes of all molecules are vertical and parallel to q , respectively.

Since v_{\parallel} is determined by the intramolecular interaction and is large compared with v_{\perp} , which is mainly due to the intermolecular effect, as an approximation, Moseley has assumed $v_{\parallel} = \infty$, and simplifies Eq. (2)

$$\frac{1}{v^2} = \frac{1 - \langle \cos^2 \theta \rangle}{v_{\perp}^2} = \frac{3}{2} (1 - \langle \cos^2 \theta \rangle) / v_u^2 \quad (3)$$

where in obtaining the second equality of Eq. (3) use is made to relate to the velocity of the unoriented sample v_u by $v_{\perp}^2 = \frac{3}{2} v_u^2$. In terms of Hermans' orientation factor α , Equation (3) can be written as,

$$\alpha = \frac{1}{2} (3 \langle \cos^2 \theta \rangle - 1) = 1 - \left(\frac{v_u}{v} \right)^2 \quad (4)$$

On the basis of this equation we have determined α , using the data given in Fig. 2 and have summarized the results in Table I. As clearly as the

PVF₂ film is stretched, the value of α increases rapidly, reaching the plateau value of 0.65 at $R=7$. The orientation factor does not show further increase at higher R . It would be useful to compare α obtained in the present work with the results obtained using other techniques such as optical birefringence or Raman scattering, NMR, or X-ray diffraction methods. But no such data are available at the present time.

It should be pointed out, however, that the wavelength of the hypersonic wave involved in the Brillouin scattering experiment is of the order of 1000 Å. In order for the well defined Brillouin peaks to be associated with different regions of the polymer sample, it is expected that the homogeneity in the micro-structure in PVF₂ films must exist on a scale exceeding 1000 Å. This is in contradiction to the morphology data of the much studied polyethylene¹⁶ from which a microscopic structure model is contracted for PVF₂. The current structure of PVF₂ is that of alternating layers of crystalline lamellae and amorphous materials with each having a thickness of about 10 to 20 Å. The 1000 Å homogeneous region as detected by Brillouin scattering must indicate that the region of alternating crystalline-amorphous layers is optically homogeneous and implying that the polymer molecule with a length of the order of 50-100 times the lamellae thickness must pass many times through the crystalline and amorphous layers and free to assume flexible configurations in the amorphous layers. Therefore, the bottom curve of Fig. 2 previously associated with the scattering from the amorphous region is better described as originating from the hypersonic response of the amorphous region together with the neighboring crystalline lamellae layers. Since it is well known that the sound velocity in polymer films is relatively insensitive to the degree of crystallinity,¹⁷ the calculated Hermans' orientation factor

as given in Table I should be taken to be a measure of the average orientation of all molecules in the amorphous and crystalline region.

This interpretation does not contradict with the interpretation of the upper curve of Fig. 2 as due to scattering from the crystalline region. As previously mentioned, PVF₂ films are highly birefringent media. In Fig. 3 the hypersonic frequencies are plotted as a function of the tilt angle the film makes with the incident beam. Although over the 50° range the change of frequency in the upper curve amounts to only 3% compared with 7% in the lower curve, the intensity variation associated with the high frequency Brillouin component was quite evident. This result is a clear indication of the effect due to the birefringence of crystalline solids. As shown by Hope,¹⁹ in the birefringent media, the Bragg laws derived for Brillouin scattering must be modified to accommodate the propagation of the extraordinary electromagnetic wave in materials manifesting a strong acoustoelectric effect. Moreover, the effect due to internal reflection in the birefringent crystalline solid must also be included in consideration. While the result of Hope¹⁹ is expressed in terms of integral equations and is difficult to apply to the PVF₂ film, the general result is that the intensity of the Brillouin spectrum in an optically anisotropic medium is altered substantially, depending upon the direction of the scattering vector with respect to the surface of the polymer film. This is consistent with the experimental result found for the high frequency components.

As pointed out above, the thickness of crystalline lamellae is only of the order of 10-20 Å, the fact that the high frequency Brillouin component manifests clearly the feature of a birefringent solid indicate that the scattering is due mainly to the extraordinary component of the electro-

magnetic wave, as the extraordinary component responds only to the micro-structure of the crystalline region and not to the amorphous region of the polymer film.

Finally we consider the temperature dependence of the hypersonic frequencies of the PVF₂ film in form I stretched to $R_g=4$, corresponding to the orientation parameter $\alpha = .54$. In this state, the Brillouin spectra of the PVF₂ film are essentially identical to those of form II stretched to the same draw ratio. Shown in Fig. 4 is the temperature dependence of the two Brillouin frequencies. The temperature dependent data are significant because of the acoustic mode contribution to the pyroelectric coefficient due to the thermal expansion of the piezoelectric polymer (secondary pyroelectricity). It is clear from Fig. 4, in addition to the decrease of hypersonic frequencies with increasing temperature, the data show a considerable dispersion at 50°C. This temperature corresponds well to the observed maximum in the piezoelectric constant in the direction perpendicular to the film surface induced by stretching along the direction of the film.¹⁰ The dispersion at 50°C is attributed to the primary relaxation process of the polymer associated with the motion of the main chain segments in the amorphous phase.¹⁰

SUMMARY AND CONCLUSION

We have studied for the first time the Brillouin scattering spectra of polyvinylidene fluoride films subject to mechanical deformation by stretching. We have assigned two Brillouin peaks of the VV spectrum as rising from the crystalline and the amorphous regions of the polymer film. The high frequency peak which has lower intensity than the low frequency peak is partially polarized and has an intensity variation strongly dependent on the directions of the incident and scattering beams with respect to the surface of the polymer film. These results are qualitatively consistent with the predicted intensity behavior of a birefringent medium. While the high frequency peak shows a slight shift to a lower frequency value, the low frequency peak shifts rapidly to higher frequency value as the polymer film is stretched. We have associated the frequency variation due to stretch with total molecular orientation. From the frequency versus draw-ratio data, we have obtained Hermans' orientation parameter as a function of draw ratio. The temperature dependent study of the film in form I has also been carried out. An anomaly in frequency shift which occurs at 50°C arising from the amorphous region has been detected. This result supports the observation of the earlier temperature dependence piezoelectric resonance study.

Acknowledgement

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FIGURE CAPTIONS

Figure 1. Brillouin spectra of PVF₂ films in form I and II with draw ratio equal to 4 at room temperature obtained in the UV scattering configuration. The phonon propagation direction (q) is along the draw direction of the PVF₂ film.

Figure 2. The Brillouin frequencies observed in the VV configuration as a function of draw ratio. The broken lines are drawn to indicate trends and have no theoretical meanings.

Figure 3. The Brillouin frequency of form I PVF₂ film plotted as a function of the tilted angle of the film with respect to the incident beam.

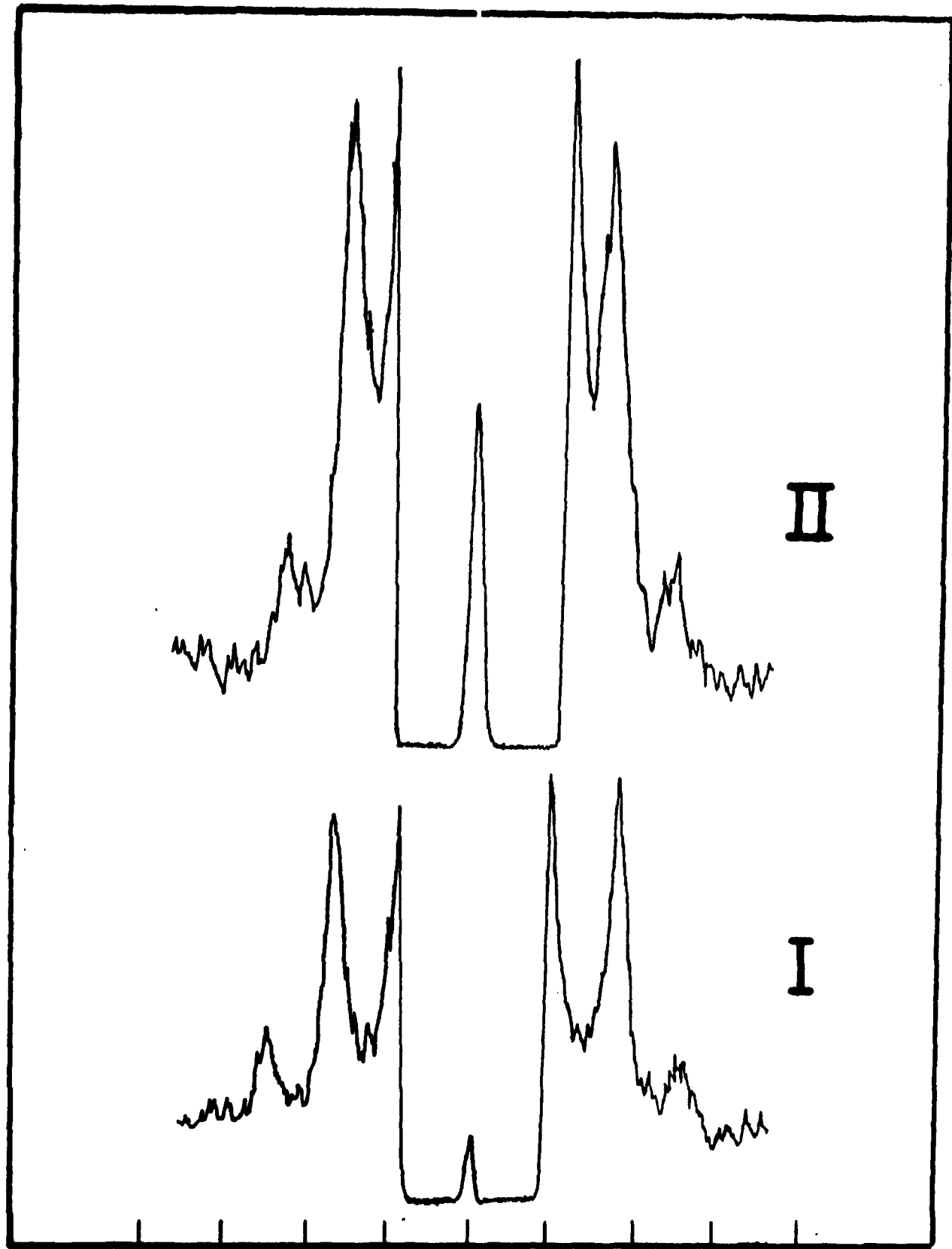
Figure 4. The temperature dependence of the Brillouin frequencies. Note the effect at 50°C.

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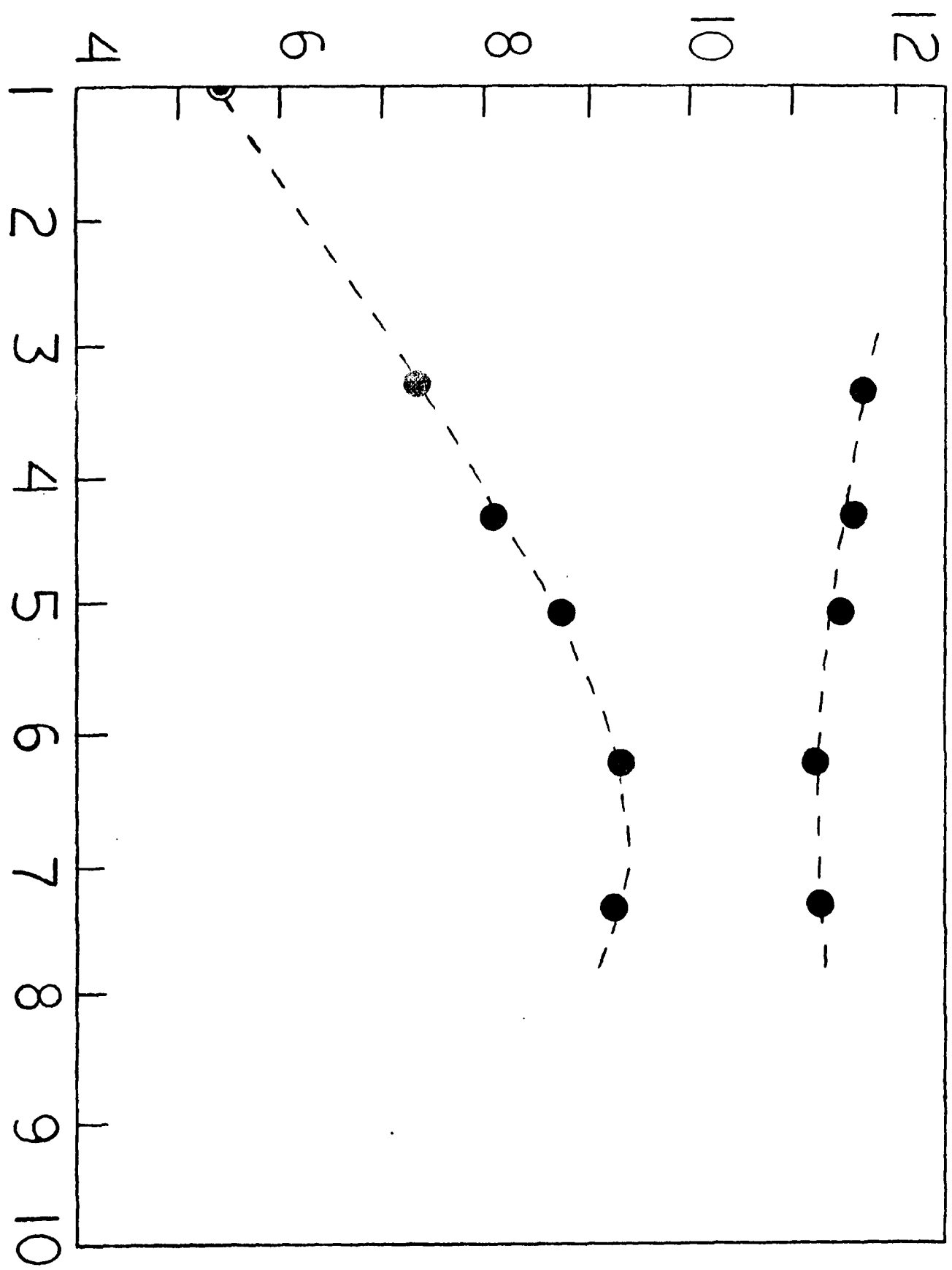
TABLE I

<u>R_s</u>	<u>V_s (km/sec)</u>	<u>α</u>
1.0	1.315	---
3.4	1.774	.45
4.3	1.948	.54
5.1	2.106	.61
7.4	2.225	.65



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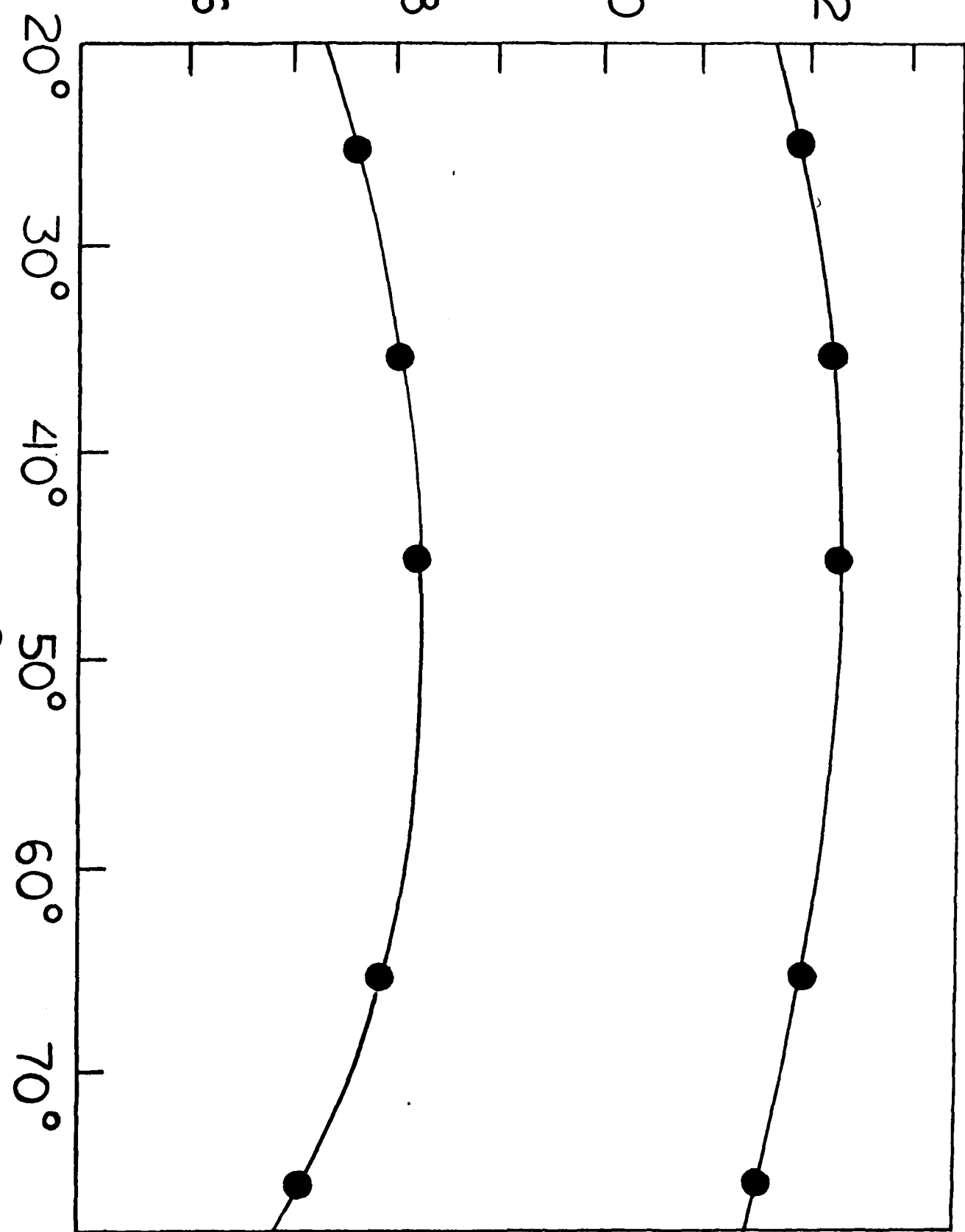
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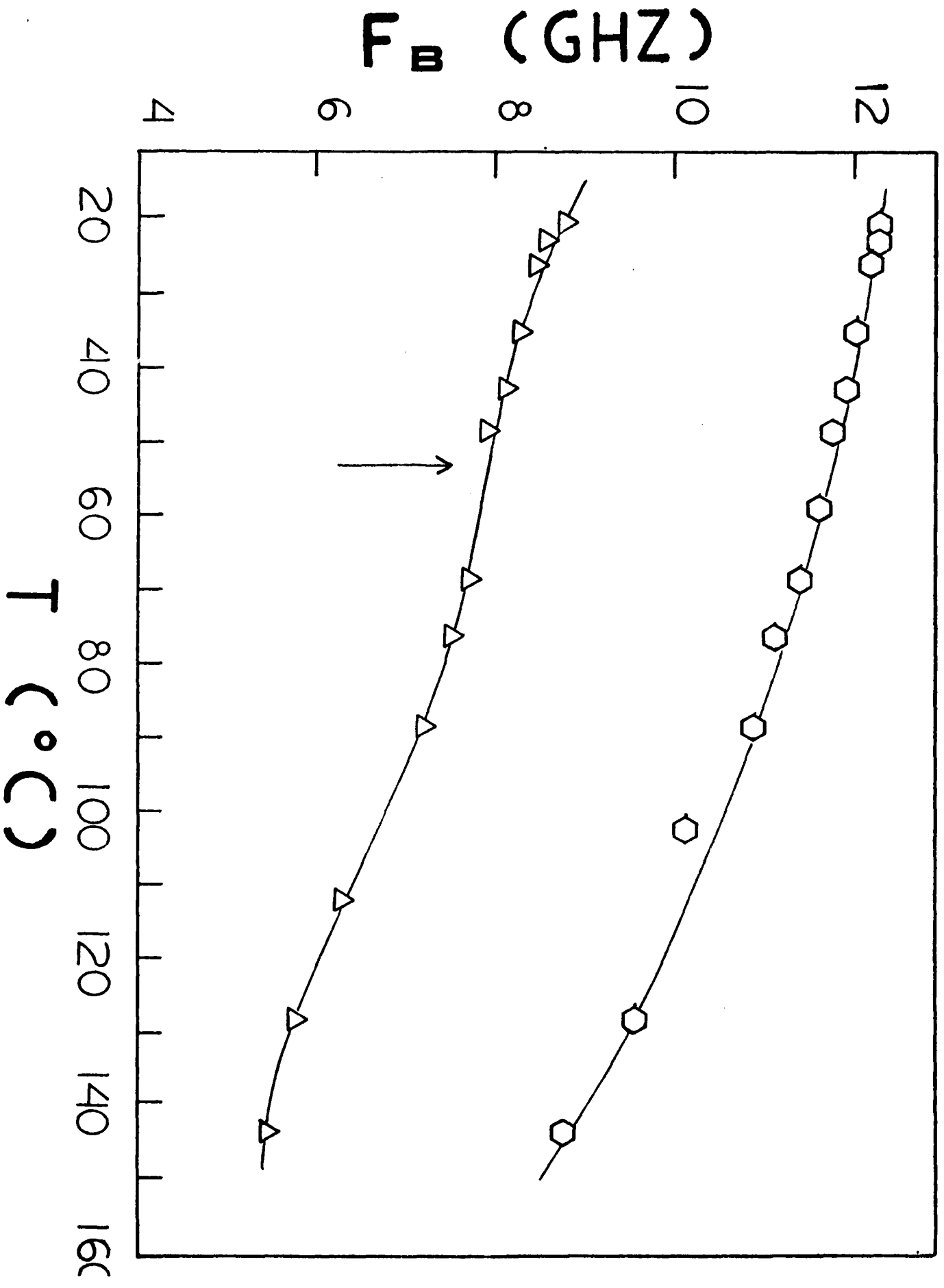
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