

THE ROLE OF CONTACTS IN THE GENERATION OF  
CHARGE-DENSITY-WAVE-CONDUCTION NOISE

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Experiments have been performed to investigate the physical origin of the narrow band noise which accompanies charge density wave conduction in the trichalcogenides  $\text{MX}_3$ . In one experiment we show by current shunting that silver paint contacts seriously perturb the non-Ohmic current. The sample is segmented into parts which oscillate independently. The amplitude relation between frequency components from different parts of the sample suggests that the origin of the noise is at the contacts. In a series of experiments on nine samples we find that the ac voltage of the noise is independent of the length of the sample. This confirms that the noise is due to a fluctuating voltage at the contacts rather than a bulk ac current. We discuss these results in terms of phase slip at the sample ends and describe the role of phase vortices (dislocations) in noise generation.

One of the most interesting aspects of non-Ohmic conduction in the linear chain compounds  $\text{NbSe}_3$  and  $\text{TaS}_3$  and the halogenated compounds  $(\text{TaSe}_4)_2\text{I}$  is the generation of well defined voltage oscillations when a dc current is maintained in the sample<sup>1-5</sup>. These ac oscillations are commonly called charge-density-wave (DCW) conduction noise although the frequency and harmonic can be very sharply defined. The frequency of the ac signal is linear in the CDW current density<sup>2</sup> with a slope directly proportional to the order parameter of the CDW condensate. Various models have been proposed to account for this remarkable phenomenon, the simplest of which is the rigid classical model<sup>6,7</sup> which ascribes the noise to the modulation of the CDW velocity by the periodic potential of the pinning centers. This simple model is largely unsatisfactory for the following reasons: 1.) It cannot account for the strikingly sharp spectrum observed in the purest samples. The existence of domains<sup>8,9,10</sup> in the weak pinning limit implies that some mechanism is necessary to lock these oscillations in phase as well as in frequency. 2.) In real samples with one impurity per 20 host atoms along the chain such a simple model implies that the ac noise is phase incoherent and would vanish when integrated over the different domains. The other difficulties faced by this model relating to the question of how the CDW depins has been discussed in the literature and will be peripheral to the work described here. A survey of our experimental findings is given, followed by a discussion of the vortex-array model recently proposed by Ong, Verma and Maki<sup>11</sup>. Since Professor Maki<sup>12</sup> is presenting a theoretical discussion of the vortex model in these proceedings I shall focus on the experimental aspects here.

In the first experiment<sup>13</sup> we address the question of what happens to the coherence of the noise spectrum when the longitudinal component of the electric field is intentionally made inhomogeneous. To accomplish this we shunt the middle segment of a four-probe sample with a variable resistor. As shown in Fig.1 the spectrum of a four-probe sample which usually has three or more distinct components in the fundamental is altered by the shunting. One or several of the components drops in frequency by an amount proportional to the shunting conductance. The remaining components are unshifted. If we now move the shunting resistor to a different segment of the sample a different set of components moves as the shunt resistance is varied. This procedure enables us to identify components with the individual segments rather unambiguously. The ease with which the individual components are moved relative to each other implies that the three segments oscillate independently and that the frequency fundamental is determined solely by the current in a

particular segment.

An examination of the field distribution of the silver paint voltage probe contacts suggests<sup>13</sup> a mechanism for the disruption of the CDW current. The longitudinal component of the E field which determines the CDW current drops abruptly, leading to a discontinuity in the CDW velocity. Near threshold the portion of the sample covered by the paint remains pinned while the condensate in the uncovered portions slide. The way the condensate resolves this phase interruption is discussed below. In any case the current is carried by free carriers in the portion covered by the paint. This experiment explains a puzzling feature of narrow band spectra observed by some groups<sup>2,5,7</sup>. At high currents the spectrum often "cleans up" and only three strong fundamentals which are not harmonically related remain. These are the  $F_0$ ,  $F_1$ ,  $F_2$  of Richard et al<sup>7</sup>. In the light of our result these three frequencies are to be associated with the three segments which support slightly different CDW currents. A second feature is the appearance of small sub-fundamental peaks (sometimes as low as 1/10 the frequency of the strong fundamentals) at high E fields (e.g. peaks D, E, F in Ref. 5). These low frequency peaks are probably associated with the slower velocities of the covered portions which are depinned at high fields.

A second observation<sup>13</sup> is the surprising change in the relative amplitudes of the different components (again focussing only on the fundamentals) when we move the amplifier and grounding lead from contact to contact. (No other experimental parameters are changed.) For example, when both leads are on segment gamma (see Fig.2) the component arising from alpha is strongly attenuated while the gamma component is observed with large amplitude. On the other hand when the amplifier is on the alpha end the relative heights are reversed. This observation implies that the strengths of the signals are rapidly attenuated as the observation point is moved away from the source, which is rather surprising from transmission line considerations. The frequency is relatively low (1 MHz) and the length of the sample is about 2mm. This strong attenuation of the ac signal suggested to us that the source is highly localised at the contacts (whether used as voltage or current probes.) A natural deduction<sup>13</sup> then is that the phase slip caused by the abrupt drop in  $E_x$  at the contact pads is also causing the ac oscillations in the voltage when the CDW conducts.

In a third series of experiments<sup>13</sup> we explored the length dependence of

the ac voltage amplitude. On the basis of some early results we concluded that the ac voltage amplitude  $v_{ac}$  is independent of sample length  $l$ . Recently Grüner et al<sup>14</sup> have performed the same studies and obtained results different from ours. They find that the ac voltage varies as the square root of  $l$  for three samples. To address this disagreement we have increased our sample and data base. In addition we have integrated the total signal intensity throughout the entire spectrum as opposed to just examining the fundamental. Our new results confirm that  $v_{ac}$  is indeed length independent. They also suggest reasons for the experimental disagreement. In our studies a long two-probe sample was mounted in a dewar in which the thermal stability was typically 30 mK at 40 K. The ac noise was preamplified with a Trontech preamp of passband from 80 kHz to 40 MHz and 70 dB gain. the spectrum was resolved with a spectrum analyser (Hewlett Packard 8557A) which has a frequency span of 10 kHz to 330 MHz and a resolution bandwidth of 1 kHz. Beacuse  $v_{ac}$  fluctuates by as much as a factor of 2 in a time scale of several seconds we signal averaged the output of the HP 8557A with a microcomputer over 20 scans and stored the averaged spectrum. Three frequencies (200 kHz, 1 MHz, and 10 MHz) were explored. After each measurement the sample was shortened by extending the paint area and recooled to the same temperature.

In earlier runs only the area under the fundamental peak was integrated. In some samples (see Fig.3) this was found to be independent of  $l$  even in the presence of substantial broadening for the shortest sample length. In Fig.4 we have collected together the integrated area (under the fundamental) for several samples and plotted it normalised to the starting length. As may be seen the data is constant over a length variation of approximately ten. However, some samples showed a resonant behavior indicating an erratic growth in signal strength (solid circles in Fig.4). To address this feature we extended our studies to samples whose lengths could be varied by a factor of 60. In addition we captured the full spectrum rather than just the fundamental. Figs. 5 and 6 show the variation of the spectrum for selected lengths of two samples. Three main features are noteworthy. First, over a range of 60 in length (Fig. 5) the area under the full spectrum is roughly constant. Secondly, the quality of the spectrum is drastically affected by contact configuration. For example, in Fig. 5 in going from 1.04 mm to 0.30 mm the spectrum degrades to broad peaks with increased broadband noise. On further length reduction to 0.13 mm the spectrum sharpens up again with little braodband noise. Thirdly, restricting one's measurement to the fundamental alone (or worse just the amplitude at te peak of the fundamental) will lead to

a spurious length dependence (see Fig. 6). Altogether we have examined 9 samples of high-purity  $\text{NbSe}_3$  each shortened four times on the average. Our conclusion is that the ac voltage amplitude is length independent.

The second point mentioned above may be viewed as further empirical support for the contact model. During the course of studying the total spectrum of several samples we noticed that the "quality" of the spectrum (sharpness of the resonance, presence of broad band noise, etc) is drastically affected when the contacts are altered. (We checked that different cooling rates are not effective in altering the quality of the spectrum. The fine structure and harmonic content are surprisingly reproducible on cooling after warming up through the transition.) Such sensitivity to contact configuration is difficult to explain using the rigid condensate model (with a distribution of domains). In particular, it is highly implausible that changing the contacts alone can alter the bulk current (or the interdomain coupling) to the extent of sharpening the spectrum or degrading it. On the other hand the contact model is quite consistent with this observation. Depending on the way the contacts are laid down and the resulting sharpness of the interface between the covered and uncovered portions of the sample the spectrum can be made to be coherent or incoherent.

The fact that the ac voltage is independent of sample length  $l$  rules out the bulk current as a source of the ac noise. In the rigid classical model, Grüner et al<sup>6</sup> suggest that such an ac bulk current  $J_{\text{CDW}}$  arises from the velocity modulation caused by the periodic pinning potential. For a single-domain sample the ac voltage would be given by  $J_{\text{CDW}} \times R$  (where  $R$  is the sample resistance) which scales as the length  $l$ . (In a multidomain model phase incoherence may lead to a fractional power dependence.) The lack of  $l$  dependence in our result implies that the oscillating component of the voltage is associated with a small component of the voltage is associated with a small portion adjacent to the contacts. A mechanism by which the voltage of the sample is modulated by the phase slippage at the contacts is discussed below.

The most economical way for the CDW condensate to resolve the phase inconsistency at the contacts (and at the ends) is by the generation of phase vortices or dislocations in the superlattice structure<sup>11</sup>. The free energy involved in creating an array of vortices of spacing  $l_v$  in a sample of width  $w$  is

$$\Delta F_V = (\omega N_0 v_F^2 / l_V) \ln(\lambda/\xi) \quad (1)$$

where  $N_0$  is the electronic density of states,  $v_F$  is the Fermi velocity,  $\lambda$  the Fukuyama-Lee-Rice<sup>8,9</sup> (FLR) length and  $\xi$  the BCS coherence length equal to  $\hbar v_F / \Delta$  (where  $\Delta$  is the CDW gap). The FLR length is the approximate size of the domain in the weak-pinning limit. We may express the ratio  $\lambda/\xi$  in terms of directly measurable quantities as

$$(\lambda/\xi)^2 = (\Delta/6\alpha E_F)(\Delta/eE_T \lambda_{CDW}) \quad (2)$$

where  $\alpha$  is a number of order 1,  $E_F$  the Fermi energy,  $E_T$  the threshold field and  $\lambda_{CDW}$  the CDW wavelength. For the purest samples of NbSe<sub>3</sub> ( $E_T$  approx.<sup>10</sup> 10 mv/cm) we estimate  $\lambda/\xi$  to be 130, or  $\lambda$  is of the order 10 to 50 microns. Comparing the cost in Eq. 1 with the energy  $\Delta F_S$  required to drive the condensate normal in a sheet of thickness  $\xi$  at the contacts we find

$$\Delta F_V / \Delta F_S = (\xi/l_V) \ln(\lambda/\xi). \quad (3)$$

From Eq. 3 it is clear that it is always more favorable to create the vortex array to reconcile the phase mismatch instead of driving the condensate normal.

On general grounds we expect that vortices will be generated under sliding conditions in the regions of the sample where phase discontinuities occur (between domains, at the sample ends, and near strong pinning centers). The role of the vortices in removing the phase mismatch may be clarified by viewing them as edge dislocations in the superlattice (Fig. 7). The open arrow indicates the CDW velocity  $v_D$  when depinned. Each time the phase front of the CDW condensate (represented by the thin lines in Fig. 7 advances by a wavelength a dislocation (indicated by an inverted T) moves transverse to the CDW velocity. Since the dislocation carries a phase of  $2\pi$  the rate of vortex annihilation at one side of the sample is determined by the bulk velocity by

$$Qv_D = 2\pi v_S / l_V \quad (4)$$

where  $Q$  is the CDW wave vector and  $v_S$  the velocity of the vortices. It may be seen that the frequency  $f$  with which vortices arrive at the side is equal to the left hand side of Eq. 4 multiplied by  $2\pi$ , and thus equal to the washboard frequency. From Eq. 4 the observed frequency is linear in the

condensate current density  $J_{CDW}$  as experimentally established<sup>2</sup>. As shown in Maki's talk<sup>12</sup> the motion of the vortices couples into  $J_{CDW}$  given by

$$J_{CDW} = en_c Q^{-1} \dot{\phi} \quad (5)$$

through the temporal modulation of the CDW phase  $\phi$ . In Eq. 5  $n_c$  is the condensate density. Maki shows that an ac component of  $J_{CDW}$  arises from the ac modulation of the vortex velocities as follows:

$$\Delta J = -2\pi en_c Q^{-1} \sum \dot{y}_i \quad (6)$$

where  $\dot{y}_i$  is the velocity of the  $i$ th vortex. The vortex velocities are modulated by image forces at the sides of the samples. Experimentally the total current through the sample is usually held constant by an active feedback circuit, and the oscillations are modulations of the voltage drop across the sample. In the particular mechanism assumed in Eq. 6 the oscillatory part of  $J_{CDW}$  is converted to an ac voltage by an effective resistance  $R_{eff}$  of the order of  $(\sigma\omega)^{-1}$  where  $\sigma$  is the conductivity of the material. Since  $R_{eff}$  is associated only with the contacts or the ends of the sliding region the ac voltage  $v_{ac} = \Delta J R_{eff}$  is independent of length.

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#### References

1. R. M. Fleming and C. C. Grimes, Phys. Rev. Lett. 42, 1423 (1979); R. M. Fleming, Phys. Rev. B22, 5606 (1980).
2. P. Monceau, J. Richard, and M. Renard, Phys. Rev. Lett 45, 43 (1980); also Phys. Rev. B25, 931 (1982).

3. G. Grüner, A. Zettl, W. G. Clark, and A. H. Thomas, Phys. Rev. B23, 6813 (1981).
4. Z. Z. Wang, M. C. Saint-Lager, P. Monceau, M. Renard, P. Gressier, A. Meerschaut, L. Guemas, and J. Rouxel, Solid State Commun., 46, 325 (1983).
5. N. P. Ong, and C. M. Gould, Solid State Commun. 37, 25 (1981).
6. G. Grüner, A. Zawadowski, and P. M. Chaikin, Phys. Rev. Lett. 46, 511 (1981).
7. J. Richard, P. Monceau, and M. Renard, Phys. Rev. B25, 948 (1982).
8. H. Fukuyama, and P. A. Lee, Phys. Rev. B17, 535 (1978).
9. P. A. Lee and T. M. Rice, Phys. Rev. B19, 3970, (1979).
10. J. W. Brill, N. P. Ong, J. C. Eckert, J. W. Savage, S.K. Khanna, and R. B. Somoano, Phys. Rev. B23, 1517 (1981).
11. N. P. Ong, G. Verma, and K. Maki, unpublished.
12. K. Maki, reported at this conference.
13. N. P. Ong, and G. Verma, Phys. Rev. B27, 4495 (1982); G. Verma, and N. P. Ong, unpublished.
14. George Mozurkewich and George Grüner, unpublished manuscript.

#### Figure Captions

Figure 1 Identification of components of the fundamental in the power spectrum of NbSe<sub>3</sub> at 50 K. By shunting each of the three segments  $\alpha$ ,  $\beta$ ,  $\gamma$  in turn it is possible to identify that segment with the components that are shifted. For example in the middle trace the largest spike is shifted down when the middle segment is shunted. This spike is then labelled  $\beta$  in the bottom trace.

Figure 2 Variation of the relative strengths of different components



when the amplifier leads are moved. The paired numbers m,n indicate the leads that are connected to the preamplifier. E.g. 32 means that the active end is at contact 3 and the ground is at 2. The data show that when the amplifier is at the  $\gamma$  end ( 32, 43 ) the  $\gamma$  lines are enhanced, whereas when the preamplifier is at the  $\alpha$  end (12,13) the  $\gamma$  lines are attenuated.

Figure 3 The insensitivity of the ac voltage to sample length ( Sample A4.). In these four traces of the fundamental the sample length is shortened from 1.19 mm to 0.13 mm. The integrated area under the fundamental is approximately constant, although the width increases drastically at the shortest length. The vertical scale is proportional to the ac voltage, not power.

Figure 4 The plot of the integrated area under the fundamental frequency versus the sample length for six samples. For a given sample the area is measured each time the length is reduced and the result is plotted normalised to the area for the longest length. The vertical axis is linear in the Fourier component of the ac voltage at the fundamental. Note that except for some anomalous points the signal is independent of length.

Figure 5 The full spectrum of  $\text{NbSe}_3$  for various lengths of Sample B3. The length of the sample is shortened from 7.8mm to 0.13 mm. The insensitivity of the integrated area under the spectrum to length variation is clearly demonstrated by these traces. The vertical scale is identical for all traces. For the length 0.30 mm the spectrum becomes dominated by broadband noise, which increases the integrated area, but decreases the height of the fundamental. We interpret this behavior as due to different contact geometry.

Figure 6 The full spectrum of  $\text{NbSe}_3$  for various lengths of Sample B2. As in Fig. 5 the area under the full spectrum is roughly independent of length. Note that as in Fig. 5 some lengths show a preponderance of broad band noise (the shortest in this case).

Figure 7 Schematic representation of the role of phase vortices ( drawn as edge dislocations ) in inducing phase slip near the paint contacts. The condensate under the paint is pinned while sliding occurs in the uncovered portion (open arrow in panel a.) Each time the bulk phase advances by one CDW wavelength a dislocation moves across the width of the sample (solid arrow in panel a.) The hatched stripes identify two CDW phase fronts that are brought

closer together as vortices are annihilated at the top edge of the sample  
(panels b,c,d).

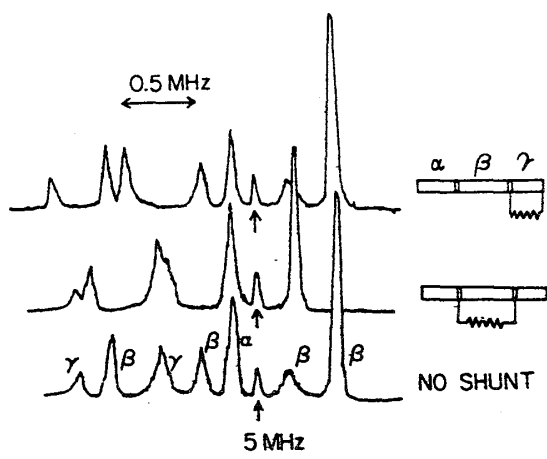


Fig. 1

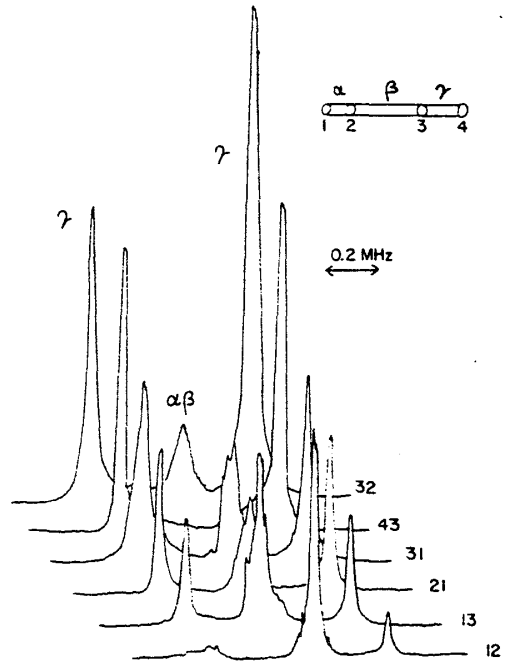


Fig. 2

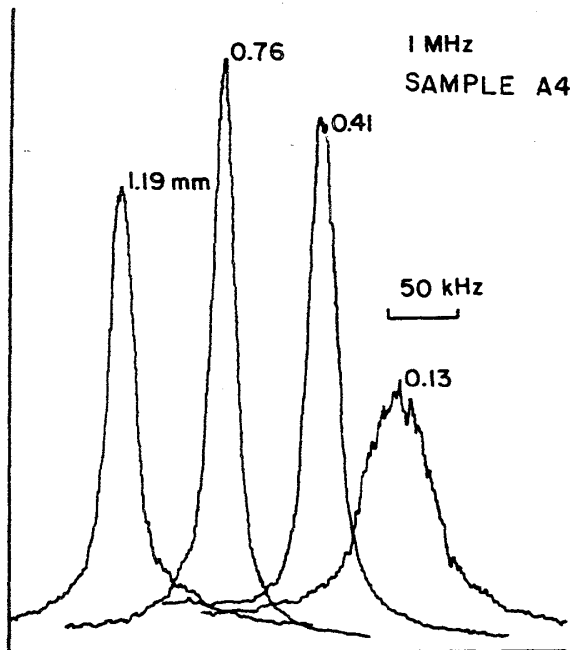


Fig. 3

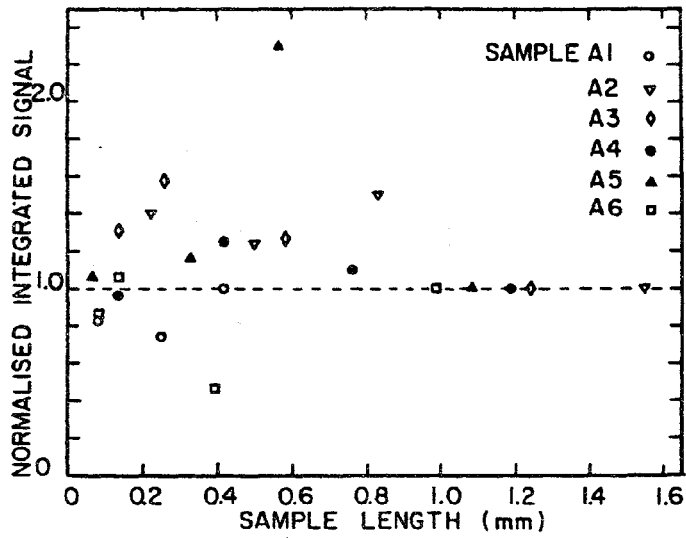


Fig. 4

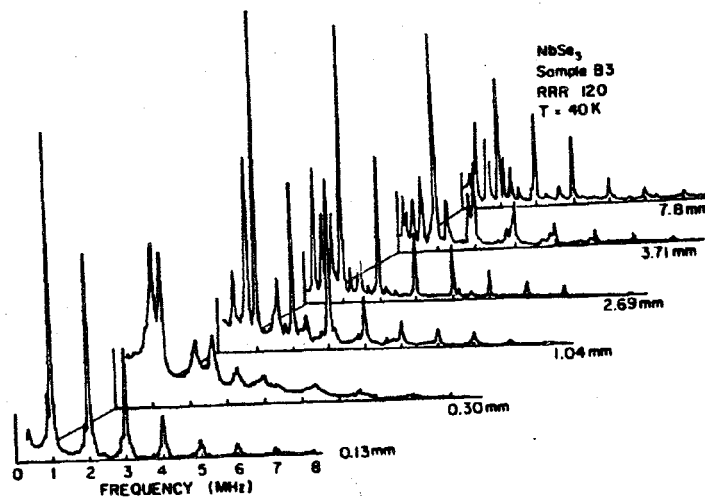


Fig. 5

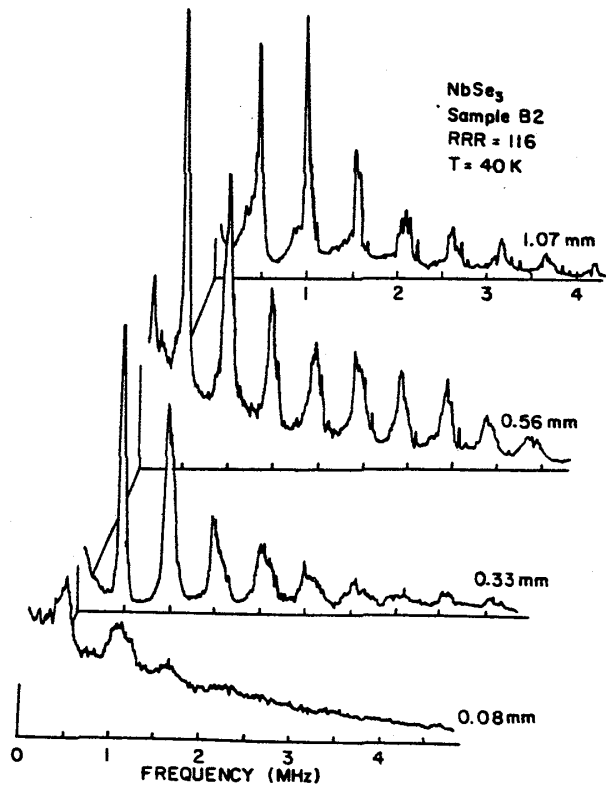


Fig. 6

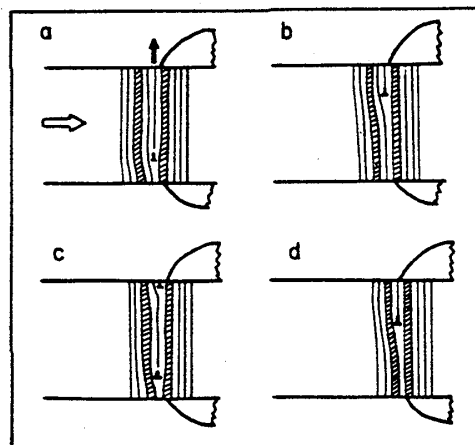


Fig. 7