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CLASSICAL MODEL OF CHARGE DENSITY WAVE TRANSPORT

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A classical model is applied that describes charge transport by pinned and depinned charge density waves (CDW). At low frequency, charge is assumed to be displaced in a potential that is the sum of strong end and weak distributed potentials. In this range the CDW may be treated as a damped harmonic oscillator. Above a critical force the CDW is depinned and moves with resistance from viscous damping and dry friction. By taking the forces of standing and dry sliding friction to be equal and assuming an exponential distribution of coherent CDW regions we obtain the reported form of current vs applied field. By assuming the CDW's to be overdamped we fit the observed frequency-dependent conductivity. We calculate the effect of a dc field on the ac conductivity and the effect on the dc conductivity of large amplitude ac fields. We apply the classical model to an analysis of CDW noise and suggest the excitation of phase modes to be the origin of the observed narrow band noise components.

INTRODUCTION

The striking charge density wave (CDW) transport behavior observed in NbSe_3 ¹, TaS_3 ², and possibly ZrTe_5 and HfTe_5 ³ has been analyzed in terms of two quite different models. The first model, proposed by Lee and Rice⁴, treats the electric field depinning of CDW's stuck at impurities. As we show, their theory leads with certain assumptions to the dependence of current on electric field reported by Fleming⁵

$$j = \sigma_a E + \sigma_b (E - E_T) e^{-E_0/(E - E_T)} \quad (1)$$

where the second term in (1) is operative only above the threshold field E_T . A second model, advanced by Bardeen⁶, considers CDW transport to arise from Zener tunneling across the gap introduced by impurity pinning. This model gives a current of the form

$$j = \sigma_a E + \sigma_b (E - E_T) e^{-E_0/E} \quad (2)$$

for $E_T < E_0$ where the threshold field E_T arises from the finite correlation distance of the CDW. The field E_0 as discussed below follows from the Zener theory. Although (1) and (2) differ in the form of the exponential, the behavior near threshold is dominated by the prefactor and present experiments have not distinguished between the two expressions.

In what follows we first apply the Lee-Rice model to the dc, ac, and coupled conductivities under certain special assumptions. Next we examine the Bardeen model. Finally, we discuss the studies of CDW noise and their implications for modeling of CDW transport. Based on the Lee-Rice model we propose a new explanation of the observed narrow-band noise.

APPLICATION OF THE LEE-RICE MODEL

Lee and Rice⁴ consider the pinning of CDW's by both strong and weak impurities with quite different pinning properties. We assume with Fukuyama and Lee⁷ that the strong impurities are randomly distributed leading to a distribution of separations ℓ :

$$P(\ell) = (1/\bar{\ell}) e^{-\ell/\bar{\ell}} \quad (3)$$

where $\bar{\ell}$ is the mean separation between strong impurities. We take the weak impurities to be present in higher concentration, which we treat on the average. The depinning force is expected to be of the form:

$$F_\ell = F_0 + (\ell/\bar{\ell}) F_1 \quad (4)$$

where F_0 is the depinning force for strong impurities, which pin the CDW at the ends. The number of weak impurities on the CDW will be proportional to ℓ with F_1 the depinning force for a CDW of mean length $\bar{\ell}$. Writing for

the depinning electric fields:

$$E_0 = F_0/\bar{q} \quad E_1 = F_1/\bar{q} \quad E_\ell = F_\ell/q_\ell \quad (5)$$

with $q_\ell/\bar{q} = \ell/\bar{\ell}$ we obtain for (4):

$$E_\ell = (\bar{\ell}/\ell) E_0 + E_1 \quad (6)$$

Alternatively, at a given electric field E there is a critical CDW length ℓ_E such that CDW's longer than ℓ_E are depinned and CDW's shorter than ℓ_E remain pinned where from (6) we have:

$$\ell_E = [E_0/(E - E_1)] \bar{\ell} \quad (7)$$

Dc Conductivity

Concerning the velocity of depinned CDW's, we take the suggestion of Lee, Rice, and Anderson⁸ that the behavior of a CDW moving in the presence of impurities may be treated by dry (or stick-slip) frictional forces. Assuming that the forces of static and dry moving friction are equal, we write for the velocity of the moving CDW:

$$v_\ell = \mu(E - E_\ell) \quad (8)$$

Finally, we obtain for the current in the presence of an electric field E :

$$j(E) = \sigma_a E + \sigma_b \int_{\ell_E}^{\infty} (E - E_\ell) (\ell/\bar{\ell}) P(\ell) d\ell \quad (9)$$

where σ_a is the conductivity of the uncondensed carriers and $\sigma_b = ne\mu$ is a measure of the CDW conductivity where n is the concentration of condensed carriers. The integral in (9) is readily performed to yield as anticipated by Lee and Rice:

$$j(E) = \sigma_a E + \sigma_b (E - E_1) e^{-E_0/(E - E_1)} \quad (10)$$

which is just the form suggested by Fleming⁵ on empirical grounds. On this basis we take E_1 to be the threshold field E_T in (1). Since σ_b evidently just about makes up for the conductivity contribution lost from σ_a we conclude that the mobility μ of the CDW must approximately equal that of the precondensed carriers.

We conclude the discussion of dc conductivity based on the Lee-Rice model by comparing the observed values of E_0 and E_T with the estimates of Lee and Rice for strong and

weak pinning respectively. Minimum values of E_0 for NbSe_3 below the lower transition at $T_2 = 58$ K range from 100 mV/cm as observed by Ong and Monceau¹ down to about 10 mV/cm as reported by Fleming⁵ on evidently purer material. Lee and Rice⁴ estimate for the depinning field at strong impurities:

$$\lambda E_0 = (\Delta/e) b/\bar{\ell} \quad (11)$$

where 2Δ is the Peierls gap, λ is the CDW wavelength, b is the lattice constant, and $\bar{\ell}$ is the mean distance between impurities. Taking $\lambda = 13.2$ Å from x-ray scattering studies¹⁰ with $b = 3.478$ Å and estimating $\Delta = .009$ eV we obtain $E_0 \bar{\ell} \approx 2.3$ mV. On comparison with the measured values of E_0 we obtain for the mean separation between strong impurity centers $\bar{\ell} \approx .02$ to 0.2 cm. The measured threshold fields E_T are typically one-half E_0 although Fleming and Grimes⁵ initially reported values of the threshold field as small as one-quarter E_0 .

Lee and Rice estimate for the field for weak depinning:

$$\lambda E_1 = (\Delta/e) (\Delta/\epsilon_F) \times 10^4 n_1^2 \quad (12)$$

We take $2k_F = 2\pi/\lambda$ and $v_F = \hbar k_F/m = 2.8 \times 10^7$ cm/sec using the free electron mass. The Fermi energy is then $\epsilon_F = \hbar^2 k_F^2/2m = 0.22$ eV. In (12) n_1 is the fractional concentration of weak impurities. Using the same values as in the evaluation of (11) we obtain $E_1 \approx 4 \times 10^7 n_1^2$ suggesting a weak impurity concentration of approximately one in 10^5 . Although these estimates are approximate, they suggest the range in which we are working with very long CDW's and a somewhat higher concentration of weak than strong impurities.

Ac Conductivity

Continuing to apply the Lee-Rice model we now discuss the response of CDW's to an alternating electric field. We analyze the response of the CDW in terms of phase modes. The uniform mode ($n = 0$) is one in which the CDW phase is displaced more or less rigidly in the potential of weak and strong impurities. We have for the square of the resonance frequency of the uniform mode:

$$\omega_0^2 = k/M \quad (13)$$

where M is the CDW mass and k is the force constant.

We relate the force constant k to the electric field for depinning by writing

$$q E_{\ell} = A k \lambda / 2 \quad (14)$$

The quantity A is a numerical parameter expected to be less than one that characterizes the softening of the potential. For a sinusoidal potential, for example,¹¹ we have $A = 1/\pi$. Substituting into (13) we obtain:

$$\omega_0^2 = (2/\lambda A) (e/m^*) E_{\ell} \quad (15)$$

where m^* is the Frohlich mass and is about 10^3 times the electron mass.

For higher modes we take the ends to be pinned with the CDW an integral number n half waves long. The square of the mode frequency is

$$\omega_n^2 = (n\pi v/\ell)^2 + (2/\lambda A) (e/m^*) E_1 \quad (16)$$

where the first term represents propagation of a phase mode along the CDW and the second term arises from displacement in the potential of the weak impurities. The velocity v has been calculated by Fukuyama¹² and is given by

$$v = (m/m^*)^{\frac{1}{2}} v_F \quad (17)$$

Using typical values for NbSe_3 we obtain with $A = 1/\pi$

$$\omega_0^2 = 1.8 \times 10^{18} (1 + 2 \bar{\ell}/\ell) \quad (18)$$

or $\nu_0 = 210$ Mhz for $\ell \gg \bar{\ell}$. For the higher modes we estimate $v = 8.7 \times 10^5$ cm/sec to obtain

$$\omega_n^2 = 2.9 \times 10^{15} (n \bar{\ell}/\ell)^2 + 1.8 \times 10^{18} \quad (19)$$

where we have taken $\bar{\ell} = .05$ cm. Comparing (18) and (19) we see that the frequencies are dominated by the impurity potential. Because the first term in (19) is small we may largely neglect it and regard the CDW as displaced in the potential of the weak impurities only with the ends pinned. The equation of motion of the pinned CDW is then approximately

$$m^* d^2x/dt^2 + (m^*/\tau) dx/dt + m^* \omega_1^2 x = eE e^{-i\omega t} \quad (20)$$

with

$$\omega_1^2 = (2/\lambda A) (e/m^*) E_1 \quad (21)$$

Integrating over the CDW distribution we obtain for the conductivity

$$\sigma(\omega) = \sigma_a + \frac{\sigma_b}{1 + i(\tau/\omega)(\omega_1^2 - \omega^2)} \quad (22)$$

From (16) and the expression for the CDW conductivity

$$\sigma_b = n e \mu = n(e^2/m^*)\tau \quad (23)$$

we obtain

$$\omega_1^2 \tau = (2/\lambda A) \mu E_1 \quad (24)$$

Taking $6.7 \times 10^3 (\Omega\text{-cm})^{-1}$ for the CDW conductivity¹³ at 42K and Gruner et al's value¹³ of $\omega_1^2 = 2.5 \times 10^8$ as determined from the frequency dependence of the conductivity we obtain from (24) a mobility $\mu = 230 \text{ cm}^2/\text{V-sec}$ and from (23) a density of carriers condensed at T_2 of $n = 1.8 \times 10^{20}/\text{cm}^3$ (ten times Ong's value¹⁴ and about one-fifth Wilson's estimate¹⁵). Assuming a Frohlich mass 10^3 times the free mass we obtain from (23) for the CDW relaxation time $\tau = 1.3 \times 10^{-10} \text{ sec}$. With this value for the relaxation time and (19) we expect a CDW resonance with $Q = 0.18$. The frequency dependence of the conductivity of NbSe_3 has been studied by Gruner et al¹³ and by Longcor¹⁶ and has been fitted with an overdamped characteristic, consistent with this estimate.

Reduced Ac Conductivity

Longcor¹⁶ and Gruner et al¹⁷ have measured the ac conductivity of NbSe_3 as a function of frequency in the presence of dc fields up to E_0 in magnitude. The application of a dc field reduces the frequency-dependent component of the conductivity as expected since some of the CDW's are depinned and contribute to the frequency-independent conductivity. The dependence on frequency of the remaining pinned CDW's seems relatively little affected by the dc field. This observation supports our conclusion that the frequency of pinned CDW's depends very little on ℓ as indicated by (19).

Ac-Induced Dc Conductivity

Gruner et al¹⁸ have measured the dc conductivity of NbSe_3 in the presence of an ac field of variable frequency and amplitude. They find that the dc conductivity increases with

increasing ac amplitude although by a decreasing amount at higher frequency. At low frequency the threshold depinning field of a CDW is E_T as given by (6). At high frequency the displacement of the CDW is limited by inertia and damping. The depinning condition is displacement through a distance $\lambda/2$. Using (20) and neglecting the potential we obtain for the high frequency depinning field:

$$E_c = (\lambda\omega/2) (m^*/e) (\omega^2 + 1/\tau^2)^{1/2} \quad (25)$$

Using (21) we may write (25) as:

$$E_c = (\omega/\omega_1^2) (\omega^2 + 1/\tau^2)^{1/2} E_T/A \quad (26)$$

We connect the low and high frequency behavior with the phenomenological expression:

$$E_c(\omega) = E_T [1 + (\omega/A\omega_1^2)^2 (\omega^2 + 1/\tau^2)]^{1/2} \quad (27)$$

Gruner *et al*¹⁸ have fitted the threshold of ac-induced dc conductivity with (27) and obtain good agreement for $A\omega_1^2\tau \approx 0.45 \times 10^{-8}$. Taking $\omega_1^2\tau = 2.5 \times 10^8$ as determined from the frequency dependence of the conductivity¹³ these experiments indicate $A = 0.18$, a substantial softening of the harmonic potential.

TUNNELING THEORY OF CDW DEPINNING

Early experiments¹⁹ of nonlinear transport in NbSe₃ indicated a current of the form:

$$j = \sigma_a E + \sigma_b E e^{-E_0/E}$$

Such a form is suggestive of Zener tunneling across a gap. Tunneling of individual carriers across the Peierls gap 2Δ , however, yields values of the field E_0 far larger than observed. Bardeen has proposed⁶ that the observed current arises from the tunneling of entire CDW's across an impurity gap. To adapt the Zener theory to CDW tunneling it is useful to replace the electron charge e by an effective charge e^* with

$$e^*/e = m/m^* \approx 10^{-3} \quad (29)$$

The field E_0 is then given by

$$E_0 = \varepsilon_g^2 / 4 \hbar e^* v_F \quad (30)$$

where ε_g is the impurity gap.

The Zener theory assumes that the tunneling distance is short compared with the electron mean free path. With a finite correlation length L for CDW's we may expect tunneling only for $e^* E L > \epsilon_g$ leading to a threshold field

$$E_T = \epsilon_g / e^* L \quad (31)$$

As Bardeen argues, tunneling can occur only for $E > E_T$ and then only over a fraction $(1 - E_T/E)$ of L . So long as the CDW correlation length L is long compared with the CDW coherence distance

$$\xi_0 = 2 \hbar v_F / \pi \epsilon_g \quad (32)$$

we expect a current of the form

$$j = \sigma_a E + \sigma_b (E - E_T) e^{-E_0/E} \quad (33)$$

already given as (2). For L short compared with ξ_0 (30) must be modified to

$$E_0 = \epsilon_g / e^* L = E_T \quad (34)$$

and the current is characterized by a single parameter:

$$j = \sigma_a E + \sigma_b (E - E_T) e^{-E_T/E} \quad (35)$$

Taking the ratio of (30) and (31) and using (32) we obtain

$$E_0/E_T = L/2 \xi_0 \quad (36)$$

With E_0 typically twice E_T for NbSe_3 the CDW correlation length should be about four times the coherence distance and (33) should apply.

Taking 50 mV/cm for E_0 we estimate from (30) an impurity gap $\epsilon_g = 1.9 \times 10^{-6}$ eV. This value is less than one percent of the binding energy to strong impurities calculated by Lee and Rice.⁴ From the observed threshold field we obtain from (31) a correlation length $L = .02$ cm, which is about half the value of \bar{L} calculated from the Lee-Rice theory using obtained values of E_0 . On the other hand, for the same value of ϵ_g Lee and Rice obtain values of E_0 substantially lower than expected from tunneling.

CHARGE DENSITY WAVE NOISE

One of the most remarkable results of the study of CDW transport has been the observation of narrow components in the noise spectrum. Such components were discovered by

Fleming and Grimes⁵, who showed that they appeared above the threshold field E_T as a fundamental frequency and harmonics. The Q of these components is about forty. By increasing the current all components move to higher frequency at first slowly and then approximately linearly with current. Fleming and Grimes observed that even in the linear range the frequencies were surprisingly low. Associating the observed frequencies with the motion of CDW's across impurities we expect from (8)

$$v = p v_Q / \lambda = (p \mu / \lambda) (E - E_Q) \quad (37)$$

where p is an integer. Below the T_2 transition in $NbSe_3$ we expect from (37) $dv/dE \approx 1.7 \times 10^{-9} \text{ Hz/V-cm}^{-1}$. The observed sweep rate is lower by a factor of fifteen. Weger et al²⁰ have extended the measurements of Fleming and Grimes to higher frequency and to various temperatures with qualitatively similar results.

Monceau et al²¹ have measured the differential resistance while the sample carried a large ac current of variable frequency plus a dc current just above threshold. They found peaks in the differential resistance at frequencies corresponding to the narrow components in the noise. Ong and Gould²² have studied the upper transition in $NbSe_3$ at $T_1 = 144 \text{ K}$. They observe as at the T_2 transition that the sweep rate dv/dE is substantially lower than expected if the CDW wavelength λ is the characteristic distance.

Gruner et al¹¹ have used the CDW dc dielectric constant and the threshold field to obtain $n\lambda$, the product of the condensed carrier concentration and the CDW wavelength. From (22) we have for the dc dielectric constant:

$$\epsilon(0) = 1 + 4\pi n e^2 / m^* \omega_1^2 \quad (38)$$

Substituting from (21) for ω_1^2 we obtain:

$$\epsilon(0) = 1 + 2\pi n \lambda e A / E_T \quad (39)$$

With $A = 1/\pi$ for a sinusoidal potential a charge density of 2.0×10^{20} is obtained, equal to our value. From the sweep rate observed by Weger et al²⁰ a concentration of 3.6×10^{20} is indicated, about twice our value.

Gruner et al²³ have observed narrow noise components associated with CDW transport in TaS_3 . They obtain a sweep rate $dv/dI = 0.1 \text{ MHz}/\mu A$ with

$$dv/dI = v/\lambda I = 1/n e \lambda A \quad (40)$$

where $A = 3.3 \times 10^{-7} \text{ cm}^2$ is the sample cross-section. Substituting into (40) and taking²⁴ $\lambda = 4 a_0 = 13.3 \text{ \AA}$ Gruner et al²³ obtain $n = 1.4 \times 10^{21}/\text{cm}^3$. From galvanomagnetic

measurements Ong²⁵ has determined the room temperature concentration to be $1 \times 10^{22}/\text{cm}^3$. Assuming that all carriers condense out at the CDW transition at 215 K we have general agreement between (40) and the observed sweep rate.

The observed narrow noise components would not be expected on the basis of a model in which the CDW's are depinned and move independently and where the noise is generated by all moving CDW's. Although the tunneling mode ℓ appears more attractive in that all CDW's are expected to tunnel with the same probability and thus give the same mean current, even here there are problems. The current from a single CDW is not uniform in time and there must be strong coupling between CDW's if the current is to be smoothed. In the following section we suggest an alternative mechanism for the origin of noise components that is compatible with independently depinned CDW's.

Origin of Narrow Noise Components

All investigators who have observed narrow noise components in CDW transport have associated them with the motion of CDW's past impurities. There are two problems with this explanation. First, the rate of frequency sweep with field is too low. The second problem is the sharpness of the observed components, which have a Q as high as forty. We propose here a mechanism for the origin of narrow noise components that may avoid both these problems.

We take a CDW sliding with velocity v_ℓ to generate noise components at frequencies:

$$\nu_p = p v_\ell / \lambda = (p \mu / \lambda) (E - E_\ell) \quad (41)$$

as given by (37) with E_ℓ given by (6) in our model of non-linear dc transport. What is new is that we require for a frequency component to be detected that it be resonant with a phase mode of the sliding CDW with

$$\nu_n = n v / 2 \ell \quad (42)$$

as given by (16) except that we assume that the weak impurity potential is averaged out by sliding. Equating (41) and (42) we find for the length ℓ of resonant CDW's:

$$\ell = \frac{n \lambda v / 2 + p \mu E_0 \bar{\ell}}{p \mu (E - E_T)} \quad (43)$$

The resonant frequencies are given by

$$v = n v/2 \ell = \frac{n p \mu (E - E_T) v}{n \lambda v + 2 p \mu E_0 \bar{\ell}} \quad (44)$$

If the first term in the denominator were dominant we would obtain $v = p\mu(E - E_T)/\lambda$ which is the standard result (41). However, taking $v = 9 \times 10^5$ cm/sec from (17) and $\lambda = 14 \text{ \AA}$ we obtain for the first term in the denominator .12 cm²/sec. For the second term in the denominator we take for NbSe₃ $\mu = 230$ cm²/V-sec, $E_0 = 50$ mV/cm and $\bar{\ell} = .05$ cm to obtain 1.2 cm²/sec, a factor of ten larger than the first term. This gives a sweep rate one tenth what would be obtained if the first term in the denominator were dominant. In this limit we obtain for (44)

$$v = (n v/2 \bar{\ell})(E - E_T)/2E_0 \quad (45)$$

independent of μ and in improved agreement with the measured sweep rates. The reason for the reduced sweep rate is that the resonant CDW's are always just above threshold. From (43) we write using (7):

$$\ell = (1 + n \lambda v/2 p \mu E_0 \bar{\ell}) \ell_E \quad (46)$$

which is ten percent above threshold with a velocity ten percent of μE as indicated by (8).

Accounting for the extreme narrowness of the observed spectral components must be somewhat speculative. We expect for the phase modes a relaxation time $\tau = 1.3 \times 10^{-10}$ or shorter as estimated earlier. This translates to a line width $\Delta v = 1.2$ GHz, orders of magnitude larger than what is observed. We believe that the explanation of the narrow observed spectral components lies in the observations of Monceau et al²¹, who showed that when a sample is driven at the frequency of one of the spectral components the dc differential resistivity is substantially increased. This observation suggests a positive feedback mechanism in which the excitation of a phase mode increases energy transfer to the mode, thus narrowing the line. We treat the process phenomenologically by writing a differential equation²⁶ for the excitation of a phase mode:

$$d^2 \phi/dt^2 + (1/\tau) d\phi/dt - v^2 d^2 \phi/dx^2 + \omega_1^2 \sin \phi = (2\pi/\lambda m^*) F_\ell \quad (47)$$

where ϕ is the phase, v is the phase mode velocity (17) and

$$F_\ell = e(E - E_\ell) + f_p e^{-i\omega t} - (\lambda m^* v/2\pi\tau) d\phi/dx \quad (48)$$

is the force with which phase is driven along the CDW by an applied electric field E . The second term in (48) arises from driving the CDW over impurities with

$$\omega = (2 \pi p / \lambda) v_{\ell} \quad (49)$$

which is essentially the same as (37). The third term is strictly phenomenological and has been constructed in such a way as to produce the required coupling between the electric field E and the phase mode. For constant F_{ℓ} (47) yields for the depinned CDW

$$\phi = (2 \pi / \lambda) v_{\ell} t \quad (50)$$

which gives the usual CDW contribution to the dc current. Allowing for excitation by the second term in (48) and assuming a phase mode of the form

$$\phi(x, t) = \phi e^{i(\omega_n x/v - \omega t)} \quad (51)$$

with

$$\omega_n = n \pi v / \ell \quad (52)$$

which is (42) we obtain

$$\phi = \frac{(2\pi\tau/\lambda m^*) f_{pn}}{(\omega_n^2 - \omega^2)\tau - i(\omega - \omega_n)} \quad (53)$$

where f_{pn} is the appropriate Fourier component of f_p . Note that the resonance in ϕ has a singularity at $\omega = \omega_n$. Thus a coupling of the form of (48) can produce the unusually narrow resonance lines that are observed.

We may also see from the form of (48), at least in a phenomenological way, how CDW motion may be blocked. We see that an advance of phase alone the CDW produces a force that reduces v_{ℓ} . Under resonance conditions we may write from (48):

$$v_{\ell} = \omega_n \lambda / 2 \pi p - (\omega \lambda \ell / 2 \pi^2 n) \overline{d\phi/dx} \quad (54)$$

For $v_{\ell} = 0$ we require

$$\overline{d\phi/dx} = (n/p) \pi / \ell \quad (55)$$

so that an additional phase advance of $(n/p)\pi$ along the CDW is sufficient to inhibit CDW drift, leading to the increase in differential resistivity observed by Monceau *et al.*²¹ Such an advance is suggestive of the excitation of solitons on the CDW.

CONCLUSIONS

We have discussed the Lee-Rice model of electric field depinning and the Bardeen adaptation of tunneling theory to CDW transport. Either theory seems capable of accounting for the observed conductivity.

The observed narrow components in the noise are suggested to arise from CDW phase modes. The fact that the observed frequencies are a factor of ten lower than expected from the imposed field does appear to require CDW's that at all field values are traveling substantially slower than the CDW mobility would indicate. Although we have suggested a mechanism of line-narrowing, it is clear that further investigation is required, particularly in the nonlinear regime.

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