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Charge-density-wave transport under pulsed electric fields in $NbSe_3$: Step structure in sliding distance

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The oscillatory response of a charge-density wave (CDW) to pulsed electric fields was investigated for various values of pulse height and pulse width in the q_2-CDW regime of $NbSe_3$. It was found that the number of oscillations observed within a pulse is always integral, regardless of pulse height or pulse width, and the CDW slides as a whole by its wavelength within each period of the oscillation. When the pulse width is increased in the pulse train, the number of oscillations observed within a pulse first becomes fractional, and then becomes integral again after about ten pulses with the new width. It was also found from snapshots of pulse responses that the virgin CDW, cooled down from the q_2-CDW transition temperature without any field, exhibits no oscillatory behavior under the first pulse, while it grows gradually under following pulses. After about 20 pulses, the pulse response of the CDW becomes typical with an integral number of oscillations. The results are discussed in terms of phase slip at strong pinning centers.

I. INTRODUCTION

In the charge-density-wave (CDW) state of many quasi-one-dimensional conductors, the large conductivity due to sliding CDW is observed above a well-defined threshold field E_T.[1-8] One of the most intriguing phenomena associated with the sliding CDW is the appearance of narrow-band noise (NBN) with many higher-harmonic components above E_T.[2,6-8] Its frequency is proportional to the current carried by the sliding CDW.[9] Under constant-current pulses, the narrow-band noise can be observed easily in the time domain as an oscillation of voltage developing across a sample.[10,11] The appearance of the narrow-band noise has been considered as a manifestation of coherent CDW motion over the macroscopic region of a crystal.

Recently an interesting response of the CDW to pulsed field was found in the crystal of $NbSe_3$ whose noise spectrum under dc bias exhibits only one fundamental component of the NBN with a sharp profile.[12,13] For repetitive pulsed fields just above E_T, the number of oscillations m observed within a pulse width is always integral regardless of pulse width and changes by one each time pulse width is changed by a constant value. Furthermore, when pulse width is changed within the range where m is constant, the CDW velocity varies so that the CDW displacement L within a pulse width may remain constant. At the pulse width where m changes by one, L jumps by the q_2-CDW wavelength λ. Such a steplike dependence of L on pulse width means that the CDW in a crystal is able to slide in a body over the whole crystal under repetitive pulsed fields. A similar pulse-width dependence of CDW displacement has been proposed to account for the pulse duration memory effect observed in blue bronze $K_{0.3}MoO_3$ (Refs. 14 and 15) and discussed in terms of mode locking. It is well known that mode locking occurs under a combined ac-dc field and many investigations on the mode locking have been carried out by now.[16-18] Because repetitive pulsed fields can be thought of as a combined ac-dc field, mode locking may occur in pulsed experiments. Mode locking under pulsed fields has been demonstrated in numerical simulations based on the Fukuyama-Lee-Rice model.[19,20] The numerical simulations have been performed for the CDW system with the impurities which cause weak pinning of the CDW.[21,22] The sliding distance of the CDW takes just an integral multiple of the CDW wavelength $n\lambda$ when the relaxation process to a pinned state after a pulse is taken into account. However, there exists a serious difference between the result of the pulsed experiment on $NbSe_3$ and that of the numerical simulation. In the experiment the relaxation current after a pulse, which appears appreciably in the simulation, is not observed at all. In the experiment, the CDW velocity depends largely on pulse width and sliding distance always takes $n\lambda$ just within a pulse, as mentioned above. In previous numerical simulations such a dependence of CDW velocity on pulse width has not been reported. Quite recently, a discernible dependence of the CDW velocity on pulse width has been demonstrated in the numerical simulation based on the Fukuyama-Lee-Rice model.[23] When pulse width is increased (decreased) within the range of widths where the CDW displacement takes the value of $n\lambda$ after a pulse, the CDW displacement within the first pulse with new width takes a larger (smaller) value than within a preceding pulse. The phase configuration of the pinned state after the first pulse gets advanced (delayed) parts in comparison with the old phase configuration. It was shown in the recent numerical simulation[23] that such advanced (delayed) parts in the phase configuration reduce (enhance) CDW velocity under the next pulse. As the following pulses are applied, the advanced (delayed) parts grow and then the CDW velocity becomes smaller (larger). Such a process repeatedly occurs under each pulse until the CDW drops into the same pinned state as the preceding one after removing a pulse. In this numerical simulation, however, the CDW velocity does not change so remarkably as the experiment and appreciable relaxation current also appears after a pulse.

In the present study, the CDW response to repetitive pulsed fields just above E_T is further investigated in the q_2-CDW regime (below $T_2=58$ K) of $NbSe_3$. The CDW displacement L within a pulse is confirmed to exhibit a steplike dependence on pulse height, too. It is found from a snapshot of pulse response that when pulse width is increased after a certain pulse in a pulse train, the CDW velocity decreases by degrees under following pulses. After about ten pulses with the new width the CDW velocity takes the new preferred value with which the CDW slides by just an integral multiple of λ within a pulse. It is also found that the virgin CDW, cooled down from a temperature $T>T_2$ without any field, exhibits no oscillatory behavior under the initial pulse while it appears gradually under the following pulses. After about 20 pulses, pulse response of the CDW becomes typical with an integral number m of oscillations and L takes the value of $m\lambda$. On the basis of the present results, the sliding manner of the CDW is discussed in terms of phase slip at strong pinning centers.

II. EXPERIMENTS

Crystals of $NbSe_3$ were synthesized by reacting the elements in an evacuated quartz tube at 750 °C for ten days. The conductivity measurements were performed on single crystals with thickness less than 1 μm by using the four-probe method. Electrical contacts with a strip shape were made by evaporating gold on the surface of a crystal. Through preparation of the contacts, considerable care was taken in order to make equipotential surfaces around the contacts homogeneous. Such preparation of the contacts, together with preparation of very thin single crystals, is very important for observation of noise spectra exhibiting only one fundamental component of the NBN with a sharp profile. In the measurements under repetitive pulses voltages developing across the sample and across a standard resistor were amplified with differential amplifiers and fed to channels A and B of a digital boxcar integrator. The conductivity was calculated by operating the B/A mode of the boxcar integrator. A snapshot of voltage developing across the sample under constant-current pulses was taken by using a storage oscilloscope with a function of digitizing the signal displayed on a CRT.

III. RESULTS AND DISCUSSIONS

A. CDW response to repetitive pulsed fields

Pulsed voltages developing across the sample and across the standard resistor are shown in Fig. 1, which displays measurements made by changing pulse height but using the same pulse width. Large oscillation, namely, the narrow-band noise, is observed in the sample voltage when the current voltage is nearly constant. It is no-

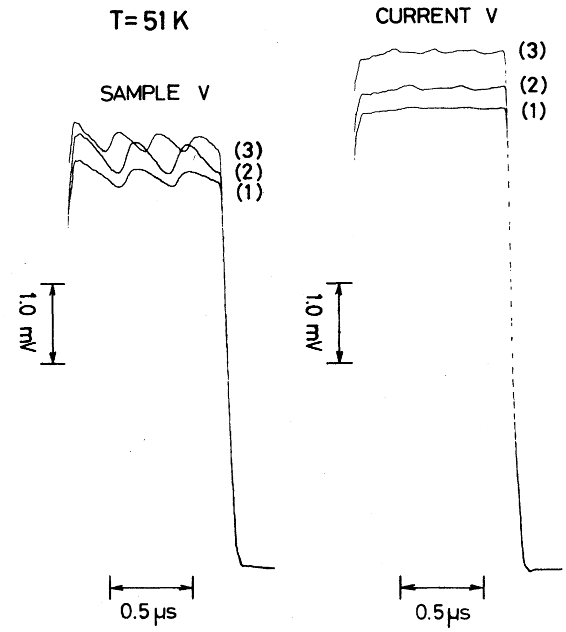

FIG. 1. Pulsed voltages developing across the sample (sample V) and across the standard resistor (current V) with different pulse heights but the same pulse width. Pulse height increases from (1) to (3). The threshold voltage V_T of the sample is 3.5 mV at $T=51$ K. Value of the standard resistor is 50.7 Ω.

ticed that the oscillation always starts from its top (the conductivity minimum) and always ceases at its bottom (the conductivity maximum) regardless of pulse height. Therefore the number of oscillations contained within a pulse width is always integral, counting the part of the initial descent as one. As pulse height is increased, the number of oscillations m observed within a pulse width will increase by one at a certain height. Similar results have been observed in the experiment in which pulse width is changed but the pulse height is kept the same, as shown in the inset of Fig. 2. In Fig. 3 the CDW currents $J_{\rm CDW}(t)$ are shown for different pulsed voltages. $J_{\rm CDW}(t)$ is obtained here by subtracting the single-particle current $\sigma_0 E(t)$ from the total current $J_{\rm tot}(t)$ measured,

$$J_{\rm CDW}(t)=J_{\rm tot}(t)-\sigma_0 E(t)\ . \tag{1}$$

It should be noticed that the CDW current averaged over the pulse width, namely, the averaged CDW velocity, remains constant regardless of pulsed voltages, as long as m is constant. This result shows that mobility of the CDW depends on pulse height, just as it does on pulse width in the case of changing pulse width.

The displacement of the CDW L within a pulsed field with a given width T can be calculated as

$$L=\int_0^T V_{\rm CDW}(t)dt=\frac{1}{eN}\int_0^T J_{\rm CDW}(t)dt\ , \tag{2}$$

where $V_{\rm CDW}(t)$ and N are CDW drift velocity and number of single carriers condensed into the CDW, respectively. As shown in Fig. 4, even if pulse height is varied

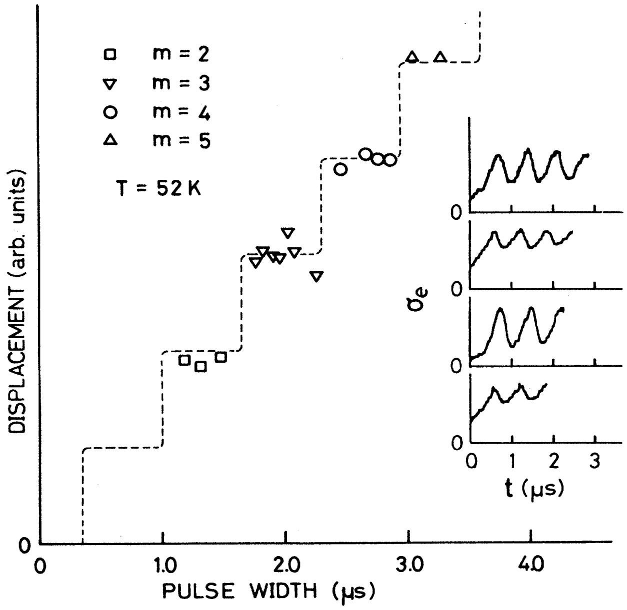

FIG. 2. Displacement of the CDW within a pulsed field as a function of the pulse width (arbitrary unit). The dotted line is a guide for the eye. Inset: Dependence of the extra conductivity due to sliding CDW on the pulse width.

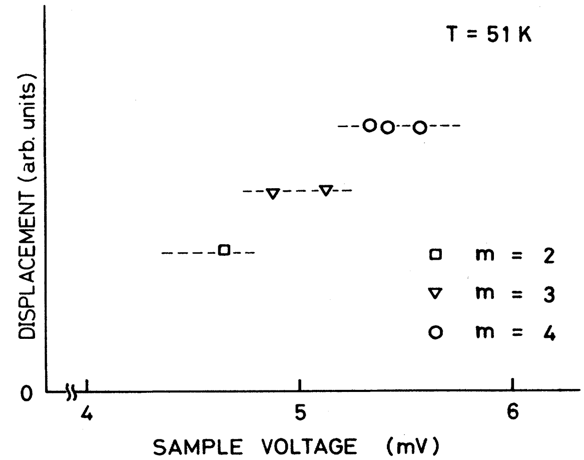

FIG. 4. CDW displacement within a pulse with a given width $T=0.9$ μs as a function of the sample voltage averaged over the pulse width (arbitrary unit). The dotted lines are guides for the eye.

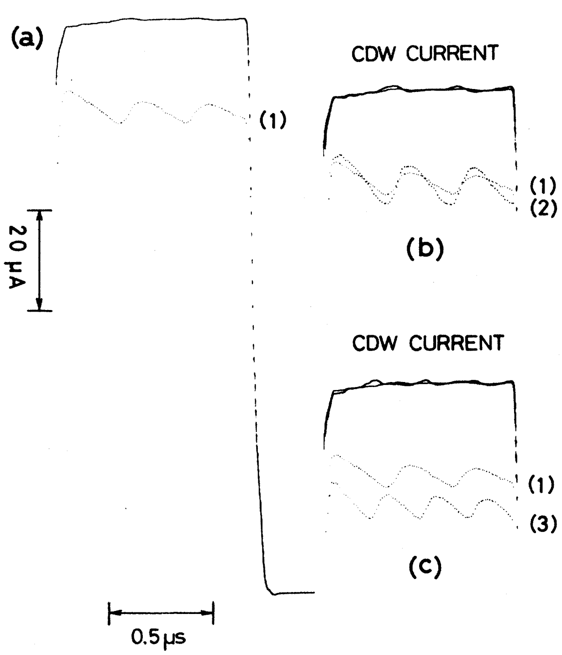

FIG. 3. (a) The total current measured (the solid line) and the single particle current (the dotted line) for the pulsed voltage indicated by (1) in Fig. 1. The difference between them corresponds to CDW current $J_{\mathrm{CDW}}(t)$. (b),(c) Comparison between CDW currents for pulsed voltages with different heights shown in Fig. 1.

within a range, the displacement of the CDW L remains constant as long as the number of oscillations observed within a pulse width is constant. It steps up by a constant value L_0 each time m increases by one with increasing pulse height. Furthermore, sliding distance within any one period of the oscillation always takes the constant value L_0. Such results are the same as those observed in the case of changing pulse width (Fig. 2). To calculate L_0, we assume that the number of single carriers condensed into the q_2-CDW is approximately half of that in the normal state.[24] The latter has been estimated in the band calculation[25,26] as 4×10^{21}. These values give $L_0\approx 12$ Å for the present sample, and $L_0\approx 10$ Å for the sample in Ref. 12. These values are of the same order with the wavelength (≈ 13.2 Å) of the q_2-CDW. This result therefore means that the CDW displaces as a whole by its wavelength λ within each period of the oscillation observed in sample voltage.

As mentioned above, the oscillation always ceases at its bottom (the conductivity maximum) regardless of pulse width or pulse height. Such a phenomenon is, of course, not observed in snapshots of pulse responses taken just after pulse width or pulse height is changed. As shown in Fig. 5, the oscillation no longer ceases at its bottom under the first pulse ($N=1$) after increasing pulse width. Its period and amplitude are, of course, equal to those observed under the preceding pulse. Under following pulses ($N\geq 2$) period and amplitude of oscillation increase by degrees and the oscillation ceases at its bottom again after about ten pulses with the new width. The CDW exhibits the same pulse response repeatedly for further pulses with the new width. It is considered that the phase configuration of the metastable pinned state into which the CDW drops after removing a pulsed field influences CDW response to the next pulse. Present results therefore suggest that the CDW drops into a metastable pinned state different from the preceding one before the tenth pulse while after the tenth pulse the CDW

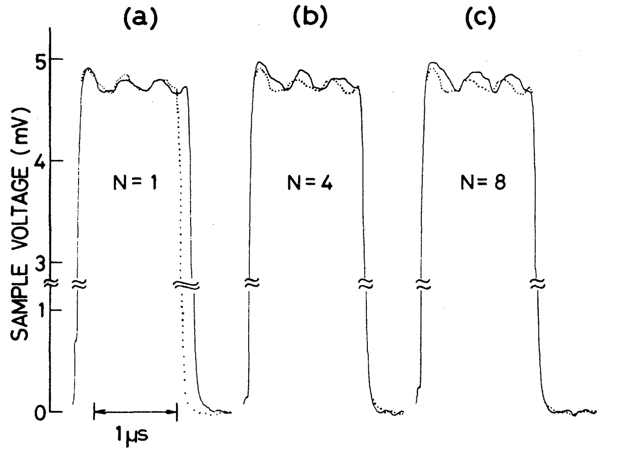

FIG. 5. (a) Comparison between snapshots of the CDW response to $N=1$ pulse (solid line) and to a pulse with the old width (dotted line). (b),(c) Comparison between the CDW response to $N=4$ ($N=8$) (solid line) and $N=1$ (dotted line). The measurements are carried out by using nearly constant current pulse.

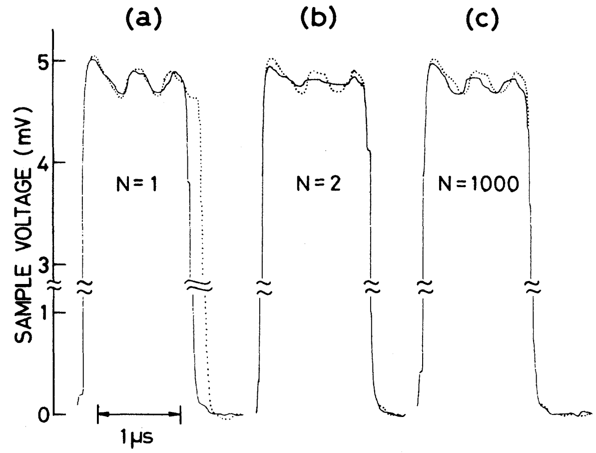

FIG. 6. Comparison between snapshots of the CDW response to the $N=1$ pulse (solid line) and to a pulse with the old width (dotted line). (b),(c) Comparison between the CDW response to $N=2$ ($N=1000$) (solid line) and $N=1$ (dotted line).

drops into the same metastable pinned state. The CDW should slide by just $n\lambda$ at each point of the sample under a pulsed field in order to drop into the same pinned state as the preceding one. Once the CDW drops into such a metastable pinned state after a pulsed field, it drops always into the same one after the following pulses. In fact, in the present experiments the CDW always slides by just $n\lambda$ within a pulse sufficiently after pulse width was varied, as shown in Fig. 2. It is stressed here that after pulse width is increased in a train of pulses, the CDW experiences only ten pulses with new width before the CDW drops into the same pinned state as the preceding one, as shown in Fig. 5. This fact seems to indicate that after increasing pulse width the pinned metastable state evolves straight to one whose phase configuration gives a sliding distance of just $n\lambda$ for a pulse with new width.

In Fig. 6, snapshots of CDW responses taken after pulse width was decreased are shown. It is noticed that under the second pulse ($N=2$) after decreasing, oscillation becomes rather indistinct, which means that CDW velocity becomes inhomogeneous over the crystal. Such a phenomenon is not observed in the case of increasing pulse width. However, under following pulses ($N\geq 3$) the oscillation grows gradually and becomes again the typical one ceasing at its bottom after 200 or 300 pulses, which is much later than the case of increasing pulse width.

B. Pulse response of the virgin CDW

Sequential responses of the virgin CDW, cooled down from a temperature $T > T_2$ without any field, to a train of constant-current pulses are shown in Fig. 7. It is noted first that no oscillatory behavior is observed under the initial pulse ($N=1$). Under the second pulse CDW velocity becomes slightly smaller, while oscillatory behavior is still not observed. Oscillatory behavior appears gradually after several pulses have been applied. Sufficiently after the initial pulse the oscillation becomes large and ceases at its bottom, as observed in the response to the $N=1000$ pulse shown in Fig. 7. It should be noticed that the voltage at the bottom of the oscillation observed under the $N=1000$ pulse is comparable to the voltage observed under the initial one. (To be precise, the former is slightly smaller than the latter.) Since pulsed currents used are constant, this fact shows that the maximum drift velocity

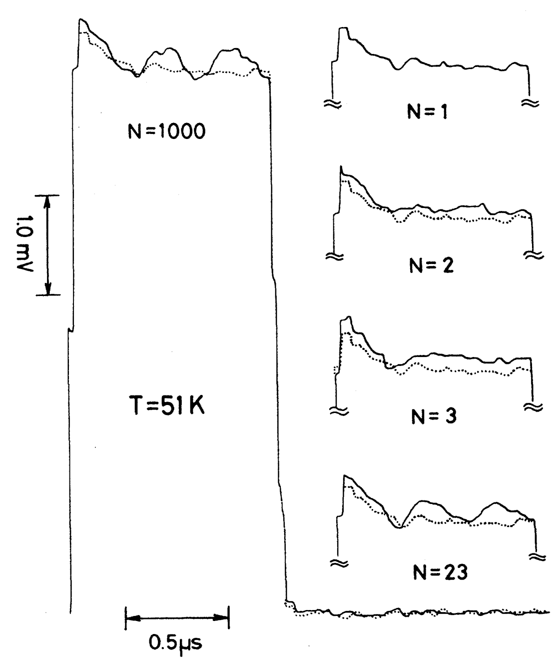

FIG. 7. Sequential responses of the virgin CDW to a train of constant current pulses (solid lines). The response to the initial pulse is shown for comparison by dotted lines.

of the CDW under the $N=1000$ pulse is almost the same as the velocity under the initial one.

On the basis of the Fukuyama-Lee-Rice model, CDW motion under a train of pulsed fields above E_T has been numerically examined on the CDW system which has experienced no electric field.[22] The CDW system treated in the numerical simulation has weak pinning centers. Under the initial pulse of the pulse train, local velocity of the CDW is rather different among different regions of the CDW system and current oscillations are canceled out due to interference over the whole CDW system. On the other hand, sufficiently after the initial pulse, CDW velocity becomes fairly homogeneous over the whole CDW system, which is due to elastic strains stored between regions with different local velocities, and so well-defined current oscillation appears. Such features in the numerical simulations are in agreement with those of the oscillation in the present results for the virgin CDW. The numerical simulations, however, do not reproduce the experimental result that the CDW velocity differs sufficiently after the initial pulse from that under the initial pulse. The CDW drift velocity averaged over a pulse is always constant in the numerical simulations.

It has been suggested from doping experiments that strong pinning centers play an important role in CDW sliding of nondoped $NbSe_3$.[27,28] In a real CDW system there will exist various strong pinning centers with different strengths. Under a train of narrow pulses just above E_T, the CDW including strong pinning centers with various strengths is expected to slide as follows. When the initial pulse is applied to the virgin state of the CDW, CDW phase at strong pinning centers will remain pinned down for sufficiently narrow pulse. However, parts of the CDW, except for small regions surrounding the pinned centers, will be able to displace rather freely because of deformability of the CDW. In such a case, if the number of the pinned centers is small, fairly large CDW current is expected to be observed under the initial pulse. On the other hand, around the pinned centers the CDW will be compressed (stretched) during the initial pulse. The CDW distortions around the pinned centers will grow during the second pulse as long as the CDW phase at the pinned center remains pinned down. The CDW will no longer be able to displace freely over such distorted regions under electric field. Therefore the CDW current observed under the second (or third) pulse is expected to be smaller than that under the initial (or second) pulse, as observed in the present experiment. As following pulses are applied, the phase slip will occur in sequence at the pinned center around which strain energy due to CDW distortion attains a critical value. Under pulses sufficiently after the initial one phase slips will occur repeatedly at each pinned center and the CDW slides as a whole. Phenomenologically it appears in the CDW sliding sufficiently after the initial pulse that the phase slips occur collectively to some extent among the whole pinned centers rather than independently, as illustrated schematically in Fig. 8(a). In this case, large oscillatory behavior will appear in the pulse response. Especially at the times indicated by arrows in Fig. 8(a), CDW phase has just slipped at a great many pinned centers and fairly large parts of the CDW will be moving. Such a situation is similar to that of CDW sliding under the initial pulse, and so the CDW drift velocity is expected to become comparable to the velocity under the initial pulse, just as observed. When pulse width is increased, the minimum value of the CDW drift velocity becomes small while the maximum one changes only slightly, as shown in the inset of Fig. 2. On the basis of the present proposition, such a pulse-width dependence of the CDW drift velocity suggests that phase slips may become more collective as pulse width is increased, as shown schematically in Fig. 8(b).

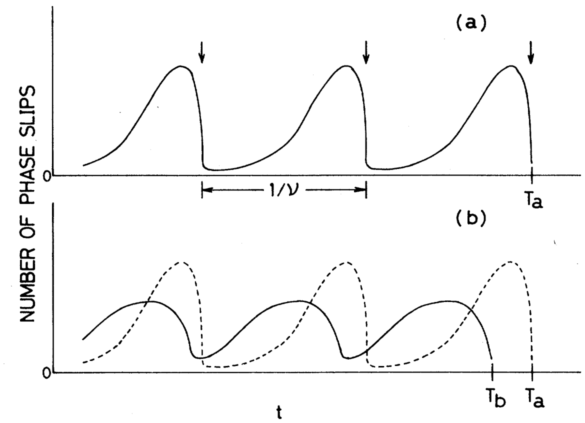

FIG. 8. (a) Number of phase slipping events as a function of time within a pulse of width T_a. Phase slip occurs once at each pinned center within a period $1/\nu$, corresponding to that of the oscillation observed in the pulsed voltage developing across the sample. At the time indicated by an arrow CDW phase has slipped at a great many of the pinned centers within a period. (b) Number of phase slipping events as a function of time for different pulse widths T_a and T_b $(T_a > T_b)$.

IV. SUMMARY

It was confirmed that for repetitive pulsed fields just above E_T the CDW displacement within a pulse always takes just integral multiples of the CDW wavelength $n\lambda$ regardless of pulse width or pulse height. The pulse responses of the virgin CDW indicate that the CDW slides through phase slips at strong pinning centers. Sufficiently after the initial pulse phase slips appear to occur collectively among the whole pinned centers rather than independently. Phase slips may become more collective with decrease of CDW velocity when pulse width is increased within a range of widths where the CDW displacement within a pulse remains constant. Such a redistribution of phase slips for varying pulse width will relate closely to the fact that the CDW displacement within a pulse is restricted to integral multiples of the CDW wavelength. However, it is not sufficiently explained at present why the CDW displacement within a pulse is always restricted to integral multiples of the CDW wavelength regardless of pulse width or pulse height. It is strongly hoped that the sliding manner of the CDW with strong pinning centers with various strengths will be examined by numerical simulations, which may give a clue to clarify the origin of such a restriction on CDW sliding under repetitive pulses just above E_T.

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