SERIALLY ARRANGED MODE-LOCKED CHARGE DENSITY WAVE DOMAINS IN NbSe₃

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ABSTRACT

We examine the local Shapiro step spectrum produced by NbSe₃ crystals that exhibit multiple narrow band noise (NBN) fundamentals either intrinsically or due to applied temperature gradients. In either case, differential resistance traces reveal multiple and co-existing families of Shapiro step spectra. Local dV/dI measurements show that each set of Shapiro steps stems from charge density wave (CDW) mode locking in unique sample sections, indicating that in NbSe₃ the CDW breaks into serially arranged velocity-coherent domains in samples with multiple NBN fundamentals.

The two independent charge density waves (CDWs) that form in NbSe₃ can be forced to slide by directing an electric field exceeding a depinning threshold field $F_T$ along the CDW chain axis.¹ The sliding CDW carries both a dc current and a small amplitude ac "narrow band noise" (NBN) signal characterized by a frequency $f_{NBN}$.² $f_{NBN}$ is linearly related to both the CDW velocity and current.³ When driven by combined ac and dc electric fields, a depinned CDW can be mode-locked to the external ac signal.⁴ When mode-locked, Shapiro steps form in the sample I-V characteristic and peaks appear in the differential resistance over an appreciable range of dc bias. The Shapiro steps occur whenever the externally applied frequency $f_{ex}$ is related to the internal NBN frequency by $nf_{ex} = n f_{NBN}$,⁵ where $n$ (the Shapiro step order) is a ratio of two integers.

Research investigating both the NBN and mode-locking phenomena in NbSe₃ has centered predominately on an examination of those samples which display a single NBN fundamental. In many samples, though, multiple NBN
fundamentals are evident in the NBN spectrum. Additionally, NbSe₃ samples which normally exhibit a single NBN fundamental can be made to display two or more NBN fundamentals by applying a longitudinal temperature gradient to them. This temperature gradient splitting of a NBN peak results from the strong temperature dependence of the ratio $I_{CDW}/I_{NBN}$ in the lower CDW state of NbSe₃.

We have used a non-perturbing voltage probe technique to examine the local Shapiro step spectrum produced by NbSe₃ crystals that display more than one NBN fundamental. Regardless of whether a sample exhibits multiple NBN fundamentals intrinsically or due to the application of a thermal gradient, dV/dI measurements on these samples show multiple and coexisting families of Shapiro step spectra. By measuring dV/dI traces in different sample segments, we find that each set of Shapiro steps can be attributed to CDW mode-locking in a unique portion of the crystal, indicating that in samples with more than one NBN fundamental, the sliding CDW breaks into independent, serially arranged velocity-coherent domains.

We first examine the Shapiro step spectrum of an NbSe₃ sample (sample 1) whose NBN spectrum intrinsically (ΔT=0) contains two fundamental peaks. The three-terminal contact arrangement employed to examine sample 1 is shown in Fig. 1. The dV/dI Shapiro step traces obtained from this contact configuration are presented in Fig. 2. The measurement made across the entire sample (trace 1-3) shows the presence of two n=1 Shapiro steps near $I_{bias} = \pm 100$ μA. The four pairs of dV/dI vs. $I_{bias}$ traces shown in Fig. 2 demonstrate that the sample is composed of two serially arranged mode-locked CDW domains, denoted α and β in Figs. 1 and 2. This is most clearly seen in the traces made with contact 2 at position b' only the dV/dI trace in section 1-2 contains the $\pm 90$ μA Shapiro step, while only the trace in section 2-3 shows the $\pm 10$ μA Shapiro step. Hence, position b resides at the interface between two mode-locked domains, with domain α locking at $\pm 90$ μA and domain β locking at $\pm 10$ μA. The traces made with probe 2 at positions a, c, and d are consistent with this domain configuration. With contact 2 at position c, for example, both Shapiro steps appear in trace 1-2, while only the $\pm 10$ μA step appears in trace 2-3.

We next turn to an examination of the Shapiro step spectrum of an NbSe₃ sample (sample 2) whose NBN spectrum contains three fundamental peaks for $I_{bias} > 0$ when subjected to a temperature gradient of 6K. The three-terminal contact arrangement used to examine sample 2 is shown in Fig. 3. The dV/dI traces obtained from this contact configuration are presented in Fig. 4. The top trace, made across the entire sample with no applied gradient
Fig. 1: Three-terminal contact arrangement used to examine sample 1's Shapiro step spectrum. The four measurement positions for the movable probe (contact 2) are denoted a, b, c, and d. The data in Fig. 2 indicate that sample 1 is composed of two domains, denoted \( \alpha \) and \( \beta \).

Fig. 2: Local Shapiro step spectrum for NbSe\(_3\) sample 1; \( V_{sh} = 5 \) mV, \( f_{sh} = 1 \) MHz. The two numbers to the right of each trace indicate between which two contacts the response measurement was made (1-3, 1-2, or 2-3). The letter to the left of the trace (a, b, c, and d) specifies the position of probe 2 (see Fig. 1). The dashed lines are guides to the eye and denote the position of the \( n+1 \) Shapiro steps for the two CDW domains (\( \alpha \) and \( \beta \)).
Fig. 3: Three-terminal contact arrangement used to examine sample 2's Shapiro step spectrum in a temperature gradient. The temperature of contact 3 was held fixed at $T_0=48K$ while contact 1 was warmed to $T_0+\Delta T$. The three measurement positions for the movable probe (contact 2) are denoted a, b, and c. The data in Fig. 4 indicate that sample 2 is composed of three domains (α, β, and γ) for $I_{\text{bias}}>0$ and $\Delta T=6K$.

Fig. 4: Local Shapiro step spectrum for NbSe$_3$ sample 2 in a temperature gradient; $V_f=5$ mV, $f_{\text{ex}}=1$ MHz. $\Delta T=0$ for the top trace and $\Delta T=6K$ for all other traces. The arrows indicate the progression of the n=1 Shapiro steps when the $\Delta T$ is applied. The two numbers to the right of each trace indicate which contacts the measurement was made. The letter to the left of each trace (a, b, or c) specifies the position of probe 2 (see Fig. 3). The dashed lines are guides to the eye and denote the position of the n=1 Shapiro steps for the three CDW domains with $I_{\text{bias}}>0$ (α, β, and γ), and the single domain (α') for $I_{\text{bias}}<0$. 
shows a clear single-domain Shapiro step spectrum with large n-1 and n-1/2 peaks for both positive and negative biases. When a 6K gradient is applied to the sample, the interference peaks move to lower current biases because Tc decreases with increasing temperature below ~53K. Additionally, the n-1 step for $T_{bias} > 0$ splits into three separate peaks; no splitting occurs for $T_{bias} < 0$. Similar ΔT results have been previously reported in other NbSe$_3$ samples.

The three sets of dV/dI traces shown in Fig. 4 clearly suggest that for $T_{bias} > 0$, the sample consists of three independent, serially arranged mode-locked CDW domains, denoted α, β, and γ in Figs. 3 and 4; for $T_{bias} < 0$ the sample is composed of a single domain, denoted by α' in the figures. This is most evident in the traces made with contact 2 at position b; only the minimum-current Shapiro step appears in trace 1-2, while the other two steps appear only in trace 2-3. This indicates that position b resides at the interface between two domains (α and β). The other interface (between β and γ) can be roughly located via the traces made with contact 2 in position c. Here, the minimum-current Shapiro step and a portion of the medium-current step appear in trace 1-2, while trace 2-3 shows evidence only of the medium- and high-current steps. From this it can be inferred that position c roughly divides the domain responsible for the medium-current Shapiro step (domain β) in half. These traces are thus consistent with the domain configuration depicted in Fig. 3. The spatial ordering of the domains and the relative bias currents at which they mode-lock are also consistent with the fact that $V_{CDW}(I_{bias})$ is an increasing function of temperature above $T_c$ = 46K. Hence, mode-locking should (and does) occur first in the hot domain (α), then in domain β, and lastly in the cold domain (γ).

The observation of mode-locked domain formation in the intrinsically split sample (sample 1) is consistent with the spatial NNN measurements of Brown and Minsky. The observation of similar domain formation in samples split via a thermal gradient provides concrete evidence that whether split intrinsically or by a temperature gradient, the CDW in NbSe$_3$ forms serially arranged dynamical domains. Each domain oscillates at a unique NNN frequency and mode-locks at a unique current bias. Hence, the dynamic CDW breaks into velocity-coherent domains, with each domain traveling at a unique drift velocity. Because a CDW is a coupled elastic wave, the velocity difference between neighboring domains requires that the interface between them be composed of a phase slip center.

In closing, we briefly consider the nature of the mechanism responsible for domain splitting in CDW samples. Because a CDW is a coupled wave, the
velocity at which it moves must be spatially uniform. Additionally, the
CDW velocity is a strong function of temperature below \( T_{\mathrm{c}} \). Hence, in
a temperature gradient, the CDW velocity would vary continuously along the
length of a CDW sample in the absence of CDW coupling. Because the CDW
coupling requires that \( v_{\mathrm{CDW}} \) be independent of position, the CDW velocity is
held spatially uniform in a \( \Delta T \) by the formation of energetically costly
phase distortions.\(^{11}\) Past a critical \( \Delta T \), it becomes energetically
favorable for the CDW to split into two sub-domains, with a phase slip
center created at the domain interface. The data in Fig. 4 indicate that
the phase-strain and phase-slip energies are strong functions of both the
bias current magnitude and direction. In samples which are intrinsically
split, impurity distribution variations presumably create a "chemical
gradient" which serves the same purpose as a temperature gradient. In both
cases, the variation of CDW parameters along the length of the crystal
cause the CDW to form independent, serially arranged phase-velocity
coherent domains. In "wide" samples of the CDW conductors \( \mathrm{K}_{0.3}\mathrm{MoO}_{3} \) or
(\( \mathrm{TaSe}_{4} \)\)\(_{2}\)I, CDW parameter variations could also lead to the formation of CDW
domains arranged in a parallel fashion.

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