Line splitting and frequency locking of narrow-band emission in short TaS_3 samples

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A shift in the emission spectrum of one sector of a TaS_3 sample has been observed when an additional current is passed through a neighboring sector. Line splitting and frequency locking are also observed.

Considerable interest has recently been attracted to the mechanism for the narrow-band emission in quasi-1*D* conductors such as $NbSe_3$ and TaS_3. It has been shown experimentally[1–5] and theoretically[3,6–8] that the narrow-band emission which accompanies the motion of a charge density wave probably comes, not from the interior of the conductor, as was previously believed, but from a region near a contact. The experiments which we are reporting in this letter show that changing the current flow conditions near a contact by passing an additional current through a sector of the sample adjacent to this contact causes a shift of certain lines in the emission spectrum in long samples, without affecting the positions of others. In short samples we find a splitting of lines and also a mutual synchronization of the oscillations at different frequencies, i.e., a frequency locking.

We studied the narrow-band emission in orthorhombic TaS_3 samples with small transverse dimensions (the cross-sectional area is 0.2–1.5 μm^2). Contact stripes 30 ± 5 μm wide with a minimum separation $L \simeq 20$ μm (the distance between their edges) are deposited through special masks on the samples, on a quartz glass substrate. The resistance of the contacts at 118 K does not exceed 100 Ω. The threshold field E_T increases with decreasing distance between the contacts, beginning at $L \simeq 60$ μm, from 1.5 to 5 V/cm at 20 μm (Refs. 1 and 9). In a given-current regime, periodic oscillations of the voltage arise across the sample at $I > I_T$. The amplitude of these oscillations is sufficient for direct detection (without an averaging step, as in Ref. 10) of their frequency dependence by means of a spectrum analyzer and an x, y recorder. Emission has been observed over the frequency range from 0.1 to 100 MHz, in a spectrum consisting of one or several fundamental frequencies and their harmonics. The average width of the emission lines is $\simeq 100$ kHz, but this width reaches $\lesssim 10$ kHz in the highest-quality samples; at this level, the width is determined by the noise in the circuit of the sample. As the current through the sample is increased, the frequencies of all the spectral lines increase in direct proportion to the current (I_1) of the charge density wave. Deviations from a linear dependence $f \sim I_1$ are usually observed at low frequencies, up to a few megahertz, and at high frequencies ($f \gtrsim 50$ MHz). At high frequencies the linear increase in f slows by an amount reaching 2% at 100 MHz, because of the heating of the sample which begins at high currents. On the whole, however, the relative positions of the fundamental frequencies remain the same, and curves of these frequencies versus I_1 do not intersect.

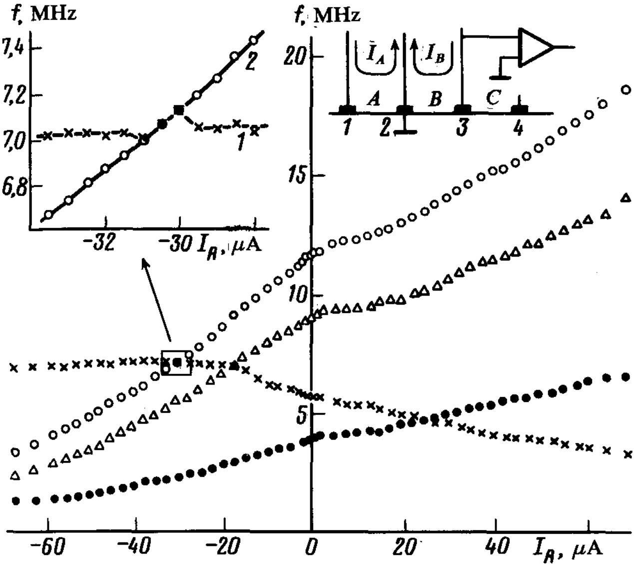

FIG. 1. The fundamental frequencies of the narrow-band emission of sample 1 ($L \simeq 20$ μm, $I_B = 8.0$ μA) versus the control current I_A. Part of the dependence near $I_A = -30$ μA is shown in larger scale. The temperature is $T = 118$ K. The inset shows the measurement circuit.

To determine the mechanism for the emission and the positions of the regions in which the oscillations are nucleated, we studied the change in the emission spectrum during a local perturbation of a region of the sample near a contact. In contrast with Refs. 4 and 5, where a temperature gradient was established between the contacts to the sample, we passed an additional (control) current I_A through one of the sectors of the sample, e.g., sector A, adjacent to a given sector B (Fig. 1). The current I_A also flowed through common contact 2, changing the conditions for the flow of the main current, I_B, near this contact.[1,11] We found that in long samples ($L \gtrsim 100$ μm) the current I_A shifts the frequency of one group of lines in the emission spectrum while not affecting the positions of a second group (within the line width). The application of the control current I_A to the other contact of sector B has the opposite effect: It shifts the previously fixed lines, without affecting the positions of the first group of lines. The effectiveness of this control over the emission spectrum decreases with increasing length of the contacts and with decreasing contact resistance. In short samples ($L \sim 20$–40 μm) a change in I_A causes a change in the positions of essentially all the lines, and these changes are nonlinear in I_A (Fig. 1). The frequencies of the fundamental group of lines and of their harmonics increase 5–15 MHz as I_A is changed from -60 to $+80$ μA. However, the spectrum has a line, and harmonics of this line, whose frequencies decrease under these conditions. The lines also exhibit a characteristic splitting when I_A is changed. Figure 2 shows in detail how one of the lines ($f \simeq 5$ MHz) initially shifts only slightly with increasing current I_A but at $I_A \gtrsim 3$ μA splits into two lines, which move apart. A similar line splitting was recently observed in $NbSe_3$ upon the imposition of a temperature gradient.[5] As we varied I_A, we also observed inverse processes: the coalescence and intersection of some of the lines with

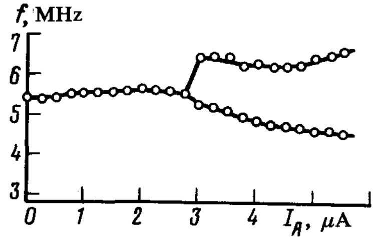

FIG. 2. Splitting of the emission lines of sample 2 ($L = 35$ μm, $I_B = 1.5$ μA) upon a change in the control current I_A ($T = 118$ K).

originally ($I_A = 0$) different frequencies, not harmonically related (Fig. 1). Figures 1 and 3 show in detail a region in which two emission lines intersect. When the lines are sufficiently close together, we observe an abrupt change in the frequency of line 1, with the result that the frequencies of the oscillations corresponding to lines 1 and 2 become equal (within the line width, $\simeq 100$ kHz). As I_A is subsequently varied $\sim 3\%$, the frequencies of the lines remain the same, and the width of the composite line does not exceed the width of either of the original lines (Fig. 3). The lines then move apart. Similar effects have been observed for other intersecting lines, in all the samples. In some cases, the coalescence of two lines is preceded by an instability of their amplitudes and frequency positions; in certain cases, a third line appears between the two. All these features are characteristic of the frequency locking of two, coupled, self-excited, oscillatory systems, which has been studied previously.

Our results can be explained in the following way. In quasi-1D conductors with $E = 0$, an incommensurate charge density wave is pinned at impurities and strong inhomogeneities, primarily near contacts. When a current $I < I_T(L)$ begins to flow, the charge-density wave initially becomes deformed; a steady-state electric field distribution is then established in the presence of the immobile charge-density wave.[7,12] At $I > I_T(L)$ the growing phase gradient of the charge density wave decreases periodically

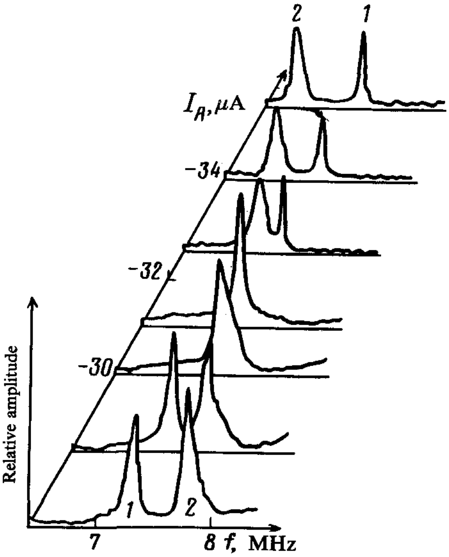

FIG. 3. Change in the emission spectrum of sample 1 caused by a change in the control current I_A near -30 μA ($T = 118$ K).

through a local abrupt change of 2π in the phase ("local" here means at a distance x_0 from the contact).[6–8] In short samples, where the pinning of the charge-density wave is determined by the contacts, this excitation of oscillations and also the appearance of a nonlinearity on the I-V characteristic become possible only when x_0 is shorter than the length of the sample, L. Estimates based on Ref. 7 show that in the short samples in the present experiments, with L ranging from 20 to $\sim 100\ \mu$m, the value of $x_0(E_T)$ is indeed on the order of L at $E \gtrsim E_T(L)$.

The passage of a control current I_A shifts the emission lines (Fig. 1). The change in the emission frequency due to the heating of the contact region by the current I_A or its branching into the circuit of sector B does not exceed 100–200 kHz according to our estimates, and it cannot explain the large frequency shifts that are observed. The most probable explanation is that the control current exerts its effects through current-conversion mechanisms at a contact[1,2,7,12] and a change in the boundary conditions for the charge density wave, in particular, a shift of the pinning boundary for the charge density wave in sector B (Ref. 1). The effect of the control current passed through various contacts of sector B on the various emission lines observed by us in the long samples is evidence that the emission is localized near the contact.

In short samples, with $x_0(E_T) \lesssim L$, the emission regions near the contacts apparently overlap significantly, and under certain conditions the self-excited oscillations in these regions may become synchronized. The application of a control current to one of the contacts causes a splitting of the originally synchronized frequencies of the oscillations in different regions (Fig. 2) or, on the contrary, a mutual locking of the frequencies of oscillations which originally occurred at different frequencies (Figs. 1 and 3).

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