Measurement of the Sliding Charge-Density-Wave Phase Velocity in $Rb_{0.3}MoO_3$

A. Jánossy,[1,2] C. Berthier,[1] P. Ségransan,[1] and P. Butaud[1]

[1]*Laboratoire de Spectrométrie Physique, Université Scientifique Technique et Médicale de Grenoble, 38402 Saint Martin d'Heres, France*

[2]*Central Research Institute for Physics, H1525 Budapest 114, Hungary*

(Received 6 April 1987)

To determine the phase velocity of the charge-density-wave voltage, noise and ^{87}Rb NMR spectra at an electric field of about $15E_T$ were measured together with I-V characteristics on a $Rb_{0.3}MoO_3$ crystal between 40 and 60 K. NMR spectra are reproduced by use of a velocity distribution determined by the noise spectrum. A ratio of the average noise frequency to j_{CDW} of between 12.8 and 17.7 kHz/A·cm^{-2} and a ratio of the average local-field oscillation frequency to j_{CDW} of 11 ± 1 kHz/A·cm^{-2} are found, values close to 12.5 kHz/A·cm^{-2} from the electron density. NMR sidebands show that local-field oscillations are coherent.

PACS numbers: 71.45.Lr, 76.60.Cq

In a number of solids with a quasi one-dimensional electronic structure, a nonlinear electric current is observed[1] below the Peierls transition termed sliding charge-density waves (CDWs) or Fröhlich conduction.[2] The CDW wavelength is incommensurate with the periodicity of the underlying lattice in these materials and thus the energy is independent of the CDW phase relative to the lattice. An electric field may then induce a sliding CDW which moves together with the periodic lattice distortion. Impurities or other defects pin the CDW so that Fröhlich conduction appears only above a threshold field E_T. This extra current is accompanied by large voltage fluctuations referred to as "noise,"[3] the origin of which is not satisfactorily understood: It may arise from impurities hindering the CDW motion[4] or from an instability at the boundaries between pinned and unpinned regions.[5] In both cases, the voltage noise frequency ν_n is related to the drift velocity of the CDW v by

$$\nu_n = v/\lambda, \tag{1}$$

where λ is the CDW wavelength.

Up to now, the measurement of ν_n as a function of the CDW current j_{CDW} has been the only available method to verify quantitatively the—long ago assumed[4]—relation between j_{CDW} and the CDW drift velocity v,

$$v = j_{CDW}/en, \tag{2}$$

where, at low temperatures, n is the density of electrons condensed into the CDW. The drift velocity is related to the winding rate of the CDW phase:

$$\nu_d = v/\lambda = (1/2\pi)d\phi/dt. \tag{3}$$

Various authors[6] suggested that in Eq. (1), λ should be replaced by $\lambda/2$ and Bardeen[7] proposed that $\nu_d = 2\nu_n$.

In this paper we report the first direct measurement of ν_d as a function of j_{CDW} from an analysis of NMR line shapes based on the fact that ν_d is also the frequency of the modulation of the local field due to the CDW at a nucleus. Previous NMR works[8-10] have already demonstrated the motion of the CDW but failed to give quantitative information on the drift velocity v. Our determination of ν_d relies only upon one assumption, namely, that the voltage noise spectrum is proportional to the spatial distribution of v. We verify relation (1) by a factor much better than 2. Direct comparison of the voltage noise spectra with j_{CDW} leads to similar agreement. We also report the first observation of sidebands in the NMR spectra which demonstrate that the temporal modulation of the local field due to the CDW at a nucleus is periodic rather than stochastic. The position of these sidebands allows a straightforward determination of ν_d, establishing unambiguously that ν_d and ν_n are equal.

The $Rb_{0.3}MoO_3$ single crystal, kindly supplied by J. Marcus, was 4.0 mm along the highly conducting b axis and had faces 0.175×2.1 mm^2 perpendicular to b. Contacts were copper plated onto the polished faces. I-V characteristics and noise and NMR spectra were taken under the same conditions *in situ* in the NMR coil. Eleven current-voltage characteristics were taken between 40 and 59 K. Joule heating was negligible under the conditions of the experiments. The CDW current is inhomogeneous over the sample cross section and the measured density j_{CDW} is an average value. The threshold field at which a nonlinear current first appeared was $102+4$ mV/cm within the investigated temperature range in agreement with Dumas *et al.*[11] For high electric fields, where, as the NMR shows, in nearly all the crystal the CDW is depinned, both the CDW current and the normal current are activated as reported by Fleming *et al.*[12] The activation energy $U_{CDW}=754$ K is substantially larger than the normal current activation energy $U_n=488$ K (Fig. 1), in agreement with Jánossy *et al.*[13] The value of U_n may be extrinsic.

The voltage noise spectra were taken at the same applied field of 1.51 V/cm at ten temperatures between 40 and 59 K. In the following, the voltage power spectra

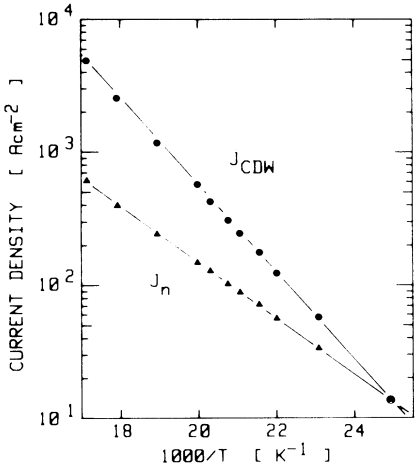

FIG. 1. CDW (circles) and normal (triangles) current densities at a field of 1.5 V/cm ($E/E_T \simeq 15$) as functions of inverse temperature. The normal current is extrapolated from low-field data. Activation energies are 754 and 488 K for the CDW and normal currents, respectively.

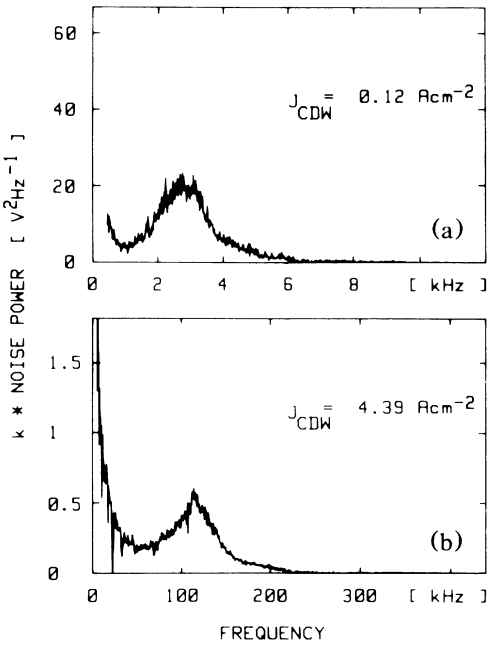

FIG. 2. Noise power spectra at (a) 59.2 K and (b) 45.3 K. The dc field is $E = 1.5$ V/cm ($\simeq 15E_T$) in both cases. A small correction has been made to subtract the first-harmonic component from the raw data. Note the similarity of the spectra despite the change of a factor of 40 of the frequency scale. The voltage power $|V(\nu_n)|^2$ changes inversely to the mean frequency so that the total intensity remains constant. k is the same parameter for the two spectra. The curve in (a) was used to determine the velocity distribution for the calculation of the NMR spectra of Fig. 3.

(i.e., the square of the voltage noise) is analyzed. All noise spectra consist of a broad peak and a background becoming large at low frequencies. The first harmonic of the broad peak has an intensity of 10% of the total, independently of the temperature, and was subtracted from the spectra.

The most important feature is that spectra taken at different temperatures with peak positions varying by 2 orders of magnitude can be scaled onto each other by changes of the frequency and intensity scales only (Fig. 2). Thus, to a good approximation, the noise power spectra are proportional to the same function $f(\nu_n/\bar{\nu}_n(T))$ at all temperatures where $\bar{\nu}_n(T)$ is the first moment of the spectra. The CDW current increases with temperature as $\bar{\nu}_n$. Both the noise spectrum and the CDW current are determined with the best precision at 59.2 K where we find

$$\bar{\nu}_n/j_{CDW} = 13.4\ \text{kHz/A}\cdot\text{cm}^{-2},$$

and, in general, for temperatures between 40.6 and 59.2 K we find ratios of $\bar{\nu}_n/j_{CDW}$ between 12.8 and 17.7 kHz/A$\cdot$cm^{-2}. The good agreement of these values with the expectation from the electron density of 12.5 kHz/A$\cdot$cm^{-2} together with the similarity of the spectra (Fig. 2) support our view that the noise intensity spectrum reflects the velocity distribution of the CDW and that this distribution scaled to its average value is little dependent on temperature. We assume in the following that the velocity distribution is proportional to the noise spectrum. This implies that the voltage oscillation intensity of a single domain with a uniform CDW velocity is independent of the velocity far above threshold.[13,14] We find that while the resistance changes by 2 order of magnitude (Fig. 1) between 40 and 59 K the total noise intensity $|V(t)|^2$ stays constant within a factor of 2. Although this is contrary to the expectations of Ong and Maki[15] based on the vortex model, we do not believe it settles the question of the origin of the noise.

NMR spectra of ^{87}Rb (site 2) were obtained by a Brucker model CXP 100 spectrometer at 80.13 MHz. Experimental details can be found in Ref. 9. The spectra shown in Fig. 3 were taken at approximately the same temperatures and with the same dc electric field (1.51 V/cm) applied to the sample as for the noise spectra. A current source was used to minimize the effect of small fluctuations of the temperature on j_{CDW} during the runs, which lasted typically 12 h. The pinned CDW NMR spectrum is independent of temperature[16] in the investigated range. At $T = 40$ K, the application of 1.51 V/cm does not alter the NMR spectrum, although the current-voltage characteristics and the noise spectrum show that the CDW is depinned. Clearly, the CDW velocity is too small to induce a change in the line shape. As j_{CDW} increases with increasing temperature the edges of the spectrum broaden and diminish in intensity and a peak emerges at the center frequency of the static spectrum.

On further increase of j_{CDW} (by increase in T) the central peak becomes the most prominent part of the spectrum. At the intermediate current of $j_{CDW}=0.405$ A/cm^2, additional steps arise on both sides of the spectrum *outside* the frequency range of the static line (Fig. 4).

An analysis of the NMR line shape of blue bronze[16] shows that the width of discommensurations, if they exist, is of the same order as their separations. Thus we treat the CDW like a plane wave. The line shape was calculated in the following way: In the static case, the spatial modulation of the $(\frac{1}{2},-\frac{1}{2})$ resonance frequency is[9]

$$\Delta\nu_{1/2,-1/2}(R)=(\omega_1/2\pi)\cos(qR+\phi)+(\omega_2/2\pi)\cos^2(qR+\phi), \tag{4}$$

where $q=2\pi/\lambda$, and the line shape $G_s(\omega)$ was calculated numerically[9] with the parameters $\omega_1/2\pi=5.2$ kHz and $\omega_2/2\pi=0.6$ kHz. The NMR line shape in the presence of a CDW sliding with a velocity v constant in time and uniform in space is[9,17]

$$G(\omega,v)=\sum_p J_p^2(\omega_1/qv)\delta(\omega-(\omega_2/2)-pqv)G_D(\omega), \tag{5}$$

where J_p is the Bessel function of order p. The spectrum consists of a central line $p=0$ (as in usual motional narrowing by a stochastic process) and sidebands at $\pm\nu_d$ which are a consequence of the periodicity (coherence) of the temporal modulation of the local quadrupole coupling. The only sources of broadening of the central line are the dipolar interaction, the crystal imperfections, and the temporal fluctuations of v which are all taken into account in $G_D(\omega)$. On the other hand, sidebands are broadened in addition to these factors by the spatial distribution of v. Extra broadening due to the temporal fluctuations of v is expected to be the same as for the central line.

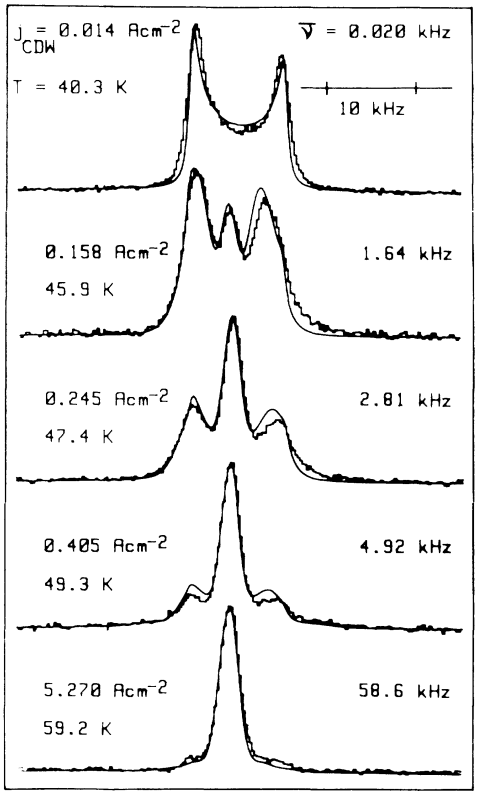

FIG. 3. Variation of the NMR line shape and computer-fitted curves as functions of the CDW current j_{CDW}. The field is fixed at $E=1.5$ V/cm ($\simeq 15E_T$) and the temperature is varied to change j_{CDW}. For the computer-simulated spectra the relative CDW velocity distribution is determined from the experimental noise spectrum at 59.2 K. For each simulated spectrum only one parameter, the average local-field oscillation frequency $\bar{\nu}_d$, was fitted.

We assume our sample to consist of a large number of domains each having a uniform CDW velocity. The spatial distribution $f(v/\bar{v})$ of the relative velocity was taken to be proportional to the experimental noise spectrum at 59 K, $\bar{v}$ acting as a temperature-dependent scaling factor. The NMR spectra were calculated by our folding (5) with the distribution $f(v/\bar{v})$. Only for very low velocities of $v/\lambda<0.17$ kHz was (5) replaced by the asymmetric static spectrum $G_S(\omega)$.

The parameter of most importance, the average drift velocity $\bar{v}=\lambda\bar{\nu}_d$, was obtained for each temperature by our finding the calculated NMR spectrum which best fitted the observed one. The best fits shown in Fig. 3 reproduce quite well the main features. We find the ratio

$$\bar{\nu}_d/j_{CDW}=11\pm 1\ \text{kHz/A}\cdot\text{cm}^{-2}$$

by averaging over fits to NMR spectra at seven temperatures with the error given by 1 standard deviation. This value is in close agreement with the value of 12.5

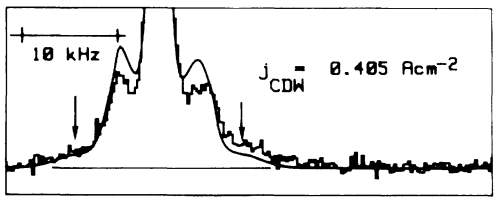

FIG. 4. $p=\pm 1$ sidebands, around ± 8.8 kHz in the NMR spectrum recorded at 49.3 K. These sidebands are evidence of a coherent periodic variation of the local hyperfine field. Solid line is the computer-simulated spectrum.

kHz/A·cm^{-2} expected from the electron density in $Rb_{0.3}MoO_3$. In Fig. 4 are shown the $p = \pm 1$ sidebands observed at 49.2 K. The appearance of sidebands is evidence for the temporal periodicity of the CDW motion. Their positions and widths are well reproduced by the computer simulation. The temporal coherence at a given position is longer than 1 ms since the width of the sidebands is well described by the spatial inhomogeneity of the current. Moreover, an additional broadening due to a temporal correlation time shorter than 1 ms would not only add to the width of the sidebands but would be inconsistent with the width of the central peak which remains less than 2 kHz at all temperatures where it was observed. It may be tempting, but incorrect, to analyze the noise spectra in terms of a "narrow-band" Gaussian peak, due to a quasiuniformly moving CDW, broadened by temporal fluctuations, and a "broad-band" component with high intensity at low frequencies with a different origin. Taking the velocity distribution which corresponds to the Gaussian peak only led to a disagreement with the observed line shape for any average velocity at all intermediate temperatures.

In conclusion, we found the following: (i) The average noise frequency $\bar{\nu}_n$ generated by a current j_{CDW} is close to that expected from both classical impurity and vortex models; (ii) the noise intensity $|V(t)|^2$ is nearly temperature independent; (iii) the NMR spectra under current are well reproduced by standard theory if the *relative* velocity distribution is obtained from the noise spectra; (iv) the frequency ν_d of local hyperfine field oscillations, a direct measure of the CDW drift velocity, has a value equal to the noise frequency ν_n within an experimental accuracy much better than a factor 2; and (v) the observed NMR sidebands provide direct proof for the coherent nature of the sliding motion.

Laboratoire de Spectrométrie Physique is Laboratoire No. LA08 associé au Centre National de la Recherche Scientifique.

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