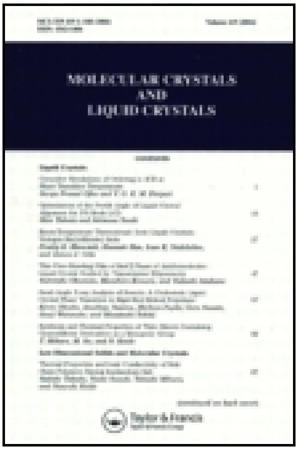

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EVIDENCE FOR MICROWAVE PHOTON-ASSISTED CDW TUNNELING IN NIOBIUM TRISELENIDE

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Abstract Nonlinear conduction has been observed in $NbSe_3$ at 22 K with microwave fields of amplitudes an order of magnitude both above and below the threshold field for non-linear dc conduction. The experimental method has been phase resolved harmonic mixing. The observations can quantitatively be explained by Bardeen's model of tunneling charge density waves and are in contrast to the model of a classical over-damped oscillator. The additional phase shift due to the tunneling process has been observed over a large temperature range and shows a correlation with the temperature dependence of the dielectric constant. This correlation is considered to be another indication for a tunneling process. The origin of the "narrow-band noise" is shown to be due to the decreasing dielectric displacement with increasing electric field strength for values larger than the threshold field. The zero-field dielectric constant ε is calculated from the conductivity applying the Kramers Kronig relations. For ε there is no scaling of the field dependence with the frequency dependence.

1. INTRODUCTION

Most of the transport properties of niobium triselenide, $NbSe_3$, reported so far can be explained equally well by a classical damped-oscillator model[1] and by a tunneling model of charge density waves (CDW).[2] These experiments have been performed with frequencies up to the 100-MHz range. As the tunneling phenomena under ac conditions is known as "photon-assisted tunneling" and the photon character of electromagnetic radiation is more pronounced at higher frequencies, it is to be expected that at microwave frequencies changes are much better to decide which of the two models yields a correct description of the experimental results. Data obtained by mixing of a 9.5 GHz radiation with its second harmonic at the nonlinear I-V characteristic of $NbSe_3$ at low temperatures have been reported by us.[3] They clearly are in favor of the quantum mechanical model as proposed by Bardeen.[2] In the present paper we show the contribution of the dielectric displacement current to the conduction phenomena to be of significance. In particular, the "narrow-band noise" turns out to be due to a negative differential dielectric constant. Also, a calculation of the zero-field dielectric constant from the conductivity is presented.

2. PHASE RESOLVED HARMONIC MIXING

In our experiment the voltage applied to the filamentary sample consists of a fundamental wave and its second harmonic, with a variable phase shift ϕ between the two:

$$E(t) = E_1 \cos(\omega t + \phi) + E_2 \cos(2\omega t) \qquad (1)$$

As we have shown before,[4] for an amplitude sum $E_1 + E_2$ less than the threshold field E_{th} for the onset of the nonlinearity in the current-field characteristic, there should be no dc current $\bar{j}^t$ (time dependent current averaged over t) for any consideration based on classical physics: $\overline{E}^t = 0$ and hence, for $j(t) \propto E(t)$, also $\overline{j}^t = 0$. Only at $E_1 + E_2 > E_{th}$, the non-linear j(E) characteristic should yield a signal $\overline{j}^t \neq 0$ which for small deviations from Ohm's law is proportional to $E_1^2 E_2 \cos 2(\phi + \psi)$ where ψ is a contribution to the phase by the conduction process. For large deviations from Ohm's law, powers higher than the second in E_1 and the first in E_2 should prevail, together with the occurrence of higher terms in the Fourier expansion of $\overline{j}^t$ as a function of ϕ. For the classical oscillator model, the threshold field E_{th} increases with the frequency ω, approximately as ω^2. For microwave frequencies which are higher than the pinning frequency of the charge density wave by two orders of magnitude, it should therefore be impossible to observe a signal without considerably raising the sample temperature by Joule heating.

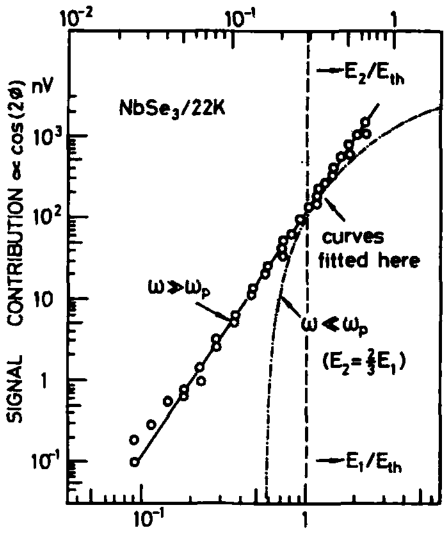

FIGURE 1 Experimental $\cos(2\phi)$ dependent mixing signal as a function of the field amplitudes E_1 and E_2; dash-dotted curve: calculated according to the classical construction of $\overline{j}^t$ from the dc characteristic.

The experimental result, however, is different. Fig.1 (reproduced from ref.4, Fig.3 for one sample only) shows the signal which is a dc voltage since we suppress the dc current $\vec{j}^t$ by applying a high-impedance voltmeter instead of an ammeter, as a function of E_1 and E_2 in a double-log plot. The straight line through the data points represents a dependence according to the $E_1^2E_2$ law mentioned above. The vertical dashed line indicates the field strengths E_1 and E_2 where $E_1 + E_2$ is equal to the dc threshold field. The signal (at least 97 % of it) is proportional to $\cos\{2(\phi + \psi)\}$. It is remarkable that there is a signal even for $E_1 + E_2$ less than E_{th} by an order of magnitude. The $E_1^2E_2$ behavior is observed both below E_{th} and above over four orders of magnitude in the signal, i.e. even at large deviations from Ohm's law in the dc characteristic. The dash - dotted curve has been calculated by the classical construction of the signal from the dc characteristic and fitted to the data at one point. There is no agreement between the data and this curve.

We can, however, beautifully represent the data by Bardeen's theory without any fitting parameter: we get the observed functional dependence on E_1, E_2, and ϕ. The signal below threshold is explained by photon assisted tunneling. While in the usual band theory of solids the photon character of electromagnetic radiation shows up only at energies where there are significant changes in band structure, the very small Peierls gap caused by charge density wave formation yields a photon effect already at microwave frequencies. The situation is somewhat similar to Josephson tunneling of Cooper pairs in superconductivity, and, in fact, the mathematical procedure to calculate phase resolved harmonic mixing in the present case is the same.[5] Since our frequencies are large compared to the pinning frequency and since we measure only the time-independent part of the signal, the calculation is simplified very much leading to the simple functional dependence given above. Even quantitative agreement is obtained: it is within a factor of 3 if we take for the effective sample lenght only the part of the sample inside the waveguide. Because of the extremely high refractive index of the sample it is probably more appropriate to take the complete sample length into account which brings the data to even better agreement with the Bardeen theory.

According to a suggestion by Bardeen, we have also observed the phase shift due to the tunneling process. For this purpose we placed a reference sample (n-Ge at 300 K) before the $NbSe_3$ sample. In the former it is the "hot-electron" effect which provides a mixing signal. Taking into account the phase shift due to the waveguide between the two samples, Fig.2 shows the additional phase shift ψ as a function of temperature. A comparison with the dielectric constant ε for various frequencies as observed by Grüner et al.[6] shows a strong correlation. According to Thornber et al.[7],

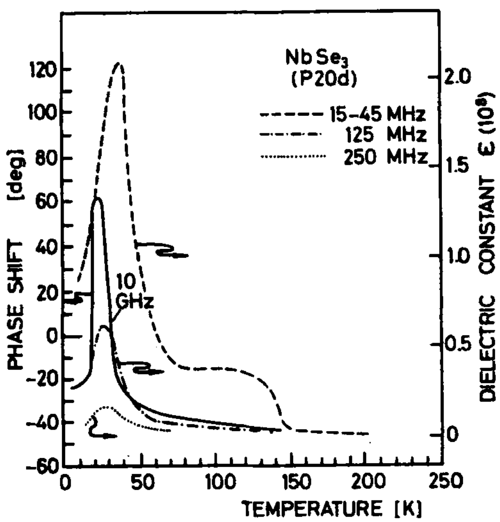

FIGURE 2 Observed phase shift ψ due to dielectric relaxation, as a function of temperature (full curve); dielectric constant at various frequencies (other curves).

the tunneling time of an electron in a crystal lattice is the dielectric relaxation time τ_d. One may expect for CDW tunneling $\tan(2\psi) \approx \omega\tau_d = 2\pi\nu\varepsilon\varepsilon_o/\sigma_b \approx 0.3$ for $\varepsilon \approx 10^7$ at $\nu \approx 10^{10}$ Hz. This is about the right order of magnitude.

3. NARROW BAND NOISE AND THE DIELECTRIC CONSTANT

Finally, we propose a theory for the Fleming-Grimes instability.[8] Even though only a dc current is applied to the sample there is a noise field E(t) from which a time dependent current j(t) and a displacement current dD/dt may arise. While usual instabilities

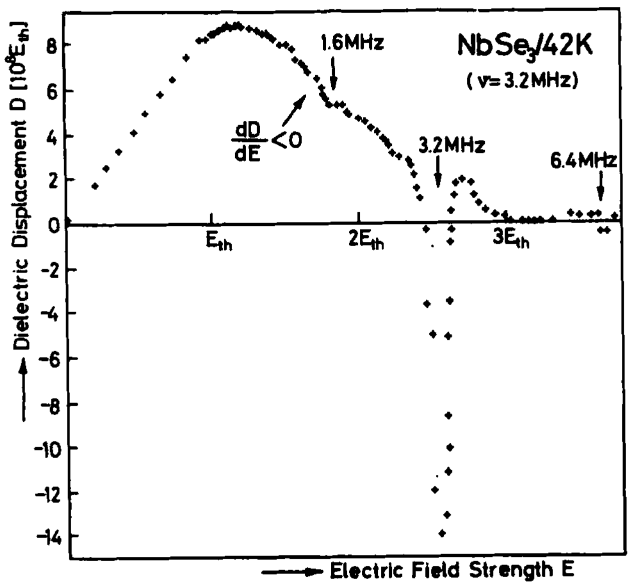

FIGURE 3 Dielectric displacement D vs electric field strength E for $NbSe_3$ at 42 K (after ε(E) data by Grüner et al[9]). The resonances indicated by 1.6 MHz, 3.2 MHz, and 6.4 MHz are related to the frequency of 3.2 MHz of measurement and can be disregarded.

such as the Gunn oscillations arise due to a negative differential conductivity $dj/dE < 0$ ("NDC") the present instability arises because $dD/dE < 0$ (negative differential dielectric constant, "N-double D - C"). The dielectric relaxation time τ_d as obtained from a combination of Poisson's equation $dD/dt = e\Delta n$ with the continuity equation $ed\Delta n/dt + dj/dx = 0$ is given by $(dD/dE)/(dj/dE)$. From Grüner and Zettl's data[9] on $\varepsilon(E)$ one can easily construct $D = \varepsilon E$ (see Fig.3) and notice that $dD/dE < 0$ for $E > E_{th}$ while $dj/dE > 0$ in this range. From now on the theory is formally very similar to that for the Gunn diode[10] except that the velocity $v = $ const.

We have also calculated the frequency dependent low-field dielectric constant $\varepsilon(\omega)$ from the frequency dependent conductivity

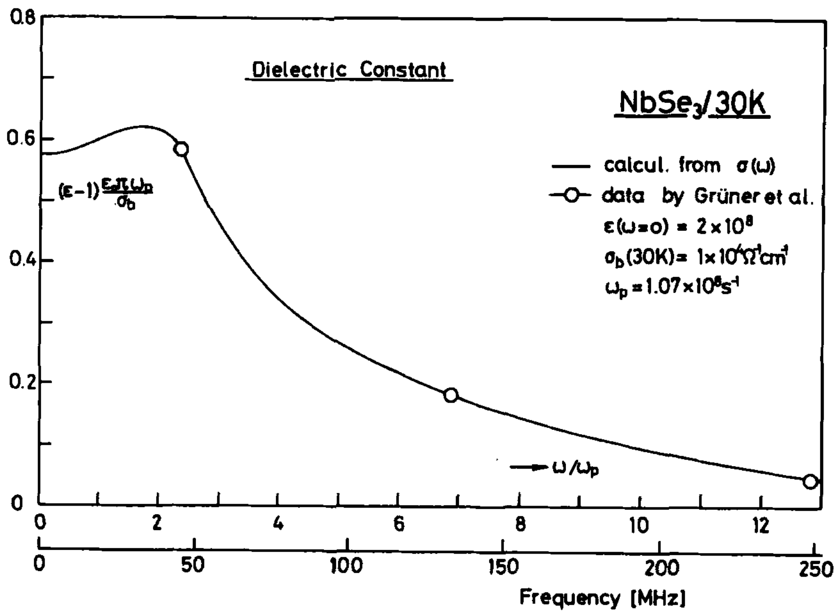

FIGURE 4 Dielectric constant vs. frequency at 30 K calculated from the frequency dependent CDW conductivity (curve), and experimental data by Grüner et al.[6]

$\sigma(\omega)$ as described by the Bardeen formula using the well-known Kramers Kronig relations for a conducting dielectric.[11] The result is shown in Fig.4, together with experimental data by Grüner et al.[5]: there is excellent agreement. In contrast, however, to the behavior of the conductivity, there is no scaling of the field dependence with the frequency dependence of the dielectric constant. It is tempting to assume that the decrease of ε with E for $E > E_{th}$ is due to the depinning of CDW's: only pinned CDW's contribute to the polarization. However, a quantitative fit is not possible with this assumption taking Bardeen's equation for the field-dependent number of depinned CDW's ito account, and it remains unclear why a moving CDW should not contribute also to polarization.

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