Effect of impurities on the threshold field of a commensurate charge-density wave

S. N. Artemenko and A. N. Kruglov

Institute of Radio Engineering and Electronics, USSR Academy of Sciences
(Submitted 11 March 1982)
Zh. Eksp. Teor. Fiz. **83**, 1134–1139 (September 1982)

The effect of an impurity on the threshold electric field E_T of a commensurate charge density wave in a quasi-one-dimensional conductor is investigated theoretically. Equations describing the smooth fluctuations of the electric potential φ and phase χ of the charge density wave are obtained on the basis of the microscopic theory by taking the impurities into account. It is concluded that the fluctuations of χ due to the impurities decrease E_T.

PACS numbers: 71.55. – i

It has been established in the last few years that the motion of a charge-density wave (CDW) in quasi-one-dimensional conductor below the Peierls-transition temperature contributes to the conductivity of the crystal.[1–3] Examples are such substances are $NbSe_3$ and TaS_3. In weak electric fields E, the conductivity of such substances is constant and is determined by the free electrons with energies larger than the Peierls gap. In this case the CDW is at rest, being pinned by the impurity and (or) by the crystal lattice (in the case of a commensurate CDW). When the field E rises above a certain threshold, the CDW begins to move and the crystal conductivity increases, with the CDW motion accompanied by nonstationary effects.[2,3] To describe the contribution of the CDW to the conductivity, both a phenomenological approach[4–6] and a microscopic theory[7–9] were used. The threshold field E_T was calculated both for an incommensurate CDW, when the pinning is by the impurities,[4,5] and for a commensurate CDW, when the pinning is connected with the commensurability of the periods of the CDW and of the initial lattice.[5,9] The threshold field of the incommensurate CDW increases with increasing impurity density. In a commensurate CDW, in the limit of a pure substance, E_T should not depend on the impurities. Yet in orthorhombic TaS_3, where the CDW is commensurate, the threshold field is different in different samples. Thus, in Ref. 10 E_T reaches 100 V/cm, and in Ref. 11 E_T amounts to 10–40 V/cm, while in Ref. 3 the samples investigated has $E_T = 2.2$ V/cm. This raises the question of the cause of such a scatter of E_T. It might be assumed that the impurities produce an additional mechanism of pinning of the commensurate CDW and increase the threshold field. We shall show that this is not the case: addition of impurities to a pure material lowers of the threshold field of the commensurate CDW as a result of the order-parameter phase fluctuations produced by the impurities.

We use the theory developed in Refs. 8 and 9, where equations were obtained for the Green's functions that describe the kinetics of a quasi-one-dimensional conductor with CDW. The equations were obtained for matrices of Green's functions and are similar to the equations used to describe kinetic phenomena in superconductors. These Green's function determine the state density, the order parameter $\tilde{\Delta} = \Delta e^{i\chi}$, and the electron distribution function; they can be used to calculate the current density and the charge density.

In the equations of Refs. 8 and 9, averaging was carried out over the locations of the impurities, and no account was taken of the CDW order-parameter fluctuations due to the impurities. Yet the fluctuations are quite substantial, and a particularly important role is played by long-wave fluctuations of the phase χ, which can lead to violation of the long-range order even in the case of a commensurate CDW.[12]

To take into account the phase fluctuations, we shall not average over the impurities in the Green's functions of Refs. 8 and 9, and obtain an equation for the phase with allowance for the fluctuations. We assume that the potential of the impurities is concentrated at distances of the order of interatomic, which are small compared with the macroscopic lengths of the problem. The equations for the Green's functions, with momenta near the Fermi surface, will contain then only Fourier components of the impurity potential

$$U(\mathbf{r}) = \sum_i v(\mathbf{r}-\mathbf{r}_i)$$

with wave vector $\mathbf{q} = 0, \pm\mathbf{Q}$, where $\mathbf{Q}$ is the wave vector of the CDW. The Fourier components with $\mathbf{q} = 0$, which describes the renormalization of the chemical potential and is not connected with the phase fluctuations, will be disregarded. The Fourier components

$$U_{\pm\mathbf{Q}} = \sum_i V e^{\pm i\mathbf{Q}\mathbf{r}_i}, \qquad V = \int U(\mathbf{r})\, e^{i\mathbf{Q}\mathbf{r}}\, d\mathbf{r}$$

relate the diagonal Green's functions that describe the current and charge densities with the off-diagonal components that describe $\tilde{\Delta}$. The equations for the Green's functions in the stationary state are of the form

$$-iv\frac{\partial \check{g}}{\partial x} - [\varepsilon\hat{\sigma}_z + \hat{\Delta} + \varphi\hat{\sigma}_z - \eta\hat{\sigma}_z, \check{g}]_-$$
$$= V\delta(\mathbf{r}-\mathbf{r}_i)\,[\hat{\sigma}_y \cos \mathbf{Q}\mathbf{r} + \hat{\sigma}_x \sin \mathbf{Q}\mathbf{r}, \check{g}]_-, \tag{1}$$

where x is the coordinate along the conducting filaments, φ is the electrostatic potential, $v = Q_m/2m$ is the Fermi velocity,

$$\check{g}=\begin{pmatrix} \hat{g}^R & \hat{g} \\ 0 & \hat{g}^A \end{pmatrix}, \quad \hat{g}=\begin{pmatrix} g & f \\ -f^+ & \bar{g} \end{pmatrix}, \quad \hat{\Delta}=\begin{pmatrix} 0 & \tilde{\Delta} \\ -\tilde{\Delta}^* & 0 \end{pmatrix},$$

$$\hat{g}=(\hat{g}^R-\hat{g}^A)\operatorname{th}\frac{\varepsilon}{2T}+\hat{g}^{(a)},$$

$\hat{g}^R, \hat{g}^A, \hat{g}^{(a)}$ are the retarded, advanced, and anomalous Green's functions, $\eta(\mathbf{p}_\perp)$ describes the dependence of the band spectrum of a quasi-one-dimensional conductor on the transverse momentum $\mathbf{p}_\perp$. We shall assume below that $\eta \ll \Delta$. The function $g(\mathbf{r}_\perp, \mathbf{r}'_\perp)$ depends on the energy ε and on the two transverse coordinates (and in the momentum representation on the two transverse momenta $\mathbf{p}_\perp$ and $\mathbf{p}'_\perp$);

$$[\check{A}, \check{g}]_- = \check{A}(\mathbf{r}_\perp)\check{g}(\mathbf{r}_\perp, \mathbf{r}_\perp') - \check{g}(\mathbf{r}_\perp, \mathbf{r}_\perp')\check{A}(\mathbf{r}'_\perp).$$

We consider a sufficiently pure conductor, so that the mean free path of the electron exceeds the coherence length $\hbar v/\Delta$. In this case we can disregard the influence of the impurities in the size of the energy gap Δ. We confine ourselves also to the case of low temperature $T \ll \Delta$. This is just the situation in TaS_3, where $T_p = 218$ K and $\Delta = 740$ K even several degrees below T_p. In this case the number of free electrons with $\varepsilon > \Delta$ is small, and we can disregard $g^{(a)}$, which determines the change of the distribution function of the excitations.

For convenience, we separate the phase factors in $\tilde{\Delta}$ and in the functions $\hat{g}$. This corresponds to transformation $\hat{g} \to \hat{S}^+ \hat{g}\hat{S}$, where

$$\hat{S}=\begin{pmatrix} e^{i\chi/2} & 0 \\ 0 & e^{-i\chi/2} \end{pmatrix}.$$

We seek the solution for $\hat{g}^R$ and $\hat{g}^A$ in an approximation linear in φ/Δ and $(\mathbf{v}\times\nabla\chi)/\Delta$:

$$\hat{g}^R=\hat{g}_0{}^R+\hat{g}_1{}^R, \quad \hat{g}_0{}^R=g_0{}^R\hat{\sigma}_z+f_0{}^R i\hat{\sigma}_y,$$

$$g_0{}^R=\frac{\tilde{\varepsilon}}{\xi_{\tilde{\varepsilon}}{}^R}, \quad f_0{}^R=\frac{\Delta}{\tilde{\varepsilon}}g_0{}^R, \quad \tilde{\varepsilon}=\varepsilon-\eta(\mathbf{p}_\perp),$$

$$\xi_\varepsilon^{R(A)}=[(\varepsilon\pm i0)^2-\Delta^2]^{1/2}(\xi^R_{\varepsilon>\Delta}>0).$$

From (1) we obtain an equation for the Fourier component $\hat{g}_1^R$ with respect to the coordinates:

$$k_\parallel v\hat{g}_1{}^R-\xi_+{}^R\hat{g}_+{}^R\hat{g}_1{}^R+\hat{g}_1{}^R\hat{g}_-{}^R\xi_-{}^R$$
$$=-{}^1/{}_2 i\chi(\eta_+-\eta_-)(\hat{g}_+{}^R-\hat{g}_-{}^R)-(\varphi+ik_\parallel v\chi/2)(\hat{g}_+{}^R\hat{\sigma}_z-\hat{\sigma}_z\hat{g}_-{}^R)$$
$$+V\sum_i e^{i\mathbf{k}\mathbf{r}_i}[\cos(\mathbf{Q}\mathbf{r}_i-\chi)(\hat{\sigma}_y\hat{g}_-{}^R-\hat{g}_+{}^R\hat{\sigma}_y)$$
$$+\sin(\mathbf{Q}\mathbf{r}_i-\chi)(\hat{\sigma}_x\hat{g}_-{}^R-\hat{g}_+{}^R\hat{\sigma}_x)],\tag{2}$$
$$\hat{g}_\pm{}^R=\hat{g}_0{}^R(\mathbf{p}_\perp\pm\mathbf{k}_\perp/2), \quad \eta_\pm=\eta(\mathbf{p}_\perp\pm\mathbf{k}_\perp/2).$$

Using the orthogonality relation

$$(\hat{g}_0{}^R)^2=1, \quad \hat{g}_+{}^R\hat{g}_1{}^R+\hat{g}_1{}^R\hat{g}_-{}^R=0,$$

we easily obtain from (2) a solution for $\hat{g}_1^A$:

$$\hat{g}_1{}^R=\frac{(\xi_+{}^R+\xi_-{}^R)\hat{g}_+{}^R+k_\parallel v}{(\xi_+{}^R+\xi_-{}^R)^2-k_\parallel{}^2v^2}\Big[\frac{i}{2}\chi(\eta_+-\eta_-)(\hat{g}_+{}^R-\hat{g}_-{}^R)$$
$$+\left(\varphi+ik_\parallel v\frac{\chi}{2}\right)(\hat{g}_+{}^R\hat{\sigma}_z-\hat{\sigma}_z\hat{g}_-{}^R)-$$
$$-V\sum_i e^{i\mathbf{k}\mathbf{r}_i}[\cos(\mathbf{Q}\mathbf{r}_i-\chi)(\hat{\sigma}_y\hat{g}_-{}^R-\hat{g}_+{}^R\hat{\sigma}_y)$$
$$+\sin(\mathbf{Q}\mathbf{r}_i-\chi)(\hat{\sigma}_x\hat{g}_-{}^R-\hat{g}_+{}^R\hat{\sigma}_x)]\Big].\tag{3}$$

The solution for $\hat{g}_1^A$ is obtained from (3) by interchanging the indices R and A,

$$\hat{g}_1=(\hat{g}_1{}^R-\hat{g}_1{}^A)\operatorname{th}(\varepsilon/2T).$$

The self-consistency condition obtained in Refs. 8 and 9 for the functions $\hat{g}$ with a singled-out phase, with allowance for the dependence of χ on $\mathbf{r}_\perp$, takes the form

$$\frac{\omega_Q{}^{2''}\Delta}{\omega_Q{}^2}\nabla^2\chi+\frac{i\lambda}{2}\int \operatorname{Sp}\hat{\sigma}_x\hat{g}\,d\varepsilon\frac{d\mathbf{k}_\perp}{S}+\Delta\gamma\sin m\chi=0,\tag{4}$$
$$\omega_Q{}^{2''}=(\partial^2\omega_q{}^2/\partial\mathbf{k}_\perp{}^2)_{\mathbf{q}=\mathbf{Q}}, \quad \gamma\sim(\Delta/\varepsilon_0)^{m-2}\ll1,$$

where ω_Q is the frequency of the phonons with wave vector $\mathbf{q}=\mathbf{Q}$, and λ is the dimensionless electron-phonon interaction constant. The last term describes the influence of the commensurability, m is the order of the commensurability, ε_0 is an energy of the order of the width of the conduction band (at $m = 4$, just as for TaS_3, we have $\varepsilon_0 = \varepsilon(3Q/2)$).

Substituting the solution (3) in (4) and assuming $k_\parallel v \ll \Delta$ and $k_\perp v_\perp \ll \Delta$ we obtain an equation for the smooth fluctuations of the phase χ:

$$-a_\parallel\frac{\partial^2\chi}{\partial x^2}-a_\perp\frac{\partial^2\chi}{\partial r_\perp{}^2}+\frac{\gamma}{\lambda}\sin m\chi+\sum_i b\delta(\mathbf{r}-\mathbf{r}_i)\sin(\chi-\mathbf{Q}\mathbf{r})$$
$$=\frac{e\hbar v}{\Delta^2}\frac{\partial\varphi}{\partial x};$$

$$a_\parallel=\frac{\hbar^2v^2}{2\Delta^2}, \quad a_\perp=\frac{\omega_Q{}^{2''}}{\lambda\omega_Q{}^2}-\frac{\hbar^2\overline{v_\perp{}^2}}{2\Delta^2};\tag{5}$$

$$b=\frac{V}{\Delta\lambda}; \quad \overline{v_\perp{}^2}=\int\left(\frac{\partial\eta}{\partial\mathbf{p}_\perp}\right)^2\frac{d\mathbf{k}_\perp}{S}.$$

It can be seen from this equation that the fluctuations of the phase χ are connected with the fluctuations of the electrostatic potential φ. If the quasi-one-dimensional metal consists of chains of different type with electron and hole conductivity, the term in the right-hand side of (5) does not lead to significant effects. If only chains of the same type are present, we must find the connection between the fluctuations of φ and χ with the aid of the Poisson equation, by expressing the charge density in the terms of the trace of $(\hat{\sigma}_z\hat{g})$.[9] For the smooth fluctuations we obtain

$$\varepsilon_\parallel\frac{\partial^2\varphi}{\partial x^2}+\varepsilon_\perp\frac{\partial^2\varphi}{\partial r_\perp{}^2}=k_0{}^2\left[(1-N_s)\varphi-\frac{\hbar^2v^2}{6\Delta^2}\frac{\partial^2\varphi}{\partial x^2}-\hbar\frac{v}{2}\frac{\partial\chi}{\partial x}\right],\tag{6}$$

where $\varepsilon_\parallel^{1/2}/k_0$ and $\varepsilon_\perp^{1/2}/k_0$ are the screening lengths of the electric fields along and across the strings in the normal metallic state (with $\Delta = 0$); $\varepsilon_\parallel$ and $\varepsilon_\perp$ are the dielectric constants along and across the filaments without allowance for the contribution of the conduction electrons. The term with $\partial^2\varphi/\partial x^2$ in the right-hand side of (6) describes the renormalization of $\varepsilon_\parallel$:

$$\varepsilon_\parallel \to \varepsilon_\parallel + \hbar^2 v^2 k_0^2/6\Delta^2.$$

The quantity $1 - N_s = (8\pi\Delta/T)^{1/2} e^{-\Delta/T} \ll 1$ determines the contribution of the free electrons to the charge density.

We eliminate the potential φ from (5) with the aid of (6). Returning to Fourier components, we obtain

$$L\chi = \sum_i b\delta(\mathbf{r}-\mathbf{r}_i)\sin(\chi - \mathbf{Q}\mathbf{r}) + \frac{\gamma}{\lambda}\sin m\chi = \frac{e\hbar v}{\Delta^2}E,$$

$$L = -a_\perp k_\perp^2 - a_\parallel k_\parallel^2 \left[1 + \left(1 - N_s + \frac{\varepsilon_\parallel^2 k_\parallel^2}{k_0^2} + \frac{\varepsilon_\perp^2 k_\perp^2}{k_0^2}\right)^{-1}\right]. \quad (7)$$

Here E is the homogeneous part of the field, which is different from zero if a voltage is applied to the sample. In the limit $v_\perp = 0$ and $T = 0$, this equation goes over into the equation obtained in Ref. 12 for the phase, and neglecting the fluctuations (the term with b) and the dependence of χ on $\mathbf{r}_\perp$ it goes over into the equation of Ref. 9, which was used for the analysis of solitons.

We shall consider hereafter a case in which $a_\perp > 0$ in Eqs. (5) and (7). This condition is easily satisfied if the electron spectrum is strongly one-dimensional and the phonon spectrum is weakly one-dimensional. The ratio of the first and second terms in $a_\perp$ is of the order of

$$\frac{\Delta^2}{\lambda\omega_Q^2}\frac{s^2}{v_\perp^2} \propto \frac{m^*}{m}\frac{s^2}{v_\perp^2},$$

where s is the speed of sound (we assume satisfaction of the adiabaticity condition $m^*/m \gg 1$, which ensures smallness of the one-dimensional thermal and quantum fluctuations).

To find the threshold field E_T we must ascertain the largest value of E at which a stationary solution of Eq. (7) is possible. Neglecting the fluctuations, such a solution is possible at

$$E < E_{T0} = (\Delta^2/e\hbar v)\lambda/\gamma.$$

In fields $E > E_{T_0}$ the equation for the phase has a solution only when account is taken of nonstationary effect (this case was analyzed in Ref. 9).

We obtain now E_T with allowance for the fluctuations. We consider the case when the fluctuations are small and can be taken into account by perturbation theory. We represent the solution (7) in the form $\chi = \bar{\chi} + \delta\chi$, where $\bar{\chi} = \langle\chi\rangle$, and $\langle\ldots\rangle$ is statistical averaging. Averaging (7) in the lowest approximation in $\delta\chi$, we obtain

$$\sin m\bar{\chi}(1 - \langle\delta\chi^2\rangle m^2/2) = E/E_{T0}. \quad (8)$$

We subtract (8) from (7) and obtain in the linear approximation in $\delta\chi$ an equation for $\delta\chi$:

$$\left(L - \frac{\gamma m}{\lambda}\cos m\bar{\chi}\right)\delta\chi = -\sum_i b\delta(\mathbf{r}-\mathbf{r}_i)\sin(\mathbf{Q}\mathbf{r}_i - \bar{\chi}). \quad (9)$$

If the impurities are randomly distributed we obtain

$$\langle\delta\chi^2\rangle = \frac{b^2 n}{2}\int\frac{d^3\mathbf{k}}{[L - (\gamma m/\lambda)\cos m\bar{\chi}]^2}, \quad (10)$$

where n is the impurity density. Substituting (10) in (8) we obtain the dependence of the mean value of the phase $\bar{\chi}$ on the field E and the decrease of E_T due to the fluctuations caused by the impurities. The quantity $\langle\delta\chi^2\rangle$ depends on the relation between the small parameters γ/λ, $1 - N_s$ and $\varepsilon_\perp/a_\perp k_0^2$. In the limit

$$1 - N_s \gg \frac{\gamma}{\lambda}, \quad 1 - N_s \gg \left(\frac{\varepsilon_\perp}{a_\perp k_0^2}\right)^{1/2} \ln^2\frac{\varepsilon_\perp}{a_\perp k_0^2}\frac{\gamma}{\lambda},$$

$$\langle\delta\chi^2\rangle = \frac{\pi^2 n b^2[\lambda(1-N_s)]^{1/2}}{2a_\perp a_\parallel^{1/2}(\gamma m\cos m\bar{\chi})^{1/2}}, \quad (11)$$

$$E_T = E_{0T}\left[1 - \frac{\pi^2 n V^2\omega_Q^2}{\Delta\hbar v\omega_Q^{2''}}\left(\frac{1-N_s}{2\lambda\gamma m}\right)^{1/2}\right].$$

In order of magnitude, the decrease of the threshold field as a result of the impurities is

$$\Delta E_T \sim E_{T0}\frac{\hbar(1-N_s)^{1/2}}{\Delta\tau(\lambda\gamma)^{1/2}}, \quad (11')$$

where τ is the time of momentum scattering by the impurities. Since the denominator of (11′) contains the small commensurability parameter γ, it is seen that E_T can be substantially lowered even in relatively pure samples at $\Delta \gg 1/\tau$. If the Coulomb screening of the fluctuations is negligible because of the presence of chains with different types of conductivity, the result is obtained by leaving out the factor $1 - N_s$ from (11) and (11′).

We shall not consider the case $E > E_T$. It is natural to expect at $E > E_T$, when the slippage of the CDW begins, the radiation from the sample to contain, besides the monochromatic radiation due to the motion of the CDW under the influence of the forces of pining to the lattice,[9] also a noise part due to the presence of phase fluctuations due to the impurities. The stronger the lowering of the field E_T, the more appreciable the noise component in the radiation.

It would be of interest to investigate the case of larger densities of the impurities, when $\langle\delta\chi^2\rangle \gtrsim 1$. However, the solution of (7) in this case is quite difficult. In the limiting case $\langle\delta\chi^2\rangle \gg 1$, according to Ref. 12, the impurities disturb the long-range order of the commensurate CDW.

We are grateful to A. F. Volkov for helpful advice and a discussion and also to K. B. Efetov and A. I. Larkin for a discussion of questions connected with the disturbance of the long-range order.

[1]P. A. Lee, T. M. Rice and P. W. Anderson, Sol. St. Commun. **14**, 703 (1974).
[2]N. P. Ong and P. Nonceau, Phys. Rev. **B16**, 3443 (1977).
[3]A. Zettl, G. Grüner, and A. H. Thompson, Sol. St. Commun. **39**, 899 (1981).
[4]P. A. Lee and T. M. Rice, Phys. Rev. **B19**, 3970 (1979).
[5]J. Bardeen, Phys. Rev. Let. **45**, 1978 (1980).
[6]G. Grüner, A. Zavadovski, and P. M. Chaikin, Phys. Rev. Lett. **46**, 511 (1981).
[7]L. P. Gor'kov and E. P. Dolgov, Zh. Eksp. Teor. Fiz. **77**, 396 (1979).
[8]S. N. Artemenko and A. F. Volkov, Zh. Eksp. Teor. Fiz. **80**, 2018 (1981).
[9]S. N. Artemenko and A. F. Volkov, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 155

(1981) [JETP Lett. **33**, 147 (1981)]; Zh. Eksp. Teor. Fiz. **81**, 1872 (1981) [Sov. Phys. JETP **54**, 992 (1981)].
[10]T. Takoshima, M. Ido, K. Tsutsumi, and T. Sambongi, Sol. St. Commun **35**, 911 (1980).
[11]S. N. Zhilinskii, M. E. Itkis, F. Ya. Nad', I. Yu. Kal'nova, and V. B. Preobrazhenskii, Élektrofizicheskie svoĭstva odnomernykh kristallov TaS_3 (Electrophysical Properties of TaS_3 One-Dimensional Crystals), Inst. of Radio and Electronics, 1981.
[12]K. B. Efetov and A. I. Larkin, Zh. Eksp. Teor. Fiz. **72**, 2350 (1977) [Sov. Phys. JETP **45**, 1236 (1977)].

Translated by J. G. Adashko