New Soliton Equation for Dipole Chains

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A simple dipole chain is the main building block of many physical models in solid state physics. Here, just such a chain of essentially identical dipoles is investigated by considering nearest-neighbor interactions of the individual charges in the dipoles. A three-dimensional model is found. In a restricted, two-dimensional picture, a nonlinear, second-order partial differential equation, more general than sine-Gordon, results. Soliton-kink solutions are found.

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Phenomena involving molecular chains are sometimes investigated by looking at models of arrays of dipoles. Examples are furnished by polymer chains and ferroelectric crystals such as $NaNO_2$, to name two [1,2]. Instead of considering a chain of dipoles as an oscillator system, or else considering a special interaction energy between dipoles, as Pouget and co-workers do for their theory of $NaNO_2$, we will look at central interactions between individual charges comprising neighboring dipoles.

We consider a chain of identical dipoles, each comprised of charges q^+ and q^-, with masses m^+ and m^-, a fixed distance μ_0 apart. Initially, the spacing between dipoles is R_0. The dipoles are free to move away from their initial positions, but such that departures are much smaller than R_0. We also assume $\mu_0 \ll R_0$. However, no restriction is put on the dipole rotations.

In our theoretical treatment, $\mathbf{x}^+$ and $\mathbf{x}^-$ denote the positions of the charges comprising the dipole at $\mathbf{x}$. Thus $\boldsymbol{\mu}=\mathbf{x}^+-\mathbf{x}^-$ is a constant-magnitude (μ_0) vector. The equations of motion of the two charges comprising our dipole have the form

$$m^+\ddot{\mathbf{x}}^+=q^+\mathbf{E}(\mathbf{x}^+)+\lambda(\mathbf{x}^+-\mathbf{x}^-)\,,\qquad m^-\ddot{\mathbf{x}}^-=q^-\mathbf{E}(\mathbf{x}^-)-\lambda(\mathbf{x}^+-\mathbf{x}^-)\,. \tag{1}$$

Here $\mathbf{E}$ is the external electric field of all the other dipoles, and λ is the Lagrange multiplier ensuring the constancy of $|\boldsymbol{\mu}|$. Eliminating λ, we find

$$m\ddot{\mathbf{x}}=q^+\mathbf{E}(\mathbf{x}^+)+q^-\mathbf{E}(\mathbf{x}^-)\,, \tag{2}$$

$$\boldsymbol{\mu}\times[\ddot{\boldsymbol{\mu}}-q^+(m^+)^{-1}\mathbf{E}(\mathbf{x}^+)+q^-(m^-)^{-1}\mathbf{E}(\mathbf{x}^-)]=0\,, \tag{3}$$

where

$$\mathbf{x}^+=\mathbf{x}+(m^-/m)\boldsymbol{\mu}\,,\quad \mathbf{x}^-=\mathbf{x}-(m^+/m)\boldsymbol{\mu}\,,\qquad m=m^++m^-\,,\quad M=m^-m^+/m\,. \tag{4}$$

The force acting on a charge q^+ at $\mathbf{x}^+$ and due to a dipole at $\boldsymbol{\xi}$ is, in terms of the electric field potential ϕ,

$$\mathbf{E}(\boldsymbol{\mu}_\xi\to\mathbf{x}^+)=-\nabla_{\mathbf{x}^+}[q^+\phi(\boldsymbol{\xi}^+-\mathbf{x}^+)+q^-\phi(\boldsymbol{\xi}^--\mathbf{x}^+)] \tag{5}$$

and a similar expression for $\mathbf{E}(\boldsymbol{\mu}_\xi\to\mathbf{x}^-)$. This leads to the following form of (2) and (3):

$$m\ddot{\mathbf{x}}=-\nabla_{\mathbf{x}}\sum_{\xi}\Phi_{\xi,\mathbf{x}}\,, \tag{6}$$

$$\boldsymbol{\mu}\times\left(\ddot{\boldsymbol{\mu}}+M^{-1}\nabla_{\boldsymbol{\mu}_x}\sum_{\xi}\Phi_{\xi,\mathbf{x}}\right)=0\,, \tag{7}$$

where

$$\Phi_{\xi,\mathbf{x}}=(q^+)^2[(q^+)^2\phi(\boldsymbol{\xi}^+-\mathbf{x}^+)+q^-q^+\{\phi(\boldsymbol{\xi}^--\mathbf{x}^+)+\phi(\boldsymbol{\xi}^+-\mathbf{x}^-)\}+(q^-)^2\phi(\boldsymbol{\xi}^--\mathbf{x}^-)]\,. \tag{8}$$

We now use our approximations that departures of the centers of mass of individual dipoles from their initial positions, and also the charge separation in the dipoles, are both small quantities. For nearest neighbors we have

$$\boldsymbol{\xi}-\mathbf{x}=\mathbf{R}_0+\delta\boldsymbol{\xi}-\delta\mathbf{x} \tag{9}$$

and, for individual positive charges in neighboring dipoles,

$$\boldsymbol{\xi}^+-\mathbf{x}^+=\mathbf{R}_0+\delta\boldsymbol{\xi}-\delta\mathbf{x}+(m^-/m^+)(\boldsymbol{\mu}_\xi-\boldsymbol{\mu}_x)\,, \tag{10}$$

where the four last terms are small. There will be similar expressions for $\boldsymbol{\xi}^+-\mathbf{x}^-$, $\boldsymbol{\xi}^--\mathbf{x}^+$, and $\boldsymbol{\xi}^--\mathbf{x}^-$. Denoting

$$\mathbf{u}=\delta\boldsymbol{\xi}-\delta\mathbf{x}\,,$$

we expand $\phi(\boldsymbol{\xi}^+-\mathbf{x}^+)$, etc., in a Taylor series, neglecting terms higher than quadratic in $\mathbf{u}$ and $\boldsymbol{\mu}$. We also neglect

$\sim[\nabla\phi]_{\mathbf{R}_0}$, as these terms will cancel when added up over an infinite array of dipoles. We are left with

$$\phi(\xi^+ - \mathbf{x}^+) = \phi(R_0) + \tfrac{1}{2}[\mathbf{uu} + 2(m^-/m)\mathbf{u}(\boldsymbol{\mu}_\xi - \boldsymbol{\mu}_x) + (m_-/m)^2(\boldsymbol{\mu}_\xi - \boldsymbol{\mu}_x)(\boldsymbol{\mu}_\xi - \boldsymbol{\mu}_x)] : [\nabla\nabla\phi]_{R_0}, \tag{11}$$

etc. When these expressions are introduced into (6) and (7) and nearest-neighbor interaction assumed ($x = x_n$, $\xi = x_{n-1}$), we obtain

$$\delta\ddot{\mathbf{x}}_n - \omega_{(1)}^2 \overleftrightarrow{\mathbf{C}} \cdot [\Delta^2(\delta\mathbf{x}_n) + \alpha\Delta^2(\boldsymbol{\mu}_n)] = 0, \tag{12}$$

$$\boldsymbol{\mu}_n \times [\ddot{\boldsymbol{\mu}}_n - \alpha^{-1}\omega_{(2)}^2 \overleftrightarrow{\mathbf{C}} \cdot \{\Delta^2(\delta\mathbf{x}_n) + \alpha\Delta^2(\boldsymbol{\mu}_n) + \beta(\boldsymbol{\mu}_n)\}] = 0, \quad |\boldsymbol{\mu}_n| = \mu_0, \tag{13}$$

where

$$\Delta^2(\delta\mathbf{x}_n) = \delta\mathbf{x}_{n+1} - 2\delta\mathbf{x}_n + \delta\mathbf{x}_{n-1}, \quad \overleftrightarrow{\mathbf{C}} = [\nabla\nabla\phi]_{R_0}, \quad \omega_{(1)} = m^{-1/2}(q^+ + q^-),$$
$$\omega_{(2)} = m^{-1}M^{-1/2}(q^+m^- - q^-m^+), \quad \alpha = (M/m)^{1/2}\omega_{(2)}/\omega_{(1)}, \quad \beta = 2q^+q^-/\alpha(q^+ - q^-)^2. \tag{14}$$

Assuming ϕ to depend on distance only, we have

$$\overleftrightarrow{\mathbf{C}} = [\nabla\nabla\phi]_{R_0} = (\phi'/R_0)(\overleftrightarrow{\mathbf{I}} - \mathbf{R}_0\mathbf{R}_0/R_0^2) + (\mathbf{R}_0\mathbf{R}_0/R_0^2)\phi''. \tag{15}$$

Assuming the chain of dipoles to be initially situated along the x axis ($x = x_1$) we have that $\overleftrightarrow{\mathbf{C}}$ is diagonal: $C_{11} = \phi''$, $C_{22} = C_{33} = \phi'/R$. Table I gives values of C_{11} and C_{22} for some choices of ϕ.

If we now confine considerations to two-dimensional displacements and rotations (motion in the x-y plane),

$$\mathbf{R}_0 = (R_0, 0), \quad \mathbf{x}_n = (v_n, w_n), \tag{16}$$

$$\boldsymbol{\mu}_n = (\mu_n^1, \mu_n^2) = \mu_0(\cos\theta_n, \sin\theta_n), \tag{17}$$

the equations of motion become, from (12) and (13),

$$\ddot{v}_n - \omega_{(1)}^2 C_{11}\Delta^2(v_n + \alpha\mu_n^1) = 0, \tag{18}$$

$$\ddot{w}_n - \omega_{(1)}^2 C_{22}\Delta^2(w_n + \alpha\mu_n^2) = 0, \tag{19}$$

$$\mu_n^1\ddot{\mu}_n^2 - \ddot{\mu}_n^1\mu_n^2 + \beta\alpha^{-1}\omega_{(2)}^2(C_{11} - C_{22})\mu_n^1\mu_n^2 + \omega_{(2)}^2[C_{11}(\Delta^2\mu_n^1)\mu_n^2 - C_{22}(\Delta^2\mu_n^2)\mu_n^1]$$
$$+ \alpha^{-1}[C_{11}(\Delta^2 v_n)\mu_n^2 - C_{22}(\Delta^2 w_n)\mu_n^1] = 0. \tag{20}$$

An important special case is obtained for neutral dipoles, for which $q^+ + q^- = 0$ and $\omega_{(1)}^2 = 0$, $v_n = w_n = 0$. Taking the continuum limit, replacing Δ^2 by $R_0^2\partial/\partial x^2$, θ_n by θ, and using (17) in (20), we have

$$\ddot{\theta} - \tfrac{1}{2}R_0^2\omega_{(2)}^2\{[C_{11} + C_{22} + (C_{22} - C_{11})\cos 2\theta]\theta_{xx} + (2/R_0^2)(C_{22} - C_{11})(1 - \tfrac{1}{2}R_0^2\theta_x^2)\sin 2\theta\} = 0. \tag{21}$$

We now rescale the variables and introduce a new constant,

$$x \to 2x/R_0, \quad t \to 2^{1/2}(C_{11} - C_{22})^{1/2}\omega_{(2)}t,$$

$$\beta = 2\theta, \quad \sigma = \frac{C_{11} + C_{22}}{C_{11} - C_{22}},$$

and obtain (21) in a compact form:

$$\frac{\partial^2\beta}{\partial t^2} + (\cos\beta - \sigma)\frac{\partial^2\beta}{\partial x^2} + \left(1 - \frac{\beta x^2}{2}\right)\sin\beta = 0. \tag{22}$$

TABLE I. Diagonal components of C_{ij}.

$\phi(R)$	C_{11}	$C_{22} = C_{33}$	Comments
$1/R$	$2/R_0^3$	$-1/R_0^3$	Coulomb potential
$\ln(R/R)$	$1/R_0^2$	$-1/R_0^2$	Infinite stack of dipoles
$\phi_0(r^{12} - r^6)$ ($r = \bar{R}/R$)	$18\phi_0/R_0^2$	0	Lennard-Jones potential; R_0 taken at the minimum, $2^{1/6}R$

Equation (22) is the equation for (twice) the alignment of a dipole at x when all motions are restricted to the x-y plane. It is our most important result and differs from the ubiquitous sine-Gordon equation by two terms.

For the infinite stack of dipoles ($\sigma = 0$), we are describing the effect of the coupling of infinite, rigid chains (stacks) of dipoles. Each such chain is allowed to move in the plane perpendicular to its axis and is coupled with its two nearest neighbors in this plane. All motion along the chain has been neglected. In the remaining models ($\sigma > 0$), the coupling is between individual dipoles and coupling of neighboring chains is neglected (in particular out-of-plane motion is neglected).

The Lagrangian for (22) is

$$L = \frac{1}{2}\left(\frac{\partial\beta}{\partial t}\right)^2 + \frac{1}{2}(\cos\beta - \sigma)\left(\frac{\partial\beta}{\partial x}\right)^2 + \cos\beta$$

and the two conserved densities following from Noether's theorem are

$$\beta_t L_{\beta_t} - L = \tfrac{1}{2}\dot{\beta}^2 - \tfrac{1}{2}(\sigma - \cos\beta)\beta_x^2 - \cos\beta\,, \quad \beta_x L_{\beta_t} = \beta_x \beta_t\,.$$

The constant σ is $\frac{1}{3}$ for the Coulomb potential, 0 for an infinite stack of dipoles (logarithmic potential), and 1 for all Lennard-Jones-type potentials.

Equation (22) does not pass the Painlevé test as extended to partial differential equations [3]. When the transformation

$$u - \cos(\beta/2)$$

is used, a movable essential singularity appears. Thus, in what follows, we consider our equation to be nonintegrable.

Exact solutions to (22), representing traveling waves and solitons (kinks) are easily found. Take

$$\beta_0 = \beta_0(\xi), \quad \xi = x - vt\,, \tag{23}$$

to obtain

$$(v^2 - \sigma + \cos\beta_0)\frac{d^2\beta_0}{d\xi^2} + (1 - \tfrac{1}{2}\beta_{0\xi}^2)\sin\beta_0 = 0\,. \tag{24}$$

When this equation is multiplied by $d\beta_0/d\xi$ and then integrated, we get

$$\left(\frac{d\beta_0}{d\xi}\right)^2 = \frac{2(C + \cos\beta_0)}{v^2 - \sigma + \cos\beta_0}, \tag{25}$$

$$v^2 - \sigma - 1 > 0, \quad 1 \geq C \geq -1\,.$$

From now on we assume $v^2 - 1 - \sigma > 0$. When $v^2 - \sigma = C$, the solution is $\beta_0 = 2^{1/2}(x - vt) + \bar{\beta}_0$.

In contradistinction to the case for the sine-Gordon equation, we have a lower bound on v for kinks to exist which is $(1+\sigma)^{1/2}$. This, in terms of the original variables, is $R_0 C_1^{1/2}\omega_{(2)}$, and for equal masses and charges in a Coulomb potential,

$$v_{\text{crit}} = 2\,2^{1/2} q/m^{1/2} R_0^{1/2}\,,$$

and for a logarithmic potential, it is this without the factor $2^{1/2}$. Plasma physicists will note the similarity of v_{crit}/R_0 to the plasma frequency.

Phase-plane analysis, in which we draw $(d\beta_0/d\xi)^2$ as a function of β_0 for different possible C, will help us explore the possibilities. For C such that $-1 < C < 1$, a segment of the right-hand side between $\beta_0 = -\arccos(-C)$ and $+\arccos(-C)$ will be non-negative and this corresponds to a periodic wave structure $\beta_0(\xi, C)$. The special case $C = -1$ corresponds to a constant solution $\beta_0 = 0$ and the segment has shrunk to a point. In physical terms, the dipoles are aligned along x. If we perturb around this solution, taking

$$\beta = \delta\beta e^{i(kx - \omega t)}$$

and linearizing, we obtain the dispersion relation

$$\omega^2 = 1 - (1 - \sigma)k^2\,,$$

stable for $\sigma = 1$. If $\sigma < 1$, instability can theoretically set in if $k > (1-\sigma)^{1/2}$. However, the possibility of this is limited by the spacing of the dipoles, $k \ll \pi$.

The most interesting case, however, is $C = 1$. Now the range of β_0 is largest and is $-\pi, \pi$. At the two end points of this interval, we have that

$$\frac{\partial(\beta_{0\xi})^2}{\partial\beta_0} = 0\,.$$

This situation corresponds to a kink (see Chap. 6 of Ref. [4]).

There is also an isolated pair of constant solutions for $C = 1$, $\beta_0 = -\pi$. As $\theta = \beta_0/2$, these solutions correspond to all dipoles perpendicular to the x axis and uniformly polarized. These solutions are in principle unstable, as an analysis similar to the above for $\beta = 0$ will show. This is the case for both $\beta_0 = \pi$ and $\beta_0 = -\pi$, taking $\beta = \beta + \delta\beta$:

$$\omega^2 = -1 + (1 + \sigma)k^2\,.$$

Instability sets in if $k < (1+\sigma)^{-1/2}$. However, small k (long-wave perturbation) may be excluded by the physical limitations of the system.

Here we will just remark that the kink solution mentioned above, to be discussed in more detail in what follows, "connects" these two constant solutions in the far field (say, all dipoles pointing down on the far left $\beta_0 = -\pi$, and all pointing up on the far right $\beta_0 = \pi$).

For the soliton-kink case, $C = 1$, the equation for $\beta_{0\xi}$ as a function of β_0, Eq. (25), can be integrated. We have

$$\left(\frac{d\beta_0}{d\xi}\right)^2 = \frac{4\cos^2(\beta_0/2)}{v^2 - \sigma + 1 - 2\cos^2(\beta_0/2)}, \quad v^2 - \sigma - 1 > 0 \tag{26}$$

(there is also a similar solution for $C = -1$, $v^2 - \sigma + 1 < 0$, but we see from the numerical values of σ that it is unphysical). Thus

$$\frac{d\theta}{d\xi} = \frac{\pm\cos\theta}{(v^2 - \sigma + 1)^{1/2}(1 - k^2\sin\theta)^{1/2}}\,, \qquad k^2 = \frac{2}{v^2 - \sigma + 1} < 1, \quad \theta = \frac{\beta_0}{2}\,. \tag{27}$$

Straightforward integrations yield the solution in parametric form:

$$\xi - \xi_0 = \pm(v^2 - \sigma + 1)^{1/2}\left\{k\arcsin(k\eta) - \tfrac{1}{2}(1 - k^2)^{1/2}\ln\frac{(1-\eta)[1 + k^2\eta + (1-k^2)^{1/2}(1 - k^2\eta^2)^{1/2}]}{(1+\eta)[1 - k^2\eta + (1-k^2)^{1/2}(1 - k^2\eta^2)^{1/2}]}\right\}, \tag{28}$$

$$\theta = \arcsin\eta\,.$$

From a mathematical point of view ξ_0 could be complex, giving some interesting new solutions [5]. However, in our picture θ is an angle in real space and so ξ_0 is real. Take $\xi_0=0$. We have two solutions. One is such that

$$\theta(-\infty)=-\pi/2,\quad \theta(0)=0,\quad \theta(+\infty)=\pi/2\,,$$

and the other is its mirror image [$\xi\rightarrow-\xi$ is a symmetry of (22)]. Thus θ represents a kink, connecting a far-field configuration in which all dipoles are pointing down to one in which all are pointing up, and θ undergoes a half-twist through π. The soliton character of the solution is seen by looking at $d\theta/d\xi$, the steepness of the kink. It is seen from a small-ξ expansion that

$$\theta=\pm\xi/(v^2-\sigma+1)^{1/2},\quad \xi \text{ small}\,, \tag{29}$$

and so $d\theta/d\xi$ is $-(v^2-\sigma-1)^{-1/2}(v^2-1)^{-1/2}$ for the sine-Gordon soliton (see Chap. 7 of Ref. [4]). Thus kinks for which $v^2-\sigma-1$ is small but positive are very steep, the $d\theta/d\xi$ soliton having a large amplitude.

To investigate the in-plane stability of our solution (28), we will not use its exact form, but just the fact that, for $C<1$, $\theta(\xi)$ is periodic. Chapter 8 of Ref. [4] or else Ref. [6] should be consulted for the procedure. Here we will just give the bare outline. We work in ξ,τ coordinates and revert to $\beta_0=2\theta$,

$$\xi=x-vt,\quad \tau=t,\quad \partial_x=\partial_\xi,\quad \partial_t=\partial_\tau-v\partial_\xi\,, \tag{30}$$

and treat the soliton (kink) as a limit of a series of periodic waves with wavelengths tending to infinity as $C\rightarrow 1$. Next we linearize (22) around one of these periodic, nonlinear wave structures. Thus

$$\beta=\beta_0(\xi,C)+\delta\beta(\xi,\tau,C),\quad C<1\,, \tag{31}$$

and $\delta\beta$ satisfies

$$\begin{aligned}[\partial_\tau^2-2v\partial_{\xi\tau}^2+(v^2-\sigma+\cos\beta_0)\partial_\xi^2-\beta_{0\xi}\sin\beta_0\partial_\xi\\ +\cos\beta_0(1-\tfrac{1}{2}\beta_{0\xi}^2)-\beta_{0\xi\xi}\sin\beta_0]\delta\beta=0\,.\end{aligned} \tag{32}$$

We now assume

$$\delta\beta=e^{i(k\xi-\bar\omega\tau)}\overline{\delta\beta}(\xi),\quad \overline{\delta\beta}\ \text{periodic}\,, \tag{33}$$

$$\bar\omega=\omega-kv\,, \tag{34}$$

take k to be small, and expand $\bar\omega$ and $\overline{\delta\beta}$; thus,

$$\bar\omega=\bar\omega_1 k+\bar\omega_2 k^2+\cdots,\quad \overline{\delta\beta}=\overline{\delta\beta}_0+k\overline{\delta\beta}_1+\cdots\,.$$

This procedure leads to a hierarchy of equations. In zero order we find $\overline{\delta\beta}_0=d\beta_0/d\xi$. First order gives $\overline{\delta\beta}_1$ in terms of known functions and $\bar\omega_1$. In second order we obtain a consistency condition for the removal of secular terms. This condition is, in the laboratory system and without the order suffix,

$$\begin{aligned}\omega^{(1,2)}/k=v\pm\alpha(1-C)^{1/2}\ln(1-C)\\ +(\gamma_r+i\gamma_i)(1-C)[\ln(1-C)]^2\cdots\,.\end{aligned}$$

Here $1-C$ is assumed small at the very end of the calculation and α, γ_r, and γ_i are constants involving v and $v^2-\sigma$ but not C. Thus in the soliton-kink limit $C\rightarrow 1$, we have $\omega^{(1,2)}\rightarrow kv$, and this proves marginal stability with respect to long-wave and slow perturbations. The nonlinear waves, for which $C<1$, are unstable [4,6,7].

For $\sigma=0$ (infinite line dipoles) we can extend (22) phenomenologically, substituting $\partial_x^2+\partial_z^2$ and $\beta_x^2+\beta_z^2$ such as to include out-of-plane, z dependence of β (which is still planar). The basic kink structure remains the same, but in the stability analysis we now have $k^2=k_\xi^2+k_z^2$. Stability is still obtained in the kink limit.

The whole stability analysis outlined above is limited to small k. A general analysis for arbitrary k has not been performed for this problem. Previous work on mathematically similar problems [8] leads us to expect that, although in theory these equilibria tend to be unstable, in fact growth rates tend to zero in the kink limit.

A completely different problem, not considered by us, is that of stability with respect to out-of-plane perturbations when our dipole chain is a component of a full, three-dimensional crystal structure and $\sigma=0$. When interactions between chains could introduce new instabilities, our analysis should be considered as just a first step towards treating the full problem; however, it is quite apt as it stands when treating the single-chain problem.

In any case, even when considering a crystal, a proper understanding of the dynamics of one chain must precede a full, three-dimensional array treatment.

In conclusion, ferroelectric crystals, one-dimensional organic polymer chains, and other ordered structures can be studied at the molecular level by looking at a simple dipole chain model.

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