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Vortex arrays and Landau–Ginzburg modelling of the onset of coherence in ceramic superconductors

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Abstract. A Landau–Ginzburg formulation is presented to describe a series of superconducting islands interacting via Josephson weak links in the presence of a magnetic field. It is demonstrated that the structure of superconducting islands (and the lattice of magnetic vortices), described by the order parameter's envelope, may be commensurate or incommensurate with a superimposed array of weak links via the order parameter's phase for each island. A discussion is provided in terms of a Kosterlitz–Thouless transition in the latter structure. A description of space modulation of these phases is given through sine–Gordon equations.

1. Introduction

Since the discovery of high-temperature superconductors by Bednorz and Müller (1986) a substantial amount of experimental knowledge about these ceramic superconductors has been accumulated leading to further progress in obtaining higher transition temperatures (Ehrenreich and Turnbull 1989). Proliferation of theoretical models has also been in evidence but so far there appears to be no consensus on the nature and type of mechanisms responsible for these phenomena. There are a number of intriguing features that appear to be in contrast with those of low-temperature standard superconductors. Obviously, the 1–2–3 compounds (Y–Ba–Cu–O) and La–R–Cu–O materials possess a number of specific features. Nevertheless, the main qualitative behaviour is quite common in both cases. In both these compounds it is crucial to emphasise the effects leading to the existence of oxygen vacancies, which seem to be mainly concentrated in the Cu–O planes. Below the critical temperature, they have been found to form a regular lattice (Inoue *et al* 1987, Aligia *et al* 1988, Schmahl *et al* 1988). It is also important to note that the phase diagrams of these compounds have regions of an insulating antiferromagnetic phase (Hass 1989) as well as a spin glass phase (Aharony *et al* 1988). It appears that, due to doping, mobile charge carriers are first produced, which may subsequently form a conduction band and, eventually, through strong interactions, a superconducting phase. Hall effect measurements unambiguously indicate that these charge carriers are holes (Ong *et al* 1987, Uchida *et al* 1987) in most of the compounds investigated.

Very important differences between the ceramic and standard superconductors are exemplified by the characteristic length scales associated with the two types of system. In the new superconductors the coherence length in the *ab* plane is ≃30–50 Å, while

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along the c axis it may be 5–8 Å or less, and the penetration depth varies from 1500 to 2500 Å (Aeppli *et al* 1987). In standard superconductors the coherence length is much longer and the penetration depth substantially shorter (White and Geballe 1979). The Meissner effect experiments seem to reveal that the new superconductors are not ideal diamagnets but expel up to 80% of the external magnetic field, thus indicating that the superconducting phase is either a mixture of coexisting phases or a modulated type of superconducting phase in which the order parameter is highly position dependent (Golovashkin 1987). In addition to the levitation effects found in standard superconductors, high-T_c ceramics exhibit effects of suspension in a magnetic field (Gregory and Johnson 1988). This indicates that the partial penetration inside the sample may be thermodynamically favourable and such a state may have inherent stability.

Furthermore, a number of structural effects are worth mentioning. The ceramic superconductors are highly anisotropic with a copper–oxygen structure giving rise to either two-dimensional rectangular lattices or a quasi-one-dimensional chain-like structure in other layers (Beyers and Shaw 1989). As a consequence of the anisotropy, much lower conductivity along the c axis is exhibited in both the normal and the superconducting state. Thus, it is believed that the planes formed by Cu and O are responsible for the superconducting properties of the crystals. Other structural anomalies are also present. These are evident from experiments that measure an anomalous thermal expansion coefficient as a function of temperature, strong pressure dependence of the critical temperature in the a–b plane and a nearly negligible effect along the c axis (Karpinski *et al* 1989, Golovashkin 1987). The granular nature of the new superconductors can be seen through a number of properties: anomalous voltage excursions as a function of temperature and magnetic field (Cai *et al* 1987), logarithmic time decays of magnetisation (Mota *et al* 1988), flux trapping (Müller *et al* 1987) and tails of resistivity versus temperature due to boundary resistance between grains (Tsuneto 1988). The list of experimental facts and observations is very long at the present time and for a more detailed account the reader is referred to extensive reviews.

Based on the above observations we wish to explore the following possibility for the onset of bulk coherence. The effect appears to be gradual and associated with a broad (over 20–30 K) temperature range over which resistivity drops to zero. The theoretical model that we wish to put forward is based on the existence of and the role played by the oxygen vacancies. The depletion or excess of oxygen ions first of all leads to the creation of additional charge carriers (Dharma-Wardana 1987). Since the latter are holes with a high effective mass, compared with that of electrons, the degree of localisation of excess charge is expected to be much greater than in standard superconductors. Moreover, the underlying electrostatic potential of the crystal lattice may play a greater role in such effects as pinning.

Our initial step is to follow the ideas of Deutscher and Müller (1987) and envisage regions of a size, at least initially, smaller than the mean grain size. Their average size and spacing can be affected by such factors as temperature, impurities, magnetic fields and currents. We shall investigate these factors separately in later sections.

2. The model

Deutscher and Müller (1987) pioneered the investigations into glassy behaviour of high-T_c superconductors. They pointed to a number of properties that appear to be due to a glassy state: microwave response of point contacts, extrinsic critical currents in single crystals, gapless characteristics of tunnelling spectroscopy results, etc. They proposed a model based on the typical Hamiltonian for superconductor–insulator–superconductor (SIS) junctions, i.e.

$$H_0 = -\sum_{ij} J_{ij} \cos(\varphi_i - \varphi_j - A_{ij}) \tag{1}$$

where A_{ij} is a phase factor due to the presence of external magnetic fields which is responsible for randomness and frustration and φ_i is the phase of the local order parameter. Here, the coupling constant for SIS structures is given by the Ambegaokar–Baratoff (1963) formula

$$J_{ij} = \frac{\hbar}{2e} \frac{\pi \Delta(T)}{2e\rho d_{ij}} \tanh\left(\frac{\Delta(T)}{2kT}\right) \tag{2}$$

where $\Delta(T)$ is the energy gap, ρ is the normal state resistivity and d_{ij} is the island–island separation. They stress that behaviour consistent with that predicted by the model has been seen in the I–V characteristics of such Josephson junctions (Estevé *et al* 1987). They also emphasise that the observed logarithmic time dependence of magnetisation decay confirms the glassy state hypothesis. A very crucial role in the formation of a network of coupled superconducting domains is played by the short coherence length. Subsequently, Sahling and Sahling (1989) observed glassy behaviour in 1–2–3 compounds through heat release experiments with a broad distribution of relaxation times typical of amorphous solids and structural glasses. A very strong argument in support of the glassy behaviour model has been recently given by Koziol *et al* (1989) who maintain that, in addition to T_c, there is another transition temperature T_J related to the occurrence of bulk superconductivity induced by Josephson junction interactions. Below T_J quantum phases of local order parameters freeze and the Hamiltonian, equation (1), becomes inadequate to describe the system. They also maintain that the presence of frustration results in a pinning potential. Energy barriers must be overcome in order to generate flux motion. The existence of T_J and its smooth decrease with the applied field H has been experimentally demonstrated by Barbara *et al* (1988) through measurements of the imaginary part of the susceptibility function.

A very elaborate calculation of a granular model of a superconductor based on (1) was recently published by Fishman (1989) who concentrated on phase fluctuations using a $1/z$ expansion technique and concluded that correlations of phase fluctuations enhance short-range order of granular superconductors.

To balance this, so far one-sided, discussion on the merits of the glassy model we should mention opposite views expressed by Malozemoff *et al* (1988) who believe that a flux pinning picture rather than the spin glass one is more adequate to explain remanent magnetic moment measurements. They do admit, however, that a strong glassy flux pinning is also possible to explain the observations; this seems to be confirmed by Koziol *et al* (1989). In fact, it has been experimentally confirmed that boundaries between grains act like Josephson junctions (Chaudhari *et al* 1988). In this connection Stankowski *et al* (1987) found, through EPR studies, that Josephson loops of sizes in the range of 0.65–0.81 μm should exist in the 1–2–3 compound; in order to explain the observed oscillations in the spectra. Our model will be principally based on the Landau–Ginzburg (LG) theory since, in the past, this has provided a unified and extensive description of various aspects of the superconducting phenomenon. We do not intend to speculate on the particular mechanisms that may bring about superconductivity within each island but, rather, point to the importance of short-range interactions which may, at least initially, play the dominant role.

The LG model has already been rather fruitfully employed in the investigations of various aspects of high-temperature superconductivity such as, e.g., twinning (Danilov and Safonov 1988), multilayered structures (Eab and Tang 1988, Theodorakis and Tesanovic 1988, Tarento 1988), coupled order parameters (Chela-Flores *et al* 1988, Choy *et al* 1988) or even the connection to the RVB model (Nakamura and Matsui 1988). In addition, a direct connection has been recently found between a microscopic second-quantised Hamiltonian and the LG order parameter picture (Tuszynski and Dixon 1989a, b). Thus, our present modelling may help in future *ab initio* calculations.

The theoretical framework within which we construct our model involves assigning a complex superconducting wavefunction as the (localised) order parameter $\psi_i(r_i)$ for each island, centred at the position r_i, with its own LG free-energy expansion. The islands are then allowed to interact via weak-link Josephson junctions using the Lawrence–Doniach (LD) (1971) expression which is slightly more general than the form in (1). We will subsequently discuss the minimisation of the postulated free energy under various distinct conditions, such as the presence or absence of magnetic fields and the effect of superconducting currents, in order to find the critical values of these two quantities. We will also draw extensively on a number of interesting connections linking the XY model, the Kosterlitz–Thouless (KT) model and the LG model in the continuum limit. In contrast to similar analyses done in the past, we intend to concentrate on non-linear aspects inherent in the model.

The starting point for our development is the adoption of the idea of Deutscher and Müller (1987) that domains of superconductivity are being formed throughout the sample. These regions of superconductivity may have finite spatial dimensions and can be separated by regions of normal phase. We shall refer to these superconducting domains as islands whose spatial array structure can perhaps be visualised as being stabilised by the oxygen defect/excess (Dharma-Wardana 1987). Since, at least initially, i.e. close to the onset of superconductivity, these islands are well separated, the interactions between them are rather weak and may be modelled using the weak-link approximation as in arrays of Josephson junctions (Martinoli *et al* 1987).

Based on the general information above we postulate to represent the free-energy density of a ceramic material, in the vicinity of its critical temperature, as the sum of individual LG terms for each of the islands i with interactions between them in the LD form. We realise that the LD term has been initially employed to describe interactions between superconducting layers. However, it is applicable also to arrays of Josephson junctions within a plane (Raboutou *et al* 1987, Choi and Doniach 1985, Shih *et al* 1984). In general, it appears in situations where superconducting grains are well separated by regions of normal order. The free energy f is therefore represented as the sum of three terms below

$$f = f_1 + f_2 + f_3 \tag{3}$$

where

$$f_1 = \sum_n f_{1n} = \sum_n \left[A_2 |\psi_n|^2 + A_4 |\psi_n|^4 + \frac{\hbar^2}{2m^*} \left| \left(-\mathrm{i}\nabla_n - \frac{2e}{\hbar c} A \right) \psi_n \right|^2 \right] \tag{3a}$$

$$f_2 = \tfrac{1}{2} \sum_{nl} b_{nl} \left| \psi_n \exp\left(-\frac{2e\mathrm{i}}{\hbar c} \int_n^l A \cdot \mathrm{d}l \right) - \psi_l \right|^2 \tag{3b}$$

and

$$f_3 = H^2/8\pi \tag{3c}$$

where the first term f_{1n} is a standard Ginzburg–Landau (1950) free-energy expansion for the local order parameter characterising the nth island. The spatial extent of the order parameter $\psi_n = \psi_n(r_n)$ is restricted to be within the distance between the impurities and is therefore dependent on doping concentration, x. The coefficient A_4 in f_1 is assumed to be only weakly temperature dependent so we can take it to be constant. The second term, f_2, i.e. in (3b), is in the form of the LD interaction where the path integral involves a trajectory linking the centres of the two islands n and l and A is the spatially dependent vector potential. The parameter b_{nl} is a distance-dependent intergrain coupling constant. Following Landau and Lifshitz (1980) we assume that $A_2 = a(T - T_c)$ where a is positive and we assume this is independent of the particular island. In the interaction terms, i.e. f_2, the temperature dependence would usually enter through an entropy term due to the possible distributions of vortices. However, we shall assume initially that the vortices are pinned in such a way that f_2 becomes virtually temperature independent.

To simplify the problem we make a number of physically motivated approximations. First, representing ψ_n in the usual way as $\psi_n = \eta_n \exp(i\varphi_n)$ we assume the amplitudes η_n, for each island, to be virtually identical, i.e. $\eta_n = \eta_m \equiv \eta$, but not necessarily homogeneous, resulting in a sort of periodicity requirement. Thus, both η_n and φ_n are seen as localised functions of their position variable r_n (centred at different points in space) which tend to their asymptotic values on the nth island's boundary. We also assume that only the nearest neighbour islands are effectively interacting and that the phase difference between them is sufficiently small to invoke the so-called weak-link approximation. As a consequence of these assumptions the free energy becomes

$$f_1 = \left(A_2 + \frac{2e^2A^2}{m^*c^2}\right) N\eta^2 + A_4 N\eta^4 + \frac{\hbar^2}{2m^*} N|\nabla\eta|^2 + \frac{\hbar^2}{2m^*}\sum_n \left(|\nabla\varphi_n|^2 - \frac{4eA}{\hbar c}\cdot\nabla\varphi_n\right)\eta^2 \tag{4}$$

$$f_2 = b\eta^2 \sum_{\langle n,l\rangle}\left[1 - \cos\left(\varphi_n - \varphi_l - \frac{2e}{\hbar c}\int_n^l A\cdot dl\right)\right] \tag{5}$$

where b is the value of b_{nl} for nearest neighbour interactions and the sum over $\langle n, l\rangle$ is for nearest neighbours only. Minimising the free-energy functional

$$F = \int (f_1 + f_2 + f_3)\, d^3r$$

with respect to η, φ and A we obtain

$$\frac{\delta F}{\delta\eta} = 0 = 2N\left(A_2 + \frac{2e^2A^2}{m^*c^2}\right)\eta + 4A_4N\eta^3 - \frac{\hbar^2}{m^*}N\nabla^2\eta + \frac{\hbar^2}{m^*}\eta\sum_n\left(|\nabla\varphi_n|^2 - \frac{4eA}{\hbar c}\cdot\nabla\varphi_n\right) + 2b\eta\sum_{\langle n,l\rangle}\left[1 - \cos\left(\varphi_n - \varphi_l - \frac{2e}{\hbar c}\int_n^l A\cdot dl\right)\right] \tag{6}$$

$$\frac{\delta F}{\delta\varphi_n} = 0 = -\frac{\hbar^2}{m^*}\eta^2\nabla^2\varphi_n + \eta^2\frac{\hbar e}{m^*c}\nabla\cdot A + b\eta^2 \times \sum_{\langle l\neq n\rangle}\sin\left(\varphi_n - \varphi_l - \frac{2e}{\hbar c}\int_n^l A\cdot dl\right) \tag{7}$$

$$\frac{\delta F}{\delta A} = 0 = \frac{4e^2 A}{m^* c^2} N\eta^2 - \frac{\hbar e}{m^* c}\eta^2 \sum_n \nabla\varphi_n - \frac{2e}{\hbar c} b\eta^2 \times \sum_{\langle n,l\rangle} \hat{d}_{nl} \sin\left(\varphi_n - \varphi_l - \frac{2e}{\hbar c}\int_n^l A\cdot \mathrm{d}l\right) + \nabla\times(\nabla\times A) \tag{8}$$

where $\hat{d}_{nl}$ is the unit vector from site n to site l.

In order to simplify this set of coupled equations we can choose a gauge in such a way that $\nabla\cdot A = 0$. In addition, we replace the integral

$$\frac{2e}{\hbar c}\int A\cdot \mathrm{d}l$$

by a function $\varphi_0(r)$ whose numerical value depends on the distance between islands as well as the nature of the magnetic field penetration.

3. The approximation of non-interacting islands

In order to understand properly the full complexity of the problem described by (6)–(8) we shall first briefly review the situation in the absence of interactions between superconducting islands, i.e. assume $b = 0$. Our interest here is in a small and isolated region of space where the superconducting order nucleates without the inclusion of interactions with other such domains of superconductivity. We assume here that these domains are sufficiently far apart and superconductivity weak enough to ignore any correlation effects between them. The two generic situations to consider in this context are the onset of superconductivity in a single island in the absence of external magnetic fields and the problem of magnetic field penetration of a superconducting domain. These two cases have been recently analysed in detail by Tuszynski and Dixon (1989a, b) and Vos *et al* (1990). As one of these papers has not, as yet, been published a short account of their main results is given below. The first case allows for either quasi-linear or cylindrical solutions of the equations of state assuming that the temperature is below T_c. At and close to $T = T_c$ spiral order parameter structures can be found in addition to those above. Within this general description we also found two special forms of solution depending on the presence or absence of superconducting currents in the sample. The general form of the envelope represents an elliptic function whose type depends on the temperature region, the magnitude of the superconducting current and the sign of the effective mass m^*. It is important to note that when $m^* > 0$ the lowest-energy solution is a mean field separated by a gap from the next lowest state which is localised (either a 'kink' for zero current or a 'cusp' otherwise). These solutions are then followed by a continuum of elliptic waves. On the other hand, when $m^* < 0$, the situation is completely different and can be interpreted as a modulational instability—that is, the lowest-energy solution is a periodic function with the highest frequency and amplitude allowed by the crystal structure. An expression for the associated period λ has also been given in terms of the free-energy expansion coefficients and an integration constant. In general, this expression involves an elliptic integral. However, the periodicity of the 'cut-off' solution (lowest energy) corresponds to twice the lattice spacing d_0 of the oxygen vacancy network, $\lambda_{\min} = 2d_0$. These solution forms happen to be asymptotically correct in the two-dimensional case. The behaviour of the solutions close to the centre of an island can be found by linearisation about the mean-field solution. It is worth emphasising that this approach also seems to predict the correct temperature dependence of the critical current density $J_c(T) \propto (T - T_c)^{3/3}$ in standard superconductors and gives a variety of possible forms of $J_c(T)$ for high-T_c superconductors.

The second case of magnetic field penetration is much more difficult to study using exact methods. Nevertheless, it has been possible to demonstrate the existence of quasi-linear and cylindrical solutions (vortices) for arbitrary tempratures below T_c as well as spiral metastable solutions in the vicinity of T_c. The quasi-linear solutions are generally expressed in terms of elliptic functions with the limiting cases of 'bumps' and 'kinks' referring to nucleation centres and magnetic field penetration, respectively. The physical interpretation of the elliptic-type solutions for both the order parameter's envelope η and the vector potential A is the setting up of magnetic vortices in a periodic array with vortex cores coinciding with the envelope's zeros with the intensity of the magnetic field decaying radially from their centres. The solutions discussed here are obtained in free space and boundary conditions may then be conveniently imposed to select such solutions that satisfy the desired boundary requirements. For detail calculations and analysis the reader is referred to the two papers mentioned above.

4. Preliminary discussion of the role of island–island interactions

In the previous section we have discussed the role of f_1 and f_3 assuming that the superconducting islands are either non-interacting or that there is only one such island in the system. In the present section we wish to address the question of the role played by term f_2 responsible for island–island interactions. In the first instance we will simply review the theoretical results concerning the vortex dynamics in two-dimensional arrays of weak links as described by term f_2 alone. In the next step we shall investigate the effective influence of island ordering on the remaining degrees of freedom.

There exists a large body of literature that deals with the analysis of superconducting weak links based on the interaction energy of (5) (Martinoli *et al* (1987) and references therein). First of all, in zero magnetic field the nearest neighbour interaction energy between a pair of islands n and l is given by

$$E_{nl} = (\hbar i_c(T)/2c)[1 - \cos(\varphi_l - \varphi_n)] \tag{9}$$

where $i_c(T)$ is the critical current of the isolated junction in the absence of thermal fluctuations (directly proportional to the square of the order parameter's envelope). It is most interesting that (9), when summed over i and j, maps directly onto the XY model for classical spins with a temperature-dependent exchange constant $\bar{J} \equiv \hbar i_c(T)/2e$. The latter model has been extensively studied by Kosterlitz and Thouless (1973). The main result of their analysis is the possibility of a new type of phase transition occurring at $T = T_J$ given by

$$T_J \simeq \pi J/1.12\, k_B. \tag{10}$$

With T_c denoting the temperature at which a single island becomes superconducting, and, making an approximation consistent with the assumption made in (9) that

$$i_c(T) \simeq i_c^0(T_c - T) \tag{11}$$

we find our expression giving T_J in terms of T_c:

$$T_J = [\alpha/(1 + \alpha)]T_c \tag{12}$$

where $\alpha = (\hbar i_c^0)/(4.48e\, k_B)$. The choice of symbol T_J for the ordering transition temperature is dictated by the apparent similarity with the phenomenon discussed by Koziol *et al* (1989). If α is sufficiently large the two transitions may coincide at $T \simeq T_c$. The first transition leads to the nucleation of superconducting islands and the second to the creation of topological order in the phase of the order parameter. This latter situation is realised by the presence of bound-vortex–antivortex pairs separated by a distance r. Then the energy of a vortex–antivortex pair is given by KT as

$$U(r) = (\hbar i_c(T)/2e) \ln(r/a). \tag{13}$$

Hence, above T_J the superconducting islands possess uncorrelated phases whereas below T_J their phase geometry can be described by a vortex–antivortex regular lattice structure. Note that the relationship in (13) implies that making b temperature dependent, so that $b = \beta(T - T_J)$, effectively incorporates the seemingly missing entropy term in f_2 as can be seen from (10) and (11) of KT. It is also important to note that the periodicity of the vortex–antivortex array (based on the order parameter's phases φ_i) and that of the island lattice structure (related to the order parameter envelope η) may not be the same as or even commensurate with each other as we shall explore later in the paper. As $i_c(T)$ is a local current that is a decreasing function of r, $U(r)$ in (13) remains finite. Thus, our model is free from the undesired divergence of the flux energy. Although in their original paper KT precluded the application of their model to superconductors, their argument was based on two main assumptions. The first was the use of the mean-field description of the superconducting envelope and the second that the interaction referred to magnetic flux lines penetrating the superconducting phase. We hope to apply our approach to the new high-T_c superconductors where the use of a mean-field description is highly questionable because of the short coherence length. The interaction term refers here, not to magnetic vortices, but rather the phases of the order parameter field φ_i centred at different islands. In this respect this is similar to the earlier work on arrays of Josephson junctions (Martinoli *et al* 1987). Furthermore, a similar picture has already been advocated for the explanation of glassy behaviour in high-T_c superconductors (Cai *et al* 1987, Tsai *et al* 1987, Schneider *et al* 1988, Morgenstern *et al* 1988, Kampf and Schön 1988, Sonin 1988). Recently, Zwerger (1987a, b, 1988) studied networks of Josephson junctions including quantum fluctuations and dissipation and found the possibility of two superconducting phases: one with long-range order and the other with local phase coherence.

In the presence of a perpendicular magnetic field $\boldsymbol{B}$ the interaction energy of a pair of weak-linked islands is

$$E_{nl} = (\hbar i_c(T)/2e)[1 - \cos(\varphi_l - \varphi_n - A_{nl})] \tag{14}$$

where A_{nl} is proportional to the line integral of the vector potential $\boldsymbol{A}$ from site n to site l. This form of interaction is isomorphic to a frustrated XY (spin glass) model with a number of interesting properties (Ebner and Stroud 1985). In particular, for a square lattice, the superconducting current distribution in the ground state consists of a series of staircase supercurrents (Hasley 1985). Moreover, the vortex lattice induced by the magnetic field interacts with the pinning potential created by the periodic array of islands. With the degree of frustration defined by $\sigma = \Phi/\Phi_0$, where Φ is the magnetic flux for a unit cell and Φ_0 is the superconducting flux quantum, there are competing periodicities related to the fixed array structure and the vortex structure determined by σ. This leads to a sequence of commensurate and incommensurate vortex phases. The former ones are characterised by the formation of a superlattice. At low temperatures the commensurate phases are pinned by the periodic potential due to the array structure, in contrast to the incommensurate phase in which the vortex lattice is free to slide. This latter property is characteristic of glassy behaviour which was attributed to high-T_c

superconductors and discussed in the frame of the frustrated XY model by Morgenstern *et al* (1988) and Stroud and Ebner (1988). Stroud and Ebner (1988) also discovered the possibility of a transition between a spin glass and a ferromagnetic phase within this model induced by a sufficiently large magnetic field.

In these papers several of the properties obtained appear to be in qualitative agreement with experiment. These include the presence of a spin glass phase on the phase diagram with its property of irreversibility, the field dependence of the critical line and the temperature dependence of the susceptibility. We add to this list the results of experimental papers showing frequency jumps on voltage–temperature plots for varying magnetic fields, the jump frequency being largest at low temperatures and disappearing at high fields (Cai *et al* 1987, Tsai *et al* 1987). This seems to be consistent with the frustrated XY model picture. Some authors (Cai *et al* 1987, Teitel and Jayaprakash 1983) have suggested that the effective coupling constant, b_{ij}, varies as $\exp(-\mu d_{nl}T)$ where μ is a constant and d_{nl} is the distance between sites n and l, implying the correct behaviour at low temperatures. Experiments on the irreversible magnetisation of the 1–2–3 compounds have been interpreted as indicating the existence of intrinsic geometrical boundaries for the critical currents, which could be related to the decomposition of these non-stoichiometric crystals in regions of high and low oxygen concentration (Sulpice *et al* 1988). This might be used to strengthen our argument for the relationship between the doping fraction and the size and separation between the superconducting islands.

5. Further insights into spatial modulation

We first assume that f_1 and f_3 in the free energy are by far the most dominant as they define the array of superconducting islands separated by magnetic vortices. This structure has been analysed in an earlier paper by Vos *et al* (1990) where it has been demonstrated that in the absence of island–island interactions and assuming the magnetic induction $\boldsymbol{B}(x)$ to have orientation normal to the sample along x, the order parameter envelope can be expressed as

$$\eta = (\alpha \mathrm{E}(\tau x, k) + \beta)/(\gamma \mathrm{E}(\tau x, k) + \delta) \tag{15}$$

where α, β, γ, δ, τ are appropriately chosen constants and E is any one of the elliptic functions sn, cn or dn. The corresponding vector potential must be linearly related to η for proper compatibility of the equations involved. Therefore, the most important conclusion from this analysis is that both the superconducting order parameter's envelope η and the vector potential $\boldsymbol{A}$ (and hence the magnetic induction $\boldsymbol{B}$) may be periodic elliptic functions with the *same* periodicity. To account properly for the Meissner effect the two quantities should be phase shifted with respect to each other. Moreover, if the effective mass m^* is negative then the lowest-energy solution is chosen as the Jacobi cn function with the shortest wavelength allowed, i.e. corresponding to the defect/excess oxygen mean separation. This then defines the primary (non-topological) structure of the superconductor. The inclusion of weak-linked *phase* interactions can be done through the analysis of (7) taken alone with $\boldsymbol{A}$ determined by (6) and (8) as outlined above. The solution of (7) in terms of φ_i may then be fed back into (6) and (8) as an effective dressing term. In particular, we note that the $\nabla\varphi_i$ terms in (4) are multiplied by η^2 and therefore can be reinterpreted as redefining the local temperature via the first term in (4). More specifically, in regions where $\nabla\varphi_i$ is large, i.e. at the edges of the ith island, the effect will be to diminish the envelope while inside the island the effect will be virtually negligible.

We now analyse (7) in more detail in order to investigate the ordering effect in *phase space*. First of all, we can divide (7) by η^2 assuming that $\eta \neq 0$ as is the case in a superconducting region, and, as already stated, use a gauge in which $\nabla \cdot A = 0$. Assuming that φ_0 is an integer multiple of 2π, i.e. $\varphi_0 = 2n\pi$, when $b < 0$ or has the form $\varphi_0 = (2n+1)\pi$ when $b > 0$, then (7) takes a special form of the sine–lattice equation (Takeno and Homma 1986), i.e.

$$\sin(u_{n+1} - u_n) - \sin(u_n - u_{n-1}) - \ddot{u}_n = g \sin u_n \tag{16}$$

with $g = 0$ where $u_n = \varphi_n$ and is a real dependent variable depending on an island label n and an independent variable z found by scaling (7) appropriately. This, of course, is a one-dimensional approximation to the problem, which may be of special importance in strongly anisotropic situations. The so-called SLO equations, which is (16) with $g = 0$, have been recently studied by Homma (1987) who found that the approximate π- and 2π-kink solutions are

$$u_n(\Lambda) = \begin{cases} 2\tan^{-1}(\exp(\Lambda)) & \pi\text{-kink} \\ 4\tan^{-1}(\exp(\Lambda)) & 2\pi\text{-kink} \end{cases} \tag{17}$$

with $\Lambda = kx + \omega z + \Lambda_0$ and the φ_n is the phase at the nth island at position x where k and ω satisfy the dispersion relation

$$\omega^2 = 4\sinh^2(k/2). \tag{18}$$

These π-kink solutions have been found to be stable and possess soliton-like qualities on collision. The 2π-kinks eventually break up into a pair of oppositely moving π-kinks.

In the next stage of the development we notice that, assuming a sort of periodicity condition, i.e. that each neighbouring phase $\varphi_l = \nu\varphi_n$ where ν is a constant other than unity (in particular, $\nu = -1$ if we expect a vortex-antivortex lattice), and with the same assumptions as before for φ_0, equation (7) takes the form of a sine–Gordon equation (SGE). Hence

$$-(\hbar^2/m^*)\nabla^2\varphi_n + bN\sin(1-\nu)\varphi_n = 0 \tag{19}$$

where N is the number of nearest neighbours. The extent of the spatial argument of φ_n is limited to within the nth island. This may be clearly rewritten, by choosing a new dependent variable $(1-\nu)\varphi_n = \varphi$ and scaling the independent variables appropriately, as

$$\nabla^2\varphi = \sin\varphi. \tag{20}$$

Equation (20) is a two-dimensional, time-independent version of an SGE whose standard form represents an integrable non-linear equation. The solutions of the latter have been well investigated in the past (see for example Dodd *et al* 1982). The multi-dimensional SGE has been analysed using the symmetry reduction method by Grundland *et al* (1982). Among the reductions obtained we find two which are also applicable in our case, namely the quasi-linear variable $\xi = \alpha x + \beta y$ and the radial variable $\rho = \sqrt{x^2 + y^2}$. In the first case a complete set of solutions can be obtained by direct integration and the most interesting solutions are in the form of 'kinks'. The one-kink solution is given by

$$\varphi(\xi) = 4\tan^{-1}(\exp\xi). \tag{21}$$

More importantly for our purposes an N-kink solution can be found following Caudrey *et al* (1975) and takes the form

$$\varphi = \cos^{-1}[1 - 2(\mathrm{d}^2/\mathrm{d}\xi^2)\ln f(\xi)] \tag{22}$$

where $f(\xi) = \det|\mathbf{M}|$ and $\mathbf{M}$ is the $N \times N$ matrix whose elements are

$$M_{ij} = [2/(a_i + a_j)] \cosh[(\theta_i + \theta_j)/2]. \tag{23}$$

The coefficients are $a_i = \mp 1$, $\theta_i = \pm \xi + \xi_{0i}$ and ξ_{0i} is an arbitrary constant. This solution represents N well separated kinks with an arbitrary distribution of asymptotic values and it could be taken as a global solution to our problem provided it is chosen in such a way as to satisfy our initial requirement that $\varphi_l = \nu\varphi_n$. As we shall see later this phenomenon is closely related to multiple-stepped staircases.

The other reduction, $\varphi = \varphi(\rho)$, leads to the equation

$$\varphi_{\rho\rho} + (1/\rho)\varphi_\rho = \sin\varphi \tag{24}$$

which has the same asymptotic behaviour as in the previous case and close to the origin it can be conveniently analysed by expanding it about a mean-field solution to obtain a Bessel equation. For an in-depth analysis of this equation the reader is referred to a recent paper by Malomed (1987). To conclude this short review of the solutions of (20) we see that the phase pattern of the array of weak links in the absence of magnetic fields can be looked upon as a two-dimensional structure with staircase properties given by the arrangement of multi-kink solutions. It should be emphasised that these solutions may indeed exhibit periodicity which may or may not be commensurate with the periodicity of the underlying islands (order parameter's envelope structure). The interplay between these two distinct structures and their periodicities will be the subject of our discussion that follows. Obviously, the global solutions obtained here must be eventually subjected to the consistency check that $\phi_1 = \nu\phi_n$ as stipulated earlier.

We now intend to examine (7) with the presence of magnetic fields. Allowing for spatial inhomogeneities on going from site n to l we may write the argument of the sine in (7) as

$$(1-\nu)\phi_n + \mu\hat{\boldsymbol{d}}_{nl}\cdot\nabla\phi_n - \phi_0(r) \tag{25}$$

so we may approximate and write (7) assuming that $\mu\hat{\boldsymbol{d}}_{nl}\cdot\nabla\phi_n$ is relatively small as

$$(\hbar^2/m^*)\nabla^2\phi_n \simeq Nb\sin[(1-\nu)\phi_n] + (\mu\hat{\boldsymbol{d}}_{nl}\cdot\nabla\phi_n - \phi_0)\,Nb\cos[(1-\nu)\phi_n] \tag{26}$$

where again N is the number of nearest neighbours. On rescaling dependent and independent variables we can recast (26) in the form

$$\nabla^2\phi = \sin\phi + (\boldsymbol{\gamma}\cdot\nabla\phi - \phi_0)\cos\phi \simeq \sin\phi + \boldsymbol{\gamma}\cdot\nabla\phi \qquad \text{for } \phi \simeq 0 \tag{27}$$

where the damping constant $\boldsymbol{\gamma} = +\mu\sqrt{(m^*Nb)/[\hbar^2(1-\nu)]}\,\hat{\boldsymbol{d}}_{nl}$. Since $\phi_0(r)$ is related to $\boldsymbol{A}(\boldsymbol{r})$, it basically has the periodicity of the array of islands which is the same as the magnetic vortex periodicity (η and $\boldsymbol{A}$ are linearly dependent due to compatibility relations used by Vos *et al* 1990). Equation (27) is a damped driven SGE and the reader is referred to the following papers for details, Lawrence *et al* (1985), Lomdahl (1985), Büttiker (1986) and Overman *et al* (1984). The periodicity of the phase field is, of course, independent of that of the islands and may or may not be commensurate with it. If it happens to be commensurate then the effects of driving, due to $\phi_0(r)$, and damping (or dissipation), due to the term proportional to γ, may be mutually compensating as has been described by Eilbeck (1983) and lead to a steady-state kink solution. The motion of such a kink solution would be subject to a pinning potential which would have to be overcome in order to set the vortex array in motion. The general case of (26), i.e. the damped driven SGE, has received considerable attention in the past few years. Largely numerical studies indicate a number of interesting phenomena such as the radiation effects of SG kinks (Malomed 1987) and perhaps more interestingly a transition to stochastic, or indeed chaotic, behaviour (Eilbeck 1983). In a recent letter Olsen *et al* (1985) reported the existence of pattern selection and the onset of low-dimensional chaos in a two-dimensional damped driven SGE. Depending on the strength of the forcing term the behaviour of the field undergoes a number of changes including periodic, running periodic and chaotic. The glassy behaviour linked previously in the dynamics of two-dimensional arrays of Josephson's junctions (Martinoli *et al* 1987) to a commensurate–incommensurate transition in vortex phases may indeed, in the present formulation, be related to the various running periodic and chaotic regimes above. This type of behaviour in high-T_c materials has been discussed by Deutscher and Müller (1987) and Morgenstern *et al* (1988) and studied by Müller *et al* (1987). It may also be related to the experiments on thermally activated flux creep (Ebner and Stroud 1985). The presence of the commensurate phase may herald flux pinning effects in some high-T_c superconductors (Aeppli *et al* 1987, Kes 1988).

6. Discussion and conclusion

In this paper we have presented a preliminary description of the onset of coherence in high-temperature superconductors in the framework of LG free-energy expansions. We have modelled a typical high-temperature superconductor by a quartic expansion in terms of the order parameter for each of a set of superconducting islands in the presence of vector potentials and including the interactions between neighbouring islands in the form of Lawrence–Doniach weak Josephson links. The main idea for island–island interaction follows the pioneering work of Deutscher and Müller (1987). We have derived the equations of state and discussed the régime of uncorrelated superconducting islands first followed by the inclusion of magnetic field penetration. Finally, with a network of superconducting islands forming a square lattice, we have discussed the effect of subsequent interactions between the islands. The main conclusion from the first part of the development is that the short coherence length and inherent periodicity achieved through the doping process may be reconciled theoretically through the concept of modulational instability in the system implying the ground state, for high-T_c superconductors, is not the mean field but a modulated one (elliptic wave) with its periodicity determined mathematically by an elliptic integral and physically by oxygen lattice periodicity. With an external magnetic field present a lattice of magnetic vortices is envisaged to separate the islands with its cores located at the zeros of the order parameter's envelope.

Turning our attention to the order parameter's phases, we have first presented a static picture following earlier papers on that topic. A connection has been made with the XY model and a Kosterlitz–Thouless transition which is predicted to occur close to T_c and leads to the establishment of an array of vortex–antivortex pairs in the space of order parameter phases. These should not be confused with the magnetic vortices and indeed the periodicities of the two lattices may not only be different but incommensurate with each other. We have subsequently analysed the space modulation of the island phase structure and have demonstrated that the relevant equation is closely related to the SG equation. As a result, with no magnetic field present, the N-kink SG solution, which seems appropriate in our case, may be described by what it has been referred to in earlier papers as a sequence of staircase supercurrents. In this case the commensurate periodicities of the two lattice structures seem to imply the presence of a pinning potential. The application of a magnetic field has been shown to give rise to a damped driven SGE with a number of fascinating properties and especially its transition to chaos at a certain value of the driving term which derives from the vector potential. Many of the qualitative features found in our model appear to be exhibited by experimental results. These include a glass-like behaviour (or even the existence of a spin glass phase), flux pinning and flux creep phenomena. With the presence of damping in the SGE there may be a critical value of the mean distance between neighbouring islands (directly related to both the temperature and the oxygen stoichiometry parameter) below which Josephson tunnelling may take place, leading to the onset of bulk superconductivity as opposed to just the presence of superconducting nucleation centres. We intend to investigate this in future studies which may give insight concerning the phase diagram of these high-T_c superconductors.

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