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On the triangular incommensurate phase in γ-brass: II. Deformation of the triangular pattern

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Abstract. An electron microscopic study has revealed the presence of a strong deformation of the triangular IAPD pattern in γ-brass. Using the free-energy functional associated with the phase variables of the pattern, we demonstrate the invariance of the topology of the pattern. We can regard the pattern system to be a medium, which we call the 'phase medium', and we can also obtain the effective elastic constants of the medium. The observed deformation of the pattern has been qualitatively reproduced by calculating the strain field of the phase medium under a proper boundary condition.

1. Introduction

Cu_xZn_{1-x} alloys crystallise with a somewhat complicated structure, conventionally called the γ-phase, in the narrow concentration range $0.57 < x < 0.66$ (Morton 1978). The structure belongs to a non-centrosymmetric space group T_d^3. Morton first found an interesting long-period triangular pattern using electron microscopy and then showed that the pattern is due to inversion antiphase domains (IAPD) (Morton 1975, 1976, 1977).

A recent paper discussed the origin of the stability of the γ-phase by assigning the amplitude of a particular phonon mode as the order parameter. Allowing spatial modulation of the order parameter, a phenomenological treatment of the formation of the triangular IAPD pattern in γ-CuZn was developed (Yamada and Koh 1987; hereafter referred to as part I).

Morton (1975) observed further interesting deformations of the regular array of the triangular pattern. As shown in figure 1 (see also figure 3 of part I, or that originally given in Morton's paper), the triangular pattern is deformed heavily near the boundaries surrounding a grain having a somewhat different alloy concentration, which corresponds to the region showing the striped pattern (Morton 1974). It is noticeable that at the boundaries the striped pattern keeps its rigid straight-line structure, while the triangular pattern is heavily deformed from a regular pattern. As is pointed out in part I, these characteristics lead us to imagine that the triangular pattern (hereafter called simply 'the pattern') is drawn on an extremely deformable elastic continuum.

In order to give a physical basis to this hypothesis, we proceed as follows. Let us assume that the perturbation of the pattern can be expressed by the change of the *phase*

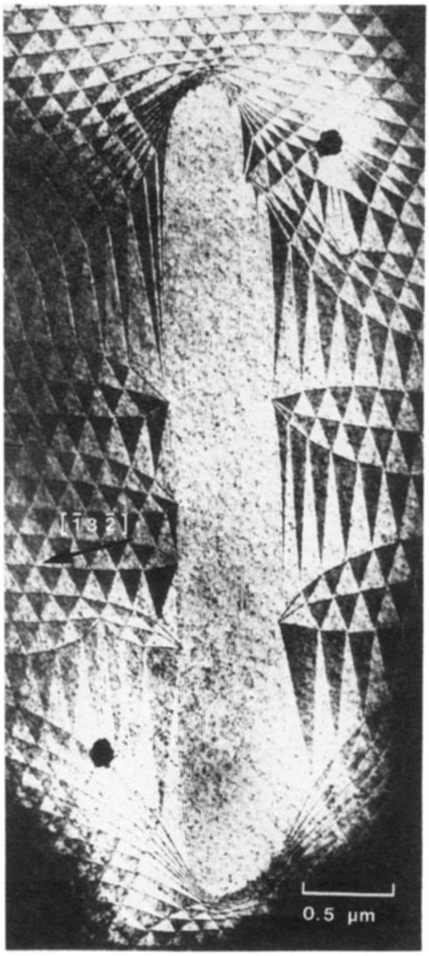

Figure 1. Observed deformation of the triangular pattern in the vicinity of the grain boundary of the striped phase, which has grown within the triangular matrix (Morton 1975).

of the modulation of the order parameter (phase mode approximation) (Kawasaki 1985):

$$\xi(\boldsymbol{r}) = \sum_{\nu=1}^{3} \left(\xi_{\nu 0}(\boldsymbol{r}) \exp(\mathrm{i}\Delta\varphi_{\nu}(\boldsymbol{r})) + \mathrm{cc}\right) \tag{1}$$

where $\xi_{\nu 0}(\boldsymbol{r})$ is the order parameter of the unperturbed state given by equation (14) of part I and cc is the complex conjugate. The regular triangular pattern is determined by the three phases of $\xi_{\nu 0}(\boldsymbol{r})$, where $\nu = 1, 2, 3$.

In this approximation, the energy associated with the deformed system should be given by the same free-energy functional as introduced in part I. The deformation energy is then simply given by the φ-dependent part of the free energy. It would then be quite possible that the lowest-order term of the deformation energy has formally the same expression as the elastic energy of an equi-phase line. It will be shown that the restricted condition $\Sigma_{\nu}\Delta\varphi_{\nu} = 0$ leads to the invariance of the topology of the pattern. Therefore the terms have the same expression as the elastic energy of a continuous elastic medium characterised by certain *effective elastic constants* which are expressed in terms of the parameters included in the free-energy functional. These considerations seem to suggest the validity of using the elasticity analogue to describe the deformation of the pattern around the grain boundary. In this paper, we will develop a treatment to discuss the observed deformation of the pattern based on the idea introduced above.

2. The free energy

In part I we discussed the formation of the triangular IAPD pattern in terms of a spatial modulation of the order parameter $\xi(\boldsymbol{r})$, which is the local amplitude of the frozen A_{2u} optical mode, and is also accompanied by a strain $\varepsilon(\boldsymbol{r})$. The pattern is obtained by minimising the free-energy functional, which is given by

$$F = F_1 + F_2 + F_3 + F_4 = \int (f_1 + f_2 + f_3 + f_4)\, \mathrm{d}^3\boldsymbol{r} \tag{2}$$

where

$$f_1 = a\xi^2 + b[(\partial\xi/\partial x)^2 + (\partial\xi/\partial y)^2 + (\partial\xi/\partial z)^2] + c(\partial^2\xi/\partial x^2 + \partial^2\xi/\partial y^2 + \partial^2\xi/\partial z^2)^2 + \lambda(\varepsilon_{yz}^2 + \varepsilon_{zx}^2 + \varepsilon_{zy}^2)$$

$$f_2 = \delta(\varepsilon_{xy}\, \partial\xi/\partial x + \varepsilon_{yz}\, \partial\xi/\partial y + \varepsilon_{zx}\, \partial\xi/\partial z)$$

$$f_3 = \mathrm{d}\xi^4 + e\xi^6$$

$$f_4 = f(\partial^4\xi/\partial x^4 + \partial^4\xi/\partial y^4 + \partial^4\xi/\partial z^4 + \partial^4\xi/\partial y^2\partial z^2 + \partial^4\xi/\partial z^2\partial x^2 + \partial^4\xi/\partial x^2\partial y^2)^2 + g[(\partial^4/\partial x^4)(\partial^2/\partial y^2 - \partial^2/\partial z^2)\xi + (\partial^4/\partial y^4)(\partial^2/\partial z^2 - \partial^2/\partial x^2)\xi + (\partial^4/\partial x^4)(\partial^2/\partial y^2 - \partial^2/\partial z^2)\xi]^2.$$

F_1 corresponds to the quadratic part with respect to $\xi(\boldsymbol{r})$ and $\varepsilon(\boldsymbol{r})$, F_2 to the bilinear coupling energy between $\xi(\boldsymbol{r})$ and $\varepsilon(\boldsymbol{r})$, and F_3 and F_4 to the higher-order terms with respect to ξ and K^0. The signs of these coefficients are chosen to be $a < 0$, $b, c > 0$, $d, e, f, g > 0$ and $\lambda > 0$ (see part I).

First, we eliminate $\varepsilon_{ij}(\boldsymbol{r})$ by minimising the free energies F_1 and F_2 with respect to $\varepsilon_{ij}(\boldsymbol{r})$. This procedure simply renormalises the parameter b to $(b - \delta^2/4\lambda)$. Thus the free energy to be discussed is given by

$$F = \int \mathrm{d}\boldsymbol{r}\left\{a\xi^2 + \left(b - \frac{\delta^2}{4\lambda}\right)\left[\left(\frac{\partial\xi}{\partial x}\right)^2 + \left(\frac{\partial\xi}{\partial y}\right)^2 + \left(\frac{\partial\xi}{\partial z}\right)^2\right] + c\left(\frac{\partial^2\xi}{\partial x^2} + \frac{\partial^2\xi}{\partial y^2} + \frac{\partial^2\xi}{\partial z^2}\right)^2 + d\xi^4 + e\xi^6 + (f_4 \text{ term})\right\}. \tag{3}$$

For the following development to give the deformation energy, we only take F_1, F_2 and F_3 into account and ignore the higher-order term, F_4. The calculation and discussion of the F_3 and F_4 terms are given in the appendix.

As shown by equation (10) in part I, the regular state (undeformed state), obtained by minimisation of F, is expressed by

$$\xi_0(\boldsymbol{r}) = \frac{1}{\sqrt{3}} \sum_{\nu=1}^{3} \xi_{0\nu}(\boldsymbol{r}) \tag{4a}$$

$$\xi_{0\nu}(\boldsymbol{r}) = (\hat{\xi}/\sqrt{2})(\exp[\mathrm{i}(\boldsymbol{K}_\nu^0 \cdot \boldsymbol{r} + \varphi_\nu^0)] + \mathrm{cc}) \tag{4b}$$

and

$$\sum_{\nu=1}^{3} \varphi_\nu^0 = (n + 1/2)\pi \tag{5}$$

where the $\boldsymbol{K}_\nu^0$ ($\nu = 1, 2, 3$) are the k-vectors pointing in the three equivalent $\langle \bar{2}11 \rangle$ directions within the (111) plane:

$$\boldsymbol{K}_1^0 \parallel \tfrac{1}{3}(1, 1, \bar{2}) \qquad \boldsymbol{K}_2^0 \parallel \tfrac{1}{3}(\bar{2}, 1, 1) \qquad \boldsymbol{K}_3^0 \parallel \tfrac{1}{3}(1, \bar{2}, 1)$$

and

$$|\boldsymbol{K}_\nu^0| = (1/\sqrt{2c})\sqrt{(b - \delta^2/4\lambda)}.$$

In this state, the equi-phase lines of $\xi_0(\boldsymbol{r})$ simply form the regular triangular pattern.

To discuss the deformation of the pattern we use the same free-energy functional as previously introduced. In contrast, however, we adopt the 'phase mode approximation' for the order parameter $\xi(\boldsymbol{r})$. That is, we assume the deformation can be expressed solely by the additional phase factors and we express $\xi(\boldsymbol{r})$ under the external perturbation in the form:

$$\xi(\boldsymbol{r}) = \frac{1}{\sqrt{3}} \sum_{\nu=1}^{3} \xi_\nu(\boldsymbol{r}) \tag{6a}$$

where

$$\xi_\nu(\boldsymbol{r}) = (1/\sqrt{2})(\xi_{0\nu}(\boldsymbol{r}) \exp(\mathrm{i}\Delta\varphi_\nu(\boldsymbol{r})) + \mathrm{cc}). \tag{6b}$$

Later we denote the total phase of $\xi_\nu(\boldsymbol{r})$ as $\Phi_\nu(\boldsymbol{r}) = \boldsymbol{K}_\nu^0 \cdot \boldsymbol{r} + \varphi_\nu(\boldsymbol{r}) \equiv \boldsymbol{K}_\nu^0 \cdot \boldsymbol{r} + \varphi_\nu^0 + \Delta\varphi_\nu(\boldsymbol{r})$. At this stage, we consider that the perturbation can be expressed by the additional phase factor $\Delta\varphi_\nu(\boldsymbol{r})$ alone; however, below we will discuss the relationship between the $\Delta\varphi_\nu(\boldsymbol{r})$s.

We use the same free-energy functional, equation (2) (or equation (3)). Substituting $\xi(\boldsymbol{r})$ of equation (6) into equation (3) and neglecting terms proportional to $(\partial^2\varphi(\boldsymbol{r})/\partial \boldsymbol{r}^2)$ gives the free energy in the deformed state:

$$F \simeq \tfrac{1}{3}\hat{\xi}^2 \int \mathrm{d}\boldsymbol{r}\left(a + B \sum_{\nu=1}^{3} \left|\boldsymbol{K}_\nu^0 + \frac{\partial}{\partial \boldsymbol{r}}\Delta\varphi_\nu(\boldsymbol{r})\right|^2 + c \sum_{\nu=1}^{3} \left|\boldsymbol{K}_\nu^0 + \frac{\partial}{\partial \boldsymbol{r}}\Delta\varphi_\nu(\boldsymbol{r})\right|^4\right)$$
$$+ \tfrac{5}{2}d\hat{\xi}^4 L^3 + e\hat{\xi}^6 \int \mathrm{d}\boldsymbol{r}\left(\tfrac{85}{9} + \tfrac{5}{3}\cos[2(\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}))]\right) \tag{7}$$

where

$$B \equiv b - \delta^2/4\lambda$$

and L is the size of the system.

Using the equilibrium value of $|\boldsymbol{K}_\nu^0| = \sqrt{(-B/2c)}$ given in equation (7) and expanding $\Delta\varphi$ around $\boldsymbol{K}_\nu^0$, we obtain the following compact formula for the free energy:

$$F \simeq \left[\left(a - \frac{B}{12c}\right)L^3 + \frac{4c}{3}\sum_{\nu=1}^{3}\int \mathrm{d}\boldsymbol{r}\left(\frac{\partial\Delta\varphi_\nu}{\partial\boldsymbol{r}}\cdot\boldsymbol{K}_\nu^0\right)^2\right]\cdot\xi^2$$
$$+\tfrac{5}{2}d\xi^4L^3 + e\xi^6\int \mathrm{d}\boldsymbol{r}\left(\tfrac{85}{9} + \tfrac{5}{3}\cos[2(\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}))]\right). \tag{8}$$

Omitting the constant term, we can write the free energy as

$$F \sim \int f\left(\varphi_\nu(\boldsymbol{r}), \frac{\partial\Delta\varphi_\nu(\boldsymbol{r})}{\partial\boldsymbol{r}}\right)\mathrm{d}\boldsymbol{r}$$
$$= \int\left[\frac{4c}{3}\xi^2\sum_{\nu=1}^{3}\left(\frac{\partial\Delta\varphi_\nu}{\partial\boldsymbol{r}}\cdot\boldsymbol{K}_\nu^0\right)^2\right.$$
$$\left. + \tfrac{5}{3}e\xi^6\cos[2(\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}))]\right]\mathrm{d}\boldsymbol{r} \equiv F_{\mathrm{ela}} + F_{\mathrm{phase}}. \tag{9}$$

Thus we find that F is a functional of $\varphi_\nu(\boldsymbol{r})$ and $(\partial\varphi_\nu(\boldsymbol{r})/\partial\boldsymbol{r})$ $(\nu = 1, 2, 3)$. The first term is the 'one-dimensional' elastic term in each ν-direction, while the second term is the phasing-relation term.

3. Phase medium

We must discuss the equation

$$\delta F = \delta(F_{\mathrm{ela}} + F_{\mathrm{phase}}) = 0 \tag{10}$$

to obtain the solution which has the minimum of F. On the other hand, a typical solution is obtained by firstly setting $\delta F_{\mathrm{phase}} = 0$, which gives us a relationship between the $\varphi_\nu(\boldsymbol{r})$, and secondly by setting $\delta F_{\mathrm{ela}} = 0$†. This relationship has the following significance, although its solution does not contain the ground-state free energy. The minimisation of F_{phase} is obtained by using the Euler–Lagrange equation

$$\delta F_{\mathrm{phase}} = -\tfrac{10}{3}e\xi^6\int \mathrm{d}\boldsymbol{r}\sum_{\nu=1}^{3}(\sin[2(\varphi_1 + \varphi_2 + \varphi_3)])\delta\varphi_\nu = 0 \tag{11}$$

which gives

$$\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}) = (n + 1/2)\pi\ddagger \tag{12}$$

or

$$\Delta\varphi_1(\boldsymbol{r}) + \Delta\varphi_2(\boldsymbol{r}) + \Delta\varphi_3(\boldsymbol{r}) = 0.$$

This leads to a concept which we will call 'phase medium'.

Let us consider a periodic pattern in two-dimensional space, characterised by a periodic function $f_0(\boldsymbol{r})$. If a perturbation results in a deformation of the pattern, the

† This procedure will be justified and the calculation will be performed in § 4.

‡ $$\sum_{\nu}^{3}\varphi_\nu(\boldsymbol{r}) \equiv \sum_{\nu}^{3}\varphi_\nu^0 + \sum_{\nu}^{3}\boldsymbol{K}_\nu^0\cdot\boldsymbol{r} + \sum_{\nu}^{3}\Delta\varphi_\nu(\boldsymbol{r}) \equiv (n + \tfrac{1}{2})\pi + 0 + \mu(\boldsymbol{r})$$

where each term on the right-hand side corresponds to the respective term in the middle expression.

periodic function consequently changes from $f_0(\boldsymbol{r})$ to $f(\boldsymbol{r})$. If the topology of the pattern is invariant, we can express the deformed function $f(\boldsymbol{r})$ by

$$f(\boldsymbol{r}) = f_0(\boldsymbol{r} + \boldsymbol{u}(\boldsymbol{r})). \tag{13}$$

That is, the 'phase' of the original pattern at $\boldsymbol{r}$ has been shifted to $\boldsymbol{r} + u(\boldsymbol{r})$ after the deformation. In this case, it is natural to consider the system as being a continuous medium associated with the 'phase' of the pattern (phase medium) and the deformation of the pattern due to the perturbation as being the elastic deformation of this medium.

In the present system the order parameter $\xi(\boldsymbol{r}) = \Sigma_\nu \xi_\nu(\boldsymbol{r})$ represents the function $f(\boldsymbol{r})$. In § 2 we assumed the 'phase mode approximation' for *each* constituent wave:

$$\xi_\nu(\boldsymbol{r}) = \xi_{\nu 0}(\boldsymbol{r} + u_\nu) \qquad \nu = 1, 2, 3 \tag{14}$$

$$\boldsymbol{K}_\nu^0 \cdot \boldsymbol{u}_\nu = -\Delta\varphi_\nu. \tag{15}$$

However, this does not mean that the phase mode representation of $\xi(\boldsymbol{r})$ is always satisfied.

Here we use the constraint in equation (12),

$$\sum_\nu \Delta\varphi_\nu \equiv \mu(\boldsymbol{r}) = 0 \tag{16}$$

is satisfied during the deformation process. Using equation (18), it is easy to show that the constraint gives the necessary condition to allow the phase mode representation of $\xi(\boldsymbol{r})$:

$$\xi_\nu(\boldsymbol{r}) = \xi_{\nu 0}(\boldsymbol{r} + \boldsymbol{u}^{(\mathrm{ph})}) \qquad \nu = 1, 2, 3. \tag{17}$$

That is, there exists a common vector $\boldsymbol{u}^{(\mathrm{ph})}$ which gives the phase shift of each $\xi_\nu(\boldsymbol{r})$ irrespective of ν. Then we have

$$\begin{aligned} \xi(\boldsymbol{r}) &= \sum_\nu \xi_{\nu 0}(\boldsymbol{r} + \boldsymbol{u}^{(\mathrm{ph})}) \\ &= \xi_0(\boldsymbol{r} + \boldsymbol{u}^{(\mathrm{ph})}). \end{aligned} \tag{18}$$

The geometrical interpretation of the above equation is that, even after the deformation, the three equi-phase lines intersect each other. That is, the topological characteristic of the pattern is maintained during the deformation process. We see that this situation is almost consistent with experiment (see figure 1).

From these considerations, we can take such a system with 'a' periodic pattern to be 'a' '*phase medium*'. We can imagine that the uniform phase medium has elastically 'strained' under an external perturbation so that the point $\boldsymbol{r}$ embedded on the undistorted phase medium has been displaced by $\boldsymbol{u}^{(\mathrm{ph})}$.

The value of $\boldsymbol{u}^{(\mathrm{ph})}$ is easily obtained from

$$\boldsymbol{K}_\nu^0 \cdot \boldsymbol{u}^{(\mathrm{ph})}(\boldsymbol{r}) = -\Delta\varphi_\nu(\boldsymbol{r}). \tag{19}$$

Defining an effective strain tensor $\varepsilon_{ij}^{(\mathrm{ph})}$ of the phase medium, in analogy to the definition of the ordinary strain of an elastic medium, by

$$\varepsilon_{ij}^{(\mathrm{ph})} = \frac{\partial u_i^{(\mathrm{ph})}}{\partial x_j} \qquad (i, j = x, y, z) \tag{20}$$

we rewrite equation (22) as

$$\partial \Delta\varphi_\nu(\boldsymbol{r})/\partial \boldsymbol{r} = -\boldsymbol{\varepsilon}^{(\mathrm{ph})} \cdot \boldsymbol{K}_\nu^0. \tag{21}$$

This gives the expression of the effective strain tensor $\boldsymbol{\varepsilon}^{(\mathrm{ph})}$ explicitly in terms of $\Delta\varphi_\nu(\boldsymbol{r})$.

Referring to a specific orthogonal reference frame $(\bar{x}, \bar{y})$ in the $\{111\}$ plane, where the $\bar{x}$ axis is taken parallel to one of the wavevectors of the three modulation waves, $\boldsymbol{K}_1^0$, equation (24) is expressed as follows:

$$\begin{bmatrix} \partial\varphi_\nu/\partial\bar{x} \\ \partial\varphi_\nu/\partial\bar{y} \end{bmatrix} = -\begin{bmatrix} \partial u_{\bar{x}}^{(\mathrm{ph})}/\partial\bar{x} & \partial u_{\bar{y}}^{(\mathrm{ph})}/\partial\bar{x} \\ \partial u_{\bar{x}}^{(\mathrm{ph})}/\partial\bar{y} & \partial u_{\bar{y}}^{(\mathrm{ph})}/\partial\bar{y} \end{bmatrix} \cdot \begin{bmatrix} \boldsymbol{K}_{\nu x}^0 \\ \boldsymbol{K}_{\nu y}^0 \end{bmatrix}. \tag{22}$$

4. Minimisation of the free energy

We will investigate the solution $\{\varphi_\nu(\boldsymbol{r})\}$ which has the minimum free energy. The solution is obtained using the equation $\delta F = \delta(F_{\mathrm{ela}} + F_{\mathrm{phase}}) = 0$ (equation (10)), which leads to the Euler–Lagrange equation:

$$\sin[2(\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}))] = -3\alpha\,|\boldsymbol{K}_\nu^0|^2\,\partial^2\Delta\varphi_\nu(\boldsymbol{r})/\partial q_\nu^2 \qquad (\nu = 1, 2, 3) \tag{23}$$

where q_ν is the coordinate in the direction of $\boldsymbol{K}_\nu^0$† and $\alpha \equiv 4c/15e\xi^2$ is the ratio of the coefficients of F_{ela} and F_{phase}. Here we note that equation (23) includes the equations

$$\begin{aligned} \partial^2\Delta\varphi_1(\boldsymbol{r})/\partial q_1^2 - \partial^2\Delta\varphi_3(\boldsymbol{r})/\partial q_3^2 = 0 \\ \partial^2\Delta\varphi_2(\boldsymbol{r})/\partial q_2^2 - \partial^2\Delta\varphi_3(\boldsymbol{r})/\partial q_3^2 = 0. \end{aligned} \tag{23'}$$

We can see from equation (23) that

$$\Delta\varphi_1(\boldsymbol{r}) + \Delta\varphi_2(\boldsymbol{r}) + \Delta\varphi_3(\boldsymbol{r}) \to 0 \quad \text{as} \quad \alpha \to 0. \tag{24}$$

Thus if α is small, the system is approximately the phase medium. Hereafter, in contrast to the approximate 'phase medium,' we will denote the system in which $\Delta\varphi_1(\boldsymbol{r}) + \Delta\varphi_2(\boldsymbol{r}) + \Delta\varphi_3(\boldsymbol{r}) \equiv 0$ as the complete phase medium. The experimental result shows that the phase-medium picture explains the deformation process quite well. Therefore we can consider that $|\alpha| \ll 1$.

On the other hand, the equation $\delta(F_{\mathrm{phase}} + F_{\mathrm{ela}}) = 0$ can be rewritten as

$$\delta F = \frac{4}{3}c\xi^2\left[\delta\int \mathrm{d}\boldsymbol{r}\sum_{\nu=1}^{3}\left(\frac{\partial}{\partial\boldsymbol{r}}\Delta\varphi_\nu\cdot\boldsymbol{K}_\nu^0\right)^2 + \left(\frac{1}{3\alpha}\right)\delta\int \mathrm{d}\boldsymbol{r}\cos[2(\varphi_1+\varphi_2+\varphi_3)]\right] = 0. \tag{25}$$

This can be interpreted as Lagrange's method of undetermined multipliers where F_{ela} is minimised under the restricted condition that F_{phase} is stationary, i.e. $\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}) = (n + 1/2)\pi$‡ and the undetermined multiplier is $(1/3\alpha)$. Needless to say that α is not an undetermined number, rather it is a deterministic number which is the ratio of the coefficients in the Ginzburg–Landau expansion. However the fact that $|\alpha| \ll 1$ (or $|1/3\alpha| \gg 1$) from equation (23) and the experiment may allow a treatment in which α is regarded as an undetermined number. Equation (25) then leads again to equation (23), in which $(1/3\alpha)$ is also interpreted as the undetermined multiplier. Substituting the restricted condition that $\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}) = (n + 1/2)\pi$ into

† $q_\nu = \bar{x}\cos\theta_\nu + \bar{y}\sin\theta_\nu$. The coordinate axis is parallel to $\boldsymbol{K}_\nu^0 = (|\boldsymbol{K}_\nu^0|\cos\theta_\nu, |\boldsymbol{K}_\nu^0|\sin\theta_\nu)$. $(\bar{x}, \bar{y})$ is a coordinate on the observed plane.

‡ We cannot consider another relation, $\Sigma_\nu\varphi_\nu = n\pi$, from the experimental result and the physical viewpoint.

equation (23), the multiplier $(1/3\alpha)$ becomes infinite. This result is consistent with the above fact that $(1/3\alpha) \gg 1$ from $\delta(F_{\mathrm{ela}} + F_{\mathrm{phase}}) = 0$ and the experimental result. Therefore the use of Lagrange's method to obtain the solution $\{\varphi_\nu(\boldsymbol{r})\}$ which gives the complete phase medium can be justified.

We now examine Lagrange's method in detail, using the three following steps.

(i) The restricted condition is:

$$\delta F_{\mathrm{ela}} = \tfrac{10}{3}\, e\xi^6 \int \mathrm{d}\boldsymbol{r} \sum_{\nu=1}^{3} (\cos[2(\varphi_1 + \varphi_2 + \varphi_3)])\delta\varphi_\nu = 0. \tag{26}$$

This leads to the phasing relation $\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}) = (n + 1/2)\pi$ (or $\Delta\varphi_1(\boldsymbol{r}) + \Delta\varphi_2(\boldsymbol{r}) + \Delta\varphi_3(\boldsymbol{r}) \equiv 0$).

(ii) F_{ela} under the above restricted condition is given by

$$F_{\mathrm{ela}} = \tfrac{4}{3} c\xi^2 \int \mathrm{d}\boldsymbol{r}\left[\left(\frac{\partial}{\partial \boldsymbol{r}}\Delta\varphi_1 \cdot \boldsymbol{K}_1^0\right)^2 + \left(\frac{\partial}{\partial \boldsymbol{r}}\Delta\varphi_2 \cdot \boldsymbol{K}_2^0\right)^2 + \left(\frac{\partial}{\partial \boldsymbol{r}}(0 - \Delta\varphi_1 - \Delta\varphi_2)\cdot \boldsymbol{K}_3^0\right)^2\right]. \tag{27}$$

Here we note that the number of independent functionals becomes two, for example $\varphi_1(\boldsymbol{r})$ and $\varphi_2(\boldsymbol{r})$, as a result of the restricted condition.

Substitution of equation (21) into equation (27) gives the deformation energy of the pattern in a quadratic form with respect to $\varepsilon^{(\mathrm{ph})}$ as follows:

$$F_{\mathrm{ela}} = \frac{4c}{3}\xi^2 \sum_{\nu=1}^{3} \int \mathrm{d}\boldsymbol{r}\{\boldsymbol{K}_\nu^0 \cdot \boldsymbol{\varepsilon}^{(\mathrm{ph})} \cdot \boldsymbol{K}_\nu^0\}^2. \tag{28}$$

We can easily show that the formalism is not changed by using the conventional symmetrical strain tensor $\{\hat{\varepsilon}^{(\mathrm{ph})}\}$†. Using the tensor components of $\hat{\varepsilon}^{(\mathrm{ph})}$, the energy (28) is reformulated with the same form as the elastic strain energy:

$$F_{\mathrm{ela}} = \tfrac{1}{2}\int \mathrm{d}\boldsymbol{r} \sum_{i,j,k,l} C_{ijkl}^{(\mathrm{ph})}\, \hat{\varepsilon}_{ij}^{(\mathrm{ph})}\, \hat{\varepsilon}_{kl}^{(\mathrm{ph})}. \tag{29}$$

Here, $C_{ijkl}^{(\mathrm{ph})}$ represents the effective 'elastic constant'. We have thus established that the system under consideration is equivalent to an elastic medium characterised by the effective elastic constants $\{C_{ijkl}^{(\mathrm{ph})}\}$.

Comparing equations (28) and (29), and substituting the components of $\boldsymbol{K}_\nu^0$ into equation (28), we obtain the following values as the components of $C_{ijkl}^{(\mathrm{ph})}$:

$$\begin{aligned} C_{\bar{x}\bar{x}\bar{x}\bar{x}}^{(\mathrm{ph})} &= C_{\bar{y}\bar{y}\bar{y}\bar{y}}^{(\mathrm{ph})} = \tfrac{9}{4}|B|\,\xi^2 \\ C_{\bar{x}\bar{x}\bar{y}\bar{y}}^{(\mathrm{ph})} &= C_{\bar{y}\bar{y}\bar{x}\bar{x}}^{(\mathrm{ph})} = \tfrac{3}{4}|B|\,\xi^2 \\ C_{\bar{x}\bar{y}\bar{x}\bar{y}}^{(\mathrm{ph})} &= C_{\bar{y}\bar{x}\bar{y}\bar{x}}^{(\mathrm{ph})} = \tfrac{3}{2}|B|\,\xi^2 \\ C_{(\mathrm{otherwise})}^{(\mathrm{ph})} &= 0. \end{aligned} \tag{30}$$

The expressions for the components of the $C^{(\mathrm{ph})}$ tensor show that the 'phase medium'

† The symmetrical tensor components $\hat{\varepsilon}_{ij}^{(\mathrm{ph})}$ are defined as

$$\hat{\varepsilon}_{ij}^{(\mathrm{ph})} = \tfrac{1}{2}(\partial u_i^{(\mathrm{ph})}/\partial x_i + \partial u_j^{(\mathrm{ph})}/\partial x_j) \qquad (x_i, x_j = \bar{x} \text{ or } \bar{y}).$$

is elastically isotropic. The contribution from the higher-order terms, considered to be a small perturbation, will be discussed in the appendix.

We further note that the tensor components are proportional to $|B| = b - \delta^2/4\lambda$. In part I it was shown that the long periodicity of the pattern in γ-CuZn is due to the near cancellation of b and $\delta^2/4\lambda$. Hence, we see that the pattern will be in fact very deformable. The long periodicity and the ease of deformability of the pattern are interrelated.

(iii) Minimisation of F_{ela} under the restricted condition yields:

$$\delta F_{\mathrm{ela}} = \frac{8}{3} c\xi^2 \, |\boldsymbol{K}_\nu^0|^2 \int \mathrm{d}\boldsymbol{r} \left[\left(\frac{\partial^2 \Delta\varphi_1}{\partial q_1^2} - \frac{\partial^2 \Delta\varphi_3}{\partial q_3^2} \right) \delta\varphi_1 + \left(\frac{\partial^2 \Delta\varphi_2}{\partial q_2^2} - \frac{\partial^2 \Delta\varphi_3}{\partial q_3^2} \right) \delta\varphi_2 \right] = 0. \tag{31}$$

Then we obtain the equations

$$\partial^2 \Delta\varphi_1(\boldsymbol{r})/\partial q_1^2 - \partial^2 \Delta\varphi_3(\boldsymbol{r})/\partial q_3^2 = 0 \qquad \partial^2 \Delta\varphi_2(\boldsymbol{r})/\partial q_2^2 - \partial^2 \Delta\varphi_3(\boldsymbol{r})/\partial q_3^2 = 0 \tag{32}$$

under the restricted condition at each point $\boldsymbol{r}$. This equation is the same as equation (23′), except that there is no phasing relation in equation (23′).†

Using the relationship between $\varphi_\nu(\boldsymbol{r})$ and $\varepsilon^{(\mathrm{ph})}$ from equation (21) in § 3, we can easily show that equation (32) is equivalent to the force balance equation in elasticity theory:

$$\sum_{j,k,l} \left(\frac{\partial}{\partial x_j} C_{ijkl}^{(\mathrm{ph})} \varepsilon_{kl}(\boldsymbol{r}) \right) = 0. \tag{33}$$

This equation also results from the minimisation of the elastic formula for F_{ela}, equation (29).

We solve the equation under the following boundary condition: a small circular hole is cut at the origin of the free continuum, and then the edge of the circle is expanded to form a large ellipsoid keeping the outer boundary of the continuum fixed (see figure 2). This situation is more clearly understood by introducing the phase soliton picture (Bak 1978, Bak and Emery 1976).

This problem has been already studied for the case of an ordinary elastic system. Following the established treatment, we introduce Airy's stress function $\varphi(r, \theta)$ of the two-dimensional system which satisfies the double harmonic equation

$$[\partial^2/\partial r^2 + (1/r)(\partial/\partial r) + (1/r^2)(\partial^2/\partial\theta^2)]^2 \varphi = 0 \tag{34}$$

expressed in a cylindrical coordinate system (r, θ).

We expand the θ-dependent part of φ with respect to $\cos(m\theta)$ and $\sin(m\theta)$ and take the terms $m \leqslant 4$ into account. The components of the stress tensor $(\sigma_{i,j})$ are then expressed by

$$\begin{aligned} \sigma_{rr}^{(\mathrm{ph})}(r, \theta) &= -P_0(1/r^2) - 4P_2(\cos(2\theta)/r^2) - 18P_4(\cos(4\theta)/r^4) \\ \sigma_{\theta\theta}^{(\mathrm{ph})}(r, \theta) &= P_0(1/r^2) + 6P_4(\cos(4\theta)/r^4) \\ \sigma_{r\theta}^{(\mathrm{ph})}(r, \theta) &= -2P_2(\sin(2\theta)/r^2) - 12P_4(\sin(4\theta)/r^4) \end{aligned} \tag{35}$$

† In the limit $\alpha \to 0$, equation (23′) approaches equation (32) and also equation (33). For finite α, each expression in equation (32) shows the 'phase medium' in a narrow sense which consists of only two groups of the equi-phase lines.

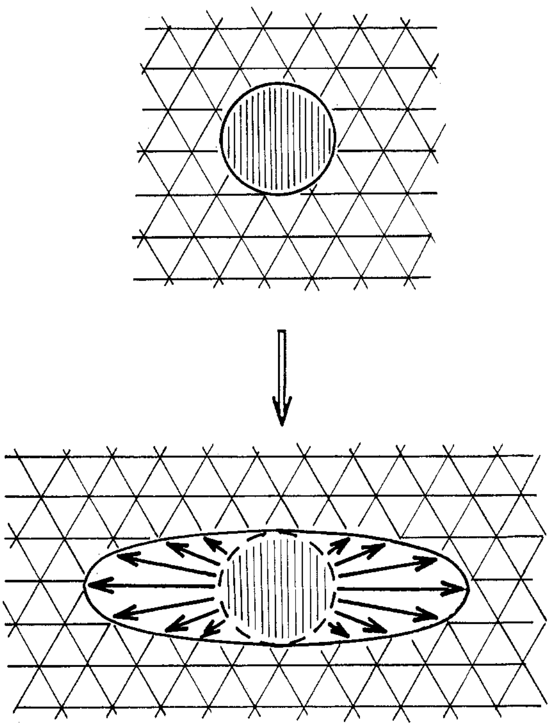

Figure 2. Schematic description of the boundary condition used for the calculation in § 3. A circular hole (broken circle) is opened in an elastic medium and then expanded to form a larger ellipsoid. The total mass of the medium is conserved during the process.

where P_0, P_2 and P_4 are the parameters to be determined by the boundary conditions. Using Young's modulus and Poisson's ratio of the elastic medium, the strain components are given by

$$\begin{aligned}
\varepsilon_{rr}^{(\mathrm{ph})} &= \partial u_r/\partial r = [(1+\sigma)/E][(1-\sigma)\sigma_{rr}^{(\mathrm{ph})} - \sigma\sigma_{\theta\theta}^{(\mathrm{ph})}] \\
\varepsilon_{\theta\theta}^{(\mathrm{ph})} &= (1/r)(\partial u_\theta/\partial\theta) + u_r/r = [(1+\sigma)/E][(1-\sigma)\sigma_{\theta\theta}^{(\mathrm{ph})} - \sigma\sigma_{rr}^{(\mathrm{ph})}] \\
\varepsilon_{r\theta}^{(\mathrm{ph})} &= \tfrac{1}{2}[(1/r)(\partial u_r/\partial\theta) - u_\theta/r + \partial u_\theta/\partial r] = [(1+\sigma)/E]\sigma_{r\theta}^{(\mathrm{ph})}.
\end{aligned} \tag{36}$$

In the present case, the Young's modulus E and Poisson's ratio σ associated with the effective elastic constants $\{C_{ijkl}^{(\mathrm{ph})}\}$ are given as $\sigma = 1/4$ and $E = (15/16)|B|\hat{\xi}^2$. The absolute value of $\varepsilon^{(\mathrm{ph})}$ is irrelevant because E only normalises the scale of P_i $(i = 0, 2, 4)$.

Finally, the displacement of the phase medium at (r, θ), $u^{(\mathrm{ph})}(r, \theta)$, is expressed by the following simple analytic functions:

$$\begin{aligned}
u_r^{(\mathrm{ph})}(r,\theta) &= [P_0(1+\sigma)/Er] - [4P_2(1+\sigma)(1-2\sigma)/E]\cos(2\theta)/r \\
&\quad + [2P_4(1+\sigma)(3-2\sigma)/E]\cos(4\theta)/r^3 \\
u_\theta^{(\mathrm{ph})}(r,\theta) &= -[2P_2(1+\sigma)(1-2\sigma)/E]\sin(2\theta)/r \\
&\quad + [4P_4(1+\sigma)\sigma/E]\sin(4\theta)/r^3
\end{aligned} \tag{37}$$

These values give the deformation of a pattern drawn on the medium during the process where a circular area has been expanded to a larger ellipsoidal region. The results of the

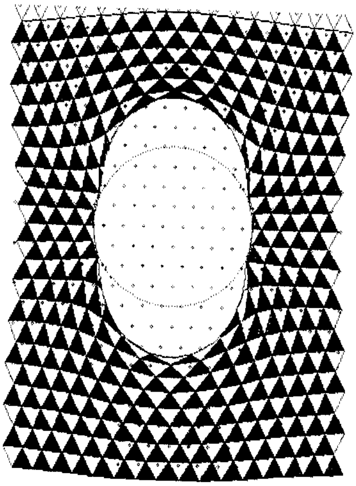

Figure 3. Calculated pattern when the intitial hole (broken circle) is enlarged to the ellipsoidal hole within the phase medium. The pattern qualitatively reproduces the observed deformed triangular pattern.

calculation of the deformation of the triangular pattern are given in figure 3, where $P_0 = 8$, $P_2 = 2$ and $P_4 = 2$. It is seen that the calculated pattern seems to reproduce the essential characteristics of the observed deformation (Morton 1975).

In the above discussion we considered the complete phase medium in which $(1/3\alpha) = \infty$ (or $\alpha = 0$); however, this value is large enough but is finite in the real system which is the approximate phase medium. Finally we estimate the deviation in the free energy of the complete phase medium from the ground state, i.e. the appropriate phase medium ($\mu(\boldsymbol{r}) \neq 0$):

$$\Delta F = F\{\mu(\boldsymbol{r}) = 0\} - F\{\mu(\boldsymbol{r}) \neq 0\}. \tag{38}$$

Note that $\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}) \equiv (n + 1/2)\pi + \mu(\boldsymbol{r})$, or $\Delta\varphi_1(\boldsymbol{r}) + \Delta\varphi_2(\boldsymbol{r}) + \Delta\varphi_3(\boldsymbol{r}) \equiv \mu(\boldsymbol{r})$ in the ground state. Note also that $|\mu(\boldsymbol{r})|$ and $|\alpha|$ are of the same order (from equation (23)).

Substituting this relation into equation (9), the ground-state free-energy density can be rewritten as

$$\begin{aligned} f(\boldsymbol{r}) &= (4c/3)\xi^2\{[(\partial\Delta\varphi_1/\partial\boldsymbol{r})\cdot\boldsymbol{K}_1^0]^2 + [(\partial\Delta\varphi_2/\partial\boldsymbol{r})\cdot\boldsymbol{K}_2^0]^2 \\ &\qquad + [(\partial/\partial\boldsymbol{r})(0 + \mu(\boldsymbol{r}) - \Delta\varphi_1 - \Delta\varphi_2)\cdot\boldsymbol{K}_3^0]^2\} + \tfrac{5}{3}\,e\xi^6(1 - 4\mu^2 + \ldots) \\ &= \tfrac{5}{3}e\xi^6[\![1 - 3\alpha\{[(\partial\varphi_1/\partial\boldsymbol{r})\cdot\boldsymbol{K}_1^0]^2 + [(\partial\varphi_2/\partial\boldsymbol{r})\cdot\boldsymbol{K}_2^0]^2 \\ &\qquad + [(\partial/\partial\boldsymbol{r})\,(0 - \Delta\varphi_1 - \Delta\varphi_2)\cdot\boldsymbol{K}_3^0]^2\} \\ &\qquad + \{6\alpha[(\partial\mu(\boldsymbol{r})/\partial\boldsymbol{r})\cdot\boldsymbol{K}_3^0]\cdot[(\partial/\partial\boldsymbol{r})(0 - \Delta\varphi_1 - \Delta\varphi_2)\cdot\boldsymbol{K}_3^0] \\ &\qquad - 4\mu^2(\boldsymbol{r})\} + \ldots]\!]. \end{aligned} \tag{39}$$

These terms can be classified by the order

$$F \simeq \frac{5}{3}e\xi^6\int \mathrm{d}\boldsymbol{r}(o(1) + o(\alpha) + o(\alpha^2) + \ldots). \tag{40}$$

Obviously the free-energy density of the complete phase medium corresponds to the term $(o(1) + o(\alpha))$. Thus, the shift of the energy in the complete phase medium from the ground-state energy is represented by

$$\Delta F \simeq \frac{5}{3} e\xi^6 \left| \int \mathrm{d}\boldsymbol{r}(o(\alpha^2) + o(\alpha^3) + \ldots) \right| \tag{41}$$

that is, more than α^2-order. We consider these terms to be negligible.

5. Conclusions and discussion

We have shown that the deformation of the triangular IAPD pattern can be understood in terms of an elastic continuum model of the phase medium. The regular arrangement of the equi-phase lines in the normal IAPD pattern defines the undistorted phase medium. Using the free-energy functional, we could introduce quantities to discuss the deformation of this 'phase medium', such as the effective strain, $\varepsilon^{(\mathrm{ph})}$, the effective stress $(\sigma_{i,j}^{(\mathrm{ph})})$, etc, and the effective elastic constants, $C_{ijkl}^{(\mathrm{ph})}$. The observed deformation has been qualitatively reproduced by calculating the displacement field under the proper boundary condition.

We have derived the important sum rule (equation (12))

$$\sum_{\nu=1}^{3} (\Delta\varphi_\nu(\boldsymbol{r})) = 0.$$

This result has two important implications from an experimental viewpoint.

(i) The sum rule has been deduced using equation (23) and the assumption $|\alpha| \ll 1$. However, in the region where the pattern is heavily deformed, this condition may not be satisfied in spite of the small α. We expect the sum rule to be broken near the grain boundary where the deformation is very strong. In fact, we can see in the observed electron micrographs that the vertices of the full triangles are linked completely in the non-deformed or weakly deformed region, while they sometimes fall apart near the grain boundary. The difference is very subtle, and can only be seen in the original figure presented in Morton's (1975) paper.

(ii) It is convenient to introduce the 'local wavevector' to interpret the deformed pattern. Let us expand $\varphi(\boldsymbol{r})$ with respect to $\boldsymbol{r}$ around $\boldsymbol{r}_0$:

$$\varphi_\nu(\boldsymbol{r}) = \varphi_\nu(\boldsymbol{r}_0) + (\partial\varphi_\nu/\partial\boldsymbol{r}) \cdot (\boldsymbol{r} - \boldsymbol{r}_0). \tag{42}$$

The order parameter $\xi_\nu(\boldsymbol{r})$ around $\boldsymbol{r}_0$ is given by

$$\begin{aligned} \xi_\nu(\boldsymbol{r}) &= \hat{\xi}(\exp[\mathrm{i}(\boldsymbol{K}_\nu^0 \cdot \boldsymbol{r} + \varphi_\nu(\boldsymbol{r}))] + \mathrm{cc}) \\ &\simeq \hat{\xi}(\exp\{\mathrm{i}[\boldsymbol{K}_\nu^0 \cdot \boldsymbol{r} + \varphi_\nu(\boldsymbol{r}_0) + (\partial\varphi_\nu/\partial\boldsymbol{r})|_{\boldsymbol{r}_0} \cdot \boldsymbol{r}]\} + \mathrm{cc}) \\ &= \hat{\xi}(\exp[\mathrm{i}(\boldsymbol{K}'_\nu(\boldsymbol{r}_0) \cdot \boldsymbol{r} + \varphi_\nu(\boldsymbol{r}_0))] + \mathrm{cc}) \end{aligned} \tag{43}$$

where $\boldsymbol{K}'_\nu \equiv \boldsymbol{K}_\nu^0 + \partial\varphi_\nu/\partial\boldsymbol{r}$. This means that the spatial modulation in a coarse-grained region around $\boldsymbol{r}_0$ may be approximated by a sinusoidal modulation with the wavevector

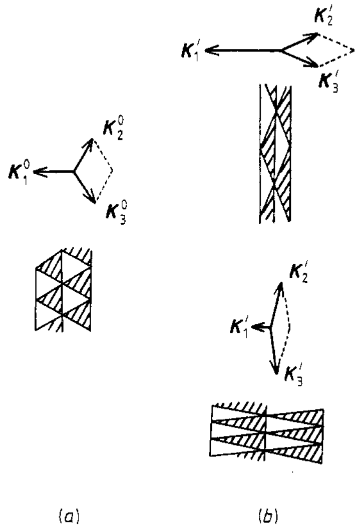

Figure 4. 'Local-k' representation of the triangular pattern: (a) the regular pattern and the corresponding set of $\{K_\nu^0\}$; (b) two typical deformed patterns and the corresponding sets of $\{K_\nu'\}$, The local free-energy minima in k-space may be considered to have displaced from $\{K_\nu^0(r)\}$ to $\{K_\nu'(r)\}$ under local stress.

$K_\nu'(r_0)$ which we call the 'local wavevector'. Due to the sum rule given in equation (15), the local wavevectors $\{K_\nu'\}$ satisfy:

$$\sum_\nu K_\nu'(r) = 0 \tag{44}$$

which is the necessary condition for the three waves $(\exp(\mathrm{i}K_\nu' \cdot r))$ to form a triangular pattern. We may interpret the characteristics of the deformed triangle around r_0 as being represented by the set of local wavevectors $\{K_\nu'(r_0)\}$ (see figure 4).

The local-k representation of the deformation is useful for the following discussion. As discussed in part I, $\{K_\nu^0\}$ is determined by the minimum points of the free-energy surface in k-space. Similarly, $\{K_\nu'(r)\}$ must correspond to the new minimum points of the local free energy when the system has been brought under the effective stress. If the energy surface has a shallow minimum in k-space, the minimum point is easily displaced, whence the pattern is strongly deformed under a small perturbation.

In § 1 we noted that at the grain boundary the triangular pattern is subjected to strong deformation while the striped pattern apparently shows no appreciable changes. This situation may be understood from the above viewpoint as follows. The free-energy surface of the present system has basically two kinds of minima in k-space. One is at $|k_s| \simeq 10^{-2}$ Å^{-1}, representing the stabilisation of the striped phase. The other is at $|k_t| \simeq 10^{-3}$ Å^{-1} corresponding to the triangular phase. We consider that the minima at k_t is very shallow while that at k_s is well defined. Under the same effective stress field, the triangular pattern is easily deformed but the striped pattern will remain relatively rigid.

Appendix

Here we discuss the higher-order term F_3 and F_4 in equation (2). In this appendix, we specify a term in F denoted by $F^{(*)}$, where the asterisk stands for the coefficients e, f, g, . . ., of the specified term and we also denote $k_\nu \equiv K_\nu^0$ for simplicity. The terms $F_3^{(d)}$ and $F_3^{(e)}$ are isotropic and the terms $F_4^{(f)}$ and $F_4^{(g)}$ are anisotropic in terms of the direction of the wavevectors. In addition, we will discuss the term $F_3^{(h)}$ which is also discussed in the appendix of part I.

It is evident that $F_3^{(d)}$, whose integrand consists of ξ^4, gives the same constant value $(5/2)d\xi^4$ as in the regular triangular state in part I. On the other hand, $F_3^{(e)}$ contributes to give the sum rule

$$\sum_{\nu=1}^{3} \Delta\varphi_\nu(\boldsymbol{r}) = 0. \tag{A1}$$

This condition determines the relative phase relation between the three waves. That is, $F_3^{(e)}$ is essentially controlling the 'relative phase locking' condition. The term

$$F_3^{(h)}(\boldsymbol{r}) = h \int \frac{\partial\xi}{\partial x}\frac{\partial\xi}{\partial y}\frac{\partial\xi}{\partial z}\,\mathrm{d}\boldsymbol{r} \tag{A2}$$

gives essentially an identical contribution. Using the same argument as for the $F_3^{(e)}$, we have

$$F_3^{(h)}(\boldsymbol{r}) = h\,\xi^2 \int h \sin(\varphi_1(\boldsymbol{r}) + \varphi_2(\boldsymbol{r}) + \varphi_3(\boldsymbol{r}))\,\mathrm{d}\boldsymbol{r}. \tag{A3}$$

The functional derivative of this term results in the same relation for $\Delta\varphi_\nu(\boldsymbol{r})$.

Now we discuss F_4, which represents the anisotropic part of the free energy:

$$F_4^{(f)} = f[\partial^4\xi/\partial x^4 + \partial^4\xi/\partial y^4 + \partial^4\xi/\partial z^4 + \partial^4\xi/\partial y^2\partial z^2 + \partial^4\xi/\partial z^2\partial x^2 + \partial^4\xi/\partial x^2\partial y^2]^2 \tag{A4}$$

$$F_4^{(g)} = g[(\partial^4/\partial x^4)(\partial^2/\partial y^2 - \partial^2/\partial z^2)\xi + (\partial^4/\partial y^4)(\partial^2/\partial z^2 - \partial^2/\partial x^2)\xi + (\partial^4/\partial x^4)(\partial^2/\partial y^2 - \partial^2/\partial z^2)\xi]^2. \tag{A5}$$

First we calculate $F_4^{(f)}$. Substituting equation (6) into (A4), we obtain

$$\begin{aligned} F_4^{(f)} \simeq f\frac{\xi^2}{6}\int \mathrm{d}\boldsymbol{r}\Bigg(\sum_{\nu=1}^{3}\bigg[&(k_{\nu x}^2 + k_{\nu y}^2 + k_{\nu z}^2 - 2k_{\nu y}k_{\nu z} - 2k_{\nu z}k_{\nu x} - 2k_{\nu x}k_{\nu y}) \\ &+ 4k_{\nu x}(k_{\nu x}^2 - k_{\nu y}^2 - k_{\nu z}^2)\frac{\partial\varphi_\nu}{\partial x} + 4k_{\nu y}(k_{\nu y}^2 - k_{\nu z}^2 - k_{\nu x}^2)\frac{\partial\varphi\,\nu}{\partial y} \\ &+ 4k_{\nu z}(k_{\nu z}^2 - k_{\nu x}^2 - k_{\nu y}^2)\frac{\partial\varphi_\nu}{\partial z}\bigg]\exp[\mathrm{i}(\boldsymbol{k}_\nu\cdot\boldsymbol{r} + \varphi_0)] + \mathrm{cc}\Bigg)^2. \end{aligned} \tag{A6}$$

We take the three equilibrium directions of $\boldsymbol{k}_\nu$, $\boldsymbol{k}_1 \parallel [1, 1, -2]$, $\boldsymbol{k}_2 \parallel [-2, 1, 1]$, $\boldsymbol{k}_3 \parallel [1, -2, 1]$ in the (x, y, z) framework. Using the above $\boldsymbol{k}_\nu$ ($\nu = 1, 2, 3$), the first term of equation (A6) is equal to zero as shown in part I. However, the other terms remain finite.

Let us use the new reference frame $(\bar{x}, \bar{y}, \bar{z})$ as in § 2. The transformation matrix is given by

$$\begin{bmatrix} \partial\varphi_\nu/\partial x \\ \partial\varphi_\nu/\partial y \\ \partial\varphi_\nu/\partial z \end{bmatrix} = \begin{bmatrix} 1/\sqrt{2} & 1/\sqrt{6} & 1/\sqrt{3} \\ -1/\sqrt{2} & 1/\sqrt{6} & 1/\sqrt{3} \\ 0 & -2/\sqrt{6} & 1/\sqrt{3} \end{bmatrix} \begin{bmatrix} \partial\varphi_\nu/\partial \bar{x} \\ \partial\varphi_\nu/\partial \bar{y} \\ \partial\varphi_\nu/\partial \bar{z} \end{bmatrix}. \tag{A7}$$

Then we have

$$F_4^{(f)} \simeq \frac{32}{27} f|\boldsymbol{k}|^6 \xi^2 \sum_{\nu=1}^{3} \int \mathrm{d}\boldsymbol{r} \left(\frac{\partial\varphi_\nu}{\partial \bar{z}}\right)^2 \tag{A8}$$

within the order of $(\partial\varphi/\partial\boldsymbol{r})$. Corresponding to equations (15) and (16), the derivation $\{\partial\varphi_i/\partial\bar{z}\}$ along the $\bar{z}$ direction is given by

$$\begin{bmatrix} \partial\varphi_1/\partial \bar{z} \\ \partial\varphi_2/\partial \bar{z} \\ \partial\varphi_3/\partial \bar{z} \end{bmatrix} = \begin{bmatrix} 0 & 1 & 0 \\ -\sqrt{3}/2 & -1/2 & 0 \\ \sqrt{3}/2 & -1/2 & 0 \end{bmatrix} \begin{bmatrix} \partial u_x^{(\mathrm{ph})}/\partial \bar{z} \\ \partial u_y^{(\mathrm{ph})}/\partial \bar{z} \\ \partial u_z^{(\mathrm{ph})}/\partial \bar{z} \end{bmatrix}. \tag{A9}$$

Finally we obtain the $F_4^{(f)}$ term, as expressed in the $(\bar{x}, \bar{y}, \bar{z})$ reference frame, as

$$\begin{aligned} F_4^{(f)} &\simeq \frac{16}{9} f|\boldsymbol{k}|^6 \xi^2 \sum_{\nu=1}^{3} \int \mathrm{d}\boldsymbol{r} \left[\left(\frac{\partial u_x}{\partial \bar{z}}\right)^2 + \left(\frac{\partial u_y}{\partial \bar{z}}\right)^2\right] \\ &= \frac{16}{9} f|\boldsymbol{k}|^6 \xi^2 \sum_{\nu=1}^{3} \int \mathrm{d}\boldsymbol{r} [(\varepsilon_{\bar{x}\bar{z}}^{(\mathrm{ph})})^2 + (\varepsilon_{\bar{y}\bar{z}}^{(\mathrm{ph})})^2]. \end{aligned} \tag{A10}$$

Obviously, this term does not contribute to the deformation energy within the $\{111\}$ plane.

We also calculate the $F_4^{(g)}$ term by substituting equation (6) into (A5). This gives

$$\begin{aligned} F_4^{(g)} \simeq g\frac{\xi^2}{6} \int \mathrm{d}\boldsymbol{r} \Bigg(\sum_{\nu=1}^{3} \Bigg\{ & [k_{\nu x}^4(k_{\nu y}^2 - k_{\nu z}^2) + k_{\nu z}^4(k_{\nu z}^2 - k_{\nu x}^2) + k_{\nu z}^4(k_{\nu x}^2 - k_{\nu y}^2)] \\ & + k_{\nu x}(k_{\nu y}^2 - k_{\nu z}^2)(k_{\nu y}^2 + k_{\nu z}^2 - 2k_{\nu x}^2)\frac{\partial\varphi_\nu}{\partial x} \\ & + k_{\nu y}(k_{\nu z}^2 - k_{\nu x}^2)(k_{\nu z}^2 + k_{\nu x}^2 - 2k_{\nu y}^2)\frac{\partial\varphi_\nu}{\partial y} \\ & + k_{\nu z}(k_{\nu x}^2 - k_{\nu y}^2)(k_{\nu x}^2 + k_{\nu y}^2 - 2k_{\nu z}^2)\frac{\partial\varphi_\nu}{\partial z} \Bigg\} \\ & \times \exp[\mathrm{i}(\boldsymbol{k}_\nu \cdot \boldsymbol{r} + \varphi_\nu^0)] + \mathrm{cc} \Bigg)^2. \end{aligned} \tag{A11}$$

Since the first term is eliminated because of the symmetry properties of k_ν, we concentrate on the remaining terms. Following a similar procedure to that adopted for $F_4^{(f)}$, we obtain $F_4^{(g)}$ to lowest order of $(\partial\varphi/\partial\boldsymbol{r}')$ as follows:

$$\begin{aligned} F_4^{(g)} \simeq \frac{1}{192} g|\boldsymbol{k}|^{10}\xi^2 \int \mathrm{d}\boldsymbol{r} \Bigg[& 2\left(\frac{\partial\varphi_1}{\partial\bar{x}}\right) \exp[\mathrm{i}(\boldsymbol{k}_1 \cdot \boldsymbol{r} + \varphi_1^0)] \\ & + \left(-\frac{\partial\varphi_2}{\partial\bar{x}} + \sqrt{3}\frac{\partial\varphi_2}{\partial\bar{y}}\right) \exp[\mathrm{i}(\boldsymbol{k}_2 \cdot \boldsymbol{r} + \varphi_2^0)] \end{aligned}$$

$$+\left(-\frac{\partial\varphi_3}{\partial\bar{x}}-\sqrt{3}\frac{\partial\varphi_3}{\partial\bar{y}}\right)\exp[\mathrm{i}(\boldsymbol{k}_3\cdot\boldsymbol{r}+\varphi_3^0)]+\mathrm{cc}\Bigg]^2. \tag{A12}$$

Substituting equations (15) and (16) into (A12), we get

$$F_4^{(g)}\simeq\frac{3}{64}g|\boldsymbol{k}|^{10}\xi^2\int\mathrm{d}\boldsymbol{r}\left[\left(\frac{\partial u_{\bar{x}}}{\partial\bar{x}}-\frac{\partial u_{\bar{y}}}{\partial\bar{y}}\right)^2+\left(\frac{\partial u_{\bar{y}}}{\partial\bar{x}}+\frac{\partial u_{\bar{x}}}{\partial\bar{y}}\right)^2+2\left(\frac{\partial u_{\bar{y}}}{\partial\bar{x}}-\frac{\partial u_{\bar{x}}}{\partial\bar{y}}\right)^2\right]. \tag{A13}$$

We ignore the last term which represents the rotation of the total system. Finally, we obtain the free energy of the pattern deformation using the new coordinate system:

$$F_4^{(g)}\simeq\frac{3}{64}g|\boldsymbol{k}|^{10}\xi^2\int\mathrm{d}\boldsymbol{r}\,(\hat{\varepsilon}_{\bar{x}\bar{x}}^{(\mathrm{ph})2}+\hat{\varepsilon}_{\bar{y}\bar{y}}^{(\mathrm{ph})2}-2\hat{\varepsilon}_{\bar{x}\bar{x}}^{(\mathrm{ph})}\hat{\varepsilon}_{\bar{y}\bar{y}}^{(\mathrm{ph})}+4\hat{\varepsilon}_{\bar{x}\bar{y}}^{(\mathrm{ph})}). \tag{A14}$$

The effective elastic constants are given by

$$\begin{aligned} C_{\bar{x}\bar{x}\bar{x}\bar{x}} &= C_{\bar{y}\bar{y}\bar{y}\bar{y}}=\tilde{g}\\ C_{\bar{y}\bar{y}\bar{y}\bar{y}} &= -\tilde{g}\\ C_{\bar{x}\bar{y}\bar{x}\bar{y}} &= 2\tilde{g}\\ C_{(\mathrm{others})} &= 0 \end{aligned} \tag{A15}$$

where

$$\tilde{g}\equiv\frac{3}{64}g|\boldsymbol{k}|^{10}\,\xi^2.$$

These tensor components correspond to the elastic constants of a system with an isotropic symmetry. However, they have an anomalous elastic nature, i.e. the Poisson's ratio becomes infinite. We must add these elastic constants to the main terms in equation (22), then these components will give minor contributions because they are higher order in $|\boldsymbol{k}|$ compared with the isotropic terms.

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