

# Alignment of primary Al<sub>3</sub>Ni phases in hypereutectic Al-Ni alloys with various compositions under high magnetic fields

WANG Qiang<sup>†</sup>, WANG ZhongYing, LIU Tie, WANG ChunJiang, ZHANG Chao & HE JiCheng

Key Laboratory of Electromagnetic Processing of Materials (Ministry of Education), Northeastern University, Shenyang 110004, China

**Al-Ni hypereutectic alloys with various compositions were solidified under various magnetic field conditions to investigate the alignment of primary Al<sub>3</sub>Ni phases. The results showed that the application of high magnetic fields could improve the homogeneity of the primary Al<sub>3</sub>Ni phase distribution and induce the alignment of primary Al<sub>3</sub>Ni phases in the direction perpendicular to the magnetic field direction to form chain-like structures. However, the alignment was different from the orientation of the Al<sub>3</sub>Ni phases. Furthermore, the degree of the alignment decreased with the increasing concentration of Ni element. This can be attributed to the combination effects of high magnetic field and alloy composition on the concentration field around the crystallized primary Al<sub>3</sub>Ni crystals.**

high magnetic fields, alloy composition, primary Al<sub>3</sub>Ni phase, alignment, solidification

It has been found that a high magnetic field, as a useful directional physical field, can induce crystal orientation and phase alignment. This indicates that it is possible for the alteration of the structures and improvement of the properties of materials with the aid of high magnetic fields during their fabrication process<sup>[1-7]</sup>. With the development and the advance of superconducting magnet technologies, the application of a high magnetic field during the solidification process of metal materials to achieve aligned microstructure has been recognized as one of the useful technologies in materials processing<sup>[8-11]</sup>. Al-Ni alloys with non-eutectic compositions normally contain a wide range of volume percent eutectic which can meet higher technical demands on material properties and have been an important branch of research on functionally graded materials processing<sup>[12-15]</sup>. Studies on the mechanical properties of fiber-reinforced composite suggested that for Al-Ni alloys with an aligned-Al<sub>3</sub>Ni structure, their tensile and fatigue properties were remarkably improved in the direction along which the Al<sub>3</sub>Ni phases were aligned<sup>[16]</sup>.

Therefore, it is a promising method to obtain Al<sub>3</sub>Ni-aligned microstructure so as to improve the properties of the Al-Ni alloys by controlling the formation process of the structure in the alloys. In 1981, Mikelson and Kar-klín<sup>[17]</sup> firstly reported that high magnetic fields of 0.5–1.5 T could induce the directional growth of primary Al<sub>3</sub>Ni phases in Al-10 wt.%Ni alloy in the direction perpendicular to the field direction. Wang et al.<sup>[18]</sup> observed that with high magnetic fields the primary Al<sub>3</sub>Ni phases in Al-10 wt.%Ni alloy were oriented to *c*-axis along the magnetic field direction. Li et al.<sup>[19]</sup> investigated the effects of magnetic flux density and temperature on the degree of the orientation of primary Al<sub>3</sub>Ni phases in Al-8 wt.%Ni and Al-10 wt.%Ni alloys.

Previous works have mainly focused on the orientation of Al<sub>3</sub>Ni crystals. However, little work has been

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<sup>†</sup>Corresponding author (email: wangq@epm.neu.edu.cn)

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investigated on the alignment of primary  $\text{Al}_3\text{Ni}$  phases. Considering the fact that there are essential differences between crystal orientation and phase alignment, it is necessary to study the alignment of primary  $\text{Al}_3\text{Ni}$  phase with a magnetic field. In this work, the effects of high magnetic fields on the alignment of primary  $\text{Al}_3\text{Ni}$  phase in hypereutectic Al-Ni alloys with various compositions and their dependence on the concentration of Ni element have been investigated. The relationship between the crystal orientation and phase alignment of  $\text{Al}_3\text{Ni}$  phases was discussed.

## 1 Experimental

Three kinds of Al-Ni alloys with different compositions were prepared by induction-melting 99.99% Al and 99.999% Ni in graphite crucibles under vacuum. The alloy compositions were chemically analyzed to be Al-6.8 wt.%Ni, Al-7.4 wt.%Ni and Al-8.5 wt.%Ni, which were all hypereutectic according to the Al-Ni phase diagram as shown in Figure 1. The obtained ingots were machined into cylindrical specimens of 9 mm in diameter and 15 mm in length.

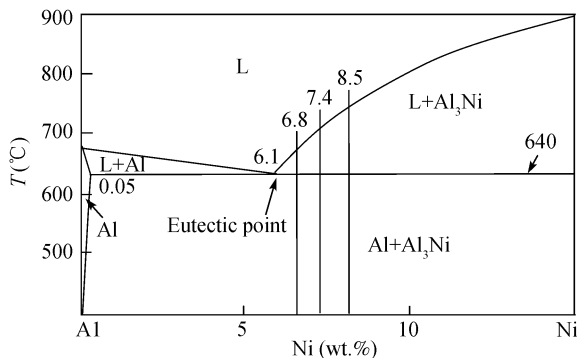


Figure 1 Phase diagram of Al-Ni system.

The schematic view of experimental apparatus is shown in Figure 2. A superconducting magnet can generate a uniform axial magnetic field with an adjustable magnetic flux density up to 12 T at the centre of a bore of 100 mm in diameter. A resistance furnace, in which the temperature can reach 1200°C, was installed in the bore of the magnet for melting and solidifying specimens. The temperature of the furnace was controlled by an R-type thermocouple. The specimens in alumina crucibles were fixed at the centre of the furnace, heated to 800°C at a heating rate of 5°C/min and held at the same temperature for 30 min to ensure its homogeneity. Then, the temperature was cooled down to 550°C at a cooling

rate of approximately 10°C/min. Finally, the specimens were cooled to the room temperature by turning off the DC power source. The magnetic field conditions are summarized in Table 1. The magnetic field-treated specimens were cut along longitudinal section which was parallel to the magnetic field direction, polished and etched with a hydrofluoric acid solution for metallographic examination. To investigate the crystal orientation of primary  $\text{Al}_3\text{Ni}$  phase in the alloys, X-ray diffraction (XRD) with  $\text{Cu K}\alpha$  irradiation was also performed on this section.

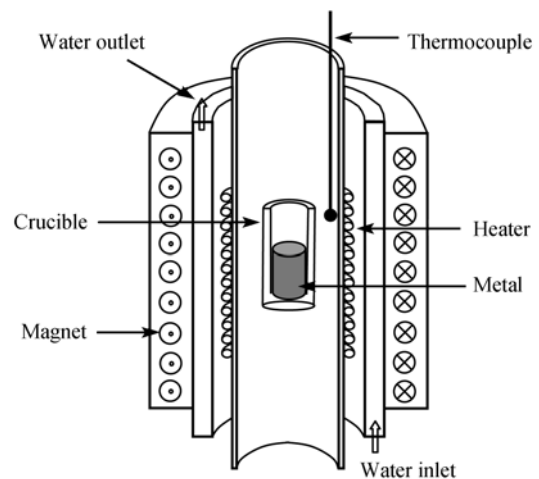


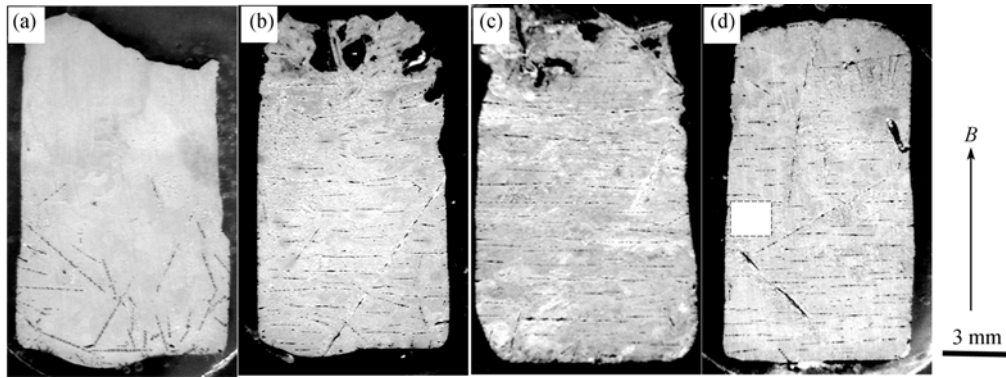
Figure 2 Schematic illustration of the experimental apparatus.

Table 1 Experimental conditions of high magnetic fields

Magnetic flux density, $B$ (T)	0	4.4	8.8	11.5
Al-6.8 wt.%Ni	√	-	√	√
Al-7.4 wt.%Ni	√	√	√	√
Al-8.5 wt.%Ni	√	√	√	√

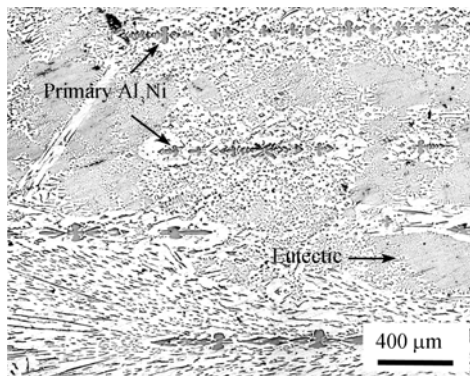
## 2 Results

The as-solidified macrostructures on the longitudinal section of Al-6.8 wt.%Ni alloy specimens solidified under various magnetic field conditions are shown in Figure 3. In these photographs, only primary  $\text{Al}_3\text{Ni}$  phases (dark strip-like one) and Al- $\text{Al}_3\text{Ni}$  eutectic (bright one) can be observed, although there are some holes (dark nubby ones) can be found at the upper regions of the specimens. It can be found from Figure 3(a) that without a high magnetic field present, primary  $\text{Al}_3\text{Ni}$  phases are mostly gathered at the lower part of the specimen, due to the rather larger difference in densities between the precipitated  $\text{Al}_3\text{Ni}$  ( $\rho_{\text{Al}_3\text{Ni}}=3950 \text{ kg/m}^3$ ) phases and melt ( $\rho_{\text{Al}}=2360 \text{ kg/m}^3$ ) surrounding it. Meanwhile, the  $\text{Al}_3\text{Ni}$  phases show a randomly aligned structure. When



**Figure 3** Macrostructures of Al-6.8wt.%Ni alloy specimens solidified under various magnetic field conditions. (a) 0 T; (b) 4.4 T; (c) 8.8 T; (d) 11.5 T.

imposing high magnetic fields, the primary  $\text{Al}_3\text{Ni}$  phases are homogeneously distributed in the specimens and aligned in the direction perpendicular to the imposed magnetic field. Furthermore, the high magnetic field shows an increasing effect on the alignment with the increase of magnetic flux density (Figures 3(b)–(d)). Figure 4 shows the typical microstructure of the specimen treated at 11.5 T (indicated by the dashed rectangle in Figure 3(d)), in which the primary  $\text{Al}_3\text{Ni}$  phase is clearly aligned with its plane perpendicular to the magnetic field direction.

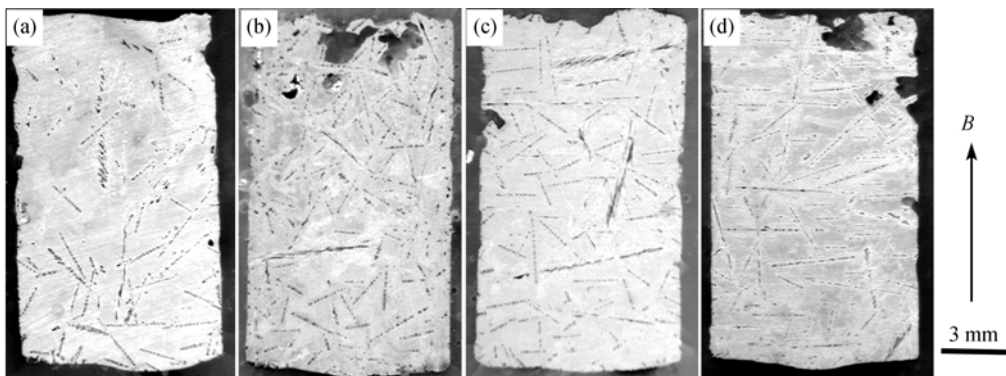


**Figure 4** Microstructure of the Al-6.8 wt.%Ni alloy indicated by the dashed rectangle in Figure 3(d).

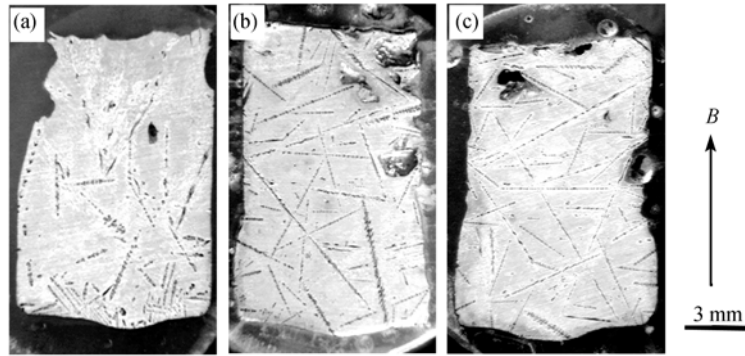
The macrostructures on the longitudinal section of Al-7.4 wt.%Ni alloy specimens solidified under various magnetic field conditions are shown in Figure 5. It can be seen that similar to their Al-6.8 wt.%Ni counterparts, the primary  $\text{Al}_3\text{Ni}$  phase is distributed from nonhomogeneously to homogeneously in the specimens and aligned gradually in the direction perpendicular to the imposed magnetic field with the application of high magnetic fields. However, the degree of magnetic alignment of Al-7.4 wt.%Ni alloy is much weaker than that of Al-6.8 wt.%Ni alloy treated under the same magnetic field conditions.

Figure 6 shows the macrostructures on the longitudinal section of Al-8.5 wt.%Ni alloy specimens solidified under various magnetic field conditions. The application of high magnetic fields shows the similar effects on the distribution of the primary  $\text{Al}_3\text{Ni}$  phases. But the high magnetic field gives a weaker effect on the alignment for the case of Al-8.5 wt.%Ni alloy than the other two cases, with only a few phases being aligned with the magnetic fields.

From the results as mentioned above, it can be concluded that the application of high magnetic fields can



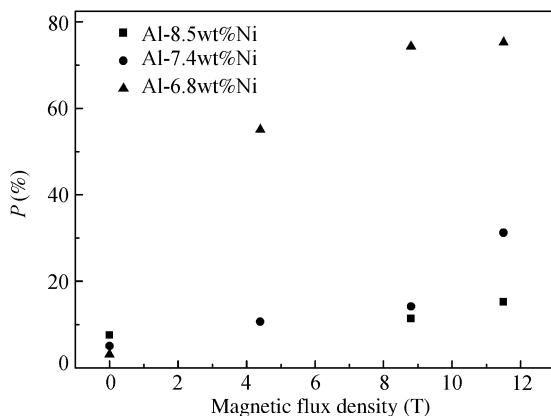
**Figure 5** Macrostructures of Al-7.4 wt.%Ni alloy specimens solidified under various magnetic field conditions. (a) 0 T; (b) 4.4 T; (c) 8.8 T; (d) 11.5 T.



**Figure 6** Macrostructures of Al-8.5 wt.%Ni alloy specimens solidified under various magnetic field conditions. (a) 0 T; (b) 8.8 T; (c) 11.5 T.

produce similar effects on the distribution but various effects on the alignment of the primary  $\text{Al}_3\text{Ni}$  phases with changes in alloy composition. This indicates that the composition of Al-Ni alloys can influence the effects of high magnetic fields on the alignment of the primary  $\text{Al}_3\text{Ni}$  phase. In order to evaluate the degree of alignment of the primary  $\text{Al}_3\text{Ni}$  phases, an estimate of the alignment extent can be made by  $P=N_p/N$ , where  $N_p$  and  $N$  are the number of aligned primary  $\text{Al}_3\text{Ni}$  phases and the total number of the  $\text{Al}_3\text{Ni}$  phases per  $\text{cm}^2$ , respectively (generally, a primary  $\text{Al}_3\text{Ni}$  strip can be treated as an aligned one if the angle between its plane and the direction perpendicular to the magnetic field direction is below  $5^\circ$ ).

The changes of  $P$  in the alloys of Al-6.8 wt.%Ni, Al-7.4 wt.%Ni, and Al-8.5 wt.%Ni as a function of magnetic flux density are shown in Figure 7. For a certain alloy, with the increase of magnetic flux density, the degree of alignment of the primary  $\text{Al}_3\text{Ni}$  phase increases. However, this increase is strongly dependent on the composition of the alloys. For example, under the magnetic field of 11.5 T, the value of  $P$  decreases from



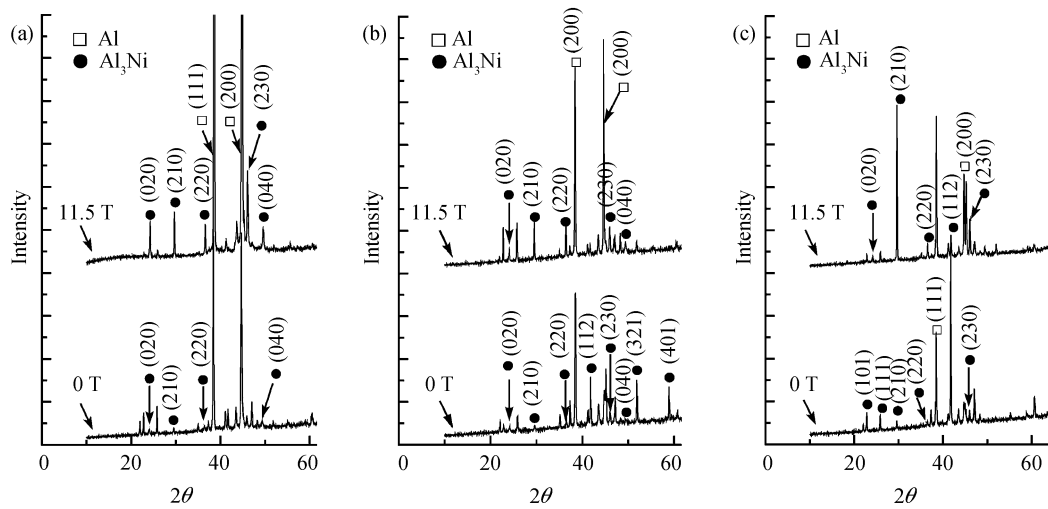
**Figure 7** Changes of  $P$  in Al-6.8 wt.%Ni, Al-7.4 wt.%Ni, and Al-8.5 wt.%Ni alloys as a function of magnetic flux density.

about 75% for Al-6.8 wt.%Ni to about 30% for Al-7.4 wt.%Ni, and to about 10% for Al-8.5 wt.%Ni. This indicates that the effect of high magnetic field on the alignment of the primary  $\text{Al}_3\text{Ni}$  phases in hypereutectic Al-Ni alloys increases with the decrease of the alloy composition.

For determination of the effects of alloy composition on the alignment of the primary  $\text{Al}_3\text{Ni}$  phases, XRD was performed on the longitudinal section for all three composition alloys without or with a high magnetic field of 11.5 T present and the obtained XRD patterns are shown in Figure 8. It can be observed from Figure 8 that the diffraction peaks of the  $\text{Al}_3\text{Ni}$  phases in all alloys obtained at 0 T exhibit almost all crystalline faces and indicate that the primary  $\text{Al}_3\text{Ni}$  phase at 0 T has a randomly oriented structure. In the cases of 11.5 T, the intensities of the (020), (210), (220), (230) and (040), i.e. ( $hk0$ ) planes are more or less enhanced, together with that the intensities of other peaks are suppressed systematically and even disappear in comparison with that of the 0 T cases. This indicates that the  $c$ -axis of  $\text{Al}_3\text{Ni}$  crystal is oriented to the magnetic field direction. The application of high magnetic fields can induce both the alignment and orientation of primary  $\text{Al}_3\text{Ni}$  phases, but the former is dependent on and the later is independent of the alloy composition. Therefore, it can be deduced that there should be different mechanisms to explain the phenomena of alignment and orientation induced by high magnetic fields.

### 3 Discussion

From the results of XRD, it has been found that the application of high magnetic fields can promote the orientation of primary  $\text{Al}_3\text{Ni}$  phase with its  $c$ -axis along the field direction. Meanwhile, this orientation is independ-



**Figure 8** X-ray diffraction patterns of Al-Ni alloys on the section parallel to the magnetic field direction. (a) Al-6.8wt%Ni; (b) Al-7.4wt%Ni; (c) Al-8.5wt%Ni.

ent of the alloy composition. When a substance is subjected to a uniform magnetic field, the energy for magnetization of the substance is described by<sup>[6]</sup>

$$E = -\frac{1}{2\mu_0} \chi V B^2, \quad (1)$$

where  $\mu_0$  is the vacuum permeability,  $V$  is the volume of the substance,  $B$  is the magnetic flux density and  $\chi$  is the magnetic susceptibility per unit volume, respectively. If the substance has a magnetic anisotropy where the magnetic susceptibility is different in each crystal direction, anisotropy energy will arise due to the difference of the magnetization energy along various crystal directions<sup>[2]</sup>.

$$\Delta E = -\frac{1}{2\mu_0} \Delta\chi V B^2, \quad (2)$$

where  $\Delta\chi$  is the anisotropy of the paramagnetic susceptibility. From eq. (2), it can be found that high magnetic fields can drive the crystal to a stable orientation so as to decrease the magnetization energy. According to the analysis<sup>[19]</sup>,  $\text{Al}_3\text{Ni}$  is a paramagnetic material and the  $c$ -axis of it has the biggest value of magnetic susceptibility. During the solidification of a hypereutectic Al-Ni alloy under a high magnetic field, the primary  $\text{Al}_3\text{Ni}$  phase of the alloy will be oriented to the  $c$ -axis along the magnetic field direction. From eq. (1) it can be found that the alloy composition has no effect on the magnetization energy, and, therefore, does not affect the orientation of  $\text{Al}_3\text{Ni}$  phase. This further confirms the experimental results mentioned above.

Under high magnetic fields, the primary  $\text{Al}_3\text{Ni}$  phases are distributed homogeneously in the alloys in spite of alloy composition. This can be attributed to the braking effects of high magnetic fields on the convection in the melt in terms of Lorentz force<sup>[20]</sup>.

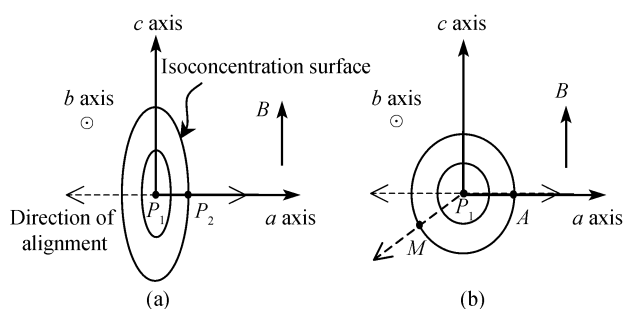
During the solidification of a hypereutectic Al-Ni alloy, as a primary phase, the firstly precipitated  $\text{Al}_3\text{Ni}$  crystal  $P_1$  crystallizes from the melt and could grow along the  $b$ -axis of the crystal, because the  $[010]$  direction is the preferred crystallographic growth direction<sup>[21]</sup>. In this case, a concentration gradient would appear near the region around the  $b$ -axis of crystal  $P_1$ . In the case of without a magnetic field present, because the convective velocity ( $\approx 10^{-6}$  m/s) is greater than the diffusion velocity ( $\approx 10^{-7}$  m/s)<sup>[22,23]</sup>, the solute redistribution at solid-liquid interface should be dominated by convective mass transfer. This means that the concentration field around the crystal  $P_1$  can be rapidly eliminated by the convection. Consequently, the followed crystals would mainly nucleate in the upstream direction similar to dendrite growth mode, and grow also with their  $[010]$  direction as the preferred crystallographic growth direction. In this work, differing from directional solidification, the convection in the melt is inordinate and thus results in a randomly aligned primary  $\text{Al}_3\text{Ni}$  structure.

When a high magnetic field is applied, the mass transfer in the melt changes from convection-controlled to diffusion-controlled due to the braking effect of the high magnetic field on the convection<sup>[24]</sup>. The diffusivity coefficient of charged particle under magnetic field can be identified as<sup>[25]</sup>

$$D_{\perp} \approx D / (1 + \omega^2 \tau^2), \quad (3)$$

$$D_{\parallel} = D, \quad (4)$$

where  $\tau$  is the mean collision time of the charged particles,  $D$  is the diffusion coefficient without magnetic field,  $\omega = qB/m$ ,  $q/m$  is charge-mass ratio of the charged particle and subscripts  $\perp$  and  $\parallel$  indicate the directions parallel and perpendicular to the magnetic field, respectively. From eqs. (3) and (4), it can be found that the diffusion is suppressed in the direction perpendicular to the magnetic field, but does not change in the direction parallel to the magnetic field. Thus, an elliptic concentration field with its long axis parallel to the magnetic field direction would be expected, as shown in Figure 9(a). According to the nucleation theory, nucleation generally occurs in the region where there are enough concentration fluctuations, thus, the followed crystals such as  $P_2$ , would mostly nucleate in the short axis of the ellipse of concentration field which is perpendicular to the magnetic field direction (Figure 9(a)). With this trend, a chain-like structure resulting from the alignment of primary  $Al_3Ni$  in the direction perpendicular to the magnetic field direction as discussed above is obtained.



**Figure 9** Schematic illustrations of concentration gradient around crystallized primary  $Al_3Ni$  crystals in Al-Ni alloys under high magnetic fields. (a) Lower Ni concentration; (b) higher Ni concentration.

Using above discussed mechanism, the alignment of primary  $Al_3Ni$  phases under high magnetic field conditions can be successfully explained. However, the experimental results shown in Figures 3–6 illustrate that this alignment strongly depends on the alloy composition, suggesting that the alloy composition is another key factor that can affect the alignment of primary  $Al_3Ni$  phase other than high magnetic field and must be taken into account. As can be seen in Figure 9(a), under high magnetic field conditions, the concentration field around a crystallized crystal is ellipsoid. However, with the increase of Ni concentration, the difference in lengths of the long and short axes of the ellipse decreases gradually,

that is, the high magnetic field shows a decreasing effect on solute redistribution (Figure 9(b)). It can be deduced from eqs. (3) and (4) that with the increase of Ni concentration, the mean collision time  $\tau$  decreases and thus results in an increase of  $D_{\perp}$  to values near  $D_{\parallel}$ . In this case, the followed crystals will crystallize randomly at the site either in the long or short axis, such as sites marked in Figure 9(b) as A and M, and reduce the degree of the alignment of primary  $Al_3Ni$  phases. From the discussion above, it can be concluded that for fabricating aligned functionally materials using high magnetic fields, alloy composition is another key parameter as well as magnetic flux density to control the fabricating process.

## 4 Conclusions

The effects of high magnetic fields on the alignment of primary  $Al_3Ni$  phases in hypereutectic Al-Ni alloys with various compositions and their dependence on the concentration of Ni have been investigated. Under high magnetic field conditions, in spite of alloy composition, the primary  $Al_3Ni$  phases are distributed homogeneously in the alloys due to the braking effects of Lorentz force on the convection and oriented to its  $c$ -axis along the magnetic field direction. Furthermore, the primary  $Al_3Ni$  phases are aligned in the direction perpendicular to the imposed magnetic field, but this alignment strongly depends on the alloy composition, i.e., the degree of the alignment decreases with increasing Ni concentration.

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