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Magnetic structure of γ-Fe precipitates in Cu: II. Transverse spin component

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Abstract. The 100 magnetic diffuse peak of γ-Fe precipitates in Cu is re-examined by means of neutron diffraction. The distribution of the diffuse scattering intensity extends along the [010] direction. A magnetic structure is proposed with transverse spin components along the *c* axis in addition to the coexistence of the double-Q spin structure and periodic lattice distortion on the *c* plane previously reported.

1. Introduction

The magnetic structure of FCC (γ)-Fe precipitates in Cu was first investigated by Abrahams *et al* [1] by means of neutron diffraction. They observed the 110 and 001 magnetic peaks at 4.2 K and proposed a longitudinal-first-kind antiferromagnetic (L-AF-1) structure, in which the magnetic moments on the (001) plane are oriented along the [001] direction and they couple ferromagnetically with other moments in the same plane. The moments on different (001) planes couple antiparallel along the [001] axis. Since the 001 magnetic peak was very weak and is prohibited for the L-AF-1 structure, they explained this peak as a spin component inclined from the [001] axis. After that first investigation, the 001 magnetic peak, which is mostly diffuse scattering, was commonly observed for the FCC systems with the L-AF-1-type spin structure such as Fe–Ni–Cr [2], Fe–Mn [3], Co–Mn [4], Mn–Cu [5] and Mn–Ni [6] alloys. Since the magnetic structure of γ-Fe alloys (Fe–Mn and Fe–Ni–Cr) was reported to be the same as that of the γ-Fe precipitates in Cu, no one doubted this magnetic structure for many years. However, Ehrhart *et al* [7] found a reduction in the lattice symmetry below the Néel temperature for γ-Fe precipitates in Cu. Subsequently one of the present authors and a colleague [8] made more precise x-ray measurements of the lattice structure and found that the FCC lattice undergoes a structural phase transition to a sinusoidally modulated lattice structure at the onset of antiferromagnetic ordering. In this low-temperature phase, the atomic positions on the *c* plane are described by a shear wave propagating along the [110] direction. Along the *c* axis, the lattice slightly contracts uniformly by 0.17%. Since the lattice deformation on the *c* plane is far larger (about 2.5%) than that along the *c* axis, the local lattice structure is roughly orthorhombic ($a^+ > c > a^-$).

This lattice structure is incompatible with the magnetic structure reported by Abrahams *et al* [1] and compelled us to reinvestigate the magnetic structure of γ-Fe precipitates in Cu [9]. We do not repeat here the detailed analysis of the data, but, using the peculiar diffraction patterns of the 110, 120 and 210 magnetic peaks, a magnetic structure superimposed on the periodic lattice modulation was determined in our previous paper, as shown in figure 1. In this figure, which is a schematic illustration, the magnetic moments are on the *c* plane. This is essentially the double-Q wave structure which is composed of two components of the L-AF-1 structures; one propagating along the [100] axis and the other along the [010] axis. The magnetic moments are also modulated in space along the [110] direction because of the coupling with the periodic lattice distortion [10]. However, in our previous paper we did not take the 001 magnetic peak into consideration to determine this magnetic structure because this peak is very weak and comes from the transverse-first-kind antiferromagnetic (T-AF-1) structure in which the direction of moments is perpendicular to the propagation vector of the antiferromagnetic wave. It is necessary to explain the 001 magnetic peak without inconsistency with the basic double-Q spin structure and periodic lattice distortion on the c plane determined above. To do this, the 001 magnetic peak was carefully re-examined by means of neutron diffraction. From these data, we propose a new explanation of the 001 magnetic diffuse peak of γ-Fe precipitates in Cu.

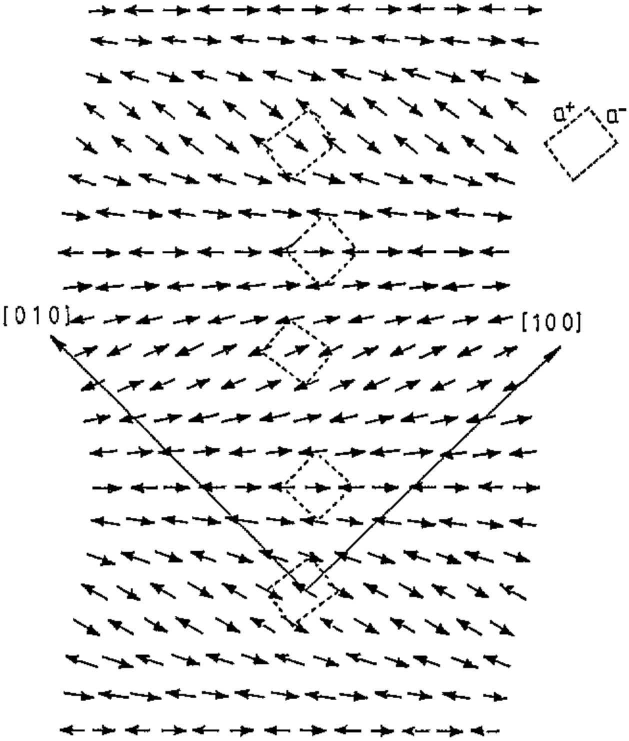

Figure 1. The periodic lattice distortion and magnetic structure of γ-Fe precipitates on the c plane determined in our previous studies.

2. Measurement

A single crystal of a Cu–Fe supersaturated alloy with 2.8 at.% Fe was grown by the Bridgman method. After the homogenization anneal at 1050 °C for 16 h, the specimen was quenched into water; then a precipitation anneal at 650 °C for 72 h was performed. The mean diameter of the precipitates is estimated to be 60 nm using the empirical equation determined by Borrelly *et al* [11]. Because the lattice parameter of γ-Fe (0.3577 nm at 70 K) is close to that of Cu (0.3603 nm), the Fe precipitates are coherent with the Cu lattice and, therefore, the cubic axes of each precipitate are parallel to those of the Cu matrix and those of every other precipitate. Thus, if we grow the precipitates in a Cu(Fe) single crystal, we can regard all the precipitates as a single crystal of γ-Fe. However, when the structural phase transition takes place, normally all possible variants are equally probable. In order to simplify the problem, the present measurements were performed under a uniaxial stress of about 300 kgf cm^{-2} loaded along the direction perpendicular to the scattering plane. Under this condition, a variant with the *c* plane perpendicular to the uniaxial stress direction grows predominantly [12]. In the present measurements, about 85% of the precipitates have the *c* plane in the scattering plane. This value was estimated from the intensity ratio of the 110 and 210 magnetic peaks.

Neutron scattering measurements were performed at the triple-axis spectrometer HQR installed at the thermal guide of the JRR–3, JAERI, Tokai. Some data were taken at the HB-1 triple-axis spectrometer at HFIR, Oak Ridge. In order to minimize the higher-order contamination from the Cu Bragg peaks, a thick PG filter was used for both measurements.

3. Experimental data and analysis

3.1. *Confirmation of the basic magnetic structure on the c plane*

In our previous paper, the magnetic structure was determined using the data for a multi-variant specimen. Under uniaxial stress along the [001] direction, the variant with the *c* plane perpendicular to the uniaxial stress direction grows predominantly below T_N. There are still two variants: one with the periodic lattice wave propagating along the [110] direction and the other along the $[1\bar{1}0]$ direction, both on the *c* plane. However, this condition is still far simpler for re-examining the magnetic structure.

In figure 2, the observed scattering intensity maps around the 110, 210 and 120 reciprocal-lattice points (RLPs) are given. Peak intensities at 210 and 120 are higher than that at 110 in spite of the fact that the magnetic form factor of the latter is about twice that of the former. This is due to the anisotropic distribution of the variants under the uniaxial stress. As shown in our previous paper [9], the 110 magnetic peak comes from the variants with the *c* plane perpendicular to the scattering plane. Uniaxial stress diminishes the volume fraction of these variants, resulting in the weak 110 magnetic peak. Both the 210 and the 120 peaks are composed of components extending along the [110] and $[1\bar{1}0]$ directions, indicating that each variant has a double-Q wave structure in which the L-AF-1-type structures propagating along the [100] and [010] directions are coexistent. (See the data analysis in [9].) Elongated diffraction patterns consist of satellite reflections centred around the 210 and 120 RLPs although each peak is not resolved owing to the poor resolution. However, the intensity of the $[1\bar{1}0]$ component is extremely asymmetric with respect to the RLP. Actually, only the satellite peaks of one side are observable. This is explained as follows. If only the periodic lattice distortion exists, the satellite peak intensities of both sides should be equal as in the case of x-ray diffraction data [8]. However, when the periodic lattice distortion and spin modulation coexist, satellite peak intensities of both sides become asymmetric. Let us write the atomic positions and spins as

$$r_n = na + \Delta \sin(Q \cdot na) \qquad S_n = [S_0 + \sigma \cos(q \cdot na)] \cos(n\pi).$$

Then the neutron scattering amplitude $F(\boldsymbol{K})$ is given as [9]

$$\begin{aligned} F(\boldsymbol{K}) = (\tfrac{1}{2}S_0)\Big(& J_0(\boldsymbol{K}\cdot\boldsymbol{\Delta})\delta(\boldsymbol{K}\pm\boldsymbol{t})+\sum J_n(\boldsymbol{K}\cdot\boldsymbol{\Delta})[\delta(\boldsymbol{K}\pm\boldsymbol{t}+n\boldsymbol{Q})+(-1)^n\delta(\boldsymbol{K}\pm\boldsymbol{t}-n\boldsymbol{Q})]\Big) \\ &+ (\tfrac{1}{4}\sigma)\Big(J_0(\boldsymbol{K}\cdot\boldsymbol{\Delta})\delta(\boldsymbol{K}\pm\boldsymbol{t}\pm\boldsymbol{q})+\sum J_n(\boldsymbol{K}\cdot\boldsymbol{\Delta})[\delta(\boldsymbol{K}\pm\boldsymbol{t}\pm\boldsymbol{q}+n\boldsymbol{Q}) \\ &+ (-1)^n\delta(\boldsymbol{K}\pm\boldsymbol{t}\pm\boldsymbol{q}-n\boldsymbol{Q})]\Big) \end{aligned}$$

where S_0 indicates the L-AF-I spin component without spin modulations, σ and $\boldsymbol{\Delta}$ are amplitudes of the modulation waves in spin and lattice, and $\boldsymbol{q}$ and $\boldsymbol{Q}$ are the wavevectors of spin and lattice modulation, respectively. (Experimental data show that $\boldsymbol{q}=\boldsymbol{Q}$.) $\boldsymbol{K}$ and $\boldsymbol{t}$ indicate the scattering vector and a half the RL vector; J_n is the nth-order spherical Bessel function. The intensity ratio of the first satellites on both sides is approximately written as

$$\frac{I(+\boldsymbol{Q})}{I(-\boldsymbol{Q})} \simeq \frac{[\frac{1}{4}\sigma J_0(\boldsymbol{K}\cdot\boldsymbol{\Delta})-\frac{1}{2}S_0J_1(\boldsymbol{K}\cdot\boldsymbol{\Delta})]^2}{[\frac{1}{4}\sigma J_0(\boldsymbol{K}\cdot\boldsymbol{\Delta})+\frac{1}{2}S_0J_1(\boldsymbol{K}\cdot\boldsymbol{\Delta})]^2}.$$

At the 120 and 210 RLPs, the argument of the J_n, i.e. $\boldsymbol{K}\cdot\boldsymbol{\Delta}$, is far larger for the wave propagating along the $[1\bar{1}0]$ direction than for that along the $[110]$ direction because the lattice has a transverse wave. ('For the wave propagating along the $[1\bar{1}0]$ direction, $\boldsymbol{K}\cdot\boldsymbol{\Delta} = |K\Delta|\cos(18.435) = 0.95|K\Delta|$ while, for the $[110]$ direction, $\boldsymbol{K}\cdot\boldsymbol{\Delta} = |K\Delta|\cos(71.565) = 0.32|K\Delta|$, where $|K\Delta| \simeq 1.76$.') Then the ratio $J_1(\boldsymbol{K}\cdot\boldsymbol{\Delta})/J_0(\boldsymbol{K}\cdot\boldsymbol{\Delta})$ which contributes mainly to the anisotropy of the satellite intensities is about four times larger for the $[1\bar{1}0]$ direction than for the $[110]$ direction.

In the previous experimental results for the multi-variant specimen, there were additional components which extended along the [010] direction for 210 and the [100] direction for 120. These were again the contributions from the variants with the c plane perpendicular to the scattering plane as well as for 110 '(see figure 9(b) in [9])'. These variants are diminished to about 15% of the sample volume by the uniaxial stress in the present measurement, and an intensity contribution of about 20% of the 110 peak is expected. This is too weak to be observed under the coexistence with the strong magnetic peak of predominant variants.

Thus, these data obtained on a sample under uniaxial stress are fully consistent with the magnetic structure on the c plane as given in figure 1 which was determined by the previous experiment on the multi-variant specimen.

3.2. The 100 magnetic diffuse peak

The index of 001 for this peak is commonly used. Actually, in the case of the Mn–Cu alloy, the magnetic diffuse peak is observed at 001 if we take the tetragonal axis as the c axis. In the present paper, the periodic lattice modulation propagates on the c plane. Since the c axis of the predominant variant is perpendicular to the scattering plane, the observed diffuse peaks should be indexed as 100 and 010 peaks in the present case.

The scattering-intensity contour map observed at 12 K around the 100 RLP is given in figure 3. The diffraction line profile does not indicate a well defined Bragg peak, but rather a broad diffuse peak. In order to see this, the instrumental resolution determined using the Bragg peak of the Cu host is shown. (In x-ray diffraction data, the Bragg peak of γ-Fe precipitates with this size has a broader linewidth by about 30% than that of the Cu host, probably for several reasons such as the smallness of the size, the particle

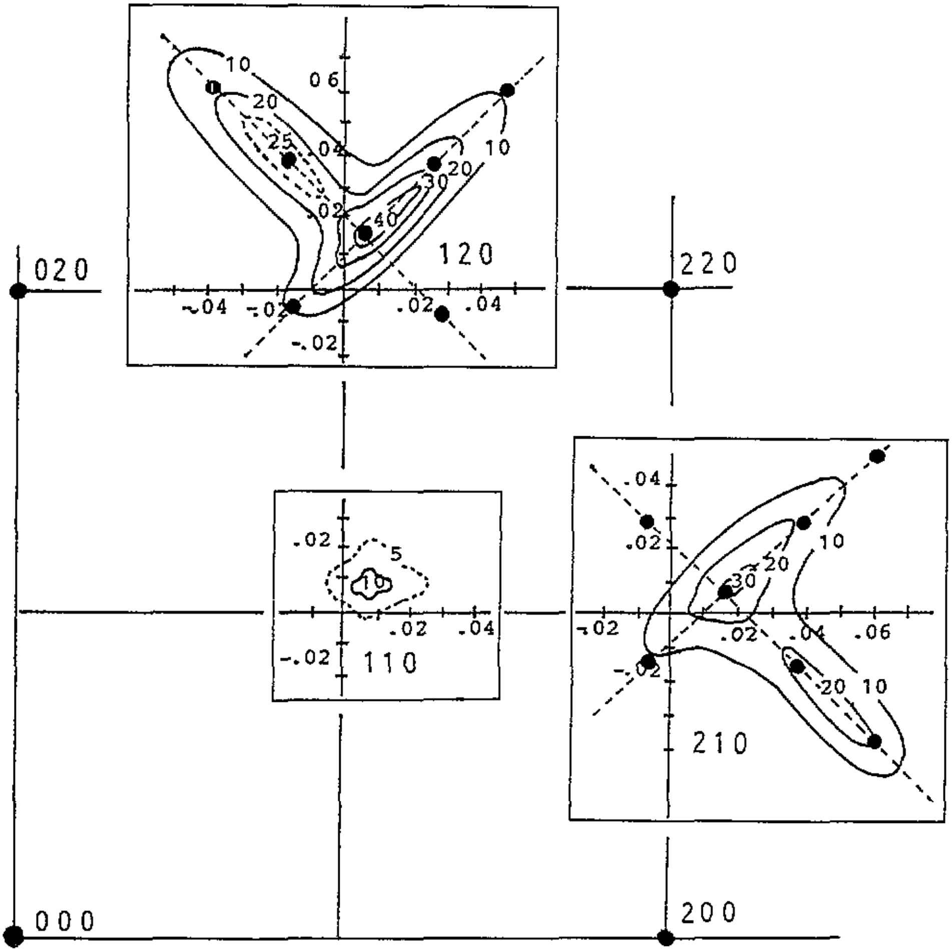

Figure 2. Scattering intensity contour maps of 110, 120 and 210 magnetic Bragg peaks observed under uniaxial stress loaded along the [001] direction. Maps are drawn in a RL frame of a Cu matrix with an enlarged scale around the RLPs. The full circles indicate the satellite peak position estimated from the x-ray data.

size distribution and the strains at the interface region between precipitate and host.) The intensity distribution is roughly symmetric with respect to the 100 RLP although all the other magnetic Bragg peaks are extremely asymmetric. This is another reason why we consider this peak separately. One peculiar feature of this diffraction pattern is an extension of the intensity along the [010] direction [13].

Diffuse scattering around the 010 RLP was also studied. Almost the same diffraction contour map which has the extension axis along the [100] direction was observed.

The diffuse peak intensity decreases monotonically with increasing temperature and almost disappears at around T_N. This behaviour is just the same as those for the 120 and 210 peaks and confirms that all peaks observed here are of magnetic origin.

The inelastic scattering of neutrons around the 100 RLP was also investigated. No appreciable inelastic scattering was observed at the lowest temperature (10 K). Therefore we believe that the 100 diffuse peak is essentially elastic in origin.

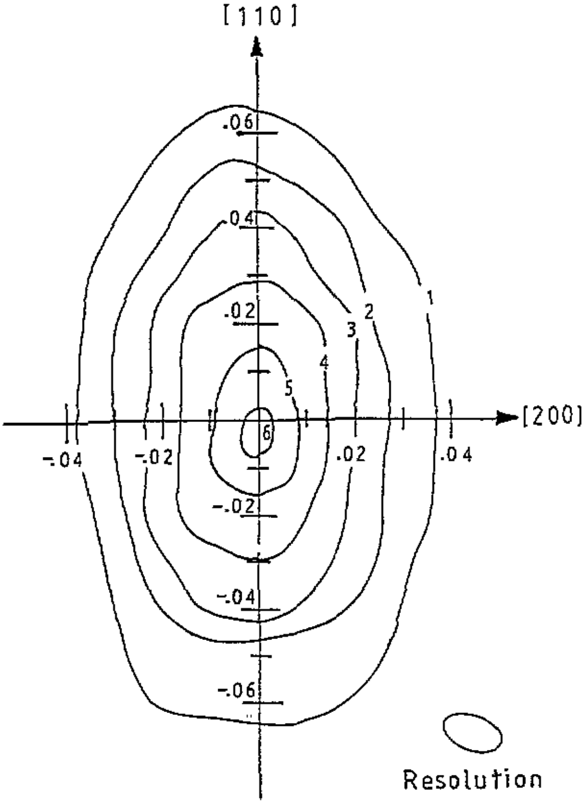

Figure 3. Observed intensity contour map of the 100 diffuse peak. Instrumental resolution determined using the Bragg peak of the Cu host is shown.

3.3. Model of the 100 diffuse scattering

In the previous measurements using the multi-variant specimen, the 100 diffuse peak intensity was very weak compared with that of the 110 magnetic peak. The observation of a far stronger 100 diffuse peak intensity in the present measurement indicates that most of 100 diffuse scattering comes from the predominant variants. Since the 100 magnetic peak is prohibited for the L-AF-1-type structure, this indicates the existence of the T-AF-1 spin component. There are two choices for the T-AF-1-type structure which contributes to the 100 magnetic peak of the predominant variant. One is that the magnetic moments are parallel to the [010] axis and the other is that they are parallel to the [001] axis (c axis). However, if we adopt the former, both the L-AF-1 and the T-AF-1-type structures are coexistent on the c plane and we have to consider that not only the moment direction but also the moment magnitude are modulated on the c plane and that atoms in equivalent lattice sites have different sizes of magnetic moments.

It is rather natural to assume that the spin is along the c axis for this lattice structure. This is deduced from an analogy to the γ-Mn alloy. The γ-Mn–Ni alloy with relevant Ni concentration has an orthorhombic lattice structure ($a > b > c$). The magnetic structure of this phase was recently determined conclusively by Takeuchi [6] to be the L-AF-1 structure with double-Q waves which have propagation directions along the b and c axes. The magnitude of each spin component closely relates to the atomic distances of the orthorhombic lattice, i.e. $|m_c| > |m_b| > |m_a| = 0$. In the case of γ-Fe precipitates, the lattice has the orthorhombic structure in the node of the sinusoidally modulated phase ($a^+ > c > a^-$). Then from the analogy to the γ-Mn–Ni alloy, γ-Fe should have the c-axis spin component at the node of the periodic lattice wave. However, as pointed out in the previous paper, the special feature of the diffraction pattern of the 110 Bragg peak is that there is no scattering intensity extending along the $[1\bar{1}0]$ direction. This definitely indicates that there is no c-axis spin component with the L-AF-1 structure. Thus, the c-axis spin component presumably has the T-AF-1-type structure.

Now let us apply a rather reasonable rule which is common to the systems with the AF-1-type spin structure; the antiferromagnetic (ferromagnetic) spin coupling is favoured in the plane with the shorter (longer) atomic distance. If we apply this rule to the c-axis spin component, the spin configuration shown in figure 4 is obtained for the periodically modulated lattice. In this figure, the larger circles are the atoms on the c plane and the small circles are on the lower half of the c plane. Open circles indicate the up spins, and full circles the down spins. When we travel parallel to the [100] axis, averaged magnetic moments on successive (100) planes are zero in region A but finite in region B. Each precipitate includes several sets of these regions.

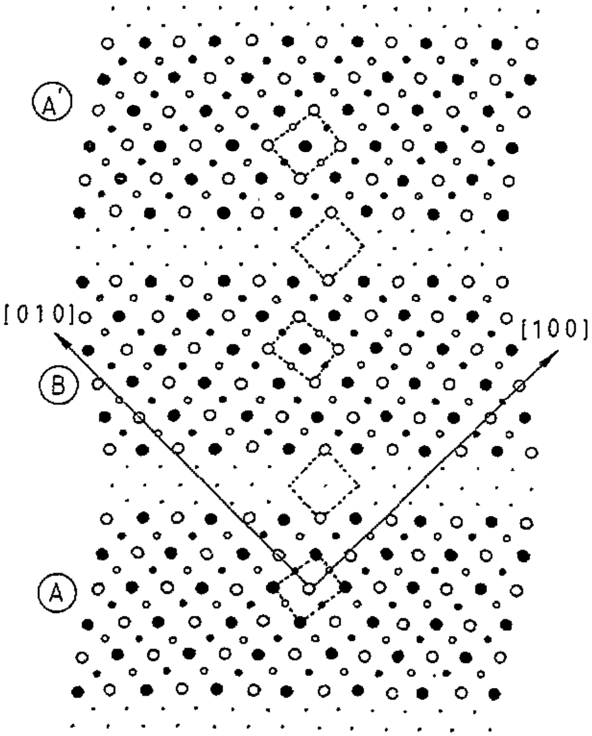

Figure 4. A model of the c-axis spin component which explains the 100 and 010 magnetic diffuse peaks.

The diffraction pattern expected from this spin configuration is as follows. Along the [100] direction, each (100) plane couples antiferromagnetically in region B. Within a (100) plane, the magnetic moments are periodically modulated along the [010] direction, i.e. in regions A (zero), B (up spin), A′ (zero), B′ (up spin) and so on. Thus, magnetic peaks appear at 100 and $1 \pm \delta 0$ as satellite reflections from regions B and B′. Along the [010] direction, regions A and A′ can be considered in the same way, and the 010 and $\pm\delta 10$ satellite peaks result. The modulation wavevector is estimated to be $\delta \simeq 0.025$ of the RL vector from the wavelength of the periodic lattice modulation.

This figure assumes an in-phase relation between B and B′. If regions B and B′ have opposite phases '(i.e. up spin at B and down spin at B′ within the (100) plane)', each region in a precipitate works rather as an independent magnetic lamella and a diffuse peak at 100 is expected owing to the small thickness of each lamella. The linewidth is estimated to be $\Gamma = 0.04$–0.05 in RL units for this case. As an actual case, if the phase relation between each region is random, diffuse peaks at 100 and $1 \pm \delta 0$ are expected, resulting in the diffuse peak elongated along the [010] direction around the 100 RLP. At the same time, a diffuse peak elongated along the [100] direction is expected at the 010 RLP. This is consistent with the diffraction patterns observed around 100 and 010.

4. Discussion

The 001 magnetic diffuse peak is commonly observed for the FCC antiferromagnetic systems with the L-AF-1-type structure such as Fe–Mn, Fe–Ni–Cr, Co–Mn and Mn–Cu. The reason why the T-AF-1 component appears is not well understood. Adachi *et al* [4] studied the 100 magnetic diffuse peak of a $Co_{52}Mn_{48}$ alloy and reported an almost circularly shaped intensity distribution, but no origin for the diffuse peak was discussed. Ishikawa *et al* [2] found the anisotropic distribution of the 001 diffuse peak for a γ-Fe–Ni–Cr alloy. They explained it by proposing a periodically modulated short-range spin correlation with the T-AF-1 spin structure. However, the origin of the periodic modulation was not given. Since the periodic lattice distortion has not been reported for the Fe–Ni–Cr system, this case seems to be different from the present γ-Fe precipitates. Recently, Hicks [14] explained the transverse spin component of γ-Mn–Cu alloy as an effect of magnetic defects. Long and Bayri [15] also discussed the 001 diffuse peak of γ-Mn alloys from the same standpoint as Hicks. The γ-Fe precipitates do not seem to be the same as this case. The impurity concentration in γ-Fe precipitates is very small; Cu atoms in γ-Fe are considered to be less than 1 at.%. Furthermore, in the cubic phase of γ-$Fe_{97}Co_3$ alloy precipitates, for which a helical spin ordering based on the AF-1 structure is stabilized and satellite reflections are observed at $1 \pm \delta 0$ ($\delta \simeq 0.12$), but no diffuse peak at the 100 RLP.

An important difference between the 001 diffuse peaks of γ-Fe precipitates and other systems such as Fe–Ni–Cr and Mn–Cu alloys is the temperature dependence of the diffuse peak intensity. In the former, the 100 diffuse peak intensity monotonically decreases with increasing temperature and disappears at around T_N. Thus, the T-AF-1 component of γ-Fe precipitates behaves like an order parameter. On the other hand, the 001 diffuse peak in the latter shows a maximum intensity at T_N and the intensity gradually decreases above T_N. The origin of the latter seems to be static and/or dynamic spin fluctuations due to the distribution of the magnetic defects.

Recently, one of the present authors found that the structural phase transition is suppressed if the precipitates are small ($d < 15$ nm) or if the precipitates are diluted with Co, e.g. γ-$Fe_{97}Co_3$ even with $d \simeq 100$ nm. As mentioned above, in the cubic phase of γ-Fe and γ-Fe–Co precipitates, a helical spin arrangement is stabilized [16]. In this spin structure, the T-AF-1- and L-AF-1-type spin couplings appear alternately in space along the propagation direction of the helix (see figure 11 of [16]), indicating that the T-AF-1 and

L-AF-1 spin structures are energetically degenerate in the cubic phase of γ-Fe. Then, under the periodic lattice distortion, the degeneracy might be removed owing to the reduction in crystal symmetry and it is suspected that the T-AF-1-type spin configuration would be stabilized for the c-axis spin component because of the coupling with the lattice distortion.

It is hard to estimate the integrated intensities of these peaks with sufficient accuracy because all magnetic peaks extend to rather a wide range in q space. In order to estimate the magnitude of the c-axis spin component approximately, we use the maximum peak intensity of each magnetic peak. Then, we can estimate the c-axis spin component to be about 7% of the moment on the c plane.

As pointed out previously, the 120 and 210 magnetic peaks are extremely asymmetric with respect to the RLPs. This is due to the coexistence of the periodic lattice distortion and the modulated spin structure. For the present model of the c-axis spin component, the periodic lattice distortion and modulated spin structure again coexist and an asymmetric intensity distribution might be expected. The main contribution to the asymmetric intensity distribution comes from the relative value of the first- and zeroth-order spherical Bessel functions in the expression for the scattering intensity. At the 100 RLP, the value of $\boldsymbol{K} \cdot \boldsymbol{\Delta}$ is a third of that at 120 and 210 and the relative value of the first- and zeroth-order spherical Bessel functions is negligibly small, resulting in an almost symmetric intensity distribution for the 100 diffuse peak.

In conclusion, for γ-Fe precipitates in Cu, the 100 magnetic diffuse peak was carefully re-examined by means of neutron diffraction. This peak was originally observed by Abrahams *et al* and explained by them as an inclined spin component superimposed on the AF-1 structure. In the present experimental study, the magnetic structure of the T-AF-1 spin component was reconsidered under the coexistence of the periodic lattice distortion and double-$\boldsymbol{Q}$ wave L-AF-1 spin structure on the c plane. We propose a new model for the magnetic structure of the c-axis spin component.

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