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Authors
Miyasato, S.
Thomas, G.

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Presented at the 5th International Aluminum-Lithium Conference, Williamsburg, VA, March 28–31, 1989

Microstructural Evolution in Al-Li and Al-Li-Cu-Mg Alloys

S. Miyasato and G. Thomas

March 1989

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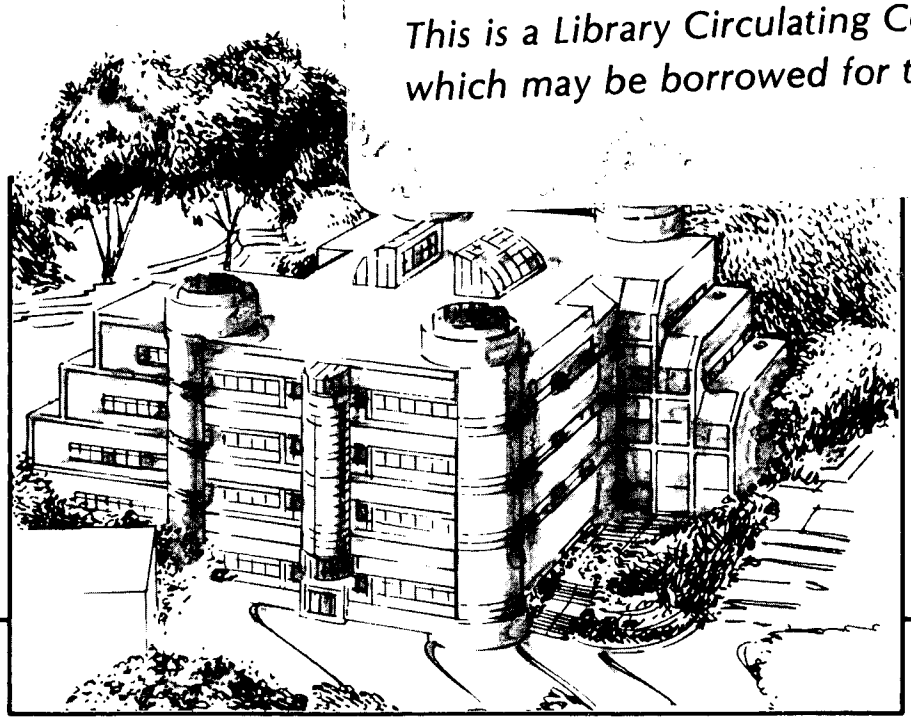

Materials and Chemical Sciences Division

Lawrence Berkeley Laboratory • University of California

ONE CYCLOTRON ROAD, BERKELEY, CA 94720 • (415) 486-4755

Prepared for the U.S. Department of Energy under Contract DE-AC03-76SF00098

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MICROSTRUCTURAL EVOLUTION IN Al-Li AND Al-Li-Cu-Mg ALLOYS

S. Miyasato and G. Thomas*

to be presented at the Fifth International Aluminum-Lithium Alloy Conference, Williamsburg, VA, March 28-31, 1989.

**Center for Advanced Materials and National Center for Electron Microscopy, Materials and Chemical Sciences Division, Lawrence Berkeley Laboratory, University of California, Berkeley, CA 94720*

SYNOPSIS

Since subsequent microstructures and properties depend upon initial decomposition processes, the early developments in a binary Al-Li and a quaternary Al-Li-Cu-Mg alloy were observed by high resolution transmission electron microscopy. Interpretation of high resolution images involved appropriate image simulations. The as-quenched microstructure was found to consist of small ordered domains which were unaffected by the presence of Cu and Mg alloying additions, β' (Al_3Zr) dispersoids and grain boundaries, and are thus interpreted as forming by spinodal decomposition. The ordered domains, ~4 nm in diameter, were often observed in antiphase with each other and were separated by regions of disordered matrix. No discrete δ' (Al_3Li) particles were observed for natural aging times between 24 hours and 3 weeks.

INTRODUCTION

The δ' (Al_3Li) phase in Al-Li based alloys is responsible for the poor ductility and low fracture toughness as well as the high specific strength which makes these alloys attractive for aerospace applications [e.g. 1,2]. The mechanism of δ' formation is still under discussion, and several thermodynamic models have been proposed [3-5]. The most recent model, by Khachaturyan et al., involved superposed static concentration waves to obtain a metastable two-phase ($\alpha + \delta'$) field with δ' composition varying from that of stoichiometry [5]. Under certain conditions, they predicted that a cascade of phase transformations would occur. Upon rapid quenching a

disordered solution (α) would order on the $L1_2$ lattice, decompose into two ordered phases of differing Li content, and finally, the low-%Li phase would disorder. The microstructure would then consist of ordered δ' particles in disordered matrix.

Experimental evidence for an ordering process after quenching was detected by Noble and Thompson [6] and Ceresara et al. [7] using resistivity measurements. Differential scanning calorimetry studies have shown heat evolution stages which precede δ' formation during accelerated aging [8-10]. Conventional imaging by transmission electron microscopy has failed to detect any precursor phases to δ', although the $L1_2$ ordered lattice is evinced by electron diffraction [11-13]. However, high resolution lattice fringe imaging has proved to be an effective method of observing the as-quenched microstructure [14-16]. Regions of relative degrees of order and disorder may be distinguished, although site specific atom positions may not be determined. Sato et al. imaged ordered domains, but included only superlattice reflections in the objective aperture, and so could not distinguish the structure of the material separating the domains [14]. Radmilovic et al. used a larger objective aperture size and complemented the high resolution electron microscopy (HREM) with x-ray diffraction for measurements of degrees of ordering [15,16]. Both HREM studies were performed on aged material. In this work, emphasis was placed upon imaging the structure before significant aging had occurred. Also, image simulations were performed to characterize the effect of foil thickness on the observed image.

PROCEDURE

Two alloys, binary Al-Li(-Zr) and quaternary Al-Li-Cu-Mg(-Zr), were used in this study. The binary alloy was received in plate form from ALCOA, and the rapidly solidified quaternary alloy was received as extruded bars from Allied Signal, Inc. Nominal compositions are given in Table 1.

Table 1 -- Nominal Alloy Compositions

		Al	Li	Cu	Mg	Zr
binary	wt %	97.6	2.4	---	---	0.1
	at%	*91.3*	*8.7*	---	---	*0.03*
quaternary	wt %	95.4	2.6	1.0	0.5	0.5
	at %	*89.5*	*9.5*	*0.4*	*0.5*	*0.1*

From each alloy, small blocks were cut, solution heat treated at 550°C for 2 hours in air, and quenched in agitated iced water. An Isomet diamond saw was used to obtain slices 300 μm (0.013 in) thick well away from Li depleted surfaces. Foils were jet electropolished in 25% nital at -30°C, 15V and stored in a desiccator at room temperature. Aging times before microscopy ranged from 12 hours to 3 weeks. A JEM 200CX with beam divergence semiangle of 0.9 mrad, and objective apertures of 0.65 and 0.74 nm^{-1}, was used to obtain lattice fringe images in symmetric low index orientations. Image simulations were performed using the NCEMSS multislice programs [17]. In the simulations, the composition of the ordered domains was assumed to be $Al_3(0.6Al+0.4Li)$, which corresponds to 10 at.% Li. No variation in composition across a domain was used.

RESULTS

Microscopic examination of foils aged overnight at room temperature revealed ordered structures similar to those observed by Sato et al. [14] and Radmilovic et al. [16]. Figure 1 is a representative image from the binary alloy taken very near the edge of the foil. Lattice fringe spacings of 0.4 nm and 0.2 nm may be observed in different areas in the highly magnified image in Figure 1(b). Image simulations (e.g. Figure 2) show that at foil thicknesses and defocus values for which 0.4 nm periodicities are observed in the ordered domains, the 0.2 nm periodicities correspond to the disordered aluminum lattice. Thus, areas with 0.4 nm spacing are interpreted as having ordered $L1_2$ structure, while areas with 0.2 nm spacing correspond to disordered, low Li content regions.

Ordered regions labelled A and B in Figure 1(b) are observed to be in antiphase with one another. Regions B and C appear to be of the same variant, however, not all antiphase relations are visible in this <100> zone axis.

After aging at room temperature for 3 weeks the modulated order/disorder structure was still observed, as shown in Figure 3. This agrees with the results of Sato et al., who preserved large areas of modulation after aging a binary alloy for 24 hours at 50°C [14]. The change in composition required for nucleation and growth is expected to be assisted by "diffusion pipes" at heterogeneities such as grain boundaries and matrix/dispersoid interfaces. However, imaging at grain boundaries and interfaces (Figure 4) revealed no discrete δ' formation.

DISCUSSION

Spinodal Decomposition

The presence of microstructural heterogeneities such as grain boundaries, dislocations, and matrix/dispersoid interfaces did not affect the modulated order/disorder structure (Figure 4). If the ordered domains had formed by a nucleation and growth process, their size or number would increase near heterogeneities, which assist the diffusion necessary for stable embryo. Spinodal decomposition, on the other hand, requires only short range diffusion and is affected little by these diffusion pipes. Hence, the lack of a difference in observed degree of order in grain interiors and at grain boundaries, etc. is taken as evidence of the spinodal mechanism.

Antiphase Boundaries

The observed antiphase domains in Figure 1(b) suggest that congruent ordering during a rapid quench occurs as predicted. There are four variants to the $L1_2$ lattice and atomic arrangement may occur on all four. The antiphase boundaries which form will influence the spinodal wave, as they have higher energy and thus lower the limit of stability of the high Al content ordered phase. The disordered Al regions separating ordered antiphase regions (Figure 1(b)) may thus be considered a wide APB. All the disordered regions in the modulated structure separate antiphase domains. This may be checked using a microscope with large tilting capability to observe two different <100> zone axes. The high concentration of antiphase boundaries in the as-quenched structure should increase the strength and decrease the ductility of the material.

Degree of Order

The degree of long range order in as-quenched Al-Li-based alloys should depend on the atom fraction of Li and the atom fraction of Cu, Mg, etc. solute atoms. As the fractional amount of Li increases, the overall alloy composition is shifted closer to the δ' side of the two-phase field of the phase diagram, and consequently the volume of δ' must increase. Since δ' is preceded by the Li-rich ordered domains, it also follows that the degree of order in quenched alloys must also increase. The growth of disordered Li-lean regions occurs at antiphase domain boundaries by diffusion of Li into the ordered domain, and of Cu and Mg solute and excess Al atoms away from the ordered domain. The diffusion rate limiting process is expected to be slow diffusion of Cu atoms. Hence, the larger the atom fraction of Cu in the alloy, the higher the degree of order. Another possible effect of Cu and Mg additions is the reduction of solubility of Li in Al, which may also serve to increase the rate of the spinodal decomposition. No significant difference in degree

of order is apparent by HREM in the Al-Li and Al-Li-Cu-Mg alloys, however. Insufficient semi-quantitative metallography or aging times may be culprits. Further HREM and x-ray diffraction work to measure the long range order parameter immediately after quenching, and after short artificial aging times is in progress.

Foil Thickness Considerations

The domain size varies from 2 to 6 nm in diameter and the foil thickness is estimated to be between 10 and 20 nm, so several domains may be stacked upon each other in the direction normal to the beam. This effect has not been considered thus far. Simulations of ordered regions by Radmilovic et al. assume only one variant of ordered phase with no disordered phase present. Extensive image simulations have been performed to study the effects of multiple variants and aluminum interlayers on the image. Results show that stacking 2 or 3 different variants does affect the image, but under optimum defocus conditions, the 0.4 nm spaced superlattice fringes may still be identified. It has also been shown that interlayers of aluminum 3 nm thick still result in superlattice fringes, but of reduced intensity. Thus, the regions which appear ordered, may actually contain disordered material in the thickness direction. HREM can distinguish only regions of relative order and disorder in the thickness direction, but cannot quantify this in any way. A few highly ordered domains separated by thick disordered layers will project the same image as less ordered domains separated by thin disordered layers.

CONCLUSIONS

A high resolution electron microscopy study of the development of the initial microstructure of quenched aluminum-lithium based alloys resulted in the following observations:

The modulated order/disorder domain structure was observed in both binary and quaternary alloys after room temperature aging for less than 24 hours; i.e. ordering is not suppressed by quenching.

Heterogeneities such as grain boundaries and interfaces did not influence the modulated structure, which therefore must form by spinodal mechanism and not by nucleation and growth.

Computer image simulations showed the effect of multiple variants and aluminum interlayers through the foil thickness. Stacking two different $L1_2$ variants still resulted in visible ordered structure. The 3 nm aluminum interlayers did not affect the image significantly.

ACKNOWLEDGMENTS

The authors thank Prof. J. W. Morris, Jr., J. Glazer, T. Lindsay, and A. G. Fox for their interest and helpful comments. This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences Division, of the U. S. Department of Energy under Contract No. DE-AC03-76SF00098.

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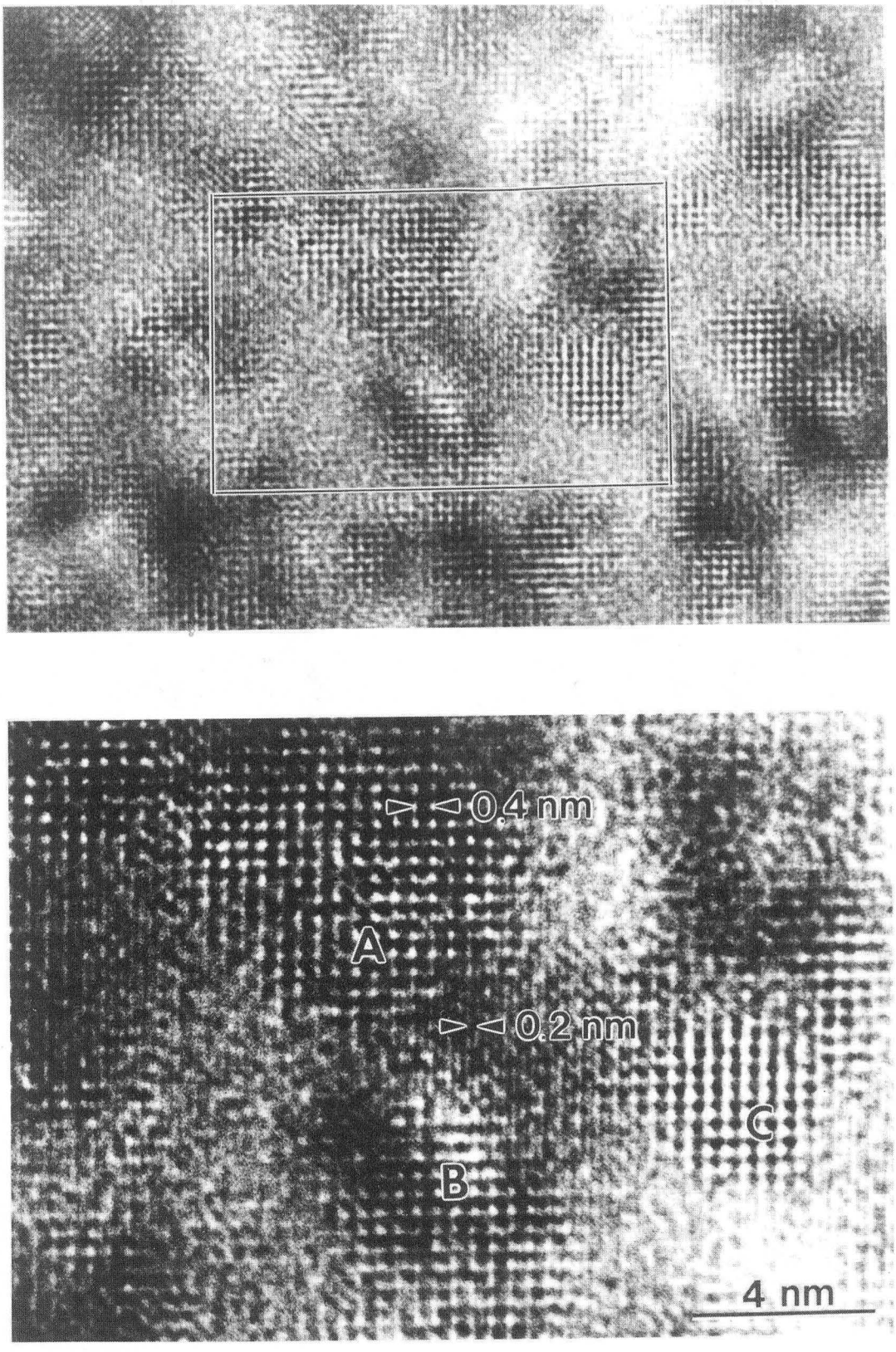
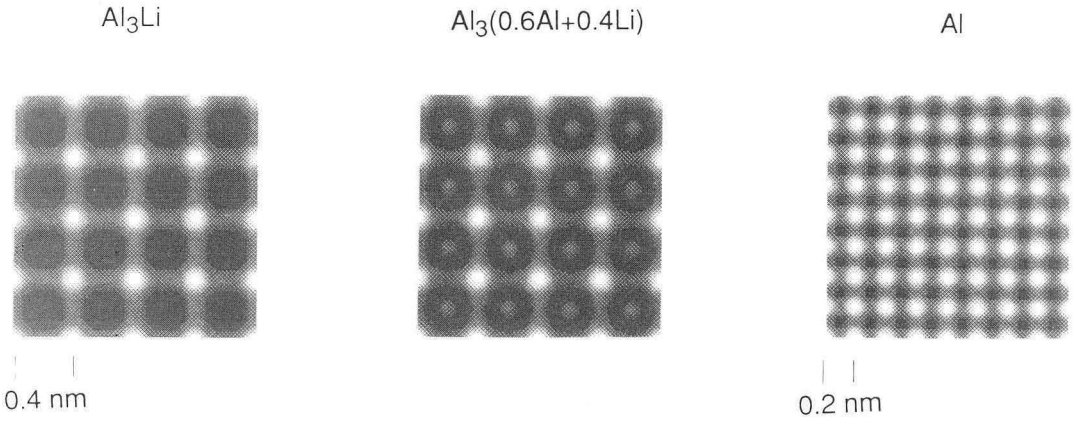


Figure 1. HREM image of Al-Li alloy after quenching and room temperature aging for 24 hours. The modulated order/disorder structure is shown in (a). Antiphase domains, A and B, are shown in (b), a high magnification image of (a). The disordered aluminum matrix may be considered an antiphase domain boundary.

XBB 893-1742

Identification of Modulated Order/Disorder Domains



By matching experimentally obtained images with calculated images, the ordered and disordered regions may be identified.

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Figure 2. Computer image simulation of stoichiometric δ' (Al_3Li), ordered domain with 10% Li, and aluminum. Foil thickness is 10 nm and objective lens defocus is -60 nm. Ordered structure is identified by 0.4 nm fringe spacing and the aluminum FCC structure is identified by 0.2 nm fringe spacing, for this foil thickness and defocus setting.

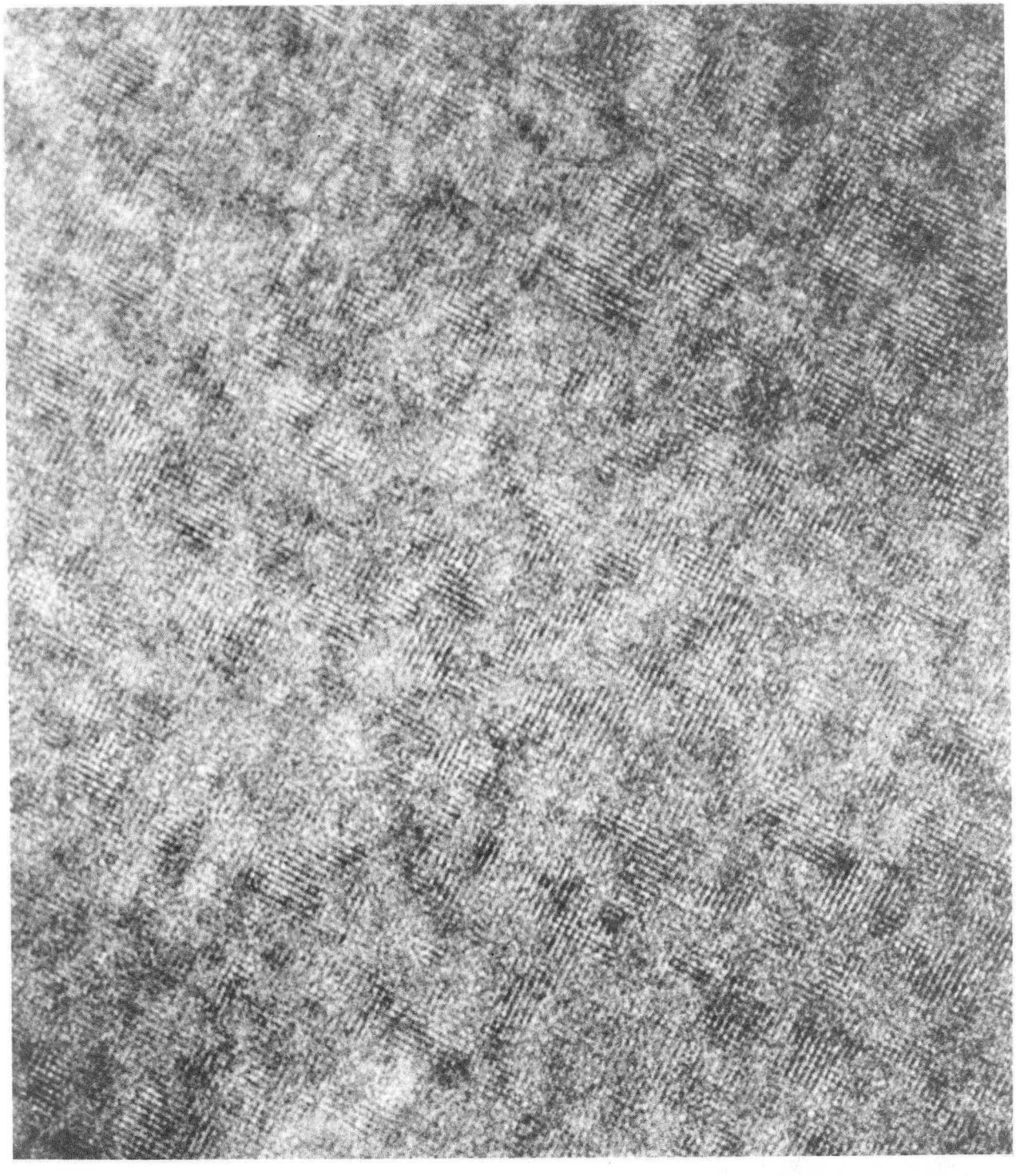

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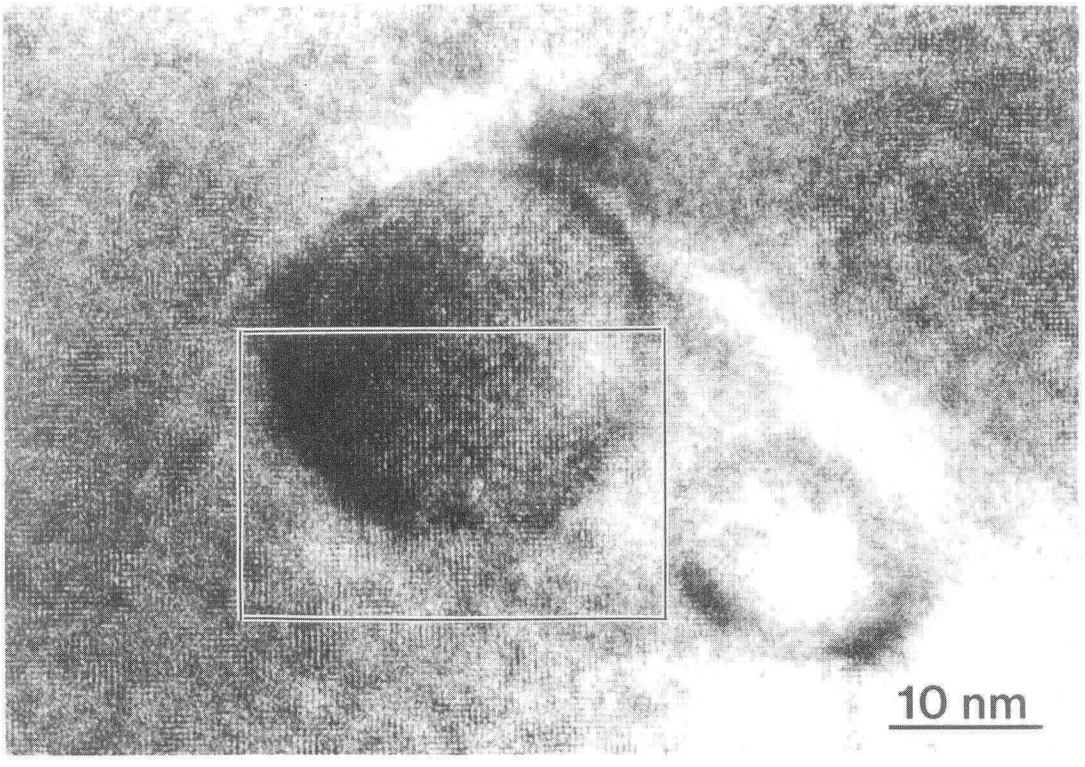
Figure 3. HREM image of Al-Li-Cu-Mg alloy after quenching and room temperature aging for 3 weeks. Modulated order/disorder structure is observed over large areas.



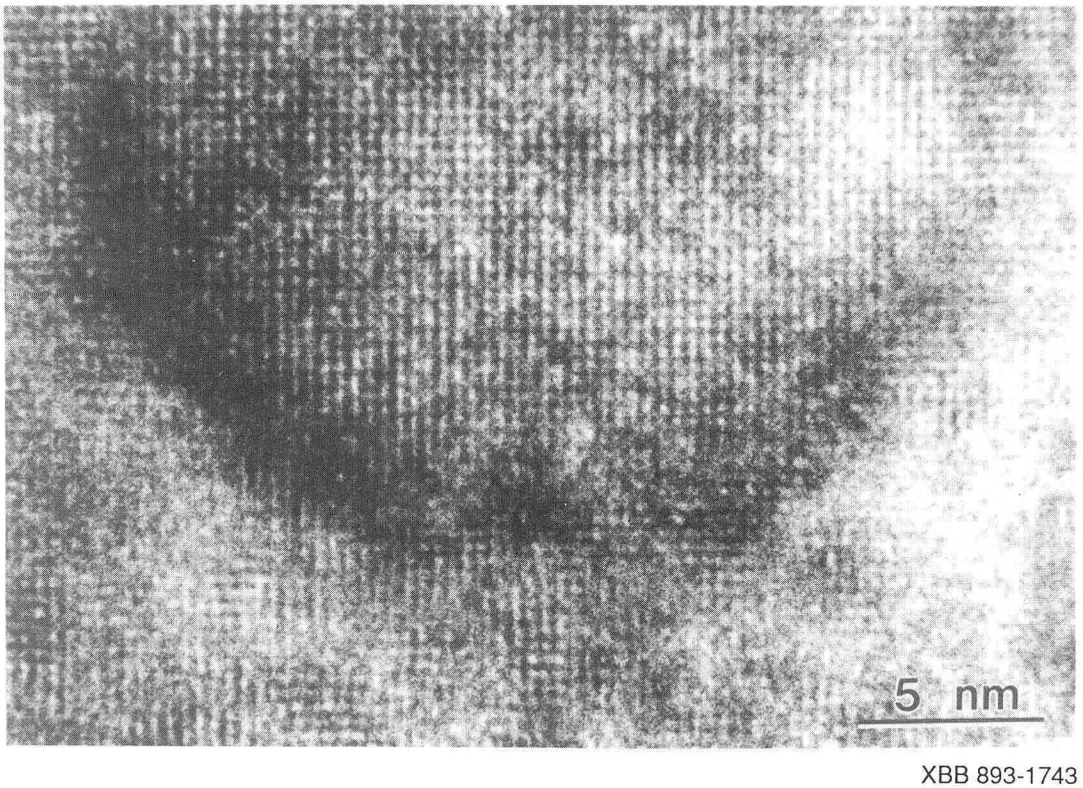


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Figure 4. HREM image of Al-Li-Cu-Mg alloy after quenching and room temperature aging for 3 weeks. Modulated order/disorder structure continues up to dispersoid interface.

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1 CYCLOTRON ROAD
BERKELEY, CALIFORNIA 94720