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Limits of kinematic modelling of superlattice period due to Laue function convergence [1]

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Abstract

Thin alternating Au, Ag and Cu wedge-shaped films were sputtered onto Formvar-coated grids. X-ray diffraction patterns of the samples revealed satellites in the vicinity of (111) reflections, which are well fitted in the kinematic approximation for imperfect superlattices, and are sensitive to the fluctuations of the superlattice period, as long as the superlattice Laue function does not converge.

Keywords: Superlattices; X-ray diffraction; Kinematic approximation

Thin films of alternating Au, Ag and Cu wedge-shaped metal layers were magnetron-sputtered onto Formvar-coated Mo grids in order to develop a method for investigation of multicomponent systems. The total thickness of each sample was about 360 nm with superlattice period of 12 nm. X-ray diffraction patterns of all as-deposited samples revealed satellites in the vicinity of (111) reflections, resulting from the formation of composition-modulated artificial superlattices. The experimental spectra are well explained by the kinematical diffraction theory for imperfect superlattices. Computer simulations have demonstrated an extremely high sensitivity (0.5%) to the fluctuations $\Delta H/H$ of the superlattice period H. As a result, a rapid and simple method of $\Delta H/H$ determination was proposed [1]. This method, however, is limited by the fast convergence of the superlattice Laue function, leading to an inability to distinguish between cases where fluctuations of a superlattice period exceed a certain limit. In the current work, the limiting value of 2.5% has been established.

For an ideal superlattice, the diffraction intensity can be calculated by means of direct summation of waves (with corresponding phases) scattered by the array of atomic planes. In the case of binary superlattices, such calculations have been performed by several authors [2,3]. In our previous work, we calculated the diffracted intensities for the case of *an imperfect ternary* superlattice [1]. The imperfections can be divided into four major categories: interdiffusion, layer thickness fluctuations, crystallinity and crystalline orientation. Realistically speaking, the distinction between the particular type of imperfection and its effect on diffraction spectra is not so sharp [4]. The square of the structure factor (proportional to the diffracted intensity) consists of the lattice function summing the waves scattered by atomic planes of elemental components comprising the superlattice,

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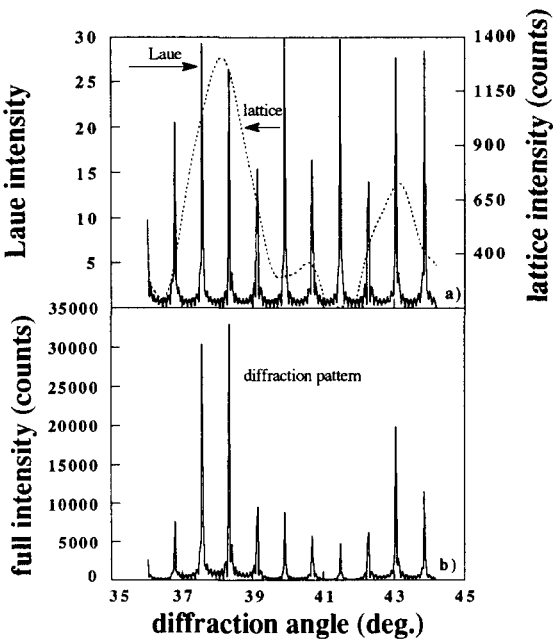

Fig. 1. (a) The lattice and the Laue functions for the *perfect* superlattice composed of 22 at% Au, 24 at% Ag and 54 at% Cu. (b) The resulting diffraction pattern.

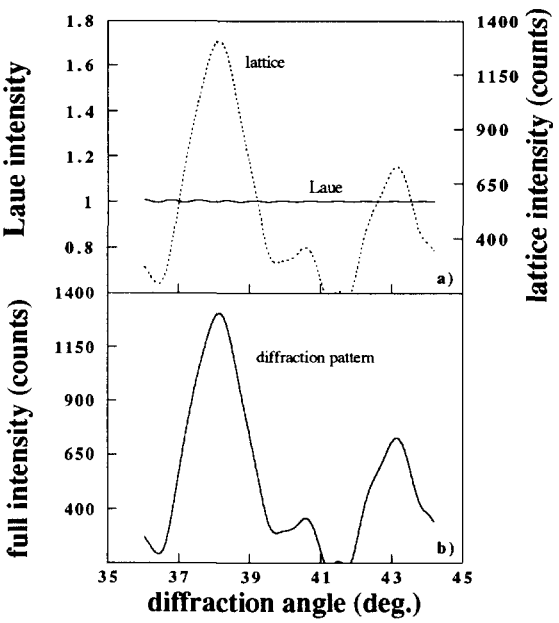

Fig. 2. (a) The lattice and the Laue functions for the superlattice composed of 22 at% Au, 24 at% Ag and 54 at% Cu, with period fluctuations of $\Delta H/H = 2.5\%$. (b) The resulting diffraction pattern.

multiplied by the superlattice Laue function which takes into account the newly formed superperiodicity which, in addition, may fluctuate around some average value H [5]. Such fluctuations cause non-cumulative disorder, which is expressed by the Gaussian distribution, as shown in the expression below:

$$L(Q,\sigma) = \frac{1 + \exp(-N\sigma^2Q^2/2) - 2\exp(-N\sigma^2Q^2/4)\cos(NHQ)}{1 + \exp(-\sigma^2Q^2/2) - 2\exp(-\sigma^2Q^2/4)\cos(HQ)},$$

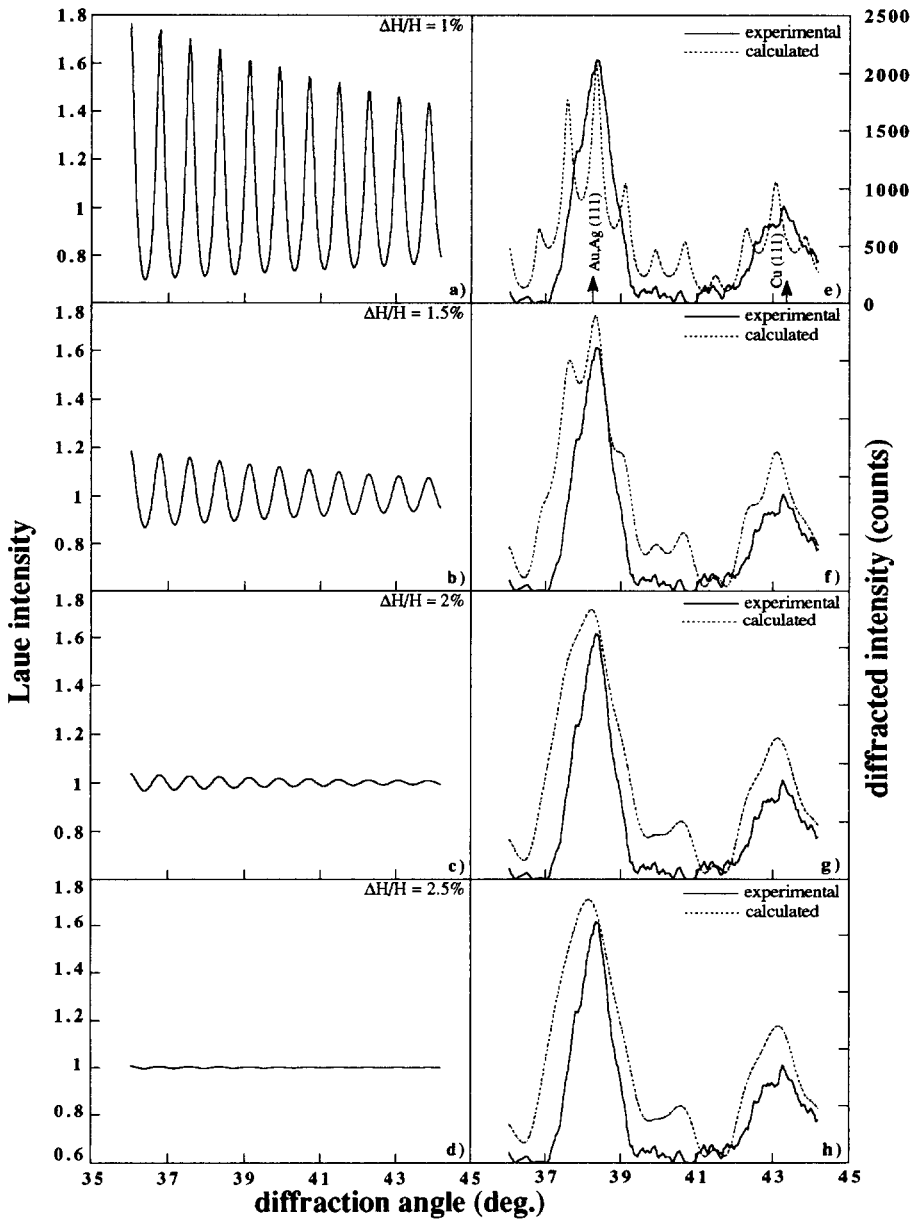

Fig. 3. (a–d) Dependence of the Laue function for the superlattice composed of 22 at% Au, 24 at% Ag and 54 at% Cu on superperiod fluctuations. (e–h) Variation of the diffraction profile due to variation of the Laue function.

where H is the superperiod thickness, N is the number of superperiods (trilayers) in a total deposited thickness, Q is the diffraction vector and σ reflects the fluctuations, $\Delta H/H$, of the superperiod.

Fluctuations can be also introduced through the layer structure factor, in two possible ways: fluctuations of number of elemental layers, or interface thickness fluctuations. However, these fluctuations represent an effective Debye–Waller factor, which mainly affects the peak intensity but not its width. On the other hand, comparing Figs. 1 and 3, one can clearly observe significant differences in peak widths, between the sharp ideal pattern (Fig. 1) and the broad experimental one (Fig. 3). Irregularities in the superperiod thickness destroy the

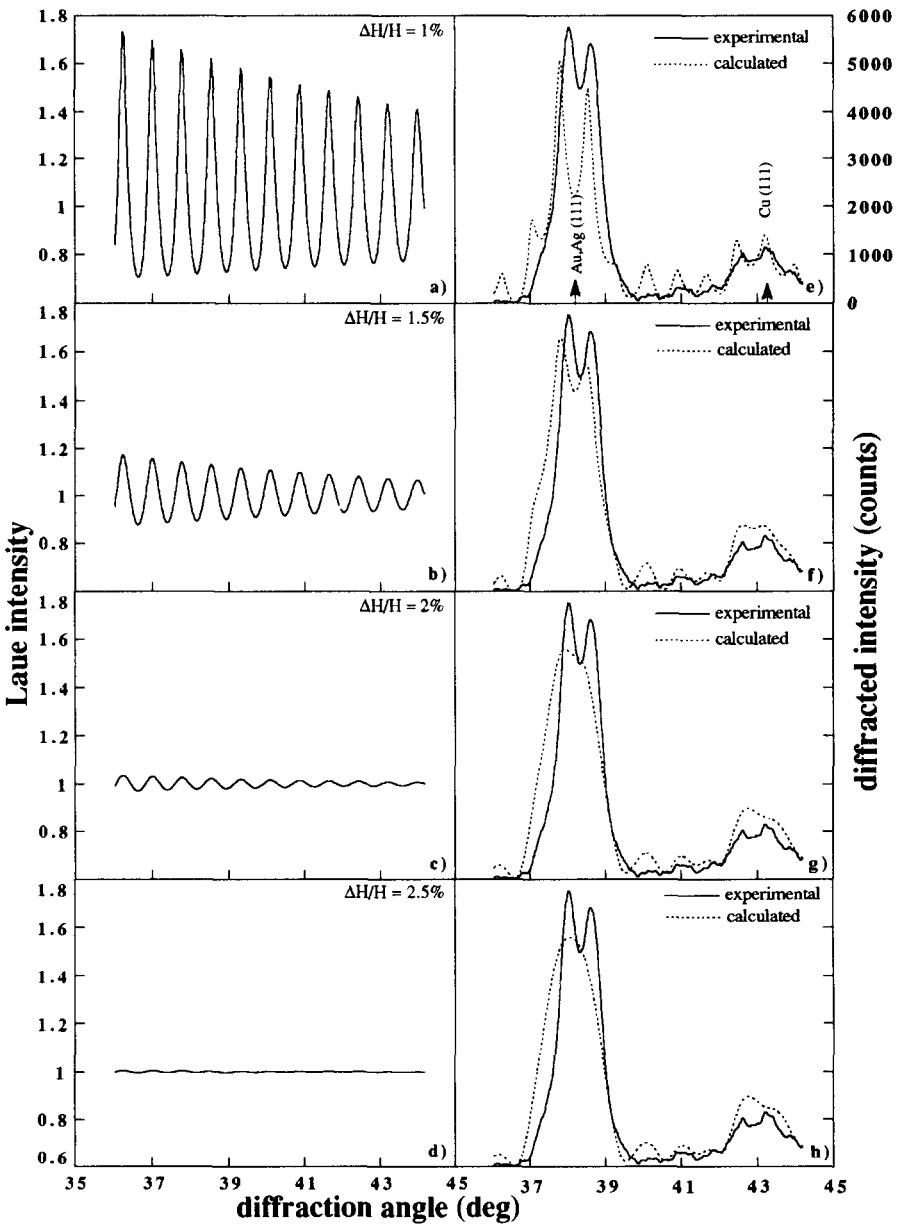

Fig. 4. (a–d) Dependence of the Laue function for the superlattice composed of 35 at% Au, 21 at% Ag and 44 at% Cu on superperiod fluctuations. (e–h) Variation of the diffraction profile due to variation of the Laue function.

phase relations between the otherwise coherently diffracted X-ray beams, manifesting themselves via peak broadening [4,5]. Thus the approach taken seems reasonable and is supported by the good quality fits. The more precise fitting procedure takes into account the average layer thickness, lattice spacings of elemental components including strain profiles near each interface and additional disorder mechanisms, such as discrete atomic layer thickness variations and fluctuations of interface lattice spacings [6–8]. However, such a procedure is extremely complex when applied to ternary superlattices.

The full expression for the ternary case is not shown here for the sake of brevity, but is given in our previous work [1]. In its derivation a complete alternation of chemical compositions at interfaces between adjacent layers ('step model') was assumed. The diffraction patterns shown in Figs. 3 and 4 represent some of the best fits from our previous work [1]. However, the current work demonstrates the limiting cases, which were not analysed then. The advantage of these relatively high-Q portions of diffraction patterns is that they bear direct information on the intralayer structure (crystallinity and orientation), unlike small-Q regions. The diffraction pattern for the *perfect* superlattice composed of 22 at% Au, 24 at% Ag and 54 at% Cu [1] is shown in Fig. 1b. It is a product of the lattice and the ideal Laue functions ($\sigma = 0$) shown in Fig. 1a. It is obvious from the expression for the Laue function that the larger the fluctuations, the faster the Laue function converges to unity. In fact, starting from $\Delta H/H = 2.5\%$, the effective value of the function is unity, and the shape of the diffraction pattern is identical to the shape of the lattice function, as demonstrated in Fig. 2. Intermediate cases are shown in Fig. 3, undoubtedly demonstrating the best fit for the case where $\Delta H/H = 1.5\%$. Thus when $\Delta H/H \geq 2.5\%$, diffraction patterns no longer differ in appearance (being identical to the lattice function which does not depend on $\Delta H/H$ at all) and it is impossible to deduce the superperiod fluctuations from them using such a simple modelling. Similar results have been obtained by us for Au–Ag–Cu superlattices of different compositions [1]. An example of the 35 at% Au, 21 at% Ag and 44 at% Cu composed sample is shown in Fig. 4. It is apparent that apart from a phase shift the intensity of the Laue function is the same as in the previous case, yielding similar conclusions.

The fact that the (111)-lattice spacings of Au and Ag are contracted over their bulk values, as arrowed in Figs. 3 and 4, while essentially equal in the case of Cu, not necessarily indicates the presence of strains, since the diffraction pattern from the intralayer bulk structure is affected by the relative magnitude of lattice spacings from the components. In order to verify the coherency strains the additional, in-plane scan, is required. However, the asymmetric appearance of the same order-satellites about the main peak, as about the (111) Au, Ag reflection in Fig. 3, is an additional confirmation of strains present in the Au and Ag layers [5]. The Au/Ag lattice mismatch is quite small ($\approx 0.2\%$), unlike the Au/Cu and Ag/Cu mismatches ($\approx 11\%$). These strains probably cause defect formation on the Au/Cu and Ag/Cu interfaces, resulting in the observed superperiod fluctuations.

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