Breakdown of Continuum Elasticity Theory in the Limit of Monatomic Films

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We demonstrate that the predictions of continuum elasticity theory fail in the ultimate limit of monolayer films. We directly measure the lattice distortion of ultrathin InAs layers in GaAs by high-resolution electron microscopy. For InAs films of 3 monolayer thickness, the observed tetragonal distortion agrees with the prediction of elasticity theory. For single InAs monolayers, however, the measured strain is much higher than expected. The InAs unit cell in this case is strained such as to conserve the bulk bond length at the interface

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The progress in crystal growth techniques has made it possible to synthesize artificial layered materials built up of structurally and chemically dissimilar constituents. The accurate control of the growth processes allows the thickness of the individual layers to be scaled down to the atomic regime. The successful synthesis of such structures led to the exciting opportunity to address a fundamental question of solid-state physics; namely, at which scale the bulk properties of the constituent materials are established and serve as an adequate description of the heterostructure, and, on the other hand, at which scale the properties of the structure are dominated by the local atomic configuration of the interface [1–3]. Much effort is hence currently devoted to the understanding of the formation and atomic configuration of semiconductor heterointerfaces [4–6].

In this Letter, we demonstrate a novel method to determine the atomic configuration at the interface between crystalline materials. By analyzing high-resolution lattice images of coherently strained InAs films in GaAs, we directly measure the tetragonal distortion of the InAs unit cell. The distortion of 3 monolayers (ML) InAs agrees with continuum elasticity theory. In contrast, the elasticity theory fails for 1 ML InAs, where the lattice distortion is much larger than expected. In this case, the distortion of the unit cell is consistent with the conservation of the bond length at the interface. Our study thus demonstrates that the continuum case establishes rapidly, but significant deviations occur in the ultimate limit of a monatomic layer.

The investigated structures consist of single InAs films buried in GaAs and are synthesized by solid-source molecular-beam epitaxy on semi-insulating (100) GaAs substrates. The nominal thicknesses of the InAs films are either 1 or 3 ML. A novel growth procedure has been developed to ensure the controlled buildup of both InAs/GaAs and GaAs/InAs interfaces [7]. This technique allows us to synthesize atomically smooth InAs films of well-defined thickness even in the monolayer regime. For the high-resolution electron microscopy (HREM) experiments, cross-sectional samples along the ⟨110⟩ direction are prepared by conventional ion milling. The specimen thickness is then between 10 and 20 nm, as determined by image simulations. Lattice images are taken in a JEOL 4000FX electron microscope operating at 400 kV.

Qualitative information about the interface morphology and the strain of the InAs film is obtained by double-crystal x-ray diffraction. The experimental diffraction patterns are analyzed by the dynamical x-ray diffraction theory [7]. These experiments demonstrate the exceptional structural perfection of the InAs/GaAs heterointerfaces created by the inserted InAs film. Measurements around asymmetric reflections reveal the commensurate state of the InAs/GaAs interfaces for an InAs layer thickness below 3 ML, i.e., the InAs unit cell is biaxially strained to fit the GaAs lattice parallel to the interface. This in-plane strain in turn results in an elastic tetragonal distortion $\epsilon_\perp$ of the InAs unit cell perpendicular to the interface, the magnitude of which cannot, however, be determined independently from these experiments. The tetragonal distortion of the unit cell is thus usually assumed to be given by continuum elasticity theory: $\epsilon_\perp = -(2C_{12}/C_{11})\epsilon_\parallel$, where $\epsilon_\perp$ and $\epsilon_\parallel$ denote the strain components with respect to the unstrained cubic crystal perpendicular and parallel to the interface, respectively, and C_{ij} are components of the elastic stiffness tensor.

In principle, HREM is the ideal tool for measuring both thickness and strain state of ultrathin films separately. However, several imaging artifacts in general complicate the interpretation of contrast variations in high-resolution lattice images. Long-range contrast changes at interfaces are thus by no means a reliable probe of the atomic configuration of the interface. The approach we describe in the following is based on the direct measurement of lattice distortions caused by the lattice mismatch between the constituent materials. This technique is inherently free from imaging artifacts caused by thickness and defocus fluctuations as well as by the spherical aberration of the objective lens.

We first outline our technique by applying it to an image simulation. Figure 1(a) shows the image simulation

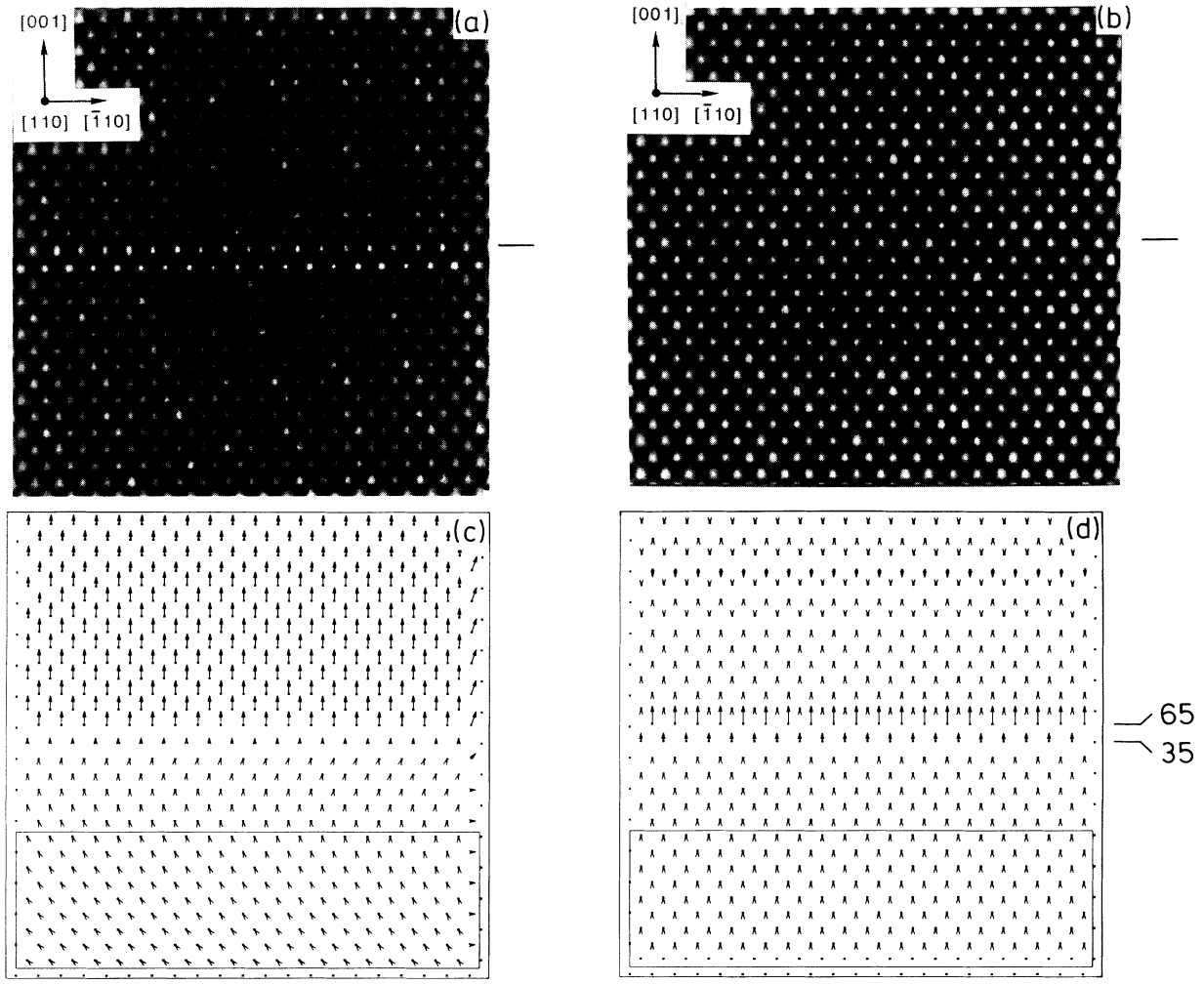

FIG. 1. (a) Image simulation of a single InAs monolayer (ML) in GaAs with a tetragonal distortion of 7.26%. (b) Lattice image after Fourier filtering. The bars indicate the plane containing In. (c) Vector representation of the shift in atom positions of the lattice image shown in (b) with respect to the GaAs lattice (the rectangle surrounds the reference area used for generation of the GaAs lattice). The magnitude of the vectors is amplified by a factor of 3. (d) Vector representation of the relative shift in position of each plane with respect to its immediate neighbor. Numbers denote the relative magnitude of the displacement vectors in percent.

for 1 ML InAs, whose lattice constant parallel to the plane equals that of GaAs. Perpendicular to the plane, the InAs unit cell is assumed to be tetragonally distorted by 7.26%, the value predicted by elasticity theory. This lattice image is Fourier transformed and frequency filtered by an aperture in reciprocal space, transmitting spatial frequencies between 2 and 4.5 nm^{-1} ($1/e$ values). This frequency range includes the {111} and {200} beams and is therefore sufficient to resolve the distance between the (100) planes in GaAs [8]. After Fourier filtering, the lattice image is transferred back to real space. It is finally processed by an algorithm which maximizes the local contrast variations. In Fig. 1(b), the lattice image is depicted after Fourier filtering and contrast enhancement. The image is now free from any background contrast which may lead to misleading information of the atom configuration at the interface.

In the next step, an ideal GaAs lattice is fitted to an unperturbed portion of the experimental lattice image by a recursive formalism, stopping when an accuracy of 10^{-4} in both magnitude and direction of the basis vectors is reached. This calculated lattice is now extrapolated over the entire investigated lattice. The difference in position of the dumbbells in the calculated and the investigated lattice is determined and represented by a two-dimensional vector [9]. In Fig. 1(c), we show the resulting two-dimensional vector field obtained by analyzing Fig. 1(b). This picture represents directly the absolute shift of the (100) lattice planes in the simulated image with respect to the one of the unperturbed GaAs lattice, caused by the tetragonal distortion of the InAs unit cell. The shift is determined to be 0.42 ± 0.01 Å [10], corre-

sponding to a strain of $\epsilon_\perp = (7.29 \pm 0.3)\%$, which is in excellent agreement with the input value of 7.26%.

For a direct identification of the InAs ML, we calculate the relative shift of each lattice plane in the [001] direction with respect to its immediate neighbor. The resulting vector field is shown in Fig. 1(d). It is important to note that *one* strained ML leads to the observation of *two* displacement vectors in the lattice image, representing the two strained In-As bonds connecting the (100) In lattice plane with each of the sandwiching (100) As lattice planes. Under our experimental conditions, the whole tetragonal distortion $\epsilon_\perp$ is then shared among two lattice planes with a ratio of about 35:65 [11].

Next, we apply the above presented technique to experimental lattice images obtained from fabricated InAs/GaAs structures. The results shown in the following are representative for samples containing InAs films of 3 and 1 ML thickness. In Fig. 2(a), we show the Fourier-filtered lattice image of the sample with an InAs layer thickness of 3 ML. The relative shift of the (100) lattice planes is shown in Fig. 2(b). The overall shift of the lattice in [001] direction is determined to be 1.24 ± 0.01 Å. This distortion is shared among four lattice planes [Fig. 2(b)], corresponding to an InAs film of 3 ML. The overall strain is distributed in the lattice image according to 20:40:30:10. Assuming a homogeneous distribution of the strain over each of the 3 ML [12], the strain per ML

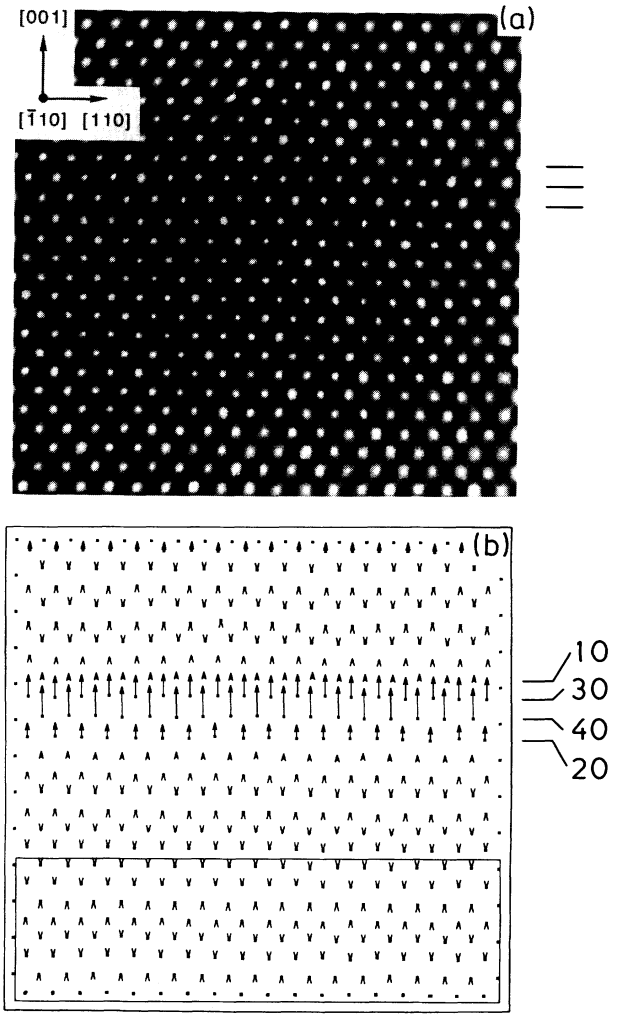

FIG. 2. (a) Fourier-filtered experimental lattice image of 3 ML InAs in GaAs. Bars indicate planes containing In. (b) Vector representation of the relative shift in position of each ML with respect to its immediate neighbor. The magnitude of the vectors is amplified by a factor of 5. Numbers give the relative magnitude of the displacement vectors in percent.

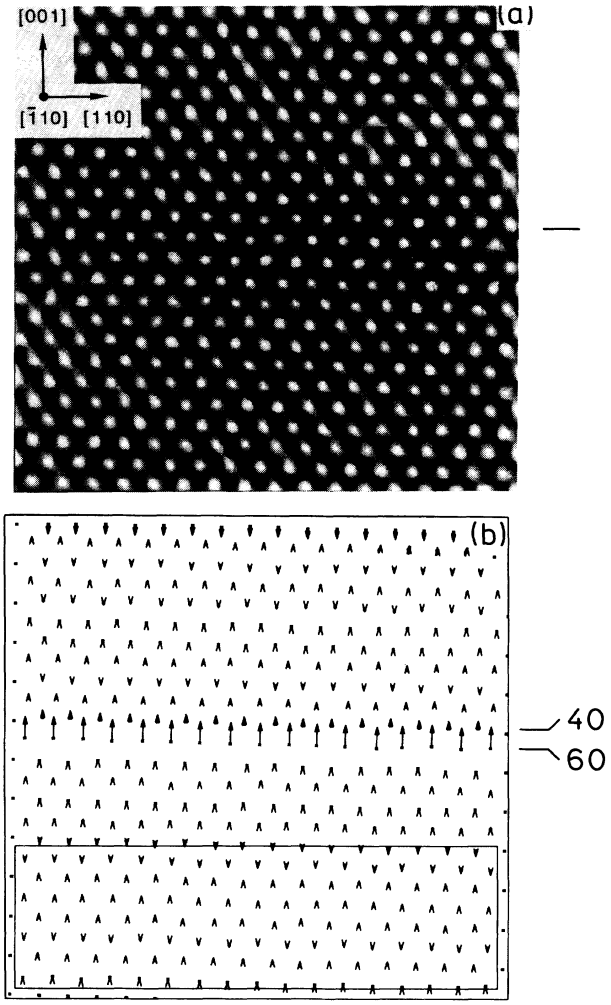

FIG. 3. (a) Fourier-filtered experimental lattice image of 1 ML InAs in GaAs. The bar indicates the plane containing In. (b) Vector representation of the relative shift in position of each ML with respect to its immediate neighbor. The magnitude of the vectors is amplified by a factor of 5. Numbers give the relative magnitude of the displacement vectors in percent. The cross section is oriented along [1$\bar{1}$0] instead of along [110], resulting in the inversion of the length ratio of the two displacement vectors at the interface with respect to the image simulation.

is determined to be $(7.06 \pm 0.1)\%$, close to the value expected from elasticity theory.

In Fig. 3(a), we show the Fourier-filtered lattice image of the sample with an InAs layer thickness of 1 ML. The relative shift of the lattice planes is shown in Fig. 3(b). The overall shift of the lattice in [001] direction is determined to be 0.58 ± 0.01 Å. This distortion originates from 1 ML, as demonstrated by the fact that two displacement vectors with a length ratio of 60:40 are detected [Fig. 3(b)]. It corresponds to a strain of $(12.6 \pm 0.3)\%$, significantly larger than that expected from elasticity theory.

The above shown results demonstrate that a 3-ML-thick film can already be considered as an elastic continuum, i.e., the bulk case establishes quite rapidly. The same has been found for ultrathin Ge films on Si [13,14]. However, a significant deviation from the elasticity theory occurs in the ultimate limit of a monoatomic layer. In this case, the measured tetragonal distortion of the unit cell corresponds to an In-As bond length of 2.62 Å, exactly equal to that of unstrained bulk InAs. This experimental result confirms recent *ab initio* total-energy calculations [15]. By explicitly considering the present case of a single In lattice plane buried in a GaAs matrix, these calculations predict an In-As bond length that closely resembles the one of the unstrained bulk crystal. The In-As bonds directly at the InAs/GaAs interface are thus stretched in order to conserve their bulk bond length [16]. The tendency towards conservation of the unstrained bulk bond length has in fact been suggested for rather different chemical environments [17–21]. Thus, while films of a few ML thickness are well described by macroscopic arguments, the atomic configuration at the interface can be understood only by considering the local properties of the crystal lattice, namely, the chemical bond.

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[8] Only spatial frequencies required for the determination of the atom positions are thus transmitted, whereas those responsible for long-range contrast fluctuations and those creating the internal structure of the individual GaAs dumbbells in [110] projection are cut.
[9] In general, the resulting vector field represents a Moiré structure and can easily be analyzed by the well established mathematical formalism developed for such patterns.
[10] It should be pointed out that the sensitivity of our technique is mainly limited by the size of the matrix representing the digitized lattice image. At present, we are restricted to 512×512 pixels, yielding an uncertainty in the *individual* dumbbell position of 0.15 Å. For the determination of the total shift we take the *statistical* average over all dumbbells within an area containing n dumbbells (equivalent to that used for the generation of the GaAs reference lattice), which reduces the uncertainty by $\sqrt{n}$.
[11] Note, that the In-As dumbbell is not resolved; i.e., the maximum intensity in the lattice image coincides with the center-of-gravity of the dumbbell, which is apparently different for the two bonds.
[12] The observed length ratio is close to that expected for a homogeneously strained film (22:33:33:12).
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[16] It is illustrative to compare this result with Keating's phenomenological valence-force-field approach. In the frame of this model, the elastic response of the crystal is described by two microscopic parameters, namely, the bond-stretching (α) and bond-bending (β) force constants. The ratio β/α is a measure for the relative change of bond angle and bond length induced by external stress. InAs has in fact one of the smallest β/α values of all III-V semiconductors, which means that strain is accommodated preferentially by bond bending instead of bond stretching. See R. M. Martin, Phys. Rev. B **1**, 4005 (1970) and A. Continenza, S. Massidda, and A. J. Freeman, Phys. Rev. B **41**, 12013 (1990).
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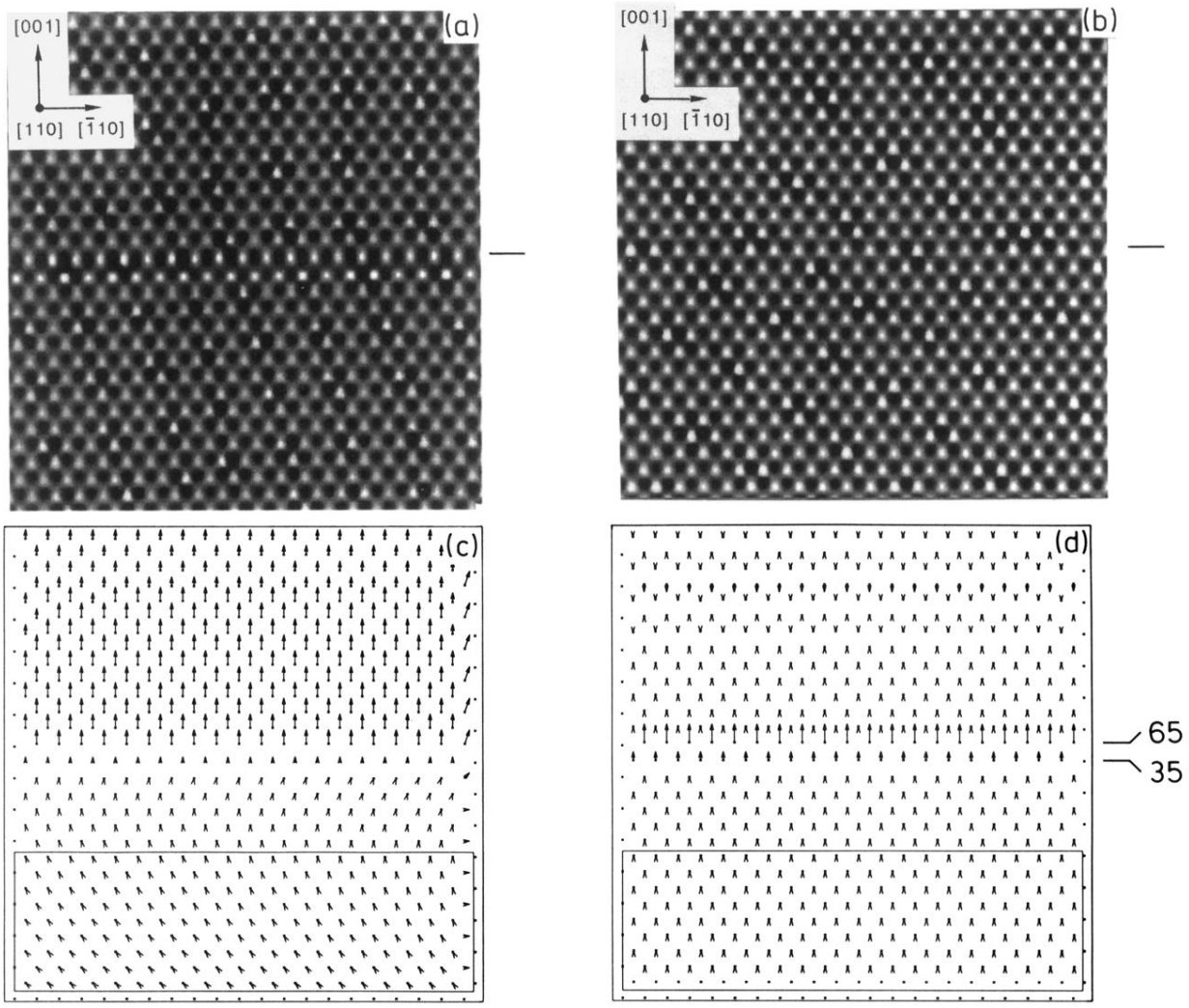

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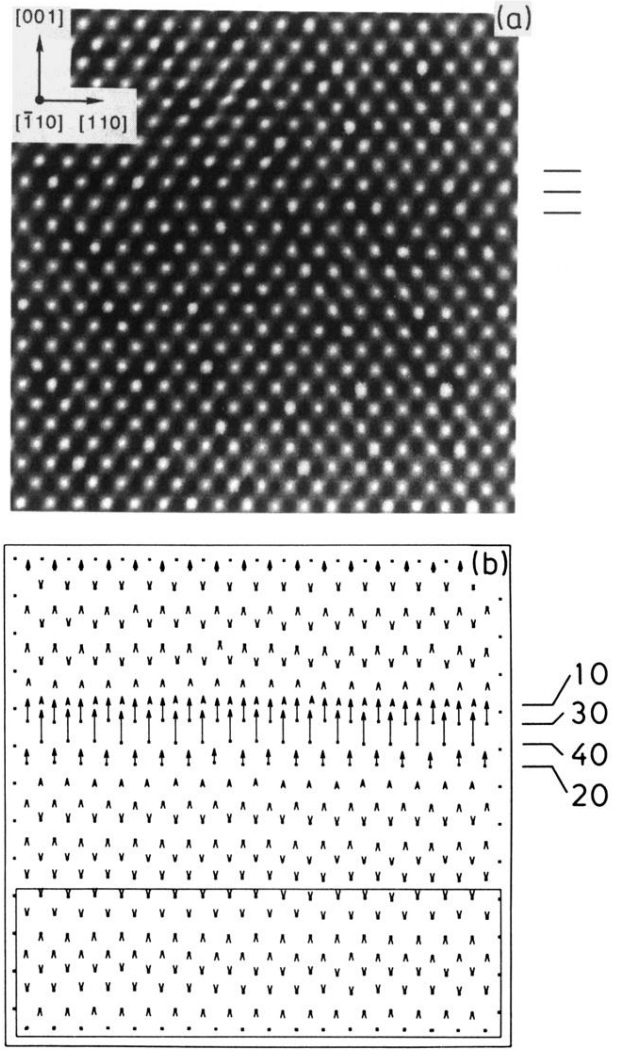

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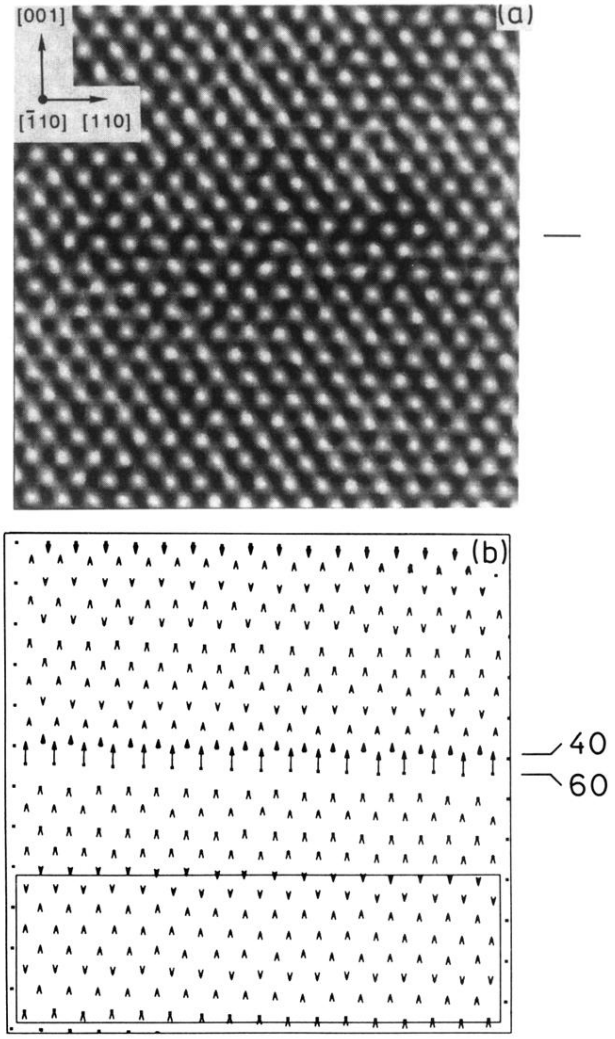

FIG. 3. (a) Fourier-filtered experimental lattice image of 1 ML InAs in GaAs. The bar indicates the plane containing In. (b) Vector representation of the relative shift in position of each ML with respect to its immediate neighbor. The magnitude of the vectors is amplified by a factor of 5. Numbers give the relative magnitude of the displacement vectors in percent. The cross section is oriented along [1$\bar{1}$0] instead of along [110], resulting in the inversion of the length ratio of the two displacement vectors at the interface with respect to the image simulation.