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Auxetic properties of cubic metal single crystals

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The behaviour of the Poisson's ratio is investigated on the basis of derived by the authors explicit equations determining the components of the compliance tensor in an arbitrarily rotated coordinate frame. Clear analytical expressions describing the angular dependencies of the auxetic properties have been found and the crystallographic regions possessing such auxetic behaviour have been revealed for single crystals of elemental metals. This was completed with the straightforward expressions for calculation of the maximum values of auxeticity. The connection between the auxetic properties and the level of anisotropy of single crystals as well as the Cauchy relation has been analysed for various metals with BCC and FCC lattices.

1 Introduction Materials having an intriguing property of negative Poisson's ratio are called auxetics [1], from Greek αὐξητικός – the one which tends to increase. If such material is stretched along a certain direction, it expands, rather than compresses along some axis in the plain perpendicular to the tension. In addition to auxetic materials occurring in nature (e.g. minerals [2, 3] or metals discussed below), a real explosion of new types and examples has come from material engineering. This is led by the pursuit of creating more materials with superior mechanical properties, such as higher resilience [4], increased shear stiffness and plane strain fracture toughness [5]. Starting from the first demonstration of auxetic foams [4], various porous [6, 7], composite [8, 9] and granulated [10, 11] auxetic materials have been reported, complemented by auxetic polymers [12] and semiconductors [13]. Such developments called for a vital need for understanding of this counter-intuitive phenomenon. Following the first mechanical model of a perfect auxetic [14], both pioneering works on numerical demonstration of auxeticity in isotropic thermodynamically stable material [15] and a rigorous analytical solution of the model of static auxetic phase [16] have been reported. These studies were succeeded by a model of rotating squares relevant to the auxeticity in inorganic crystalline materials [5] and a model describing the auxeticity mechanism in nematic liquid crystalline polymers [12]. Special interest was given to both experimental [17] and theoretical [18]

investigation of auxetic phenomena near phase transitions. Concluding the overview, in metals auxeticity presents a particular interest due to numerous practical applications. For the most recent developments in the auxetics research we refer the readers to a special section in *Physica Status Solidi B*, guest-edited by Alderson et al. [19] (see also references therein), which underlines the ongoing activity in these areas.

At a first sight, negative values of Poisson's ratios is an exotic phenomenon. Nevertheless, the possibility of such mechanical reaction to the external stimuli is confirmed in previous research and is in agreement with known relations of an isotropic elastic theory, connecting the Poisson's ratio ν with Young's modulus E, bulk modulus K and shear modulus G. From these relations it follows that if the conditions $2G > E$, $2G > 3K$ are satisfied, the negative values of the Poisson's ratio are possible; the range of possible values is given by $-1 \le \nu \le 0.5$. It has been shown that in the case of the polycrystalline materials negative Poisson ratio within the stability region of the original crystal is generally possible [20], but such materials, as well as crystalline materials that are auxetic in all the directional combinations, seem to be extremely rare in nature [21–23]. For the isotropic polycrystalline metals the nonauxetic behaviour can be easily seen from the positiveness (to the best of our knowledge of the reported data at normal conditions)

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Phys. Status Solidi B 251, No. 11 (2014)

Original

Paper

2315

of their elastic constants C_{11} and C_{12}:

$$\nu = \frac{C_{12}}{C_{11} + C_{12}} > 0. \qquad (1)$$

At the same time, in anisotropic metal single crystals it is not possible to assume *a priori* that these conditions are not satisfied and hence rule out the possibility of negative Poisson's ratios. Indeed, the negative Poisson's ratios have been observed in several metal single crystals [24–26], including those with BCC and FCC crystalline lattices [27, 28]. In fact, it has been theoretically shown that 69% of the cubic elemental metals show auxetic behaviour when stretched along the [110] direction [29]. This called for extensive theoretical research in this area. In the works by Brańka et al. [21, 30, 31] and Jasiukiewicz et al. [20] it have been proposed interesting approaches of mapping the auxetic behaviour of the cubic single crystals in the parametric coordinate frame, where the x and y axes represent combinations of the elastic constants of the materials. The areas of complete auxetics (materials that are auxetic for all the directional combinations), partial auxetics (materials that are auxetic for some stretch–strain directional combinations) and nonauxetics (not showing auxetic properties for any directional combination) have been demonstrated. Also, in the works of Brańka et al. [21, 30, 31] and Norris [32], the crystallographic directions for which the maximal auxeticity is achieved have been shown.

The studies reported above give the answer to the general question of the existence of auxeticity in a cubic single crystal. Furthermore, a general criterion of auxeticity was stated, expressed directly through the material's elastic constants, which gives the necessary and sufficient condition for the existence of auxetic behaviour for at least one crystallographic direction [23, 29, 33, 34] (complete or partial). However, they do not say anything about the range of directions for which the auxetic properties exist. At the same time, the range of such directions along with the range of the directions perpendicular to them for which the auxetic behaviour can be observed are of extreme interest. In several works addressing the auxetic directions, only a single crystallographic direction [110] [3, 28, 29, 35], or directions lying in a single crystallographic plane (110) [3, 36] were considered, without a comparative analysis of auxetic characteristics for other directions. All the directions of the applied stretch were considered in Refs. [3, 37], however the strain direction along the axis perpendicular to them was fixed to one particular angle [3], the auxetic behaviour studied in Ref. [37] was averaged over all perpendicular directions. Alternatively, an accurate numerical parametric 3D mapping of the angular space (two defining angles for the stretch direction and one defining the transverse strain direction) was presented in Ref. [22], but whilst giving an interesting overall picture, this findings might be difficult to interpret – an analytical theory on this subject would be of a great interest.

In this paper, we discuss the auxetic properties for all possible crystallographic directions in single crystals with cubic symmetry. In clear and explicit analytical expressions we reveal the domain of crystallographic directions possessing auxeticity as well as necessary and sufficient conditions of their existence. Furthermore, for each auxetic direction, we also determine the range of strain directions perpendicular to it for which the auxeticity can be observed. These explicit analytical results help to develop a clear picture of the angular dependencies describing the auxetic properties, and in a very straightforward way expose the role of the elastic constants. Finally, the correlations of auxeticity with the type of the cubic crystalline lattice and its level of anisotropy have been analysed, which was followed by the discussion of its link to the level of fulfilment of the Cauchy's relation.

2 Theoretical approach Our approach is based on the conversion of the compliance tensor in the main crystallographic axes xyz into a new one in coordinate system $x'y'z'$, rotated in respect to the former:

$$S'_{ijkl} = \alpha_{ip}\alpha_{jq}\alpha_{kr}\alpha_{ls}S_{pqrs}, \qquad (2)$$

where S'_{ijkl} are the components of the tensor in the new coordinate system $x'y'z'$, S_{pqrs} are the components in the old system xyz and α_{ip}, α_{jq}, α_{kr}, α_{ls} are the cosines of the angles between the corresponding coordinate axis of the new and the old coordinate systems (e.g. [27]). This transformation allows to find the tensor components S'_{ijkl}, required to determine the Poisson's ratio for an arbitrary crystallographic direction:

$$
\begin{aligned}
S'_{3333} = S'_{33} &= S_{11} - 2(S_{11} - S_{12} - S_{44}) \\
&\quad \times \left(\alpha_{31}^2\alpha_{32}^2 + \alpha_{31}^2\alpha_{33}^2 + \alpha_{32}^2\alpha_{33}^2 \right), \\
S'_{3322} = S'_{32} &= S_{12} + (S_{11} - S_{12}) \\
&\quad \times \left(\alpha_{31}^2\alpha_{21}^2 + \alpha_{31}^2\alpha_{22}^2 + \alpha_{33}^2\alpha_{23}^2 \right) \\
&\quad + S_{44}(\alpha_{31}\alpha_{21}\alpha_{32}\alpha_{22} + \alpha_{31}\alpha_{21}\alpha_{33}\alpha_{23} + \alpha_{32}\alpha_{22}\alpha_{33}\alpha_{23}).
\end{aligned}
$$
$$(3)$$

In the forthcoming analysis the investigated crystallographic direction is conventionally defined by the Euler angles, connected with α_{ij} in a straightforward way. The notations for the Euler angles are adopted as: α (rotation around z), β (rotation around y') and γ (rotation around z') (see inset of Fig. 1). In this format, α and β are the azimuthal and polar angles, respectively, defining the direction of the z'-axis in the original coordinate system, while γ signifies the direction of the Poisson's ratio determination in the plane perpendicular to the chosen z'. The latter angle is defined with respect to line of nodes N.

In the general case, the Poisson's ratio for the arbitrary chosen crystallographic direction z' can be expressed as

$$\nu(\alpha, \beta, \gamma) = -\frac{S'_{3j}(\alpha, \beta, \gamma)}{S'_{33}(\alpha, \beta)}, \qquad (4)$$
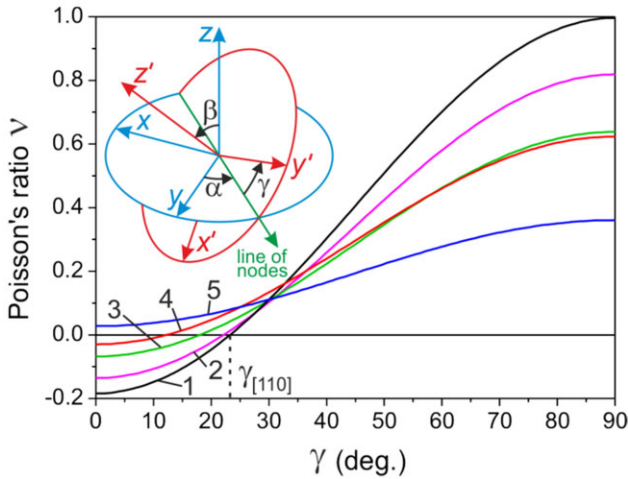
Figure 1 Dependencies of $\nu(45°, 90°, \gamma)$ for various metal single crystals: 1 – Pb, 2 – Cu, 3 – Ni, 4 – α-Fe, 5 – Ge. The inset shows the definitions for the Euler angles.

where $S'_{3j}(\alpha, \beta, \gamma)$ and $S'_{33}(\alpha, \beta)$ are the components of the compliance tensor for this direction. It should be noted that the rotation around z' by the angle of $\gamma = 90°$ results in change in the index of S'_{3j}:

$$S'_{3j}(\alpha, \beta, 0°) = S'_{32}(\alpha, \beta),$$
$$S'_{3j}(\alpha, \beta, 90°) = S'_{31}(\alpha, \beta). \tag{5}$$

In the framework of the coordinate system transformation described above, in a cubic single crystal the explicit expression for these components were derived as

$$S'_{33}(\alpha, \beta) = S_{11} - 2S\big[\sin^2(\alpha)\cos^2(\alpha)\sin^4(\beta) + \sin^2(\beta)\cos^2(\beta)\big], \tag{6}$$

$$S'_{3j}(\alpha, \beta, \gamma) = S_{12} + 2S\big[\sin^2(\alpha)\cos^2(\alpha)\sin^2(\beta)\cos^2(\gamma)$$
$$+ \frac{1}{8}\sin(4\alpha)\sin(\beta)\sin(2\beta)\sin(\gamma)\cos(\gamma)$$
$$+ \big(1 - \sin^2(\alpha)\cos^2(\alpha)\big)\sin^2(\beta)\cos^2(\beta)\sin^2(\gamma)\big], \tag{7}$$

where S_{11}, S_{12}, S_{44} are the components of the compliance tensor in the main crystallographic axis of the single crystal xyz, coinciding with unit cell edges, and $S = S_{11} - S_{12} - S_{44}/2$.

3 Discussion Since for conventional materials the value of $S'_{33}(\alpha, \beta)$ is always positive ($S'_{33}(\alpha, \beta) = 1/E$), the sign of the Poisson ratio is defined by the sign of the numerator in Eq. (4). Expressing this statement explicitly, the Poisson ratio will be negative if

$$S'_{3j}(\alpha, \beta, \gamma) > 0, \tag{8}$$

which is a necessary and sufficient condition for the existence of auxetic behaviour for single crystals of any symmetry.

The analysis of $\nu(\alpha, \beta, \gamma)$ based on Eqs. (4) and (7) allows to make some conclusions about the auxetic properties of overwhelming majority of cubic single crystals (with $S'_{33}(\alpha, \beta) > 0$, as explained above and $S_{12} < 0$ [23], otherwise see Refs. [21] and [32]). Firstly, if the auxetic properties exist (see the condition below, automatically setting $S > 0$), the highest negative value of the Poisson ratio is achieved for [110] crystal direction (and the directions analogous to it) at $\gamma = 0°$, the latter corresponding to determination of the Poisson ration in [1$\bar{1}$0] direction. This can be obtained by numerical minimisation of $\nu(\alpha, \beta, \gamma)$ in respect to all angle variables, which is equivalent in the considered case to the maximisation of the expression in square brackets in Eq. (7). The latter can be done by standard methods of mathematical analysis or by a straightforward numerical approach. For the above maximal Poisson ratio it was adopted a notation of $\nu(110, 1\bar{1}0)$ [29]. In notations of this form, which will be used henceforward, the first group of Miller indexes represents the direction of the uniaxial tension, while the second represent the direction of the Poisson's ratio determination. Thus, $\nu(110, 001)$ corresponds to the stretching in the same [110] direction, and observation along [001] ($\gamma = 90°$). These ($\nu(110, 1\bar{1}0)$ and $\nu(110, 001)$) are the extreme values, while the Poisson ratio experience a smooth monotonic evolution in the region between them with a change of γ (Fig. 1). It can be stressed that the values of $\nu(110, 001)$ are anomalously high in the presence of the auxetic effect. Furthermore, the higher they are, the more negative values of $\nu(110, 1\bar{1}0)$ are achieved. The extreme values of Poisson's ratios are determined by the relations

$$\nu(110, 1\bar{1}0) = -\frac{2S_{12} + S}{2S_{11} - S}, \tag{9}$$

$$\nu(110, 001) = -\frac{2S_{12}}{2S_{11} - S}. \tag{10}$$

Secondly, for the crystallographic directions gradually deviating from [110] either along α (keeping $\beta = 90°$) or β (keeping $\alpha = 45°$), the highest negative values of the Poisson ratio will remain to be at $\gamma = 0°$ (i.e. along y'), but their values will decrease, finally disappearing, which can be seen from a straightforward simplification of Eq. (7) for these cases. More importantly, the boundaries of the regions, comprising the crystallographic directions possessing auxetic properties, are defined by the condition of the numerator of Eq. (4) being zero:

$$S'_{3j}(\alpha, \beta, \gamma_{\max}) = 0, \tag{11}$$

where $\gamma = \gamma_{\max}$ maximises this function with respect to γ:

$$\left.\frac{\partial S'_{3j}(\alpha, \beta, \gamma)}{\partial \gamma}\right|_{\gamma=\gamma_{\max}} = 0, \quad \left.\frac{\partial^2 S'_{3j}(\alpha, \beta, \gamma)}{\partial \gamma^2}\right|_{\gamma=\gamma_{\max}} < 0. \tag{12}$$

Phys. Status Solidi B 251, No. 11 (2014)

2317

Original

Paper

These conditions can be rewritten explicitly as

$$\tan(2\gamma_{max}) = \frac{1}{8}\sin(4\alpha)\sin(\beta)\sin(2\beta)$$

$$\div \left[\sin^2(\alpha)\cos^2(\alpha)\sin^2(\beta)\right.$$

$$\left. - \left(1 - \sin^2(\alpha)\cos^2(\alpha)\right)\sin^2(\beta)\cos^2(\beta)\right],$$

$$\frac{1}{8}\sin(4\alpha)\sin(\beta)\sin(2\beta)\sin(2\gamma_{max})$$

$$+ \left[\sin^2(\alpha)\cos^2(\alpha)\sin^2(\beta)\right.$$

$$\left. - \left(1 - \sin^2(\alpha)\cos^2(\alpha)\right)\sin^2(\beta)\cos^2(\beta)\cos(2\gamma_{max})\right] > 0. \quad (13)$$

The second condition here defines the root of the first equation, which needs to be selected. The regions of auxeticity can be found semi-analytically from the above equations or directly via numerical maximisation of $S'_{3j}(\alpha, \beta, \gamma)$ over γ and calculating the sign of this parameter at found γ_{max}. Such regions are presented in Fig. 2 in the form of the first quarter of a stereographic projection plotted for single crystals of Pb, Cu, Ni and α-Fe. In the coloured areas all the directions, defined by the azimuthal angle α and polar angle β possess the auxetic behaviour (for a certain range of γ, see below). All the directions which are beyond these regions have positive Poisson ratios at any γ, correspondingly. Small black circles in the projection mark the special direction cases with the given Miller indexes. Here, it needs to be noted, that although in the stereographic projections of the regions with auxetic behaviour around [110], [011] and [010] directions have different configurations due to inequivalence of α and β in Euler angle definitions, they have the same configuration is spatial representation (projected on a sphere of spatial angles) due to the equivalence of the above crystallographic directions.

Thirdly, if present for given (α, β), the negative Poisson ratios exist only for the certain values of γ in the interval from $-\gamma_1$ to γ_2, the values of which correspond to the roots of

$$S'_{3j}(\alpha, \beta, \gamma) = 0. \quad (14)$$

The region of auxeticity observation over γ in a form of a continuous interval is stipulated by the fact that $S'_{3j}(\alpha, \beta, \gamma)$ can be represented as a periodic function given by first harmonics of 2γ, which leads to the above conclusion about the regions of its negative values (if present). Furthermore, the periodicity over 2γ represents physically clear symmetric response in respect to the stretching axis. Applied to the [110] direction, the above condition takes the explicit form

$$\cos^2\left(\gamma_{[110]}\right) = -\frac{2S_{12}}{S}. \quad (15)$$

For all $|\gamma| < \gamma_{[110]}$ the auxetic effect is observed ($\nu < 0$, Fig. 1).
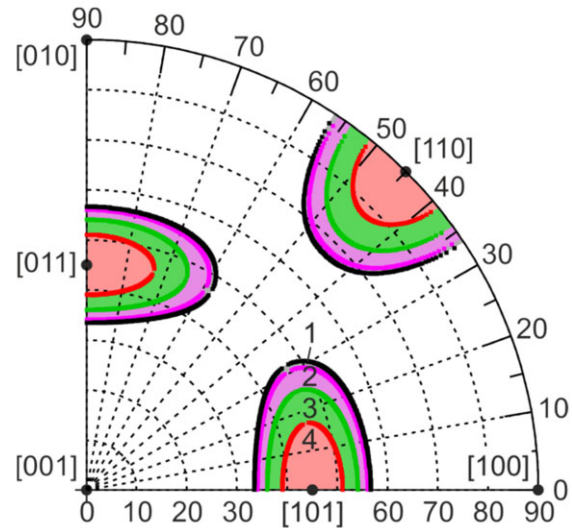

Figure 2 Stereographic projection of the regions with auxetic behaviour for single crystals of: 1 – Pb, 2 – Cu, 3 – Ni, 4 – α-Fe. The black line corresponding to lead encloses slightly larger area than the magenta line corresponding to copper.

Concluding the analysis, it is possible to formulate the conditions for existence of auxetic behaviour in metal single crystals with cubic lattice symmetry. The condition $S'_{3j}(\alpha, \beta, \gamma) > 0$ derived above is both necessary and sufficient for observation of negative Poisson's ratio for a given (α, β, γ) combination, but is too general and rather difficult to be used in estimations. However, it can be represented in a more specific and convenient form, separating the necessary and sufficient parts. As it follows from Eqs. (8) and (7) the necessary condition for the existence of directions with the auxetic behaviour is given by the inequality [23, 29, 33, 34]

$$S_{11} + S_{12} - S_{44}/2 > 0, \quad (16)$$

obtained from the global maximisation of expression in the square bracket in Eq. (7) over all the angles, returning the value of 1/4, which after the substitution in Eq. (8) gives $S_{12} + 2(S_{11} - S_{12} - S_{44}/2) \cdot 1/4 > 0$. The fulfilment of this condition means the existence of directions with negative Poisson ratios in the single crystal, but does not specify them. On the other side, the sufficient conditions were found above as

$$S'_{3j}(\alpha, \beta, \gamma_{max}) > 0, \quad (17)$$

$$-\gamma_1 < \gamma < \gamma_2. \quad (18)$$

The first inequality determines the region of crystallographic directions with auxetic behaviour, given by α and β while the second condition defines the interval of directions in the plane perpendicular to the chosen crystallographic direction, in which the auxetic properties reveal themselves.

To examine the possibility of auxetic behaviour in various BCC and FCC metals, the Poisson ratios were analysed for all the crystallographic directions for more than 30 single crystals, most of which were found to possess the auxetic behaviour. The results are summarised in Table 1. In the 3rd column the type of the crystallographic lattice is stated. Column 4 presents the maximal negative value of the Poisson's ratio, corresponding to [001] crystallographic direction (calculated using Eq. (9)), while column 5 gives angles $\gamma_{[110]}$ defining the region where the negative Poisson's ratio is observed in the plane perpendicular to this direction (calculated using Eq. (15)). The 6th column presents the maximal positive value of the Poisson's ratio for the investigated metals (calculated using Eq. (10)). In addition to the characteristics described above, the following parameters were introduced. The 7th column returns the values of the two-dimensional Poisson ratio, defined in terms of a change of the area of the perpendicular cross-section of a rod with [110] major axis under tension in this direction:

$$
\begin{aligned}
\nu_s = -\frac{\Delta s/s}{2\Delta l/l} &= -\frac{S'_{3j}(\alpha,\beta,0°)+S'_{3j}(\alpha,\beta,90°)}{2S'_{33}} \\
&= \frac{\nu(110,1\bar{1}0)+\nu(110,001)}{2}.
\end{aligned}
\tag{19}
$$

The 8th column gives the numerical values for the level of anisotropy of the considered single crystals, related to the maximal value of Young's modulus:

$$
A_E = \frac{E_{\max}-E_{\min}}{E_{\max}} = \frac{E_{[111]}-E_{[001]}}{E_{[111]}} = \frac{2S}{3S_{11}}.
\tag{20}
$$

This parameter returns $A_E = 0$ when applied to isotropic materials, while e.g. for a single crystal of tungsten, which is nearly isotropic, $A_E = 2 \times 10^{-4}$. For the cubic single crystals both positive ($E_{[111]} > E_{[001]}$) and negative ($E_{[111]} < E_{[001]}$) values of A_E are possible. In the 9th column the ratios C_{12}/C_{44} are given, reflecting how closely the material satisfies the Cauchy relation, which for cubic single crystals with central interatomic forces states $C_{12}/C_{44} = 1$. The deviation of C_{12}/C_{44} from unity is usually considered to be the evidence for non-central character of forces between the atoms, which can be a good approximation for crystalline media, though one have to note that strictly it is valid only in the limit of zero temperature and pressure (for counter-example see e.g. Ref. [40]).

The analysis of the calculation results listed in Table 1 allows to draw several conclusions about the auxetic properties of the considered metals:

– the auxetic behaviour is found in the metals with positive values of the anisotropy levels, for which $E_{[111]} > E_{[001]}$;

Table 1 Comparative analysis of the auxetic properties of cubic metal single crystals. The elastic constants along main crystallographic axes were taken from [38, 39].

| metal	group number	lattice type	$\nu(110,1\bar{1}0)$	$\gamma_{[110]}$ (deg.)	$\nu(110,001)$	ν_s	A_E	C_{12}/C_{44}
alkali metals								
Li	1a	BCC	−0.555	33	1.31	0.38	0.87	1.16
Na	1a	BCC	−0.525	34	1.16	0.32	0.84	0.79
K	1a	BCC	−0.412	31	1.16	0.37	0.81	1.42
Rb	1a	BCC	−0.399	30.5	1.15	0.38	0.81	1.49
Cs	1a	BCC	−0.362	33.5	0.83	0.24	0.72	0.66
transition metals								
Cu	1b	FCC	−0.136	22	0.819	0.34	0.65	1.64
Ag	1b	FCC	−0.095	18.5	0.821	0.36	0.64	2.03
Au	1b	FCC	−0.037	12	0.876	0.42	0.64	3.73
α-Fe	8a	BCC	−0.049	15.5	0.623	0.30	0.53	1.38
Co	8a	FCC	−0.149	24	0.762	0.31	0.64	1.24
Ni	8a	FCC	−0.068	18	0.638	0.29	0.55	1.17
Pd	8a	FCC	−0.100	18.5	0.877	0.39	0.66	2.48
alkali earth metals								
Ca	2a	FCC	−0.275	29	0.863	0.30	0.71	1.02
Sr	2a	FCC	−0.264	29	0.851	0.29	0.70	1.04
lanthanide metals								
Ce	3a	FCC	−0.154	26.5	0.626	0.24	0.58	0.82
Yb	3a	FCC	−0.330	34	0.715	0.19	0.71	0.56
actinide metals								
Th	3a	FCC	−0.223	28	0.764	0.27	0.67	1.02
post-transition metals								
Pb	4b	FCC	−0.184	23	0.996	0.41	0.72	2.72

Phys. Status Solidi B 251, No. 11 (2014)

2319

Original

Paper
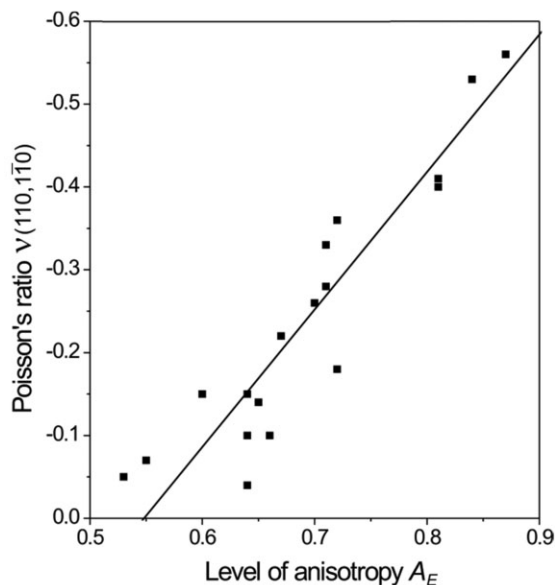
Figure 3 Correlation of $\nu(110, 1\bar{1}0)$ with the level of anisotropy A_E.

– the auxetic properties do not demonstrate a deterministic relation to the type of the crystal lattice, both BCC and FCC single crystals can possess them;

– all the considered metal single crystals from the first (1a and 1b) and 8a groups of the periodic table demonstrate the auxetic behaviour. In other groups this happens for the metal single crystals positioned at the lower part of the periodic table;

– the two-dimensional Poisson ratio ν_s for auxetics is always positive, which corresponds to the decrease of the area of perpendicular cross-section under applied tension;

– the highest negative values of the Poisson ratio, defined by $\nu(110, 1\bar{1}0)$, raise with the increase of the single crystal anisotropy level A_E. In the first approximation with a correlation coefficient $r = 0.94$, this relation can be considered as linear (Fig. 3);

– on the other hand, in the considered metal single crystals no correlation is observed between $\nu(110, 1\bar{1}0)$ and the level of fulfilment of the Cauchy relation, which calls for more cautious application of models considering only central forces (e.g. [29]) for explanation of auxeticity in a general case.

4 Conclusions In conclusion, for cubic crystalline lattices using realistic assumptions and a rigorous theoretical approach, explicit analytical expressions describing the behaviour of the Poisson's ratio for an arbitrary combination of spatial angles were derived. This allowed to reveal the areas of the spatial directions possessing negative Poisson's ratio and furthermore for any given direction in these areas, to find the directions in a plane perpendicular to it exposing such auxetic properties. Finally, the analysis of auxeticity in various metal single crystals was performed, their key auxetic characteristics were determined and a link between them and the level of the elastic anisotropy was demonstrated.

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