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Elastic and piezoresistive properties of nickel carbides from first principles

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The nickel-carbon system has received increased attention over the past years due to the relevance of nickel as a catalyst for carbon nanotube and graphene growth, where nickel carbide intermediates may be involved or carbide interface layers form in the end. Nickel-carbon composite thin films comprising Ni$_3$C are especially interesting in mechanical sensing applications. Due to the metastability of nickel carbides, formation conditions and the coupling between mechanical and electrical properties are not yet well understood. Using first-principles electronic structure methods, we calculated the elastic properties of Ni$_3$C, Ni$_2$C, and NiC, as well as changes in electronic properties under mechanical strain. We observe that the electronic density of states around the Fermi level does not change under the considered strains of up to 1%, which correspond to stresses up to 3 GPa. Relative changes in conductivity of Ni$_3$C range up to maximum values of about 10%.

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I. INTRODUCTION

Nickel-carbon compounds and composite thin films containing amorphous carbon are of high interest for various applications. Thin films have been investigated for their piezoresistive properties [1] and as low friction solid lubricants [2]. The metastable Ni$_3$C has been frequently observed in such films [3–5] and was suggested to cause piezoresistive behavior [1]. This carbide has been reported to be hard to distinguish from hcp nickel, where a study [5] suggests that hcp nickel is only stable in the presence of carbon and with some carbon content. A metastudy on this subject can be found in Ref. [4]. A recent study [6] confirmed that Ni$_3$C in such films only decomposes at temperatures well above 250°C.

The nickel-carbon system is also of interest for the catalytic production of carbon nanotubes (CNTs) and graphene. CNT growth was achieved both using nickel nanoparticles as a catalyst [7,8] and on carbon-nickel nanocomposite thin films [9]. While studies suggest that carbides do not form during CNT growth from Ni nanoparticles [10], Ni$_3$C has been observed in nanoparticles after CNT growth by plasma-enhanced chemical vapor deposition was stopped [11]. A more recent study [12] confirmed that Ni/Ni$_3$C core-shell structures can indeed be produced. In such a setup, the carbide could act as an advanced contact material for CNT junctions with properties similar to those demonstrated for Mo$_2$C [13]. The advantage would be that the Ni$_3$C-CNT unit can be grown bottom-up. Ni$_3$C does also occur as a parasitic by-product of carbon nanofiber growth on nickel foam [14].

Graphene [15] and graphene-type interfacial layers [16] can be produced by metal-induced crystallization and layer inversion as well as by epitaxial growth on transition metals, such as nickel [17]. In the latter case, one study [18] excluded the occurrence of crystalline Ni$_3$C on a polycrystalline Ni surface by XRD measurements. Others observed an interface layer between {111} nickel and graphene with the stoichiometry Ni$_2$C by Auger spectroscopy [19,20]. In both cases, mechanical details, especially of carbide intermediates, require further study.

The stability of a range of nickel carbides has been investigated by density functional calculations [21], yet neglecting the influence of elastic deformations which we address here. The study confirmed that, without externally induced strains, Ni$_3$C in space group 167 structure, Fig. 1(c), is the least unstable carbide and suggests that Ni$_5$C is most stable in orthorhombic structures of space groups 058 (Pnnm) and 060 (Pbcn); see Figs. 1(a) and 1(b).

All in all, especially the phase Ni$_3$C has potential technical applications in heterojunctions consisting of nickel and carbon allotropes, including (CNTs), acting as electrical circuit elements, for example piezoresistive sensors [22]. In these applications, the mechanical and piezoresistive properties of a potential carbide layer between nickel and the carbon structure can become relevant when the device is being strained during operation or when the layer is under constant epitaxial stress which may be caused the large surface tension of nickel [23].

The present work focuses on investigating the elastic properties of the three nickel carbides NiC, Ni$_2$C, and Ni$_3$C in their most stable crystallographic structures. Ground state properties of the carbides are compared in Sec. III A; the obtained elastic properties are discussed in Sec. III B. For the experimentally most relevant carbide, Ni$_3$C, the influence of strain on the electronic transport properties is discussed in Sec. III C.
When calculating ground state properties of carbides the Brillouin zone was sampled with a Monkhorst-Pack grid of \(12 \times 12 \times 12\) \(k\) points. Thermal smearing of Fermi-Dirac type \[33\] was fixed to about 27 meV(\(1 \times 10^{-5}\) Ha). Since the unit cells of nickel and diamond are smaller, denser grids of \(32 \times 32 \times 32\) and \(16 \times 16 \times 16\) \(k\) points, respectively, were required in order to get comparable sampling accuracy.

The ground state formation energies per f.u. for the carbides were calculated according to

\[
\Delta E_f = E_{\text{Ni}_x C_y} - x E_{\text{fcc Ni}} - y (E_{\text{diamond}} - 25 \text{ meV}),
\]

where \(E_i\) is the total energy of compound \(i\). Diamond was calculated as the carbon reference structure instead of graphite because the employed method is not capable of correctly calculating van der Vaals interactions. An empiric correction of \(\Delta E_C = 25 \text{ meV per carbon atom, also used in Ref. [21], was applied to obtain formation energies with respect to graphite.}"

**II. COMPUTATIONAL METHODS**

**A. Electronic structure calculations**

NiC was calculated in rocksalt (B1) structure, for \(\text{Ni}_x C_y\) the structures proposed by Gibson et al. \[21\] were used, and for \(\text{Ni}_x C_y\) the rhombohedral (bainite, space group 167) structure, which was experimentally found by Nagakura \[24\], was assumed [see Fig. 1(c)].

All results presented here were obtained applying (DGT), in the (GGA) in the Perdew-Burke-Ernzerhof (PBE) parametrization \[25\] as the exchange-correlation functional, which is known to give good results for bulk mechanical properties when comparing to experiments \[26\]. The plane-wave implementation in the ABINIT package \[27–29\] was used, employing the (PAW) method \[30\]. The PAW atomic data sets treat 3\(d^8 4s^2\) and 2\(s^2 2p^2\) as valence for nickel \[31\] and carbon \[32\], respectively.

For numerical accuracy, the plane-wave cutoff was converged to \(E_{\text{cut}} \sim 980\) eV (\(\approx 36\) Ha). Only, for calculations of fcc nickel, the cutoff was set to about 1360 eV (50 Ha) in order to reach a convergence of total energy below about 2.7 meV(\(1 \times 10^{-5}\) Ha) per atom. At this point energy differences under strain are converged to below about 0.27 \(\mu\)eV(\(1 \times 10^{-8}\) Ha), which is far more accurate than required for structural relaxation and the calculation of elastic properties. The stronger total energy criterion was chosen with regard to calculating formation enthalpies.

**B. Frozen phonon calculations**

Within the linear regime, elastic properties can be described by the elastic tensor \(\mathbf{\hat{C}}\), which gives the stress response \(\mathbf{\sigma}\) of a material proportional to a deformation \(\mathbf{\varepsilon}\):

\[
\sigma_i = \sum_j c_{ij} \cdot \varepsilon_j.
\]

Here, Voigt’s notation is used to write the stress and deformation tensors as six-vectors \((11 \rightarrow 1; 22 \rightarrow 2; 33 \rightarrow 3; 23 \rightarrow 4; 13 \rightarrow 5; 12 \rightarrow 6)\), with entries corresponding to three axial strains \((1–3)\) and shear strains \((4–6)\). In this way the elastic tensor can be written as a \(6 \times 6\) matrix from which all elastic properties can be derived. The bulk modulus is given by

\[
B = \frac{1}{3} (\langle c_{11} \rangle + 2 \langle c_{12} \rangle),
\]

where \(\langle c_{11} \rangle\) denotes an average over the diagonal axial strain entries and \(\langle c_{12} \rangle\) an average over the off-diagonal axial strain entries.

The entries of the elastic tensor were calculated using the (FP) method, where the stress response was derived from ground state calculations of the deformed primitive cell. A more detailed explanation can be found in \[34\]. The six primitive deformations were applied separately with magnitudes ranging up to 1%. All elastic constants were then determined using Eq. \(2\). The diagonal entries of the tensor can also be determined from the total energies of the same calculations:

\[
E_{\delta} = E_0 + \frac{V_0}{2} \sum_i c_{ii} \cdot \varepsilon_i^2,
\]

where \(E_0\) and \(V_0\) are the total energy and volume of the unstrained cell. The calculated tensors were checked for consistency by comparing the results of Eqs. \(2\) and \(4\). The calculation parameters were converged until the difference between the diagonal tensor elements from both equations was less than 2GPa. This criterion called for using a \(48 \times 48 \times 48\) \(k\)-point grid for the deformed cell of NiC; for the other materials it was met by using the aforementioned simulation parameters.
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If the material’s unit cell exhibits internal degrees of freedom, performing a ground state calculation of the deformed cell without relaxation of the ion positions yields entries of the so-called clamped-ion elastic tensor $C^c$. To obtain the more physical relaxed-ion elastic tensor $C^r$, the internal atomic coordinates were relaxed using the Broyden-Fletcher-Goldfarb-Shanno algorithm as implemented in ABINIT until all forces were below $5 \times 10^{-4}$ eV/Å.

C. Electronic transport

Electronic transport was calculated assuming constant relaxation time $\tau$ within the Boltzmann formalism where the conductivity tensor at zero temperature is given as

$$\sigma_{ij} = \tau \frac{e^2}{(2\pi)^3} h \sum_n \int_{E(E_{Fermi})} dS \frac{\nu^n_i(k)\nu^j_n(k)}{|\nu^a_n(k)|},$$

with $\nu^n_i(k) = \frac{1}{\hbar} \nabla_{k} \varepsilon^n_i(k)$, \(i, j\) denote Cartesian vector components.

Off-diagonal elements of $\sigma_{ij}$ are zero by symmetry. For the relaxation time $\tau$ no specific value is assumed, though it might be anisotropic ($\tau_{xx} \neq \tau_{yy} = \tau_{yy}$) in the case of Ni$_3$C due to its rhombohedral structure [36]. The integrals on the right-hand side of Eq. (5) reflect the anisotropy of the band structure of the unperturbed, but eventually strained, systems at the Fermi level. Assuming that $\tau$ remains constant under strain in the linear regime, since no new scattering centers are created, predictions can be made about the change of conductivity under strain.

For strained cells, the band structure with relaxed ion positions was used as basis for these calculations.

III. RESULTS AND DISCUSSION

A. Ground state results

The lattice parameters of the investigated materials are available in the literature, some even from experiments. The obtained lattice parameter for fcc nickel in the present work ($a_{\text{Ni}} = 3.524$ Å) agrees very well with values found in the literature [21,37]. The obtained lattice parameter for diamond ($a_{\text{diamond}} = 3.577$ Å) is only slightly larger than the experimental value of 3.567 Å [38]. Lattice parameters obtained for the carbides as well as formation enthalpies will be given for comparison, the latter with respect to fcc Ni and graphite.

(a) NiC. Assuming rocksalt structure, the lattice parameter $a_{\text{NiC}} = 4.073$ Å was obtained, which is in good agreement with Ref. [21] (4.077 Å) and other numerical studies cited therein. The calculated formation enthalpy of $\Delta E_f,\text{NiC} = 49.7$ kcal/mol of f.u. also agrees with Ref. [21] (48.6 kcal/mol).

(b) Ni$_3$C. The calculated values for the two investigated structures are summarized in Table I. The lattice parameters are in excellent agreement with Ref. [21]. Only, the formation enthalpies stated therein disagree with the present results (see Table I, values in parentheses). However, there is agreement on the prediction that both structures are essentially degenerate, with the variant of space group 060 being less than 5 meV lower in total energy.

(c) Ni$_3$C. The obtained lattice parameters $a = 4.60$ Å and $c = 13.00$ Å are in good agreement with Ref. [21] ($a = 4.49$, $c = 13.02$ Å) and electron diffraction measurements [39] ($a = 4.553$, $c = 12.92$ Å). A formation enthalpy of $\Delta E_f,\text{Ni}_3\text{C} = 6.3$ kcal/mol was obtained, which is identical to the value reported in Ref. [21] and reflects the thermal decomposition observed in [40].

For the relaxed, strain-free geometries, all carbides of nickel investigated here are metastable at $T = 0$ K, nonmagnetic, and metallic as observed previously [21]. Ni$_2$C exhibits a very low (DOS) around the Fermi energy. The DOS for the investigated carbides and the reference phases are plotted in Fig. 2.

In all carbides the C 2s band is located below the conduction band. It is shifted to lower energies (shifted left in Fig. 2) for Ni$_2$C and Ni$_3$C in comparison to the carbide with higher carbon content, NiC, indicating that a deeper potential well for electrons is provided by the carbon atoms. They are also more strongly negatively charged than in NiC. The Ni 3d states are located below the Fermi energy above about $-5$ eV for Ni$_2$C and Ni$_3$C; for NiC they are spread over a broader energy range, starting at around $-6$ eV. The Ni 3d orbitals do contribute to the DOS at the Fermi level, but much less than in fcc nickel, where the 3d DOS of the minority spin peaks at the Fermi level.

![FIG. 2. Total and projected DOS of carbides compared to those of the reference phases. For display, Gaussian smearing of about 270 meV (1x10⁻² Ha) was applied. Atomic orbitals are indicated as obtained from calculations of projected DOS; filled areas under curves indicate the fraction of DOS attributed to C sites. The arrows indicate the bonding band with C 2p and hybrid Ni states.](image-url)
level. Ni 4s and C 2p states also contribute to the DOS at the Fermi level. The part of the conduction band below about −5 eV is predominately composed of C 2p states hybridizing with Ni states; see arrows in Fig. 2.

B. Elastic constants

As a reference, the elastic tensors of fcc Ni and diamond were calculated and the nonzero, not symmetrically equivalent elements are provided in Table II. The calculated bulk modulus for diamond is identical to earlier theoretical works [41] and also the tensor components agree with earlier literature data [42]. The bulk modulus for Ni is within about 10 GPa of experimental results [43]. This deviation is predominantly attributed to the approximations involved in DFT. The following predictions for the elastic properties of nickel carbides can be expected to have about the same accuracy.

All carbides exhibit a larger bulk modulus than nickel and a much lower one than diamond, as apparent from the last column of Table II. Being the carbide with the highest carbon content, NiC shows the largest bulk modulus of the carbides. Evidently, the bulk modulus increases with increasing carbon content; that is, the substances become harder. Table II lists all calculated nonzero and not symmetrically equivalent elastic constants. The carbides Ni$_2$C and Ni$_3$C exhibit less symmetric unit cells, resulting in more independent entries in the elastic tensor.

(a) Ni$_2$C. Both investigated hypothetical forms of Ni$_2$C are predicted to be equally hard and even show quite similar anisotropies, probably due to the fact that both are orthorhombic. The elastic properties of the sample should not depend on the relative prevalence of these two phases. Still, judging by the elastic tensors, deforming one cell into the equilibrium shape of the other and allowing the atoms to rearrange into the other structure by relaxation requires overcoming a large potential barrier [44]. Thus, even under stress, both structures can be expected to coexist in one sample.

(b) Ni$_3$C. Judging by the obtained bulk moduli, a macroscopically isotropic polycrystalline sample of Ni$_3$C is predicted to be about as hard as the less stable Ni$_2$C. Even for the most extreme simulated deformations of 1% stresses were found to be still in the linear regime. Using the calculated value for $c_{11}$, a compression in the $e_1$ direction of this magnitude corresponds to applying a pressure of about 2.7 GPa, which by far exceeds pressures achievable in most experiments.

Investigating the DOS and band structure of deformed cells, no qualitative difference with respect to that obtained for the equilibrium geometry was found [44]. For purely axial strains and compressions [i.e., $e_1$, $e_2$, and $e_3$; see Fig. 3(b)] bands move slightly closer to the Fermi level under strain and farther away

![FIG. 3. Band structure of equilibrium Ni$_3$C for a large energy range (a) and for a small interval around $E_{\text{Fermi}}$ in comparison with 1% uniaxially distorted cells [(b) and (c)]. The displayed deformation directions are the ones corresponding to the largest strain (b) and shear (c) components of the relaxed elastic tensor. Bands of positively deformed cells are plotted in red dash-dotted lines, negative deformations in blue dashed lines.](image-url)
under compression. This can be attributed to changing overlaps between atomic orbitals. This difference is marginal close to and at the Fermi level, the region most relevant to transport properties. For pure shear deformations no significant changes are observed; see Fig. 3(c). Band structures shown in Fig. 3 for deformed cells use relaxed ion positions. Clamped cells show qualitatively identical changes; only the shift of bands for strains and compressions is larger.

### C. Electronic transport under strain

A closer analysis of the Ni$_3$C band structure yields a density of states of $2.83 \pm 4\%$ per f.u. and anisotropic averaged Fermi velocities of $v_x = v_y = 0.90 \times 10^6$ m/s and $v_z = 1.10 \times 10^6$ m/s. This results in an in-plane/out-of-plane conductivity anisotropy of about 0.67.

The respective effect of axial strains in the $x$ and $z$ directions ($e_1$ and $e_3$) on electronic transport was investigated. For strains up to $\pm 1\%$ the DOS remains unaffected within the precision of the calculation. Table III summarizes relative changes under strain in both conductance and Fermi velocities. The strongest changes in conductance can be observed for strains along $e_3$, where the parallel conductance in the $z$ direction changes by $10\%$; the anisotropy increases slightly. For small strains along $e_1$ the in-plane isotropy ($xx, yy$) is unaffected, while the in-plane/out-of-plane anisotropy changes slightly.

Computation and analytical investigations of the piezoresistivity of semiconducting (CNTs) suggest an increase in resistance of well above $50\%$ under longitudinal strains of about $0.3\%$ [45,46]. To exert the required stress on a Ni$_3$C-contacted CNT, the contact material would be strained by about $1\%$, given the ratio of the established Young’s modulus of (CNTs) of about 1000 GPa [47] and the elastic coefficients we obtained for axial strains in Ni$_3$C. This would cause a change in conductivity of $3\%$ to $10\%$ in the contact material, which is significantly smaller than the effect observed in the CNT. Thus, the piezoresistive properties of a device with Ni-contacted (CNTs) as functional structure are dominated by the electronic response of the (CNTs) to mechanical deformations.

### IV. CONCLUSIONS

The complete sets of elastic constants of nickel carbides have been calculated in a way that they can be expected to be within 10 GPa of experimental values. The electronic structure and electronic transport properties of bulk Ni$_3$C under stress have been investigated. Assuming a constant relaxation time $\tau$, changes in conductivity not exceeding about $4\%$ in-plane and about $10\%$ out-of-plane for stresses below $3GPa$ are predicted. As a contact material in sensing applications these changes are of minor significance. These results also show that Ni$_3$C does not contribute significantly to the piezoresistive effects observed in nickel-carbon thin films by Uhlig and coworkers [1].

For the formation enthalpy of both Ni$_3$C variants, the absolute values obtained here differ quantitatively from those by Gibson et al. [21], but qualitatively both studies agree on the relative ordering with respect to the other carbide phases. On the enthalpy difference between the ground state Ni$_2$C variants the agreement is excellent. The studies also agree on the formation enthalpies of the other carbides.

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