### Models for H isotope and He retention in irradiated tungsten

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### Outline

- Conceptual framework
- Overall objectives
- Immediate objectives
- Relation to experimental observations





### Conceptual framework: multiscale modelling



### Limitations of the "multiscale modelling" concept

Microstructure, mechanical properties

**Atomic structure** 

**Electronic structure** 



<u>Non-collinear</u> magnetism in Fe-Cr alloys, α-γ-δ phase transitions, high-T elastic instability of defects and dislocations

Effect of magnetism on zero-T phase stability of Fe and Fe alloys

Structure and magnetism of defects and dislocations

The time-dependent dynamics of noncollinear magnetism

The origin of magnetism: spontaneous symmetry breaking at electronic level



# Limitations of the "multiscale modelling" concept

Microscopic parameters – specifically hydrogen and helium trapping energies – control macroscopic experimental observations.



Please see the presentation by M. Mayer *et al.* given earlier

- (i) Desorption is controlled by certain binding energies through the Arrhenius law:  $exp(-E_b/kT)$ . The origin of  $E_b$  is quantum-mechanical.
- (ii) There appear to be not too many binding energy values required to describe the TDS curves, the curves do not appear to exhibit many features





### Current position with modelling

DFT calculations can now predict, at the ~0.1 eV level of accuracy, the energies of formation and migration of SMALL objects (~500 atom simulation cells, defect configurations containing less than ~10 atoms).

Advances made over the last 15 years are spectacular – DFT calculations generate real knowledge of phenomena where performing experimental observations is often not even possible.

The drawback: small system size. This is fundamental, and is related to the fact that a DFT calculation necessarily involves the investigation of electronic structure, even if this information is not used for any purpose.

DFT calculations have not yet been carried out for dislocations that may be relevant to the interpretation of TDS observations





### The Damage and Recovery of Neutron Irradiated Tungsten<sup>†</sup>

By M. W. Thompson

Atomic Energy Research Establishment, Harwell, Berks.

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#### ABSTRACT

Residual electrical resistivity has been used as an index of the damage present in tungsten irradiated in a nuclear reactor at  $4^{\circ}$ k and  $77^{\circ}$ k. A comparison was made between four different types of specimen prepared from cold-drawn wire respectively annealed before irradiation at 2000, 1500, 800°c and not at all. A fifth type of specimen was prepared from wire in a different state of purity.

Four stages of recovery have been identified, the first occurring below  $-170^{\circ}$ c, the second between -170 and  $350^{\circ}$ c with an activation energy rising from 0.25 to 1.7 ev, the third between 350 and 450°c with a single activation energy of 1.7 ev and the fourth above 450°c. Second-stage recovery was enhanced by the presence of cold-work, affected by impurities and in well-annealed samples after light irradiations it was suppressed.

A comparison of recovery with that in other metals leads to the conclusion that vacancies in tungsten migrate at  $400^{\circ}$ c with an activation energy of 1.7 ev. The general suggestion is advanced that vacancy migration occurs in all metals

### Vacancy formation and migration energies

	Al	Cu	Au	Ni	Pd	Pt	Pu
$E_f$	0.580 <sup>i</sup>	1.04 <sup>d</sup>	$0.782^{i}$	1.37, <sup>e</sup> 1.43, <sup>r</sup>	1.70 <sup>j</sup>	1.18 <sup>j</sup>	1.31, 1.36, 1.08 <sup>t</sup>
				1.65 <sup>r</sup>			
$E_m$	0.57 <sup>m</sup>	0.72 <sup>d</sup>	_	1.285, <sup>e</sup> 1.08 <sup>r</sup>	_	1.51 <sup>j</sup>	_
	V	Nb	Ta	Cr	Мо	W	Fe
$E_{f}$	$2.51^{1}$	2.99 <sup>1</sup>	$3.14^{1}$	2.64 <sup>1</sup>	2.96, <sup>j</sup> 2.96 <sup>l</sup>	3.56 <sup>1</sup>	2.02, <sup>b</sup> 2.07, <sup>k</sup> 2.15 <sup>l</sup>
$E_m$	0.621	0.91 <sup>1</sup>	1.48 <sup>1</sup>	0.91 <sup>1</sup>	1.28 <sup>1</sup>	$1.78^{l}$	0.65, <sup>b</sup> 0.67, <sup>k</sup> 0.64 <sup>l</sup>
	С	Si	Ge	Be	Ti	Zr	Hf
$E_{f}$	8.2 <sup>f</sup>	3.17, <sup>c</sup> 3.29 <sup>g</sup>	2.3 <sup>h</sup>	0.81, <sup>n</sup> 1.09 <sup>o</sup>	1.97, <sup>p</sup> 2.13 <sup>q</sup>	2.17, <sup>q</sup> 1.86 <sup>s</sup>	2.22 <sup>q</sup>
$E_m$	1.7 <sup>f</sup>	0.4 <sup>g</sup>	_	0.72B,	0.47B,	0.51B,	0.79B, 0.91NB <sup>q</sup>
				0.89NB°	$0.61 \mathrm{NB^{p}}$	$0.67 \mathrm{NB}^{\mathrm{q}}$	

DFT calculations are able to predict vacancy migration and formation energies for materials where there are no experimental data available. Values derived from DFT calculations are free from the effect of impurities. It is also possible to perform calculations and determine vacancy-impurity interaction energies.



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## Formation and migration energies of SIAs

	(111)	(110)	(100)	Tetrahedral	Octahedral	$E_m$
Fe	4.66, <sup>b</sup> 4.45 <sup>c</sup>	3.94, <sup>b</sup> 3.75 <sup>c</sup>	5.04, <sup>b</sup> 4.75 <sup>c</sup>	4.26 <sup>c</sup>	4.94 <sup>c</sup>	0.34 <sup>c</sup>
V	3.37, <sup>d</sup> 3.14 <sup>e</sup>	3.65, <sup>d</sup> 3.48 <sup>e</sup>	3.92, <sup>d</sup> 3.57 <sup>e</sup>	3.84, <sup>d</sup> 3.69 <sup>e</sup>	3.96, <sup>d</sup> 3.62 <sup>e</sup>	
Nb	5.25 <sup>d</sup>	5.60 <sup>d</sup>	5.95 <sup>d</sup>	5.76 <sup>d</sup>	6.06 <sup>d</sup>	
Ta	5.83 <sup>d</sup>	6.38 <sup>d</sup>	7.00 <sup>d</sup>	6.77 <sup>d</sup>	7.10 <sup>d</sup>	
Cr	5.66 <sup>d</sup>	5.68 <sup>d</sup>	6.64 <sup>d</sup>	6.19 <sup>d</sup>	6.72 <sup>d</sup>	
Mo	7.42, <sup>d</sup> 7.34 <sup>e</sup>	7.58, <sup>d</sup> 7.51 <sup>e</sup>	9.00, <sup>d</sup> 8.77 <sup>e</sup>	8.40, <sup>d</sup> 8.20 <sup>e</sup>	9.07, <sup>d</sup> 8.86 <sup>e</sup>	
W	9.55 <sup>d</sup>	9.84 <sup>d</sup>	11.49 <sup>d</sup>	11.05 <sup>d</sup>	11.68 <sup>d</sup>	
Al	1.959 <sup>f</sup>	1.869 <sup>f</sup>	1.579 <sup>f</sup>	1.790 <sup>f</sup>	$1.978^{f}$	0.084 <sup>f</sup>
Ni	4.69 <sup>g</sup>	4.99 <sup>g</sup>	4.07 <sup>g</sup>	4.69 <sup>g</sup>	4.25 <sup>g</sup>	0.14 <sup>g</sup>
Si	3.84 <sup>h</sup>	3.80 (hexagonal)	3.85 (caged)	4.07 <sup>h</sup>	4.8	0.18 <sup>h</sup>

DFT calculations prove particularly useful, in fact irreplaceable, in the treatment of selfinterstitial atom (SIA) defects. The formation energies of SIA defects are much larger than the formation energies of vacancies, and SIAs do not form at temperatures below 1000°C. They do form under irradiation (a Frenkel pair = a vacancy + a SIA), making DFT an essential tool for the treatment of radiation damage phenomena.



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## Current position with modelling

DFT calculations now can predict, accurately, at the ~0.1 eV level of accuracy, the energy of formation and migration of SMALL objects (~500 atom simulation cells, defect configurations containing less than ~10 atoms).

MD simulations rely on semi-empirical potentials that have lower accuracy (sometimes <u>very</u> low accuracy). Well benchmarked semi-empirical potentials are either not available, or are under development, for the W-hydrogen system. Some potentials are suitable for defects, some for surfaces – potentials appear to be application specific. Transferability of potentials is the key issue, and also the key difficulty impeding applications.

For the W-He case the position is slightly better, since the physics of He-metal interaction is clearer.

kMC simulations – the high quality ones – are based on direct input from DFT, less often MD (the accuracy issue).

MD simulations are necessary for modelling large-scale systems, e.g. dislocations, presently there appears to be no other way.



Rate theory – also relies on DFT input, MD plays an important role (vacancy clusters). 🚦

# Immediate objectives, relation to experiment (I)

- Threshold displacement energies for defect production in tungsten, can be found from simulations, experiments could be done using HVEM – could this be actually accomplished?
- Binding and trapping energies, involving not only single defects but also defect complexes – e.g. vacancy clusters: it is clear that accurate values can only be derived from DFT. TDS experiments show that there are only a few binding energy values that characterise the desorption curves.
- Interpretation of macroscopic TDS experimental data necessarily requires the use of rate theory type, or kMC, or both approaches. So far information about defect, dislocation and grain boundary microstructure is not included in such simulations.
- Relating TDS measurements to microstructure is probably a reasonable overall objective for the modelling effort.





# Immediate objectives, relation to experiment (II)

- Standard tungsten samples
- Standard preparation of the samples: annealing, implantation, followed by characterization (PAS), for the comparison of TDS curves – possibly for hydrogen and helium
- Comparison of TDS experiments, accompanied by comparison of interpretations using various codes (TMAP, kMC). Are the effective trapping/activation energies derived from various experimental TDS curves going to be the same?
- Comparison of software for the interpretation of TDS experiments. Need a "standard" TDS curve, for hydrogen, and for helium desorption.
- TDS experiments a range of experiments with variable desorption rates.
- Depth distribution of hydrogen and deuterium and tritium as a likely source of uncertainties
- Specifically a modelling task: the development of potentials for MD of tungsten, comparison of potentials with DFT, for various configurations.



