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Tracer diffusion and PAC measurements of ^{111m}Cd tracer atoms in A_3B compounds

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

S.V. Divinski

D. Gaertner, G. S. Collins[2], M. Zacate[3], J. Schell[4,5], K. Johnston[4], T. T. Dang[5], D. C. Lupascu[5], T. Hickel[6,7]

Institute of Materials Physics, Münster University, Germany

[2]Pullman Professor of Physics, Washington State University (WSU), Pullman, WA, USA
[3]Physics and Geology Department, Northern Kentucky University Highland Heights, KY, USA
[4]European Organization for Nuclear Research (CERN), CH-1211 Geneva, Switzerland
[5]Institute for Materials Science and Center for Nanointegration Duisburg-Essen (CENIDE), University of Duisburg-Essen, 45141 Essen, Germany
[6]Department of Computational Materials Design, Max-Planck-Institut für Eisenforschung GmbH, Max-Planck-Straße 1, 40237 Düsseldorf, Germany
[7]BAM Bundesanstalt für Materialforschung und -Prüfung, Richard-Willstätter-Str. 11, 12489 Berlin, Germany

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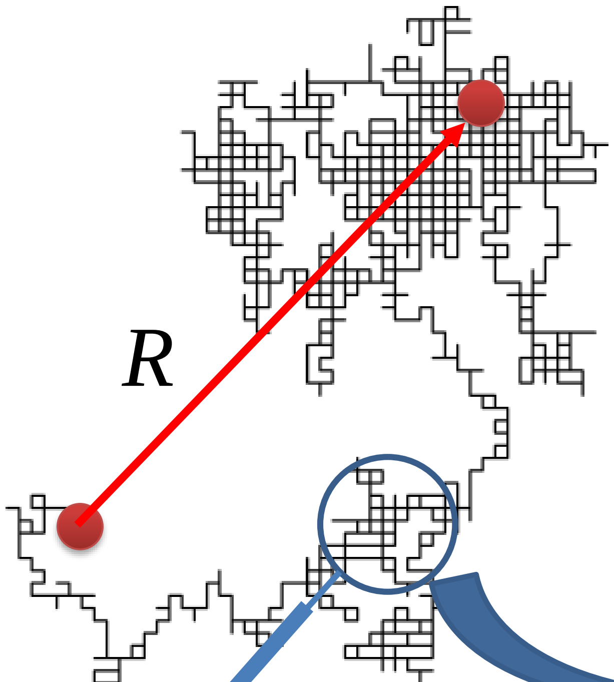
Correlation factor in diffusion

$$D^* = \frac{\langle R^2 \rangle}{4t}$$

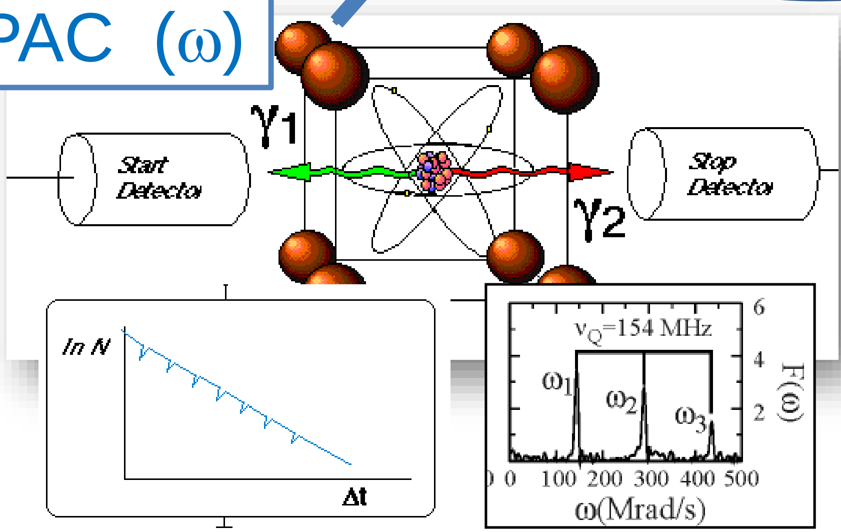
$$D_{random} = \frac{n\lambda^2}{4n\tau} = \frac{\lambda^2}{4}\omega$$



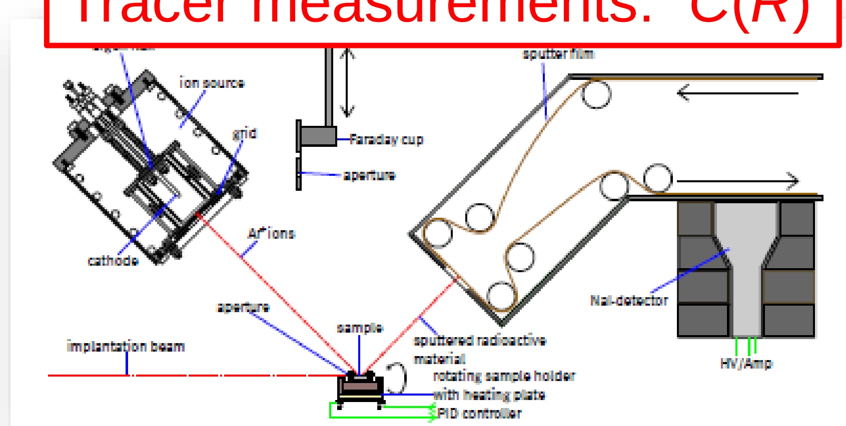
$$f \equiv \frac{D^*}{D_{\mathrm{random}}}.$$

$$f = \lim_{n\to\infty} \frac{\langle R^2 \rangle}{\langle R^2_{\mathrm{random}} \rangle} = 1 + 2 \lim_{n\to\infty} \frac{\sum_{i=1}^{n-1} \sum_{j=i+1}^{n} \langle r_i r_j \rangle}{\sum_{i=1}^{n} \langle r_i^2 \rangle}$$

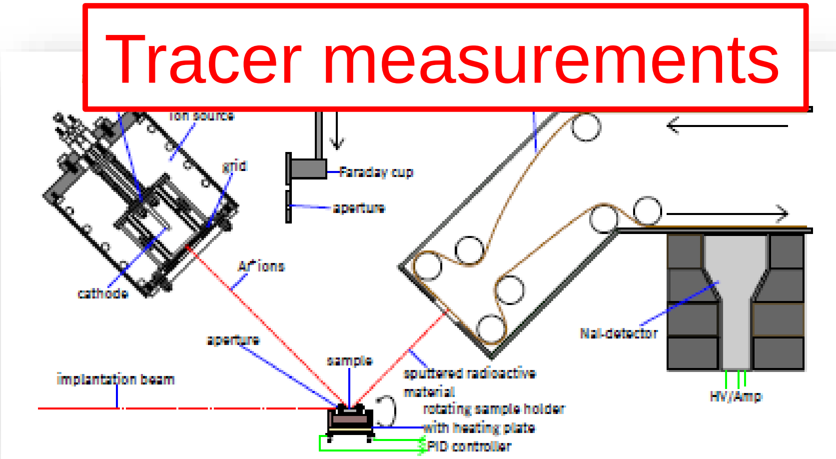
PAC (ω)



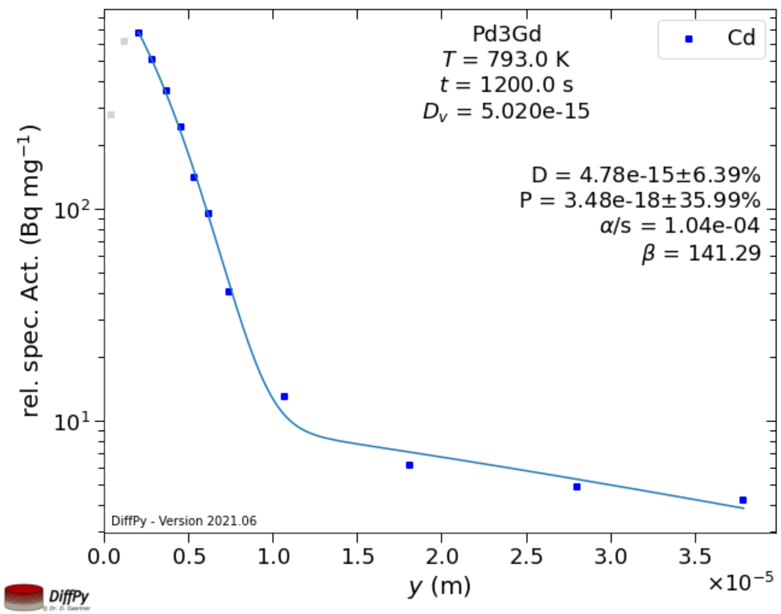
Tracer measurements: $C(R)$



Correlation factor in diffusion (tracer diffusion)

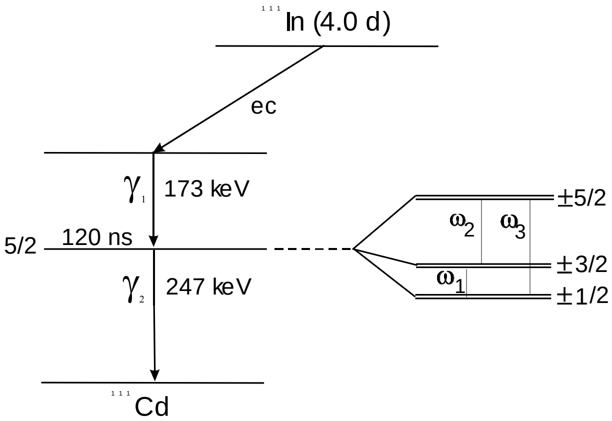


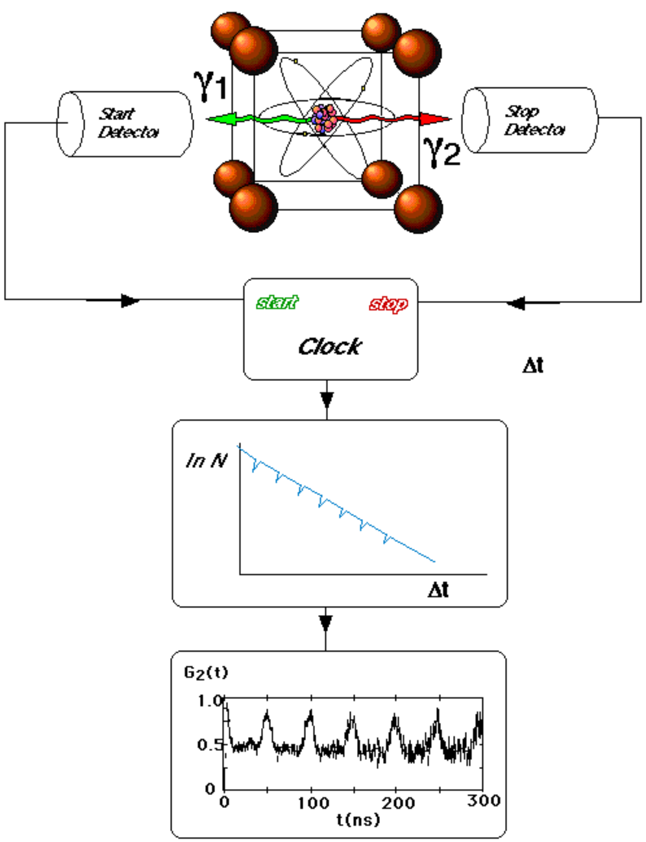
On-Line Diffusion Chamber facility at ISOLDE

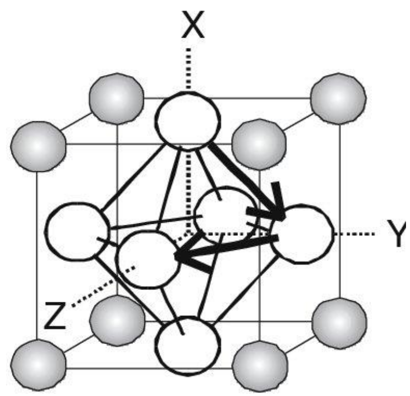


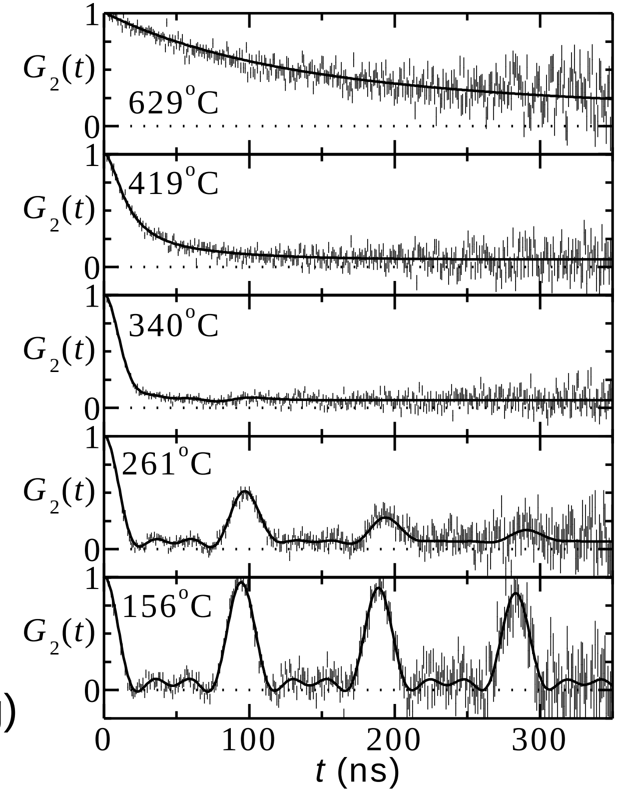
The penetration profile for ^{111m}Cd diffusion in Pd_3Gd at 793 K. The measured relative specific radioactivity was corrected for the short life time of the isotope. Two contributions, namely bulk and grain boundary ones, could be distinguished. The measurements were performed using On-Line Diffusion Chamber at ISOLDE.

Perturbed angular correlation of gamma-rays (PAC)





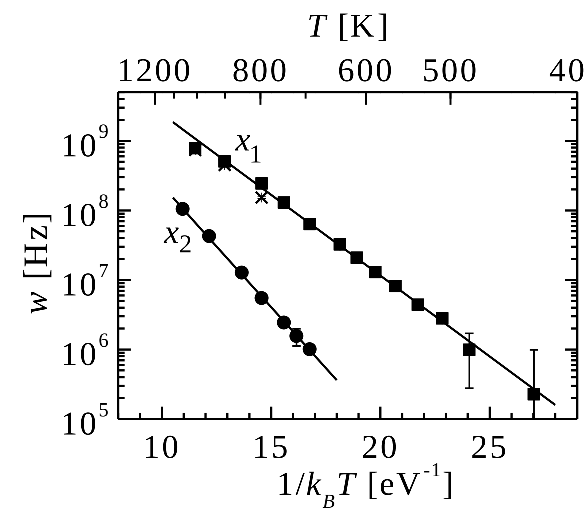




In_3R crystal (Cu_3Au)

Jumps on In-sublattice reorient electric-field gradients by 90°, decoherence (damping) of precessions. Fit to obtain jump-frequency w. Arrhenius plot gives activation enthalpy. Relation to diffusivity:

$$D = \frac{1}{6}\ell^2 fw$$



Crucial issues of the proposal:

Choice of In3R phases:

PAC: "frequency window" (1-10 MHz) translates to ~600-1000°C

Radiotracer: 10^{-20} $m^2/s < D^* < 10^{-14}$ m^2/s

Diffusion mechanism: In-sublattice mechanism for ^{111m}Cd ($L1_2$ structure)

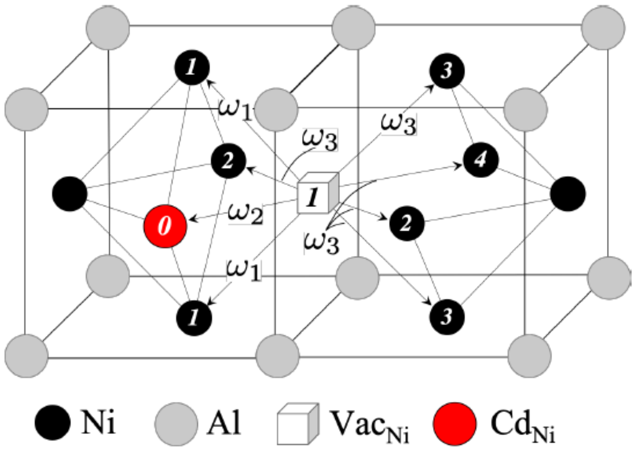
Full characterization of the samples: XRD, SEM (+EBSD analysis), EDS, HR-TEM (including analytical TEM)

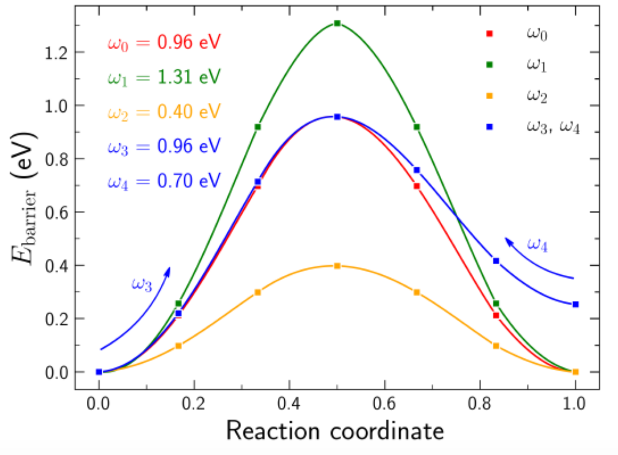
Temperature calibration: via measurements of ^{64}Ni diffusion in pure Ni by SIMS

Follow-on measurements: phases In3Lu, In3La and In3Nd are considered (different ratio of intra- and inter-sublattice jumps)

DFT calculations: state-of-the-art calculations of the jump frequencies and the correlation factor

Correlation factor for Cd in Ni3Al (DFT-informed study)

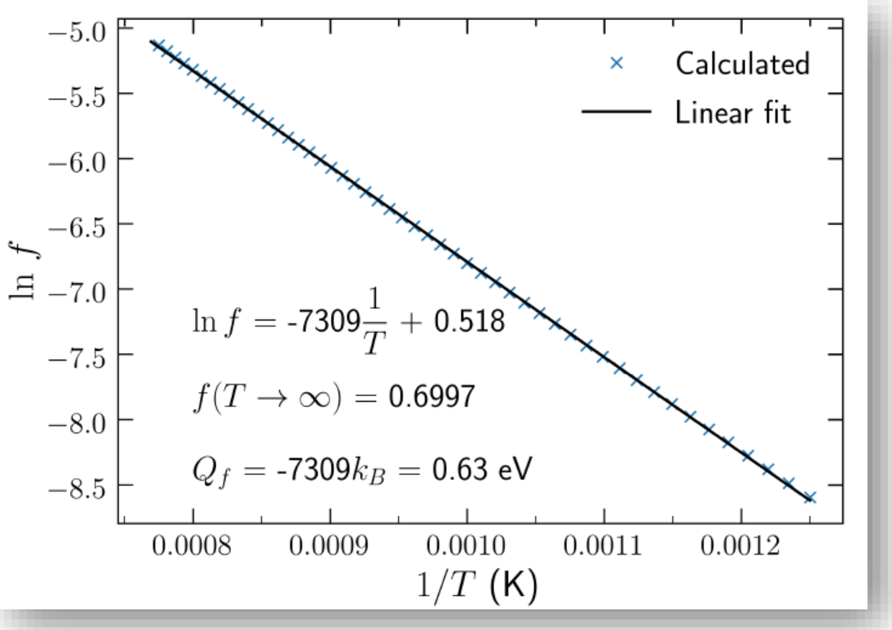




$$f = \frac{H}{2\omega_2 + H}$$

$$H = \frac{10\omega_3 F^z(2\omega_1 + 5\omega_3 F^x)}{2\omega_1 + 5\omega_3(F^x + F^z)}$$

$$w_i = \nu_i \exp(-\Delta H_m / k_B T)$$



Conclusions

Correlated measurements of individual atom jumps (PAC) and of long-range diffusion (radiotracer) will **for the first time** provide an experimental access to the correlation factor of diffusion.

In fact, the correlation factor was never measured so far by any technique.

- For the $L1_2$ A_3B systems, the intra-subblattice (the A-sublattice) jumps of Cd could be followed by PAC and they contribute simultaneously to long-range diffusion of Cd
- The isotope ^{111m}Cd, suitable for both PAC and tracer diffusion, can be purely produced at ISOLDE-CERN;
- Both PAC arrangement and the On-Line Diffusion Chamber facility are available at ISOLDE

DFG Deutsche Forschungsgemeinschaft