Quadrupole Interaction of ^{100}Rh in Rh_3M Alloys

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For d-elements the electric field gradients in metals are still not well understood, partly due to scatter in the experimental data. For such studies the value of the nuclear quadrupole moment Q has to be available in order to extract the efg from the interaction frequencies measured. For the 75keV 2^+ state in ^{100}Rh, used extensively in PAC research, only rough estimates of Q were available to date.

In recent years the calculation of efgs for simple lattices has become rather straightforward with modern ab-initio band structure methods [1]. Thus a measurement of the quadrupole coupling constant for ^{100}Rh in a simple Rh compound, when compared with such a calculation, would allow to determine Q in a reliable way. Since rhodium metal has a cubic site symmetry, a binary alloy would be the simplest matrix. In particular there exist several alloys Rh_3M crystallizing in the Cu_3Au structure that are suitable candidates. The purpose of the present work was to compare theoretical efg calculations for this series of alloys with experimental measurements of the quadrupole coupling by PAC and thus hopefully determine Q.

The theoretical calculations of the efgs were performed using the full potential linearized augmented plane wave method based on density functional theory. The computer code WIEN93 has been demonstrated to yield reliable results for the band structure of various solids, in particular also for efgs [1]. The calculations for the alloys selected were performed at the experimentally known lattice constants. The obtained results are summarized in Table 1.

For the experiments the alloys were prepared by melting Rh (4N) with the corresponding amount of the second metal by an electron beam in vacuum. For better mixing the ingots were remelted several times after turning in the copper heath. Slices of 0.5mm thickness and about 5mm diameter were cut out of the spheres with a low speed diamond saw. The ^{100}Pd isotope was produced in-situ by irradiating the disks with 50 MeV protons via the ^{103}Rh(p,4n)^{100}Pd reaction. The samples were subsequently annealed at about 1600°C in vacuum between two alumina covered tungsten boats for one hour and slowly cooled to room temperature.

The perturbation functions measured in a 4-detector NaI(Tl) setup showed that in all three alloys studied the Rh nuclei see a unique field gradient of close to axial symmetry. From the spectra the nuclear quadrupole coupling constants given in Table 1 were determined. Since an axially symmetric EFG is what one expects for the Rh lattice site, there can be no doubt that the interaction measured is characteristic for ^{100}Rh nuclei at the substitutional site.

It is straightforward to obtain the absolute value of Q from the measured interaction frequencies and the calculated field gradients for the Rh_3M alloys. It is comforting that the extracted values given in Table 1 are very close to each other, with an average of $|Q|=.153b$. An estimate of the error is difficult. The experimental error is neglegible, and the effect of lattice vibrations is still expected very small at room temperature. For the theoretical calculation we rely on the experience gained from other systems and conservatively estimate an uncertainty of 10%.

Our value for Q is a factor of 2 larger than the former qualitative estimation from the quadrupole interaction measured at Rh impurities in various metallic matrices [2].

Table 1: Results for some Rh alloys with Cu_3Au structure.

Alloy	a[Å]	V_{ZZ}[V/Å^2]	e^2QV_{ZZ}/h[MHz]	$Q_{calc}(^{100}Pd)$
Rh_3Ti	3.823	-34.9		
Rh_3Zr	3.926	-43.2	15.75(6)	.151b
Ru_3Hf	3.912	-38.1	14.3(1)	.155b
Rh_3Nb	3.857	-31.2	11.6(1)	.154b

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