

## Nuclear-orientation study of PtTe<sub>2</sub>

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Nuclear-orientation measurements at temperatures down to  $\sim 10$  mK were performed on <sup>199</sup>Au, <sup>191</sup>Pt, and <sup>131</sup>I in PtTe<sub>2</sub>. The electric-field gradients of Au, Pt, and I in PtTe<sub>2</sub> were determined to be  $+5.6(0.6)\times 10^{17}$ ,  $+10(5)\times 10^{17}$ , and  $-3.7(1.7)\times 10^{17}$  V/cm<sup>2</sup>, respectively.

### I. INTRODUCTION

The nuclear-orientation (NO) technique has become a standard technique for the determination of the hyperfine splitting of radioactive nuclei in environments which provide large magnetic and/or electric hyperfine fields. The aim of such experiments may be twofold: (1) measurement of the nuclear moments of the short-lived radioactive isotopes and (2) measurement of magnetic and electric hyperfine fields. For the determination of the magnetic moments of radioisotopes, many experiments have been performed with the ferromagnetic host lattices Fe, Co, Ni, and Gd. In many cases the hyperfine splitting could be measured with the highly precise method of NMR on oriented nuclei (ON) detected via the anisotropy of radiation (NMR-ON). For the measurement of quadrupole moments of radioisotopes, the quadrupole-interaction-tuned (QIT) NMR-ON technique has been developed recently, which, however, is not applicable to all isotopes of current interest.<sup>1</sup> Especially, the high-spin  $\frac{13}{2}^+$  Pt and  $\frac{11}{2}^-$  Au isomers are not easily accessible with this technique at present. For the nonresonant determination of electric quadrupole moments, the technique of quadrupole-interaction nuclear orientation (QI-NO) has been applied successfully for a number of isotopes. (For a review, see Refs. 2 and 3.) Mostly noncubic metal matrices have been used up to now. As the electric-field gradients (EFG's) in pure metals are "only" of the order of magnitude up to several  $10^{17}$  V/cm<sup>2</sup>,  $\gamma$  anisotropies of the order of  $< 10\%$  are obtained at temperatures of  $\sim 10$  mK, limiting the accuracy of QI-NO experiments to  $\approx 10\%$ . Thus (metallic) matrices with larger EFG's would be desirable. In principle, dichalcogenide layer compounds could fulfill this condition. For <sup>182</sup>Ta in 2H-TaS<sub>2</sub>,  $\gamma$  anisotropies of  $\geq 50\%$  have been observed.<sup>4,5</sup> Data on the systematics of EFG's in dichalcogenide layer compounds are, however, not yet available. Here we report on the measurement of the EFG's of Au, Pt, and I in PtTe<sub>2</sub>.

### II. NUCLEAR ORIENTATION

The angular distribution of  $\gamma$  rays emitted in the decay of radioactive nuclei oriented by quadrupole interaction

at low temperatures is most conveniently written as<sup>6</sup>

$$W(\theta) = 1 + \sum_{k=2,4} A_k B_k (h\nu_Q/k_B T) P_k(\cos\theta) Q_k. \quad (1)$$

Here the parameters  $A_k$  depend on the characteristics of the nuclear decay; they are products of the normally used angular correlation coefficients  $U_k$  and  $F_k$ , which, e.g., are tabulated in Ref. 7. The  $P_k(\cos\theta)$  are Legendre polynomials,  $\theta$  being the angle between the quantization axis (here the  $c$  axis of the single crystal) and the direction of observation. The  $Q_k$  are solid-angle correction coefficients; they are normally near unity. The parameters  $B_k$  describe the degree of orientation; they depend on  $\nu_Q/T$ , where  $\nu_Q$  is the usually quoted quadrupole splitting frequency and  $T$  is the temperature of the system.  $\nu_Q$  is usually defined as

$$\nu_Q = e^2 q Q / h, \quad (2)$$

where  $eq$  is the EFG and  $Q$  is the spectroscopic quadrupole moment of the nucleus. In the "high-temperature" region,  $h\nu_Q < k_B T$ , the orientation parameter  $B_4$  is much smaller than  $B_2$ . This means that only the  $k=2$  contribution in Eq. (1) has to be considered. Moreover,  $B_2(h\nu_Q/k_B T)$  can be expanded in powers of  $h\nu_Q/k_B T$ , which yields

$$B_2 = -c_I h\nu_Q / k_B T. \quad (3)$$

Here  $c_I$  is a positive constant depending on the spin  $I$  of the oriented state, which is given by

$$c_I = \left[ \frac{I+1}{80I(2I-1)} \right]^{1/2}. \quad (4)$$

The  $\gamma$  anisotropy is then given by

$$W(\theta) - 1 = -c_I A_2 P_2(\cos\theta) Q_2 h\nu_Q / k_B T. \quad (5)$$

Because the almost linear dependence of the anisotropy on  $1/T$  according to Eq. (5) holds in a remarkably large temperature range, only the product  $A_2 Q_2$  can be determined from QI-NO measurements in general. It is reasonable to measure the angular distribution for  $\theta=0^\circ$  and  $90^\circ$ ,  $W(0^\circ, T)$  and  $W(90^\circ, T)$ , as then the following combination can be analyzed:

TABLE I.  $\gamma$  anisotropies and quadrupole splittings measured at the average temperature of 8.9(5) mK with the annealed and unannealed PtTe<sub>2</sub> samples.

Isotope	$E$ (keV)	$A_2$	$W_2$	$\nu_Q$ (MHz)	$\nu_Q^{\text{av}}$ (MHz)	$W_2$	$\nu_Q$ (MHz)	$\nu_Q^{\text{av}}$ (MHz)
<sup>199</sup> Au	158	-0.396	+0.037(1)	annealed +70(6)		+0.020(1)	unannealed +40(6)	
	208	+0.763	-0.078(2)	+77(6)	+74(5)	-0.046(2)	+47(6)	+44(5)
<sup>131</sup> I	284	-0.470	+0.012(7)	+31(20)		+0.011(7)	+28(22)	
	636	-0.420	+0.018(9)	+45(29)	+36(16)	+0.026(10)	+74(35)	+41(21)
<sup>191</sup> Pt	538	+0.422	+0.088(21)	-155(43)		+0.054(14)	-97(33)	

$$W_2(T) = \frac{3W(0^\circ) - 8W(90^\circ) + 5}{7} = A_2 B_2(T) Q_2, \quad (6)$$

from which  $A_2 \nu_Q$  can be determined completely unambiguously. Up to now we have tacitly assumed that 100% of the impurity nuclei are subject to one unique hyperfine interaction. Often, the assumption is made that a fraction  $f$  is subject to the full undisturbed hyperfine interaction, while the fraction  $1-f$  is subject to a negligibly small, i.e., zero, hyperfine interaction. It is obvious that  $f$  has to be known for the interpretation of QI-NO experiments. This means that  $A_2$  in the above formulas has to be replaced by  $fA_2$  and that only the product  $fA_2 eQV_{zz}$  can be determined actually.

### III. EXPERIMENTAL DETAILS

Single crystals of PtTe<sub>2</sub> were grown with the technique of chemical vapor transport. Combinations of platinum, phosphorus, sulfur, and tellurium in the molar ratios 1:1:3:2 were used as the reactants. (For details, see Ref. 8.) Single crystals with an area of 7–17 mm<sup>2</sup> and a thickness of  $\sim 10$   $\mu\text{m}$  were obtained, the  $c$  axis always being perpendicular to the crystal surface. Two crystals were irradiated at the reactor in Munich, FRM, for 16 h in a neutron flux of  $2 \times 10^{13}$  neutrons/(cm<sup>2</sup> s), to produce <sup>199</sup>Au ( $I^\pi = \frac{3}{2}^+$ ,  $T_{1/2} = 3.1$  d) as the decay product of 30 m <sup>199</sup>Pt, <sup>191</sup>Pt ( $I^\pi = \frac{3}{2}^-$ ,  $T_{1/2} = 2.8$  d) and <sup>131</sup>I ( $I^\pi = \frac{7}{2}^+$ ,  $T_{1/2} = 8.0$  d) as the decay product of 25 m <sup>131</sup>Te. After the irradiation one of the crystals was annealed for 20 h at 500°C and continuously cooled to room temperature. It was soldered with GaIn to one side of the copper cold finger of a demagnetization cryostat.<sup>9</sup> For thermometry a <sup>60</sup>CoCo(hcp) thermometer was soldered to the other side of the cold finger. After cooling down to 8.5(1) mK, a magnetic field of 2 kG was applied to keep the solder normal conducting. The  $\gamma$  rays were detected with two Ge(Li) detectors placed at 0° and 90° with respect to the  $c$  axis of the PtTe<sub>2</sub> single crystal. After a measurement time of 16 h at the average temperature of 8.9(5) mK, the cryostat was warmed up and normalization spectra were taken for 22 h at 1.5 K. Similar measurements were performed with the second crystal, which had not been annealed after the neutron irradiation, in order to get information on the radiation damage.

### IV. RESULTS AND DISCUSSION

Figure 1 shows the  $\gamma$  anisotropy  $W_2$  for the 158-keV transition of <sup>199</sup>Au. The solid line is the result of a least-squares fit according to Eq. (6), demonstrating the almost linear behavior of  $W_2$  on  $1/T$ . Because of this nearly complete linearity, we did not measure additional anisotropies at higher temperatures, as the obtainable accuracy would not have been improved. Our results of the measurements on the annealed and unannealed samples are compiled in Table I. The ratios of quadrupole splittings for the annealed to unannealed samples are 1.7(2), 1.6(7), and 0.9(6) for Au, Pt, and I, respectively, with an average value 1.6(2). At present, we cannot state whether this ratio is different for the Pt and Te sites. However, a similar ratio has been observed for <sup>182</sup>Ta in 2H-TaS<sub>2</sub>. In this case it has been shown that the lattice damage introduced by the neutron irradiation could be healed completely by a proper annealing step.<sup>4,5</sup> Thus we conclude that the results for our annealed sample can be interpreted with the assumption that all impurity nuclei of one species are subject to the same undisturbed EFG. Taking the quadrupole moments as  $Q(^{199}\text{Au}) = +0.55(3)$  b,<sup>10</sup>  $Q(^{191}\text{Pt}) = -0.64(26)$  b,<sup>11</sup> and  $Q(^{131}\text{I}) = -0.40(1)$  b,<sup>12</sup> the field gradients are deduced to be, for Au in PtTe<sub>2</sub>,

$$eq = +5.6(6) \times 10^{17} \text{ V/cm}^2,$$

for Pt in PtTe<sub>2</sub>,

$$eq = +10(5) \times 10^{17} \text{ V/cm}^2,$$

and for I in PtTe<sub>2</sub>,

$$eq = -3.7(1.7) \times 10^{17} \text{ V/cm}^2,$$

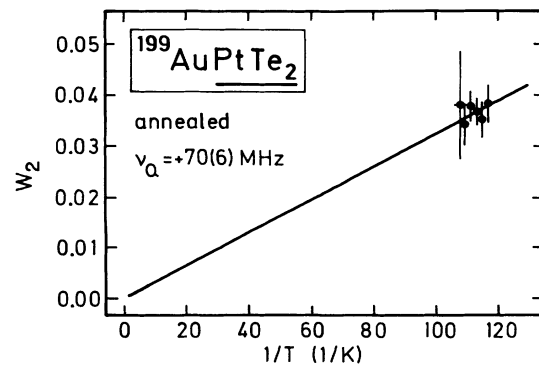


FIG. 1.  $\gamma$  anisotropy of the 158-keV transition of <sup>199</sup>Au.

which are smaller in magnitude, as could have been expected according to data on other layered compounds [5]. The relatively large error for the field gradient of Pt in PtTe<sub>2</sub> is mainly due to the uncertainty of the quadrupole moment. In this case no stable  $I > \frac{1}{2}$  isotope exists for which the quadrupole moment could be determined with muonic x-ray techniques. Thus it is impossible to calibrate the EFG of Pt in any matrix. The quadrupole moment of <sup>191</sup>Pt has been determined by a measurement of the quadrupole splitting of <sup>191</sup>Pt in hcp Os and by estimating the EFG of PtOs from the experimental systematics of a series of 5d elements in Os. Recently, laser spectroscopy measurements on several Pt isotopes have been reported.<sup>13</sup> These authors quote  $Q(^{191}\text{Pt}) = -0.98(5)$  b, however, *without* Sternheimer correction, the calculation of which seems to be difficult, and by which the "actual" quadrupole moment could possibly be reduced by up to  $\approx 30\%$ .<sup>13</sup> Hilberath *et al.*<sup>14</sup> quote  $Q_S^{\text{corr}}(^{191}\text{Pt}) = -0.87(4)$  b, applying a Sternheimer correction  $R = -0.1$ . To resolve this uncertainty, it might be possible that, by precise measurements of the EFG's of a series of 5d elements in PtTe<sub>2</sub>, the EFG of Pt

in PtTe<sub>2</sub> can be estimated to a better precision from the functional dependence of the EFG on the impurity charge. Together with such systematic measurements in other matrices, it might be possible to derive quadrupole moments with better precision, which would be of current interest for the interpretation of the absolute values of the nuclear deformation of neutron-deficient Pt isotopes via quadrupole moments.

Although the present accuracy is limited for Pt and I, the sign determination is unambiguous. Our data suggest that the EFG is positive for impurities at the Pt site and negative for impurities at the Te site. In 2H-TaS<sub>2</sub> the sign of the EFG at the Ta site is negative, i.e., opposite to the present case. No conclusions on this fact can be drawn at present because of the lack of additional data.

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