Electric field gradient of Ag in Zn

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(Received 12 October 1982)

The electric quadrupole interaction of ^{110}Ag^m implanted in a zinc single crystal has been studied by means of low-temperature nuclear orientation. An electric-field-gradient value of $+1.71(11) \times 10^{17}$ V/cm^2 is deduced. This measurement provides a first inclusion of a one-valent probe into the systematics.

The electric field gradient (EFG) at a probe nucleus in a noncubic metallic lattice can be written as[1]

$$V_{zz} = V_{zz}^{\text{latt}}(1 - \gamma_\infty) + V_{zz}^{\text{el}} \ . \tag{1}$$

V_{zz}^{latt} is the contribution to the EFG from the metal ions, γ_∞ is the Sternheimer antishielding factor, and V_{zz}^{el} arises from the conduction electrons. As the ionic part of the total EFG, i.e., $V_{zz}^{\text{ion}} = V_{zz}^{\text{latt}}(1 - \gamma_\infty)$, is well understood and the experimental V_{zz} value is measured, the conduction-electron part of the EFG can be calculated.

Some empirical relations have been proposed between the different components which are believed to determine the EFG. Besides the "universal" correlation of Raghavan,[2] other relations have been suggested, such as the correlation between the electronic enhancement factor $K = -V_{zz}^{\text{el}}/V_{zz}^{\text{ion}}$ and the impurity valence Z,[3] and the empirical fit between $\kappa = V_{zz}/V_{zz}^{\text{latt}}$ and the temperature T.[4] However, in a Zn host none of these relations has been tested for a one-valent probe. Silver, with only one s electron outside closed shells, becomes therefore an interesting impurity for electric-quadrupole-interaction studies; hence this experiment completes the systematics.

The method of nuclear orientation enables one to determine both sign and magnitude of the quadrupole hyperfine interaction parameter QV_{zz}, in contrast to other hyperfine techniques which normally only measure the strength of the quadrupole interaction. If the sign and the magnitude of the nuclear quadrupole moment Q are known the experimental V_{zz} value follows in a straightforward way. The knowledge of the sign of V_{zz} is an absolute necessity to separate the ionic and electronic contributions to the EFG.

In our experiments the data points on several γ transitions were fitted to $\epsilon = W(0°,T)/W(90°,T)$ with

$$W(\theta,T) = 1 + \alpha \sum_k Q_k U_k F_k B_k(T,QV_{zz},I)P_k(\cos\theta) \ , \tag{2}$$

the radiation distribution function assuming an axially symmetric quadrupole interaction. The Q_k are solid angle corrections and were taken to be 1. The U_k and F_k are the radiation parameters of the unobserved, observed transitions, respectively, and the P_k are the Legendre polynomials. The B_k orientation parameters depend on the temperature, the quadrupole interaction parameter QV_{zz}, and the nuclear spin I. For γ radiation, k is even and, in practice, only the $k = 2,4$ terms are taken into account. The factor α indicates the substitutional fraction of implanted ions in the host matrix.

The ^{110}Ag^m ($t_{1/2} = 250$ d) activity was produced by neutron activation of ^{109}Ag in the Br2 reactor at Mol (Belgium). It was implanted at 85 keV in a Zn single crystal up to a dose of 5×10^{13} atoms/cm^2 at the Leuven Isotope Separator. The crystal was soldered with Woods metal to the copper cold finger of a ^{3}He-^{4}He dilution refrigerator[5] together with a ^{57}CoFe thermometer. The base temperature of the cryostat is 8 mK. An external magnetic field of 0.2 T was applied to polarize the thermometer. We placed one detector at 0° and one at 90° with respect to the c axis of the single crystal. Single γ spectra were recorded for fixed time intervals at different temperatures.

As an angular correlation measurement on Zn with the known ^{111}Ag as probe ion indicated a fully substitutional implantation,[2] we took α to be 1. The $U_k F_k$ parameters are taken from the experiment done by Johnston and Stone.[6] The quadrupole moment of the ground state of ^{110}Ag^m has been measured as $+1.65(10)$ b.[7]

Table I gives the energy, the intensity, the experimental $U_k F_k$ coefficients,[6] and the fitted QV_{zz} value for eight γ transitions in the decay of ^{110}Ag^m. The listed quadrupole interaction parameter values for the different γ rays yield a weighted mean value of $+2.78(8) \times 10^{-7}$ V. Taking into account the error on the quadrupole moment we derive an EFG value of $+1.71(11) \times 10^{17}$ V/cm^2 for Ag in Zn. As an example, the plot for the 884-keV γ ray is displayed in Fig. 1. The error bars on the figure are purely statistical.

If one assumes that the two outer s electrons of Zn occupy the conduction electron band, the point-charge contribution to V_{zz}^{latt} for the hcp Zn^{2+} ions can be calculated using the formula given by Das and

TABLE I. Different γ energies in the decay of ^{110}Ag^m used to determine the electric field gradient in Zn. The $U_k F_k$ values were taken from Ref. 6. A weighted mean value of $+2.78(8) \times 10^{-7}$ V includes only the statistical errors on the data.

γ energy (keV)	Relative intensity	$U_2 F_2$	$U_4 F_4$	$\dfrac{QV_{zz}}{(10^{-7} \text{V})}$
657	100	−0.320(10)	−0.100(50)	+2.84(36)
707	17	+0.965(10)	+0.114(30)	+3.04(30)
764	24	−0.395(5)	−0.195(20)	+3.00(55)
818	75	+0.181(10)	+0.015(20)	+4.49(109)
884	75	−0.361(5)	−0.119(30)	+2.99(15)
937	36	−0.365(5)	−0.143(20)	+2.30(21)
1384	28	+0.900(10)	+0.076(20)	+2.54(12)
1505	14	+0.795(10)	+0.175(30)	+2.99(20)
				+2.78(8)

Pomerantz[8] as

$$V_{zz}^{\text{latt}}(\text{point}) = \frac{Ze}{a^3}\left[0.0065 - 4.3584\left(\frac{c}{a} - 1.633\right)\right] ,$$

(3)

where c and a are the lattice parameters of Zn, and Z is the charge of the lattice ions. The values at 0 K of these parameters can be extrapolated from the known room-temperature values by considering the experimental temperature dependence of the linear expansion coefficients along the c and a axes.[9] The effect of quadrupole polarization of the host ion by its core electrons on the V_{zz}^{latt} (point) value is smaller than 1% and can be neglected.[10−12]

The empirical relations are based on data points taken at, or normalized to, room temperature. This makes it necessary to normalize our EFG value measured at 0 K towards 300 K. The temperature dependence of the EFG for AgZn can be estimated using the empirical $T^{3/2}$ law: $V_{zz}(T) = V_{zz}(0)(1 - BT^{3/2})$.[13]
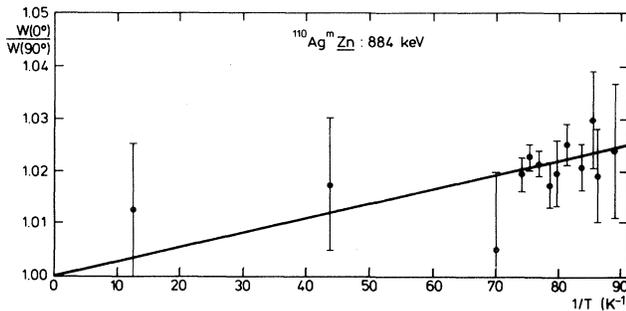
FIG. 1. Anisotropy of the 884-KeV γ ray in the decay of ^{110}Ag^m. The error bars are purely statistical.

As a one-valent probe in a Zn matrix has not yet been measured, the slope B cannot be obtained with the method described in Ref. 14. As for seven different impurities measured in Zn, the rate $V_{zz}(300$ K$)/V_{zz}(0$ K$)$ varies between 0.92 and 0.95 (the only exception is F with a rate of 0.82),[1,15] we assume the same limiting values of $V_{zz}(300$ K$)/V_{zz}(0$ K$)$ for the AgZn system. We then obtain an EFG value varying between $+1.57(10) \times 10^{17}$ V/cm^2 and $+1.62(10) \times 10^{17}$ V/cm^2.

Another possibility to determine $V_{zz}(300$ K$)$ is offered by the relation found by Krien et al.[4] These authors define an electronic enhancement factor $\kappa(T) = V_{zz}(T)/V_{zz}^{\text{latt}}(T)$ normalized to room temperature. Empirically they fit a straight line through the reduced enhancement factors for different impurity-Zn systems plotted as a function of temperature. A ratio $\kappa(0$ K$)/\kappa(300$ K$)$ of 1.18 can be found from the plot. This, together with our $V_{zz}(0$ K$)$, and the calculated $V_{zz}^{\text{latt}}(0$ K$)$ and $V_{zz}^{\text{latt}}(300$ K$)$ values, gives

$$V_{zz}(300 \text{ K}) = +1.63 \times 10^{17} \text{ V/cm}^2 ,$$

which correponds with the previous result.

The only factor left in (1), necessary to calculate the electronic contribution of the EFG, is the Sternheimer antishielding factor $\gamma_\infty(\text{Ag}^+)$. This has been computed by several authors. The calculations of Sternheimer[16] and of Feiock and Johnson[17] take into account the interaction of the external field gradient with the impurity electronic states, i.e., a zeroth-order interaction, respectively, without and with relativistic corrections. Their values, $[\gamma_\infty(\text{Ag}^+)]_{\text{St}} = -34.9$ and $[\gamma_\infty(\text{Ag}^+)]_{\text{FJ}} = -35.8$, clearly show that the difference due to relativistic corrections is not large for silver. Schmidt et al.[18]

TABLE II. Values of different parts in $V_{zz} = V_{zz}^{latt}(1 - \gamma_\infty) + V_{zz}^{el}$ tabulated for 0 and 300 K, using two different values for γ_∞ (Refs. 17 and 18). As no errors on V_{zz}^{latt} and γ_∞ are available, we did not quote an error on V_{zz}^{el} and K. For V_{zz} (300 K) we took $(1 - BT^{3/2}) = 0.95$, as this value corresponds with the result, we found using the empirical relation found by Krien et al. (Ref. 4).

T (K)	V_{zz} (10^{17} V/cm^2)	V_{zz}^{latt} (10^{17} V/cm^2)	$\gamma_\infty(Ag^+)$	V_{zz}^{ion} (10^{17} V/cm^2)	V_{zz}^{el} (10^{17} V/cm^2)	K
0	+1.71(11)	−0.131	−43.14	−5.78	+7.49	1.30
			−35.82	−4.82	+6.53	1.35
300	+1.62(10)	−0.146	−43.14	−6.44	+8.06	1.25
			−35.82	−5.38	+7.00	1.30

compute, besides the zeroth-order effect (which is in good agreement with Ref. 16), the contribution to the antishielding factor from the interactions between the electronic shells, however, without including relativistic corrections. Their result for Ag$^+$, i.e., $[\gamma_\infty(Ag^+)]_{SSDW} = -43.14$, significantly demonstrates the importance of this first-order contribution. The relevant parts of (1) calculated with the Sternheimer factors of Refs. 17 and 18 are listed in Table II. The electronic contribution to the effective-field gradient in the AgZn system is larger and, as expected, opposite in sign to V_{zz}^{ion}.

In the original paper on the "universal" correlation Raghavan et al. define an electronic enhancement

factor $K = -V_{zz}^{el}/V_{zz}^{ion}$.[2] In Fig. 2 we plotted our result (for the different γ_∞ values used) in the modified "universal" correlation of Raghavan et al.[19] Although with this first measurement of a one-valent probe one cannot prove the existence of a nominal valence $Z = 1$ line, Fig. 2 shows our value to be in agreement with the proposed systematics in Ref. 19. Soares et al.[3] already concluded in their analysis that K is almost constant for a given impurity in different hosts and depends strongly on the valence Z of the impurity atom. Now a one-valent probe is added to their plot in Fig. 3 and, as can be seen, the K value for silver in zinc fits excellently in this picture.

Theoretically, the empirically established systematics cannot be explained. Microscopical calculations for impurity-host systems would be very interesting but are too complicated; up to now, calculations of the EFG in metals have been carried out only for pure systems.[1,20,21]
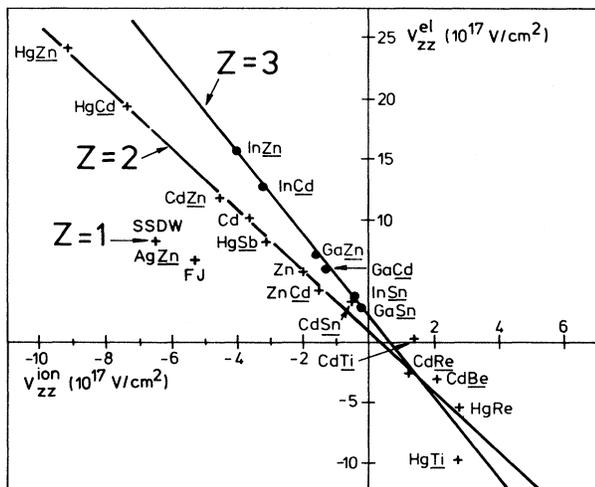
FIG. 2. Plot of V_{zz}^{ion} vs V_{zz}^{el} (Ref. 19). The points with indices FJ and SSDW indicate the data we found using, respectively, $[\gamma_\infty(Ag^+)]_{FJ}$ (Ref. 17) and $[\gamma_\infty(Ag^+)]_{SSDW}$ (Ref. 18). Our data point possibly shows the existence of a $Z = 1$ line.
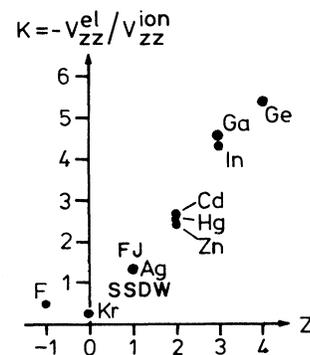
FIG. 3. Plot of the electronic enhancement factor K vs the impurity valence Z for different impurities in a Zn matrix.[3] The points with indices FJ and SSDW indicate the data we found using, respectively, $[\gamma_\infty(Ag^+)]_{FJ}$ (Ref. 17) and $[\gamma_\infty(Ag^+)]_{SSDW}$ (Ref. 18). On this plot these data points cannot be resolved.

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