Temperature dependence of the electric field gradient of indium in cadmium

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(Received 21 December 1973)

The temperature dependence of the electric field gradient for indium as an impurity in cadmium metal has been measured using the time-differential γ-γ angular correlation technique. The results, combined with previous work on the In-Cd system, strongly support the assumption that the electric field gradient for this system is determined by the host lattice.

INTRODUCTION

Electric field gradients (EFG's) in metals are very sensitive to the total electron distribution in the surroundings of the probe atom and not, as for example in the case of macroscopic transport properties and Knight shift, just those at the Fermi surface. They should, therefore, provide a good test of the theoretical electronic wave functions in metals. Unfortunately, the EFG is a product of several contributing factors which are difficult to separate. Among these are the very often unknown nuclear electric quadrupole moment, bonding effects, Sternheimer factor in solids, as well as the conduction-electron distribution. One can hope that by studying the systematics of EFG's a clearer understanding will emerge. It is with this expectation that the following work was undertaken.

EXPERIMENTAL

The samples of cadmium metal were produced by the reduction, under hydrogen, of 97.2% isotropically enriched ^{116}CdO[1] in a quartz tube. The chemical impurities are stated to be less than 0.2%. After preparation, samples were sealed in the quartz tube in a residual atmosphere of hydrogen gas.

Three different samples were prepared and used interchangeably for the measurements. Samples were irradiated for 20 min in a thermal neutron flux of 1.2×10^{12} neutrons/cm^2 sec.

The nuclear level of interest is that at 659 keV in ^{117}In with $t_{1/2}=53.5$ nsec, $I=\frac{3}{2}$. It was studied via the 90–345-keV γ-γ cascade. It is reached after the β^- decay of ^{117}Cd ($T_{1/2}=2.5$ h) which is produced by the thermal neutron reaction $^{116}\mathrm{Cd}(n,\gamma)^{117}\mathrm{Cd}$.[2]

Since the theory and practice of time-differential perturbed γ-γ angular correlations (DPAC) have been well described elsewhere[3] we will restrict ourselves to only the details of our specific case. The apparatus consisted of a conventional "fast-slow" system with two NaI (Tl) detectors at a fixed angle of 180°. For the 90-keV photon we used a 0.5×1.5-in.-diam crystal and for the 345-keV photon, a 1.5×1.5-in.-diam crystal.

The DPAC function for this case is[3]

$$W(\theta,t)=[1+A_{22}G_2(t)P_2(\cos\theta)]e^{-t/\tau},$$

which with $\theta=\pi$ reduces to $[1+A_{22}G_2(t)]e^{-t/\tau}$, where τ is the lifetime of the intermediate state (in this case 77 nsec), A_{22} is the angular correlation coefficient, and $G_2(t)$ is the perturbation factor. For the case of a pure axially symmetric, randomly oriented EFG and nuclear spin $I=\frac{3}{2}$, the perturbation factor $G_2(t)$ is given by

$$G_2(t)=\tfrac{1}{5}(1+4\cos\omega_0 t)\quad,$$

where $\omega_0=3e^2Qq/2I(2I-1)\hbar$ and eq is the EFG at the nucleus and the quadrupole coupling constant $e^2Qq/h=\omega_0/\pi$.

The experiment was performed to determine e^2Qq/h for various temperatures. The data analysis was carried out as follows: From the raw $W(\theta,t)_{\mathrm{expt}}$ data from the multichannel analyzer, a constant background due to the accidental coincidences was subtracted to give, roughly, a straight line with slope $1/\tau$ on a logarithmic plot. The data were then divided by the function $I_0e^{-t/\tau}$ giving the experimental $A_{22}G_2(t)_{\mathrm{expt}}$. This was then least-squares fitted to a function of the form

$$A_{22}G_2(t)_{\mathrm{expt}}=\tfrac{1}{5}a[1+4\cos\omega_0(t-t_0)]+b,$$

with a, ω_0, t_0, and b as parameters.

For the low-temperature experiments a glass Dewar vessel was used and the sample was immersed in the cryogenic fluid. For the experiments above room temperature, a double-walled, Dewar-type furnace was constructed. On the inner glass wall a Nichrome heating wire was wound whose electrical power came from a constant-current power supply. The samples were attached with wire to one of a set of seven normal thermometers, covering the range of (0–350)°C, which was inserted into the furnace. The source-to-detector distance was 3.5 cm for both detectors in all experiments.

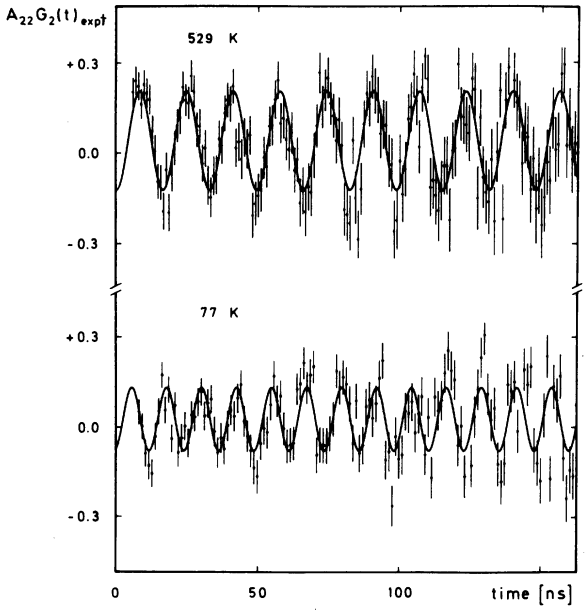

FIG. 1. Typical time spectra analyzed as described in the text to obtain $A_{22}G_2(t)_{expt}$.

RESULTS

Some typical time spectra are shown in Fig. 1. In this obviously favorable case it is possible to obtain a value of e^2Qq/h with 1% accuracy in about 2-h measuring time.

In Table I the results for all our measurements are summarized. The errors given include those from the uncertainty in the least-squares fit and systematic errors in the time calibration and are understood to be standard deviations. The room-temperature result is in very good agreement with previous measurements of Haas and Shirley[4] and of Raghavan and Raghavan.[5] The quadrupole coupling constant extrapolated to 0 °K is found to be $e^2Qq/h = 163.3$ MHz.

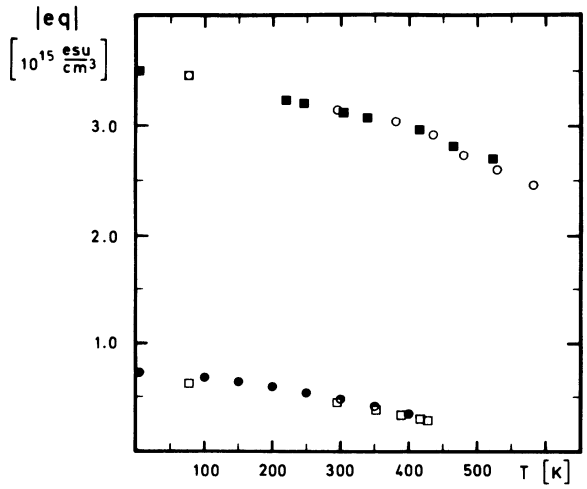

FIG. 2. Electric field gradients in the In-Cd system under the assumption $eq(Cd^{111}Cd, 80\ °K) \equiv eq(Cd^{117}In, 77\ °K)$. This gives $Q(^{111}Cd) = 0.54$ b. □, $In^{111}Cd$, Ref. 6; ●, $In^{115}In$, Ref. 16; ■, $Cd^{111}Cd$, Ref. 10; and ○, $Cd^{117}In$, this work.

DISCUSSION

The following discussion will be, of necessity, qualitative, as a complete discussion of the various systems can only be carried out in the light of a band-structure calculation. As the problem of EFG's at impurities in metals has not been treated extensively, we hope that our discussion may stimulate some efforts in that direction.

In order to compare the EFG's in the four systems *Cd*In, *Cd*Cd, *In*In, *In*Cd one needs to know the quadrupole moments of the various nuclei. Since that of ^{111}Cd has not been experimentally determined, we have chosen to follow the assumptions of Brandt and Rosenblum[6] and Bodenstedt *et al.*[7] that the EFG's depend only on the host lattice.[8] We have normalized the data by assuming that the EFG for In in Cd is equal to the EFG for Cd in Cd at 77 °K. This gives $|Q(^{111*}Cd)| = 0.541$ b. With this value and the known quadrupole moments of $Q(^{115g}In) = +0.863$ b[9] and $|Q(^{117*}In)| = 0.64$ b[5] we obtain the temperature dependence for the four cases, as shown in Fig. 2.

This argument can be carried even further in regard to the sign of the EFG as a result of recent experimental and theoretical work. For Cd metal the sign of the EFG has been determined to be positive from both the theoretical[11] and experimental side.[12]

For In the situation is not quite so clear. From arguments about the systematics of the EFG in In alloys, Thatcher and Hewitt[13] conclude $eq < 0$. However a circular-polarization–directional-correlation measurement by Behrend and Budnick[14] gives $eq > 0$ under the assumption that $Q(^{111*}Cd) > 0$.[5] It would be very useful if this difference could be resolved since this would allow determination of the sign of other quadrupole moments. Because of this discrepancy we have only plotted $|eq|$ for Cd and In.

TABLE I. Quadrupole coupling constants of ^{117}In in cadmium.

Temperature (°K)	e^2qQ/h (MHz)
77	160.5(19)
294(3)	146.0(15)
380(3)	140.9(17)
435(3)	135.3(19)
480(3)	126.6(16)
529(3)	120.5(15)
582(3)	114.1(13)

From this figure it is clear that the assumption that the EFG is characteristic of the host lattice is strongly confirmed. For both the In[6,16] and Cd[17] lattice, the EFG arising from the ion cores is much too small to explain the measured quadrupole interaction. This is a clear indication that the conduction-electron component is the determinative one. This has been confirmed in the case of Cd in a recent pseudopotential calculation by Mohapatra, Singal, and Das[11] in which they found that the electronic contribution is 3.5 times larger than the ionic contribution.

ACKNOWLEDGMENTS

We would like to thank the staff at the Forschungs- und Messreaktor der Physikalisch-Technischen Bundesanstalt in Braunschweig for their help with the irradiations. We are also grateful for the interest of Dr. E. Recknagel in this work and for financial support from the Bundesministerium für Forschung und Technologie.

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