

Temperature dependence of the hyperfine interactions of ^{111}Cd in germanium

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^{111}In was implanted into intrinsic germanium. After removal of the radiation damage by thermal annealing of the samples the hyperfine interaction of the daughter nucleus ^{111}Cd was studied as a function of temperature in the range 20–300 K by use of the time-differential perturbed angular correlation technique. While at room temperature all the probes are in sites of nearly cubic symmetry, at low temperature almost all of them are subject to hyperfine interactions. These interactions can be understood in terms of a fluctuating electric field gradient. These fluctuations can be described with the Abragam and Pound model. The attenuation parameter depends linearly on $1/T$. The present results are compared with those reported for similar experiments carried out in silicon.

I. INTRODUCTION

In recent years the system ^{111}In implanted in Si has been intensively studied¹ by means of the time-differential perturbed angular correlation (TDPAC) technique. Initially interest was centered in the lattice defects produced by the implantation and their subsequent annealing behavior. No electric field gradient (EFG) could be expected in the lattice sites of a perfect diamond structure, so it was possible to monitor the presence of defects or lattice distortion through the electric quadrupole interaction on the radioactive probes. Recently a new aspect has received attention: TDPAC spectra taken after removal of the radiation damage depend strongly on temperature. Probe atoms which are not perturbed at room temperature feel the influence of extranuclear fields at low temperature.^{1–3} This fact has been attributed to “after effects” of the electron-capture decay of ^{111}In to ^{111}Cd by Deicher *et al.*² On the other hand, Kemerink *et al.*³ concluded that aftereffects are important only below 10 K and explain their results (for high-resistivity silicon) in terms of a charge state Cd^- and Jahn-Teller distortions. We have reported¹ a very well-defined temperature dependence of the hyperfine parameters of ^{111}Cd in intrinsic silicon and we have tentatively assigned the static quadrupole interactions to the presence of electronic holes, more or less bound to the radioactive acceptor ion. Holes in the valence band are produced by the electron-capture decay of ^{111}In .

Now, in order to obtain a broader base for a conclusive explanation of this phenomenon we have extended this kind of study to indium implanted in germanium. As is well known, Ge and Si, both with diamond structure, have qualitatively similar physical properties. But concerning indium implantation in both materials, there is a striking difference: no significant indium interstitial component is produced in germanium while in silicon this component can be as high in concentration as the substi-

tutional one.⁴ This different behavior has been recently associated⁵ with the fact that indium is a deep impurity in silicon but a shallow impurity in germanium. In the experiments reported in this paper only hyperfine interactions that can be assigned to impurities in substitutional sites were observed. We give a model to explain the measured interactions and discuss their differences from those reported for silicon.

II. EXPERIMENT

A. Sample preparation

The samples were prepared by implanting 80-keV ^{111}In ions into Ge polycrystals (99.9999% purity, provided by Ventron (GmbH) to a dose of 10^{13} ions/cm². The projected range of the ions was roughly 25 nm with a depth distribution of 10 nm,⁴ leading to a concentration of impurities of about 10^{19} atoms/cm³.

After implantation the samples were encapsulated in quartz tubes and annealed in a vacuum ($p \leq 10^{-5}$ kPa). We have used a two-step annealing process like the one recommended for silicon, but at lower temperatures on account of the lower melting point of Ge: 450°C for 30 min followed by 650°C for 30 min. There was a high loss of activity ($\sim 95\%$).

B. Technique

The hyperfine interactions of ^{111}Cd were observed by means of the TDPAC technique. The decay of the excited level of ^{111}Cd , populated by the electron-capture decay of ^{111}In , occurs by a γ - γ cascade which is very appropriate for TDPAC experiments. The angular correlation of these rays is perturbed if there are external fields acting on the intermediate nuclear level. These fields appear in the perturbation factor $G_2(t)$, where t is the time spent by the nucleus in the intermediate level. We have deter-

mined this perturbation factor using a standard four-NaI(Tl) detector system⁶ with the detectors arranged in a plane at 90° angular intervals. Eight coincidence spectra between both γ rays, $C_{ij}(t)$, were simultaneously recorded. $C_{ij}(t)$ is the number of events in which the first γ ray enters detector i and after a time t the second enters detector j . After correction for accidental coincidences the eight spectra were combined

$$C(90^\circ, t) = [C_{12}(t) + C_{21}(t)]^{1/2} [C_{34}(t) + C_{43}(t)]^{1/2} \quad (1)$$

and

$$C(180^\circ, t) = [C_{13}(t) + C_{31}(t)]^{1/2} [C_{24}(t) + C_{42}(t)]^{1/2}$$

and from these spectra the following ratio was obtained:

$$R(t) = \frac{C(180^\circ, t) - C(90^\circ, t)}{2C(90^\circ, t) + C(180^\circ, t)} \quad (2)$$

This is related to the perturbation factor $G_2(t)$, which will be described below by

$$R(t) = A_2 G_2(t) \quad (3)$$

The factor A_2 depends on the geometry of the source detectors involved and on the anisotropy of the γ cascade.

C. Expressions for the perturbation factor

The form of the perturbation factor $G_2(t)$ that is relevant for the content of this article will be described here. In order to interpret the observed spectra we will consider the situation in which the EFG jumps randomly among axes X , Y , and Z . These fluctuations occur in the case of a dynamic Jahn-Teller effect or in the case of jumps of a defect in the neighborhood of the probe atom. Unfortunately, in these cases there is no analytic expression for the perturbation factor, and numerical techniques^{7,8} are necessary to compute $G_2(t)$. But in the case of fast-relaxation processes, when the time of correlation τ_c is very short, $G_2(t)$ is given by the Abragam and Pound limit:⁹

$$G_2(t) = \exp(-\lambda t) \quad (4)$$

where λ is proportional to $\langle \omega^2 \rangle \tau_c$, ω being the average interaction frequency.

III. RESULTS

Figure 1 shows a TDPAC spectrum taken after implantation. This spectrum is like those reported for indium implantation in silicon.¹ It corresponds to a wide distribution of quadrupole frequencies arising from radiation damage and surface effects. Figure 2(a) shows a TDPAC spectrum obtained at room temperature after annealing. All the probe atoms occupy sites of almost cubic symmetry. Figure 2(b) shows a spectrum obtained at 20 K. As can be seen, lowering the measurement temperature gives rise to a strong effect. The solid lines are fits of theoretical functions

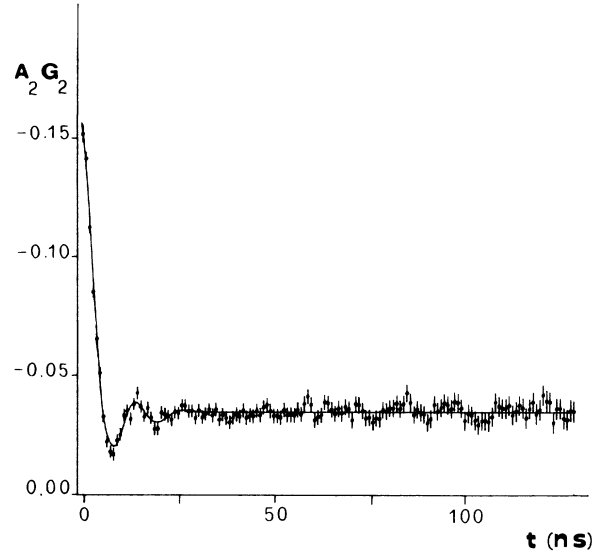


FIG. 1. TDPAC spectrum obtained after implantation.

$$A_2 G_2(t) = A_2 [f_0 + f_1 \exp(-\lambda t)] \quad (5)$$

to the experimental data (with $f_0 + f_1 = 1$). Here f_1 is the fraction of probe atoms experiencing time-dependent interactions and f_0 corresponds to a fraction of “unperturbed” probes. All the spectra taken between room temperature and 20 K can be fitted using (5) with the same values of f_0 and f_1 (9% and 91%, respectively). The dependence on temperature of the relaxation parameter λ is shown in Fig. 3.

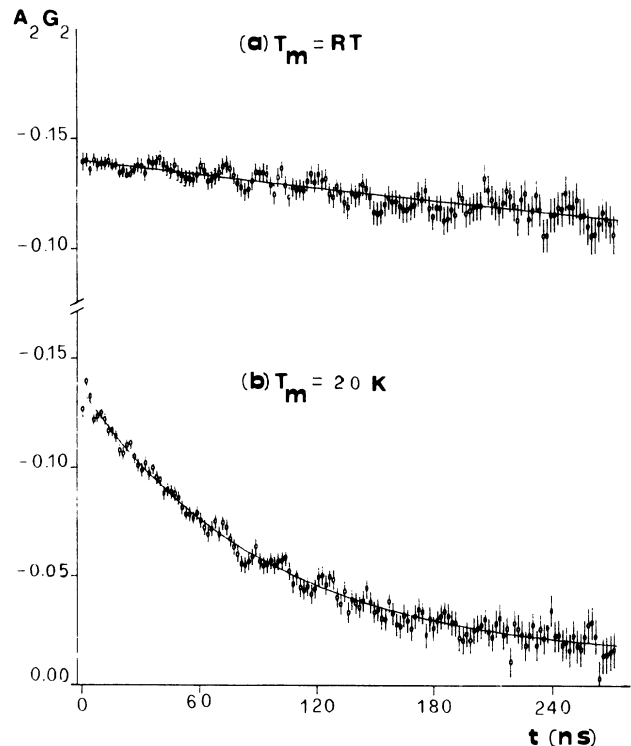


FIG. 2. TDPAC spectra obtained after annealing. (a) Measured at room temperature. (b) Measured at 20 K.

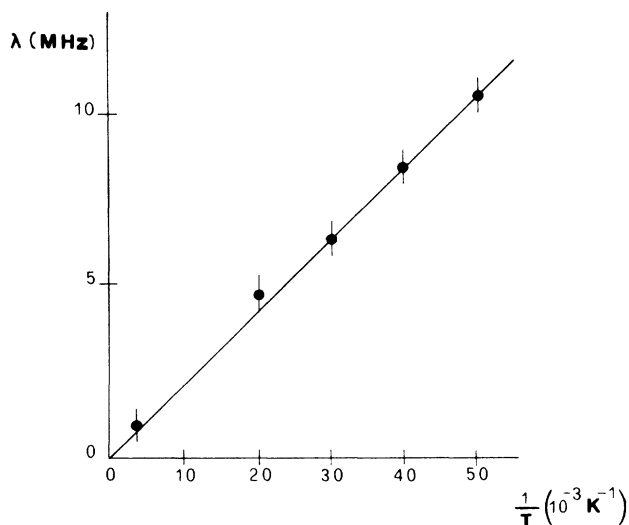


FIG. 3. The attenuation parameter is shown as a function of temperature. The solid line corresponds to $\lambda = (211.6/T)$ K MHz.

IV. DISCUSSION AND CONCLUSION

As was expected, an influence of the measurement temperature on the hyperfine interactions similar to that reported for ^{111}Cd in silicon was found. But there are interesting differences which will be discussed further. The result obtained at room temperature after annealing shows that all the probes are in sites of almost cubic symmetry. In the diamond lattice this symmetry exists at the substitutional sites as well as the tetrahedral interstitial sites. If both sites are populated, it would be possible to expect a "splitting" in the measured interaction at low temperatures due to the different influence of the electron-capture aftereffects at both sites. But this effect does not appear in the temperature range studied. Therefore, we can conclude that all the probes are in the same type of site. We conclude that this site is the substitutional one due to the absence of a strong attenuation at low temperature which is expected for probes at interstitial sites.¹ On the other hand, channeling studies have given evidence that indium implantation does not produce an interstitial component in germanium.⁴ This conclusion may appear to be in contradiction to our explanation of the hyperfine interactions observed for ^{111}Cd in silicon.¹ In that case we have reported static quadrupole interactions for probes in substitutional lattice sites. The electric field gradients were associated with a distortion of the cubic symmetry in the neighborhood of the probes. We have described this distorted configuration as one produced by holes more or less bound to the probes. The present experiments on germanium can be understood within the framework of the following scheme: The quadrupole interactions are produced by a distorted configuration in the neighborhood of the probes. The dynamic character is due to fluctuations of the distorted configurations between adjacent energy wells.

We will assume that the distorted configuration has the

same structure for Cd in both Ge and in Si. In both host lattices the substitutional Cd impurity causes local lattice distortion because of the difference between the ionic radius of the impurity and the host atoms. Also it could be possible that the localization of holes in the neighborhood of the impurity will produce an additional displacement. So, the distorted configuration consists of a cloud of atoms a little off from its lattice site and of electronic holes bound to it.

This distortion can change and reorientate with temperature, and can jump between wells of more or less the same energy. Due to the different physical properties of Si and Ge, namely, ionic radius, lattice parameters, force constants, etc., it is possible to expect a difference in the potential barrier which separates equivalent configurations. For this reason, the hyperfine interaction at low temperature is static for Cd in silicon but dynamic for Cd in germanium.

It is not possible to get the value of the activation energy for these jumps from the temperature dependence of the attenuation parameter λ , because the correlation time τ_c and the quadrupole frequency ω cannot be resolved. Both parameters are temperature dependent. τ_c depends on the activation energy for jumps between wells and ω on the geometry of the distorted configuration, which may change with temperature (as occurs in silicon). On the other hand, a decrease of the electric field gradient with temperature may be expected from the temperature dependence of the dielectric function. The fraction f_0 of probes at sites of cubic symmetry, even at low temperature, could be connected with sites where the holes are not bound, perhaps due to the presence of deeper traps in the surroundings. Then the distortions may still conserve the lattice symmetry. However, part of the f_0 value could also be produced by a shift of the baseline in the spectra due to absorption in the sample.

We conclude that the distortions in the neighborhood of Cd substitutional impurities in silicon and germanium are qualitatively similar. The difference in the activation energy for fluctuations can be ascribed to the different elastic and electronic properties of both hosts. More experimental evidence is necessary in order to decide if a Jahn-Teller distortion is involved or not. Furthermore, the question arises whether the Jahn-Teller distortion is induced by the electron-capture decay of ^{111}In . New information could be obtained by TDPAC experiments using the radioactive nuclei ^{111m}Cd . This metastable state with a half-life of 48 min decays to the Cd ground state through the same cascade studied here, without producing holes and electrons in the host bands.

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