

## Electric quadrupole interactions of $^{111}\text{Cd}$ nuclei in the cubic Ag lattice doped with Sn impurity\*

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 (Received 23 February 1976)

The electric quadrupole interactions produced by near-neighbor impurity atoms of Sn on  $^{111}\text{Cd}$  probe nuclei in a cubic Ag lattice were studied by the time-differential perturbed angular correlation technique. The effects of concentration of the impurity and of temperature have been investigated. The results show the presence of a high-frequency interaction  $\omega_h$  superimposed to a smeared out low frequency  $\omega_l$ . Both  $\omega_h$  and  $\omega_l$  increase with the impurity concentration, but the ratio  $\omega_h/\omega_l$  is constant for all the concentrations.

Considerable interest has recently been focused on the time-differential perturbed angular correlation (TDPAC) investigations of electric field gradients (EFG) produced in cubic metals by impurities<sup>1</sup> as well as by radiation damage.<sup>2</sup> The present work describes, for the first time, a systematic TDPAC study of the effect of nearest-neighbors (nn) Sn impurity atoms in a cubic Ag lattice. The radioactive probe nuclei were obtained by the reaction  $^{109}\text{Ag}(\alpha, 2n)^{111}\text{In}$ , where the  $^{111}\text{In}$  nuclei decay to  $^{111}\text{Cd}$  by electron capture. The TDPAC technique was used to measure the interaction of the quadrupole moment of the  $I = \frac{5}{2}$  247-keV state in  $^{111}\text{Cd}$  with the EFG's produced by the impurity. The Sn impurity was introduced into Ag by melting the constituents (with purity  $\geq 99.99\%$ ) together under argon atmosphere and subsequently annealing the alloy also in argon during some hours. The sample in our experiment contains three components, the matrix of Ag, the solute atoms of Sn and the radioactive probe  $^{111}\text{In}$ . Both the concentration of the impurity atoms and the temperature of the sample were changed in the present work.

Assuming only static quadrupole interaction in a random polycrystalline sample the angular correlation attenuation coefficient can be expressed by

$$G_{hh}(t) = \sum_n \sigma_{hn} \cos(\omega_n t) e^{-(\delta/2)nt} \quad (1)$$

where the amplitudes  $\sigma_{hn}$  and the frequencies  $\omega_n$  are related to the nuclear quadrupole frequency  $\omega_Q$  and to the asymmetry parameter  $\eta$ .<sup>3</sup> The term  $e^{-(\delta/2)nt}$  gives the Lorentzian distribution of frequencies having a width  $\delta$ .

The results for different amounts of Sn impurity measured at room temperature are shown in Fig. 1 together with the results for 3 at.% of Sn mea-

sured at temperatures of 90 and 673 °K. The  $G_{22}(t)$  curves show very clearly that the  $^{111}\text{Cd}$  nuclei are exposed not only to a smeared out low quadrupole frequency but in addition a certain percentage of them experience a sharp quadrupole frequency, denoted by the small peaks superimposed to the gradually declining curves. Also in Fig. 1 is shown the result for an annealed sample of pure irradiated Ag, which as it is expected corresponds to the unperturbed  $G_{22}(t) = 1$  value.

The analysis of the data was performed assuming that a part  $a_h$  of the probe nuclei are subjected to a high frequency  $\omega_h$ , a second part  $a_l$  to a distribution of low frequency  $\omega_l$ , and a third part  $a_0$  of unperturbed probe nuclei which do not feel any field. The solid lines in Fig. 1 are computer fits and Table I displays the obtained parameters for the different concentrations of impurity and temperatures.

Several interesting facts can be drawn from the analysis of the results shown in Table I:

(i) The  $^{111}\text{Cd}$  nuclei are exposed not only to a distribution of low quadrupole frequencies around  $\omega_l$ , but also a part of them experience a high frequency  $\omega_h$ . In previous experiments on the influence of impurities in Ag (Ref. 1) and in the tetragonal In-Sn system,<sup>4</sup> the presence of high-frequency interactions was not observed.

(ii) The low frequency  $\omega_l$  is generated by impurities distributed at various different atomic distances from the probe nuclei and can be explained in terms of the original Kohn and Vosko approach.<sup>5</sup>

(iii) The observation of the sharp frequency  $\omega_h$  can be understood if it is assumed that the nearest sites to the probe nuclei are occupied by Sn impurity atoms. The EFG obtained for 1.5 at.% of Sn in Ag is of the order of  $2.5 \times 10^{18}$  V/cm<sup>2</sup>. On the other hand the EFG measured by NMR<sup>6</sup> at the

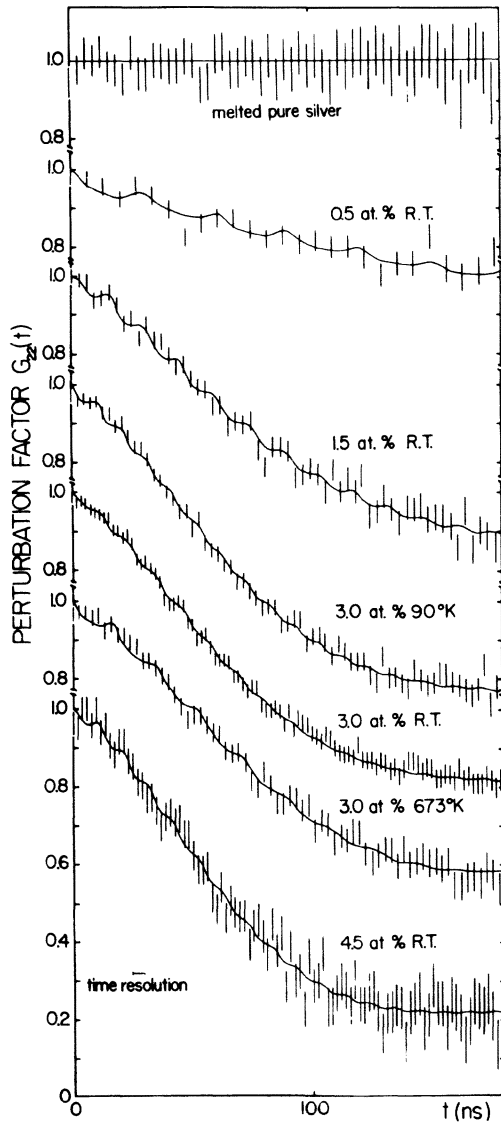


FIG. 1. Time-differential attenuation coefficients  $G_{22}(t)$  of the  $^{111}\text{Cd}$   $\gamma$ - $\gamma$  correlation observed for different amounts of Sn impurity. Also are shown the results for 3 at.% of Sn for different temperatures.

first shell around 1 at.% of Sn impurity in Cu matrix is of the order of  $10^{17}$  V/cm<sup>2</sup> or 25 times smaller. In fact, a value of  $4.5 \times 10^{16}$  V/cm<sup>2</sup> much closer to the NMR result is calculated from our low-frequency interaction  $\omega_l$  (we used  $Q = 0.77$  b for the  $\frac{5}{2}^+$  state in  $^{111}\text{Cd}$ ). For other impurities in Cu and in Al matrix the NMR results<sup>6-9</sup> also associate EFG's between  $10^{16}$  and  $10^{17}$  V/cm<sup>2</sup> as due to the nn impurity atoms. It is interesting to observe that the TDPAC results of Behar and Steffen<sup>2</sup> also give EFG's of the order of  $10^{18}$  V/cm<sup>2</sup> in the damaged Ag host.

(iv) The high frequency  $\omega_h$  increases with concentration and the ratio  $a_h/a_l$  is approximately independent of concentration. These facts are difficult to explain. A possibility is that when the concentration is increased more than one closest site to the Cd nuclei is populated. In addition an inspection of Table I shows that the rate of variation of the high frequency  $\omega_h$  with concentration is the same as for the low frequency  $\omega_l$ , resulting that the ratio  $\omega_h/\omega_l$  is remarkably constant for all the concentrations. It is very interesting to note that there is a linear relationship between the log of the concentration and both  $\omega_l$  and  $\omega_h$ , as can be observed in Fig. 2. This empirical relation is very well fulfilled for all the experimental points.

(v) The distribution  $\delta_l$  of low frequencies decreases with the increase of concentration as is expected. For the high frequency  $\delta_h$  remains approximately constant and small.

(vi) The high-frequency interaction is strongly dependent on the temperature. The three measured temperature points are very well represented by a simple  $T^{3/2}$  relation (Fig. 3) as in various noncubic hosts like Zn (hexagonal), Sn, In (tetragonal), and  $\beta$ -Ga (monoclinic). Recently, the temperature dependence of EFG's in noncubic metals was theoretically studied by Jena<sup>10</sup> who concludes that the  $T^{3/2}$  behavior is primarily due to conduction electron effects and applies to all noncubic metals. Our result shows that for a cubic metal a similar  $T^{3/2}$  relation is observed

TABLE I. Parameters obtained from the fittings. Subscripts  $l$  and  $h$  refer to low and high frequencies.

$c$ (at.% of Sn)	$T$ (°K)	$a_0$	$a_l^a$	$\omega_l$ (MHz)	$\delta_l$	$a_h^a$	$\omega_h$ (MHz)	$\delta_h$	$\frac{\omega_h}{\omega_l}$
0.5	300	$0.60 \pm 0.04$	0.36	$4.0 \pm 0.3$	$0.81 \pm 0.06$	0.04	$210 \pm 10$	0.02	$52.5 \pm 3.0$
1.5	300	$0.18 \pm 0.02$	0.75	$7.8 \pm 0.3$	$0.48 \pm 0.07$	0.07	$430 \pm 10$	0.02	$55.0 \pm 2.5$
3.0	300	$0.12 \pm 0.02$	0.82	$9.9 \pm 0.3$	$0.35 \pm 0.03$	0.06	$550 \pm 10$	0.02	$55.5 \pm 2.0$
4.5	300	$0.10 \pm 0.02$	0.83	$11.5 \pm 0.5$	$0.28 \pm 0.06$	0.07	$635 \pm 15$	0.02	$55.2 \pm 2.7$
3.0	90	$0.12 \pm 0.02$	0.81	$9.7 \pm 0.3$	$0.33 \pm 0.04$	0.07	$606 \pm 15$	0.01	
3.0	673	$0.15 \pm 0.03$	0.78	$9.2 \pm 0.3$	$0.30 \pm 0.04$	0.07	$345 \pm 10$	0.03	

<sup>a</sup> Typical errors are about 10%.

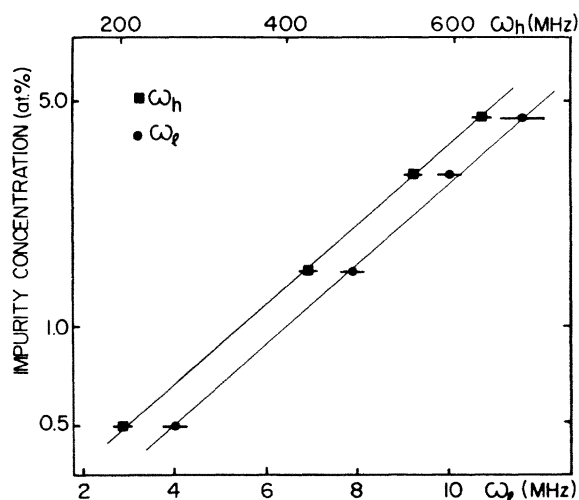


FIG. 2. Rate at which the low and the high frequencies  $\omega_l$  and  $\omega_h$  change with the concentration. As can be seen there is a linear relationship between log of the concentration and both frequencies.

for the EFG attributed to a nn impurity. On the contrary the low frequency  $\omega_l$  changes very little with the temperature and  $\delta_l$  is practically constant.

(vii) The EFG measured for 4.5 at.% of Sn is of the order of  $3.5 \times 10^{18}$  V/cm<sup>2</sup>. This value is more than three times larger than the highest field measured on Cd in noncubic metals.

The simplest interpretation of our results is that the high field is produced by the nn solute atoms and the low-field distribution is generated by impurities distributed at various atomic distances from the probe nuclei. A direct comparison of the high-frequency interactions with the theory of Blandin and Friedel or Kohn and Vosko

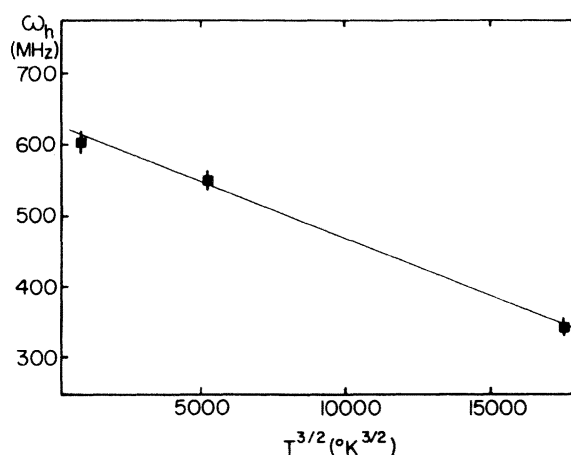


FIG. 3. Rate at which changes the high frequency  $\omega_h$  with the temperature. The three measured temperature points are very well represented by a simple  $T^{3/2}$  relation.

is not valid since this approach is an asymptotic one and no agreement is expected for the first neighbors. It is hoped that a quantitative understanding of the relative importance of the various contributions to the EFG's can be obtained by a systematic study of impurities with different valence and size in the cubic Ag metal. Such a work is in progress.

The authors are grateful for the assistance of L. Amaral and for the support of Dr. F. P. Livi. We also thank Dr. A. Vasquez for the assistance in the computer analysis. One of us (M.B.) acknowledges the financial support of the CONICET (Argentina).

\*Work supported by Banco Nacional de Desenvolvimento Econômico and Conselho Nacional de Desenvolvimento Científico e Tecnológico.

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