TCNQ[3] as a function of temperature. The one-dimensionality of these organic crystals is firmly established,[1,2] but improved descriptions of the spin dynamics would be desirable. In our opinion, the diffusion limit appropriate in TMMC (where at room temperature $kT > J$, the intrachain exchange) fails in magnetically dilute systems like PD-chloranil, TMPD-TCNQ, or TMPD-chloranil in which $kT < \Delta E_p$.

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Time-Differential Quadrupole Interactions following Recoil Implantation*

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The quadrupole interaction of the 247-keV state of ^{111}Cd recoil implanted into polycrystalline Cd metal has been measured by the time-differential perturbed angular-correlation technique, following Coulomb excitation. The resulting interaction frequencies at three temperatures are in agreement with the recently measured lattice quadrupole interaction in Cd. This observation of a usable electric field gradient for recoil nuclei suggests the possibility of measuring excited-state quadrupole moments by such time-differential studies following nuclear-reaction implantation into metals.

The knowledge of quadrupole moments of excited nuclear states represents a significant body of information in regard to the understanding of nuclear structure physics. Unfortunately, very few excited-state quadrupole moments have been measured except for those of stable nuclei that can be studied by the reorientation effect in Coulomb excitation[1] and the relatively few Mössbauer and radioactivity results.[2] A possible method for expanding quadrupole-moment measurements is the observation of a quadrupole-moment–electric-field-gradient (EFG) interaction by perturbed angular correlations (PAC) following a nuclear reaction. Various nuclear reactions would allow such quadrupole-moment measurements of many different excited states, provided the excited residual nuclei experienced an appropriate static EFG upon recoil implantation into a suitable backing material. The question of whether a usable crystal EFG can be found for a large variety of recoil nuclei is unanswered; noncubic metals, where little damage is expected from the electronic stopping of recoil ions and the beam, offer the best hope. Very recently the quadrupole-interaction frequency for the 247-keV ^{111}Cd state of nonimplantation, lattice nuclei in polycrystalline Cd metal was measured by γ-γ time-differential PAC using ^{111m}Cd radioactivity[3,4]; these results are consistent with a unique EFG that is static and axially symmetric. In the present Letter, we report on a charged-particle–γ time-differential PAC experiment in which the same quadrupole interaction was observed for the 247-keV ^{111}Cd state of nuclei that were recoil implanted into polycrystalline Cd metal following Coulomb excitation with 40-MeV ^{16}O ions. These results point to the successful measurement of excited-state quadrupole moments by time-differential PAC following nuclear reactions. Since the present results were obtained for large recoil energies, this technique should in particular be applicable to the (heavy ion, *Xn*) reactions which are known to populate numerous interesting isomers.

The quadrupole interaction of the $\frac{5}{2}^+$ 247-keV state ($\tau = 121$ nsec) of ^{111}Cd recoil implanted into Cd metal has been studied by the time-differential PAC technique. The state was populated following Coulomb excitation with 40-MeV ^{16}O ions from the Stony Brook FN tandem. The PAC measurements were made at different temperatures in order to study the temperature dependence of the perturbed amplitude and to aid in the identification of the quadrupole interaction via its temperature dependence. Measurements to date have been made at room temperature, 180°K, and 77°K with a polycrystalline target consisting of 5 mg/cm^2 of ^{111}Cd metal backed by 10 mg/cm^2 of natural Cd metal. Because of background and target-stability considerations, initially the beam was stopped in a shielded Faraday cup; for the measurements discussed here, the beam was stopped directly behind the target. Backscattered ^{16}O ions were detected in a cooled 100-μm annular silicon detector, while the 247-keV $\frac{5}{2}^+ \rightarrow \frac{1}{2}^+$ decay γ rays were detected in two 5.1-cm × 5.1-cm NaI scintillators positioned initially at 45° and 90°. For an angular correlation characterized by an A_2 and A_4 of comparable magnitude but of opposite signs, as expected for the fractional population due to direct Coulomb excitation, the most favorable angle is 90°; however, 0° is preferable for only an A_2. In these measurements the Faraday cup prevented detection at 0°; measurements at 0° are currently in progress. The charged-particle–γ time spectra were obtained from fast-slow delayed-coincidence electronics with a time resolution of ~1 nsec full width at half-maximum.

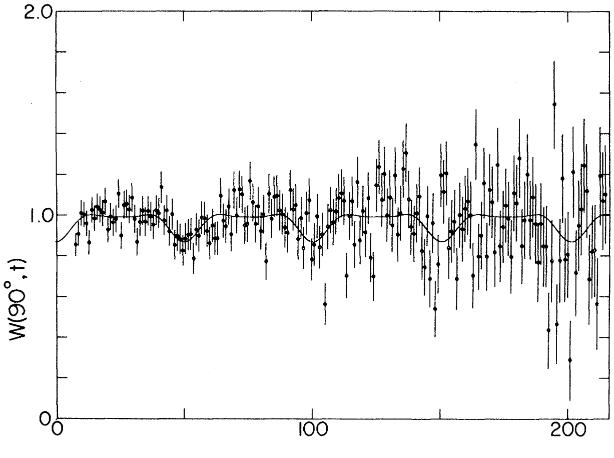

FIG. 1. Experimental $W(90°,t)$ data for the 247-keV $\frac{5}{2}^+$ state in ^{111}Cd recoil implanted into Cd metal. The solid curve is a theoretical fit to the data assuming a static, axially symmetric field gradient.

Figure 1 shows the perturbed angular-correlation data, $W(\theta, t)$, as a function of time for the γ-ray observation angle of 90° and at a temperature $T = 180$°K. The experimental $W(\theta, t)$ were extracted from the perturbed time spectra by subtracting random events and dividing out the exponential lifetime. These data were accumulated over approximately 20 h with about 25 nA of ^{16}O^{+5} ions on target. Because of evidence of target deterioration with time, the target spot was changed every 2–3 h to minimize the effect of crystal damage in the target.

The experimental $W(\theta, t)$ were fitted by least-squares with the theoretical expression $W(\theta,t) = \Sigma_k A_k G_k(t) P_k(\cos\theta)$ with the polycrystalline quadrupole perturbation factors $G_k(t) = \Sigma_n S_{kn} \cos(n\omega_0 t)$ assuming a static, axially symmetric field gradient. For $J = \frac{5}{2}$, $\omega_0 = 3eQV_{zz}/20\hbar$, where Q is the quadrupole moment, $V_{zz} = \partial E_z/\partial z$ refers to the EFG, and the S_{kn} are calculable factors. The frequency ω_0 as well as the correlation coefficients A_k were treated as variable parameters in the fits. The quadrupole interaction frequency $\nu_Q = eQV_{zz}/h$ is related to ω_0 by the equation $\nu_Q = (10/3\pi)\omega_0$. The results of the data fits at each temperature are summarized in Table I; included in the table are the quadrupole interaction frequencies ν_Q and the perturbation amplitudes, defined as $W(90°, t = 2\pi N/\omega_0) - 1.0 = -0.500A_2 + 0.375A_4$. The fitted ν_Q, which were found to be very insensitive to the values of A_k, have uncertainties as

TABLE I. Quadrupole-interaction frequencies and perturbation amplitudes for the 247-keV $\frac{5}{2}^+$ state in ^{111}Cd obtained in the present experiment at different temperatures following recoil implantation into Cd. For comparison, the interaction frequencies known for lattice nuclei are also listed at the same temperatures.

	Recoil-implanted nuclei		Lattice nuclei
T (°K)	ν_Q (MHz)	Amplitude $[W(90°, t=2\pi N/\omega_0)-1.0]$	ν_Q (MHz)
295 ± 5	124.3 ± 4.4	-0.09 ± 0.03	124.5^{a}
180 ± 5	133.2 ± 4.8	-0.13 ± 0.02	132.0^{a}
77^{+10}_{-0}	135.3 ± 3.0	-0.08 ± 0.02	136.5^{a}

[a] See Refs. 3 and 4.

small as 2.2%. Within the rather large uncertainties in the perturbation amplitudes, no strong temperature dependence of the amplitude was observed. The time spectra at 45° were also consistent with these fits although with reduced sensitivity to the ν_Q. For comparison purposes, the corresponding ν_Q for lattice ^{111}Cd nuclei are given for each temperature; these ν_Q were obtained from γ-γ PAC studies with ^{111m}Cd radioactivity[3,4]. From this comparison, it can be seen that the observed ν_Q for recoil-implanted nuclei agree within uncertainties with those for lattice nuclei.

The above analysis of the perturbation data shows that a significant number of recoil-implanted ^{111}Cd nuclei do experience a common EFG in polycrystalline Cd metal. The fact that the interaction frequency at each temperature agrees within errors with that for ^{111}Cd lattice nuclei indicates that the EFG for recoil-implanted ^{111}Cd nuclei originates at lattice sites. Furthermore, with the observation of 4 to 5 periods, the analysis indicates that the EFG is also static and axially symmetric.

A comparison of the perturbation amplitude with the initial γ-correlation amplitude can give a measure of the number of recoil nuclei that experience the unique EFG. A reduction in the perturbation amplitude is expected if a fraction of the recoil nuclei do not locate in a lattice site but rather end up interstitially where the extranuclear environment may be very different from that of a lattice site. The perturbation amplitude, however, can also be reduced from the initial correlation amplitude for other reasons. Crystal damage such as nearby vacancies or interstitials produced by the recoil ions and/or the beam could strongly relax the nuclear alignment for a number of the recoils. Other more complicated dynamic effects near the end of the stopping process are also possible. These effects, which reduce the perturbation amplitude, do not alter a time-differential frequency measurement, provided a sufficient number of the recoil nuclei see the expected EFG to give an observable perturbation amplitude. These effects do, however, interfere with time-integral PAC measurements where it is usually assumed that all the nuclei see the same EFG with full alignment. Several time-integral studies for implanted nuclei have shown these effects.[5]

In order to study the reduction in the perturbation amplitude, the initial γ-correlation amplitude had to be measured. The γ correlation expected in the present experiment is difficult to calculate because the 247-keV state is populated to a significant amount by mixed γ cascades from higher-energy levels that are also Coulomb excited. An estimate from known $B(E2)$ values suggests that the amount of direct Coulomb excitation is only on the order of 10%. To measure the initial γ correlation, the time measurements discussed above were repeated as a function of angle but with a thin ~600-μg/cm^2 ^{111}Cd target evaporated on a Cu backing. The recoil ^{111}Cd nuclei thus were stopped in the Cu where no EFG is expected because of the cubic lattice. Preliminary results from this angular-distribution measurement give an $A_2 = 0.24 \pm 0.05$ and an $A_4 = 0.09 \pm 0.07$. The perturbation amplitude at 90° corresponding to this γ correlation is -0.09, which to within ~30% uncertainties is consistent with the amplitudes listed in Table I that were measured at the different temperatures. No definite statements can be made, however, regarding small amplitude reductions. The amplitude comparisons are presently being improved by repeating the γ-correlation measurement in heavier cubic backings; the measurement is made difficult because of the thin ^{111}Cd target and the large background. Also, better statistics are being obtained for the perturbation amplitudes with measurements at $\theta = 0°$ which turns out to be a more sensitive angle with the presently measured γ correlation. The evidence of a reduction in perturbation amplitude with beam time on a particular target spot must also be further investigated.

Since a quadrupole interaction frequency is proportional to a product of the quadrupole moment and the EFG, the magnitude of the EFG must be known in order to extract a quadrupole moment from a frequency measurement. The EFG's at impurity nuclei are not generally known and cannot be reliably calculated. Thus, it is often only possible to obtain ratios of quadrupole moments from two independent measurements with a common EFG; all isotopes of a given Z are expected to have identical EFG in the same crystalline metal because of similar atomic properties. On the basis of the present measurements, where nuclei recoil implanted by a nuclear reaction experience the lattice EFG, quadrupole-interaction measurements from Mössbauer and radioactivity experiments can also be combined with those from recoil PAC experiments to give quadrupole-moment ratios. If an absolute quadrupole moment is known from other techniques, then it may be possible to obtain the absolute EFG from a quadrupole interaction measurement on the same state.

In conclusion, the present experiment in our opinion has demonstrated the feasibility of quadrupole-interaction measurements of excited states by time-differential PAC following a nuclear reaction with recoil implantation into a metal backing. To extend this technique to a large number of excited nuclear states, reactions with thin targets, where all of the recoils reach the appropriate metal backing, will have to be used. The question of whether this reaction recoil technique has wide application depends on lattice-site populations and on the EFG for different impurities in various suitable metal backings. Further time-differential PAC studies with reactions that populate and strongly align excited states of sufficient lifetimes are necessary to obtain this information. Several preliminary experiments have been made to investigate these questions. For recoil Sn in Cd, the reactions ^{114}Cd(α, $2n$)^{116}Sn and ^{116}Cd(α, $2n$)^{118}Sn with $E_\alpha = 24$ MeV were used to study the quadrupole interaction of the 2.35-MeV ^{116}Sn 5^- state ($\tau = 330$ nsec) and 2.57-MeV ^{118}Sn 7^- state ($\tau = 330$ nsec). The experimental arrangement was similar to that described above except that neutrons were detected at 0° instead of the backward detection of charged particles. No significant time-differential perturbations were observed in either time spectra. The reason for not seeing any time-dependent perturbations, whether the recoil Sn do not locate in lattice sites or whether the EFG or quadrupole moment are small, is not known at the present time. Assuming a ν_Q equal to that of the ^{111}Cd 247-keV state, the spin dependence of ω_0 would make the perturbation period equal to 450 nsec for the ^{116}Sn 5^- state and 900 nsec for the ^{118}Sn 7^- state as compared with the 50-nsec period for the $\frac{5}{2}^-$ state in ^{111}Cd. Thus, with a small ν_Q, the perturbations would be difficult to detect. PAC studies have also been made for the recoil implantation of ^{111}Cd into Zn following Coulomb excitation; preliminary results for the 247-keV state show a quadrupole interaction that is comparable to that for ^{111}Cd in Cd at room temperature.

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