ment of the TCNQ$^-$ stack. I believe that the possibility of coupling of orientational (internal) degrees of freedom with the band motion of the extended π electron wave function of π molecular planar systems is a genuinely new aspect of organic conductors.

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Detection of a Quadrupole Interaction in a Ferromagnetic Cubic Lattice by Allowed β-γ Time-Differential Perturbed Angular Correlation

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We observed the magnetic hyperfine interaction of ^{129}I in nickel, by allowed β-γ time-differential perturbed angular correlation, as a consequence of a quadrupole interaction existing in a ferromagnetic cubic lattice.

The quadrupole interaction has been a major aspect in the study of hyperfine interactions as it gives valuable information in the field of nuclear as well as solid-state physics. Recently Raghavan, Raghavan, and Kaufmann[1] demonstrated that an allowed β-γ cascade can be used, in a time-differential perturbed angular correlation (TDPAC), to detect the quadrupole interaction. Because of the quadrupole interaction, the initial polarization of the nucleus after β decay is transformed into a periodically varying alignment.[2] Moreover, by this technique the sign of the interaction can be determined. In this Letter we propose a new application of this method, which is very useful in cases where a perturbing quadrupole interaction exists in the presence of a dominating magnetic interaction. Such cases of combined interaction exist even in cubic lattices as is known from the NMR iridium experiments.[3,4] Recently[5] we found, in a Mössbauer experiment on ^{129}I in iron, also a small quadrupole interaction together with a large magnetic interaction.

It is the purpose of this Letter to show that the β-γ TDPAC technique is very suitable in this kind of study.

A general theoretical formulation for a β-γ angular correlation in the presence of a combined magnetic and electric interaction is rather lengthy. A numerical diagonalization is needed for the interaction Hamiltonian, which depends on the relative orientation of the electric field gradient to the magnetic field and their relative strengths. The perturbation factor can no longer be written in a closed and transparent form. Therefore, we would like to propose a frame for approaching the problem, with the advantage that the formulas remain interpretable. In this way we clearly show what can be expected in this particular case, without, however, giving a complete description.

The perturbed angular correlation is given as

$$W(\vec{k}_1,\vec{k}_2,t)=\sum_{\substack{k_1k_2\\N_1N_2}}(-1)^{k_1+k_2}A_{k_1}(\beta)A_{k_2}(\gamma)G_{k_1k_2}{}^{N_1N_2}(t)Y_{k_1}{}^{N_1}(\theta_1,\Phi_1)Y_{k_2}{}^{N_2}(\theta_2,\Phi_2). \tag{1}$$

In the perturbation factor $G_{k_1k_2}{}^{N_1N_2}(t)$ the terms with $N_1\neq N_2$ only occur in cases where the perturbation has nonaxial symmetry or the interaction matrix is nondiagonal as in the case of combined interaction. When the magnetic interaction dominates, the quadrupole interaction can be handled as a first-order perturbation. Then a remarkable simplification of the theory is obtained as only $N_1=N_2$ terms must be considered in Eq. (1). With the magnetic field chosen as the quantization axis, the energy level splitting is

$$E_m=-m\hbar\omega_\mathrm{B}+\hbar\omega_Q\tfrac{1}{2}(3\cos^2\beta-1)[3m^2-I(I+1)]. \tag{2}$$

In this formula, m labels the split-level term in the intermediate state with spin I. The interaction frequencies are ω_B for the magnetic and ω_Q for the electric interaction. The angle β is one of the Euler angles which specify the relative orientation of both interactions. The energy difference among the different m terms can be written as

$$\Delta E(m,m')=-N\hbar\omega_\mathrm{B}+\tfrac{1}{2}(3\cos^2\beta-1)n\hbar\omega_0, \tag{3}$$

with $N=m-m'$, $2n=m^2-m'^2$, and $\omega_0=6\omega_Q=6eQV_{zz}/\hbar 4I(2I-1)$. In the pure quadrupole case[2] the time-dependent perturbation factor of the angular correlation for k_1+k_2 odd (β-γ case) is given as

$$G_{k_1k_2}{}^{NN}(t)=\sum_{n>0}S_{nN}{}^{k_1k_2}(-i)\sin(n\omega_0t). \tag{4}$$

By adding a magnetic interaction we obtain,

$$G_{k_1k_2}{}^{NN}(t)=\sum_{n>0}S_{nN}{}^{k_1k_2}(-i)\sin(n\omega_0t)\exp(-iN\omega_\mathrm{B}t). \tag{5}$$

By using the symmetry relation $S_{n\,-N}{}^{k_1k_2}=(-1)^{k_1+k_2}S_{nN}{}^{k_1k_2}$ we write, referring to Eq. (1) and setting $\Phi=\Phi_1-\Phi_2$,

$$G_{k_1k_2}{}^{NN}e^{+iN\Phi}+G_{k_1k_2}{}^{-N-N}e^{-iN\Phi}=\sum_{n>0}S_{nN}{}^{k_1k_2}\sin(n\omega_0t)\sin N(\omega_\mathrm{B}t-\Phi). \tag{6}$$

The result is that a beating pattern appears in which the magnetic precession is modulated by the quadrupole precession. Indeed, by the quadrupole interaction the energy levels in the intermediate state are shifted in such a way that the splitting results in transition frequencies equal to $N\omega_\mathrm{B}\pm n\omega_0$, which interfere and result in a beating when ω_0 is small. Furthermore, from the sine behavior of the perturbation factor it is clear that only terms contribute where the magnetic precession function is modulated by (the harmonics of) the quadrupole interaction. This is in contrast to the γ-γ experiments where also the nonmodulating $n=0$ term appears.

In the present experiment, we detected the 1.46-MeV β, 27-keV γ cascade in the ^{129}Te decay. The activity was implanted in a nickel foil with an accelerating voltage of 75 kV and a dose of 10^{15} ions/cm^2. The β rays were detected with a thin (1 mm) plastic scintillator to avoid absorption of γ rays and γ-γ coincidences. The low-energy γ ray was detected with a thin NaI(Tl) crystal. The nickel foil was carefully magnetized in a direction perpendicular to the plane of the detectors. To avoid deviation of the electrons in the exter-

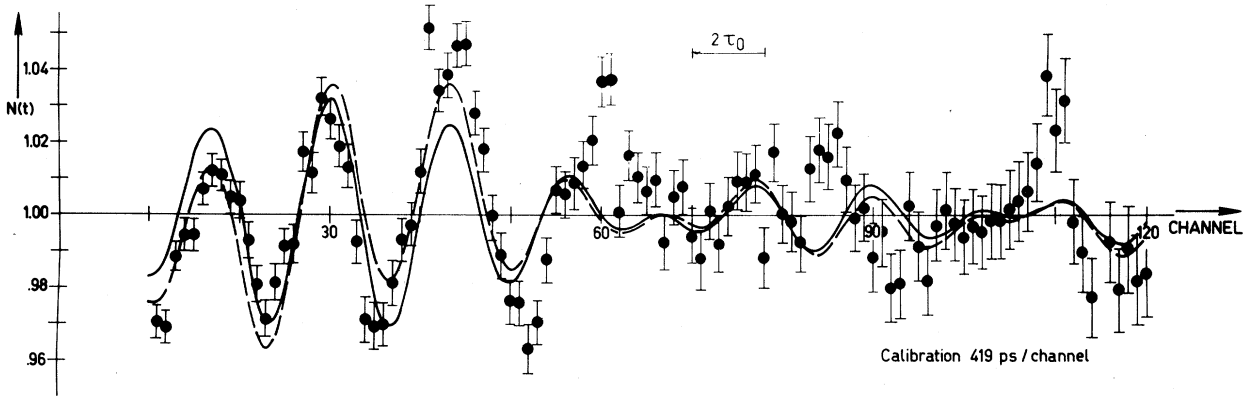

FIG. 1. β-γ TDPAC on ^{129m}Te implanted in nickel, fitted with one field site (full line) and two field sites (dashed line).

nal field the polarizing magnet was switched off before the experiment started. The time spectrum of the delayed coincidences was measured with an angle of 90° between the detectors and the time resolution of the system was about 3 nsec. As, for counting reasons, only one interdetector angle was measured, the nuclear decay could only be removed mathematically. The function fitted to the corrected experimental counting rate as a function of delay time, shown in Fig. 1, has the form

$$N(t)=1+A\sin(\omega_B t)[C\sin(\omega_0 t)+D\sin(2\omega_0 t)],$$

as is given by Eq. (5). The hyperfine parameters are $\omega_B=+(1.12\pm0.10)\times10^9$ rad/sec or $B=+205\pm20$ kG and $P_2(\cos\beta)\omega_0=+(71\pm5)\times10^6$ rad/sec or $P_2(\cos\beta)V_{zz}=-(4.6\pm0.3)\times10^{17}$ V/cm^2. A slight improvement in the fit is obtained by introducing a lower magnetic field component. From our previous γ-γ TDPAC experiments[6] different field sites in nickel at this dose are evident. The first part of the data are well reproduced by the fit, while the last part deviates. Such partial agreement could be expected because a unique angle β was assumed and the direction of the foil magnetization is not well established in this experiment. Moreover, taking also the $N_1\neq N_2$ terms into account does not improve the fit. The contribution of these terms is small compared to the $N_1=N_2$ part. The main effect of these second-order perturbation terms is that the level shift due to the quadrupole interaction becomes slightly anharmonic in the quadrupole frequency ω_0.[7]

Besides this partial agreement these data clearly demonstrate a quadrupole interaction existing in a cubic lattice such as nickel. Indeed, this quadrupole interaction is the reason that we could detect a magnetic hyperfine interaction by allowed β-γ angular correlation. This result is important as it is the first time a magnetic interaction has been seen by an allowed β-γ cascade.

The interpretation of the present result is difficult at this moment as a lot of information about the parameters, such as β, the relative ratio of the combining interactions, and the orientation of the magnetic field, is still unresolved. New data are needed in well-defined geometries and magnetic orientations. This work as well as a more general theory is in progress and will be reported in the future. However, already a few comments can be stressed. The data show that the magnetization of the foil in this experiment was not oriented perpendicular to the detector plane, as then, from the angular dependence of the spherical harmonics appearing in the full angular correlation function, the $N_1=N_2$ terms vanish. Because the external polarizing field was switched off, the resulting magnetization of the foil can be distributed around the perpendicular direction.

The value of the magnetic hyperfine field derived from these data is smaller than the 293 ± 3 kG result from Mössbauer experiments. This discrepancy probably originates in the different temperature regions for the two experiments. This fact should suggest that the hyperfine field at iodine nuclei does not follow the Brillouin curve as is found for Sn in iron.[8]

Concerning the origin of the detected quadrupole interaction, three possibilities are open: magnetostriction, vacancies, and magnetically induced quadrupole interaction. The magnetostriction creates field gradients an order of magnitude too small.[9] Also introduction of vacancies or defects in the nickel matrix by implantation seems to be improbable, as then the quadrupole interaction is random and no effects can be detected by an allowed β-γ correlation. Moreover, recent experiments with a much lower implantation dose still show the β-γ effect. In the present situation of the observations and analyses of data only the magnetically induced quadrupole interaction[9] can account for the observed effects. In this case the electric field gradients will be oriented axially symmetrically around the magnetic field, keeping the angle β fixed. We have to integrate over the azimuthal angle of the field gradient. This results in the condition that $N_1 = N_2$ and we return to the theoretical frame presented above. In conclusion, the actual experiment demonstrates that the allowed β-γ TDPAC technique is a unique tool for studying the quadrupole interaction in the presence of a large magnetic field.

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Scaling of High-Frequency Sound Propagation near the Ferromagnetic Transition of MnP

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The propagation of longitudinal sound waves at frequencies between 10 and 520 MHz is analyzed at the ferromagnetic transition of metallic MnP. In spite of the existence of highly asymmetric attenuation about T_c, it is shown that the data are consistent with a symmetric critical relaxation time, τ. It is also shown that the reduced attenuation can be scaled by functions of $\omega\tau$ which differ above and below T_c.

Previous experimental investigations have shown that sound propagation phenomena near magnetic phase transitions can be classified, somewhat unexpectedly, into two categories: those observed in insulators and those in metals. In magnetic insulators, a sound wave predominantly couples to spin-energy density fluctuations which decay most effectively by emission of a phonon of energy $\approx kT_c$.[1,2] The time scale for insulators is thus characteristic of the spin-lattice relaxation time for that material. The temperature dependence of this process is characteristic of the magnetic specific heat and leads to a weak singularity in the acoustic attenuation at T_c.[3] In magnetic metals, the sound wave couples directly to the dynamic spin fluctuations and a time scale related to the decay of correlations within the spin system is appropriate.[4] The temperature dependence of this decay rate is strong and can be predicted, for many systems, by dynamic scaling[5] or mode-mode coupling theory,[6] and is accessible to measurement by inelastic neutron scattering.[7]

Recently, theoretical and experimental attention has been directed toward the behavior of sound waves in the regime for which the sound frequency becomes equal to or greater than the characteristic order-parameter decay rate, $\omega \gtrsim \tau^{-1}$.[5,8-11] I report here an analysis of sound attenuation measurements in the ferromagnet MnP near T_c. The regime $0.1 \lesssim \omega\tau \lesssim 10$ has been experimentally explored above and below T_c. The magnitude and temperature of the attenuation maximum which occurs below T_c is observed to depend on the sound frequency. It is shown, however, that the reduced attenuation is a single-