

FIG. 2. Values of H^{-1} corresponding to maxima and minima and maxima and minima of δ , plotted against integers.

between the two values of β^*/E_0 is further evidence for a one-to-one correspondence between the various magneto-oscillatory effects in bismuth.

Further work is in progress and will be reported in a later date. We wish to acknowledge the benefit of discussions with Dr. T. G. Berlincourt.

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Multiphonon Transitions in the F Center

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KLICK¹ has shown that transitions from the excited state of an F center to the ground state occur without photon emission. The possibility of that center transferring its energy to a close by foreign atom ("sensitized luminescence") seems slight since the crystals Klick used have no known appropriate activator. One seems forced to the conclusion that the system transfers its energy directly to the lattice by a spontaneous emission of about 50 phonons. This process must occur in less than 10^{-8} sec to suppress the emission of photons. At present the mechanism by which this process occurs is not understood.

The author has constructed a model to indicate how this multiphonon process occurs. The model consists of an electron attached to a vibrating ion. Transition probabilities between state e (the electron is in a bound excited state and the ionic degrees of freedom have only their zero-point vibrational energy—this corresponding to 4°K) and state g (the electron is in the ground state, and the ionic vibrations are excited) have been calculated. The results indicate relative probability of a single, double, triple... phonon process. This is achieved by assuming that the electron energy interval between state e and g increases, requiring the emission of one, then two, then three... phonons. The model is sufficiently simplified so that one does not need special summation procedures.^{2,3} Using the Born-Oppenheimer approximation, one may show the transition probability is governed by matrix elements of the type [reference 2 Eq. (15)]:

$$\int \xi_{0e} \xi_{ng} dX \quad \text{and} \quad \int \xi_{0e} P_X \xi_{ng} dX,$$

where X is a degree of freedom of the ion; ξ_{ne} is the vibrational eigenfunction corresponding to the X degree of freedom when the electron is in the excited state and the ion has the energy $(n + \frac{1}{2})h\nu_e$; ξ_{ng} is the corresponding function for the ground state; P_X is the

momentum operator corresponding to the X degree of freedom; and ν_e is the ionic frequency in the excited state.

One might at first expect that these matrices restrict the absorption of energy per degree of freedom to zero or one phonon. This would be true if the frequency and rest position of the ion are independent of the electron state. Recently, Kubo² has indicated how the frequency changes with the electronic state. The equilibrium position also depends on the electronic distribution. For example, calculations on the self-trapped electrons⁴ indicate that the nearest neighbors shift about 0.05 Å during the trapping process. The average zero-point displacement of an ion is about 0.04 Å. Since the rest position and frequency do depend on the electronic state, a degree of freedom can absorb more than one phonon.

One would like to know if 1 ev of energy, about 50 phonons, could be absorbed rapidly by the lattice when the electron jumps from the excited state to the ground state. Kubo's frequency shift alone does not explain Klick's observation. The equilibrium position shift, however, seems to. In our model we have artificially displaced the "central ion" (which actually does not exist in an F center) instead of the neighboring ions. A 10-phonon process is more rapid than a single-phonon process. The model does not give an absolute life time for the process. If one assumes that single-phonon processes occur in 10^{-12} sec, one would expect from the calculation that a process involving up to 50 phonons might occur more rapidly than a photon mission. As the energy gap between the e and g states increases, the probability of a multiphonon process eventually decreases, and for a large enough energy the system will emit a photon. The transition point cannot be estimated from this simple model.

Huang and Rhys's calculation of a multiphonon process disagrees with Klick's results and the calculations presented here. Due to the brevity of their paper, the author is not sure where the disagreement lies. Huang and Rhys attempt to calculate absolute rates (this, it seems, would require a more accurate model of the F center than is available at present) and use a simplified summation procedure which neglects some important terms. Pekar⁵ has calculated the Stokes' shift for Klick's experiment but he did not consider the competitive multiphonon process.

Details of the calculation will be submitted for publication in the near future. The author would like to thank Professor F. Seitz for several discussions of this problem, Dr. Klick for the detailed discussion of his results, and Mr. Clavier and Mr. Bronstein for help in the calculations.

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Existence of a 3.7×10^{-8} -sec Metastable State in Pa^{233}

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A METASTABLE state of Pa^{233} having a half-life of $3.69 \pm 0.04 \times 10^{-8}$ sec has been observed by a study of delayed conversion electrons following the alpha decay of Np^{237} .

Stilbene scintillation counters were used as detectors. After amplification with Hewlett Packard 480A wide-band amplifiers, the pulses were shaped to 0.03- μ sec duration with shorted delay lines and mixed in a coincidence circuit similar to that described by Garwin.¹ Pulse delay was accomplished by introducing measured lengths of 184-ohm cable (Transradio C.22) between the cathode follower preamplifier and the first amplifier. The velocity of propagation in this cable was found to be 0.91c (1.12m μ sec/ft) by measurement of the resonant frequencies of shorted cables. Single channel pulse-height analyzers driven by separate cathode

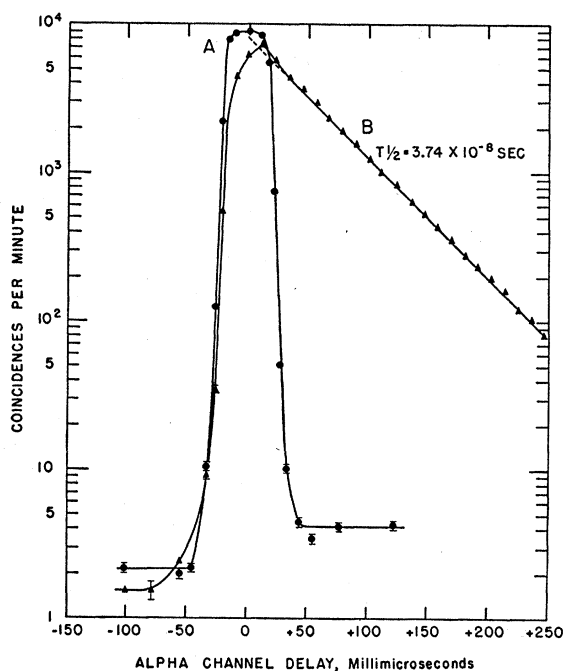


FIG. 1. Delay curve of α - e^- coincidences in U^{234} (A) and Np^{237} (B).

followers in the preamplifiers permitted the selection of alpha pulses from one detector and conversion electrons from the other. The discrimination against conversion electron pulses in the alpha detector was complete; however, some alpha pulses were counted by the conversion electron detector.

The Np^{237} samples were separated from their 27.4-day Pa^{233} daughter by solvent extraction and were evaporated onto plastic films 0.6 mg/cm² thick which were inserted between the two counters with the sample facing the conversion electron crystal. An aluminum light reflector 0.2 mg/cm² thick was located between the sample and the alpha counter. The delay curves were determined within one day after separation of the Pa^{233} .

Curve A, Fig. 1, is the delay curve for the prompt U^{234} alpha-conversion electron coincidences. The limiting slope on the positive alpha delay side corresponds to a maximum half-life of 1.4×10^{-9} sec. The delay curve for Np^{237} is given by Curve B. The slope corresponds to a half-life of 3.74×10^{-8} sec. The average half-life obtained from four such determinations was $3.69 \pm 0.04 \times 10^{-8}$ sec. The slight increase in slope near the peak of the curve is probably attributable to a small prompt coincidence contribution.

The energy of the metastable state in Pa^{233} was not determined in these measurements since the energy resolution of a stilbene counter is very poor for low energy conversion electrons. Only a single broad peak with a maximum at 30 keV and extending up to 100 keV appears in the delayed coincidence spectrum. The energy of the metastable state may be inferred from measurements of the alpha-gamma coincidence spectrum now in progress,² which indicate that the most abundant level of Pa^{233} populated by alpha decay is 87 keV above the ground state. Since few, if any, prompt coincidences or other periods are seen, the 87-keV level is probably the metastable level.

It is noteworthy that the half-life of 3.7×10^{-8} sec observed here for the metastable state of Pa^{233} is very similar to those found in Np^{237} (6.3×10^{-8} sec)³ and in Ac^{227} (4.2×10^{-8} sec),⁴ both of which are also odd- Z , odd- A nuclides. In the latter two examples, the

transition to the ground state is believed to be by electric dipole radiation.

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"Slow Beats" in Nuclear Quadrupole Induction*

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WHILE studying the behavior of spin echoes in quadrupole systems,^{1,2} a phenomenon analogous to the "slow beats" previously reported in nuclear magnetic resonance has been observed. In nuclear magnetic resonance, the term "slow beats" refers to a periodic modulation of the echo envelope, the modulation frequencies being closely related to the splitting frequencies in the hyperfine structure of the resonance.

In nuclear quadrupole induction, a similar modulation of the echo envelope appears when a small magnetic field is applied. While the explanation of nuclear magnetic resonance "slow beats" required an interaction between neighboring nuclear spins, the "slow beats" thus far observed in quadrupole induction can be explained solely in terms of the splitting and mixing of the degenerate electric quadrupole energy levels by the interaction of the nuclear magnetic moment with the external magnetic field. By means of theory outlined previously,^{1,2} it may be shown that for two rf pulses at the quadrupole resonant frequency separated by a time τ , the voltage induced in the coil at $t=2\tau$ is given by

$$V(2\tau) \propto \sin(\Omega_1 t_\omega) \sin^2\left(\frac{\Omega_1 t_\omega}{2}\right) \left\{ \frac{f^2+1}{2f^2} + \frac{f^2-1}{2f^2} [\cos(3\Omega_0\tau \cos\theta_0) + \cos(\Omega_0\tau f \cos\theta_0) + \cos(3\Omega_0\tau \cos\theta_0) \cos(\Omega_0\tau f \cos\theta_0)] \right\}$$

+ terms identified with free induction signals following each of the pulses. Here $\Omega_1 = \sqrt{3}\gamma H_1 \sin\theta_1$; $\Omega_0 = \gamma H_0$; $f = (1 + 4 \tan^2\theta_0)^{1/2}$; H_1 and H_0 are the amplitudes of the rf and static magnetic fields, respectively; θ_1 and θ_0 are the angles they make with the symmetry axis of the electric field gradient ∇E ; t_ω is the duration of the pulses; γ is the nuclear gyromagnetic ratio.

The identification of this expression as the echo was made by introducing in the theory a distribution of $|\nabla E|$ corresponding to a line width $\approx \Delta\omega$ and averaging the calculated signal over this distribution for $k \ll 2\pi/\Delta\omega$. The distribution of $|\nabla E|$ therefore has the same function in quadrupole induction as an inhomogeneous magnetic field in magnetic induction.

The term independent of τ in Eq. (1) represents the ordinary echo studied by Hahn and Herzog² in $NaClO_3$. The other terms give rise to the "slow beats" and should decay with a time constant $\approx 2\pi/\gamma H_{loc}$, where H_{loc} is the average value of the internal magnetic fields at the nuclear sites. Note that these terms vanish

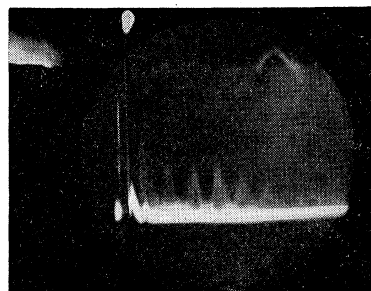


FIG. 1. Multiple exposure showing echo envelope for single crystal of $NaClO_3$. H_0 in (0,0,1) direction and equal to 15.5 gauss. Sweep length is 1500 μ sec.