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Saturation of Nuclear Electric Quadrupole Energy Levels by Ultrasonic Excitation*

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We have observed the decrease in the population difference between the degenerate $m=\pm\frac{1}{2}$ and the $m=\pm\frac{3}{2}$ quadrupole energy levels of Cl^{35} in $NaClO_3$ following a long pulse of ultrasonic excitation at the transition frequency. The experiment was performed at the temperature of liquid nitrogen, for which the transition frequency is 30.63 Mc/sec and the thermal relaxation time 0.94 sec.

The population difference was measured by the amplitude of the transient nuclear induction signal following a short (50 μsec) pulse of radio-frequency magnetic flux at the transition frequency.[1] In our experiment, the signals were induced in a second coil, a receiver coil, perpendicular to the exciting or transmitter coil, as suggested by Dean.[2] The sodium chlorate crystal, about 1 cm^3 in volume, located between the above coils, received ultrasonic energy in the (1,0,0) direction across a polished face in contact with the polished face of a long, narrow halite crystal. The latter was joined similarly to a second halite crystal, joined in turn to an X-cut quartz crystal, used as an ultrasonic transducer. Rubber-vaseline vacuum grease was used as an interface medium. The halite crystals, about 1 cm^2 in cross section, were each about 4 cm long, separating the quartz transducer from the $NaClO_3$ sample by about 8 cm. The transducer was excited by a second transmitter of variable frequency; current reached the transducer through a coaxial cable, of which the outer, grounded conductor was flared out at the end to enclose the quartz transducer completely and make contact with the silver coating of its outside face, thus preventing magnetic fields from originating from the transducer to a great extent.

The ultrasonic pulse was 0.3 second in duration; the rf power supplied to the transducer was about 5 watts. After a delay of about 0.03 second, the population difference was examined. The cycle was repeated at 1-second intervals. Depending upon a number of variables, it was observed that the amplitude of the transient was diminished to 20 percent or less of its equilibrium amplitude only when the transducer was excited at the transition frequency. To separate the ultrasonically induced quadrupole transitions from a spurious effect which would be obtained by dipole transitions caused by magnetic flux leaking into the sample region, the resonant transmitter and receiver coils were short-circuited by relays during the ultrasonic excitation period. Further, after a small gap ($\sim\frac{1}{2}$ mm) was introduced between the sample crystal and its neighboring halite crystal, interrupting the path of ultrasonic energy while providing a geometry and transducer loading for which one would expect almost identical leakage fluxes, no attenuation was discernable. A further possible spurious effect, due to the generation of a temperature gradient in the sample crystal, is not likely since the transducer heating should not be frequency-dependent; the transducer, driven at the third harmonic, tuned broadly when loaded.

Quantitative measurements are now under way. The experiment was performed in order to be able to measure the direct and Raman process contributions to the thermal relaxation time.[3]

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Experimental Evidence for Thermal Spikes in Radiation Damage*

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Theoretical work[1–3] and experimental measurements of stopping power indicate that toward the ends of their ranges in solids, energetic atoms lose $\sim 10^4$ ev to the lattice at a rate of about 10^9–10^{10} ev/cm. Brooks[4] discussed the resultant temperature fluctuations (termed thermal spikes). He showed that the thermal spikes in the electronic system are small, behave nearly independently, and cannot produce marked structural effects. For the lattice, the temperature distribution would be between cylindrical and spherical

$$T\sim 3(10^{-21})(q/c)(x^2t)^{-1}\exp(-r^2/4x^2t) \quad \text{or}$$
$$10^{-21}(Q/c)(x^2t)^{-\frac{3}{2}}\exp(-r^2/4x^2t),$$

where the symbols mean: T, temperature (°C); q, rate of energy loss (ev/cm); c, volume heat capacity (cal/cm^3); x, thermal diffusivity (cm/sec$^{\frac{1}{2}}$); t, time (sec); r, radial distance (cm); Q, total significant energy loss (ev). Thus after $\sim 10^{-11}$ sec, the thermal spike encompasses $\sim 10^4$ atoms which are experiencing a temperature rise ~ 10°C in good conductors, e.g., metals, graphite, and diamond, and several hundred degrees in intermediate conductors, e.g., quartz. Effects have been attributed[5,6] to these thermal spikes in some cases where propagation or conservation mechanisms could be invoked to permit the energy of the thermal spike to be effective before it was dissipated. However, since the displacement rates, the energy loss rates, and the proper values of the physical properties are all uncertain, it has not been possible, in general, to evaluate the contribution of the thermal spikes to the irradiation effects which are observed. In a poor conductor, like vitreous silica, the corresponding temperature rises should be several thousand degrees and hence readily observable.

Primak, Fuchs, and Day[7,8] investigated the changes in properties observed in vitreous silica and quartz irradiated simultaneously in a nuclear reactor and found that the damage could be readily followed by measuring density changes. Initially (below 2 units),[9] the density of vitreous silica increased about 0.18 percent per unit, and the density of quartz decreased about 0.07 percent per unit; by 60 units, the change in vitreous silica had saturated at 2.1 to 2.7 percent while the density of quartz had decreased about 5 percent; and by 1000 units, the quartz had become completely vitreous and indistinguishable from the irradiated vitreous silica. The density changes in quartz were followed in greater detail over the range 50–200 units by Wittels and Sherrill[10,11] who found that in this range, the dilatation of quartz accelerated to a rate 0.2 percent/unit (about 3 times as great as that observed in the early stages of irradiation by Primak, Fuchs, and Day) before it began to saturate. Such a damage-dosage curve is quite unique since most substances show a simple saturation curve for their property changes on irradiation. Since the thermal conductivity (and hence the thermal diffusivity) is changing with the other properties,[12] it seems reasonable to attribute the effect to the increasing contribution of the thermal spikes to the damaging since the slowing down processes must necessarily remain much the same. This is more reasonable than Wittels' and Sherrill's[11] mechanism, which, since the density is decreasing and the structure "opening up" on irradiation, should lead to simple saturation. Further, free interstitials do not seem to be present for an differential thermal analysis there is little or no stored energy[13] found in irradiated quartz[8] and none in irradiated vitreous silica.[14] Supporting evidence for the importance of the thermal spike mechanism of damage in disordered silica structures is the character of the annealing of irradiated vitreous silica[15] and the marked decrease of coherent x-ray scattering resulting from the irradiation of quartz and absent for some other substances (e.g., diamond and silicon carbide) which show large property changes on irradiation.[7,13]

The thermal spike mechanism may be of similar importance in the metamictization of minerals, and it may be necessary to modify the views expressed in earlier articles[7,13] accordingly.

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Superfluidity in Liquid He^3†

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In a recent note, Rice[1] has suggested that, at sufficiently low temperatures, the liquid state of the helium isotope He^3 may exhibit superfluidity. This conclusion is based on the results[2] of a pair-cell model for the liquid, which leads to a spectrum for excited states, corresponding to the hindered rotational states of a pair of atoms. In this spectrum, there is a finite energy gap between the ground and first-excited states and the presence of this gap led to the suggestion of superfluidity below some temperature lower than that corresponding to the energy gap.

It is the purpose of this note to point out that a simple extension of these considerations shows that a finite gap in the spectrum cannot be expected for a liquid of Fermi particles, so that, like the ideal gas model, the cell model actually leads one to expect the absence of superfluidity in liquid He^3.

Let us consider a pair model for a quantum liquid. If the lifetime of the rotational states is not long com-