and is at least bounded for all temperatures. The quantity $\rho_s = \rho - \rho_n$ is the superfluid mass density.

At vanishing temperature this calculation reproduces the conclusions of Gavoret and Nozières[2] who make at least the same assumption; specifically it predicts $mc_{ph}^2 = \partial p/\partial n$ and (in contrast with weakly interacting Fermi systems for which $mc_{ph}^2 \approx 3\partial p/\partial n$) merges with the hydrodynamic analysis. At vanishing temperature the chief difficulty with the assumed expansion appears to be the long-range effective potential between phonons. At finite temperatures, there appears to be additional dependence on the ratio of ω and k making the expansion assumption even more unjustifiable. However, crude estimates of the nonexpandable terms on the basis of a kinetic equation[3] at temperatures of the order of 2° suggest a behavior qualitatively similar to (3). Although this estimate is hardly reliable, it reinforces the qualitative conclusions of the simplest weak-coupling approximations at finite temperatures (also applicable for $\omega\tau \gg 1$). The consensus of these calculations is that in addition to being more significantly attenuated, $c_{ph}(T)$ should be considerably reduced from the hydrodynamic first-sound velocity, $c_1(T) \approx c_{ph}(0)$, when ρ_s is significantly less than ρ. Such a decrease in slope and broadening in the dispersion relation for the density fluctuation spectrum should be clearly visible in the inelastic scattering of neutrons with small momentum transfer ($k^{-1} \sim 10$ Å) at 2°K (since the frequency dependence of sound attenuation indicates that $\omega\tau = ck\tau \gg 1$ in this region). This behavior is also illustrated in Fig. 1.

We remark finally on the identification of $\rho_n = \rho - \rho_s$ as it occurs in Eq. (3). What actually appears is the derivative of the current with respect to an external probe that couples to the current, the condensate $\langle\psi\rangle$ remaining fixed. In a system in which there is Bose condensation (and no vortices) this derivative can be shown to equal the current produced by a transverse probe (corresponding to a rotation). The two equivalent expressions for ρ_n imply that whenever there is condensation ($\langle\psi\rangle \neq 0$), there is superfluidity ($\rho_n < \rho$). They reduce to the Landau expression[4] for ρ_n when the system can be discussed as a gas of free quasiparticles, and they are consistent with the Landau stability criterion $E(p)/p > 0$ in such a case. Finally, since they imply different transverse and longitudinal responses, they also imply the existence of infinitely long-range velocity correlations.

We have had useful conversations with C. de-Dominicis, J. Gavoret, L. P. Kadanoff, P. Nozières, D. Pines, and L. P. Pitaevskii.

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RADIATIONLESS TRANSITION RATE CONSTANT DETERMINATION FROM DELAYED FLUORESCENCE*

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Recently, delayed fluorescence in anthracene was observed after excitation with ruby laser light.[1,2] The delayed fluorescence arises from the annihilation of two triplet excitons giving a singlet exciton which then decays giving the fluorescent emission. With ruby laser light ($\lambda = 6943$ Å) the triplets are populated directly by the incident radiation. We have observed de-

layed fluorescence in anthracene single crystals by populating the first excited singlet state directly with weakly absorbed 4350Å light. Some of the excited singlets decay by a radiationless transition to the triplet state. The triplet excitons then annihilate regenerating singlet excitons which then decay giving the fluorescence. Thus, measurements on the delayed fluorescence give directly the radiationless transition rate constant.

The mm-thick single crystals used in these experiments were cut from crystals grown by the Bridgman technique from Eastman X-480 anthracene. After preliminary purification, the final zone refining and growth was done in the same inert atmosphere. A 2-μsec light pulse of about 5×10^{15} photons/cm^2 incident on the crystal was obtained from a GE BH6 mercury lamp pulsed through a mercury wetted relay and filtered by a Corning-3389 color filter. This filter passed light of wavelengths longer than 4150 Å. When a 3387 filter was used (wavelengths longer than 4550 Å), no delayed fluorescence was observed, even though the intensity is reduced by only 30%. A 1P28 photomultiplier was used to observed the delayed fluorescence. The voltage to the phototube was applied 200 μsec or longer after the incident light flash in order to prevent blocking of the photomultiplier by the ordinary fluorescence. The signal was displayed on a Tektronix-555 oscilloscope. A typical delayed fluorescence signal is shown in Fig. 1.

Since the light flash is on for only a microsecond, the initial density of triplets, n_{T0}, is

$$n_{T0} \approx k_{ST} t_f t_l \epsilon I_0, \qquad (1)$$

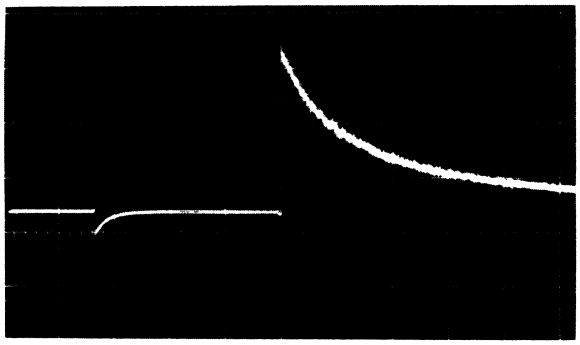

FIG. 1. Delayed fluorescence signal vs time. The time scale is 200 μsec/cm. The small pulse is a marker with the leading edge representing the time of the light flash. The signal is seen only after 680 μsec after the flash because of the gating of the photomultiplier.

where k_{ST} is the radiationless transition rate constant, ϵ is the absorption coefficient of the light; t_f is the lifetime of the excited singlet, and t_l is the time the incident light is on. The delayed fluorescence signal observed, F, is

$$F = \tfrac{1}{2}A\gamma[n_{T0}/(1+\gamma n_{T0}t)]^2, \qquad (2)$$

where A is a factor involving the geometric arrangement and the sensitivity of our equipment, and γ is the rate constant for triplet-triplet annihilation. Equation (2) holds only for times short compared with the triplet lifetime which according to Kepler[1] is 10^{-2} sec.

According to Eq. (2), a plot of $(1/F^{1/2})$ vs time gives for short times ($t<10^{-2}$ sec) a straight line with a slope of $(2\gamma/A)^{1/2}$ and whose intercept is $[n_{T0}(\tfrac{1}{2}A\gamma)^{1/2}]^{-1}$. A typical graph of our data is shown in Fig. 2.

From the ratio of the slope to the intercept, $\gamma n_{T0} = 5\times10^3$. Avakian et al.[3] and Hall, Jennings, and McClintock[2] report values of γ from 4 to 6 $\times10^{-11}$ cm^3/sec. Thus, $n_{T0} = (8 \text{ to } 12)\times10^{13}$, and from Eq. (1), $k_{ST} = 16\times10^5 - 24\times10^5$ sec^{-1} since $\epsilon = \tfrac{1}{2}$.[4] The same value of γn_{T0} was obtained from the ratio $F/(dF/dt)$.

Previous determinations of k_{ST} were made from data on fluorescence and phosphorescence efficiency by Kleinerman, Azarraga, and McGlynn.[5] Their value (10^5 sec^{-1}) is significantly smaller than ours. However, since their determinations do not take into account any mode of decay of the triplets, other than phosphorescence, their value should be viewed as a lower limit.

Finally, we looked for delayed fluorescence using strongly absorbed light ($\lambda < 4000$ Å). The

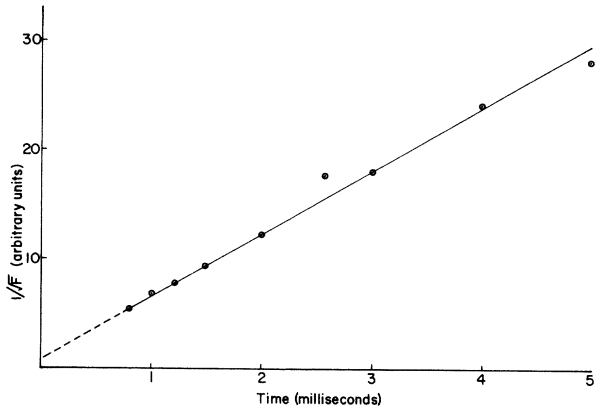

FIG. 2. Plot of reciprocal of square root of delayed fluorescence vs time.

intensity of the delayed fluorescence was considerably less ($\sim 10^{-5}$) than the value expected on the basis of a $\gamma = 5 \times 10^{-11}$. It is likely that this delayed fluorescence is due to triplets generated via bulk reabsorption of the fluorescence rather than triplet-triplet annihilation on the surface. This is consistent with Kepler's results[6] on laser-induced delayed fluorescence, which showed delayed fluorescence in crystals but not in powders where the surface-to-volume ratio is very large.

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OBLATE NUCLEAR DEFORMATIONS*

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It has been pointed out[1] that there should exist a region of nuclear deformation among neutron-deficient isotopes in the vicinity of Ba in the periodic table. We have performed calculations of nuclear shapes in this region. We find, as expected, that these nuclei have large intrinsic quadrupole moments but, in addition, these quadrupole moments are negative, i.e., the nuclei are oblate, or disc-shaped. This is an unexpected result since all strongly deformed nuclei known so far have prolate shapes, i.e., positive quadrupole moments.

Our calculations are based on the pairing-plus-quadrupole[2] model of nuclear forces. The total Hamiltonian consists of four parts: (1) A single-particle, spherical part

$$H_S = \sum_\alpha \epsilon_\alpha c_\alpha^\dagger c_\alpha,$$

in which $c_\alpha^\dagger$ creates a nucleon in state α, specified by quantum numbers $(nljm)$. The wave functions are those of an isotropic harmonic oscillator and the energies ϵ_α (independent of m) give the sequence of shell-model levels in spherical nuclei. (2) and (3) Two pairing parts with different strengths g_p and g_n, one for protons and one for neutrons, each of the form

$$H_P = -\tfrac{1}{4} g \sum_{\alpha\gamma} c_\alpha^\dagger c_{-\alpha}^\dagger c_{-\gamma} c_\gamma,$$

where $-\alpha$ is the time-reversed state of α. (4) A quadrupole part with a single strength,

$$H_Q = -\tfrac{1}{2}\chi \sum_{\alpha\beta\gamma\delta M} \langle \alpha | r^2 Y_{2M} | \gamma \rangle \langle \delta | r^2 Y_{2M} | \beta \rangle \times c_\alpha^\dagger c_\beta c_\delta^\dagger c_\gamma.$$

This Hamiltonian is treated in the Hartree-Bogolyubov approximation with a number of additional approximations, the net result being as follows.[3] First, one diagonalizes the single-particle Hamiltonian:

$$H_D = H_S - \sum_{\alpha\gamma} \langle \alpha | \sum_M D_M^* r^2 Y_{2M} | \gamma \rangle c_\alpha^\dagger c_\gamma,$$

which is essentially the Nilsson Hamiltonian,[4] for all values of the deformation parameters D_M. Then, one uses the pairing force to construct a BCS wave function, for each deformation, out of these Nilsson-like single-particle functions. Finally, one takes the expectation value of $H_S + H_P + H_Q$ for this BCS function. This is the total energy of the nucleus, which really depends on only two deformation parameters, for instance, the familiar β and γ.[5] The lowest minimum of this function, if it is at least a few MeV deep, gives the stable shape of the nucleus.

In applying this method to the region $50 < Z < 82$, $50 < N < 82$, we use essentially the ϵ_α's of Mottelson and Nilsson[6] for the protons and those of Nilsson[4] for the neutrons (see Figs. 1 and 2), except for a lowering of the $g_{9/2}$ level. We only include in the calculation the states of the two

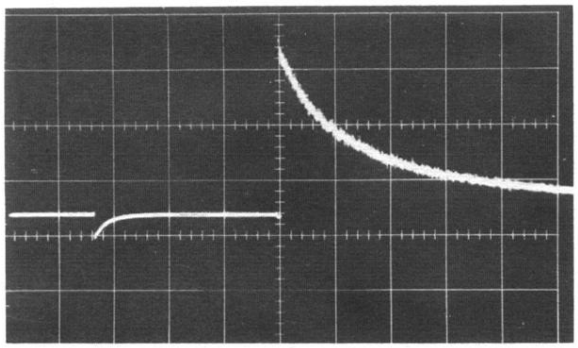

FIG. 1. Delayed fluorescence signal vs time. The time scale is 200 μsec/cm. The small pulse is a marker with the leading edge representing the time of the light flash. The signal is seen only after 680 μsec after the flash because of the gating of the photomultiplier.