High-Resolution Study of Excitations in Superfluid ^{4}He by the Neutron Spin-Echo Technique

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Neutron scattering studies of elementary excitations can now, for the first time, be extended to the microelectronvolts resolution range by the use of the newly developed neutron spin-echo method. The first such experiment is described, and its results are shown to complement substantially previous knowledge on the temperature dependence of the energy and lifetime of the roton excitation, and the suggested onset of three-phonon decay beyond the roton minimum in superfluid ^{4}He.

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Neutron spin echo[1] (NSE) represents a conceptually new approach in inelastic neutron scattering. The basic originality of the method is that it makes the energy-transfer resolution independent of the monochromatization of the incoming and outgoing beams. Thus it offers improved resolution (up to two orders of magnitude in certain cases) under good or acceptable neutron intensity conditions. NSE can most simply be applied to quasielastic scattering effects like diffusion[2] or spin relaxation.[3] The author has introduced earlier the generalized scheme for the use of NSE in the study of elementary excitations.[4] The present Letter reports the first such results.

The scheme of the experiment made at the Institut Laue-Langevin on the specially modified IN11 NSE spectrometer[5] is shown in Fig. 1. In NSE the initially polarized neutrons perform Larmore precessions during their flight through a well-defined magnetic field region both before and after the scattering. The total Larmor precession angle φ is given[1] by the difference of the outgoing and incoming precession angles:

$$\varphi = \varphi_1 - \varphi_0 = \gamma_L l H_1/V_1 - \gamma_L l H_0/V_0 = \varphi(V_0, V_1), \tag{1}$$

where $\gamma_L = 2.916$ kHz/Oe, l is the length of the field regions, H_1 and H_0 are the outgoing and incoming field strengths, v_1 and v_0 the respective velocities for the considered neutron. The basic idea of NSE is to use φ for the measurement of the neutron energy change $\hbar\omega = E_0 - E_1 \equiv \frac{1}{2}mv_0^2 - \frac{1}{2}mv_1^2 = \hbar\omega(v_0, v_1)$, while the momentum change is determined by a background spectrometer, constituted in this case essentially by the velocity selector and the graphite crystal (Fig. 1). Since φ and ω are basically different functions of v_0 and v_1, φ can only be a measure of ω locally, with respect to some average values $\bar{\varphi} = \varphi(\bar{v}_0, \bar{v}_1)$ and $\bar{\omega} = \omega(\bar{v}_0, \bar{v}_1)$,

$$\varphi - \bar{\varphi} = t(\omega - \bar{\omega}), \tag{2}$$

where t is a constant, and the bar stands for the average. [For example, when studying the roton minimum in ^{4}He, $\bar{\omega}$ will correspond to the roton energy $\Delta(T)$, i.e., to the center of gravity of the roton line.] This is the fundamental equation of NSE, which is readily found to be satisfied to first order in $\delta v_0 = v_0 - \bar{v}_0$ and $\delta v_1 = v_1 - \bar{v}_1$ if and only if

$$\hbar\gamma_L l H_0 = tm\bar{v}_0^3 \text{ and } \hbar\gamma_L l H_1 = tm\bar{v}_1^3. \tag{3}$$

In practice Eqs. (3) are used to determine the H_0/H_1 ratio, to which the spectrometer has to be set, and the parameter t. In the general case of dispersive elementary excitations we want to measure ω with respect to the dispersion relation $\omega_d(\vec{\kappa})$, where $\vec{\kappa} = m(\vec{v}_1 - \vec{v}_0)/\hbar$. Thus we will require, instead of Eq. (2), that

$$\varphi - \bar{\varphi} = t[\omega - \omega_d(\vec{\kappa})], \tag{4}$$

where, unlike Eq. (2), the right-hand side depends not only on the absolute value of v_0 and v_1, but also on their direction via $\partial\omega_d/\partial\vec{\kappa} \neq 0$. This generalized NSE condition can be met by use of precession field configurations asymmetric, "tilted" with respect to the neutron beam direction[4] (in contrast to those shown in Fig. 1), so that φ_0 and φ_1 also become dependent on the direction of $\vec{v}_0$ and $\vec{v}_1$. More details are given in Ref. 6, and this particular experiment will be described elsewhere.[7]

The conceptual difference between the classical and the NSE approach is that in the first case the measured energy transfer is given as the ference between two measured beam averages: $\frac{1}{2}m\langle\langle v_c^2\rangle\rangle - \frac{1}{2}m\langle\langle v_1^2\rangle\rangle$, whereas in NSE the beam average of a difference quantity $\langle\langle \varphi_1 - \varphi_0\rangle\rangle$ is

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measured directly.

In the *experiment* the x and y components of the precessing polarization are measured,[1] i.e., the $\langle\langle\cos\varphi\rangle\rangle$ and $\langle\langle\sin\varphi\rangle\rangle$ averages for the outgoing beam. The NSE polarization "signal" $P_{\rm NSE}$ is given as $P_0(\langle\langle\cos\varphi\rangle\rangle^2+\langle\langle\sin\varphi\rangle\rangle^2)^{1/2}$, where P_0 is the polarization efficiency of the NSE setup, and by definition $\overline{\varphi}=\arctan(\langle\langle\sin\varphi\rangle\rangle/\langle\langle\cos\varphi\rangle\rangle)\pm 2\pi n$. In practice it is possible but tedious to measure H_0 and H_1 sufficiently precisely for $\overline{\varphi}$ to be given an absolute, rather than a relative, meaning. For a Lorentzian phonon line given by the scattering function $S(\vec{\kappa},\omega)=\gamma/\{\gamma^2+[\omega-\omega_d(\vec{\kappa})]^2\}\pi$, we obtain by Eq. (4)

$$P_{\rm NSE}=P_0\int S(\vec{\kappa},\omega)\cos[t(\omega-\omega_d)]\,d\omega$$
$$=P_0\exp(-\gamma t), \qquad (5)$$

where the integration extends over the transmission function ("resolution ellipsoid") of the background spectrometer, which should be broad compared with γ (i.e., the integration can be taken from $-\infty$ to ∞) if high NSE resolution is required. In addition, a relative change in the phonon energy $\delta\omega_d$ shows up as a change in the phase angle $\delta\overline{\varphi}=t\delta\omega_d$. In the experiment $P_{\rm NSE}$ was measured versus the time parameter[1] $t\propto H_0$ at constant H_0/H_1 [cf. Eqs. (3)], and γ was determined from Eq. (5) which is represented by straight lines on a semilog plot, cf. the inset in Fig. 1. The sample NSE spectra shown for the roton are clearly consistent with Lorentzian line shape.

The raw neutron data have been corrected for background, whereas the broad multiphonon and multiple-scattering spectra cannot contribute to the finely tuned NSE signal. The spectrometer configuration shown in Fig. 1 determines the momentum transfer of the excitation with ± 0.02 Å^{-1} resolution. Parallel with the experiment Monte Carlo simulation calculations were performed in order to study the possible spurious effects due to finite momentum resolution, etc. It has been found that these effects are less than the statistical errors if the momentum of the roton minimum and the single-to-multiple phonon scattering intensity ratio do not change more between 1 and 1.4 K than ± 0.02 Å^{-1} and $\pm 15\%$, respectively. These requirements are known to be largely met.[8,9]

In this first NSE experiment of this kind a few phonon groups were studied in superfluid ^{4}He. The results obtained for the temperature dependence of the linewidth $\gamma(T)$ and of the energy $\Delta(T)$ of the roton excitation are shown in Fig. 2 together with results of previous studies. The previous, classical neutron scattering experiments were limited by insufficient resolution to temperatures where $2\gamma(T)$ and $\Delta(T)\sim 8$ K are comparable and thus the final results strongly depend

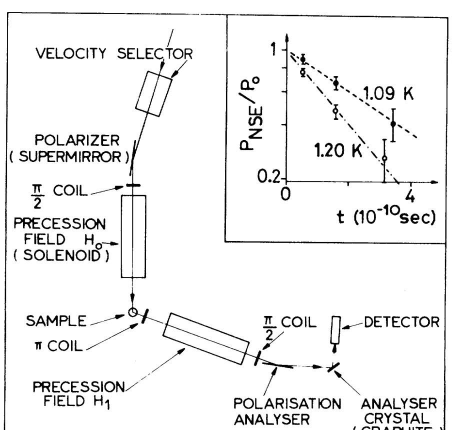

FIG. 1. Scheme of NSE experiment for the study of the roton excitation in superfluid ^{4}He. Sample NSE spectra for $T=1.09$ and 1.20 K are shown in the inset, where the straight lines correspond to Lorentzian fits with $\gamma=20$ and 34 mK, respectively.

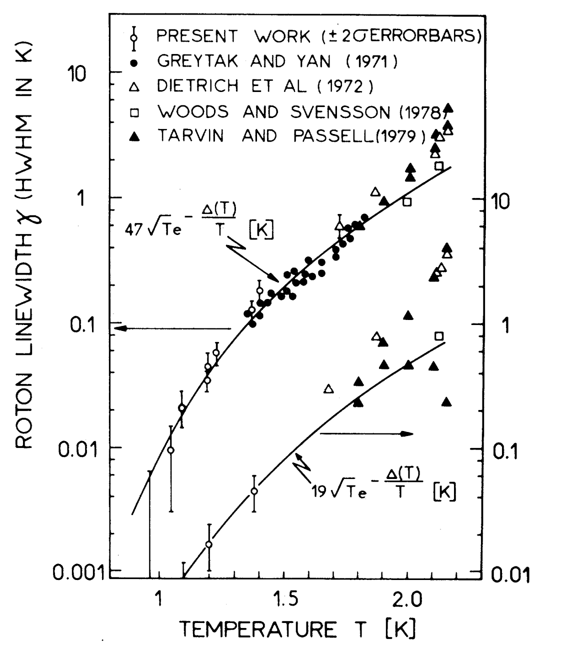

FIG. 2. Comparison of the present results on the temperature dependence of the roton energy and linewidth with previous Raman-scattering (Ref. 8) and neutron-scattering (Refs. 9–11) data, and with theoretical predictions.

on the choice of the model scattering function to which the spectra are fitted. This is the reason for the scatter of these results, as was clearly shown in Ref. 10 in which the two data points shown in Fig. 2 at each temperature were obtained by use of two different, plausible mathematical expressions for the line shape. The temperature range where $\gamma(T) \ll \Delta(T)$, and this inherent ambiguity is fully removed, could only be investigated for the first time in the present work. As can be seen in the figure, this is particularly essential for the determination of $\Delta(0)-\Delta(T)$. On the other hand it is remarkable that the Raman-scattering results[11] (dots), obtained indirectly by the use of the two-roton bound-state theory,[12] are consistent with the direct neutron-scattering observations. At temperatures below 1.1 K, however, the decay of the bound roton pair was shown[13] to be dominated by photon processes not available to the single roton. The most direct evidence for this is given by the low values of γ found in the present work around 1 K, which are indeed well below 0.027 K, the value which the $T = 0.6$ K Raman-scattering data[14] would imply (not shown in the figure).

The first–Born-approximation theory of the roton-roton interaction by Landau and Khalatnikov[15] (LK) predicts that $\gamma(T) = 47\sqrt{T}\exp[-\Delta(T)/T]$, where all quantities are measured in kelvins, and the coefficient 47 was calculated from normal-fluid viscosity data. Furthermore, it has been pointed out that within the LK framework there is a roton-roton contribution to $\Delta(T)$ too, which has the same temperature dependence. A fit to the present NSE results then gives $\Delta(0)-\Delta(T) = 19\sqrt{T}\times\exp[-\Delta(T)/T]$, while about twice as much obtained by fitting the higher-temperature neutron data[17] of Ref. 9. (As a matter of fact 0.96 K was the lowest T in this experiment, and the extrapolation to $T=0$ was made with use of this expression.) The values for $\Delta(T)$ thus obtained with the accepted value of $\Delta(0) = 8.62$ K[18] were in turn used for the calculation of the LK prediction for $\gamma(T)$ (upper curve in Fig. 2). The excellent absolute agreement over more than two orders of magnitude in γ, without any fitted parameter, indicates that the roton lifetime is indeed dominated by the roton-roton processes for the temperatures shown, with possible deviations between 2 K and the λ point. However, the magnitude of $\gamma(T)$ turns out to be much too big for the first Born approximation to be applied, and actually it is about four times bigger than the upper (unitarity) limit for single-channel scattering processes, brought about by the final-state interactions. Interestingly, the present new values for $\Delta(0)-\Delta(T)$ also turn out to be about four times bigger than the same type of limit.[17] Furthermore, within their range of validity, the Born and Hartree-Fock (HF) approximations predict that $[\Delta(0)-\Delta(T)] \gtrsim \gamma(T)$, in strong contrast to the present results. This is additional evidence that a strong-coupling theory has to be used. Following Fomin's suggestion[12] of several helicity channels, the present $\gamma(T)$ and $\Delta(0)-\Delta(T)$ data, and in particular their ratio, can be explained by a minimum of seven scattering channels with an average effective coupling constant $g_4 = -1.2\times10^{-38}$ erg cm^3, if for each channel $-10^{-38} \gtrsim g_4 \gtrsim -1.8\times10^{-38}$ erg cm^3. It is worth noticing that the $T > 1.8$ K data of Ref. 10 agree well with the present theoretical curves. This would lend support to the interpretation of the scattering spectra proposed in this Ref. 10, if in addition one could assume that both $\gamma(T)$ and $\Delta(0)-\Delta(T)$ are dominated by the roton-roton processes in the entire temperature range above 1 K, which is by no means obvious as yet.

In addition I have determined at a few temperatures the excitation linewidth at $\kappa = 1.08$, 1.72, and 2.1 Å^{-1} with 10 mK resolution, and, as in Ref. 10, excellent agreement was found with the above LK expression derived originally for the roton.

Another high-resolution problem has been touched on in this experiment, the suggested onset of phonon emission decay for phonons with momenta above 2.1–2.2 Å^{-1} (Pitaevski broadening). Similarly to earlier results[19] and in spite of the considerably improved resolution, no such effect was found. The present 70%-confidence-level upper limits for the decay rate at 1 K which I have been able to establish are 10, 20, 20, and 10 mK at 2.1, 2.2, 2.3, and 2.4 Å^{-1}, respectively. Following the analysis of Jäckle and Kehr[20] this indicates that the presumed linear section of the dispersion curve, which should start at $\kappa = 2.25$ Å^{-1}, ends below 2.4 Å^{-1}, if it exists at all.

The present experiment is the first in which energy resolutions around 1 μeV ~ 10 mK (1 part in 10^3 in energy transfer) have been attained in neutron-scattering study of elementary excitations. For the investigated cases this means a 20–40-fold improvement compared with classical methods, which should be typical for the new method, as was shown by the Monte Carlo simulation calculations I have performed to assess the higher-order terms neglected in Eq. (4). The NSE has, in addition, the interesting feature that it is selective for the slope of the dispersion curve.[4] In the present case this high resolution made possible the investigation of the temperature dependence of the roton energy and linewidth at low enough temperatures, where the linewidth is much less then the energy. This was earlier found to be a necessary condition for unambiguous data reduction, and it is in fact assumed in the existing theories. Thus the present work is the first really meaningful direct neutron-scattering study of this problem.

It appears to me that the present results convincingly show both the feasibility of the NSE method for the high-resolution study of elementary excitations (lifetime and energy-shift effects, crossing branches and hybridization, etc.) and the physical interest of such experiments.

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Raman Measurement of Lattice Temperature during Pulsed Laser Heating of Silicon

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The ratio of Stokes–to–anti-Stokes phonon Raman scattering of a probe laser pulse at 405 nm is used to obtain a direct measure of the lattice temperature in silicon within 10 nsec after a heating pulse at 485 nm. A lattice temperature rise of only 300°C is found with a heat pulse power density of ~1 J/cm^2. Since silicon melts at 1412°C, this result is direct evidence that strictly thermal melting models of laser annealing are inappropriate.

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Pulsed laser annealing has recently shown great promise for repairing the damage introduced by ion-implantation doping of semiconductors.[1,2] However, the mechanisms responsible for annealing a micron thick layer on time scales of tens of nanoseconds are not well understood. Strictly thermal models of the annealing can begin to explain the processes only if the semiconductor is assumed to melt.[2,3] Alternative non-thermal-equilibrium models,[1,4-7] on the other hand,

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