them are still degenerate) of the manifold that are infrared active. The splitting is proportional to the quadrupole moment and the relative intensity is 3 to 2 for the doublet.[18] Calculations also show that the origin of the doublet should be at six-tenths of the splitting from the low-energy[11] component of the doublet. The first-order theoretical value for this splitting derived from the theoretical quadrupole moment is 8.17 cm^{-1},[19] whereas our observed value is 6.30 cm^{-1}. The discrepancy is believed to arise from the simplification of intermolecular forces in the calculations where the contribution from zero-point motion, many-body effects, charge overlap, and the dispersion-induced forces have been ignored.

In conclusion, we have reported the first observation of the $\Delta\nu = 4$ vibrational-rotational molecular H_2 transitions in solid hcp p-H_2. The data provide much needed information on the long sought after third vibrational overtone of H_2 which may have significant impact on the interpretation of some of the unidentified features in the optical spectra of Neptune and Uranus. Equally important are the observations of the frequencies, widths, and strengths of the spectra which should stimulate extensive theoretical work on this simplest of molecular solids.

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Coupling of Spin Waves with Zero Sound in Normal ^{3}He

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A scheme is proposed whereby a coupling can be achieved between the spin-wave and zero-sound modes in a neutral Fermi liquid; the technique should permit a detailed study of the spin-wave spectrum.

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Spin waves do not propagate in normal ^{3}He (Ref. 1); however, Silin[2] showed that in the presence of a finite magnetic field such modes do exist at sufficiently long wavelengths. The so-called Leggett-Rice effect[3] implies the existence of spin waves.[4] However, a systematic technique to directly study spin-wave dispersion has been absent to date.

In this Letter I propose a method to study spin waves, in detail, by observing the coupling to zero sound occurring near a special type of mode crossing. Under ordinary circumstances there is no coupling between sound and the propagating (transverse) components of the spin density. However, suppose the static magnetization is tipped by an angle θ_H from the magnetic field direction $\hat{H}$ by the application of a suitable rf (H_1) pulse. (As we shall see, θ_H cannot be 90° since spin waves will not propagate under this circumstance.[3,4]) This sets up a precessing uniform magnetization at the Larmor frequency, $\Omega_0 = -\beta H/\hbar$, which would decay in a time T_1; this time is sufficiently long that many acoustic pulses could be propagated, thus allowing an accurate determination of any change in the zero-sound propagation characteristics. If sound with a frequency ω and wave vector k is propagated, the Silin equations (to be discussed shortly) show that various beat frequencies are generated, in particular $\omega - \Omega_0$. If the magnetic field is adjusted such that $\omega(k) = \Omega(K) + \Omega_0$, a resonant transfer of energy from the sound to the spin wave will occur, provided the spin-wave vector satisfies $K = k$; this situation is shown in Fig. 1. One may alternatively view the effect as a three-particle scattering event: The tipped magnetization fills the liquid with $K=0$ magnons (spin waves), which cause stimulated decay of the zero-sound phonons to create a final-state magnon with $K \neq 0$ in an energy- and momentum-conserving manner. The actual coupling comes about because of the energy dependence of the density of states (so-called particle-hole asymmetry). By study of various transducer harmonics the spin-wave spectrum can be determined at a discrete number of points. I note that multiple spin modes

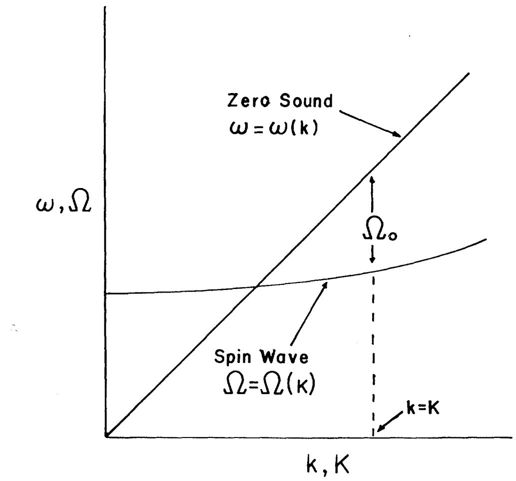

FIG. 1. Schematic dispersion relations for the coupled spin-wave (Ω)—zero-sound (ω) modes in the presence of a uniform precessing magnitization (Ω_0); a more general case where $\Omega(0) \neq \Omega_0$ is depicted.

($l \neq 0$) are expected if the quasiparticle interaction function has sufficient structure. For longitudinal zero sound, coupling is limited to $m=0$ modes; transverse zero sound would couple to $m = \pm 1$ modes. Here l and m denote spherical harmonics characterizing the Fermi-surface disturbance associated with the collective modes.

We write for the spin-density matrix, $\underline{n}$, and the "Hamiltonian," $\underline{\epsilon}$,

$$\underline{n} = n(p)\underline{1} + \vec{m}(\vec{p}) \cdot \underline{\vec{\sigma}} \tag{1}$$

and

$$\underline{\epsilon} = \epsilon(p)\underline{1} + \vec{h}(\vec{p}) \cdot \underline{\vec{\sigma}}\,. \tag{2}$$

The variation of the energy has the form

$$\delta\underline{\epsilon} = \delta\underline{\epsilon}_{\text{ext}} + \tfrac{1}{4}\operatorname{Tr}\int d\tau' [f^s(\vec{p},\vec{p}\,')\underline{1}\,\underline{1}' + f^a(\vec{p},\vec{p}\,')\underline{\vec{\sigma}}\cdot\underline{\vec{\sigma}}'\,]\,\delta\underline{n}'(p')\,. \tag{3}$$

One can perform a Galilean invariance argument where δn and $\delta\vec{m}$ are separately varied. The variation δn leads to the usual effective-mass sum rule; the variation of $\delta\vec{m}$ leads to the relation

$$\partial h_j/\partial p_i \equiv V_{ji} = \int d\tau' f^a(\vec{p},\vec{p}\,')\,\partial m_j/\partial p_i\,, \tag{4}$$

where to lowest order $|\vec{m}|$ is the magnetization $|\vec{m}_0|$ and makes an angle θ_H with H after application of the rf pulse. Defining $F^a = 2[4\pi p^2/(2\pi\hbar)^3 v] \times f^a = N(\epsilon)f^a$ and similarly for F^s, and carrying out the integrations we find $V_{ji} = \frac{1}{6}\hat{m}_{0j}\hat{p}_i[\partial(F_1{}^a v)/\partial\epsilon]\gamma H$, where $\gamma \equiv \beta/(1+F_0{}^a)$, and $F_i{}^a$ ($F_i{}^s$) are coefficients in a Legendre-polynomial expansion of F^a (F^s). We also find $\vec{m}_0(p) = -\frac{1}{2}\gamma H(\partial n_0/\partial\epsilon)\hat{m}_0$ and $\partial n_{0j}/\partial p_i = -\hat{m}_{0j}\cdot\hat{p}_i(\gamma H/2)(\partial^2 n_0/\partial\epsilon^2)v$.

If we write $n = n_0 + n'$ and $\vec{m} = \vec{m}_0 + \vec{m}'$ where n' and $\vec{m}'$ are the deviations from equilibrium, the linearized Silin-Landau transport equations may be written

$$\frac{\partial n'}{\partial t} + \frac{\partial n'}{\partial r_i}v_i - \frac{\partial n_0}{\partial p_i}\frac{\partial}{\partial r_i}\int d\tau' f^s(\vec{p},\vec{p}\,')n'(\vec{p}\,') + \frac{\partial m_{0j}{}'}{\partial r_i}V_{ji} - \frac{\partial m_{0j}}{\partial p_i}\frac{\partial}{\partial r_i}\int d\tau' f^a(\vec{p},\vec{p}\,')m_j{}'(\vec{p}\,') = 0\,, \tag{5}$$

and

$$\frac{\partial m_j'}{\partial t}+\frac{\partial m_j'}{\partial r_i}v_i-\frac{\partial n_0}{\partial p_i}\frac{\partial}{\partial r_i}\int d\tau' f^a(\vec{p},\vec{p}')m_j'(\vec{p}')+\frac{\partial n'}{\partial r_i}V_{ji}-\frac{\partial m_{0j}}{\partial p_i}\frac{\partial}{\partial r_i}\int d\tau' f^s(\vec{p},\vec{p}')n'(\vec{p}')$$
$$-\frac{\beta}{\hbar}(\vec{m}'\times\vec{H}_0)_j-\frac{2}{\hbar}\int d\tau' f^a(\vec{p},\vec{p}')\{[\vec{m}_0(\vec{p})\times\vec{m}'(\vec{p}')]_j+[\vec{m}'(\vec{p})\times\vec{m}_0(\vec{p}')]_j\}=0. \tag{6}$$

Here we have not included the collision integral which restricts the treatment to $T=0$. We may write $\vec{m}_0=\vec{m}_{0\perp}+m_{0z}\hat{z}$, where

$$\vec{m}_{0\perp}=m_{0\perp}\left[\left(\frac{e^{i\Omega_0 t}+e^{-i\Omega_0 t}}{2}\right)\hat{x}+\left(\frac{e^{i\Omega_0 t}-e^{-i\Omega_0 t}}{2i}\right)\hat{y}\right], \tag{7}$$

with $m_{0z}=m_0\cos\theta_H$, $m_{0\perp}=m_0\sin\theta_H$, and m_0 is the equilibrium magnetization $(=|\vec{m}_0|)$. We look for solutions satisfying the conditions $\omega(k)=\Omega(k)+\Omega_0$ and we neglect all other terms not having this common time dependence. Writing $m_+'=m_x'+im_y'$, $n'(\vec{p})=\nu(\theta,\varphi)\partial n_0/\partial\epsilon$, $m_+'(\vec{p})=\mu(\theta,\varphi)\partial n_0/\partial\epsilon$, and defining $a=-\frac{1}{4}\gamma H v^{-1}\partial v/\partial\epsilon$ and $b=\frac{1}{12}\gamma H v^{-1}\partial(F_1{}^a v)/\partial\epsilon$ and using $\gamma/\beta=(1+F_0{}^a)^{-1}$, we obtain after some calculation

$$(kv\cos\theta-\omega)\nu(\theta)-kv\cos\theta\tfrac{1}{2}\int_0^\pi F^s(\cos\chi)\nu(\theta')\sin\theta' d\theta'$$
$$+bkv\cos\theta\sin\theta_H\mu(\theta)-akv\cos\theta\sin\theta_H\tfrac{1}{2}\int_0^\pi F^a(\cos\chi)\mu(\theta')\sin\theta' d\theta'=0 \tag{8}$$

and

$$\left[kv\cos\theta-\Omega+\Omega_0\left(1-\frac{F_0{}^a}{1+F_0{}^a}\cos\theta_H\right)\right]\mu(\theta)-\left(kv\cos\theta+\frac{\Omega_0}{1+F_0{}^a}\cos\theta_H\right)\tfrac{1}{2}\int_0^\pi F^a(\cos\chi)\mu(\theta')\sin\theta' d\theta'$$
$$+2bkv\cos\theta\sin\theta_H\nu(\theta)-2akv\cos\theta\sin\theta_H\tfrac{1}{2}\int_0^\pi F^s(\cos\chi)\nu(\theta')\sin\theta' d\theta'=0. \tag{9}$$

Here χ is the angle between $\hat{p}$ and $\hat{p}'$. In the above we have restricted ourselves to the case of symmetrical $(m=0)$ modes only. To solve this set of coupled integral equations we expand $\nu(\theta)$ and $\mu(\theta)$ in Legendre polynomials[5] $\nu(\theta)=\sum_l \nu_l P_l(\cos\theta)$ and $\mu(\theta)=\sum_l \mu_l P_l(\cos\theta)$. We define

$$s_1=\omega/kv,\quad s_2=\frac{\Omega-\Omega_0\{1-[F_0{}^a/(1+F_0{}^a)]\cos\theta_H\}}{kv},\quad x=\cos\theta$$

and introduce the integrals[5]

$$I_{ll'}{}^{(n)}=\frac{1}{2}\int_{-1}^{+1}P_l(x)\frac{x^n}{x-s}P_{l'}(x)\,dx. \tag{10}$$

With these definitions and the fact that from the addition theorem of spherical harmonics,

$$\tfrac{1}{2}\int F^s(\chi)\nu(x')dx'=-\sum_l\frac{1}{2l+1}F_{l'}{}^s P_{l'}(x)\nu_{l'}(x),$$

Eqs. (8) and (9) become

$$\frac{\nu_l}{2l+1}+\sum_{l'}I_{ll'}{}^{(1)}(s_1)F_{l'}{}^s\frac{\nu_{l'}}{2l'+1}+b\sin\theta_H\sum_{l'}I_{ll'}{}^{(1)}(s_1)\mu_{l'}+a\sin\theta_H\sum_{l'}I_{ll'}{}^{(1)}(s_1)F_{l'}{}^a\frac{\mu_{l'}}{2l+1}=0 \tag{11}$$

and

$$\frac{\mu_l}{2l+1}+\sum_{l'}\left[I_{ll'}{}^{(1)}(s_2)+\frac{1}{1+F_0{}^a}\frac{\Omega_0}{kv}\cos\theta_H I_{ll'}{}^{(0)}(s_2)\right]F_{l'}{}^a\frac{\mu_{l'}}{2l'+1}$$
$$+2b\sin\theta_H\sum_{l'}I_{ll'}{}^{(1)}(s_2)+2a\sin\theta_H\sum_{l'}I_{ll'}{}^{(1)}(s_2)F_{l'}{}^s(s_2)\frac{\mu_{l'}}{2l'+1}=0. \tag{12}$$

For simplicity we restrict our discussion to the case $F^s(\chi)=F_0{}^s$ and $F^a(\chi)=F_0{}^a$. In this approximation we have a 2×2 problem; setting the determinant equal to zero we obtain

$$G_1(s_1)G_2(s_2)-\Gamma\Gamma'/s_1{}^2s_2{}^2=0, \tag{13}$$

where

$$G_1(s_1) \equiv 1 + I_{00}{}^{(1)}(s_1) F_0{}^s, \tag{14}$$

$$G_2(s_2) \equiv 1 + I_{00}{}^{(1)}(s_2) F_0{}^a + \frac{F_0{}^a}{1+F^a}\,\frac{\Omega_0 \cos\theta_H}{kv}\, I_{00}{}^{(0)}(s_2), \tag{15}$$

$$I_{00}{}^{(1)}(s) = 1 + \frac{s}{2}\ln\frac{s-1}{s+1} = -\frac{1}{3s^2} - \frac{1}{5s^4} - \frac{1}{7s^6}\cdots \quad (s \gg 1), \tag{16}$$

and

$$I_{00}{}^{(0)}(s) = \frac{1}{2}\ln\frac{s-1}{s+1} = -\frac{1}{s} - \frac{1}{3s^3} - \frac{1}{5s^5}\cdots \quad (s \gg 1), \tag{17}$$

and where $\Gamma \equiv 2\sin\theta_H(-aF_0{}^s + b)$ and $\Gamma' \equiv \sin\theta_H \times(-aF_0{}^s + b)$. In the absence of coupling the normal-mode frequencies are given by $G_1(s_1) = 0$ and $G_2(s_2) = 0$. To lowest order[6] $\omega = kv(F_0/3)^{1/2}$ and

$$\Omega = \Omega_0\left[1 + \frac{1}{3}\,\frac{(1+F_0{}^a)^2}{F_0{}^a\cos\theta_H}\left(\frac{kv}{\Omega_0}\right)^2\right];$$

note the latter expansion is valid only for sufficiently large values of $\cos\theta_H$ and Ω_0. Note from Eq. (15) that if $\theta_H = 90°$ the dispersion relation is identical to that of zero sound. However, no propagating solutions exist in this case since $F_0{}^a < 0$.[7]

Far off the mode crossing the frequency on one branch approximately satisfies the equation $G_i(s_i) = 0$ and we may expand to first order in $s_i - s_{i0}$:

$$G_i(s_i) = G_i(s_{i0}) + (s_i - s_{i0})\,\partial G_i/\partial s_i, \tag{18}$$

where $G_i(s_{i0}) = 0$; for the other branch G_i will in general differ greatly from zero. Confining ourselves to the sound branch only we find

$$\delta s_1 \equiv s_1 - s_{10} = \frac{\Gamma\Gamma'}{s_{10}{}^2 s_{20}{}^2 G(s_2)\,\partial G_1/\partial s_1}. \tag{19}$$

This shift is second order in $\gamma H/\epsilon_F$ and would be difficult to observe.

Near the mode crossing we expand both G_1 and G_2 and, since both of these functions vanish at the uncoupled degeneracy point, we have

$$\delta s_1 \delta s_2 = \frac{\omega\Gamma\Gamma'(1+F_0{}^a)}{\Omega_0 F_0{}^s F_0{}^a \cos\theta_H}. \tag{20}$$

Collecting factors we have

$$\delta\omega^2 = \tfrac{3}{8}\omega^2\,\frac{4\sin^2\theta_H(-aF_0{}^s + b)(-aF_0{}^a + b)(1+F_0{}^a)}{F_0{}^s F_0{}^a \cos\theta_H}, \tag{21}$$

where from before $a \equiv -\frac{1}{4}\gamma H v^{-1}\,\partial v/\partial\epsilon$ and $b \equiv \frac{1}{12}\gamma H \times v^{-1}\partial(F_1{}^a v)/\partial\epsilon$. From Eq. (21) we see that the splitting $\delta\omega/\omega$ is *first* order in $\gamma H/\epsilon_F$; for a field of ~20 kG this ratio is of order 10^{-4}. As a measure of the ease of detection of the above phenomenon I note that the coupling strength of zero sound to the real squashing mode in the superfluid B phase is of a similar order[8,9] (both arising from particle-hole asymmetry). At low temperatures the effect of collisions can be qualitatively incorporated by writing $\omega + i/\tau_1$ and $\Omega + i/\tau_2$ for ω and Ω, respectively, in Eq. (21), where τ_1 and τ_2 are the collision times appropriate to zero sound and spin waves, respectively.

Finally I note the possibility that various "echo" phenomena may be observable; however this is yet to be investigated in detail.

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[6] Terminating the expansion at F_0^s and F_0^a limits the accuracy of the resulting dispersion relations; more accurate expressions can be found in Refs. 4 and 5. Here the aim has been to examine the coupling between the modes. I also note that the expression for $\Omega(k)$ breaks down as $H \to 0$.

[7] The fact that there is no coupling for $\theta_H = 0$ is a consequence of symmetry; that the dispersion relation for $\theta_H = \frac{1}{2}\pi$ reduces to that for longitudinal spin waves

is a trivial consequence of geometry. The lack of coupling at $k=0$ in (8) and (9) is due to particle and spin conservation.

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Kinetics of the Q-State Potts Model in Two Dimensions

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An efficient Monte Carlo procedure is applied to the study of the kinetics of low- and high-Q-state Potts models quenched from an initial high-temperature ($T \gg T_c$) state to very low temperatures ($T \simeq 0$). After an initial transient period, the mean domain size, R, increases algebraically with time as $R \sim Ct^n$. The exponent n decreases from $\frac{1}{2}$ for $Q=2$ (Ising model) to 0.38 for large Q. The change in n is attributed to a coalescence process which becomes increasingly effective with decreasing Q. For large Q, the prefactor C is proportional to $Q^{1/2}$.

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The kinetics of domain growth is a subject of considerable interest in the fields of surface science[1,2] and metallurgy.[3] Both phenomenological[4-8] and field-theoretic approaches[9] have been developed to explain the growth of domains in the ordering of alloys (e.g., Fe-Al, Cu-Au, etc.) with two equivalent sublattices quenched from high to low temperatures ($T < T_c$). The kinetics of domain growth has also been studied in binary alloys by Monte Carlo (MC) techniques with a simple Ising model.[10,11] All of the above investigations showed that the characteristic length R of a domain grows (or shrinks) algebraically with time [i.e., $R(t) \sim t^{1/2}$]. This growth law has been experimentally observed in various alloys.[9] In recent years, the kinetics of domain (or island) growth of adsorbed atoms on surfaces with more than two degenerate ground states has also been investigated. In a number of MC studies on chemisorption[1,12,13] and physisorption systems, domains of multiply degenerate ground states have been reported to grow very slowly. Analytical results on simple domain geometries have also suggested that the characteristic domain size grows slowly, as a logarithmic function of time.[6,14] A similar slow evolution has been observed by Lagally *et al.*[2] for ordering of oxygen atoms on a tungsten substrate [O/W(110)]. However, it is difficult to conclude from the experimental studies whether this slow growth is due to surface heterogeneities (steps, terraces, vacancies, etc.) or to more basic topological effects. A similar situation is encountered in the experimental study of grain growth in polycrystalline materials.[15]

In this Letter, we report the results of our computer simulations on the kinetics of a ferromagnetic Q-state Potts model which is rapidly quenched from $T \gg T_c$ to $T \simeq 0$. For low Q ($Q=2,3,4,6$), Potts models provide a good approximation to many chemisorption and physisorption systems, while for high Q, this model can be used to study the kinetics of continuous (infinitely degenerate) systems such as grain growth in a polycrystalline material.

We study the Q-component ferromagnetic Potts model,

$$H = -J \sum_{\mathrm{NN}} \delta_{S_i, S_j}, \tag{1}$$

where S_i is the Q state of the spin on site i ($1 \leq S_i \leq Q$) and δ_{S_i,S_j} is the Kronecker delta. The sum is taken over nearest-neighbor spins and $J > 0$. Using MC techniques, we study the domain growth of systems originally quenched from a high temperature to a very low one. To reduce the boundary effects, we employ very large systems (200×200 sites on a triangular lattice) with periodic boundary conditions. Standard MC procedures, in which a randomly chosen spin is allowed to flip into any of the $Q-1$ other orientations, was found to be very inefficient since, for large Q, the probability of acceptance of an arbitrary spin flip is very small. We have employed a variant[16] of the efficient MC procedure of Bortz

