Brewer and Edwards is in fact the critical velocity of the modified Vinen theory. This interpretation of the data is completely consistent with previous work on the normal-fluid turbulent transition in He II[5] and should provide a proper framework for further work in the area. We emphasize that the critical relative velocity of the modified Vinen theory is one of the very few critical-velocity phenomena in He II for which there is a satisfactory theory.

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Short-Wavelength Collective Excitations in Liquid and Solid Hydrogen

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Intermolecular and second-order light scattering in the condensed phases of hydrogen at para concentrations ranging from 25% to 96% have demonstrated the existence of underdamped short-wavelength collective excitations in the liquid phase over this concentration range and their relation to phonon excitations in the solid.

Recent studies of the rare-gas solids and liquids have established the utility of higher-order inelastic light-scattering processes for the investigation of short-time, short-wavelength lattice and molecular dynamics.[1-4] For example, second-order Raman scattering in the solid and intermolecular light scattering in the liquid have been applied to all the rare-gas materials in order to study the density and temperature evolution of excitations having wavelengths on the order of interparticle separation.[2,4] The behavior of these excitations upon melting has thus been explored thoroughly.[2,4] Thus far, with the exception of helium, all liquids have exhibited intermolecular spectra which are centered at zero frequency shift and decay essentially exponentially in frequency $[I(\nu) = I_0 e^{-\nu/\Delta}]$ at a characteristic rate Δ which depends simply on density, temperature, and the parameters of the intermolecular potential.[4,5] This behavior is consistent with the interpretation that the scattering occurs from pairs of overdamped density fluctuations whose individual wave vectors are essentially equal and opposite. Indeed the intermolecular ("two-roton") spectrum in liquid He can be semiquantitatively explained as arising from the same process where the excitations are underdamped.[1,6] Until quite recently the only liquid known to exhibit long-lived short-wavelength excitations was He, and consequently it was expected to be the only liquid to exhibit a nonexponential intermolecular light-scattering spectrum. Recent inelastic neutron studies[7] have established strikingly the existence of relatively well-defined density fluctuations of very short wavelength ($K \simeq 1-2$ Å^{-1}) in pure parahydrogen ($J=0$). However, because of the large incoherent scattering from the orthohydrogen ($J=1$) species, a study of the concentration dependence of these modes—particularly their observation in normal hydrogen (75% ortho)—was not possible by neutron scattering.

In this Letter we report a variety of new aspects to the light-scattering spectra in both liquid and solid hydrogen over a concentration range between 25% and 96% parahydrogen at temperatures between 9 and 23°K. Our observations include (1) a markedly nonexponential intermolecular spectrum in liquid hydrogen, which becomes increasingly obscured by anisotropy or "Rayleigh-wing" scattering as the ortho concentration is increased; (2) two-phonon Raman scattering in solid hydrogen; (3) phonon sidebands on both the $S_0(0)$ and $S_0(1)$ rotational transitions in the solid, and their changes upon melting; and (4) the dependence of (1) and (3) upon ortho-para concentration.

Experiments were performed using a system previously described,[4] with the exception that a new sample cell with improved window configuration to minimize fluorescence was employed for most of the liquid measurements. All spectra were obtained using a 4880-Å argon laser with typically 200 and 40 mW, respectively, for liquid and solid runs. Temperature was monitored with a calibrated carbon resistor and was accurate to better than 0.1°K.

Parahydrogen was prepared by liquefying normal hydrogen in the presence of a ferrous-oxide catalyst. Concentration of the final sample was conveniently measured *in situ* spectroscopically: $R=\frac{5}{3}C/(1-C)$, where C is the para concentration and R the ratio of $S_0(0)$ to $S_0(1)$ integrated intensities.

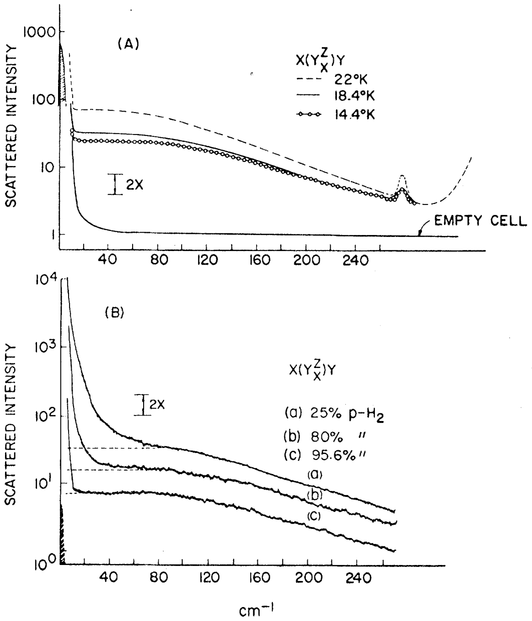

FIG. 1. Depolarized intermolecular scattering in liquid hydrogen. (a) Temperature variation in 95.6% parahydrogen. (b) Variation with ortho-para composition at constant temperature (15°K). Vertical displacements of the spectra have been made for clarity.

Figure 1 shows the low-frequency portion of the depolarized $X(Y^Z_X)Y$ spectrum of liquid hydrogen under different conditions of temperature and parahydrogen content. In Fig. 1(a) the intermolecular spectra from 95.6% parahydrogen are shown for three different temperatures. The weak line at 270 cm^{-1} is the $S_0(0)$ transition from hydrogen-deuteride impurities. In all cases an exponential tail develops out beyond 100 cm^{-1}. However, at lower frequencies the spectra fall well below exponential and at 14.4°K a hint of a decrease in intensity at low frequency can be seen. This behavior is in marked contrast to the situation in the classical rare-gas liquids,[3-5] where the exponential behavior persists down to the lowest frequencies observed (5–10 cm^{-1}). Indeed the *liquid*-hydrogen spectral shapes imply even better-defined short-wavelength excitations than were observed in the *solid* classical rare gases at temperatures above about two thirds of their melting temperatures.[4] As T is increased, the integrated intensity increases (by about 30% for a 55% increase in T). At the same time the exponent Δ decreases from 75 cm^{-1} at 14.4° to 62 cm^{-1} at 22.5°K. These values of Δ are uncertain by about ±3 cm^{-1} due to subtraction of the fluorescent background (~3% of the maximum intermolecular intensity). It is interesting to compare the observed value of Δ with that predicted by the empirical formula deduced from experiments on classical fluids[4,5]:

$$\Delta=\frac{3}{2\pi c}\left(\frac{\epsilon}{M\sigma^2}\right)^{1/2}\left(\frac{kT}{\epsilon}\right)^{1/2}\left\{1+\left(\frac{2\sigma^3}{M}\rho\right)\right\}^2, \tag{1}$$

where c is the velocity of light, and ϵ and σ are the Lennard-Jones intermolecular potential parameters. For parahydrogen at vapor pressure and 15°K, Eq. (1) predicts $\Delta = 31.6\ \text{cm}^{-1}$, suggesting that some quantum corrections are required.

Two phenomenological quantum corrections have occurred to us. First the replacement in Eq. (1) of the effective thermal velocity, $\sim (kT)^{1/2}$, by a zero-point velocity, $V_0 = \hbar/Mr_0$, expresses the physical expectation that for $kT \lesssim kT_0 = \frac{1}{6}MV_0^2$ the spectrum should be temperature independent. Second, quantum effects on corresponding-states expressions for transport coefficients have been phenomenologically expressed[8] in terms of the deBoer parameter $(\Lambda_i^*)^2 = h^2/\sigma_i^2 M_i \epsilon_i$ as

$$\frac{\alpha_1(T^*,\rho^*)}{\alpha_2(T^*,\rho^*)} = 1 + \Delta\Lambda^{*2}\left(\frac{\partial \ln\alpha}{\partial\Lambda^{*2}}\right)_{\Lambda_2^*}. \tag{2}$$

This relates two materials at the same corresponding temperature and density having different quantum behaviors. Taking Eq. (1) to apply to the hypothetical substance "classical" hydrogen (i.e., $\Lambda^* = 0$) and replacing α by Δ^{-1} we compute $\Delta_{\text{H}_2}{}^{\text{Cl}}(1+1.22)$ for temperature $T \gtrsim T_0$. We have also assumed that $\partial\ln\Delta^{-1}/\partial\Lambda^{*2}$ can be evaluated graphically as done in Ref. 8. For example, $(\partial\ln\eta/\partial\Lambda^{*2})_{\Lambda^*(\text{H}_2)} = -0.41$. Of course $\Delta\Lambda^{*2} = -\Lambda^{*2}(\text{H}_2) = -2.98$. Taking $r_0 = (\rho/M)^{-1/3}$ we find for hydrogen ($\Lambda^* = 1.73$) at 15°K at saturated vapor pressure that $T_0 = 13°\text{K}$ so the additional correction $(T_0/T)^{1/2}$ is rather small here. The result of both corrections is a calculated $\Delta_{\text{H}_2} = 65\ \text{cm}^{-1}$ which, considering the crude nature of these corrections, is in fairly good agreement with observation. Although phenomenological and semiquantitative at best, such corrections appear to extend Eq. (1) successfully into the quantum regime. Some confidence in these corrections can be gained by the application of the same procedure to the case of liquid He^4 ($\Lambda^* = 2.67$). There Surko and Slusher[9] have observed a nearly temperature independent Δ ($= 34 \pm 3\ \text{cm}^{-1}$) at constant density (0.18 g/cm^3) for temperatures between 2.2 and 21°K. Equation (1) predicts at 5°K a value $\Delta_{\text{He}}{}^{\text{Cl}} = 11\ \text{cm}^{-1}$, which, of course, should vary like $T^{1/2}$. The corrections discussed above produce a temperature independent $\Delta = 40\ \text{cm}^{-1}$, which again is in fairly satisfactory agreement with experiment.

The effect on the liquid-hydrogen spectrum of increasing orthohydrogen content is illustrated in Fig. 1(b) where 25%-, 80%-, and 95.6%-parahydrogen spectra at $T \approx 15°\text{K}$ are compared. The high-frequency behaviors are virtually identical, whereas the striking emergence of the strong low-frequency "Rayleigh wing" is seen with increasing orthohydrogen content. We note that this $\Delta J = 0$, $\Delta M_J \neq 0$ spectrum has an indistinguishably different shape from the $\Delta J = 2$, $\Delta M_J \neq 0$ transition [$S_0(1)$] centered at 586 cm^{-1}. If this single ortho-molecule orientational scattering line shape is subtracted without adjustment in intensity from the total spectrum, the dashed-line extensions in Fig. 1(b) result. This implies that the ortho molecules contribute to the intermolecular spectrum on an equal footing with the parahydrogen molecules, and, further, that their contributions to the liquid's high-frequency dynamics are essentially identical to the parahydrogen. In other words, normal hydrogen as well as parahydrogen sustains well-defined short-wavelength excitations, which are not strongly coupled to the rotational modes.

We now turn to the spectrum of solid hcp hydrogen, and confine our attention here to the essentially pure para case. Figure 2(a) shows the entire solid spectrum of interest on a logarithmic intensity scale at 11°K. The most prominent features are the E_{2g} phonon at 38 cm^{-1}, the unresolved $S_0(0)$ triplet at 355 cm^{-1}, and the $S_0(1)$ line at 586 cm^{-1}. All of these have previously been observed[10,11] and will not be discussed further here.[12] The new features of interest are the two-phonon

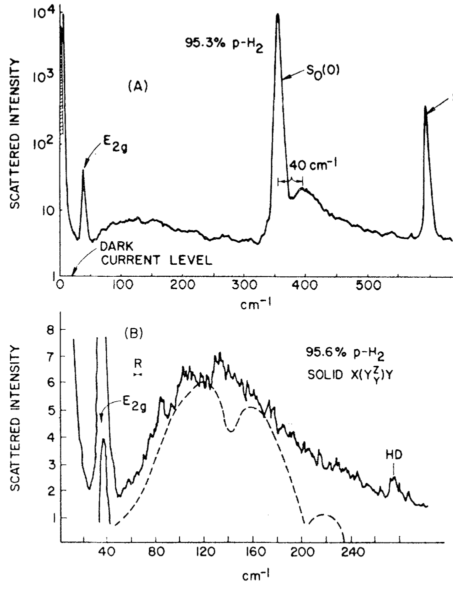

FIG. 2. Solid-parahydrogen Raman spectrum at 11°K. (a) Logarithmic intensity scale for comparison of features differing greatly in intensity. (b) Linear-scale spectrum for one- and two-phonon processes. Dashed curve, theory for polycrystal as described in text.

spectrum and the phonon sidebands to the $\Delta J = 2$ transitions. The $S_0(0)$ phonon sideband peaks at 40–50 cm^{-1} above the center of the $S_0(0)$ transition and extends out at least for another 100 cm^{-1}. A similar sideband to the $S_0(1)$ transition can be seen in normal hydrogen.[12] The sideband shapes are not simply those expected from the one-phonon density of states in hcp hydrogen. A peak at 40–50 cm^{-1} in the one-phonon density of states would arise from T' zone-boundary TA phonons (according to the neutron scattering[13] in hcp hydrogen), but should be accompanied, if not overshadowed, by higher frequency features. Whether selection rules governing the simultaneous excitation of one phonon and one $\Delta J = 2$ rotational transition can explain the observed shapes is a matter for future theoretical consideration.

Of greater interest is the two-phonon spectrum shown in more detail on a *linear* intensity scale in Fig. 2(b). This spectrum is somewhat similar to that in hcp He^4, for which both calculations[14] and observations[2] are available. Inelastic neutron scattering in hcp He^4[15] and hcp p-H_2[13] reveal very similar phonon dispersion curves. We have invoked this similarity to produce the dashed theoretical curve in Fig. 2(b), where we have reproduced the calculated two-phonon spectrum[14] for polycrystalline hcp He^4 scaled in frequency by approximately a factor of 4 [the ratio of the observed E_{2g} phonon (38 cm^{-1}) frequency in hydrogen to that (9.5 cm^{-1}) in helium[2]]. The agreement is quite satisfactory—indeed better than the agreement between the same theory and the observed He^4 spectra.[2] For example, the spectral strength lies dominantly in the correct position (80–180 cm^{-1}) and the high-frequency excess scattering (above the predicted 24-cm^{-1} cutoff) is much smaller than in He.[2] There is even a reasonable suggestion of the predicted dip in intensity near 120 cm^{-1}. The fact that hydrogen is so much stronger a scatterer than He suggests that it might be a more convenient testing ground for theories of lattice dynamics in quantum crystals and that experimental effort directed toward single-crystal spectroscopy, particularly under pressure, would be worthwhile.

Finally, we compare the intermolecular scattering in the liquid and the second-order scattering in the solid parahydrogen. Above 150 cm^{-1} the two spectra are essentially indistinguishable. This is quite similar to the previous observations on melting the classical rare-gas solids.[4] As with them, the intensity in the low-frequency portion of the spectrum increases in hydrogen upon melting—but not by so much as to impart an exponential shape to the spectrum. Instead the effect of melting in parahydrogen is intermediate between the classical Ar[4] and the fully quantum He.[2] It is entirely consistent with the broadening, but not the disappearance or overdamping, of the solid-zone-boundary phonons upon melting. Qualitatively, this behavior may be said to arise because, like He, hydrogen melts at a much lower fraction of its Debye temperature than do any of the classical materials. Pressure studies, wherein the Debye temperature may be significantly altered, should further clarify the important questions of collective modes in liquids and their relation to phonons in solids, and perhaps even point toward a microscopic understanding of melting. Finally, the qualitative, but nevertheless striking, correlation between the behavior of short-wavelength excitations and the value of the deBoer parameter strongly suggests the extension of these experiments to the study of orthodeuterium ($\Lambda^* = 1.223$).

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Mode-Coupling Saturation of the Parametric Instability and Electron Heating*

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By solving the hydrodynamic equations for the parametric instability including mode-coupling terms, we predict the saturation level and turbulent spectrum for the oscillating two-stream instability. Coupling the electron distribution function to the spectrum with the quasilinear diffusion coefficient gives the heating rate and high-energy tail formation. These predictions agree with the results of particle simulations.

A problem of considerable interest to laser fusion is the possible formation of high-energy tails on the electron distribution function due to parametric instabilities. These tails have been observed experimentally[1,2] and in simulations.[3] Energetic electrons so formed may lead to serious preheat and decoupling problems in the design of pellets.[4] We report here a one-dimensional analysis of the saturation and heating mechanism for the parametric instability at the critical density (laser frequency equal to the plasma frequency). The analysis is based upon mode-coupling saturation of the linear instability and subsequent electron heating by quasilinear diffusion due to the saturated spectrum.

Previous authors have considered saturation of the decay instability in a plasma with comparable electron-ion temperatures.[5] The dominant nonlinearity for the plasma waves is then their nonlinear Landau damping on the ions, a process which only spreads plasma wave energy to higher phase velocities. (When $T_e \gg T_i$, this process goes over into the resonant coupling of plasma waves by ion acoustic waves.) In these analyses electron-ion collisions play a central role as the process by which energy is coupled from the waves into the particles. In general, however, plasma wave energy spreads to both lower- and higher-phase-velocity waves. Coupling of energy to lower phase velocity can result in the transfer of energy from the waves into the particles by Landau damping. Hence a stationary nonlinear state is possible even when collisions are negligible. Since the energy transfer is via Landau damping, the particle heating will be characterized by the formation of high-energy tails.

The nonlinear evolution of the parametric instabilities driven by an electric field oscillating near the plasma frequency has been extensively investigated in computer simulations. They show that for laser field intensities $\eta^2 \equiv E_0^2/4\pi n T_e \gtrsim 1$, the instability saturation is simply determined by electron trapping in the unstable plasma waves (E_0 is the laser field amplitude; n is the electron density). However, for $\eta^2 \ll 1$, trapping does not occur. In this regime the plasma wave energy saturates at a level several times the pump field energy [see Fig. 1(a)]. The particles heat linearly in time, principally because of generation of suprathermal tails on the distribution function.

In order to discuss a specific instability, let us consider the case in which the laser frequency equals the electron plasma frequency. Then only the oscillating two-stream instability occurs. In this case simulations show that, after saturation, the plasma wave spectrum ($|E_k^2|$) assumes an approximate k^{-2} shape for wave numbers greater than the linearly most unstable mode (Fig. 2). The heated-electron distribution function has a central Maxwellian, an exponential tail, and then a sharp drop (Fig. 3). Our analysis will predict all these basic features observed in the simulations.

Since the fluctuating fields in the saturated state become as large as the pump field, this suggests that mode coupling may be the saturation mechanism. We apply Nishikawa's ordering scheme[6] to the hydrodynamic equations for electrons and ions, but without neglecting the products of pairs of fluctuating quantities. The following equations