Effects of Helium on Two-Level Systems in Porous Vycor Glass

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From sound-velocity measurements we found that the presence of helium in the pores of Vycor increased the relaxation rates of two-level systems in the glass. Superfluid ^{4}He caused a much larger increase than either solid ^{4}He or liquid ^{3}He. The results are interpreted in terms of additional thermal phonons in the helium, with fourth-sound modes possibly causing the faster relaxation for superfluid ^{4}He.

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The transfer of excitations between different media at low temperatures, especially between ordinary solids and liquid and solid helium, is still not well understood. We present here new results on this issue. Specifically, we report on measurements of changes in ultrasonic velocities due to helium in porous Vycor glass.[1] By filling the pores with ^{3}He or ^{4}He, the effects of helium on the nearby two-level systems (TLS) in the glass could be studied. The results indicate that the helium increases the relaxation rate of the TLS and that superfluid ^{4}He has a particularly large effect.

Our experimental method has been described elsewhere.[2,3] Transverse sound pulses were generated in a rod 1.02 cm long and 0.3 cm in diameter. Measurements were made between 20 and 194 MHz in empty Vycor and in Vycor containing liquid ^{3}He or superfluid or solid ^{4}He.

Before presenting our results, we summarize the predictions of the TLS model[4,5] for the sound velocity in a glass. At low temperature there is a resonant interaction of the sound wave with TLS which results in a change in the sound velocity of the form

$$\Delta v_{\rm res}/v = (nM^2/\rho v^2)\ln(T/T_0), \tag{1}$$

where n is the TLS density of states, M is a deformation potential, ρ is the density, and T_0 is an arbitrary reference temperature. At higher temperatures, relaxation of TLS due to interactions with thermal phonons becomes important. The relaxation time τ_1 for a TLS with energy splitting E is given by

$$\tau_1^{-1} = \left(\frac{M_l^2}{v_l^5} + \frac{2M_t^2}{v_t^5}\right)\frac{E^3}{2\pi\rho\hbar^4}\coth\frac{E}{2k_BT}, \tag{2}$$

where the subscripts l and t denote longitudinal and transverse modes. The relaxation contributes a velocity change of the form

$$\frac{\Delta v_{\rm rel}}{v} = -\int_0^\infty dE\,\mathrm{sech}^2\left(\frac{E}{2kT}\right)\frac{nD^2}{8\rho v^2}\times\frac{1}{1+\omega^2\tau_1^2}, \tag{2a}$$

where D is a deformation potential and E is the energy splitting. The relaxation process causes a decrease in the velocity as the temperature increases.[5] As a result, the velocity increases logarithmically with T at low temperature and goes through a maximum at a temperature $T_{\rm max}$ which increases with frequency.

Figure 1 shows the sound velocities in empty Vycor. From the slope at low temperature, we find $nM_t^2 \simeq 2\times10^7$ erg/cm^3 ($\rho = 1.558$ g/cm^3; $v_t = 2.19\times10^5$ cm/sec). We made some measurements on Vycor fused to remove the pores and found qualitatively similar results ($nM_t^2 \simeq 8\times10^7$ erg/cm^3 with $\rho = 2.115$ g/cm^3, $v_t = 3.55\times10^5$ cm/sec, and $T_{\rm max} \simeq 1.1$ K at 17.5 MHz and 1.9 K at 110 MHz). We conclude that the sound velocity in porous Vycor is dominated by TLS.

The addition of helium to the pores affects the sound velocity both because the density increases by an amount[1] $\phi\rho_{\rm He}$ (thus decreasing v_t) and because the elastic moduli increase (thus increasing v_t).[2,3] For liquid ^{3}He (zero shear modulus) only the change in density affects v_t. This effect is independent of frequency and is almost independent of temperature below 1 K since the density of ^{3}He is nearly constant there. Figure 2 shows the measured sound velocities when the Vycor contains ^{3}He at its saturated vapor pressure.

The increase in v_t above 2 K is due to the thermal expansion of the ^{3}He, but the velocity clearly shows an additional dependence on both temperature and frequency below 1 K. The increase in v_t at low temperature followed by a velocity maximum at

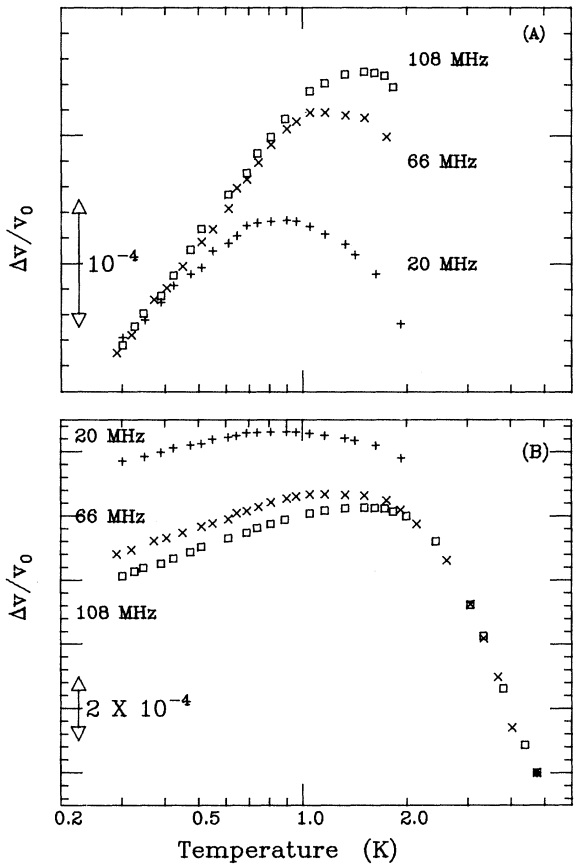

FIG. 1. Transverse sound velocities in empty Vycor. (a) Data at different frequencies plotted to agree at temperatures below the maxima. (b) Data at different frequencies plotted to agree at temperatures above the maxima. Note the difference in the scale of velocity changes between (a) and (b). The same temperature scale is used in (a) and (b).

a temperature which increases with frequency indicate that the behavior shown in Fig. 2 is also due to the glass TLS. However, the maxima occur at lower temperatures when the ^{3}He is present (by a factor of about 2.5), implying a much shorter TLS relaxation time τ_1 in the glass.

This effect is not unique to ^{3}He as we found similar behavior by filling the Vycor with ^{4}He. In addition to the decrease in v_t due to the density of the ^{4}He, we found[2] that, when the ^{4}He solidified, v_t increased as a result of the added shear modulus of the solid ^{4}He in the pores.

In Fig. 3 we show the low-temperature sound velocity in Vycor filled with solid ^{4}He. The pressure was 83 bars when the ^{4}He began to freeze. The resulting increase in shear modulus caused an increase in v_t of about 0.4% between the start of

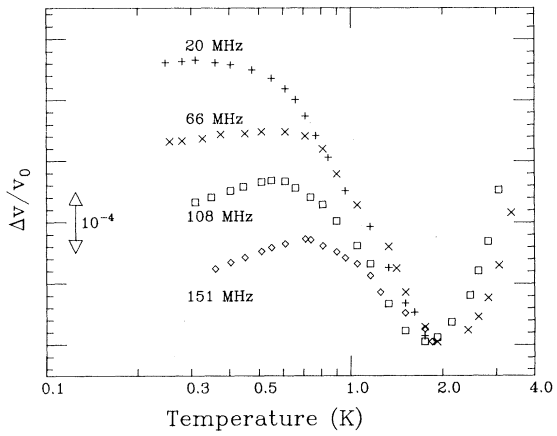

FIG. 2. Transverse sound velocities for liquid ^{3}He in Vycor (at saturated vapor pressure). Data at different frequencies have been plotted to agree at the velocity minimum near 2 K.

freezing at 2.38 K and 1 K. The modulus change due to freezing of the ^{4}He is independent of frequency and, since the shear modulus of solid ^{4}He is nearly constant below 1 K, v_t should not change in that range of temperatures. Figure 3 (inset) clearly shows a temperature and frequency dependence of v_t which is typical of TLS in glasses. The logarithmic slope at low temperatures (indicating

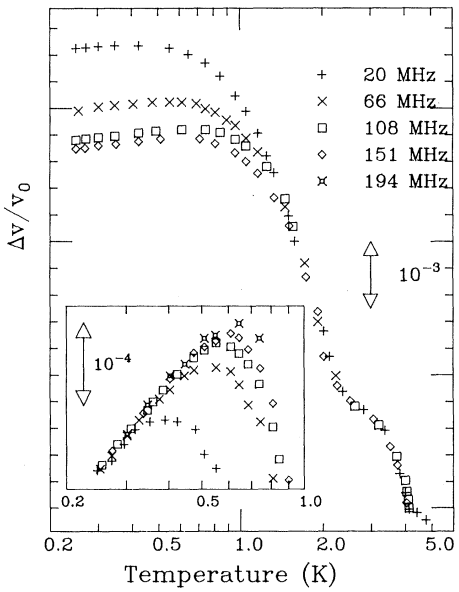

FIG. 3. Transverse sound velocities for solid ^{4}He in Vycor, at different frequencies, plotted to agree at the temperature corresponding to the onset of freezing of helium in Vycor pores ("knee" in data at about 2 K). Inset: the same data near the maximum, on an expanded scale, plotted to agree at temperatures below the maxima.

$nM_t^2 \simeq 1.7\times 10^7$ erg/cm^3) and the frequency dependence of T_{max} are similar to those found in empty Vycor (Fig. 1), but the velocity maxima are shifted to lower temperatures (again by a factor of about 2.5) compared to empty Vycor. Thus, the presence of solid ^{4}He in the pores appears to decrease the TLS relaxation time much as does liquid ^{3}He.

The effect, shown in Fig. 4, of filling the pores with ^{4}He at its saturated vapor pressure, was surprising. As previously reported[2,3] the sudden change in v_t below the λ transition (at 1.94 K in the Vycor pores) is due to the decoupling of a fraction of the superfluid from the Vycor, resulting in a frequency-independent increase in v_t. The amount of superfluid which decouples from Vycor has been studied directly in a torsional-oscillator experiment[6] and was found to change by less than 1% below 0.5 K. In addition to the decoupling effect, however, Fig. 4 shows that there is a frequency-dependent velocity variation which is significant below 1.5 K. Although we believe this low-temperature variation to be due to the TLS in the Vycor, the velocity does not go through a maximum, except perhaps around 0.3 K at 151 MHz.

The frequency dependence of v_t shown in Fig. 4 is consistent with the relaxation behavior of TLS above the velocity maxima, indicating that the velocity maxima are shifted to temperatures below 0.3 K. Thus, the superfluid ^{4}He must reduce τ_1 in the glass much more than do either solid ^{4}He or liquid ^{3}He. To check whether this was due to the superfluidity of the ^{4}He, we made measurements in which the pores were partially filled with ^{4}He in an effort to extend the normal-fluid range. Since we saw no evidence in either the velocity or the attenuation of a λ transition at temperatures down to 0.3 K, we believe that the Vycor was less than about one-third full (cf. Ref. 6) which would correspond to a film thickness of less than about 7 Å. At 108 MHz (our only measurement frequency in this case), the velocity had a logarithmic slope at low temperatures (corresponding to $nM_t^2 \simeq 0.95\times 10^7$ erg/cm^3) and went through a maximum at 0.8 K, thus showing behavior similar to that of solid ^{4}He and liquid ^{3}He.

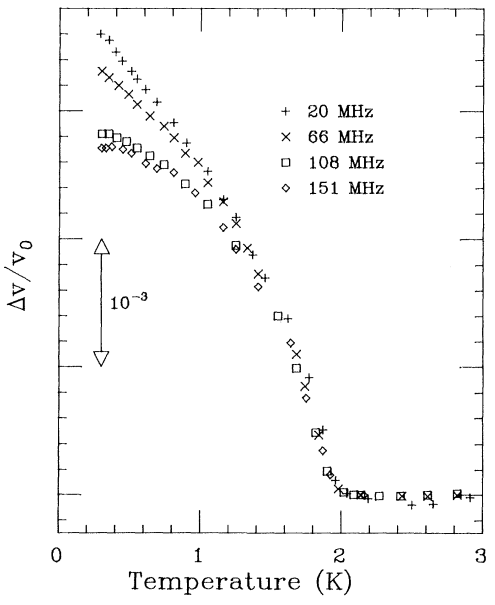

FIG. 4. Transverse sound velocities for liquid ^{4}He in Vycor (at saturated vapor pressure). The λ transition occurs at $T = 1.94$ K. Data at different frequencies have been plotted to agree above the superfluid transition temperature.

From the frequency and temperature dependence of the sound velocity in these experiments, we conclude that the presence in the Vycor pores of liquid ^{3}He, solid ^{4}He, or a film of nonsuperfluid ^{4}He increases the glass TLS relaxation rate τ^{-1} by a similar amount. We note that the temperatures of the velocity maxima in these cases decreased by the *same* factor (~ 2.5) for all frequencies covered ($\omega/2\pi$ from 20 to 151 MHz). For fused or empty Vycor, as well as Vycor containing solid ^{4}He or liquid ^{3}He, the temperatures of the maxima had the frequency dependence $T_{max} \propto \omega^{0.30\pm 0.05}$ (consistent with the $\omega^{1/3}$ dependence predicted[5] by the TLS model in the limit $\omega\tau_1 >> 1$). A crude estimate of the change in the "effective" relaxation time τ due to the helium can be obtained by assuming that the velocity maxima occur at the same values of $\omega\tau$ in all cases. Then, at the maxima, $\omega\tau = \text{const}$ and $\tau \propto \omega^{-1} \propto T_{max}^{-3}$. From the observed values of T_{max}, we find that the addition of solid ^{4}He or liquid ^{3}He caused a decrease in τ by a factor of about 15. The addition of superfluid ^{4}He, however, appears to cause a substantially faster relaxation. The data of Fig. 4 indicate that T_{max} is below about 0.3 K at all frequencies when the pores contain superfluid. This implies that τ is at least another factor of 8 smaller in the superfluid case.

In a recent experiment, Schubert, Leiderer, and Kinder[7] studied the transmission of 25-GHz phonons from quartz into a 10-Å film of ^{4}He through a 300-Å amorphous film. They found a large and saturable transmission which they attributed to TLS in the amorphous layer, giving support to the defect model of the anomalous Kapitza conductance. Without the ^{4}He film, no effects due to the TLS were observed, a result which they interpreted as indicating that the TLS relaxation was much faster

when the ^{4}He film was present. Our results support this and provide direct evidence of the effect of helium on the TLS relaxation.

Schubert, Leiderer, and Kinder suggested that TLS near the helium film are coupled to the thermal phonons in the helium, perhaps through the Van der Waals interaction between the helium and the glass surface. Thus, in addition to the relaxation rate due to thermal phonons in the glass, Eq. (2), there is a contribution from thermal phonons in the helium. Since the density and sound velocities in helium are much smaller than those of the glass, the relaxation rate due to the helium, τ_{He}^{-1}, can be larger than the empty-Vycor relaxation rate [cf. Eq. (2)], even if the TLS are only weakly coupled to the helium. Such a mechanism would adequately account for our results for solid ^{4}He and fluid ^{3}He, but would also lead us to expect a similar change for superfluid ^{4}He, in contrast to our observations.

To understand the difference between the superfluid and the other cases, we consider the effect of the small pore size on thermal phonons in the helium. Phonons with wavelengths shorter than the pore size D are similar to those in bulk helium, but, if the helium cannot move relative to the Vycor, phonons with wavelengths longer than D are hindered in their propagation. Thus, in solid ^{4}He and in liquid ^{3}He (since the ^{3}He is viscously locked to the glass) thermal phonons with wavelengths longer than D are scarce. Superfluid ^{4}He, on the other hand, does have such phonons (fourth sound) since the superfluid can move relative to the Vycor.

The main contribution to the relaxation rate comes from[4] phonons with $\hbar\omega \simeq 3k_{\mathrm{B}}T$, so that at a temperature

$$T_c = h v_{\mathrm{He}}/3k_{\mathrm{B}}D, \tag{3}$$

the important phonons have a wavelength D. Above T_c, the short-wavelength phonons in the helium cause an increase in the TLS relaxation rate but below T_c there are few thermal phonons in ^{3}He or solid ^{4}He and so the relaxation rate decreases towards the value it had in empty Vycor.

For solid ^{4}He ($v_t \simeq 2.5\times 10^4$ cm/sec at 50 bars), we find $T_c \simeq 0.7$ K and for ^{3}He ($v \simeq 1.8\times 10^4$ cm/sec), $T_c \simeq 0.5$ K. Figures 2 and 3 indicate that the relaxation rate is much larger in these cases than in empty Vycor (Fig. 1) above 0.5 K and that it decreases at lower temperatures. Our measurements with a nonsuperfluid film indicate similar behavior. In contrast, Fig. 4 indicates that superfluid ^{4}He causes a fast relaxation even at our lowest temperatures of 0.3 K, as expected since thermal phonons of arbitrarily long wavelength can propagate (as fourth sound) in the pores.

We believe that our results show that the presence of helium in the pores increases the relaxation rate of the Vycor TLS because of the thermal phonons in the helium. At low temperatures the pore size becomes important and eliminates the long-wavelength phonons except in the case of superfluid ^{4}He where the relaxation rate remains large because of fourth sound.

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[1]Vycor is a silica glass containing an interconnected network of pores (porosity $\phi = 0.28$; average pore diameter ≈ 55 Å).

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