Velocity Spectrum of Atoms Evaporating from a Liquid He Surface at Low Temperatures

K. Andres, R. C. Dynes, and V. Narayanamurti
Bell Laboratories, Murray Hill, New Jersey 07974
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Using the heat-pulse technique, we have measured the velocity spectrum of atoms evaporating from a free liquid ^{3}He and ^{4}He surface. In the ballistic regime (at sufficiently low ambient temperatures) this spectrum is found to be Maxwell-Boltzmann in nature with no effects evident owing to the quasiparticle nature of the liquid. At higher vapor pressures (higher temperatures) the transition from ballistic atomic propagation to collective adiabatic sound in ^{3}He and ^{4}He has been studied.

I. INTRODUCTION

Over the past several years, the heat-pulse technique[1] has been extensively employed for very-high-frequency sound-propagation studies in solids. With the development of sensitive superconducting bolometers with submicrosecond response times, time-of-flight measurements for very fast low-energy pulses have become possible. We have applied this heat-pulse technique to study the properties of helium atoms evaporating from the surface of a liquid film. By changing the mean-free-path λ of the evaporated atoms by controlling the vapor pressure of the gas, we have observed the transition from collective sound propagation to single noninteracting particle flow (ballistic).[2,3] In the ballistic regime, the velocity spectrum observed is a measure of the evaporation energy spectrum and to the accuracy of the observation is Maxwell-Boltzmann in nature. We see no evidence for liquid density-of-states effects in either ^{3}He or ^{4}He.

II. EXPERIMENTAL DETAILS

The experiments were performed in the temperature range from 0.1 to 3.4 K, the sample chamber being thermally attached to the mixing chamber of a dilution refrigerator. The heat pulses were generated with a 50-Ω constantan heater film, ~500 Å thick, deposited on a glass substrate, and were detected with a similarly deposited thin-film indium bolometer. The bolometer was magnetically biased at the midpoint of its resistive transition and then biased with a constant current source. Voltage pulses then signaled the arrival of the heat pulse. The size of both the heater and the bolometer was 3.7×3.7 mm and they were mounted opposite each other in a Teflon holder at a distance $L = 2.34$ mm inside the sample chamber. The voltage signals at the bolometer were recorded with a Biomation 8100 transient recorder (with 10-ns time resolution) and accumulated in a multichannel analyzer. Sufficient helium was introduced into the sample chamber to form a puddle of liquid at the bottom and a condensed film over all surfaces. The similarity of the observed heat pulses launched from ^{4}He and ^{3}He films and the absence of any anomaly at the superfluid transition of ^{4}He suggest that the only difference between condensed films of ^{4}He and ^{3}He at low temperatures is their uniformity in thickness. It is, in fact, well known that in ^{4}He above T_λ a condensed film of similar thickness to the superfluid-film forms.[4]

In an attempt to keep the pulse "temperature" as close to ambient as possible, the pulse width employed was 0.5 μs and the pulse peak powers ranged from 0.0036 to 1.44 W, which corresponds to energy dissipations of between 0.018 and 7.2 erg/pulse. In the case of ^{4}He, which we discuss first, we can assume that the heater is covered with a superfluid film of thickness ~200 Å. Assuming further a heat of vaporization of 82 J/mole, one needs about 10 erg to evaporate the entire film. Taking account of some energy loss into the glass substrate, we deduce that the liquid film remains essentially intact during evaporation and that the evaporating atoms experience the forces of a free liquid surface particularly at the lower power levels.

III. EXPERIMENTAL RESULTS

The two variables in our experiment are the pulse power and the ambient temperature. The latter governs the density and mean free path of the gas atoms between heater and bolometer. At 2 K, the vapor pressure of ^{4}He is 25 torr. Using ideal gas laws, the mean-free-path λ at this temperature can be calculated to be 2×10^{-5} mm. This is well within the hydrodynamic regime ($\lambda \ll L$) in which we expect a heat pulse to travel as an adiabatic temperature wave with the speed of sound, given by

$$v_s = (\gamma kT/m)^{1/2}. \quad (1)$$

For He, γ equals $\frac{5}{3}$. At 0.6 K on the other hand, the mean free path is about 1 mm. This is close to the propagation length L of 2.34 mm, so that at this temperature and below, the gas atoms have a high probability of reaching the detector without scattering. This transition from the sound regime to ballistic propagation in the ^{4}He gas is shown in Fig. 1, where the bolometer voltage is shown as a function of time after the emission of a heat pulse of 7.2 erg at time zero. We assume that the bolometer is an energy detector and that the amplitude of the signal at time t is approximately proportional to the number of atoms with velocity L/t that strike the bolometer, times their energy $E_0+\frac{1}{2}mv^2$, where $v=L/t$ and E_0 is the condensation energy per atom. It can be seen in Fig. 1 that above 1.1 K the heat pulse propagates in the form of sound with a well-defined velocity v_s and, in fact, the first echo (at $t=3L/v_s$) is also observed. Interestingly, the detected pulse shape does not reproduce the input pulse but eventually (after several echos) assumes the power derivative $\partial P(t)/\partial t$ of the original. The arriving signal shows a positive voltage pulse followed in time by an equally strong negative component. In the case of a point source and detector, it has been shown[5] that for a temperature wave, the response of the system is inductive in nature, yielding a signal representing $\partial P(t)/\partial t$ rather than $P(t)$. Near 1 K, the pulse broadens and speeds up until we reach the ballistic regime below 0.5 K, where the pulse becomes very broad and its shape depends only on the energy (or the temperature) of the pulse. This is shown more clearly in Fig. 2, where the observed peak velocity is plotted against temperature for different pulse energies. Above 2 K our observed velocities are in good agreement with Eq. (1) (dashed line), as

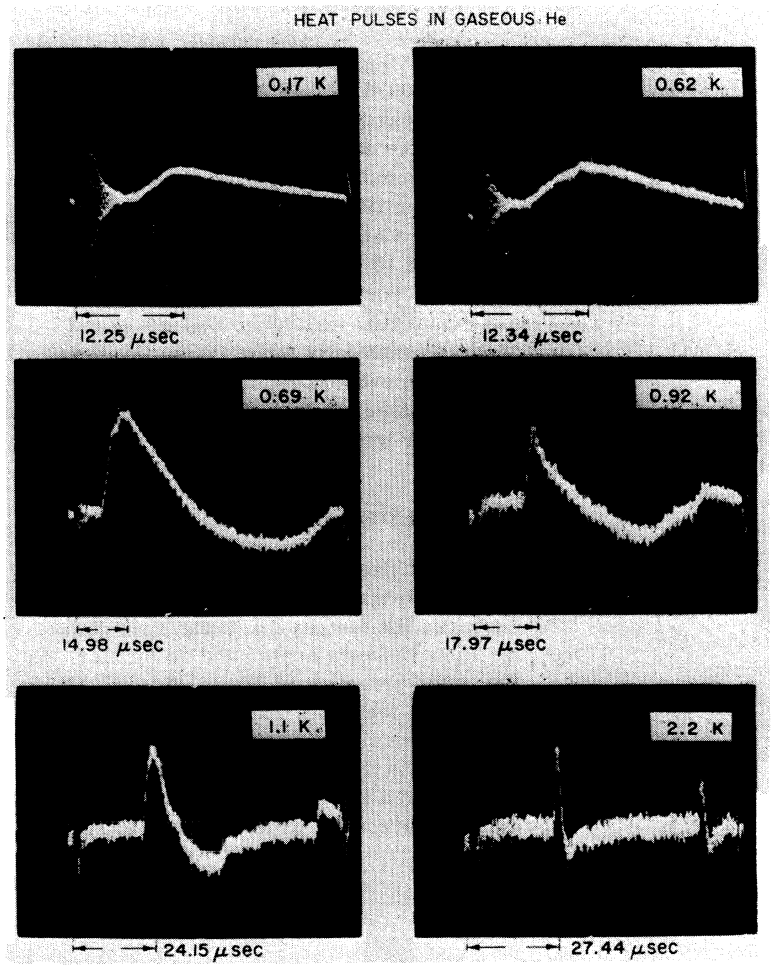

FIG. 1. Bolometer voltage vs time for various temperatures in gaseous ^{4}He. This signal displays the energy imparted to a bolometer by atoms evaporated from a He film 2.34 mm away after application of a 7.2-erg heat pulse at $t=0$.

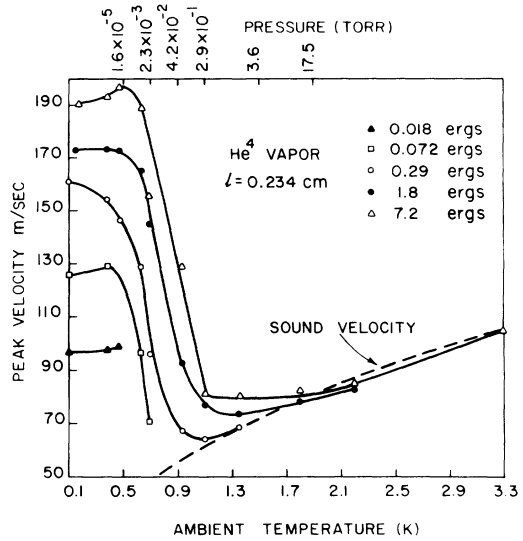

FIG. 2. Peak velocity vs temperature for varous energy pulses in ^{4}He. The dashed line is the ideal-gas sound velocity. At the lowest temperatures the pulse velocity corresponds to ballistic flow of atoms and depends on the pulse energy.

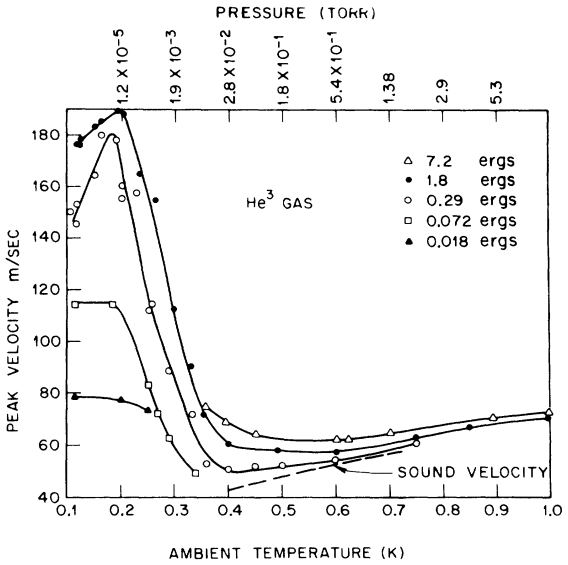

FIG. 3. Peak velocity vs temperature for various energy pulses in ^{3}He. The dashed line is the ideal-gas sound velocity.

was observed in previous work.[2] At lower temperatures, the peak velocity gradually increases and finally saturates in the ballistic regime at a value that depends only on the pulse energy.[2] In what follows, we analyze the shape of these pulses and show that they are consistent with a Maxwell-Boltzmann velocity distribution of the atoms. For the 0.018-erg pulse in the case of ^{4}He we find a pulse temperature of 1.1 K, and for the 1.8-erg pulse one of 2.9 K. Thus even for the lowest power pulses, our pulse temperature in the ballistic regime is always considerably above the ambient temperature. The results for ^{3}He are qualitatively very similar to those of ^{4}He and are shown in Fig. 3. Of course, the transition to the ballistic regime occurs around 0.25 K rather than 0.75 K due

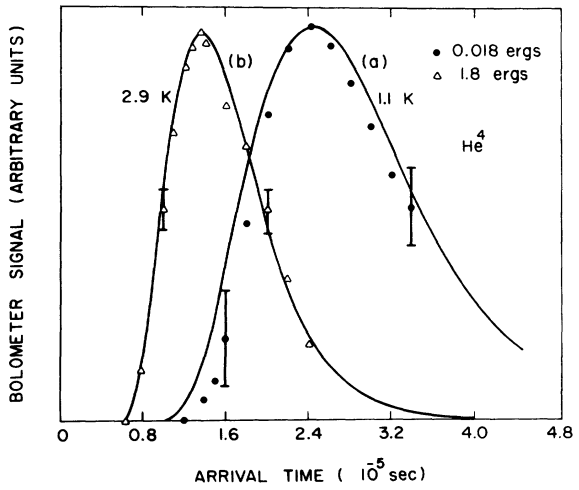

FIG. 4. Bolometer signal vs time (dots and triangles) and a best fit to a Maxwell-Boltzmann distribution (solid lines) for ^{4}He. Typical error bars are shown.

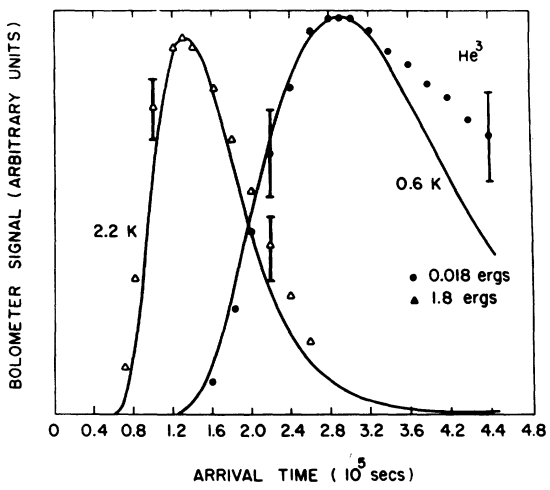

FIG. 5. Bolometer signal vs time (dots and triangles) and a best fit to a Maxwell-Boltzmann distribution (solid lines) for ^{3}He. Typical error bars are shown.

to the higher vapor pressure of ^{3}He. Again at the higher temperatures where $\lambda \ll L$, good agreement with Eq. (1) is obtained.

IV. DISCUSSION

It is intriguing to ask whether at the lowest temperatures and pulse powers the actual velocity distribution of the detected atoms reflects to any extent the excitation spectrum of the liquid. The question has been the subject of some theoretical study[6,7] following the experiments of Johnston and King[8] and, more recently, King, McWane, and Tinker.[9] It has been suggested that the anomalously high density of states in He II at the energy of the roton minimum E_r would influence the evaporation spectrum at a liquid-vapor interface. Specifically it was suggested that from a Boltzmann distribution a deviation might exist at an energy $E = E_r - E_0$, where E_0 is the binding energy per atom to the liquid surface. By this heat-pulse technique, such an anomaly would be observable in the velocity distribution measured at low temperatures.

With this in mind we have attempted to fit the actual shape of our ballistic pulses to that which one would expect from a Maxwell-Boltzmann velocity distribution. Assuming that the bolometer responds linearly with energy, for our geometry a Boltzmann distribution yields to within a constant $K(T)$ a line shape in time given by

$$S(t) = K(T) v_x^3 (\tfrac{1}{2}mv^2 + E_0) \exp[-(\tfrac{1}{2}mv_x^2 + E_0)/kT] \times \int_0^{v_x \tan\Phi} \int_0^{v_x \tan\Phi} \exp[-\tfrac{1}{2}m(v_y^2 + v_z^2)/kT]\, dv_y dv_z \,. \quad (2)$$

Here, one power of v_x appears because the bolometer detects the flux of particles striking it per unit time while the two other powers of v_x arise from the conversion from velocity space to time space [i.e., $S(t) = S(v)dv/dt$]. The term $\frac{1}{2}mv^2 + E_0$ in the prefactor represents the assumption that the bolometer responds linearly with energy as discussed earlier. The condensation energy (binding energy) E_0 is added to the kinetic energy $\frac{1}{2}mv^2$, since the bolometer is also covered with a liquid film. The double integral accounts for the fact that we also detect particles moving with y and z components of velocity up to values of $v_x \tan\Phi$ ($v_x = L/t$).

Adjusting the heat-pulse temperature T to obtain a fit to the experimental peak position in $S(t)$, the fits obtained for the 0.018- and 1.8-erg cases are shown in Fig. 4. For the low energy pulse, a fit is obtained for an effective pulse temperature of 1.1 K, while for the higher power pulse a 2.9-K distribution is needed. In both cases, however, the fit to a Maxwell-Boltzmann distribution is quite good and it would be difficult to identify any significant deviations from a purely thermal distribution that might be due to the excitations in the liquid. A similar analysis was attempted by Meyer *et al.*[2], where it was observed that the measured distribution was appreciably sharper than that of a Maxwell-Boltzmann one with the same probable velocity. This difference can be ascribed to the fact that the pulse energies employed in this earlier experiment were substantially larger (up to 300 erg compared to a maximum of 7.2 erg in our case) than those reported here. The large energies being dissipated most likely resulted in the entire film being "burnt" off and the pulse propagating as a shock wave. Note also that their received signal undergoes a wall reflection, unlike our case, which was free of wall reflections. Fits to the ^{3}He line shapes were also attempted and the results of this for two of the pulse energies are shown in Fig. 5. In this case, from heat of vaporization data, E_0 was taken to be 2.5 K. This as well as the value of 7.15 K for E_0 in ^{4}He, represents an approximation, as in both cases this value increases[4] with increasing T, rising to 5.6 and 11.4 K, respectively. These changes do not alter significantly the line shapes obtained in this calculation, but the magnitude of the absolute numbers, as will be discussed shortly, is affected. In the case of ^{3}He at low energies, there is some deviation outside the error bars for the slower components, but by suitably adjusting the magnitude of the peak, the agreement could be made substantially better. As stated earlier, the curves were fitted simply at the peak position and height. Hence again, as in the case of the ^{4}He, using the pulse temperature as an adjustable parameter, it would be difficult to argue significant deviations from a Maxwell-Boltzmann velocity distribution. An attempt was also made to fit the ^{3}He line shapes to Fermi statistics, as at these low temperatures liquid ^{3}He is a degenerate Fermi liquid. However, because $E_0 > \mu_{\text{Fermi}}$, very little difference between the two calculated distributions is observed and the fits were equally good for both statistics.

It had been observed previously in similar experiments[2] that the kinetic energy of the launched atoms varied as the fourth root of the pulse energy. Such a dependence was accounted for by the T^3 dependence of the specific heat of the heater wire and surroundings, resulting in the final heater temperature varying as (pulse energy)$^{1/4}$. It is not surprising that such an $E^{1/4}$ dependence is not observed in our geometry. First, the heater is of thin-film geometry on a glass substrate. The specific heat of glass is expected to obey the same

temperature dependence as other noncrystalline solids.[10] Below 1 K, C_v varies as T, while at higher temperatures it asymptotically approaches the usual T^3 dependence. Hence, the heater temperature should be somewhat more complicated than the $E^{1/4}$ dependence expected for a crystalline substrate and observed for a bulk heater wire by Meyer *et al.*[2] Second, for these low energies, one would not expect the temperature of the helium film to be equal to the temperature attained by the thin-film heater. The helium temperature must be determined by a dynamical balance of the heat flow in from the heater and the energy flow out via evaporation, the two flow rates determined by barrier thermal impedances. This expectation is qualitatively corroborated by the fact that for the same energy dissipations (0.018 erg) the temperature needed for a fit to the ^{3}He is 0.6 K, while for the ^{4}He it is 1.1 K. The vapor pressures of ^{3}He at 0.6 K and ^{4}He at 1.1 K (or the energy losses via evaporation) are approximately equal. Hence, one would expect that the ^{3}He film, which has a lower heat of vaporization (or E_0), to stabilize at a lower temperature than ^{4}He.

It could be argued that after evaporation of a large number of atoms, there is a strong interaction of these particles, consequent thermalization, and thus we are measuring the velocity spectrum of a thermalized gas and not the evaporating particles. If this is the case, for a pulse of width 0.5 μs, we would excite a volume $Av_s\Delta t$ of 3×10^{-4} cm^3 to 1.1 K. The total energy associated with the resultant 8×10^{14} atoms is ~0.20 erg; an order of magnitude larger than the *total* energy imparted to the heater for our lowest energy pulse (0.018 erg). Hence, we must conclude that thermalization does not take place and the atoms evaporated from the surface propagate ballistically away. In the ballistic regime from Eq. (2), we note that there is a reduction in the number of launched particles from the thermal equilibrium by the factor $e^{-E_0/kT}$ or at 1.1 K approximately 10^{-3}. This means that the number of particles launched in this case is ~10^{12} and approximately 1% of the energy dissipated goes into the evaporating atoms, the remainder presumably being taken up by the glass substrate or the helium-film flow along the surface. A similar calculation for the case of ^{3}He for a film temperature of 0.6 K suggests $\approx$ 20% of the energy is absorbed by the evaporation process. This increase in the amount imparted to the evaporating atoms seems qualitatively reasonable as there will be no superfluid-film-flow contribution to the dissipation in the case of ^{3}He.

As the energy of the pulse increases, the temperature fitted to the distribution of course increases, and it is found from similar arguments as above that the percentage of energy dissipated into the evaporating atoms also increases in both the ^{3}He and ^{4}He case. Of course, again the temperature dependence of E_0 must be properly taken into account in these considerations. At the highest energies, it is believed that a substantial percentage, although not all, of the film is evaporated during the pulse. This is supported by numerical calculations and the fact that the effective temperature of the film, which we believe is thermalized, is less than E_0.

The most straightforward explanation for such good line-shape fits is that the evaporation spectrum from the liquid is indeed Maxwell-Boltzmann. Theoretically,[6,7] the situation is by no means clear, though it has been argued by Cole[7] that this should be approximately the case. This would then be analogous to the case of electron tunneling between solids where, because of a cancellation of the density of states with the component of velocity perpendicular to the tunnel barrier, density-of-states effects, for the most part, are not observed. Experimentally,[9] it has been observed in a "dc" type of experiment using a conventional velocity selecting chopper technique in the range from 0.53 to 0.61 K that the velocity distribution is thermal with a temperature close to ambient. The present time-of-flight experiments complement these earlier results in that by varying the energy supplied into a pulsed heater, the liquid-helium temperature can be altered while still remaining in the ballistic regime in the vacuum. In this way the phonon and roton occupations in the liquid can be altered by orders of magnitude, and any influence the roton density of states has on evaporation should be more easily observable.

In a separate case, using a vertical geometry, the experiment was repeated except that the heater was immersed in *bulk* liquid ^{4}He and the liquid-gas interface was between the heater and bolometer. The experiments were performed at 0.1 – 0.2 K, well within the ballistic phonon regime,[11] and the results were similar to those reported here. Because the propagation length was approximately ten times longer, the signal to noise was not as good as that reported here and detailed line-shape analyses were not attempted. However, the qualitative results were very similar to those reported here for the same energy ranges. For even higher-energy pulses (up to 80 erg), a narrowing of the line became apparent, a phenomenon due to either partial thermalization of the evaporated atoms (initial formation of sound) or shock-wave formation. These sharper pulses were similar to the data of Meyer *et al.*[2]

In summary, we have studied the transition from

sound to ballistic atomic flow in gaseous ^{3}He and ^{4}He. In the transition and ballistic regimes the average velocity of the emitted particles is dependent upon the energy dissipated in the heat pulse. We present arguments supporting our belief, that at our low-heat-pulse powers, the measured line shape in the ballistic regime represents the velocity spectrum of atoms evaporating from the liquid-vapor interface. A line-shape analysis of these velocity distributions yields a good fit to Maxwell-Boltzmann statistics with no effects due to the density of excitations in the liquid observed.

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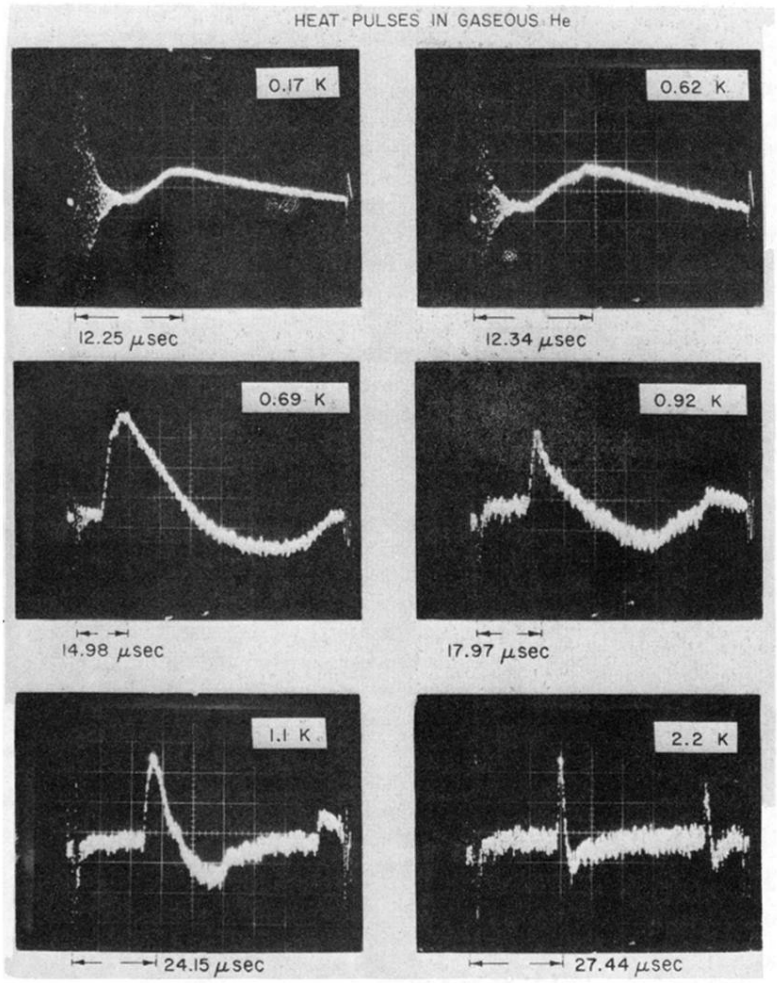

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