general results are similar: The characteristic lengths of helicity are larger than those of the energy, and the helicity again oscillates between positive and negative values at all smaller scales. The data at 1 AU were taken in a stream interaction region[13] and included both slow stream and fast stream intervals. Thus these results suggest that there is nothing atypical in the data shown in Figs. 1 and 2. A detailed analysis of the solar wind magnetic field structure at several additional locations in the heliosphere during different time periods and in different types of morphological environments is in preparation and will be presented in a more complete paper.

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Direct Measurement of Desorption Kinetics of ^{4}He at Low Temperatures

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A direct method for measuring the desorption time constant of flash-desorbed ^{4}He films ($\lesssim$ 1 monolayer), adsorbed on Nichrome or Constantan heaters, is described. A time constant τ is found which behaves as $\tau = \tau_0 \exp[E/T_s]$, where T_s is the heater temperature. The value for the characteristic lifetime τ_0 is 10^{-9}–10^{-10} sec, orders of magnitude shorter than that previously reported. The measured energy parameter E was found to be $\sim\frac{2}{3}$ of the chemical potential.

PACS numbers: 68.45.Da

If the temperature of a solid surface on which a film is adsorbed is suddenly increased, some adatoms are desorbed and the film reaches a new steady state in a characteristic time τ. Relating the desorption rate τ^{-1} to the surface temperature and the surface-adatom interaction potential is a difficult but basic problem of surface physics which has been the objective of many theoretical treatments using a variety of approaches.[1] For a weakly bound physisorbed film, theories based on simple thermodynamics and kinetic theory,[2] transition state theory,[3] as well as detailed quantum mechanical calculations[4-6] all predict a rate law of the so-called Frenkel form:

$$\tau = \tau_0 \exp(E/T_s) \tag{1}$$

in the first approximation. τ_0 and E are parameters with units of time and energy, respectively, and T_s is the substrate temperature. Although the form of the temperature dependence of τ in Eq. (1) follows from general considerations, various theories differ considerably both in the physical interpretation of τ_0 and E and in predictions of their absolute magnitudes. A study of the desorption kinetics of helium, which is the most weakly bound adsorbate, is and ideal test case for the theoretical models of the desorption process, particularly those theories which are based on a small-coupling perturbation analysis and suggest that one-phonon processes determine the desorption rate.[6]

We have, for the first time, verified directly the form given by Eq. (1) in a flash desorption experiment. For T_s between 5 and 11 K, we find $\tau_0 \lesssim 10^{-9}$ sec, 2 or more orders of magnitude shorter than values inferred from less direct measurements.[7]

The technique[8] we have developed permits us to heat and cool a substrate in approximately 10 nsec and thereby to measure the desorption time constant τ directly without using the deconvolution procedure usually necessary in more traditional flash desorption experiments.[7,9] The heating and cooling rates of 10^9 K/sec in our experiments are accomplished by using evaporated thin-film (~600-Å) Nichrome or Constantan heaters and thin (~2000-Å) Sn superconducting transition bolometer detectors on sapphire substrates. The fast thermal response arises because the thin-film devices are in intimate contact with a phonon-transparent substrate. The technology for generating and detecting nanosecond heat pulses is, in fact, adapted from previous work on ballistic phonon propagation.[10,11]

A schematic diagram of the experimental geometry is shown in the inset of Fig. 1. A controlled quantity of helium gas can be admitted to a vacuum can which is immersed in a pumped helium bath. A thin film of helium, typically less than one layer, is adsorbed on all surfaces in the cell, including the heater. A superconducting bolometer on a separate sapphire substrate is placed 1.25 mm above the heater. Both heater and bolometer have an area of 0.1 mm^2 and the heater is impedance matched to a 50-Ω cable. When a voltage pulse is applied to the heater, the bolometer detects a signal due to the helium atoms desorbed by the heat pulse. The bolometer signals, which measure the spectrum of the atomic times of flight, were observed on an oscilloscope after wide-band amplification or were averaged by a boxcar integrator. The atoms travel ballistically because the pressure in the background gas, typically between 10^{-9} and 10^{-5} Torr, is always sufficiently low that the mean free path is long compared to 1 mm. As the heater pulse width, t_p, is increased from its minimum value (~30 nsec) more and more atoms are desorbed and the bolometer signal increases until $t_p \sim \tau$. For sufficiently long pulses, however, the signal saturates and becomes independent of the pulse width as seen in Fig. 1, because the helium film reaches a steady state before the pulse is turned off.

Characterizing each spectrum by its maximum, we find empirically that data of the kind shown in Fig. 1 can be fitted by the form

$$S_m(t_p) = S_m(\infty)[1-\exp(-t_p/\tau)], \qquad (2)$$

where $S_m(t_p)$ is the height of the maximum in the bolometer signal as a function of t_p, the pulse width, and $S_m(\infty)$ is the corresponding value for very long pulses. This is shown in Fig. 2, where we have plotted $\log[1-S_m(t_p)/S_m(\infty)]$ vs t_p for the data of Fig. 1. The time τ is obtained from the slope of the resulting straight line.

In order to use the values of τ obtained in this way to test Eq. (1) and extract the parameters τ_0 and E, it is necessary to determine the heater temperature, T_s. We do so by making use of acoustic mismatch theory,[12] which is applicable to phonon transport at a metal/insulator inter-

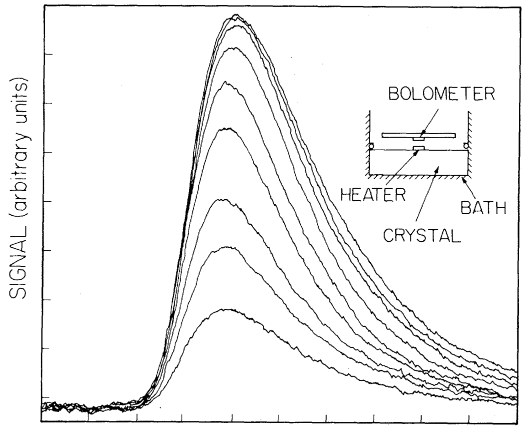

FIG. 1. Bolometer signal vs time for pulse width, t_p, in ascending order: 0.03, 0.06, 0.08, 0.15, 0.22, 0.5, 1, 1.5, and 2.5 μsec. In all cases, the heater temperature was 8.2 K, the bath temperature 3.48 K, and the chemical potential 72 K. Inset: experimental arrangement.

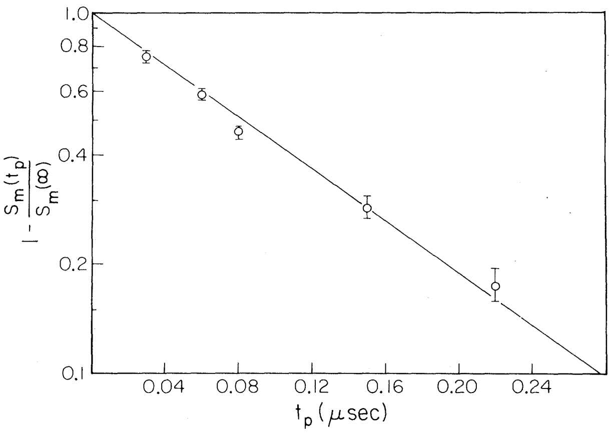

FIG. 2. $\log[1-S_m(t_p)/S_m(\infty)]$ vs t_p for data in Fig. 1. Straight line gives $\tau = 0.11$ μsec.

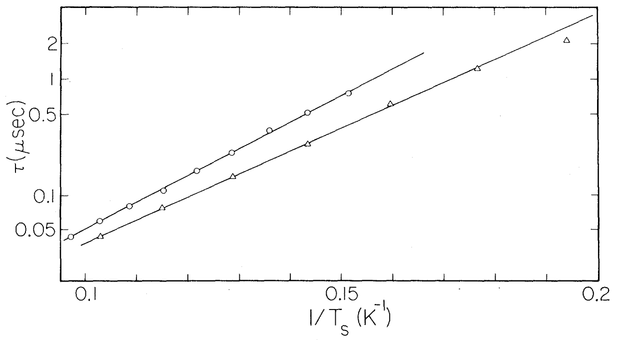

FIG. 3. Plots of τ vs $1/T_s$, verifying Eq. (1) of text. Upper and lower curves, respectively, are fifth and sixth rows of Table I.

face. Since the pulses we use are always long compared to the thermal relaxation time of the heater, the heater temperature T_s may be regarded as constant during the pulse. According to acoustic mismatch theory, T_s is given by[13]

$$T_s = (CW/A + T^4)^{1/4}, \tag{3}$$

where A is the heater area, W is the power dissipated in the heater film, T is the ambient temperature, and C depends on the heater and substrate elastic properties with $C = 6800$ K^4 mm^2/W, in our case. This relationship between phonon temperature and power has been verified in a number of previous experiments.[12-14]

An independent estimate of the heater temperature may be obtained from an analysis of the measured times of flight. Typical models of desorption suggest that the desorbed atoms should have an approximately Maxwellian velocity distribution characterized by a temperature T_a which is the same as the substrate temperature T_s.[2,5,6,15] The bolometer signal may thus be analyzed to find T_s. The precise result depends somewhat on the model used, but the values of T_s that emerge from the different models vary by only about 20%. Within these limits, the value of T_s deduced from the bolometer signal agrees with that given by Eq. (3) for each heater power.[16] This agreement supports our use of Eq. (3) for the substrate temperature.

Using the values of τ and T_s determined by the methods described above, we have tested the functional form of Eq. (1) by plotting $\log\tau$ vs T_s^{-1}. The results are very good straight lines; two examples are shown in Fig. 3. The slope and intercept of the line give the energy parameter E and the time τ_0, respectively. We cannot measure the desorption time constant for T_s above ~12 K because it becomes comparable to the thermal time constant of the heater.

In order to complete the description of the experimenal conditions, the pressure in the gas at equilibrium must be specified. Since the pressure in the gas governs the time for the film to return to equilibrium after each heat pulse, it was measured *in situ* even at the lowest pressures (~10^{-9} Torr) by observing t_{rc}, the shortest interval between pulses such that the signal was not affected. Since the film is far from equilibrium during most of its recovery, the outgoing flux is negligible compared to the incoming flux. Then, kinetic theory suggests that

$$t_{rc} \simeq \eta(2\pi m k_B T)^{1/2}/P, \tag{4}$$

where η is to be determined. This was done by measuring t_{rc} as a function of the pressure, P, in a regime in which P was high enough to be measured directly by conventional means. We found that P deduced from Eq. (4) with $\eta = 1\times10^{15}$ atom/cm^2 agrees with the directly measured pressure to within 30% for pulse powers between 0.2 and 2 W. A detailed discussion of this technique will be presented elsewhere. Because of the weak logarithmic dependence of μ, the chemical potential, on P, a 30% uncertainty in the pressure yields an uncertainty in μ of only 1%. Thus, we may regard μ as well known in these experiments.

The amount of helium adsorbed per unit area in the film is a definite, but unknown, function of T and μ. We can put an upper limit on it, however, by observing that for any T and μ, less helium will be adsorbed on the metallic heater than on the more strongly binding surface of graphite. Our thickest film had $-\mu \simeq 30$ K, corresponding

TABLE I. Summary of data for Nichrome and Constantan (*) heaters. First two columns give the temperature and chemical potential of the film before the heat pulse. Third and fourth columns give parameters in Eq. (1) for each set of initial conditions.

T (K)	$-\mu$ (K)	E (K)	$10^{10}\tau_0$ (sec)
3.75	94	60 ± 4	2
3.50	94	65 ± 2	1
3.48	94	62 ± 2	1.3
3.48	84	56 ± 2	1.6
3.48	80	54 ± 2	2.7
3.48	72	45 ± 2	5
3.27	64	33 ± 2	13
2.71	61	32 ± 2	13
1.50	30	20 ± 2	15
3.49	94*	59 ± 2*	4*
3.8	56*	43 ± 4*	2*

to 1.2 layers on graphite.[17] Thus all the data reported in this paper are for films of a monolayer or less.

The chemical potential μ can be controlled by adjusting the ambient temperature T and the amount of helium introduced into the cell. We have used this ability to study the dependence of τ_0 and E on the initial state of the film. At each T and μ, τ is determined for a sequence of heater powers, or equivalently, a sequence of values of T_s. The T_s dependence of τ is used to extract the parameters τ_0 and E, as illustrated in Fig. 3. Finally, the equilibrium state of the film is changed and the entire procedure is repeated; at each step the chemical potential is determined by measuring the critical pulse repetition rate.

The results of a series of experiments on Nichrome heaters are summarized in Table I. Essentially similar results are obtained for a Constantan heater (examples are included in the table). The most important features evident from this table are that τ_0 is of the order of 10^{-10} to 10^{-9} sec, and that the energy parameter E is strongly correlated with $-\mu$, being roughly equal to $\frac{2}{3}$ of its magnitude.[18]

The only previous experiments we are aware of which are comparable to ours are those of Cohen and King,[7] who find a value of $\tau_0 \sim 10^{-7}$ sec, more than 2 orders of magnitude larger than our result. Their experiment differed from ours in a number of respects, the most important being that their heater (a Constantan wire) could be rapidly heated but remained hot for times very long compared to τ. They deduced τ from a deconvolution analysis of their signal shapes based on the fact that the desorbing atoms arrive at retarded times if τ is sufficiently long. As a result, their procedure could not have detected time constants as short as those we observe.

In summary, we have made the first direct measurements of desorption times for He films. We find that the Frenkel form, Eq. (1), gives a good account of the results. The characteristic time, τ_0, is much smaller than previously thought, but still much longer than naive statistical mechanics estimates might suggest ($10^{-12\pm1}$ sec). The activation energy E is correlated with, but smaller than, the magnitude of the chemical potential (and hence also the binding energy, heat of adsorption, etc.). We do not know of any theory which would have led us to expect the parameter values we observe.

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Frequency Dependence of the Orientational Freezing in $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$

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Dielectric, ultrasonic, and neutron-scattering measurements in $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$ are reported for concentrations x ranging from 0.01 to 0.5. The dipolar and the quadrupolar susceptibilities exhibit frequency- and concentration-dependent cusps showing that the freezing process into an orientational glass state is a relaxational phenomenon.

PACS numbers: 64.70.Ew, 61.12.Fy, 62.20.Dc, 62.20.Mt

In KCN and the related compounds $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$ the aspherical dumbbell-shaped CN molecule resides in an octahedral Devonshire potential. The structural, elastic, and dynamic properties were explained by a coupling of the rotational excitations to the translational modes.[1,2] In the last two years there has been increasing experimental evidence that a glasslike phase exists for CN concentrations x less than 0.56.[3,4] The orientational glass state is characterized by a short-range order of the frozen-in CN molecules as suggested by the pattern of quasielastic diffuse intensity in neutron scattering experiments.[3,5] The temperatures of minimum sound velocities—determined from ultrasonic, Brillouin, and inelastic neutron scattering experiments—were regarded as freezing temperatures $T_F(x)$.[4,5] The occurrence of a glass state in these molecular systems was first predicted by Fischer and Klein.[6] It is thought to result from a frustration of the effective multipole interaction between CN pairs into which not only the orientations of the multipoles but also the bond vectors enter.

So far no attention has been paid to the question of whether the ordering in $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$ is strictly static or rather of a relaxational type. We therefore report measurements of the freezing temperatures over ten decades of frequency. In order to cover this wide frequency range we used three experimental methods: dielectric (87 Hz–100 kHz), ultrasonic (megahertz range), and neutron-scattering (terahertz range) measurements. As above, $T_F(x, \omega)$ is defined by the maximum of the appropriate susceptibility as a function of temperature.

The experiments were carried out on single crystals of $(\mathrm{KBr})_{1-x}(\mathrm{KCN})_x$ which have been grown in the crystal growth laboratory of the University of Utah from zone-refined KBr and KCN materials. Single crystals with concentrations $x = 0.01$, 0.04, 0.14, 0.25, and 0.5 were investigated. (The concentrations 0.01, 0.25, and 0.50 refer to the composition of the melt; the figures 0.04 and 0.14 have been determined by chemical analysis. The composition of the melt in these cases was 5% and 20%, respectively.) Pieces of the same single crystals were used in the different experimental setups.

The dielectric constant ϵ' measured at four frequencies is shown in Fig. 1 for $x = 0.14$. With decreasing temperature ϵ' increases in a Curie-like fashion and for all concentrations $x \geq 0.04$ passes through a maximum at temperatures $T_F(x, \omega)$. The maximum of the dielectric loss is always reached significantly below T_F.

The ultrasonic data were obtained in a conventional setup with a quartz transducer attached to the samples by using the pulse-echo-overlap method. Figure 2 shows the temperature dependence of the elastic constant c_{11} for $x = 0$, 0.01, 0.04, 0.14, and 0.25 at 10 MHz. All samples containing CN^- ions exhibit a minimum at a characteristic temperature T_F. For higher CN con-