Comment on "Critical Cone in Phonon-Induced Desorption of Helium"

In a recent Letter, Taborek[1] presented experimental results for the angular variation of the flux and velocity distribution of helium desorbing from a cryogenic surface. He found the flux to be sharply peaked toward the normal, and the velocity distributions to vary strongly with final angle. He interpreted this as resulting from the ejection of the helium atoms from the attractive well due to single-phonon annihilation.

In this Comment I point out that collisions between desorbed molecules should have occurred to the extent to largely mask their initial velocity distributions. The observed data, rather than giving information on pure phonon-adsorbate dynamics, reflects gas-phase multiple-collision effects as well.

Taborek[1] correctly points out the negligible chance of collisions between a desorbate and chamber background molecules. However, the density of desorbates from such a 100-nsec desorption is more than sufficient to allow desorbate-desorbate collisions. During the desorptions the density is similar to that of a three-dimensional (3D) gas. For a typical velocity of about 0.28 km/sec, the extent of the cloud of gas at the end of 100 nsec is about 0.003 cm above the surface. For about one-monolayer desorptions[2] at $10^{15}/\text{cm}^2$, this leads to a gas density of $3\times10^{17}/\text{cm}^3$, equivalent to 8 Torr. The collision cross section for He-He collisions over the temperature range 3 to 25 K is fairly constant at about 1.5×10^{-15} cm^2, for large-angle deflection (diffusion-theory cross section).[3] This predicts a mean free path of about 0.002 cm, implying that typically a helium atom will experience about 1.5 large-angle deflections during the 100-nsec desorption. Reference 3 discusses this further, and shows how to calculate the collisions for times greater than 100 nsec. The latter contribute around 3.5 collisions/helium typically, for a total of 5 collisions/helium. The small-angle-deflection cross section is even larger.

These collisions must occur, and will grossly perturb the initial velocity and angular distributions. This was experimentally demonstrated for the similar case of monolayer desorption of D_2 from tungsten surfaces over 30 nsec.[4]

The collisions should produce a partial adiabatic cooling, analogous to a supersonic expansion.[5] A nozzle operated continuously at 8 Torr, with area of 0.02×0.04 cm^2, would give a final Mach number of 3.[5] Though the helium desorption is not continuous and should give less cooling, the effect should be similar.[3]

The analogy with a supersonic expansion can be extended to estimate the observed velocity distribution. Assuming 1D expansion, the final flux distributions would be expected to be[5]

$$\rho(v)\,dv\,d\Omega \propto v^3 \exp\{-(m/2kT_f)[(v_z-v_f)^2+v_x{}^2+v_y^2]\}, \quad T_f=T_i(1+M^2/3)(1+M^2/3)\,(\text{for helium}), \qquad (1)$$

with v the velocity, Ω the solid angle, m the helium mass, k the Boltzmann constant, T_i the initial and T_f the final gas temperatures, and v_f the final 1D flow velocity. With the choices $M=2.2$, $v_f=0.28$ km/s, and T_i equal to the surface temperature of 8 K, a qualitative agreement can be found between Eq. (1) and Fig. 2 of Ref. 1. Both the rapid falloff of intensity, and the decrease in velocity, with increasing final angle are predicted. The qualitative features of Taborek's[1] distributions closely parallel those found for fast D_2 desorptions,[4] and the latter discusses the qualitative aspects of the effects of several collisions on velocity distributions.

In conclusion, it is clear that each helium atom should experience several major collisions, suggesting that the observed distributions of Taborek are seriously perturbed by desorbate-desorbate collisions.

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Received 7 May 1984
PACS numbers: 68.45.Da

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