Edge melting of tetramethyltin submonolayers physisorbed on graphite basal planes

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Mössbauer-spectroscopic studies of $Sn(CH_3)_4$ physisorbed on graphite show that as $T \rightarrow T_m$ (2D) an abnormal gradual decrease occurs in the average size of the solid adsorbate islands. The anomaly starts at $\sim 0.9 T_m$ (2D), where the solid two-dimensional (2D) island structure is incommensurate with the substrate. From these results we infer that edge melting preempts the 2D melting in the same way as it does in the recently reported case of $Fe(CO)_5$ on graphite.

I. INTRODUCTION

The melting of two-dimensional (2D) films absorbed on alien substrates has been extensively studied for over a decade,[1-4] and yet several aspects of this phenomenon are still controversial.[5] The KTNHY theory (Kosterlitz and Thouless,[6] Nelson and Halperin,[7] and Young[8]) predicts that 2D melting is a continuous transition. A controversially different theory by Chui[9] proposes that the melting is a first-order transition. This view has received much support from subsequent theoretical work.[10-14] The experimental situation is just as controversial and intriguing as the theoretical one. Some results show that 2D melting is apparently a first-order transition,[15-17] while other experiments indicate that the transition is continuous.[18]

Premelting effects in bulk solids were predicted a long time ago. Recent experiments have actually demonstrated the existence of *surface melting* in solids[19] and multilayer-thick adsorbed films:[20-22] When the surface of a bulk melts, a stable layer of liquid is formed that wets the solid-vapor interface at temperatures below the triple point. During the last year, evidence has been presented for the existence of *edge melting*—the "1D" analog of the surface melting in 2D physisorbed islands.[23,24]

In this paper we report on Mössbauer-spectroscopic studies of some dynamical effects associated with premelting phenomena in physisorbed films of $Sn(CH_3)_4$ on a graphite substrate.

We use the Mössbauer spectroscopy which was proved to be a sensitive tool to probe the mobile states of adsorbed films and observe changes in mobility.[16,17,25] As the melting temperature T_m is approached, a sudden drop in the Mössbauer spectral intensity, $I(T)$, from the Debye-Waller behavior is expected as the solid constituents become mobile.[16] In monomolecular-layer films the diffusion in the film plane can be detected, and the sensitivity of the method to diffusion may be affected by geometrical factors when the observation is done in the out-of-plane direction.[17]

In this paper we show that a submonolayer film of $Sn(CH_3)_4$ physisorbed on graphite deviates from the harmonic Debye-Waller dependence of $I(T)$ when the temperature of the film approaches the two-dimensional melting point T_m(2D). Above ~ 85 K, as $T \rightarrow T_m$(2D) $\simeq 95$ K, the deviation increases gradually. This resembles the behavior of the $Fe(CO)_5$-graphite system recently studied.[24] In the latter case the dynamic behavior of the adsorbed film as $T \rightarrow T_m$(2D) was explained as *edge melting*, analogous to surface melting in bulk solids.

In the following sections the experiment is described, and the possibility that edge melting actually occurs in the $Sn(CH_3)_4$ islands physisorbed on graphite basal planes is discussed.

II. EXPERIMENTAL DETAILS

The adsorbed films studied were $Sn(CH_3)_4$ (tetramethyltin, or TMT). TMT is liquid at room temperature; its vapor pressure at 296 K is 98 Torr. It freezes at 218.5 K into a bulk solid. The molecule has spherical symmetry with Sn located at the center. The dimensions and angles between bonds are specified elsewhere.[26-29]

Mössbauer absorbers were prepared for submonolayer surface densities of molecules adsorbed on exfoliated graphite (Grafoil) sheets: A 10-g stack was prepared from these sheets which allowed the γ rays to pass in the direction parallel to the Grafoil sheets, or, after 90° rotation, perpendicular to it. The stack was cleaned in the usual way (~ 950 °C, 4 h in vacuum) and transferred under helium atmosphere into the cryostat cell. The specific surface area of this stack was determined by N_2

adsorption at liquid-nitrogen (LN_2) temperature. The stack was charged at room temperature with $Sn(CH_3)_4$ to the desired surface coverage.[30] The surface density at monolayer coverage differs by only 3% for room temperature and low temperature. The sample temperature was set by controlled flow of LN_2.

In an early experiment[25] we have studied surface coverages of 0.3, 0.5, 0.6, 0.7, 0.8, and 1.0 of TMT on graphite. In the present work we repeated the experiment in more details in a limited temperature range ($85 < T < 95$ K), close to T_m(2D) and for 0.5, 0.7, and 0.9 coverages. Examples of Mössbauer spectra were shown in Ref. 25: A Mössbauer spectrum of $Sn(CH_3)_4$ displays a Lorentzian singlet as expected from the high symmetry of this molecule. With a Ba $^{119}SnO_3$ γ-ray source (~ 24 keV), we find an isomer shift of 1.22 mm/sec at 78 K relative to SnO_2 as has been reported.[31] Spectra for each coverage, temperature, and direction were accumulated for as long as was necessary to obtain $\sim 10^6$ counts per velocity channel. This resulted in an acceptable statistical spread in view of the low apparent effective thickness $t = nd\sigma_0 f_a$(2D) for the films of this adsorbate. Here n is the number of Mössbauer nuclei per cm^3; d is the absorber thickness (in cm); f_a is the Mössbauer-Lamb fraction, and σ_0 is the cross section for the nuclear process in the ^{119}Sn. The accumulated Mössbauer velocity spectra were analyzed for the spectral parameters from which the background-corrected spectral intensity $I(T)$ could be determined for each temperature in both perpendicular and parallel configurations to the substrate sheets. In Fig. 1 we illustrate the results of our spectral analysis as $\ln I$ versus T for configurations parallel and perpendicular to the sample plane, for several coverages (0.9-, 0.7-, and 0.5-layer films). The customary representation of the

Mössbauer temperature-dependent spectral intensity is based upon the harmonic Debye model: $I(T) \propto \exp(-2W)$. The intensity of the spectra measured in the perpendicular configuration [Fig. 1(a)] can be described by a simple Einstein model.[24,30] For the parallel direction [Fig. 1(b)], deviation from the harmonic Debye-Waller approximation is apparent, where $\ln I(T)$ decreases with increasing temperature in a nonlinear way. Any deviation from the harmonic behavior is generally associated with anharmonic-dynamical effects, or with diffusion.[17,32]

III. DISCUSSION

The discussion focuses on the abnormal nature of the results illustrated in Fig. 1: Figure 1(a) shows the variation of the Mössbauer spectral intensity in the perpendicular configuration with temperature variation of the mean squared displacement (MSD) in the direction perpendicular to the film. Figure 1(b) illustrates the temperature variation of the intensity when the γ-ray wavevector k is parallel to the sample surface. These results are obviously different from those depicted in Fig. 1(a): As the temperature increases, and $T \rightarrow \sim 0.9 T_m$(2D), a gradual deviation from the exponential variation commences.

In order to understand this behavior, we describe here the structure of $Sn(CH_3)_4$ film physisorbed on graphite, as inferred from our earlier x-ray-diffraction results:[25,33] For coverages above ~ 0.6 of a monolayer, one diffraction peak is observed at $2\theta = 15.6°$ for $T < 85$ K. In this case the adsorbate molecules are arranged in a triangular $(\sqrt{7} \times \sqrt{7})R 19°$ structure, commensurate with the substrate with a lattice parameter $d = 6.55$ Å. At $T \simeq 85$ K this peak disappeared and a new peak at $2\theta = 15.2°$ was observed; this was interpreted as commensurate-to-incommensurate transition. For the lower coverages below 0.6 layer film, there is no evidence of any structural transition, and from the diffraction peak at $15.2°$ it is inferred that the film at all temperatures is incommensurate with the substrate surface. In the temperature range of interest the adsorbed films at all surface coverages are in triangular and incommensurate state, with $d = 6.72$ Å resulting in a slight misfit of $\sim 3.3\%$. These results suggest that as $T \rightarrow T_m$(2D), the solid islands become gradually smaller, and consequently the observed spectral intensity weakens accordingly. Since there is no apparent Mössbauer line broadening as $T \rightarrow T_m$(2D), we believe that the transition is first order. We shall discuss the possibility that the experiment actually shows that the edges of the adsorbed islands melt as the 2D melting is approached. Melting around spots at various places across the island[34] ("pinning") would have resulted in a severe destruction of the coherence length which is evidently not the case as suggested by our x-ray results.

The temperature and configurational dependence of the Mössbauer intensity of TMT films at the proximity of the 2D-melting temperature is surprisingly similar to the recently reported behavior of iron pentacarbonyl (IPC), $Fe(CO)_5$ adsorbed on (0001) graphite basal planes.[24] As
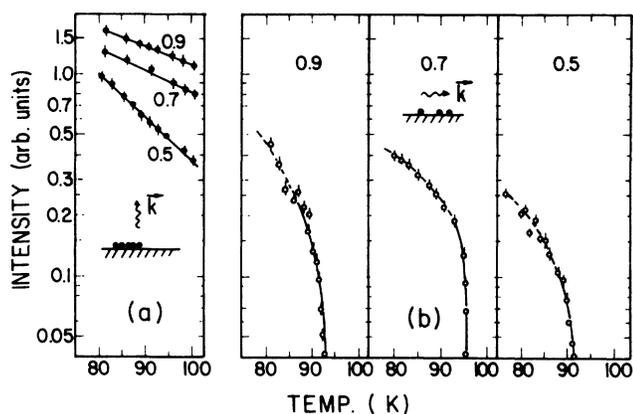
FIG. 1. The temperature dependence of the Mössbauer spectral intensity $I(T)$ above ~ 80 K is shown for 0.9, 0.7, and 0.5 coverages of $Sn(CH_3)_4$ adsorbed on (0001) graphite basal plane; (a) is in direction perpendicular to the graphite plane. The solid lines through the experimental data follow a simple harmonic model of small vibrations of the adsorbed molecule (Ref. 25). (b) is in parallel direction. Here the solid line is a result of a power-law fit and the edge-melting model described in the text.

inferred from neutron-diffraction results, close to $T_m(2D)$ the condensed state of IPC-graphite forms an incommensurate, nearly $(\sqrt{7} \times \sqrt{7})$ phase.[35] For comparison the Mössbauer results for the latter system are shown in Fig. 2. The abnormal dynamical behavior across the surface film inferred from the information depicted in Fig. 2(b), namely the enhanced nonlinear deviation from exponential behavior in the parallel configuration above $\sim 0.9 T_m$, was interpreted as resulting from edge melting.

The edge-melting phenomenon is an inherent property of a 2D solid physisorbed on a clean substrate, in analogy with surface melting in bulk solids. If the boundary between a 2D solid and its vapor is wetted by the melt liquid, the free energy of the system at $T < T_m(2D)$ is lowered. A thin peripheral band of solid liquifies and becomes the solid-vapor interface. Such an effect was clearly indicated by computer simulations of a model two-dimensional system.[36,37] The thermodynamics of this low-dimensional system is essentially the same as for surface melting.[38] As a consequence of equilibrium conditions, the variation of the thickness $L(T)$ of the liquid layer with temperature follows a power law 39.40:

FIG. 2. The temperature dependence of the Mössbauer spectral intensity $I(T)$ above 150 K for a 0.9, b 0.7, c 0.5, and d 0.3 coverages of Fe(CO)$_5$ adsorbed on (0001) graphite basal plane. (a) $I(T)$ in the direction of the γ-ray $\mathbf{k}$ vector perpendicular to the graphite plane; the solid line through the results was obtained like in Fig. 1(a). (b) $I(T)$ for $\mathbf{k}$ vector parallel to the surface film: Lateral premelting anomaly starts above $\sim 0.9 T_m(2D)$. The dashed lines indicate the Debye-Waller behavior extrapolated from lower temperatures; the solid lines through the experimental results were calculated as in Fig. 1(b).

$L \propto t^{-1/p}$, where $t = (T_m - T)/T_m$; p is the interaction exponent. In 2D systems with smooth edges and nonretarded dispersion forces, $p = 4$. Thermal fluctuations are particularly important in "one-dimensional" systems. Infinitely long 1D boundaries are rough at all finite temperatures. Roughening is expected to decrease the effective interaction exponent to $p = 3$.

The results depicted in Figs. 1 and 2 were analyzed in the following way: The solution of the diffusion equation for a 2D classical liquid, for large diffusion distances is given by the classical self-diffusion function[17,32]

$$F(\mathbf{r},t) = \frac{1}{2\pi r \sqrt{\pi D t}} \exp\left[-\frac{r^2}{4Dt} \right], \quad (1)$$

where $\mathbf{r}$ is the displacement vector in the surface plane, and D is the diffusion coefficient in 2D. $F(\mathbf{r},t)$ is substituted into the general expression for resonance cross section of a Mössbauer atom in a classical liquid [see Eq. (37), Ref. 32]. Integration over r in 2D results in the following useful expression for the on-resonance cross section for Mössbauer recoilless absorption in condensed 2D films:

$$\sigma_{\text{res}}(\theta, T) = \frac{2\hbar K}{\Gamma} \left[1 + \frac{2\hbar D k^2}{\Gamma} \sin^2\theta \right]^{-1/2}, \quad (2)$$

where

$$K = \frac{\sigma_0 \Gamma}{4\hbar} \exp\left[-\frac{\hbar^2 k^2}{8 m k_B T} \right] \quad (3)$$

is the on-resonance Debye-Waller (factor) contribution; k_B is the Boltzmann factor, $\mathbf{k}$ is the γ-ray wave vector, and θ is the angle of $\mathbf{k}$ relative to the surface-normal. σ_{res} is thus a product of two factors. Evidently, with $\theta = \pi/2$, a considerable reduction in the on-resonance cross section is expected when surface diffusion starts. At the same time $\sigma_{\text{res}}(\theta = 0)$ should behave like a regular temperature-dependent exponent (Debye-Waller factor).

In a system with N_0 physisorbed molecules, the measured Mössbauer intensity is proportional to $N_0 \sigma_{\text{res}}$. Suppose that as a consequence of a premelting process, $n < N_0$ molecules become mobile; since the diffusive broadening $(2\hbar k^2 D)$ is several orders of magnitude larger than the natural linewidth,[32] the contribution of these n molecules to the spectral intensity $I(\theta, T)$ vanishes (when observed in direction parallel to the film). Therefore, the observed spectral intensity of the Mössbauer line will be proportional to $N_0 - n(T)$:

$$I(T,\theta) \propto [N_0 - n(T)] \frac{2\hbar K}{\Gamma} + n(T) \sigma_{\text{res}}(\theta, T). \quad (4)$$

$I(T, \theta = 0)$ reflects the MSD of all N_0 molecules in the configuration perpendicular to the film plane; $I(T, \theta = \pi/2)$ is the intensity with $\mathbf{k}$ parallel to the film, due to $(N_0 - n)$ molecules. Equation (4) represents an anisotropic spectral intensity which can be used to follow the edge melting in a physisorbed solid island: This equation relates the observed Mössbauer spectral intensity to $n(T)$. The temperature dependence of the number of

molecules which have become mobile is model dependent.

For not too high diffusion rates,[17] $I(T, \theta=0)$ is proportional to $N_0(2\hbar K/\Gamma)$, and $I(T, \theta=\pi/2)$ to $[N_0-n(T)](2\hbar K/\Gamma)$: The actual temperature dependence of $n(T)$ may be complicated because the peripheral shape of an island and connectivity of the various islands for the different coverages can be rather complicated,[41] nevertheless the results in Figs. 1 and 2 can be described qualitatively by an island of a simple circular shape.[42] (The actual island periphery may be complicated and very long and may involve thermal fluctuations).

Let the island be of radius R_0 at $T < T_m(2D)$, and made of spherical molecules of radius r_0. Since the temperature range through which the anomalous deviation from the Debye-Waller (DW) temperature dependence is observed is narrow, one can assume that throughout this range $\exp(-2W) \simeq$ const (except for corrections described below). The deviation from DW due to $n(T)$ is described in the following way. The spectral intensity, $I(T, \theta=\pi/2)$ is proportional to the area occupied by molecules of the solid island: $I(T, \pi/2) \propto N_0 - n(T)$ $\propto S(T)$; $S(T)$ is the average area of an island of the radius R for a given coverage. Let S_0 be the area of the island at some $T_0 < T_m(2D)$; T_0 is the temperature at which the spectral area measured with **k** parallel to the surface area deviates from the DW behavior: $S_0 = \pi R_0^2$. Let L_j be the width of the peripheral band of the island which becomes liquid. The remaining solid will have an area $S_j(T_j > T_0) = \pi(R_0 - L_j)^2$. The temperature dependence of the spectral intensity, $I(T_j, \theta=\pi/2)$ will be proportional to $S_j(T_j)/S_0$. But even without melting, the DW factor, K_j, is temperature dependent [Eq. (3)]. The DW independent intensity is then

$$I(T_j, \theta=\pi/2) \propto \frac{S_j(T)}{S_0} = \frac{K_j}{K_0}\left[1-\frac{L_j}{R_0}\right]^2 . \qquad (5)$$

Now, $L_j = bt_j^{-\psi}$ and $l_j = L_j/R_0 = at_j^{-\psi}$: $t_j = (T_m - T)/T_m$. The curve fitting the experimental results depicted in Figs. 1 and 2 for $I_{exp}(t_j, \pi/2)$ is given by

$$I_{calc}(t_j, \pi/2) = (K_j/K_0)(1-at_j^{-\psi})^2 , \qquad (6)$$

where a and ψ are fitting parameters.

If ψ is known, a line can be drawn through the data in Fig. 1. ψ can be determined in the following way: From Eq. (6), $l_j = at_j^{-\psi} = 1 - [I_{calc}(t_j; \pi/2)K_0/K_j]^{1/2}$. Now instead of I_{calc} we express l_j by I_{exp}: From Eq. (6), $l_j = 1 - (I_{exp}K_0/K_j)^{1/2}$. $\log l_j$ can now be plotted versus $\log t_j$. ψ can now be evaluated from $d(\ln l_j)/d(\ln t_j)$ $= -\psi$. For TMT-graphite [Fig. 1(b)] the analysis yields $\psi = 0.29 \pm 0.3$ for all coverages; for the IPC-graphite system [Fig. 2(b)] this analysis yields $\psi = 0.25$ for 0.9 coverage, 0.20, 0.11, and 0.11 for the coverages 0.7, 0.5, and 0.3, respectively. Substituting ψ in Eq. (6) yields $I_{calc}(T)$ [solid line in Figs. 1(b) and 2(b)].

The reasons for deviations of ψ from the predicted value of 0.25 have been discussed above and in previous works.[23,24,38] The low value of ψ for the low coverages of the IPC films, for instance, may be due to interfacial energy term due to the quasiliquid layer around the solid is-

land, which depends on the interface peripheral length. For circular islands, the edge length decreases with increasing l_j. If the size of the island is large, it is reasonable to assume that the edge length will not change much for small l_j values and the $\psi=\frac{1}{4}$ power law holds. One must also bear in mind that such systems are not purely 2D and that the effects of substrate defects on the remaining molecules may become prominent when the island is small. The results indicate that the exponent is constant above $\sim 0.9T_m$. Unfortunately, the way ψ can be determined introduces uncertainties in the absolute value of the exponent, since t_j depends on $T_m(2D)$; the 2D melting temperature may vary slightly; for instance, substrate inhomogeneities can result in a higher melting temperature. Thus, in this analysis t_j is an adjustable parameter.[23]

Nevertheless, the results presented in Figs. 1 and 2 show that the temperature dependence of the Mössbauer spectral intensity can be represented reasonably well by a simple mean-field theory. The basic power law is derived for 2D van der Waals continua, which does not account for effects due to substrate registry, fluctuations, or intrinsic structures of the 2D solid. The experimental range of the power law corresponds to a liquid depth of several atomic distances; yet, the exponent does not change, and within the accuracy of our measurements the transition is smooth and with no evidence of discrete melting of individual "strips" of atoms. The smoothness we observe is probably due to continuous width variations.

Recently a molecular-dynamics simulation was used to examine how the pinning of edge particles affects the nature and dynamics of the melting transition for a 2D solid:[34] The adsorbed species are confined to a nearly 2D surface formed by the surface crystalline planes of a substrate. These finite-size substrate planes are small and have surface imperfections. Edges of substrate plateletes and defects can pin adsorbate molecules at fixed positions, and consequently, the 2D melting transition is expected to broaden. It seems, however, that adsorbates physisorbed on substrates and forming nonregistered 2D structures do not experience much of the pinning effects described above. For instance, neon on graphite shows edge melting:[23] This system (Ne-graphite) was selected since the 2D solid is incommensurate with the (0001) planes of the graphite,[43] and its melting transition is strongly first order.[15,44] In the other two cases where edge melting is believed to be observed, the submonolayer films were incommensurate with the substrate planes, at least close to the 2D melting point: With $Fe(CO)_5$ and $Sn(CH_3)_4$ on graphite—the first undergoes orientational disordering and becomes slightly incommensurate below $\sim 0.8T_m(2D)$, and the second undergoes a commensurate-incommensurate transition at ~ 85 K. In all these cases a first-order melting transition has been observed; a slightly broadened transition could be ascribed to pinning on account of surface steps or impurities. These effects have been discussed elsewhere:[23,25] Recent measurements of $Sn(CH_3)_4$ physisorbed on MgO show a similar behavior of the temperature dependence of the Mössbauer spectral intensity close to $T_m(2D)$.[45] The

meaning of the fact that the precursors of the 2D melting in all these examples are very similar (see Figs. 1 and 2) and practically independent of coverage lends strong evidence for edge melting.

In conclusion, it seems that edge melting occurs in "floating" films physisorbed on substrates. A dynamical view of edge melting has been presented in this paper. This premelting process occurs at the interfaces between 2D solid and 2D vapor. The effects of dimensionality of the boundaries between adsorbed (sub)monolayer phases are not clear yet and depend on the shape of the "2D" islands.

ACKNOWLEDGMENTS

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