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Rough and Vicinal Surfaces of Helium-4 Crystals. Mobility Measurements

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By studying the transmission coefficient of ultrasonic waves perpendicular to the solid-liquid interface, we have measured the mobility of vicinal surfaces of ^{4}He crystrals for different crystallographic orientations and in the temperature range from 0.38 K to 1 K. The results show an angular dependency of the vicinal mobility as predicted by theory. Further, in comparison with rough interfaces, a spectacular decrease for the mobility of vicinal surfaces due to the decreasing number of moving sites is in good agreement with preceeding results for the phonon term. It has been observed for the first time for the roton term.

1. INTRODUCTION

Crystal growth of ^{4}He is determined by the microscopic state of the crystal surface which depends on temperature and on the angle θ of the surface normal with respect to a high symmetry axis. Recent papers[1,2,3] have emphasized the different characteristics (surface tension, growth coefficient...) of a ^{4}He crystal surface as a function of its crystallographic orientation θ. At a temperature lower than the roughening transition temperature T_R, a high symmetry plane ($\theta = 0$) is an atomicaly smooth facet and has almost zero mobility. In contrast, for rough surfaces the mobility is high. Nozières and Uwaha[4] first pointed out the anisotropic behaviour of the mobility in the intermediate domaine between a rough surface and a facet. We recall that a surface tilted by small angle θ compared with a facet appears as a succession of terraces separated by well defined steps only if the step width $w(T)$ is smaller than the distance d between steps.

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Such surfaces are called stepped or vicinal. At large tilt angles, steps overlap and the surface becomes rough. More precisely, Balibar *et al.*[1] have used the roughening theory by Nozières and Gallet[5] to predict that the crossover from a facet to a rough surface occurs at a critical angle $\theta_c \sim a/3w(T)$, where a is the lattice spacing (or step height); if $\xi(T)$ is the correlation length of terraces, they defined the step width as $4\xi(T)$. From a study of crystallisation waves on ^{4}He crystal surfaces, they found experimentally[2] θ_c to be equal to $\sim 3°$ at $T = 0.2$ K. As far as the mobility is concerned, it is limited by collisions of the moving interface with the excitations of the liquid (phonons and rotons) and of the solid (phonons).[6] Thermal phonons and rotons, which scatter from a moving vicinal surface, behave as wave packets having respective sizes of $\lambda(T) = (hc/2.7k_B T)$, where c is the sound velocity and $\sim h/p_0 \sim 3$ Å, where p_0 is the momentum at the roton minimum. At low temperatures, the phonon wavelength can become comparable to d. Thus, phonons can interact with more than a single step. In this case, $\theta_c \sim a/\lambda(T)$. On the other hand, the roton wavelength is always smaller than d and $\theta_c \sim a/3w(T)$ remains a good criterion to define the mobility crossover from rough to vicinal surfaces.

1.1. Mobility. Background

Almost 15 years ago, first measurements of the mobility of rough ^{4}He crystal surfaces were made.[15] Both experiment and theory have shown that the growth resistance, the inverse of the mobility, of a rough surface can be described by the equation:

$$K^{-1} = A + BT^4 + Ce^{(-\Delta/T)} \tag{1}$$

where $\Delta \sim 7.2$ K is the roton gap. The first term is believed to depend on the quality of the crystal. For good crystals, our experiment shows that A is negligible. The second term is due to phonon scattering and is dominant for $T < 0.55$ K. Calculations[8] in the ballistic regime show that the phonon contribution to the growth resistance K_p^{-1} lies between $3.06T^4$ and $3.32T^4$ cm $\cdot$ s^{-1}. The discrepancy in these values arise from an extremely small (6%) anisotropy with the crystal orientation, due to the anisotropy of the sound velocity. The third term is attributed to roton scattering. Also in Ref. 8, the roton growth resistance far above the roughening transition is expressed as $K_r^{-1} = (2\pi p_0^4/h^3 \rho_s)(1-\xi)\exp(-\Delta/T)$, where $(1-\xi)$ represents a correction due to anormalous "Andreev" reflection of rotons at the interface. The prefactor C in Eq. (1) takes a value of 1.18×10^5 cm $\cdot$ s^{-1}, with $\xi = 0.5$.

For vicinal surfaces the situation is somewhat more complex. In a model for the mobility of vicinal surfaces at $T < 0.6$ K, Nozières and

Uwaha[4] consider the crystal surface to be made of an array of individual steps. They show that the interface growth resistance K^{-1} and the step growth resistance K_s^{-1} satisfy the equation: $K^{-1} = K_s^{-1}/(N_s a)$, where the step density per unit length along the surface $N_s = (\sin\theta)/a$. Consequently, for small θ, K goes linearly to zero as θ approaches a facet orientation. They further argue that Compton scattering of phonons by individual steps free of defects gives rise to a T^3 behaviour for K_s^{-1} (and therefore for K^{-1}) at low temperatures. For the intermediate situation, where $\lambda(T)$ is greater than d, Nozières and Uwaha give a semi-empirical interpolation relation.

In a study of the roton contribution to the growth resistance Edwards *et al.*[7] propose that rotons are scattered by kinks which are of the same size as the roton wavelength. Then the roton contribution to the growth resistance is given by:

$$K_r^{-1} = (f/u)\, N_k a/(m_4 N_s \zeta) \tag{2}$$

where f is a "force" on a kink, u is the local drift velocity of kinks, N_k and N_s are the kink and step densities respectively, and ζ is a dimensionless factor which depends on the numbers per unit length of positive and negative kinks on a step. Thus we have that K_r also tends to zero as θ approaches 0°. Further, for vicinal surfaces it is believed[9] that there can be single kink per site, irrespective of temperature, since each atom is a likely candidate to become a kink. We therefore have N_k and N_s (at large angles) to be $\sim 1/a$. By assuming that kinks on a step form a Bose gas and by including a kink-kink interaction Edwards *et al.*[7] show that the prefactor of the roton term in Eq. (1) is now only temperature dependent and varies as $C'(T) = C(1 + 1.14T)$.

2. EXPERIMENT

Experimental details have been reported elsewhere.[10] We recall that we measure the transmission of acoustic waves through the ^{4}He solid-liquid interface at 10 MHz between 0.38 K and 1 K for different crystal orientations. Our measured ultrasonic signal varies linearly with the incident power. The orientations far from that of a facet are determined from measurements of the longitudinal and transverse sound velocities in the solid.[11] Further, we use the recent results of the angular variation of the mobility[9] to determine our sample orientations close to a facet. Our crystals were grown slowly at ~ 1 K from pure ^{4}He containing less than 5 ppb of ^{3}He.

The principle of the experiment can be summarized as follows. An incident ultrasonic pressure wave, of frequency ω, creates a chemical potential difference $\Delta\mu$ between the solid and liquid phases which gives rise to a mass current flow J through the interface. The resulting motion of the interface is governed by $\Delta\mu = (K^{-1}/\rho_s)\,J + i(m_I\omega/\rho_s\rho_l)\,J$, where m_I is the surface inertia of the crystal. We note that because of the superfluid nature of the liquid and the extremely small latent heat, the thermal contribution to the chemical potential difference can be neglected.[12] The amplitude of the energy transmitted from one phase to another not only depends on the acoustic impedances of the solid and the liquid, namely z_s and z_l respectively, but also on the complex acoustic impedance of the interface $Z_i = (\rho_s \rho_l^2/\Delta\rho)\{K^{-1} + (im_I\omega/\rho_l)\}$.[10] The sound transmission coefficient is given by:

$$\tau(T, \omega) = 4z_s z_l/\{(z_l + z_s) + (z_l z_s/Z_i)\}^2 \tag{3}$$

At $T < 0.55$ K, $\tau \sim (|Z_i|)^2$. Since the excitations in each phase progressively disappears with decreasing T, the crystal surface moves more and more freely, that is, K^{-1} tends to zero. However, the surface inertia acts against the mobility of the surface, restoring partially the transmission. At low temperatures and high frequencies, the effect of m_I on τ becomes preponderant. This experiment is limited to low frequencies (10 MHz) and the effect of the surface inertia on the transmission coefficient is therefore negligible at our minimum temperature of 0.4 K. The phonon and roton terms of the mobility are determined from a measure of τ. A study of the surface inertia will be presented later.[13]

3. RESULTS AND DISCUSSIONS

In Fig. 1 we show typical measurements of the coefficient of sound transmission $\tau(T, \omega)$ from the liquid to the solid as a function of T^{-1} for samples of different crystallographic orientations. We have corrected the

TABLE I

Mobility Coefficients Determined by Fitting our Data to Eqs. (2) and (3). For Angles Greater than 10°, the Average Values Are Represented, after a Series of Measurements

Sample No.	θ (degrees)	B ($\mathrm{cm \cdot s^{-1}\,K^{-4}}$)	$C \times 10^{-5}$ ($\mathrm{cm \cdot s^{-1}}$)	$(K/K_{\mathrm{rough}})_p$ at $T = 0.4$ K	$(K/K_{\mathrm{rough}})_r$ at $T = 0.9$ K
1	~1	730	35	0.005	0.024
2	~1.8	420	19	0.009	0.045
3	~2.8	154	8	0.023	0.106
4	~6.5	10.6	1.3	0.324	0.654
5	Angles > ~10	3.5 ± 0.8	0.85 ± 0.25	1	1

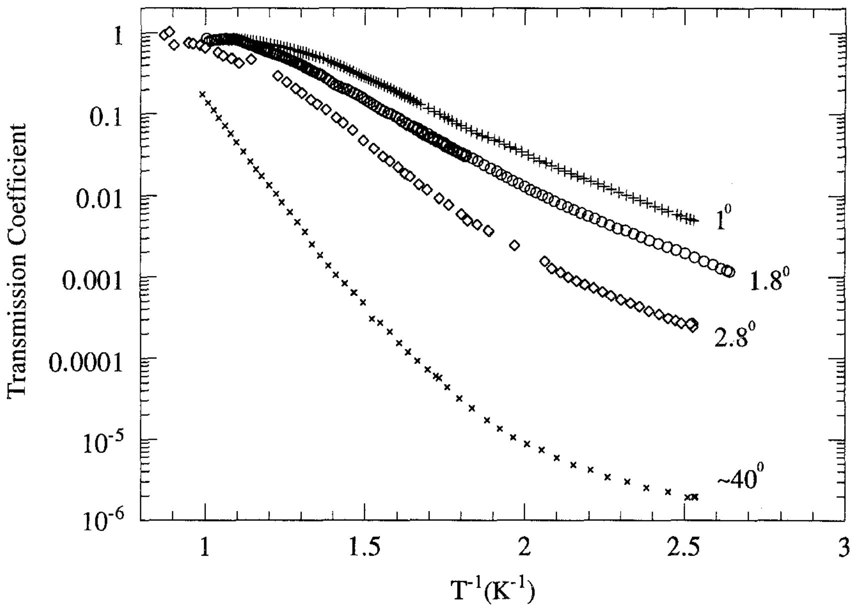

Fig. 1. Representation of the measured transmission at a frequency of 10 MHz as a function of T^{-1} for four samples. The sample corresponding to an orientation of $\sim 40°$ represents typical results of a rough surface. Samples 1, 2 and 3 represent vicinal surfaces and correspond respectively to surface orientations of $\sim 1°$, $\sim 1.8°$ and $\sim 2.8°$.

raw measurements for attenuation due to the liquid and solid bulk. The data are fitted with the Eq. (3). The coefficients obtained from our fits are summarized in Table I.

— *Mobility of Rough Surfaces:* For the phonon contribution to the growth resistance, we determined $K_p^{-1} = (3.5 \pm 0.8)\, T^4\ \mathrm{cm \cdot s^{-1}}$ above 0.38 K. This result is in good agreement with experiments of Keshishev *et al.*[15] which gives the prefactor of the phonon contribution to lie between $2.6\ \mathrm{cm \cdot s^{-1}\, K^{-4}}$ and $3.4\ \mathrm{cm \cdot s^{-1}\, K^{-4}}$ for temperatures below 0.47 K. Further, Wang *et al.*[16] determined a prefactor of $2.79\ \mathrm{cm \cdot s^{-1}\, K^{-4}}$ at temperatures close to 0.25 K. All of these results can be understood within the ballistic theory of Bowley and Edwards.[8]

In Fig. 2, we show only the roton contribution to the growth resistance as a function of T^{-1} for a typical sample. The dotted line are the experimental results of Bodensohn *et al.*[14] (also see Keshishev *et al.*[15] and Castaing *et al.*[17]). The dashed line corresponds to the theoretical equation for the roton growth resistance[8] as discussed in Sec. 1.1. We obtain a good fit to our data with $(1 - \xi) \sim 0.5$. This agrees with previously determined values.[7,8] The solid line corresponds to a fit deduced from Eq. (2) and it

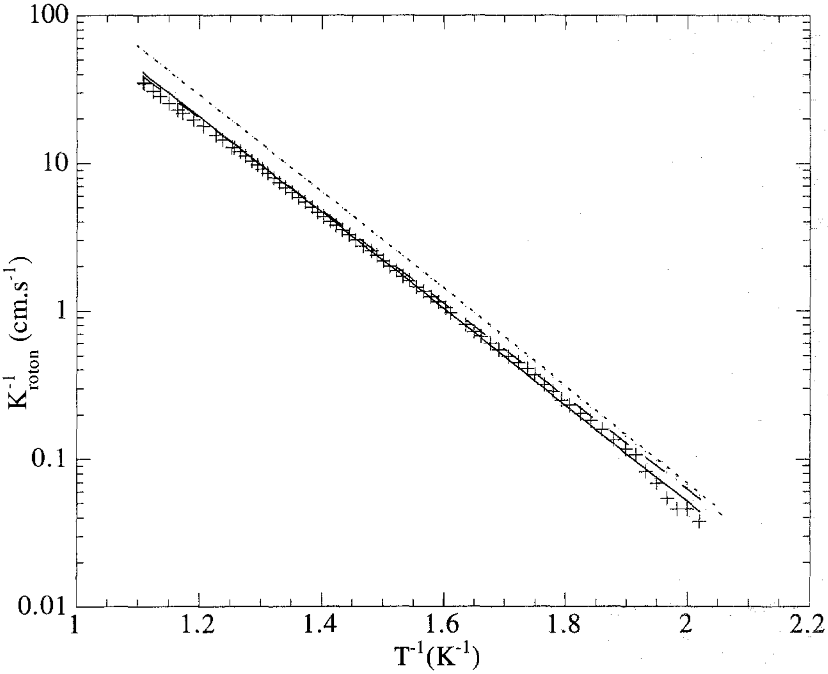

Fig. 2. Roton contribution to the mobility of a rough surface. The crosses represent the our experimental data. The dotted line, the dashed line and the solid line correspond respectively to: (a) a fit deduced from the experimental results of Bodensohn *et al.*[14] The growth resistance takes the form $K^{-1} = 2.66 \times 10^5 \exp(-7.6/T)$ cm · s^{-1}; (b) the theoretical equation of Bowley and Edwards,[8] with $\xi = 0.5$ (see text). The growth resistance is given by $K^{-1} = 1.16 \times 10^5$ $\exp(-7.2/T)$ cm · s^{-1}; and (c) the calculations of Edwards *et al.*[7] (see text, Eq. (2)). The growth resistance for rough surfaces has the form $K^{-1} = 0.606 \times 10^5(1 + 1.14T)$ $\exp(-7.2/T)$ cm · s^{-1}.

takes into account the temperature variation of $C'(T)$ described above. There is an apparent agreement between the data and the model. However, we emphasize that the temperature variation of the $C'(T)$ is small. Our results therefore do not give a convincing proof as to the validity of the temperature variations of the prefactor as predicted by the theory.

— *Mobility of Vicinal Surfaces:* Figure 3 shows the growth resistance from 0.4 K to 1 K for four orientations of vicinal surfaces. Clearly, compared to rough surfaces the mobility decreases dramatically in the whole temperature range as the surface orientation tends towards a facet. The orientations of all four vicinal samples are determined with respect to the [0001] facet for which the roughening transition temperature is 1.30 K. In determining our fits, we supposed that the phonon and roton contribution to the mobility follow respectively a T^4 and an $\exp(-\Delta/T)$ behaviour as

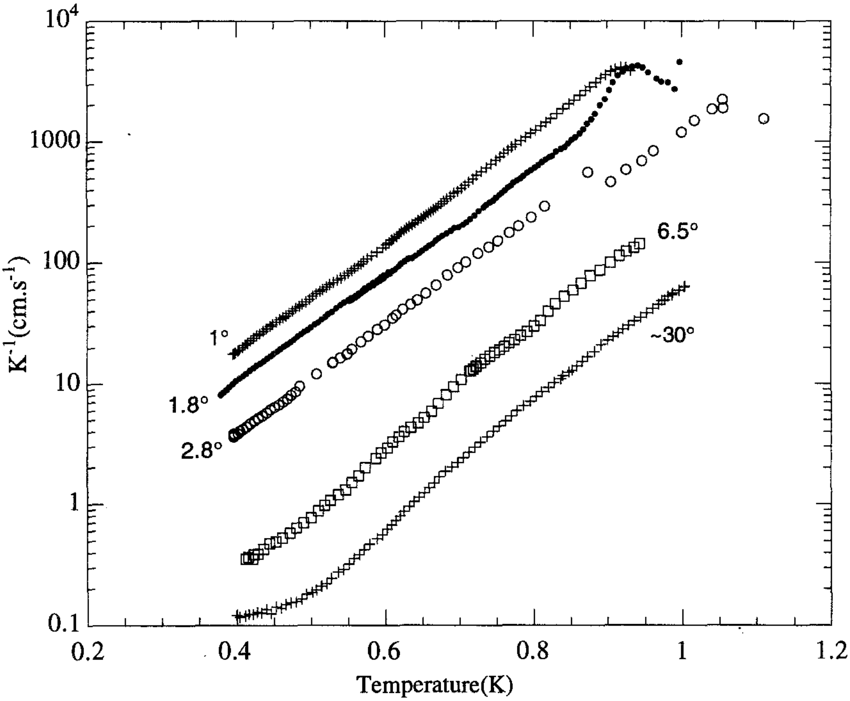

Fig. 3. Mobility of vicinal surfaces as a function of temperature for samples of different crystal orientations.

given in Eq. (1). For vicinal surfaces these behaviours in temperatures need not be necessarily be true as indicated in the Nozières and Uwaha theory. The large coefficients (see Table I) found for vicinal surfaces therefore do not necessarily correspond to a physical process. To analyse the low temperature dependence of the mobility, we trace the phonon contribution to the mobility as shown in Fig. 4. At these temperatures the roton contribution is negligible.

Our data shows that the low temperature growth resistance can also be fitted to a T^n dependency, where $n = 4.4 \pm 0.22$. Andreeva *et al.*[3] first observed a similar behaviour in the temperature range from 0.35 K to 0.45 K and for orientations as large as 9.5°. Recent experiments[9] also corroborates these temperature dependencies. It should be noted that the phonon wavelength is ~70 Å at 0.4 K. For an orientation of ~1°, the distance between steps is ~170 Å. This leads us to believe that our experiment does not fulfill the conditions of an ideal step-phonon scattering regime as described by Nozières *et al.*[4] and therefore we do not observe a T^3 behaviour.

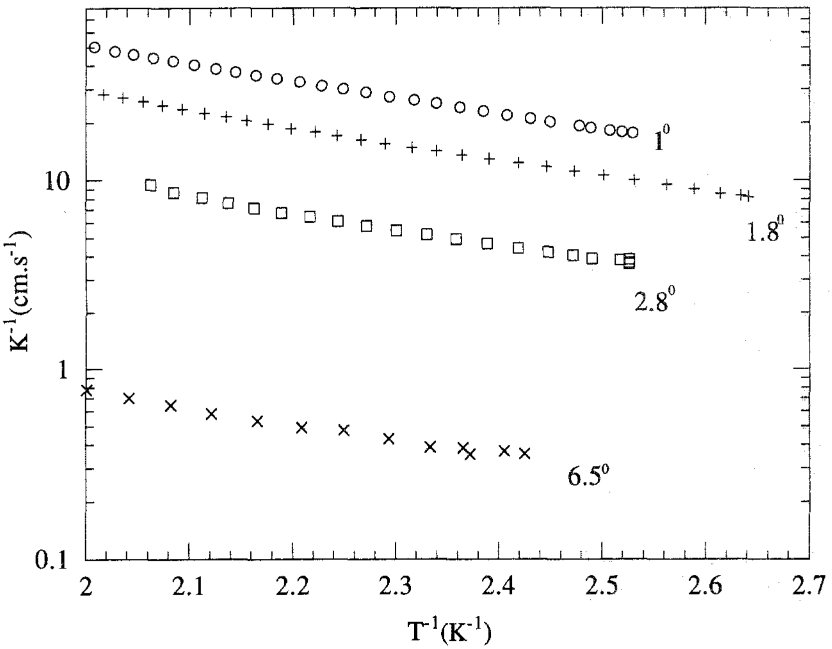

Fig. 4. Phonon contribution to the mobility of vicinal surfaces as a function of the inverse of temperature. The experimental data can also be fitted with a T^n behaviour, where $n = 4.4 \pm 0.22$ or, as suggested by Andreeva *et al.*,[3] with an $\exp(-\varepsilon/T)$ behaviour, where $\varepsilon = (1.94 \pm 0.09)$ K.

In Figs. 5a and 5b we represent the angular variation of the mobility of vicinal surfaces at different temperatures. For convenience, we have normalised the mobility of vicinal surfaces with respect to the mobility of rough surfaces. Two distinct features are put to evidence. Firstly, we shall consider the low temperature regime ($T < 0.55$ K) where rotons contribution is negligeable and thermal phonons dominate. In Fig. 5a the curve corresponding to $T = 0.5$ K is situated to the right of the curve corresponding to $T = 0.4$ K. For clarity, we have omitted the curves for intermediate temperatures. These results suggest that the departure from the mobility of rough surfaces occurs at a critical angle which increases with temperature. We can explain this with a qualitative argument. In order for a phonon to scatter of an independent step, we must have $\lambda(T)/d \ll 1$, which gives a critical angle $\theta_c \sim (a/\lambda(T)) \sim T$. However, for the temperature regime where phonons contribution is negligeable and rotons dominate, Fig. 5b shows that the critical angle decreases with temperature. This behaviour can indeed be explained using the definition $\theta_c \sim a/3w(T)$ since the step width $w(T)$ increases with temperature.

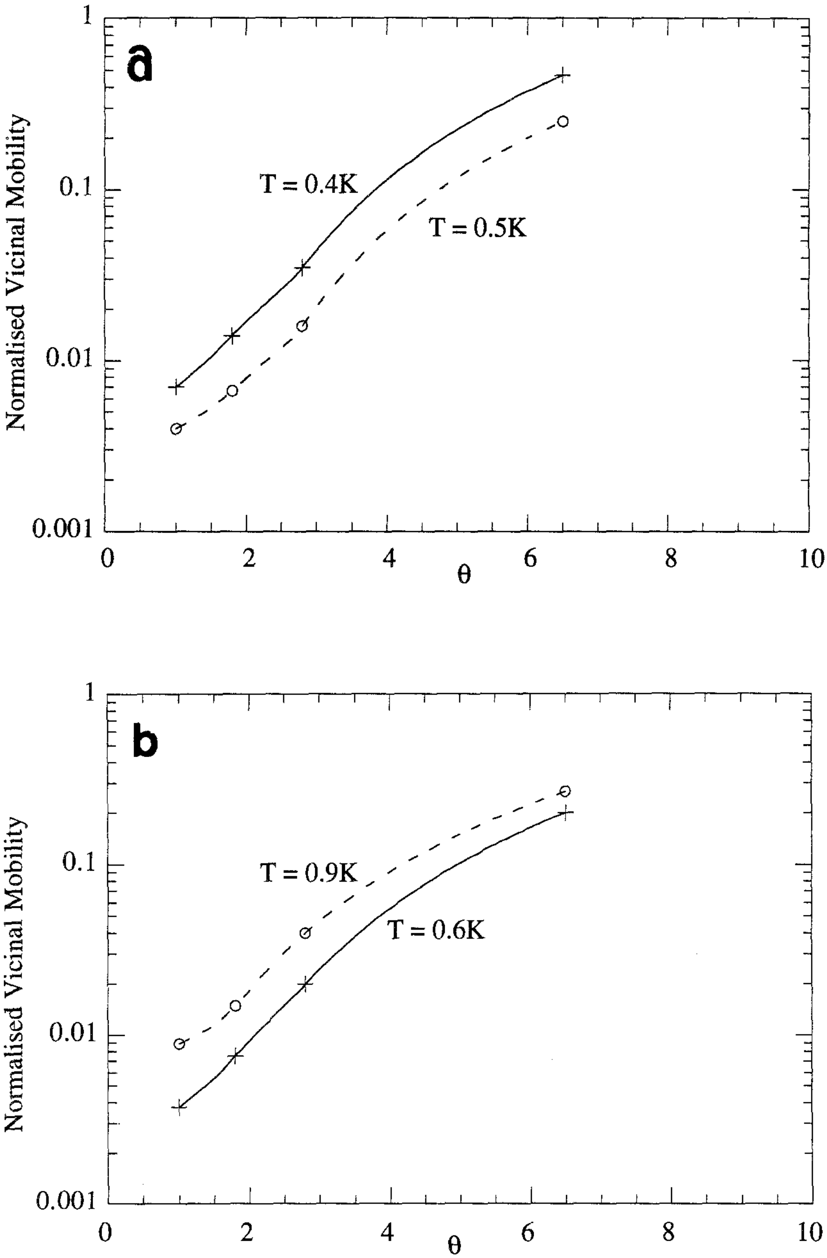

Fig. 5. Ratio of the mobility of vicinal surfaces to the mobility of rough surfaces as function of the orientation angle for $T < 0.55$ K in (a) and for $T > 0.55$ K in (b). We interpolate the data points in order to highlight temperature dependencies.

4. CONCLUSION

We have measured the mobility of vicinal surfaces for temperatures ranging from 0.4 K to 1 K. The low temperature ($T < 0.5$ K) data are in good agreement with other studies,[3,9] using different experimental techniques. Our results show that the mobility is greatly reduced in comparison with the mobility of rough surfaces in the whole experimental temperature range. We have put in evidence for the first time that the roton contribution to the growth resistance is also enhanced for vicinal surface. Further, our results indicate that the transition of the mobility from a vicinal to a rough surface takes place at a critical angle which has two-fold behaviour depending on whether phonons or rotons give a dominant contribution to the mobility. The critical angle decreases with temperature for phonons and it increases with temperature for rotons. In light of the experimental results there is a need for an elaborate theory for vicinal surfaces.

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