LETTER

Reply to Jee et al. and Israelachvili and Drummond: Lubricant films do not fluidize in intermittent stick-slip friction

Jee et al. assert (1) that the effect we measure—what happens to a lubricant film thickness in individual slip events during intermittent stick–slip sliding (2)—was already measured long ago (3, 4), and with better resolution. We are bemused by this assertion because in the works cited (3, 4) not only is intermittent stick–slip friction, with its characteristic saw-tooth pattern, not shown or measured, but their inability to measure what happens during the individual, fleeting slip events—the key finding of our own study (2)—is explicitly stated in the papers themselves (below).

The comment by Jee et al. that what was earlier measured holds "generally for other confined liquids" (1) is, however, interesting. In their papers (3, 4) these authors report an essentially identical effect for a number of simple fluids. This is surprising, because the characteristic dimensions of the molecules in these fluids differ by up to almost fourfold, so that one might have expected them to measure somewhat different effects for the different lubricants. Their unexpected finding that all effects were similar may be viewed in the light of a subsequent (2003) paper (5) by the Granick group. In this paper it is shown clearly that, because of the way mica sheets were cut in the Granick laboratory—at least before 2003—the surfaces used in their surface-forces experiments could be significantly contaminated by platinum nanoparticles (5) [a problem resolved in other laboratories, where clean surfaces were shown to be the rule (6)]. It is tempting, therefore, to attribute the results of these cited studies (3, 4) not so much to shearing of different lubricant films, as to the shearing in all cases of the platinum nanoparticles contaminating their mica surfaces [a contamination endemic at that time in the Granick laboratory (5)], whose height (approximately 2 nm) is indeed of similar magnitude to the film thicknesses they report.

Whatever was actually measured in these earlier studies (3, 4), however, their inability to shed light on what happens in the fleeting slip events in intermittent stick–slip friction is clearly stated by the authors themselves. To quote from Demirel and Granick (3): "In extending this work it will be very interesting to contrast time-resolved changes of friction and dilatancy during the course of individual stick–slip cycles. . . . Unfortunately at this point . . . it is not yet technically feasible—due to the very small amplitude of dilatancy oscillations—to acquire real-time measurements of dilatancy during the course of a single stick–slip cycle." Because measuring the dilatancy (or rather its absence) during the course of individual slip events was indeed the essential new finding and key point of our work (2), we fail to understand why Jee et al. (1) claim that our results were anticipated by these earlier studies (3, 4), in the light of the fact that they were patently incapable of making such measurements.

Israelachvili and Drummond (7) assert that we have interpreted our study, which shows clearly that no fluidization occurs during the brief slips in intermittent stick–slip friction across a model lubricant film, in a way that seems to rule out the mechanisms of wall-slip or intralayer slip, which we suggest as possible alternatives to lubricant fluidization. These authors cite in support their earlier study on sliding across squalane (8), as well as the studies noted above (3, 4), as having a bearing on the dilatancy effect in such intermittent stick–slip sliding.

Israelachvili and Drummond (7) cite studies (3, 4) that do not measure (and are not capable of measuring, as demonstrated above) what happens during the fleeting individual slip events in intermittent stick–slip friction. Because this is the key new finding of our own study, we think that they may have misunderstood and therefore misrepresented our results. Our measurements are able to follow for the first time the behavior of sheared lubricant layers during the very brief (approximately 20 ms) slip events in intermittent stick–slip sliding. We show clearly that, within our 1 Å resolution, the dilation expected from a fluidization of the lubricant film during the slip events, which would correspond to an ~10% density change arising from its shear melting, does not occur. Although we are careful to qualify our findings as applying to the model lubricant system that we study (2), it is worth recalling that for most liquids (water being a prominent exception) the solid phase is denser than the liquid phase, so that dilation on melting is the rule. Nonetheless, there is full scope—within our 1 Å resolution in the change of the lubricant film thickness—for intralayer or wall-slip to occur, as we suggest, and as qualitatively indicated earlier (e.g., reference 2 in ref. 7, with which we have no disagreement) and in simulation studies (9, 10). Such intralayer or wall-slip could readily occur within a 1 Å increase in surface separation (reference 2 in ref. 7) without the need for complete fluidization of the sheared films (which for our model lubricant would entail a dilation considerably greater than 1 Å). Israelachvili and Drummond's (7) assertion that our results preclude the mechanisms of wall-slip or intralayer slip is thus clearly incorrect.

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