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Melting anomaly and occurrence of a liquid—liquid phase transition

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A liquid—liquid phase transition is discussed in view of singularity in a melting line. Possible existence of a liquid—liquid phase transition has been discussed thus far in relation to existence of a maximum in a melting curve [1]. We claim that it is the existence of a breakpoint, rather than a maximum, that can potentially influence the occurrence of a liquid-to-liquid transition. Our in situ synchrotron x-ray diffraction measurements revealed that the molecular crystal tin tetraiodide has an unusual melting curve. It rapidly increases with a pressure up to about 1.5 GPa, at which it abruptly breaks. The melting curve becomes almost flat on the high-pressure side of the breakpoint (with a slight maximum at about 3 GPa). Kechin proposed the differential equation [2], which can be handled with an aid of Padé approximation, to capture the overall aspect of such a melting curve. We could show that the melting curve obtained as the solution to the differential equation became closer to the actual melting curve with the improvement of the degree of the Padé approximation. Kevin's proposal thus seems to be appropriate to handle the differential equation provided the slope of the melting line is everywhere continuous. We believe that this is not the case for the melting curve in question at the breakpoint, as inferred from the nature of breakdown of the Kraut—Kennedy and the Magalinskiii—Zubov relationships [3]. We propose that the breakdown of these relationships is rather a manifestation of such a qualitative change in the intermolecular interaction as electronic (bonding) transformation in the liquid state. The breakpoint may then be a triple point among the crystalline phase and two liquid phases, whose existence has been confirmed.

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