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固—液界面阴离子集团构型的密度泛函理论研究

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摘要 通过键合不同数量的H2PO4^-离子构造了H4P2O8^2-和H4P4O16^8-两种阴离子集团。利用密度泛函理论对它们的构型进行优化并计算其振动频率和喇曼散射效率。 测量了固—液界面边界层的喇曼光谱并与理论计算结果进行对比分析，进一步研究 了阴离子集团的结构、聚合及脱水等过程。实验和理论研究均表明阴离子二聚体是 KDP晶体的生长基元，他们在固液界面将进一步形成多聚体分子集团，而离子集团 的脱水过程则发生在距KDP晶体表面约50μm的固—液界面边界层中。随着生长层向 界面的推进，这些分子集团将变得越来越有序。本文的研究结果对确定晶体生长界 面动力学过程、发展和完善晶体生长理论有重要意义。

关键词 密度泛函理论 喇曼光谱法 晶体生长 构型

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Study on the Configuration of Anion Clusters in Solid-Liquid Interface of KDP Crystals with Density Functional Theory

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Abstract Two kinds of anion clusters, i.e.,H-4PO-4~- and H-4P-2O-~28 are constructed, by bonding two and four anions. The optimized configurations, vibrational frequencies and Raman activities of H-2PO- 4 , H-2PO-8~2- and are obtained by using density functional theory. The Raman scattering spectra in the solid- liquid interface boundary layer are measured , and compared with those of theoretical calculation for studying the configuration and the processes of polymerization and dehydration etc . Both the experimental and theoretical results indicate that dimmer clusters may be the growth units in crystal growth , which can polymerize further and form polymer cluster on the solid-liquid surface . The dehydrating process of dimmer and polymer clusters starts at a growth layer about 50 jura apart from the solid-liquid surface of the KDP crystal. With approaching to the crystal- liquid surface, the molecular .cluster becomes more and more orderness . The results obtained are of significance in identifying the surface kinetics process and developing more sophisticated crystal growth theories

Key words density functional theory RAMAN SPECTROMETRY CRYSTAL GROWTH CONFIGURATION

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