Fingerprints of angulon instabilities in spectra of matrix-isolated molecules

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Synopsis We use the concept of the angulon quasiparticle to simulate ro-vibrational spectra of light molecules trapped in superfluid ^{4}He nanodroplets. In this work, we demonstrate that previously predicted angulon instabilities can explain the anomalous broadening of individual spectral lines measured for CH_3 and NH_3 in experiment. Transitions to the instabilities are accompanied by a creation of one phonon with non-zero angular momentum.

The recently introduced angulon model [1] based on a quasiparticle approach to the problem of a rotating molecular impurity immersed in a many-body medium has already affirmed its applicability to ^{4}He nanodroplets. It showed a good agreement with experimental data on renormalization of rotational constants for a wide range of both light and heavy molecules [2], [3] and also allowed to describe dynamical properties of molecules in nanodroplets that were studied in femtosecond laser-induced alignment experiments [4]. A quasiparticle approach primarily simplifies a theoretical consideration of such complex systems and makes it possible to understand the main peculiarities of interactions of molecules with superfluid helium environment.

One of the most curious predictions of the angulon theory is the existence of so-called angulon instabilities [1], taking place for the angulon states with non-zero angular momentum. A transition to the instability corresponds to a transfer of angular momentum λ from the molecule to helium. Here we demonstrate that these instabilities appear exactly for those levels which are involved in the transitions showing the anomalous broadening (>50 GHz) in the experimental ro-vibrational spectra of the ν_3 band of CH_3 [5] and NH_3 [6] molecules.

Fig.1 shows the angulon spectral function illustrating a set of angulon states. Angulon levels are labeled as $L_{jk\lambda}$, where L is the total angular momentum of the system, j and k give the angular momentum of the molecule and its projection on the molecular z-axis respectively and λ is the angular momentum of helium excitations. The instability appears in the area inside the black frame. The transition to this instability (indicated by the blue arrow) from the lower angulon level leads to the creation of one phonon with $\lambda = 3$ while the molecule undergoes the forbidden transition to the molecular state 1_1. The value $\alpha = 1.12$ reproduces the following line width of the $^R R_1(1)$ line: 43 GHz for CH_3 and 46 GHz for NH_3.

Thus, this work provides a strong evidence that the angulon instabilities have already been observed in experiments on molecules in superfluid helium.

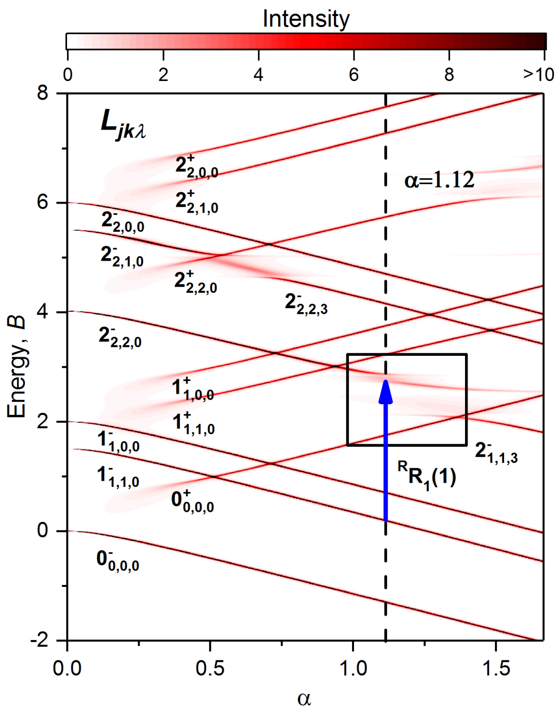

Figure 1. The angulon spectral function for CH_3 as a function of the dimensionless energy $\widetilde{E} = E/B$ (B is a rotational constant) and the α parameter regulating the strength of the CH_3-helium interaction. The blue arrow corresponds to the transition to the instability observed in the experiment and assigned as the $^R R_1(1)$ line.

References

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