ESRF	**Experiment title:** Nuclear inelastic scattering and nuclear forward scattering studies of ^{57}Fe adatom dynamics and magnetism on top of topological insulator Bi_2Se_3	**Experiment number**: HC-1667
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Report:

There is a continuous interest in the properties of electronic surface states that must set up when topological insulator (TI) and a vacuum meet. These electronic states might react with the Fe adatoms in different way than do "normal" electron states as those on top of non TI material. Indeed, they may, e.g. give rise to the enhanced magnetism of magnetic adatoms, as suggested in [Wray, Li]. They also might, possibly, be observed by Fe adatoms vibrations. This we wanted to study by means of NIS on ID18: our aim was to observe Fe adatoms vibrations on top of topological insulator Bi_2Se_3 and to compare these vibrations with those of Fe adatoms, but deposited on top of non TI material, but with similar surface structure, Bi_2S_3. Although almost no difference in Fe vibration for these materials was observed, the spectra may be separated into iron monolayer-like vibration DOS and some low energy contribution, possibly due to individual Fe atoms vibrations. No such finding was reported by now.

The samples, single crystalline Bi_2Se_3 and Bi_2S_3, were first placed in UHV and cleaved with the help of adhesive tape leaving atomically clean surface of quintuple layer. They were then cooled to T=180K, covered with submonolayer of ^{57}Fe adatoms (1Å, i.e. ca. 50% of a monolayer) and immediately transferred, all stages in situ, to the experimental chamber. NIS measurements were carried out with 2meV X ray resolution (FWHM) in the energy transfer range -10 to 50 meV and at lowest possible temperature 150K. NFS spectra were occasionally measured to ensure that the submonolayer Fe is not magnetically ordered.

The results, shown on Fig. 1, suggest that DOS calculated from the experimental data, are almost the same for both samples. Thus, in the further discussion below only DOS calculated for Bi_2Se_3, as a representative for 1Å Fe layer on quintuple layer (QL), will be used.

We have further compared our experimental DOS with the phononic density of states of those materials that can be relevant to our case, especially with iron vibration on the surface or in bulk [Slezak], or with Fe monolayer vibrations on tungsten [Stankov]. We have found that the higher energy part of our spectrum coincides with DOS of Fe surface vibrations, provided some small shift to lower energies of this Fe surface DOS is done, the procedure fully justified in view of Fe-Bi_2Se_3 lattice mismatch [Stankov]. The comparison of our results with those for Fe surface layer are shown on Fig. 2 and it is clear that although the higher energy part of our DOS fully coincides with Fe vibration spectrum, there are definitely low energy vibrations other than those specific to Fe spectrum. DOS for these vibrations was estimated by subtraction of our experimental DOS and Fe DOS, and shows characteristic dome-like peak at ca 10 meV, shown on Fig. 2.

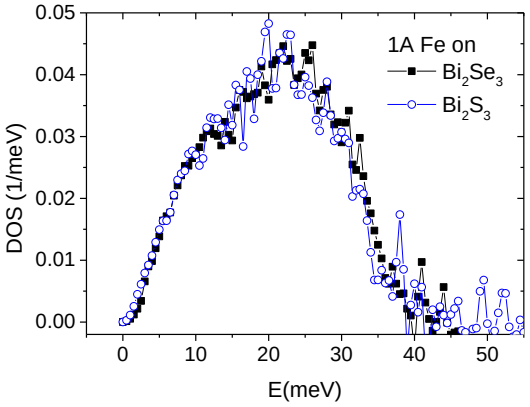

Fig1.

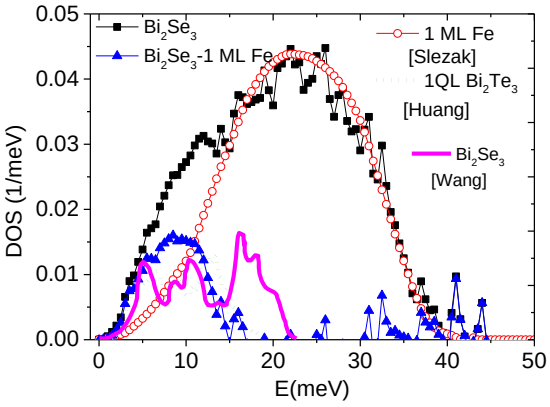

Fig. 2

Fig. 1: The results of NIS measurements on Bi_2Se_3 (TI) and Bi_2S_3 (non TI) covered with 1Å of ^{57}Fe. **Fig. 2**: Comparison of Fe on Bi_2Se_3 DOS with Fe surface vibrations (hollow circles), experimental Bi_2Se_3 (full curve) and theoretical Bi_2Te_3 surface (dotted line). The result of subtraction of Fe surface DOS from the experimental DOS is shown by triangles.

Thus, the scenario after this result might be that some part of Fe adatoms form the well defined mono-layer on part of QL and the rest Fe adatoms are either loosely clustered or in single atomic form; those remaining Fe atoms give rise to the low energy vibrations. Depending on Fe-surface coupling, either vibrations of the quasi-free Fe atoms (very small QL-Fe interaction), QL vibrations (very strong interaction: Fe vibrations would then reflect surface vibrations), or some intermediate situation occurs.

There are conflicting reports as to how the Fe adatoms behave on the TI surfaces. Although it was well established that there are two Fe surface positions, either of comparable surface energy [Honolka] or differing considerably energetically (75.8 meV) [Li], "infrequent hopping events of a single Fe atom" [Honolka] between these two positions, even at as low as 0.3K were observed. It would suggests a very limited Fe-host interactions i.e. Fe acted as free atoms, with only their outer s electrons transferred into host surface, imposing some force on Fe atoms that can be observed by Fe atom vibrations. In such a case, our low E part of DOS would describe weakly linked Fe atoms, the feature possibly irrelevant to the host properties.

On the other hand, the same authors [Honolka] report a strong Fe-d orbitals hybridization with electronic states from the surrounding crystal and influenced by the host crystal fields; it leads to an in-plane magnetic easy axis. At higher Fe coverages (> 10%), atomic multiplet features disappears due to variable size Fe clusters formation. [Honolka]. Thus, in this scenario, Fe adatoms behave as electron donors [Schlenk] rather strongly interacting with the host material. In such a case Fe vibrations might reflect QL vibrations.
Finally, this is not impossible that individual Fe adatoms migrate into QL and form some compound with e.g. Se (or S) atoms, like superconducting $FeSe_2$ [Hsu]. In fact, Fe migration below the surface was reported [Schlenk], although at higher temperature than used here.

Since neither vibration of individual Fe atoms nor surface Fe-Se phonons were reported in literature, we have only compared our spectra with measured bulk Bi_2Se_3 and the calculated, surface, Bi_2Te_3 DOS; apparently, both curves roughly coincide with our results, at least in their concentration at low energies. In any case, further studies of these effects are needed.
In conclusion, we have measured vibration spectra of 1Å thick Fe layer on top of topological insulator Bi_2Se_3 and non-TI Bi_2S_3. We have found that both spectra, are nearly identical and, in their higher energy part, coincide with the Fe surface vibration spectrum. The broad peak at low energies is either due to individual Fe atoms, Bi_2Se_3 (Bi_2S_3) vibration or the vibration of some Fe based compound. This project will be continued.

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