Evaluation of the contribution of higher-order electron-nuclear interactions to the NQR frequencies using ^{115}In spectra in InSe

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ABSTRACT

The ^{115}In NQR spectra in layered InSe single crystals for four spin transitions, the average values of which only approximately satisfied the ratio $\nu_1:\nu_2:\nu_3:\nu_4$ =1:2:3:4 were studied. It was found that for higher transitions a deviation is observed. In particular, it is shown that additional lines are observed in the multiplet groups for higher-order transitions, which, in our opinion, is associated with the presence of a hexadecapole interaction with the gradient of the crystal electric field.

1. INTRODUCTION

One of the urgent problems of interest for nuclear quadrupole resonance (NQR), which deserves the attention of some researchers, was and is a problem of assessing the contribution of the electron-nuclear interactions of higher order in the NQR frequency, particularly in hexadekapole interaction.

Based on the results of existing studies of all fundamental electrostatic interactions of ordinary substances, it can be argued that the nuclear hexadecapole bond is still the most uncertain, and this problem requires further study. Since the first observations of the interaction of the nucleus with the environment through the electric moment, many experiments have been carried out to study both static and time dependences of this phenomenon. However, despite a large number of experimental studies, none of the works on this problem has unequivocal confirmation of the hexadecapole interaction in NQR [1].

2. EXPERIMENTAL RESULTS

The hypothesis of the existence of a nuclear hexadecapole interaction was put forward by researchers even before the discovery of NQR, however, an experimental study of this phenomenon was carried out much later and continues to the present, and, it can be noted that, unfortunately, researchers still do not have unambiguity in explaining the results obtained. The main reason for this is that the hexadecapole interaction is vanishingly small and, according to a number of experimenters, is on the verge of detecting measuring technology. It should be noted that the determining condition for the existence of the hexadecapole interaction is the large value of the quadrupole moment of the nuclei under study, which, on the other hand, leads to masking of the manifestation of the small hexadecapole interaction. This is precisely the main problem for the interpretation of experimental NQR spectra of layered crystals, in particular, such as InSe.

To interpret the origin of the multiplicity of the NQR spectra in a layered InSe crystal, we studied the experimental ^{115}In NQR spectra. Layered InSe compounds are an interesting object for research, since the insignificant energy of formation of structural defects existing in them leads to the fact that, during crystal growth, appear various modifications of the crystal structure - polytypes. The latter, in turn, significantly affect the band structure of the layered compound, and phase transformations in the system of polytypes can lead to instability of the physical properties of the crystal with a change in temperature.

Atomic nuclei with a large mass number are characterized by an irregular distribution of electric charges in their volume. At a significant value of the quadrupole moment, this leads to additional excitations of the quadrupole energy levels due to the presence of the hexadecapole moment of the nucleus and its interaction with the anisotropic electric field of the crystal.

Experimental NQR spectra were obtained on pre-annealed single-crystal InSe samples grown by the Bridgman method, with a volume of 0.25 cm^3 using an NQR spectrometer with continuous scanning and Zeeman modulation. The experimental spectra were recorded at a temperature T = 290 K. In this work, we observed complex multiplet NQR spectra (Fig. 1, a-d), which differ significantly in shape for four spin transitions $\pm1/2 \leftrightarrow \pm3/2$, $\pm3/2 \leftrightarrow \pm 5/2$, $\pm 5/2 \leftrightarrow \pm 7/2$, $\pm 7/2 \leftrightarrow \pm 9/2$. The magnetic induction vector ($\mathbf{B}_1$) is directed along the crystal layers, and the modulating magnetic field induction vector $\mathbf{B}_M$ is directed along the optical axis $\boldsymbol{c}$.

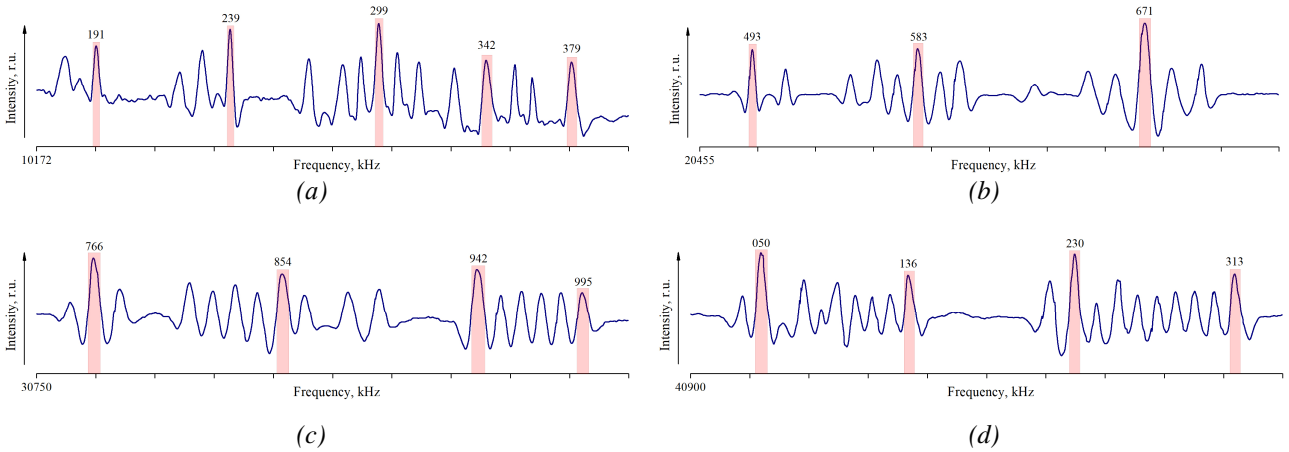

Figure 1. Form ^{115}In spectra in InSe for the transitions ±1/2 ↔±3/2 (*a*); ±3/2 ↔±5/2(*b*); ±5/2↔ ± 7/2 (*c*); ± 7/2 ↔ ± 9/2 (*d*).

The obtained spectra clearly show three multiplets, indicating the presence of a mixture of polytypes in the crystal under study [2]. The studied NQR spectra for all four transitions consist of multiplet groups with a fine structure with a constant splitting value of 10.3 ± 0.3 kHz.

Since the crystal structure of InSe has axial symmetry of the electric field gradient, which means that the asymmetry parameter $\eta = 0$, and therefore the ratio of the transition frequencies should be related as $\nu_1:\nu_2:\nu_3:\nu_4$=1:2:3:4. Experimentally measured frequencies for the characteristic lines of the spectrum of four resonance transitions showed that such a relation is fulfilled only approximately, namely, it was found that this relation is valid only for the frequencies ν_1 and ν_2, while for high frequencies (ν_3, ν_4), a deviation is observed: 10.25 ; 20.5; 30.76; 41.05.

The observed discrepancy in the NQR spectra cannot be explained by a change in the polytype structure or asymmetry parameter η [3] with an increase in the frequency of energy transitions, as well as the appearance of charge density waves in the crystal, the latter is more likely for lower temperatures than room temperature.

We believe that such a difference in the NQR spectra for resonance transitions of higher orders is due to the fact that for atomic nuclei with a large mass number and a significant value of the quadrupole moment, the hexadecapole moment is characteristic, which leads to additional excitations of the quadrupole energy levels, so there is a probability of the appearance of multiplet lines in the NQR spectra of InSe [3].

3. CONCLUSIONS

Thus, the complex structure of the NQR spectra - multiplet groups in InSe for resonance transitions of higher orders (±5/2 ↔ ± 7/2; ± 7/2 ↔ ± 9/2), characterizes not only the presence of polytypes, but also indicates the presence hexadecapole interaction is provided by *p*-, *d*- and *f*-electrons in a layered crystal and a strongly distorted figure of the charge distribution of ^{115}In nuclei. By analogy with the studies of lutetium nuclei carried out in [4], we believe that in our case the electrons of the *d*- and *f*-shell of the indium atom are excited by coordination interactions, an additional contribution to the nuclear-hexodecapole interaction may appear in the form of additional lines in the multiplet NQR spectra.

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