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COUPLED QUADRUPOLE-PHONON EXCITATIONS: INELASTIC NEUTRON SCATTERING ON VAN VLECK PARAMAGNET $PrNi_5$

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1. INTRODUCTION

In rare-earth (RE) compounds single-ion excitations such as magnetic dipole and quadrupole excitons are possible due to transitions between levels of 4f-electrons in a crystal electric field (CEF) [1]. It is well-known that the inelastic neutron scattering technique has proved to be a very powerful method of studying the spectrum of elementary excitations, especially in metallic compounds. However, whereas it is relatively easy to see magnetic dipole excitons applying to the magnetic neutron scattering, it is very difficult to induce by neutrons electronic quadrupole transitions between different CEF-levels. They can be observed only in the hybridization regime with phonons. For example, an anticrossing of the phonons and electronic branch was observed with the help of the nuclear neutron scattering in the insulator $PrAlO_3$ in the region of structural phase transition at 151K [2]. To our best knowledge, there were not such measurements in metallic systems. In the present paper we report the experimentallic investigation of coupled quadrupole-phonon excitations in the intermetallic RE compound $PrNi_5$ at temperature 8K.

2. THEORY OF COUPLED EXCITATIONS IN RE COMPOUNDS

Theory of coupled excitations in RE compounds was considered in many papers (see review [1]). Here we use a new approach developed in paper [3] on the basis of the double-time-Green-function (GF) method. The technique of differentiation with respect to the two times allows us to obtain the Dyson equation for the one-particle GF in a simple way. Comparison [3] with inelastic neutron-scattering results for $PrAl_2$ in the ferromagnetic phase shows excellent agreement between the calculated coupled magneto-vibrational modes and experimental spectrum. In the paramagnetic phase hybridization of quadrupole excitons and phonons in metallic systems was predicted.

The differential neutron cross section is expressed in terms of the phonon GF. For a standard Hamiltonian of RE metallic compounds with taking into account the interaction of RE ions with acoustic phonons (magneto-elastic interaction) the one-phonon GF in the random phase type of approximation has the form [3]:

$$D(\vec{q},\omega) = [(D^{\circ}(\vec{q},\omega))^{-1} - \sum_{mn} G_{mn}(\vec{q},\omega)]^{-1} \qquad (1).$$

In Eq.(1) $D^{\circ}(\vec{q},\omega)$ is the phonon GF in the harmonic approximation GF $G_{mn}(\vec{q},\omega)$ in paramagnetic phase describes single-ion quadrupole excitations (quadrupole excitons):

$$G_{mn}(\vec{q},\omega) = \frac{E_{mn} V_{mn} V_{nm} (f_m - f_n)^2}{\omega^2 - E^2_{mn}} , \qquad (2)$$

where E_{mn} are energies of transitions between CEF levels, $f_m = \exp(-\beta E_m)/(\sum_m \exp(-\beta E_m))$ is the occupation number of single-ion states. $V_{mn} = \langle m|V(\vec{J}_i,\vec{q})|n\rangle$ are the matrix elements of operators $V(\vec{J}_i,\vec{q})$ which are certain functions of components of $\vec{J}_i$ operators depending on the symmetry of magneto-elastic coupling (for notation see /1/). Elements V_{mn} are calculated with the use of single-ion wave functions. As is seen from Eqs.(1), (2), the hybridization of quadrupole excitons and phonons arises provided matrix elements V_{mn} are not equal to zero.

3. VAN VLECK PARAMAGNET $PrNi_5$

Intermetallic RE compound $PrNi_5$ is well-known as a Van Vleck paramagnet which is used for the adiabatic magnetic coupling. The CEF of $PrNi_5$ has been investigated by means of inelastic neutron scattering on a polycrystalline sample /4/. $PrNi_5$ has a hexagonal structure of $CaCu_5$ type. The ground state multiplet $^3H_4$ of Pr^{3+} ions splits in CEF into three singlets Γ_1, Γ_3, Γ_4 and three doublets Γ_{5A}, Γ_{5B}, Γ_6. At low temperatures ($T \lesssim 10K$) only the lowest-lying level is occupied. We will consider the low-energy transition $\Gamma_4 - \Gamma_{5A}$ at $E = (4.3 \pm 0.3)$ meV which crosses the acoustic phonon. The wave functions of single-ion states are

$$\begin{aligned} &\Gamma_4 \text{ -level:} \quad 0707/3\rangle \; -0.707/-3\rangle , \\ &\Gamma_{5A} \text{ -level:} \quad 0.2198/\mp 4\rangle \; -00.9756/\pm 2\rangle . \end{aligned} \qquad (3)$$

The symmetry of these wave functions and the magneto-elastic interaction operator allows the hybridization of quadrupolar excitons and transverse acoustic phonons in all three symmetric directions of the reciprocal hexagonal lattice. Using expressions for magneto-elastic interactions in hexagonal lattices derived in paper /5/ we obtain the operators $V(\vec{J},\vec{q})$, which are nonzero for $\Gamma_4 - \Gamma_{5A}$ transitions, in the form

$$\Delta\text{-direction } [001]: \quad V(\vec{J},q_z) \sim [e_x(q_z) q_z (J^x J^z + J^z J^x) + e_y(q_z) q_z (J^y J^z + J^z J^y)]$$

$$\text{T-direction } [100]: \quad V(\vec{J},q_x) \sim e_z(q_x) q_x (J^x J^z + J^z J^x)$$

$$\Sigma\text{-direction: } [010]: \quad V(\vec{J},q_y) \sim e_z(q_y) q_y (J^y J^z + J^z J^y),$$

where $\vec{e}(\vec{q})$ is the polarization vector for the transverse acoustic phonon branch. Using the wave functions (3) we obtain matrix elements for Eq.(2) in the form

$$\begin{aligned} &V^{(1,2)}_{\Gamma_4 \Gamma_{5A}}(\Delta) = -19.64g , \\ &V^{(1,2)}_{\Gamma_4 \Gamma_{5A}}(T) = V^{(1,2)}_{\Gamma_4 \Gamma_{5A}}(\Sigma) = -9.82g , \end{aligned} \qquad (4)$$

where $V^{2\,(1,2)}_{\Gamma_4 \Gamma_{5A}} = V^{2\,(1,2)}_{\Gamma_{5A} \Gamma_4}$ ((1,2) denote the states of doublet Γ_{5A}) and the magneto-elastic coupling constant g was introduced.

Now using Eqs. (1) and (2) we can obtain expression for the energy of coupled quadrupole-phonon excitations:

$$\omega^2_{q\pm} = \frac{E^2_{\Gamma_4 \Gamma_{5A}} + \omega^2_q}{2} \pm \left[\frac{(E^2_{\Gamma_4 \Gamma_{5A}} - \omega^2_q)^2}{4} + 2\omega_q E_{\Gamma_4 \Gamma_{5A}} (|V^{(1)}_{\Gamma_4 \Gamma_{5A}}|^2 + |V^{(2)}_{\Gamma_4 \Gamma_{5A}}|^2) \right]^{1/2} , \qquad (5)$$

where $f_{\Gamma_4} = 1$, $f_{\Gamma_{5A}} = 0$ and ω_q is the phonon energy which can be approximated by the expression: $\omega_q \simeq Kq$. The value of K can be chosen from phonon experimental data. Thus in Eq.(5) together with Eq.(4) we have one fitting parameter g (the coupling constant for the interaction of RE ions with the lattice) which can be determined from experiments. The calculated dispersion relation for the excitations given by Eq.(5) in T direction is represented in Fig.1 for the parameter $g = 0.05$ meV. In this figure the magnetic dipolar transition, the width of which is ~0.7 meV /4/, is shown also by the dashed line with the vertical line which denotes the resolution in our experiments.

4. EXPERIMENTS AND DISCUSSION

The experiments were carried out at the reactor "Saphir" in the Institute für Reactortechnic in Würenlingen using a triple axis, constant-Q, spectrometry. The measurements were performed at temperature 8K on a single crystal sample $PrNi_5$ with volume ~0.4 cm^3 which was grown by the Bridjeman method. The mosaic structure of the crystal was nearly 10′. An incident monochromatic neutron beam with an energy of 13.7 meV focused in the monochromatic by a collection of a graphite single crystals was used. The halfwidth of the elastic peak $\Gamma(\omega = 0)$ was equal to 0.7 MeV.

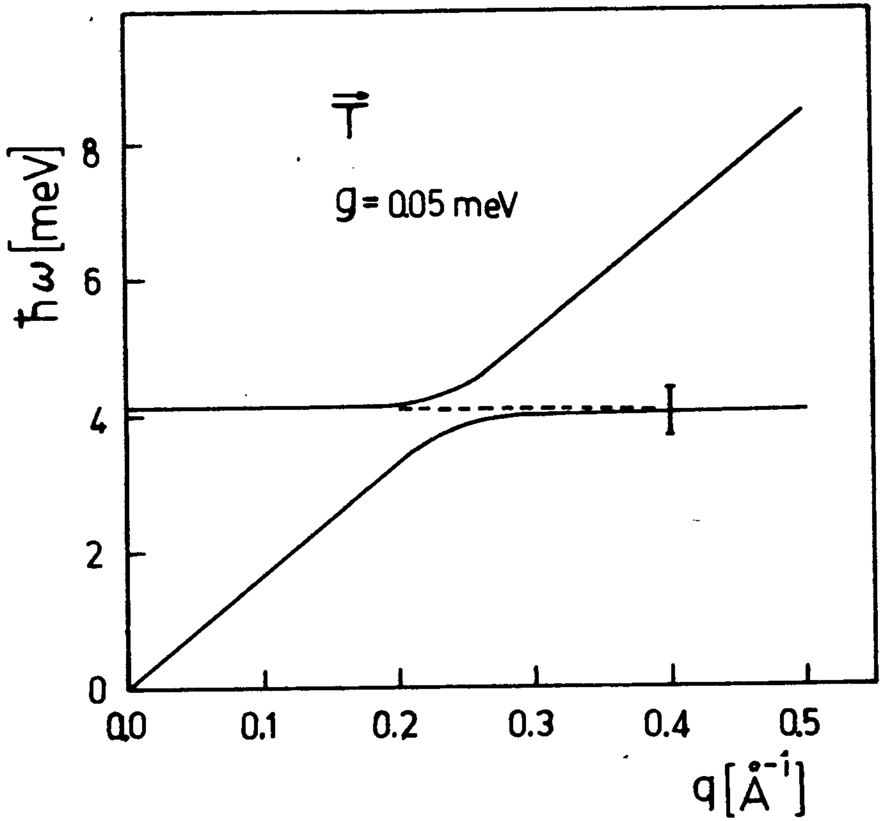

Fig.1. Calculated dispersion curves of the coupled quadrupole-phonon excitations in $PrNi_5$: [100] direction. The dashed line denotes the dipolar excitation.

A series of experiments was made in the region $0.15 \le q \le 0.5\,\mathring{A}^{-1}$ for $\vec{q}$ along the symmetric directions Λ and T. Two well-defined almost dispersionless magnetic dipole transitions were observed at 4.25 meV and 13.5 meV, respectively. The low excitation is degenerated with the quadrupole transition but only the latter mode is allowed to interact with the transversal acoustic phonon in region $0.2 \le q \le 0.35\,\mathring{A}^{-1}$. Figure 2 displays the measured intensity distribution at $\vec{Q} = (0.1, 0.1, 2.0)$. From our scans we are not able to obtain the additional peak which must appear in the anticrossing region. The main reason for this we see in the low energetic resolution of measurements.

Nevertheless, there are indirect indications of the hybridization of quadrupole and phonon excitations. In Fig.3 the q-dependence in T-direction is shown of the energy of the dipole

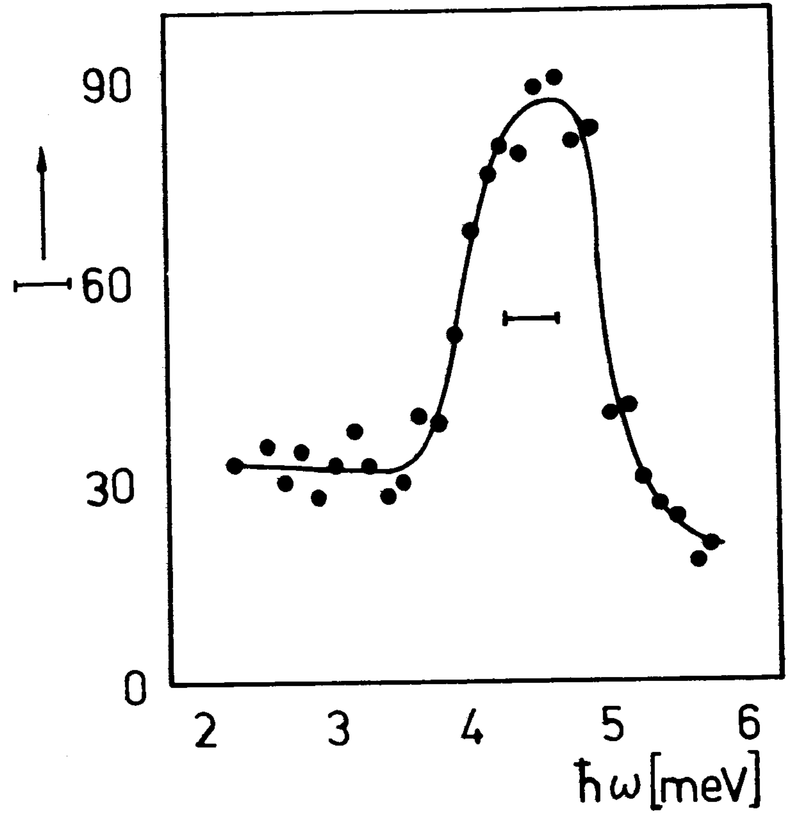

Fig.2. Energy spectra of a const. Q-scan in $PrNi_5$ at (0.1, 0.1, 2.0) R: resolution width.

magnetic (squares) and phonon (circles) excitations, which was obtained from the experimental spectra. For two points in the anticrossing region in Fig.3 we could not separate different contributions to the scattering. As one can see from Fig.3 the dipole exciton energy is higher before the crossing poin than after it. This can be interpreted as additional contributions to the cross section from the scattering on coupled quadrupole-phonon excitations which lead to the displacement of the center of mass of peaks to the region of high energy before and low energy after the crossing point.

Using Eq.(5) we can obtain the value of two-branch splitting (see Fig.1) $\Delta\omega$ at the crossing point. It has a linear dependence on the coupling constant g at small g. As we cannot resolve the splitting $\Delta\omega$ in our experiments, we can state only that $\Delta\omega < 0.07$ meV; and as a result, we obtain that $g < 0.07$ meV.

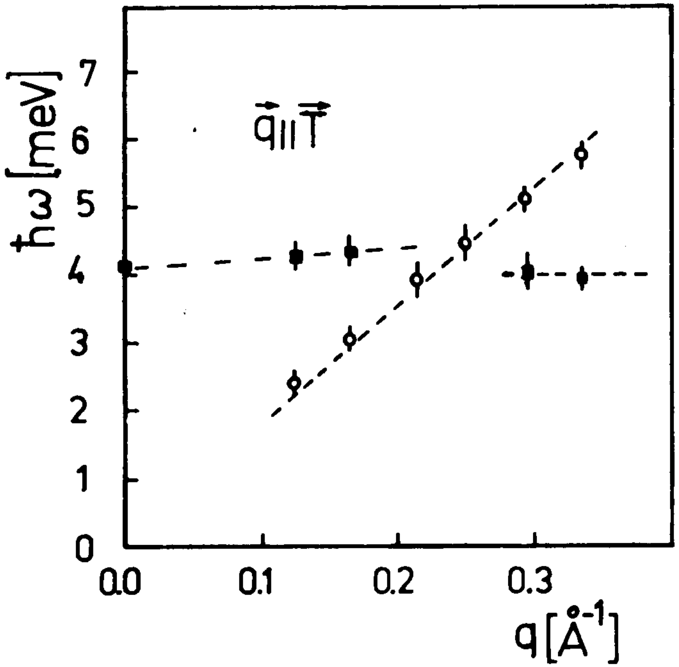

Fig.3. Excitations in $PrNi_5$ at 8K along [100] -direction.
□ - magnetic, O - transversal acoustic phonons.

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Аксенов В.Л. и др. E14-82-461

Связанные квадруполь-фононные возбуждения: неупругое рассеяние нейтронов на парамагнетике Ван Флека $PrNi_5$

Проведены эксперименты с помощью неупругого рассеяния нейтронов на монокристалле $PrNi_5$ /0,4 см^3/ при температуре образца 8К. Измерены дисперсионные кривые акустических поперечных фононов и низкоэнергетических магнитных возбуждений в Δ- и T-направлениях. Анализ области пересечения этих возбуждений показывает, что появляется дополнительная интенсивность в ядерном и магнитном дипольном рассеянии из-за магнито-упругого рассеяния. Обсуждаются гибридизированные спектры фононов и квадрупольных экситонов.

Работа выполнена в Лаборатории теоретической физики и Лаборатории нейтронной физики ОИЯИ.

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Coupled Quadrupole-Phonon Excitations: Inelastic Neutron Scattering on Van Vleck Paramagnet $PrNi_5$

Inelastic neutron scattering experiments on a single crystal $PrNi_5$ (0.4 cm^3) were performed at the sample temperature T ≈ 8K. The dispersion relations of the acoustic transverse phonons and the low-energy magnetic excitations were measured in the T and Δ-directions. An analysis of the intersection region of these excitations shows that an additional neutron intensity to the nuclear and magnetic dipolar scattering occurs due to magneto-elastic scattering. The hybridized spectra of phonons and quadrupole excitons are discussed.

The investigation has been performed at the Laboratory of Theoretical Physics and the Laboratory of Neutron Physics, JINR.

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