Neutron Scattering from Equilibrium and Non-equilibrium Phonons, Excitons and Polaritons

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Version of record first published: 14 Oct 2011.

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Neutron Scattering from Equilibrium and Non-equilibrium Phonons, Excitons and Polaritons†

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1. INTRODUCTION

To state the modern problems of physics of solids relevant to neutron spectroscopy, one has to consider new facilities available at the high flux reactors, such as the reactor at the Institute Laue-Langevin or PFR-2 (ИБР-2) at the Laboratory of Neutron Physics of the Joint Institute for Nuclear Researches at Dubna. Owing to this, the traditional treatment of the neutron spectroscopy as a new but essentially restricted technique, solving only a limited set of problems of physics of solids can be rejected. Presently, the facilities for neutron spectroscopy are vast enough to forget about the original role of the technique and to consider the neutron spectroscopy of solids from a broader viewpoint. This paper is an attempt at such a consideration.

Part I of the paper deals with traditional trends in the spectroscopy of inelastic neutron scattering (INS), that is with the investigation of the nuclear dynamics of the crystal at thermodynamical equilibrium. This trend can also be called the neutron spectroscopy of equilibrium phonons. The principal attention will be paid to the progress in this field that is due to high flux neutron sources. This part of the paper will be based on experimental results. The new problems requiring high resolution and high neutron flux intensity will be emphasized.

The next two parts will be concerned with the neutron spectroscopy of quasiequilibrium and non-equilibrium quasiparticles. Quasiequilibrium

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† The paper given at the 3rd International School in Neutron Physics, Alushta, April, 1978.

particles are meant to be the quasiparticles which are not in thermal equilibrium with the crystal, but within their allowed energy band, they manage, during their lifetime, to attain the Boltzmann distribution. No such quasi-equilibrium takes place for non-equilibrium particles. In the crystal quasi-equilibrium and non-equilibrium particles originate due to external sources (electromagnetic field, thermal pulses etc) that is, they correspond to a particular super-thermal excited state of the crystal. As will be shown below, even in the case of high intensity external sources, these quasiparticles are much fewer in number than those thermally activated. In this connection, for the neutron spectroscopy, the problem of their investigation is the problem of "low concentrations" and, therefore, the problem can only be solved under the conditions of increased experimental efficiency of modern neutron scattering investigations. The question of neutron scattering on excitons under the conditions of thermal equilibrium has also been considered. Presently, the experiments in the fields mentioned are missing. The neutron scattering in this field of problems of solids, and the analysis of its advantages over the conventional optical techniques are, however, of interest.

2. NEUTRON SPECTROSCOPY OF EQUILIBRIUM PHONONS

To analyse the state of investigations in this field, we shall confine ourselves to considering the neutron spectroscopy of phonons in molecular crystals. That we have chosen molecular crystals is not casual. This is because back in 1972 at an International Symposium on Inelastic Neutron Scattering (INS) at Grenoble the molecular crystals of complicated polyatomic cell structure were believed to be the subjects of which the INS investigations evidence to the progress in the development of these techniques. At that time as well as two years later during the Second School on Neutron Physics at Alushta, the investigations in this field were rare cases. Presently, this trend has, practically, been completely formed. A large number of crystals have been investigated: hexamethylentetramine and adamantane,[1,2] paradichlorobenzene[3,9,10] anthrachinone,[4] thiourea,[5] naphtalene,[6–10] benzene,[10–12] anthracene,[10] azoxybenzenes[10,13] and others. That the experiments are carried out at low temperatures is a very important element of the progress.

Figure 1 illustrates the spectra of inelastic coherent neutron scattering (ICNS) on a deuteronaphthalene crystal at 6 K obtained in the IN3 spectrometer at the Institute Laue-Langevin.[14] This figure is a good illustration of the ICNS spectroscopy potentialities at present. It suffices to say that the totality of spectra in different crystallographic orientations was obtained during 18 working days of the reactor. In determining phonon frequency values the measurements error did not exceed $\pm 0.5\,\mathrm{cm}^{-1}$ which is on the level of the most widely used optical techniques.
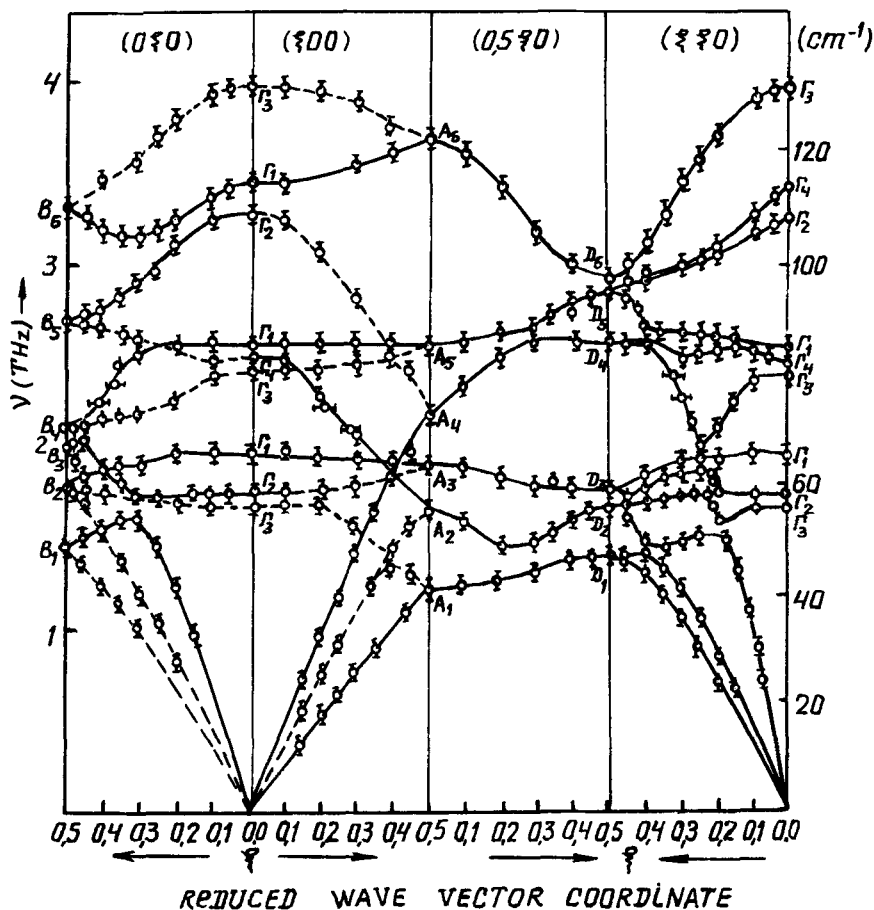
FIGURE 1 The dispersion curves of the external phonons of d_8-naphthalene crystal in the (a^*b^*) plane.[14] Solid and dashed lines are guides for the eye.

Figures 2 and 3 represent the spectra of inelastic incoherent neutron scattering (IINS) of benzene[10,11] and anthracene[10] crystals obtained at different temperatures in the inverse geometry spectrometer KDSOG-I at the PFR-I (ИБР-I) reactor at the Laboratory for Neutron Physics of JINR. The spectra represented embrace a wide range of transfered energies including both external and internal phonons (the structure of the molecular crystal phonon spectrum was discussed at the previous School).[15] In an average, it took 100 measuring hours to obtain each spectrum. It can easily be estimated that using the PFR-2 reactor will reduce this time down to ten minutes which will make the IINS spectroscopy an operative technique to
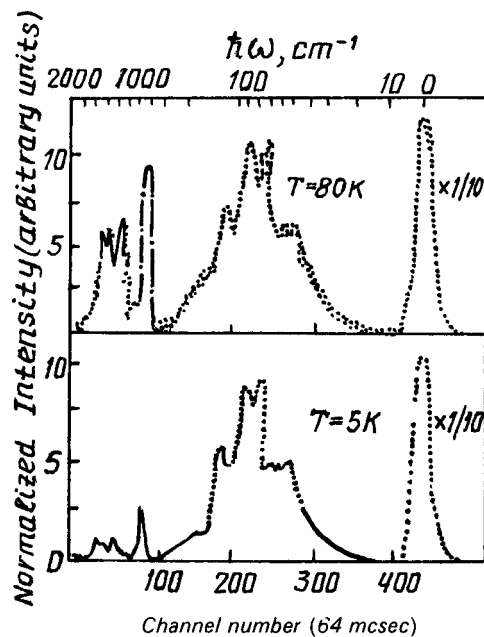
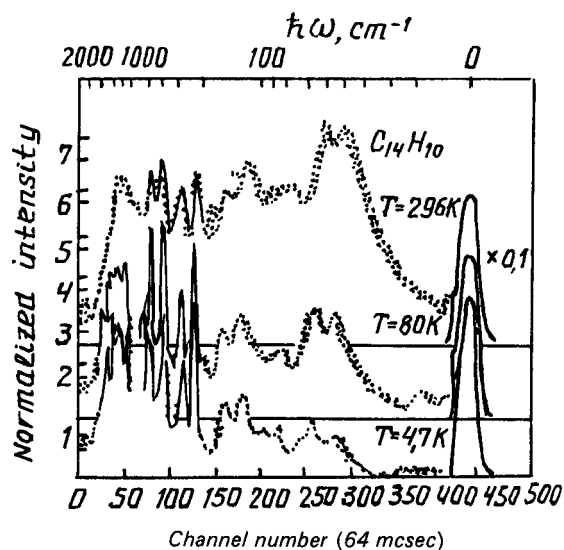FIGURE 2   The IINS experimental spectra of d_0-benzene crystal at different temperatures.[10,11]

FIGURE 3   The IINS experimental spectra of d_0-anthracene crystal at different temperatures.[10]
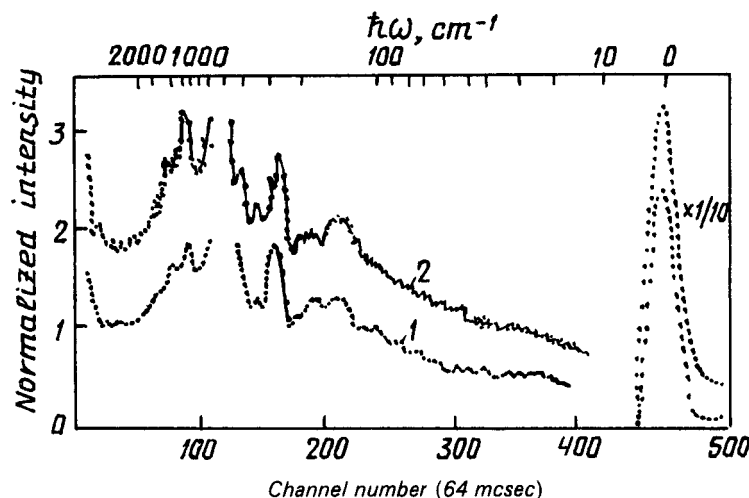
FIGURE 4 The IINS spectra of d_0-para-azoxyphenetole crystal at 80°K.[10]

solve a large number of problems within a short time as it is doing now the optical spectroscopy.

That this problem must be put forward can be demonstrated by the following example. Figure 4 represents the IINS spectra of para-azoxyphenetole obtained on two samples. Spectrum 1 corresponds to crystalline powder obtained by recipitation from benzene solution. Spectrum 2 is obtained on the same sample after it has been remelt. The difference between the IINS spectra and, hence, the difference between the phonon spectra of these crystals can readily be seen from the figure. This difference at the same temperature between the phonon spectra is a direct evidence to the existence of two crystalline phases. This has settled the bewilderment why it was that many authors obtained different structures in their crystallographic studies.[16]

A great number of works are concerned with a comparison between optical and neutron spectroscopy to study phonon spectra. In the region of external phonon modes the INS spectroscopy is undoubtedly advantageous. The ICNS and IINS spectra investigations, as is seen from Figures 1–4, permit one to obtain a complete information about the phonon spectrum whereas the optical spectroscopy is limited.

In the region of internal phonon modes which, because of their small dispersion, practically coincide with the molecular vibrations, the advantages of the INS are not so evident. Nevertheless, lack of the selection rules and a specific character of nuclear neutron scattering permit obtaining the data markedly supplementing the optical spectra. Thus, in analysing the internal phonon modes of a naphthalene crystal[17] optically inactive A_u-modes were
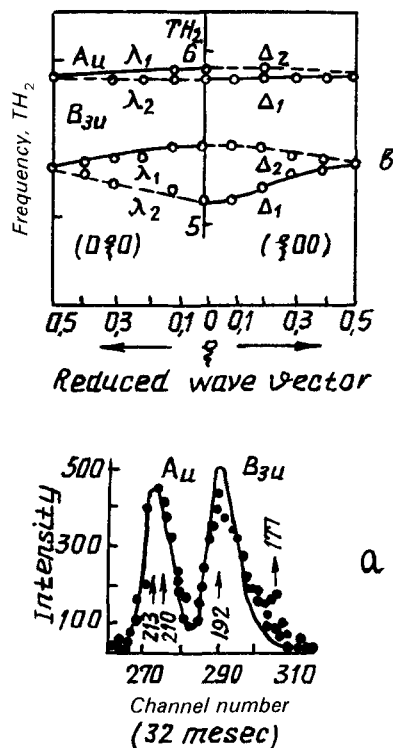
FIGURE 5   The IINS (a) and ICNS (b) spectra of d_0- and d_8-naphthalene crystals, respectively, in the region of the lowest internal modes at $T = 80°K$.[7,8] Numbers in (a) are given in cm^{-1}.

determined and the discrepancy was settled in interpreting of B_{2g}-modes. But the principal advantage of neutron spectroscopy is that it enables the dispersion of these modes to be studied. Figure 5 represents the results of the first investigations of internal phonon modes dispersion in a naphthalene crystal. First, using the IINS spectra of d_0-naphthalene (Figure 5a) the total dispersion of the two lowest phonon modes was found to be 3 cm^{-1} for A_u-mode and 18 cm^{-1} for B_{3u}-mode.[17]

Then, in the ICNS spectra of d_8-naphthalene (Figure 5b) these values were ascertained and the dispersion curves obtained.[8]

The inverse geometry techniques is quite good to investigate the INS spectra of the internal modes within a wide energy range. It is, however, essentially interior in resolution to the other techniques. The situation will noticeably be changed at an increase of the reactor power. Thus, Figure 6a shows the experimental IINS spectrum in the internal modes region of d_0-naphthalene crystal obtained in the PFR-1 reactor. Figure 6b and c
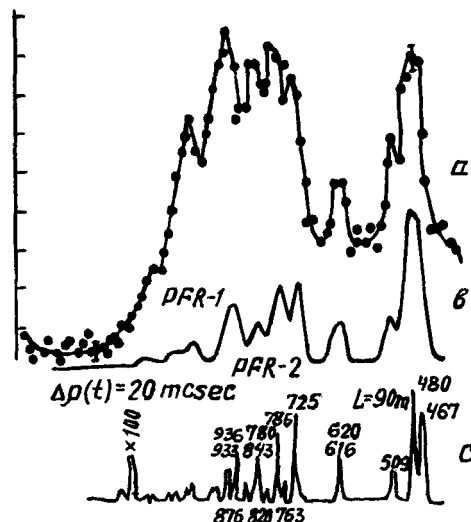
FIGURE 6(a)   The IINS experimental spectrum of d_0-naphthalene crystal in the region of the internal modes, $T = 80°$K, KDSOG-1, PFR-1, LNPh of JINR. 6(b) and 6(c). Calculated spektra of one-phonon IINS for the reactors PFR-1 and PFR-2, respectively.

represent the calculated one-phonon scattering spectra in this frequency region in PFR-1 and PFR-2 reactors, respectively.

A resolution improvement of the neutron experiment within a high frequency spectrum region suggests a possibility to investigate fine effects in the internal phonon spectrum. These comprise the fine structure investigation in the overtones region, connected with biphonon formation, as well as with two-particle state region,[18,19] and the spectrum investigation in the Fermi-resonance region.[19] Limitations imposed by symmetry selection rules impede very much biphonon and two particle states investigations by the optical techniques. The neutron scattering is free from these limitations, and the joint use of the ICNS and IINS permits obtaining a vast information on this spectrum region.

From the viewpoint of fine effects, search for neutron scattering on polarons, first considered in 1961[20] might be of interest too. The neutron-polaron interaction caused by electron-phonon coupling gives rise to different scattering types. The neutron scattering can occur in such a way that the phonon quantum number and electron state in a potential well remain unchanged in the process of scattering whereas only the polaron translation motion changes. If this scattering could be experimentally separated and its angular and energetic distributions investigated, this would enable determining such polaron characteristics as effective mass and electron density distribution in the polarization potential well.

Apart from this process, inelastic neutron scattering may occur followed by the quantum electron transition in the polaron potential well. Neutron energy undergoes great changes during these processes. Besides, the presence of polarons makes the phonon energy time finite and one-phonon ICNS peak diffuse.

All the above mentioned specific solid state effects can only be investigated by the INS when the objects have specially been selected, and under the high sensitivity and high resolution conditions of the neutron experiment.

3. NEUTRON SPECTROSCOPY OF QUASIEQUILIBRIUM QUASIPARTICLES

a. Scattering on polaritons

Typical representatives of this type of particles are polaritons. These are superpositions of photons and mechanical excitations of the crystal, that is, excitons or phonons.

First, let us consider a polariton whose mechanical part is phonon. Figure 7 shows a typical example of the polariton spectrum of an isotropic crystal. A great number of works available are concerned with the investigations of polariton dispersion, principally, using the Raman scattering technique.[21] However, these investigators have only succeeded in measuring $\omega(q)$ for the lower TO branch. The dual nature of the polariton enables its investigation by the neutron scattering technique, the information obtained, as will be shown later, being much greater than that obtained by optical techniques.

The neutron scattering on polaritons was first considered theoretically in 1974.[22] Later, many investigators have directed their attention to this problem.[23-26]

When scattering on polaritons, the neutrons interact with their mechanical part, therefore, the scattering efficiency is proportional to the phonon strength functions. This function determines the portion of the mechanical energy in polariton and equals the ratio of the atom displacement squared amplitudes in polariton and optical phonon. So, under the thermal equilibrium, after one polariton has been generated, the ICNS cross section has the form[26]

$$
\left(\frac{d^2\sigma}{d\Omega\,d\omega}\right)^c = \frac{N}{2\pi\hbar}\frac{K'}{K}\sum_{q,j}|F_j(Q)Q|^2\frac{n(\omega)+1}{2\omega_j}S_j(\omega)\delta(Q-q-2\pi\tau)
$$

$$
\times\{[\delta(\omega-\omega_-)-\delta(\omega+\omega_-)]
$$

$$
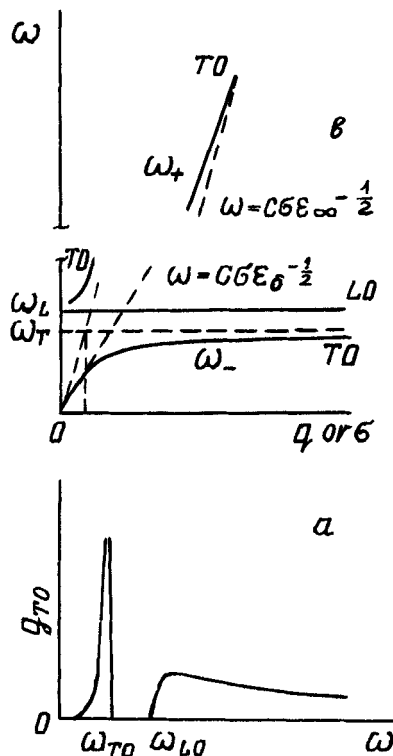+[\delta(\omega-\omega_+)-\delta(\omega+\omega_+)]f_n\},
$$

FIGURE 7(a) The density of the polariton states $g_{TO}(\omega)$. (b) The polariton spectrum in an isotropic crysta, ω_+ and ω_- are branches of TO polaritons; LO is the branch of longitudinal optical vibration; ε_∞ and ε_0 are high frequency and static values of the dielectric permeability; q and σ are the wave vectors of polariton and photon, respectively.

where

$$F_j(Q) = \sum_i M_i^{-1/2} b_i^c e^{-iQ\rho_i} e_j^i(q). \qquad (1)$$

Here, index j numbers the crystal phonon modes, i labels atoms in a primitive unit cell, f_n is the oscillator strength of the dipole excitation of the j-th mode, S_j is the phonon strength function, and ω_+ and ω_- are two branches of the polariton spectrum, shown in Figure 7b. The remaining designations are conventional in the theory of neutron scattering on phonons.

The phonon strength function $S_j(\omega)$ is

$$S_j(\omega) = \begin{cases} \dfrac{\omega \omega_p^2 \omega_j}{(\omega_j^2 - \omega^2)^2 + \omega_p^2 \omega_j^2} & \text{for dipole-active phonon modes} \\ 1 & \text{in the remaining cases.} \end{cases} \qquad (2)$$

Here, ω_p is the plasma frequency.

Under the same conditions, the IINS cross section for the cubic crystal has the form[26]

$$\left(\frac{d^2\sigma}{d\Omega\,d\omega}\right)^{inc} = \frac{N}{2\pi\hbar}\frac{K'}{K}Q^2[n(\omega)+1]\sum_i\frac{(b_i^{inc})^2}{M_i}\left(\frac{g_{TO}(\omega)}{\omega_i}+\frac{g_{ph}(\omega)}{\omega}\right). \quad (3)$$

Here $g_{ph}(\omega)$ is the density of crystal phonon states, and $g_{TO}(\omega)$ is the density of polariton states, shown in Figure 7a.

From the above equations it is seen that under the thermal equilibrium, the neutron scattering processes with polariton generation are slightly effective due to the fact that the value of $S_j(\omega)$ is small at $\omega \neq \omega_j$ and, as a result, the value of $g_{TO}(\omega)$ is small too. This disadvantage can, however, be compensated by employing high intensity optical (laser) pumping to generate quasiequilibrium polaritons of 10^{15} to 10^{17} $1/cm^3$ in number, which markedly exceeds thermally equilibrium $n(\omega)$. The scattering cross sections (1) and (3) will be determined by the values n_p representing the over an energy spectrum quasiequilibrium polariton distribution. Provided low temperatures are added the INS spectra get really measurable in the anti-Stokes region with the absorption of $n_p(\omega)$ polaritons. The selection of the crystal is also important to perform the experiment successfully. Two points are essential here. 1. The strength function $S_j(\omega)$ is proportional to the longitudinal-transverse splitting $\omega_{LO}^2 - \omega_{TO}^2$ of optically active phonon. 2. The values b_i^c and b_i^{inc} must be great enough. From these viewpoints perspective are the crystals MgF_2,[22] LiH[23], MgO[24] etc.

b.   Neutron scattering on excitons

In the case of neutron scattering on excitons two different cases must be considered. In the first case, an exciton is caused by the neutron transmitted energy in the process of scattering. The possibility of exciton generation, like in the above mentioned case of electron excitation in the polaron well[20] or impurity center[27] is due to the electron-phonon interaction.[23,27] The estimations show that in this case the complete scattering cross sections make up 10^{-2} to 10^{-3} of the one-phonon cross section. Therefore, to perform the experiment practically, one must have high flux hot neutron sources with the energy of one to several tens eV.

In the second case, neutron scattering occurs on the generated excitons. This process is analogous to that considered above for polaritons in quasi-equilibrium conditions, that is to the scattering on exciton polaritons, or, as they are called, light-excitons.[28] In molecular crystals the exciton energy band width $E(q)$ being tens to hundreds mev, high fluxes of thermal neutrons are needed to realize this scattering experimentally. The quantitative difference of neutron scattering on light-excitons from that on phonon polaritons

is the different value of the function $S(\omega)$. At neutron-exciton nuclear inter-action, this function is determined by the exciton-phonon interaction para-meters.

The neutron magnetic scattering on non-zero spin excitons may occur too. In this respect, it is of interest to consider neutron scattering on triplet excitons (TE) that are a coherent wave of molecular electronic excitation with a spin equal to unity. By the request of the authors of this paper, Dr. V. I. Sugakov has considered the common regularities of neutron scattering on TE.†

The consideration is based on the analogy of neutron scattering on magnetic excitons and magnons. However, an important difference between the case considered and scattering on magnons is the fact that exciton exists before scattering process, and scattering only leads to transitions from one exciton state to another. The consequences of this event can easily be followed in analysing Eq. (4) obtained by Dr. V. I. Sugakov for the differential cross section of neutron scattering on TE.

$$\frac{d^2\sigma}{d\Omega\,d\omega} = \frac{4}{3}\frac{K'}{K}(g_n r_0)^2 \sum_{\mu q} P_{\mu q}\left|F_{\mu q}^{\mu' q'}(Q)\right|^2$$

$$\times \sum_{\tau} \delta_{Q,\,q-q,\,+2\pi\tau}\delta(\hbar\omega - \varepsilon_{\mu q} + \varepsilon_{\mu' q'}). \qquad (4)$$

Here $g_n = 1,91$ is the neutron g-factor, $r_0 = e^2/mc^2$ is the classical radius of the electron, $P_{\mu q}$ is the wave vector distribution of TE in the μ-th exciton band, $F_{\mu q}^{\mu' q'}$ is the magnetic form-factor of TE at a change of their state from (μq) to $(\mu' q')$.

The specific nature of neutron scattering on TE reveals itself already in elastic processes at $\omega = 0$. As seen from Eq. (4) these processes are contributed by items with $\varepsilon_{\mu q} = \varepsilon_{\mu' q'}$. In the course of this, neutron and exciton vectors are changed without the energy exchange occurring between them. Thus, the TE presence gives rise to the broadening of elastic scattering peaks, and this broadening increases with temperature. In principle, this broadening might yield an information on the density of state in the TE bands.

Inelastic scattering on TE will also markedly differ from that on magnons. The excitons available in the zone will contribute scattering. As this takes place, both the processes occurring in one band $\mu = \mu'$, and those occurring in different bands $\mu \neq \mu'$ will be observed. In the general case, the F form-factor structure is complicated. A qualitative analysis of the INS on TE requires the numerical calculation of this factor, for which, in turn, the TE band model must be assigned.

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† The authors are greatly thankful to Dr. V. I. Sugakov for giving the calculation results at their disposal.

The cross section of both elastic and inelastic neutron scattering on TE is N_{ex}/N times smaller than the magnetic scattering in a ferromagnetic crystal (N_{ex} is the TE number, N is the number of crystal cells). Therefore, to experimentally observe the considered scattering even under the conditions of low temperatures and anti-Stokes processes, it is obligatory that a marked value of N_{ex} should be obtained.

c. Light-exciton generation

The lifetime of excitons in respect to radiation and leaving of a crystal is 10^{-9} to 10^{-8} sec for singlet, and in the range from 10^{-7} to seconds for triplet excitons. Alternatively, the lifetime of these excitations in respect to their relaxation, that is, to the transformation of high energy bands excitons to lower bands excitons is, usually, not longer than 10^{-10} sec. Therefore, it can actually be said that excitons accumulate only in the lowest exciton bands of the crystals (singlet or triplet).

An analysis of the experimental data obtained with various types of exciton excitation (one- and two-photon absorption at different absorption coefficients) shows that, at present, it is possible to obtain the exciton concentrations N_{ex} of 10^{14}–10^{13} 1/cm^3 under the stationary pumping conditions, and N_{ex} of 10^{18} 1/cm^3 with the pulse excitations. The external limitation from above is the necessity for providing the heat emission to prevent the crystal destruction. The internal limitations are, first of all, various non-linear effects of exciton-exciton interactions and those between excitons and the photon field arising from their recombination. These are: non-linear exciton concentration quenching, laser generation and superluminescence, induced Raman scattering, exciton plasma formation and, finally, occurrence of exciton or electron-hole condensates of various kinds. Naturally, the neutron scattering investigation, say, on exciton drops is no less interesting and promising than the proper scattering on free excitons. But, evidently, it is difficult to interpret the results with no data available on a more trivial effect.

At present, much attention is paid to the development of the resonance excitation methods for crystal exciton states. Owing to tunable lasers, it turns out possible to investigate in detail the narrow excitation region close to the exciton band bottom and to trace the change in the situation when the light is running through the crystal in the region of true transparency, and light-excitons are being formed in the direct vicinity of the band. It is to be hoped that these investigations will lead to obtaining of the controlled and sufficiently high concentrations of light-excitons in crystals.

From the viewpoint of the experimental realizations of scattering, neutron pulse sources together with optical pumping pulse techniques yielding higher

values of N_{ex} are undoubtedly advantageous. It is also evident that the most favourable observation conditions will be in the case when the light-exciton lifetime in the crystal is of the same order of magnitude as the neutron flux pulse duration.

4. NEUTRON SPECTROSCOPY OF NON-EQUILIBRIUM PHONONS

First of all, consider the formation of non-equilibrium phonon packets and fluxes. Here, three types of non-equilibrium phonon excitation might be expected to take place:

a) Injecting of thermal phonons by different means (electric heating, laser or any other light source irradiation etc), the emitter being placed next to the surface of the sample investigated.

b) IR light resonance absorption followed by optical phonon (polariton) formation close to the centre of the Brillouin zone (discussed in Section 3a).

c) Electron absorption in the crystal surface layer and the phonon formation due to the relaxation process down to the lowest electron (exciton) states, due to the luminescence exciton transitions and, due to the radiationless processes in the exciton system (for instance, Auger-processes followed non-linear quenching).

The phonons are generated most effectively in case (c). It is by the very case when great (macroscopic) phonon populations can be obtained; owing to this, the neutron experiment can be mostly successful.

Figure 8 represents the diagram experimentally confirmed for the case.[29] The principal processes of electron excitation relaxation occur in the layer $(\sim K^{-1}$ cm) wherein the excitation light is absorbed. As a result, the Brillouin zone is populated by phonons more or less uniformly. Neither the primary, nor the secondary excitons as well as optical phonons arising at the subsequent stages are able to leave this layer $(d \sim 0.5\ \mu)$, as their diffusional lengths are much shorter. Therefore, this layer can be departed from only by the acoustic phonons which have sufficiently long paths. So, this layer can be treated as an effective emitter of these phonons. Then, these phonons move into the crystal depth getting, spontaneously, approximately, halved, then halved again and again. At this stage, each phonon generation moves in a ballistic regime at the velocity of sound (strictly speaking, the appropriate several phonon groups corresponding to different acoustic branches). At a distance from the emitter corresponding to section A, there may occur either such population numbers at which the inverse merging processes will be
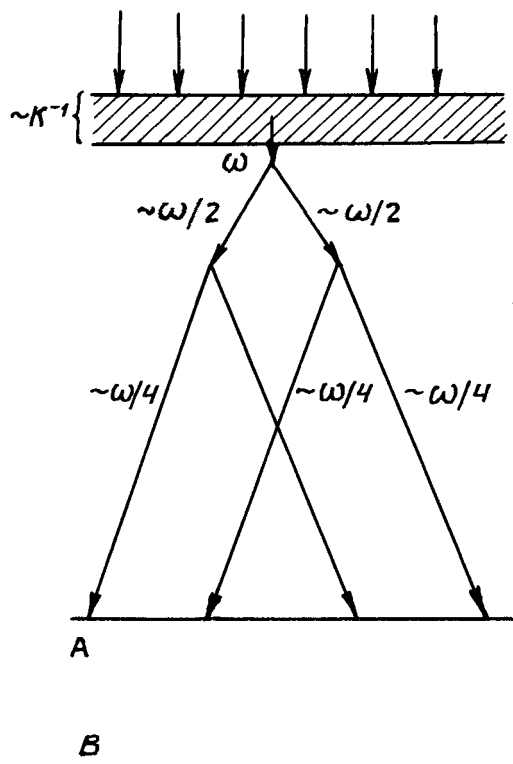
FIGURE 8   A diagramme of the generation and propagation of non-equilibrium phonons.

possible, and the hydrodynamical regime ensues (the motion of Planck-distributed phonon packets) or due to um-klapp processes, the diffusional regime ensues. There may be an intermediate case, too, when, first, hydrodynamics is formed, then, at significantly a greater depth it is transformed to diffusion. Depending on the crystal thickness, its temperature and phonon generation rate one of these types can be realized in the main crystal volume in the appropriate time intervals starting from the pulse excitation initiation. Then, in these crystals ($10-50\,\mu$) one can obtain mean concentrations of a half- or a quarter-Debye phonons of the order $10^{20}-10^{21}$ cm^{-3}.

Figure 9 shows a peculiar time pattern on phonon appearance on the surface B opposite to that of the emitter localization in thin anthracene crystals at 4.2 K. The phonon detectors were:

1) halfwidth of fluorescence bands induced by a special testing pulse,

2) relative band intensities in the given spectrum,

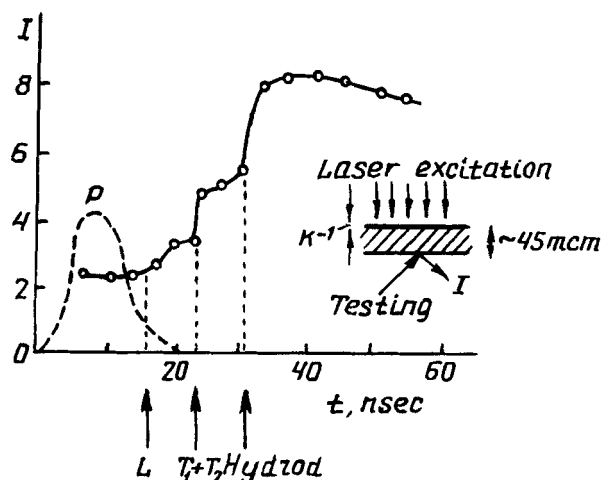3) the intensity of induced resonance Raman scattering in the crystal.

FIGURE 9   The dependence of relative intensity of 25036 and 25051 cm^{-1} bands on the delay time t at the rear surface of an anthracene crystal.[29] The crystal thickness is 45 μ, the pumping is 40 kwt/cm^3, the bath temperature is T_b = 5 K. P is the pumping pulse shape in relative units; L, T_1 + T_2 and Hidrod, are times of the occurrence of different phonon groups at the rear surface.

In the latter case, the sensitivity was maximal and responded to the scattering intensity change by a factor of 10 as the temperature increased by 0.6–0.7°. It is seen from the figure that up t = 15 ns from the pumping pulse start no change is observed in the spectrum, then, one can identify three non-equilibrium phonon groups having appeared by 15, 23 and 31 ns from the pumping pulse start at the rear of the crystal surface.

Neutron scattering fixation on these phonons occurs under the condition that their lifetimes in the crystal must agree with the neutron flux pulse duration $\tau \approx 10^{-4}$ sec. This can be achieved in comparatively thick crystals. The point is that as has been shown, at a spontaneous phonon division, their free path and the lifetime are increased at the ration $(\omega_0/\omega_f)^5$ where ω_0 and ω_f are the initial and final phonon frequencies. Even if the initial phonon time with the frequency ω_0 is 10^{-9} sec, then for the phonons with ω_f = 0.1 ω_0, this time is 10^{-4} sec. The neutron scattering, in this case, does not only allow phonons with ω_f to be fixed, but also, the precedent and subsequent fission processes and the ballistic, hydrodynamics and diffusional processes of intertransformation to be investigated.

A somewhat different technique to detect non-equilibrium phonons was used in the thermal pulse experiments.[30] Figure 10 represents the fluorescence spectrum of an organic molecule (perilene) in Spolski matrix (n-octane) under a stationary excitation (SF), and additional fluorescence (dashed line
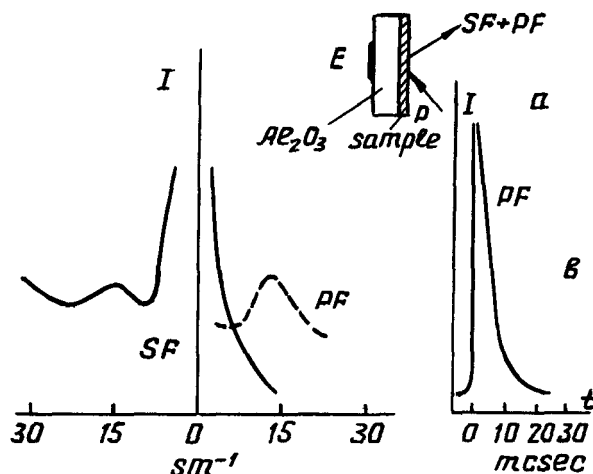
FIGURE 10   The stationary fluorescence spectrum of perilene close to the 4415.6 Å exciting line (solid line). and the fluorescence pulse spectrum in the anti-Stokes region. On the right are the experimental scheme (a) and a typical fluorescence pulse (b).

PF) occurring when the phonons from the emitter through the sapphire arrive at the Spolski layer. This additional fluorescence is in time correlation with the current pulses through the metallic emitter. The pulse fluorescence is seen to answer the anti-Stokes process which is confirmed by the phonon concentration rise in the sample at this instant of time. This experiment is an analogue to the anti-Stokes neutron scattering. It evidences that non-equilibrium phonons can be detected, which is promising in the case of neutron scattering too.

5.   CONCLUSION

We have considered a number of problems of the solid state physics which, to our belief, are of interest for the neutron spectroscopy of the days to come.

Their realization will open a new page in the development of the neutron spectroscopy of solids. One new tendency is to be noted here. Until recently, the development of the neutron spectroscopy lagged behind that of the optical spectroscopy, the delay in time being, at first, about several decades (equilibrium phonon spectroscopy). Presently, this "lag behind" is decreasing (the non-equilibrium phonon spectroscopy is the problem of today for the optics). After the above discussed problems have been realized, it will turn out quite possible that the neutron spectroscopy will develop simultaneously with the optical or, even, be ahead of it in solving the burning problems of the solid state physics.

Acknowledgment

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