

COHERENT EFFECTS IN RESONANT DIFFRACTION: THEORY

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The main results of the kinematical theory and of the dynamical theory for resonant diffraction are summarised, with special emphasis given to the coherent enhancement of the radiative channel and to the suppression of the incoherent channels.

1. INTRODUCTION

In nuclear resonance scattering the nucleus is excited by an incoming γ -quantum and decays afterwards. In the simplest case either a γ -quantum is reemitted, which is coherent with the incoming quantum, or internal conversion takes place. For low energy resonance transitions, the probability $W_{\gamma 0}$ for coherent radiative decay is usually much smaller than the probability for incoherent decay.

The question arises, whether this weak radiative decay channel of the individual nucleus can be intensified by resonant diffraction of γ -radiation, where a regularly arranged ensemble of nuclear scattering centers is excited in phase /1-9/. An enhancement of the coherent channel could be caused by constructive interference in the Bragg direction. It can also be expected, that the radiative coupling within the regular system of nuclear resonators has strong effects on the scattering parameters of the system.

These two aspects lead to the treatment of the resonant diffraction in terms of the dynamical theory. The discussion of this theory and its results will have a central place in this paper. At first, however, we will have a short look at the enhancement of the coherent channel in kinematical diffraction.

2. ENHANCEMENT IN KINEMATICAL DIFFRACTION

In the kinematical theory of diffraction, each scattering center is acted upon only by the primary wave. Multiple scattering effects are neglected.

If an individual nucleus is excited by an incoming γ -quantum, the reemitted radiation is directed into the full solid angle. However, if n nuclei in a crystal are excited by a γ -quantum incident in the Bragg direction, the coherently reemitted radiation is collimated into the forward direction and into the direction of the Bragg reflection. In these directions, the peak intensity is enhanced proportionally to n . The total probability $W_{\gamma \text{kin}}$ for the coherent re-emission of the radiation is obtained by integrating the diffracted

intensity in k-space. In the case of radiation with a broad energy distribution like with X-rays and neutrons, this integration is over the full size of the diffraction region centered at the reciprocal lattice point, which is proportional to $1/n$. As a result, there is no enhancement of the total diffracted intensity. For Mössbauer radiation, however, which is nearly monochromatic, the integration is only over a thin disc cut out of this diffraction region by an Ewald sphere with well defined radius k . Since in this way the energy distribution of the radiation is not sharpened by the diffraction, the total coherent reemission probability is enhanced as given by /1,6/

$$W_{\gamma\text{kin}} = W_{\gamma 0} 2N\pi/(ka)^2 \quad (1)$$

Here N is the number of lattice planes in the direction of the outgoing quantum, k is the length of the wave vector and a the interplanar distance. The coherent decay probability increases with N . The incoherent decay probability, by contrast, remains unchanged.

This enhancement of the coherent channel with respect to the incoherent channels is caused by the constructive interference in the radiative decay of the collective nuclear excitation of the crystal. It corresponds to the Dicke's superradiance of a symmetric state /8,19/. This state is here a single delocalised excitation, which must be specially selected by the proper in-phase excitation of the system of resonant scattering centers by a primary beam incident in the Bragg direction.

The radiative decay channel can be observed in the reflected beam. As follows from eq. (1), for sufficiently large N the total intensity of a beam reflected by the nuclei of a crystal will be larger than the intensity scattered by the same number of individual nuclei into the full solid angle. But the enhanced coherent reemission probability results also in a speed-up of the radiative decay. Thus the enhancement of the coherent decay channel in kinematical diffraction means both, an intensity increase and a speed-up in the diffracted beam.

With increasing intensity of the reflected beam, however, multiple scattering cannot be neglected any longer. Therefore the validity of eq. (1) is restricted to very small crystals, where also the enhancement effect remains small. The appropriate treatment for larger crystals has to take into account multiple scattering processes, and this is done by the dynamical theory.

3. THE DYNAMICAL THEORY: GENERAL CONCEPTION

The dynamical theory was developed by Darwin, Ewald and von Laue in the beginning of this century /10-14/. The impact of multiple scattering becomes most evident in a model introduced by Ewald /10/: Oscillators, which are placed at the lattice sites, are acted upon by all possible radiation waves in the crystal and give rise, in turn, to scattered waves. The waves are coupled by the oscillators, and the oscillators are coupled by the waves. The main idea now was to look for self-consistent solutions of this coupled system in an unbounded medium: The oscillators have to sustain just those waves by which they are driven. The eigenfunctions of this resonating system have to be such, that the incoming and the outgoing waves are matched by means of boundary conditions.

The fundamental set of equations of the dynamical diffraction theory is given by

$$2 \epsilon_j E_j = \sum_i g_{ji} E_i \quad (2)$$

where E_i is the electric field amplitude of the wave in direction i and g_{ji} are the scattering parameters from direction i into j , which are equal to $\lambda^2 \pi/V$ times the coherent scattering amplitude of the unit cell with volume V . ϵ_j is the deviation of the refractive index from unity for the wave in direction j . For simplicity, only one polarisation component is considered. Eq. (2) can be interpreted in the following way: All possible waves in the crystal, with directions i and amplitudes E_i , generate by coherent scattering (described by g_{ji}) wavelets into the direction j . The sum of all these contributions determines the refractive index of the wave in direction j . The refractive index resp. the phase velocity is used as the regulating device in order to obtain a self-consistent solution, the eigenvalues of which are ϵ_j . The simplest case of eq. (2) is well known: When there is no Bragg reflection, the refractive index n is determined by the coherent forward diffraction according to $n=1+\epsilon=1+g_{00}/2$.

In the following the discussion will be limited to the most common case, where only one Bragg reflection is activated. Then the incoming wave with wavevector \vec{k}_0 excites only two waves in the crystal, a forward wave with wavevector \vec{k}_0 and a reflected wave with wavevector $\vec{k}_1 = \vec{k}_0 + \vec{G}$, where \vec{G} is the reciprocal lattice vector. In this case the eigenfunctions are superpositions of these two coherent waves, which create each other continuously by diffraction: $[\vec{k}_0 \leftrightarrow \vec{k}_1]$. The coherent superposition of these two waves is a new physical entity, which is called wavefield or coupled pair state.

Since the wavefield eigenfunctions are formed by the coherent superposition of a wave and its reflected wave, they will always have a periodic standing wavefield structure in the direction perpendicular to the reflection planes. This structure essentially determines the refraction and absorption of the radiation in the crystal. For instance, if the scattering is isotropic, the eigenfunctions at the exact Bragg position are standing wavefields with nodes in or in between the lattice planes. Accordingly, the absorption of these wavefields will be anomalously low or anomalously high.

Up to now the general conception of the dynamical diffraction was introduced, which applies for all types of radiation and scattering processes, as long as the Bragg condition can be fulfilled. In the following the special impact of the nuclear resonant scattering process on the dynamical diffraction will be discussed.

4. NUCLEAR RESONANT DYNAMICAL DIFFRACTION

Nuclear resonance scattering is a second order, indirect scattering process, which proceeds via an intermediate state. The existence of this intermediate state has important consequences, which make up the appeal of resonant dynamical diffraction /15-25, 26 (review)/. First of all, the intermediate state can decay via coherent and incoherent channels, thereby giving the possibility to study explicitly the ratio of these two decay modes. Secondly the intermediate state has a finite lifetime, which leads to distinctive time and energy dependences of the scattering process. These characteristic properties of the individual nucleus will be drastically

changed in collective resonance scattering by an ensemble of nuclei arranged in a crystal lattice.

The coherent nuclear resonance scattering parameters for an isolated hyperfine transition (E1 or M1) are given by /23,27/

$$g_{ji}(\Delta E) = - \frac{\lambda^2}{\pi V} \frac{3}{2k} \frac{1}{\Delta E + i\alpha_c} \frac{1}{\alpha_c + 1} C^2 \eta \frac{1}{2I_g + 1} f_j f_i P_j P_i \quad (3)$$

ΔE is the distance from resonance in units of half the natural linewidth. α_c is the internal conversion coefficient, C the Clebsch-Gordon coefficient, η the enrichment and I_g the spin of the groundstate. The intranuclear incoherence due to conversion and spinflip is described by the factors $1/(\alpha_c + 1)$ and C^2 , the internuclear isotopic and spin incoherence by the factors η and $1/(2I_g + 1)$. $f_j f_i$ is the temperature factor and $P_j P_i$ the polarisation factor. Because of the factorisation of these last two terms and because of the absence of a formfactor, the complete nuclear scattering parameter can be written

$$g_{ji}(\Delta E) = g_j g(\Delta E) g_i \quad (4)$$

where g_i describes the excitation, g_j the deexcitation and $g(\Delta E)$ the energy dependence. There is no term in $g_{ji}(\Delta E)$, which directly connects the directions i and j . This complete factorisation is always possible in the case of an isolated nuclear hyperfine transition /21-23/. Eq. (4) also holds for special cases, when several hyperfine transitions coincide, e.g. for antiferromagnetic reflections /23/ and in the case of an unsplit M1-transition for the π -polarisation component of the radiation /15/. In these cases the fundamental set of dynamical equations (eq. 2) can be written

$$2 \epsilon_j E_j = g_j g(\Delta E) \left(\sum_i g_i E_i \right) \quad (5)$$

This set of equations shows, that there exist eigenfunctions with eigenvalues $\epsilon_j = 0$. They must be built up in such a way, that the total amplitude A_{exc} for the excitation of the individual nuclei in the crystal lattice vanishes completely

$$A_{exc} \sim \sum_i g_i E_i = 0 \quad (6)$$

These solutions exist at the exact Bragg position, if eq. (4) is fulfilled. Eigenvalues $\epsilon_j = 0$ mean, that the corresponding wavefield travels through the crystal without any refraction and absorption. Since the excitation of the individual nuclei is completely cancelled in this case, the incoherent decay channels are suppressed as well. This is the effect of suppression of the incoherent channels predicted by Kagan and Afanasev /15/. A sufficient condition for the suppression effect is, that the nuclear excitation amplitude vanishes. Thus the wavefield itself does not have to vanish at the nuclear sites. It is sufficient, if the wavefield is constructed in such a way, that it is exactly perpendicular to the nuclear hyperfine transition oscillators. These are circular or linear oscillators for the $\Delta m = -1$ or $\Delta m = 0$ transitions, respectively. Examples for the resulting wavefield structures are given in /23/.

At first glance it seems contradictory, that diffraction can be sustained without excitation of the nuclei. It should be noted, however, that the eigenfunctions of the dynamical theory are steady state solutions, where no transient effects are taken into account. At the exact Bragg position, the collective nuclear resonator system has just eigenfunctions which do not excite the nuclei. For a further understanding it is helpful to abandon the exact Bragg position for a moment and to have a look at the behaviour of diffraction and absorption, when the exact Bragg position is approached /28/. Near the Bragg position the total excitation amplitude A_{exc} does not vanish completely and residual resonance absorption takes place. Approaching the Bragg position, the excitation amplitude decreases, the residual absorption breaks down and the radiation penetrates deep into the crystal. The excitation amplitude must vanish to an extent, that the total diffraction, summed up coherently over the increasing crystal volume, remains constant. This is just the self-consistency condition. The excitation of the individual nuclei vanishes, but the coherent excitation of the nuclear collective remains. The incoherent channels are suppressed, because the excitation amplitude for the individual nuclei vanishes, whereas the diffraction by the collective of nuclei is fully maintained due to coherent excitation and coherent decay. Thus dynamical diffraction in spite of a vanishing excitation amplitude can be considered also as a result of the coherent enhancement of the radiative channel.

The vanishing of the excitation amplitude is independent of the energy distance from resonance. At the exact Bragg position, therefore, the diffracted intensity does not depend on frequency. As a consequence, the diffraction is instantaneous. The promptitude of the diffraction leads to an alternative interpretation of the suppression effect /19,41/. In the time picture, the incoherent channels are suppressed, because the coherent excitation of the collective of nuclei decays immediately via the coherent channels. The interpretation of the suppression effect by means of a standing wavefield with vanishing excitation amplitude, however, is the more general one, because it is closely related to the standing wavefield picture widely used in the dynamical diffraction theory for X-rays and neutrons. The spatial picture seems also to be more helpful for an understanding of the interaction of the radiation with the nuclear scattering centers in the crystal.

The temperature dependence of the suppression effect is of fundamental interest. It should be expected, that lattice vibrations always spoil the effect, because they tend to shift the nuclei into the space between the lattice planes, where the nuclear excitation amplitude does not vanish. The dynamical theory, however, predicts, that at the exact Bragg position the suppression effect is independent of temperature /15/. The excited nuclear level is so extremely sharp in energy, that the nuclear excitation by a Mössbauer γ -quantum can only proceed elastically, i.e. without simultaneous phonon creation or annihilation. If the nuclear resonance scattering process is to be elastic altogether, also the deexcitation must proceed without recoil. For this reason the temperature factor for coherent resonance scattering $f_j f_i$ is a product of an excitation and a deexcitation factor (Lamb-Mössbauer factor for scattering). As discussed above, this factorisation leads to eigenvalues $\epsilon_j=0$ at the exact Bragg position, in spite of the lattice vibrations. A handwaving explanation for this surprising result relies on the fact, that the lifetime of the intermediate state, which is of the order of 10^{-7} s, is much longer than the characteristic times of the lattice vibrations, which

are about 10^{-13} s. Because of this comparatively long lifetime of the intermediate state, the wavefield sees the nuclei only in their time averaged positions, i.e. exactly in the lattice planes, where the excitation amplitude vanishes /15/.

The set of dynamical equations (5) has four eigenvalues ϵ_0 in the case of one Bragg reflection. These eigenvalues depend on the angular deviation parameter α and on the energy distance ΔE from resonance. The full dependence on α and ΔE can be obtained analytically, if the set of dynamical equations splits into two subsets with separate eigenpolarisations. Then the general expression for the eigenvalues ϵ_0 for one eigenpolarisation component is given in the case of a crystal plate by /15/

$$4\epsilon_0^{(1,2)}(\alpha, \Delta E) = (g_{00} + \beta g_{11} - \beta \alpha) \pm \sqrt{((g_{00} + \beta g_{11} - \beta \alpha)^2 + 4\beta(\alpha g_{00} - D))} \quad (7)$$

with $\alpha = -\sin 2\theta \Delta\theta$, $\beta = \cos\theta_0 / \cos\theta_1$, θ_i angle between the surface normal and the direction i , and $D = g_{00}g_{11} - g_{10}g_{01}$. The energy dependence of the scattering parameters $g_{ji}(\Delta E)$ has been omitted in eq. (7). The eigenvalues ϵ_1 follow from $\vec{k}_1 = \vec{k}_0 + \vec{G}$. It should be noted, that the eigenvalue $\epsilon_0 = 0$ corresponding to the full suppression effect is obtained at the exact Bragg position $\alpha = 0$ only, if the determinant D vanishes. This condition, however, is always fulfilled in those cases, where the scattering parameters can be completely factorised according to eq. (4).

With this knowledge about the wavefield properties inside the crystal we are now prepared to discuss the resulting intensities of the outgoing diffracted beams.

5. DIFFRACTED BEAMS: LAUE GEOMETRY

In Laue geometry, the incoming beam and the outgoing diffracted beams are on opposite sides of a crystal plate. Diffraction in Laue geometry is especially sensitive to anomalous absorption, because the radiation becomes transmitted through the entire crystal.

Laue diffraction geometry causes the parameter β in eq. (7) to become positive. In this case the two eigenvalues $\epsilon_0^{(1,2)}$ given by eq. (7) correspond to anomalously low and anomalously high resonance absorption, respectively. The incoming beam excites in the crystal two wavefields belonging to these two eigenvalues, which are given by

$$WF^{(1)} + WF^{(2)} = (E_0^{(1)} \vec{e}_0 + E_1^{(1)} \vec{e}_1) e^{ikt' \epsilon_0^{(1)}} + (E_0^{(2)} \vec{e}_0 + E_1^{(2)} \vec{e}_1) e^{ikt' \epsilon_0^{(2)}} \quad (8),$$

where $E_i^{(1,2)} \vec{e}_i$ is the electric field vector of the wave in direction i as given in /15/ and t' is the effective crystal thickness $t / \cos\theta_0$. For instance in the case of isotropic scattering and at the exact Bragg position $\alpha = 0$, the first wavefield has nodes in the lattice planes yielding anomalously low absorption, whereas the second wavefield has the nodes in between the lattice planes yielding anomalously high absorption. Both wavefields travel along the reflection planes towards the rear side of the crystal, where they split into two diffracted beams: the Laue-reflected and the Laue-transmitted beam. In general, each one of these beams is made up by the coherent contributions of both wavefields.

If the incoming beam is at resonance, the second wavefield is extinguished on the way through a thick crystal because of anomalously high resonance absorption. In this case the outgoing beams originate only from the first wavefield which experiences the full suppression of the incoherent channels at the exact Bragg position. Thus the intensities of the reflected and of the transmitted beams are not reduced by any resonance absorption. As can be seen from eq. (8), these intensities are then determined only by the electric field amplitudes $E_0^{(1)}$ and $E_1^{(1)}$, which follow from the scattering parameters g_{ji} according to the condition, that the total excitation amplitude A_{exc} given by eq. (6) vanishes. In the case of isotropic scattering, the intensities of the reflected and the transmitted beams are equal. They can, however, strongly differ in anisotropic scattering conditions /29/. At angular positions slightly off the exact Bragg position, the suppression effect breaks down, and the intensities of the diffracted beams fall off sharply because of residual resonance absorption. The angular dependences of the Laue-reflected and the Laue-transmitted beams are Gaussians with halfwidths inversely proportional to the square root of the crystal thickness /15,22/.

With increasing energy distance from resonance, however, the resonance absorption of the individual nucleus decreases strongly according to $1/(\Delta E)^2$. For radiation several linewidths off resonance, even a thick crystal becomes transparent and the second wavefield corresponding to anomalously high resonance absorption described by the eigenvalue $\epsilon_0^{(2)}$ in eq. (8) begins to contribute to the diffracted intensities. The Laue-reflected and the Laue-transmitted beams are now composed of the coherent contributions of the two wavefields. These give rise to pronounced interference effects, the so-called Pendellösungen. The Pendellösungen depend on several parameters like crystal plate thickness, angular deviation from the Bragg position and energy distance from resonance /30/. In practice, thickness inhomogeneities of the crystal and the finite divergence of the primary beam always tend to average the Pendellösung interferences to a certain extent. In this region of intermediate and weak resonance absorption, the angular dependence of the Laue-reflected beam is still nearly Gaussian. The Laue-transmitted beam shows characteristic dark-light and light-dark intensity profiles at energy positions below and above resonance, respectively /31/.

Very far off resonance the resonance absorption can be neglected. Both wavefields given by eq. (8) reach the rear side of a thick crystal plate nearly without attenuation. At the Pendellösung maxima almost all radiation quanta incident at the Bragg position are directed into the Laue-reflected beam. This result strongly contrasts the scattering by an individual nucleus, where most of the scattered radiation is lost into the incoherent channels. This holds also far off resonance, because for the individual nucleus the ratio of the coherent and the incoherent decay rates is independent of the distance off resonance. Far off resonance the excitation amplitude for the individual nuclei vanishes because of the large distance from resonance. As a consequence, the total absorption vanishes independently of any wavefield structure. But the vanishing excitation amplitude nevertheless leads to a coherent excitation of the nuclear collective, and strong diffraction becomes possible /31/. This situation far off resonance must be distinguished from the case at resonance, where the strong resonance absorption is suppressed due to a vanishing excitation amplitude connected with a very special wavefield structure.

6. DIFFRACTED BEAMS: BRAGG GEOMETRY

In Bragg geometry, the incoming beam and the reflected beam are on the same side of the crystal. Due to this diffraction geometry, the parameter β is always negative. As a consequence, both eigenvalues $\epsilon_0^{(1,2)}$ given by eq. (7) correspond to anomalously low absorption at the Bragg position.

When the condition $\text{Im}(\epsilon_0^{(2)} - \epsilon_0^{(1)})kt' \gg 1$ is fulfilled in the case of a thick crystal, the incoming beam excites only one wavefield in the crystal, which corresponds to the eigenvalue $\epsilon_0^{(2)}$. This eigenvalue $\epsilon_0^{(2)}$ can become zero at the Bragg position, e.g. in the case of isotropic scattering conditions. The wavefield is then constructed again in such a way, that the nuclear excitation amplitude vanishes in the reflection planes giving rise to the full suppression effect. In contrast to the Laue geometry, there is only one diffracted beam in the thick crystal case. The angular dependence of the intensity of this Bragg-reflected beam shows in resonance an extremely sharp peak exactly at the Bragg position /17,32/, where even a reflectivity of 100% is reached in the case of isotropic scattering. This total reflection in resonant Bragg diffraction was predicted by Trammell /1/. The reflectivity becomes total in spite of the fact, that the wavefield penetrates infinitely deep into the crystal /28/. Total reflection at resonance is therefore also a manifestation of the suppression effect /17/.

At the exact Bragg position, the reflection is total, independent of the energy distance from resonance. In the region around the Bragg position, the reflection peaks are strongly broadened in energy. The broadening is inversely proportional to the angular deviation from the Bragg position. This dependence can be understood by considering the increase of the number of properly phased scattering centers with decreasing distance from the exact Bragg position. The closer to the Bragg position the radiation falls in, the more lattice planes are in phase, and the smaller the scattering amplitude or the larger the distance off resonance may be without causing a loss of reflectivity.

Refraction plays also an important role in the Bragg geometry diffraction. It causes angular shifts of the reflection curves. These shifts go into opposite directions at energy positions above and below resonance, because they are proportional to the real part of the coherent forward scattering parameters $g_{00}(\Delta E)$ given in eq. (3). The resulting distortion of the reflection intensity profile $I_{BR}(\alpha, \Delta E)$ can be recognised in a contour plot /32,41/.

Especially interesting is the thickness dependence of the reflected intensity. In Bragg geometry the number of reflecting planes can be changed by increasing the crystal thickness. The general expression for the reflectivity of a crystal of arbitrary thickness /17,19/ can be expanded near the exact Bragg position and yields in the case of a symmetrical reflection ($\beta=-1$) and for isotropic scattering conditions /1,19/

$$I_{BR}(\alpha \approx 0, \Delta E) \approx \left| \frac{\Gamma_{\gamma dyn}}{\Delta E + i(1 + \Gamma_{\gamma dyn})} \right|^2 \quad (9)$$

where $\Gamma_{\gamma dyn} = kt'g_{00}(0)/2$. This expression shows, that the reflected

intensity I_{BR} is broadened in energy proportionally to the crystal thickness. For very thin crystals this broadening is the same as given by the ratio $W_{\gamma_{kin}}/W_{\gamma_0}$ given in eq. (1), which was obtained from kinematical considerations. Hence eq. (9) confirms the kinematical result and shows also its limits: In order to observe the line-broadening connected with the coherent enhancement of the radiative channel, $\Gamma_{\gamma_{dyn}}$ should at least equal unity. At resonance, this yields a reflectivity of 25%, which certainly requires to take also multiple scattering into account and to use the dynamical theory.

Eq. (9) also holds for very thick crystals, as long as the deviation from the exact Bragg position is kept sufficiently small. According to eq. (9), the reflectivity reaches 100% at resonance for sufficiently thick crystals. This fact can be considered as the result of the suppression effect. Parallel to the increase of the reflectivity also the energy width $\Gamma_{\gamma_{dyn}}$ of the resonance peak becomes larger. For thickness values, where the reflectivity at resonance already goes to saturation, the energy width still increases proportionally to the number of reflection planes. This energy broadening can be interpreted as the result of the enhancement of the coherent decay probability in dynamical diffraction. Again one can realise, that the suppression of the incoherent channels occurs simultaneously with the coherent enhancement of the radiative channel.

It should be noted, however, that the discussion given above is too much idealised in order to describe the phenomena. With pure nuclear reflections, the electronic absorption is usually not suppressed and limits the penetration of the wavefield into the crystal. For this reason the peak reflectivity at resonance and the energy broadening are smaller in reality than mentioned above. Furthermore there exist no pure nuclear reflections without hyperfine splitting, which leads to nuclear interference. For nearly pure nuclear reflections, where the electronic scattering amplitude of the unit cell vanishes by chance, the interference is altogether constructive /32/. For systematically pure nuclear reflections, however, this interference is altogether destructive, because a pure nuclear reflection must vanish when all hyperfine transitions coincide. For this reason, there is no reflection intensity above and below the region of hyperfine splitting, which therefore limits the effective linebroadening /32/. Conversely, the destructive interference between close neighbouring hyperfine transitions could be used to obtain small reflection linewidths in spite of the strong energy broadening effects in resonant Bragg reflection.

A limiting case of a Bragg geometry reflection is the total external reflection in grazing incidence, which can be completely described as the diffraction arising in a certain angular region around a forward Bragg reflection /19/. Hence the basic features of resonant grazing incidence reflection can be extrapolated from the properties of resonant Bragg reflection. For a detailed discussion, however, the field is beyond the scope of this paper, and only recent progress with anti-reflection multilayer films is mentioned /33-35/.

7. TIME DEPENDENCE OF RESONANT DIFFRACTION

From the energy dependence of the resonant diffraction discussed so far, also the time dependence can be obtained by Fourier transformation. These aspects of resonant dynamical diffraction /36/ became of interest recently, when the possibility emerged to realise time resolution experiments also with the intense primary beams needed for

resonant diffraction. This time resolution can be achieved either by a fast shutter system, which instantaneously closes and opens the primary beam of a strong conventional Mössbauer source /37/, or by using the flashlike puls of a synchrotron radiation source /38/. Temporal aspects of resonant diffraction are of crucial importance for the filtering of Mössbauer radiation from the synchrotron radiation by means of a pure nuclear Bragg reflection /39-41/.

In principle, the time response of a single crystal to the synchrotron radiation puls can be directly derived from the energy response. At the exact Bragg position, the diffracted intensity is independent of the frequency, hence the diffraction is instantaneous. At small angular deviations from the Bragg position, the reflection peaks are strongly broadened, hence the diffraction is considerably speeded up. The energy broadening and the speed-up decrease with increasing distance from the exact Bragg position. The energy distribution of the diffracted beam is also determined by the energy-time correlation. The beam diffracted instantaneously is white. But after a short delay, the radiation with a strongly broadened frequency distribution has already passed, and a beam of Mössbauer quanta can be extracted from the synchrotron radiation /40/.

The speed-up in resonant dynamical diffraction occurs parallel to a total or a very high reflectivity. Due to the suppression effect, there are no absorption losses. This case must be distinguished from all other types of apparently accelerated decay or energy broadening, which can always be obtained by resonance absorption. If for instance the γ -quanta of a source are observed after they have passed through a resonant absorber, the decay of the source appears also to be accelerated, because the radiation quanta at resonance, which are preferably emitted at delayed times, are filtered out of the beam /42/.

The coherent speed-up also demonstrates a tight correlation between the spatial and the temporal properties of nuclear resonance scattering /39/. An individual nucleus scatters radiation with a characteristic decay time into the full solid angle. The ensemble of nuclei in a crystal, by contrast, diffracts the radiation much faster into the Bragg direction. With increasing number of reflection planes, the Bragg direction becomes more defined, the energy width is broadened and the diffraction is speeded up. For an infinitely thick crystal, finally, the speed-up is the stronger, the closer to the exact Bragg direction the primary beam is incident.

In detail, however, the time dependence of nuclear resonant diffraction is rather complicated. There are different laws for the speeded up decay of the collective excited nuclear state depending on the angular deviation from the Bragg position and on the time elapsed after the initial excitation /40,41/. In addition, there are special interference effects like dynamical beats /40,41/ and quantum beats /43/, which arise because the white synchrotron radiation excites a broadened hyperfine transition or several hyperfine transitions simultaneously.

8. SUMMARY

With resonant diffraction of Mössbauer radiation, the coherent radiative channel is enhanced with respect to the incoherent channels. The reflected wave in a single crystal can become so intense, that multiple scattering leads to dynamical diffraction. In the dynamical

theory for nuclear resonant diffraction, the general concept of dynamical diffraction is applied for the special case of a second order, indirect scattering process. The existence of an intermediate state in this scattering process facilitates strong incoherent decay, which gives the possibility to study explicitly the ratio of the coherent and the incoherent scattering channels. In the diffraction at resonance, a standing wavefield is built up with vanishing amplitude for the excitation of the individual nuclei, thus leading to a complete suppression of the incoherent channels, whereas the coherent excitation of the collective of nuclei and hence also the diffraction are fully maintained. The suppression effect results at the exact Bragg position in a total transmission or in a total reflection in the Laue or in the Bragg geometry, respectively. The existence of the intermediate excited state also leads to pronounced energy and time dependences of the scattering process. In resonant diffraction, an angular dependent energy broadening arises in the region of the Bragg reflection, which is directly correlated with an angular dependent speed-up in the time dependence.

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