

Resonance vibration modes of point defects and the Mössbauer effect. II

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THE PAPER constitutes the second part of a previous article dealing with the influence of the resonance modes of point defects on the Mössbauer effect. Part one concerns low concentrations at which the defect may be treated as isolated, part two deals with the influence of interaction of defects at higher concentrations. The interactions lead to resonance splitting and introduce the modulation effect which increases the recoilless emission coefficient.

Praca stanowi drugą część artykułu poświęconego wpływowi rezonansowych modów defektów punktowych na efekt Mössbauera. Pierwsza część dotyczy niskich koncentracji, gdy defekt może być traktowany jako izolowany, w drugiej natomiast rozpatruje się wpływ oddziaływań defektów przy wyższych koncentracjach. Oddziaływania te powodują rozszczepienie rezonansów i prowadzą do pojawienia się efektu modulacyjnego, zwiększającego współczynnik emisji bezodrzutowej.

Работа составляет вторую часть статьи посвященной влиянию резонансных модов точечных дефектов на эффект Мессбауэра. Первая часть касается низких концентраций, когда дефект можно трактовать как изолированный, во второй же части рассматривается влияние взаимодействия дефектов при более высоких концентрациях. Эти взаимодействия вызывают расщепление резонансов и приводят к появлению модуляционного эффекта увеличивающего коэффициент эмиссии без отдачи.

Introduction

IN THE FIRST part of this paper [1], we discussed the possibilities of investigating the dynamics of a certain class of point defects by means of the Mössbauer effect. We considered an isolated defect — i.e., the case when the concentration of the impurities is so low that we may disregard the interaction between its atoms. The other extreme case is the disordered lattice in which the number of the atoms of the impurity is close to the number of the atoms of the matrix. Since in this type of lattice the resonance lines are smeared out and usually divided into a number of wide maxima (cf. [2, 3]), it is impossible to obtain the relevant information on the dynamics of an isolated defect. On the other hand, for measurement reasons — namely in view of low intensities of the measured radiation — the case of very small concentrations is more difficult from the experimental point of view, although the information possible to be deduced is “pure” and theoretically well discernible in view of the weakness of the assumptions present in its description (cf. [1]). Thus, we shall investigate finite concentrations, sufficiently low as compared with those leading to a disordered crystal.

1. Finite concentrations of defects

In the case of finite concentrations, we cannot disregard the interactions of defects. It is known that they lead to splitting of the resonance lines, but the only detailed investigations concerned merely the case of two interacting defects [3, 4, 5] with the additional restriction to isotopic defects in a simple cubic lattice with harmonic interactions between nearest neighbours. Nevertheless, qualitative results of this model can be applied to a more realistic situation [3].

First of all, it is important that the splitting due to distant defects may be disregarded — i.e., it is sufficient to investigate configurations of comparatively close defects. In order to describe this situation more precisely, it is necessary to proceed to model configurations.

For the time being, we examine generally an arbitrary monatomic matrix and we do not assume anything concerning interatomic interactions. We are interested in configurations of impurity atoms in terms of the concentration. To this end, we choose an arbitrarily situated impurity atom (i.e., an arbitrary lattice site containing the impurity) and examine possible locations of other impurity atoms with respect to the one chosen. Assuming that the impurities distribution is spatially homogeneous, we ascertain the probability $P(n_r)$ of finding n impurity atoms in r -distant lattice sites (for nearest neighbours $r = 1$, for second neighbours $r = 2$, etc.), in accordance with Bernoulli's method:

$$(1.1) \quad p(n_r) = \binom{N_r}{n_r} c^{n_r} (1-c)^{N_r - n_r},$$

where c is the number of the impurity atoms per one matrix site (the concentration) and N_r denotes the number of r -distant sites (r -neighbours). The probability of appearance of a definite configuration $K(N_r, n_r)$ (n_r impurity atoms in r -neighbourhoods of the chosen atom) has therefore the general form:

$$(1.2) \quad P\{K(N_r, n_r)\} = \prod_r P(n_r).$$

We can now use the qualitative results known for a pair of interacting defects [3, 5] — namely the fact that splittings for defects separated by 3–4 lattice parameters are small as compared with the width of the split line. In other words, we are interested only in the situation for the first 10–20 r 's up to a certain maximum $r = R$. The probability of the relevant configuration can be written in the form:

$$(1.3) \quad P\{K(N_r, n_r; R)\} = \prod_r P(n_r) \left\{ \sum_{n_r} \prod_{R+1}^{\infty} P(n_r) \right\} = \prod_r P(n_r),$$

where the expression in the curl brackets in the central part of the equation is unity. If now $c \ll 1$, then the formula (1.1) implies immediately that $P(n_r) \propto cP[(n+1)_r]$. To within terms of the order $O(c)$, besides the physically trivial terms of the type $P(0_r)$, we can retain only terms of the type $P(1_r)$ and, moreover, since R is not greater than, say, 20, the principal contribution is due to the configurations with one non-vanishing n_r — i.e., due to pairs of the impurity atoms. The ratio of the probabilities of appearance of a con-

figuration with two terms of the type $P(1_r)$ to a configuration with only one term of this type (for small c) is as follows:

$$(1.4) \quad \frac{\left(\sum_1^R N_r\right) c^2 (1-c)^{\sum_1^R N_r - 2}}{\left(\sum_1^R N_r\right) c (1-c)^{\sum_1^R N_r - 1}} \approx \frac{1}{2} c \left(\sum_1^R N_r - 1\right) \approx \frac{1}{2} c \sum_1^R N_r.$$

We now present Tables of the first fifteen N_r 's for cubic lattices. d_r denotes here the distance between the r -th neighbour and a chosen impurity atom with coordinates $(0, 0, 0)$, in unities of the lattice parameter. The number N_r is the number of different permutations obtained from the triad of numbers in the column "type of coordinate", taking into account negative coordinates.

Table 1

r	simple cubic			b.c.c.			f.c.c.		
	d_r^2	type of coordinate	N_r	d_r^2	type of coordinate	N_r	d_r^2	type of coordinate	N_r
1	1	100	6	$3/4$	$1/2 \ 1/2 \ 1/2$	8	$1/2$	$1/2 \ 1/2 \ 0$	12
2	2	110	12	1	100	6	1	100	6
3	3	111	8	2	110	12	$3/2$	$1 \ 1/2 \ 1/2$	24
4	4	200	6	$11/4$	$3/2 \ 1/2 \ 1/2$	24	2	110	12
5	5	210	24	3	111	8	$5/2$	$3/2 \ 1/2 \ 0$	24
6	6	211	24	4	200	6	3	111	8
7	8	220	12	$19/4$	331	24	$7/2$	$3/2 \ 1 \ 1/2$	48
8	9	300, 221	30	5	210	24	4	200	6
9	10	310	24	6	211	24	$9/2$	$3/2 \ 3/2 \ 0,$ $2 \ 1/2 \ 1/2$	36
10	11	311	24	$27/4$	$3/2 \ 3/2 \ 3/2,$ $5/2 \ 1/2 \ 1/2$	32	5	210	24
11	12	222	8	8	220	12	$11/2$	$3/2 \ 3/2 \ 1$	24
12	13	320	24	$33/4$	$22 \ 1/2, \ 5/2 \ 11$	24	6	211	24
13	14	321	48	$35/4$	$5/2 \ 3/2 \ 1/2$	24	$13/2$	$5/2 \ 1/2 \ 0,$ $2 \ 3/2 \ 1/2$	24
14	16	400	6	9	300, 221	18	$15/2$	$5/2 \ 1 \ 1/2$	24
15	17	410, 322	48	10	310	24	8	220	8
		$\sum_1^R N_r =$	274		$\sum_1^R N_r =$	280		$\sum_1^R N_r =$	306

The formula (1.4) and the values of N_r from Table 1 make it possible to determine the maximum admissible configuration for which with a required accuracy we may disregard configurations with three interacting impurities. For instance, for $c = 10^{-3}$, the contribution from interactions of triads of defects is, roughly speaking, by one order smaller than the contribution due to pair interactions.

As already indicated, it is necessary to proceed to model considerations in order to investigate more exactly the splitting of the resonance lines. Thus, there arises the problem of the type of the Green function to be applied in our calculations — namely, we have

to decide whether to use the Green function for the perfect crystal (including the perturbation operator describing the pair of defects considered) or rather to investigate the pair of defects in a more realistic manner — i.e., in the crystal described by the mean Green function, taking into account deviations from the perfect lattice due to the presence of defects in the distant background.

The mean Green function for a crystal with random distribution of defects can be expanded into the perturbation series [3]:

$$(1.5) \quad \bar{R} = G + G\bar{J}G + G\bar{J}G\bar{J}G + \dots,$$

where the line above the letter denotes the averaging over the distribution of defects, G is the Green function for the perfect crystal, J the perturbation operator describing the difference between the perfect lattice and the lattice containing defects. Since \bar{J} is proportional to the concentration [3], for small c we may replace \bar{R} by G to within terms of the order $O(c)$. Thus, if we confine ourselves to interactions of pairs of defects — i.e., accuracy of the order 10^2c — then certainly we can use in calculations the Green function for the perfect lattice.

Finally, the situation is such that a part of the impurity atoms can still be regarded as isolated defects, while for the remaining part we must take into account the interactions, confining ourselves for the time being to pairs of defects. Since for an isolated defect the coefficient of the recoiles emission is given by the formula [1]

$$(1.6) \quad f(t) = f_{st}^{1+\Phi} e^{-\lambda t},$$

where f_{st} is the stationary coefficient of the recoiles emission, Φ is a parameter describing the magnitude of the vibrational excitation, λ is the width of the vibrational resonance and t is the time counted from the moment of excitation, in our case the coefficient $f(t)$ has the general form:

$$(1.7) \quad f(t) = \left[1 - \sum_r^R P(1,r) \right] f_{st}^{1+\Phi} e^{-\lambda t} + \sum_r^R P(1,r) f_r,$$

where f_r is the coefficient of the recoiles emission of the subensemble of the atoms interacting with r -neighbours.

With this general background, we are in a position to proceed to the problem of determination of the splitting and the amplitudes of the split lines.

The frequency of the split lines is determined from the condition of vanishing of the determinant of the reduced matrix [3],

$$(1.8) \quad \det \{GJ - I\} = 0,$$

where I is the unit matrix.

The resonance amplitudes can be determined directly from the system of equations for the displacement of the defect (we are interested only in the ratios of the resonance amplitudes) [3], namely:

$$(1.9) \quad u = (I - GJ)^{-1} u_{\text{pert. lat.}}$$

It is now necessary to choose a reasonable model of matrix for our case, and the model of the defect, which will represent the impurity atom considered and will provide the required values of G and J . The simplest is the model of the simple cubic lattice with harmonic interactions between nearest neighbours, for which there exist tables of the Green functions [6]. For a defect in this matrix, there are two possibilities — namely, the isotopic defect and a defect with altered coupling parameters. The second choice leads to considerable difficulties of a computational nature and yields qualitative results which do not differ from those of the isotopic defect [3]. Consequently, we shall examine the isotopic atom model of the defect.

2. Splitting of the resonance lines. Modulation effect

In view of our previous considerations, we consider a pair of isotopic defects in a simple cubic matrix. The perturbation operator for defects situated at the sites $\mathbf{0}$ and \mathbf{h} has the form [3]:

$$(2.1) \quad J_{ij}^{mn} = \varepsilon M \omega^2 \delta_{ij} \{ \delta^{om} \delta^{on} + \delta^{nm} \delta^{nn} \},$$

where $\varepsilon = (M' - M)/M$ and M and M' are the masses of the matrix atom and the impurity atom, respectively. The Eq. (1.8) can be written in the form:

$$(2.2) \quad \{ \varepsilon M \omega^2 (G_{ii}^{oo} + G_{ii}^{on}) - 1 \} \{ \varepsilon M \omega^2 (G_{ii}^{oo} - G_{ii}^{on}) - 1 \} = 0$$

for every Cartesian component i . We can determine from this equation the resonance frequencies in terms of the relative position of the impurity atoms, by means of the function G^{oh} . ε is here regarded as a known parameter.

For a simple cubic lattice, whose coupling parameters satisfy the relation $\Phi_{ij}^{mn} + 100 = \text{const } I^{mn}$ (I is the unit matrix), for which there exist tables of the Green function, we have:

$$(2.3) \quad \omega_1^2 = \frac{1}{\varepsilon M (G^{oo} + G^{on})}, \quad \omega_2^2 = \frac{1}{\varepsilon M (G^{oo} - G^{on})}.$$

The ratio of the moduli of the resonance amplitudes is obtained from the Eq. (1.9), making use of (2.1) and (2.3). It is equal to unity. Thus, $\mu_{R,ex}(\omega)$ is a superposition of two Lorentzian lines with the same heights and widths:

$$(2.4) \quad n_R(\omega) = \text{const} \left\{ \frac{1}{(\omega - \omega_1)^2 + \lambda^2/4} + \frac{1}{(\omega - \omega_2)^2 + \lambda^2/4} \right\}.$$

Strictly speaking, the widths of the two lines are different. With our model assumptions the differences can easily be calculated. It turns out then that the final result is the same, since the above differences are small and their contribution is negligible. This fact follows from the formula (2.7).

$u_{R,ex}(t)$ (the Fourier original) has therefore the form:

$$(2.5) \quad u_{R,ex}(t) = u_{R,ex}(0) e^{-\frac{\lambda}{2}t} \cos \nu t \cos \omega_R t,$$

where $\omega_R = \frac{1}{2}(\omega_1 + \omega_2)$, $\nu = \frac{1}{2}(\omega_1 - \omega_2)$. Hence,

$$(2.6) \quad \langle u_{R,ex}^2(t) \rangle_{w(n_s)} = \langle u_{R,ex}^2(t) \rangle e^{-\lambda t} \cos^2 \nu t.$$

This formula describes the time behaviour of the mean square of the perturbed part of the displacement vector of the impurity atoms, the interaction of which with another impurity is taken into account by means of terms of the type $P(1_r)$ [cf. Eq. (1.7)].

The modulation frequency ν depends of course on the distance between the interacting defects, we shall therefore write hereafter ν_r . We can now present an explicit form of the coefficients f_r entering the formula (1.7):

$$(2.7) \quad f_r = f_{st} \exp \left\{ -\kappa^2 \langle u_{R,ex}^2(0) \rangle_{w(n_s)} e^{-\lambda t} \cos^2 \nu_r t \right\}$$

and, finally, the coefficient of the recoilless emission for a finite but low isotropic concentration of the impurity of the Mössbauer atoms in the above described matrix can be written in the form:

$$(2.8) \quad f(t) = \left[1 - \sum^R P(1_r) \right] f_{st}^{1+\Phi e^{-\lambda t}} + \sum^R P(1_r) f_{st}^{1+\Phi e^{-\lambda t} \cos^2 \nu_r t} \\ = f_{st}^{1+\Phi e^{-\lambda t}} \left[1 - \sum^R P(1_r) + \sum^R P(1_r) f_{st}^{-\Phi e^{-\lambda t} \sin^2 \nu_r t} \right].$$

Since $\Phi > 0$, each of the modulation terms of the form $f_{st}^{-\Phi e^{-\lambda t} \sin^2 \nu_r t}$ is greater than unity, since always $f_{st} < 1$. The interaction of defects yielding a modulation of the resonance lines leads therefore to an increase at each instant of the coefficient $f(t)$ as compared with its value for very low concentrations. However, it is evident that this effect is important only for sufficiently large values of $\Phi e^{-\lambda t}$ — i.e., for times which do not drastically exceed λ^{-1} . On the other hand, for times belonging to this interval, important may be only these terms, ν_r , of which does not appreciably exceed λ , so that the first maximum of $\sin^2 \nu_r t$ occurs in the interval of not too small $e^{-\lambda t}$. Therefore, disregarding a priori interactions with distant defects — i.e., with defects leading to small ν_r — does not in fact influence the final value of $f(t)$.

We shall now consider numerical examples which make it possible to examine possibilities of measurements of $f(t)$.

3. Numerical examples

First, we present results for the model of the matrix and the defect described above and then we discuss their generalizations.

All times and frequencies appearing in our calculations are in the units of λ^{-1} and λ , respectively.

From the formulae (2.3) we can calculate the frequency $\frac{\omega_1^2 - \omega_2^2}{\omega_1^2 + \omega_2^2}$ in terms of G^{00} and G^{0h} . The ratio λ/ω_R is known from the considerations for an isolated defect [7,3]. Thus, we calculate ν_r/λ .

It is known that sharp resonance lines which are of interest to us have the ratio λ/ω_R of the order 10^{-2} [8, 9, 7]. For an isotopic defect, $\lambda/\omega_R = \frac{\pi}{4}(3\varepsilon)^{-1}$. It follows that the required ratio λ/ω_R must have atoms of the impurity mass exceeding by about 200 times the mass of the matrix atoms; it is therefore evident that there must appear a change in the coupling parameters of the defect, which we do not want to take into account, at least directly, in our calculations (cf. the remarks made in considering the choice of the model).

Two procedures are therefore possible. First, we can approach the real relaxation times of the resonance modes by assuming unusually large masses of the impurity atoms; next, we can take realistic values of the masses and perform calculations for wider resonance lines. The first possibility — i.e., modelling the changes of the coupling parameters by means of an unusually large mass of the defect — has a shortcoming in that it leads to lower resonance frequencies than those observed for the required ratio λ/ω_R . The second method assumes deviations from the proper λ/ω_R , but since only the ratio of λ^{-1} to the lifetime of the nuclear Mössbauer level is relevant, this shortcoming can be compensated by introducing appropriate lifetimes, shorter than real, of the Mössbauer level. However, it turns out finally that the numerical results do not markedly depend on the assumed ε . Consequently, we present results obtained for a more realistic value $\varepsilon \approx 33$ for which $\nu_r/\lambda = 25.4G^{0h}/G^{00}$.

The Green functions used here have been presented in Tables in [6] for $h = \{(0, 0, 0), (1, 0, 0), (1, 1, 0), (2, 0, 0)\}$ in terms of the frequency. The remaining required values of G^{0h} have been derived by extrapolation in accordance with a suitable curve of the continuum Green tensor for an isotropic medium.

To calculate $f(t)$, we have to know f_{st} and the parameter Φ [cf. the formulae (1.6) and (2.8)]. The maximum Φ is determined by the recoil energy transferred by recoil of the signal photon from the transition preceding the Mössbauer emission, which typical interval of the recoil energy is $10^{-1} - 10^{-3}$ eV, while the effective Φ to be used in the formulae (1.6) and (2.8) depends also on the geometry of the spectrometer, mainly by the system of the axes of the source and the signal detector and the source and the spectrometer detector, since for the value of $f(t)$ only the component of the vibration generation in the direction of the vector κ is important. For this reason, we present results for a number of values of the parameter Φ . For f_{st} , we have chosen the value 0.1, which is close to the typical values in the case of the Mössbauer nuclids considered, at low temperatures. The results are shown in Figs. 1, 2, 3.

The concentration $c = 0$ corresponds to $f(t)$ for an isolated defect. The curves for concentrations lower than $5 \cdot 10^{-4}$ of the impurity atoms per matrix site have not been drawn, so that the figures may be clear.

For other values of f_{st} , the behaviour of $f(t)$ is similar.

The graphs presented illustrate also the dependence of $f(t)$ on parameter Φ .

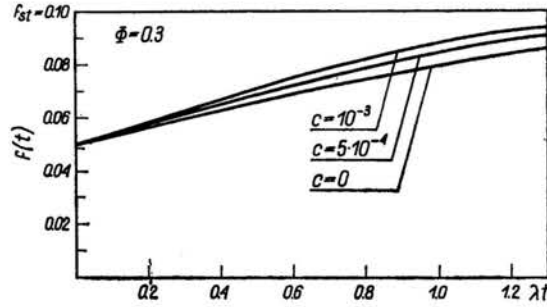


FIG. 1.

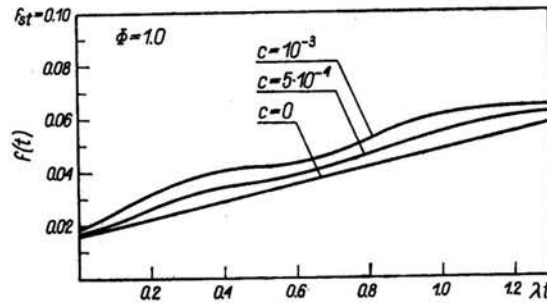


FIG. 2.

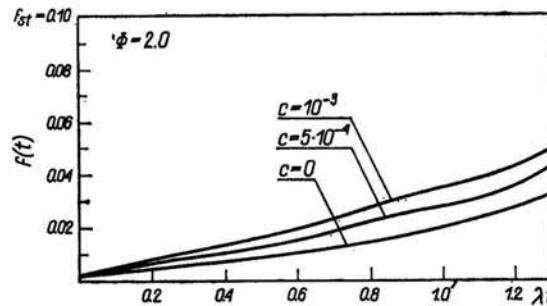


FIG. 3.

Before proceeding to certain problems referring to a possible experiment, we consider similarly to the case of an isolated defect, the most important assumptions under which the numerical results presented have been derived. These assumptions, together with those inherited from the considerations for an isolated defect [1], concern primarily the models of the matrix and the defect.

3.1. The matrix model

An application of the model of a simple cubic lattice with special values of coupling parameters yields only splitting into two lines. In the general case — i.e., for arbitrary admissible coupling parameters — the splitting into two, four or six lines is possible, with pairs of equal amplitudes moduli. The situation is similar in the case of other cubic

lattices — such as f.c.c. and b.c.c. [3, 5]. However, the time behaviour of $f(t)$ remains qualitatively the same, the only small modification occurring in the modulation effect. The calculations for a hypothetical spectrum consisting of four decomposed lines, analogous to the localized lines investigated in [5], lead to numerical deviations from $f(t)$ in Figs. 1, 2, 3 not exceeding 2%. This is due to the fact that in accordance with (1.7) $f(t)$ contains a considerable number of modulation terms with different values of the ratio $f_r/f_{st}^{1+\phi e^{-\lambda t}}$, describing the magnitude of the contribution of the modulation effect from r -distant impurity atoms. A great number of the lines leads to the appearance in the modulation terms of, instead of $\sin^2 \nu_r t$, combinations of trigonometric functions depending on the frequency differences of many lines. The greater their number, the closer the value of these combinations to zero and the smaller the modulation effect. However, the splitting into many lines has the property that only the extreme lines on the frequency axis have comparatively large frequency differences, comparable to the widths of the lines [5]. The remaining lines are concentrated near the frequency of the unsplit line. Consequently, only for small r are the modulation terms decreased due to the contribution of combinations of all split lines. On the other hand, for further r , only the contributions of the modulation of extreme lines are important and the result is similar to that obtained on the basis of the simplified model and contributes to its latter validity.

It is known that for lower frequencies the Green function of the lattice can be replaced by the corresponding continuum Green tensor [3]. This considerably extends the applicability of the results obtained for a definite matrix, since the appropriate model of the continuum leading to the continuum Green tensor considered contains in the low frequency limit a whole class of discrete models. Thus, our remarks prove that we can expect good qualitative agreement of the behaviour of $f(t)$ for real crystal lattices with $f(t)$ obtained on the basis of the simple model introduced above.

3.2. Model of a defect representing an impurity atom

The remarks preceding the numerical results emphasized the fact that in real situations the resonance lines with widths which are of interest to us must appear, owing not only to the large mass of the impurity atoms but also as a result of changes of coupling parameters of the defect with the matrix. It is known that these changes [3] may lead to new "defect" degrees of freedom resulting in a splitting into additional lines. However, it should be borne in mind that in the problem considered neither the modes the nod of which is at the impurity atom, nor the higher frequency modes the relaxation times of which are too short, are important, and this decreases somewhat the number of relevant decompositions. Anyhow, the above reasoning for the model of the matrix remains valid and, consequently, we can still expect agreement of the measurements with the derived behaviour of $f(t)$.

3.3 Assumption on a homogeneous spatial distribution of the impurity

This assumption is certainly not satisfied satisfactorily for every choice of the matrix and the impurity. In fact, if for any reason — say in view of a stronger coupling defect-defect than defect-matrix — a generation of defect aggregates is energetically favourable,

then special procedure in preparing the sample is necessary in order to obtain the most homogeneous distribution possible. It is possible, for instance, to scatter impurity aggregates by heating, and then, to cool the sample retaining the distribution of defects obtained.

4. Conclusions

We have presented in this paper a suggested application of certain results of the dynamics of point defects to the Mössbauer effect, and this application should be useful for the theory of defects, providing it with a new tool for experimental investigation of mechanical properties of defects. Strictly, we have in mind utilizing the influence of the relaxation of the excited vibrational resonance modes on the coefficient of the recoils gamma emission.

Observe that in the course of preparation of the paper, HOY and WINTFRSTEINER performed a pioneering experiment [10] confirming the qualitative dependence of the coefficient f on time. The low degree of accuracy of the existing measurement results makes a quantitative comparison impossible so far.

In the second part of the paper, we investigated finite concentrations of the impurity atoms, since for technical reasons measurements for higher concentrations are easier. The first step consisted of pairwise interactions, for which there exists a theory. These results are not based on the model assumptions, which are needed only in the performance of calculations concerning the modulation.

Next, we discussed the problem of choice of the model of the defect and the matrix. For computational reasons we chose the model of isotopic defect in a simple cubic lattice with harmonic interactions between nearest neighbours. On the basis of this model, we calculated the values of decomposition of the resonance lines in terms of the distance between the defects, and this made it possible to determine the magnitude of the modulation effect for various configurations of the impurity atoms, and finally to calculate the coefficient f for various concentrations. It turned out that the decomposition of the resonance lines leads, via the modulation effect, to an increase in the value of f as compared with small concentrations.

The numerical examples presented contain the coefficient f in terms of time for various values of the following parameters: the concentration and the quantity Φ determining the magnitude of the vibrational excitation. In discussing the numerical results, we have presented a reasoning supporting the fact of a qualitative agreement of the above effects with phenomena which may occur in real crystals. The principal reason for this agreement is a weak dependence of the modulation effect on the models of the defect and the matrix. This is the fact favourable for the measurement of the relaxation of the vibrational resonance, since already a simple model theory makes it possible to predict the perturbation introduced by an increase in concentration. On the other hand, this result leads to the fact that the modulation effect itself does not constitute a distinct test of the assumed models of the defect and the matrix.

Thus, the conclusion is as follows: the measurement of the relaxation time of resonance modes for finite concentrations is perturbed by the modulation effect and therefore in

order to determine the width of a separate resonance we must have at our disposal measurements of the coefficient f for at least two concentrations, which makes it possible to separate the modulation effect, provided we know qualitatively (in the sense described above) $f(c)$.

One more remark. The modulation effect may appear not only in direct measurements of the time dependence of a physical quantity, a typical example of which are coincidence measurements, but also in measurements of quantities already averaged over time. It is readily observed for instance that a usual measurement of the coefficient f is also modified by the modulation — namely, f in terms of the concentration of the Mössbauer-active atoms increases more rapidly than linearly in the range of concentrations in which the defect-defect interaction begins to play a role. A linear dependence would occur in the case of independent contributions of various atoms.

A similar effect is to be expected for infra-red absorption by localized modes [11] and also by sharp resonance modes. In this case, owing to the dependence of the absorption coefficient on the displacement by means of a factor of the Debye-Waller type, the modulation contributions of various atoms after averaging do not cancel each other, but rather lead to an increase of the absorption coefficient.

However, these effects are weaker than in the case of the measurement of a time dependence, since usually there occurs averaging not only over time of duration of the process but also over its initial phases [12].

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