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Abstract

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THE MÖSSBAUER EFFECT ON IMPURITY NUCLEI AT LOW TEMPERATURES

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The study of the Mössbauer effect (^{1}) on impurity nuclei of a solid solution is of considerable interest, since it opens up some possibility of changing the intensity of the Mössbauer line of one and the same γ-radioactive nucleus by placing it in different solid solvents, and of increasing the intensity of the Mössbauer line by an appropriate choice of the solid solvent.

In the general case the intensity of the Mössbauer line of an impurity nucleus depends on the ratio of the masses of the impurity and solvent atoms and on the binding energy of the impurity atom with the solvent atoms. Calculation of the intensity of the Mössbauer line in this case is possible only if the solutions of the dynamical problem of the vibrations of the solvent lattice containing the impurity are known. Attempts at such a solution for certain models were made in ($^{2-4}$); in (^{5}) an attempt was made to solve the dynamical problem for a monatomic cubic crystal, one of whose atoms is replaced by an atom with a γ-radioactive nucleus; the solutions obtained were then used to determine the Mössbauer factor (^{6}). However, along this path one encounters great difficulties connected with determining all the vibrational frequencies of a nonideal crystal.

To avoid solving the dynamical problem for a crystal lattice with impurity atoms, we shall consider here the Mössbauer effect at low temperatures ($T < 20$°K) for substitutional solutions of very low concentration, taking into account, for simplicity, only the ratio of the masses of the impurity and solvent atoms. The Hamiltonian describing the vibrations of such a lattice is written in the form

$$H = H_0 + \mu \frac{\mathbf{p}^2_{j''n''}}{2M_{j''}}, \tag{1}$$

where H_0 is the Hamiltonian operator of the ideal solvent lattice; $\mu = M_{j''}/m_{j''} - 1$; $M_{j''}$, $m_{j''}$ are, respectively, the masses of the solvent and impurity atoms; $\mathbf{p}_{j''n''}$ is the momentum operator of the impurity atom; n is the number of the elementary cell; j is the number of the site in the cell.

In the harmonic approximation the operator H_0 reduces to the form

$$H_0 = \sum_s \hbar\omega_s \left(b_s^+ b_s + 1/2\right),$$

if one sets

$$\mathbf{u}_{jn} = \sum_s \sqrt{\frac{\hbar}{2M_j N\omega_s}}\,(b_s + b^+_{-s})\;\mathbf{v}_j(s)\exp\{i\mathbf{f}\mathbf{R}_n\},$$

$$\mathbf{p}_{jn} = -iM_j \sum_s \sqrt{\frac{\hbar\omega_s}{2M_j N}}\,(b_s - b^+_{-s})\;\mathbf{v}_j(s)\exp\{i\mathbf{f}\mathbf{R}_n\},$$

where $\mathbf{u}_{jn}$ are the displacements of atoms from equilibrium positions; ω are the frequencies of the normal vibrations of the ideal lattice; $s = (\mathbf{f}, \tau)$; $\mathbf{f}$ is the wave vector of the vibration; τ is the branch number; b^+_s, b_s are the creation and annihilation operators

phonons; $\mathbf{v}_j(s)$ is the displacement from the equilibrium position of the j-th atom of the cell $n = 0$; N is the number of cells of the crystal.

The probability of transition of an impurity nucleus from the excited state l_1 to the ground state l_0 with emission of a γ-quantum of energy E_σ can be represented, according to (^{7}), in the form

$$w(E_\sigma) = \frac{2|L_{l_1 l_0}|^2}{\gamma}\,\mathrm{Re}\int_0^\infty d\tau\,B(\tau)\,e^{i\tau(E_0 - E_\sigma) - \tau\hbar\gamma/2}. \tag{2}$$

Here

$$B(\tau) = \frac{1}{Z}\,\mathrm{Sp}\left\{e^{-\beta H + i\tau H} e^{-i\chi\mathbf{u}} e^{-i\tau H} e^{i\chi\mathbf{u}}\right\};$$

$L_{l_1 l_0}$ is the matrix element of the operator of the energy of interaction of the electromagnetic field with the nucleus; E_0 is the energy difference between the ground and excited states of the impurity nucleus (we omit here its indices $j''n''$); γ is the "width" of the excited level (with allowance for the coefficient of internal conversion (^{8})); Z is the statistical sum for the phonons; χ is the wave vector of the emitted γ-quantum; $\beta = 1/kT$. Further putting

$$\mathbf{u}(\tau) = \exp(i\tau H)\,\mathbf{u}\,\exp(-i\tau H), \tag{3}$$

we represent the expression for $B(\tau)$ in the form

$$B(\tau) = \frac{1}{Z}\,\mathrm{Sp}\{e^{-\beta H} S_1\} \equiv \langle S_1\rangle,$$

where

$$S_1 = S_{\alpha=1}; \qquad S_\alpha = T \exp\left\{-i\alpha \int_{-\infty}^{\infty} V(z)\, dz\right\}; \qquad V(z) = \chi\mathbf{u}(z)\delta(z-\tau)-$$

$$-\chi\mathbf{u}(z)\delta(z);$$

T is the chronological operator.

Differentiating $\langle S_\alpha \rangle$ and using the fact that $\langle S_0 \rangle = 1$, we find

$$\langle S_1 \rangle = \exp\left\{-i \int_0^1 d\alpha \int_{-\infty}^{\infty} dz\, \frac{\langle TV(z)S_\alpha \rangle}{\langle S_\alpha \rangle}\right\}.$$

Evaluation of the last expression is carried out easily if one uses Wick' s generalized theorem ($^{9-11}$), whereby

$$\langle S_1 \rangle = \exp\left\{-\frac{1}{2} \int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} dz_1 \langle TV(z)V(z_1)\rangle\right\}.$$

Performing here the integration over dz, dz_1 and substituting the expression obtained into (2), we find

$$w(E_\sigma) = \frac{2|L_{l_1 l_0}|^2}{\gamma} \operatorname{Re} \int_0^{\infty} \exp\left\{i\tau(E_0 - E_\sigma) - \frac{\gamma\hbar}{2}\tau + \right.$$

$$\left. + \chi^\alpha \chi^{\alpha'} \langle u^\alpha(\tau) u^{\alpha'}(0)\rangle - \chi^\alpha \chi^{\alpha'} \langle u^\alpha(0) u^{\alpha'}(0)\rangle \right\} d\tau,$$

where

$$G^{\alpha\alpha'}(\tau, 0) = \langle u^\alpha(\tau) u^{\alpha'}(0)\rangle = \frac{1}{Z} \operatorname{Sp}\{e^{-\beta H} u^\alpha(\tau) u^{\alpha'}(0)\}, \qquad \alpha, \alpha' = 1, 2, 3.$$

Expression (3) determines the shape of the emission spectrum of γ-quanta by nuclei at arbitrary temperature and arbitrary form of the operator H; moreover, if the lattice possesses translational symmetry, then $\chi^\alpha \chi^{\alpha'} \langle u^\alpha(\tau) u^{\alpha'}(0)\rangle$ determines the phonon part of the emission spectrum, while $\chi^\alpha \chi^{\alpha'} \langle u^\alpha(0) u^{\alpha'}(0)\rangle$ is the Mössbauer factor. If, however, the lattice contains an impurity atom with a γ-radioactive nucleus, then owing to the appearance of local vibrations the term $G^{\alpha\alpha'}(\tau, 0)$ will also contribute to the intensity of the Mössbauer line. However, as is easy to show, the argument of the function determining this contribution will be proportional to $\sqrt{(\bar{n}_0 + 1)\bar{n}_0}$, where

$$\bar{n}_0 = [\exp(\hbar\omega_0/kT) - 1]^{-1},$$

ω_0 is the frequency of the local vibration; therefore at low temperatures this contribution will be small and the intensity of the Mössbauer line will be determined

only in the Mössbauer factor $\exp\{-\varkappa^{\alpha}\varkappa^{\alpha'}\langle u^{\alpha}(0)u^{\alpha'}(0)\rangle\}$. In what follows we shall restrict ourselves precisely to such temperatures, assuming that H is determined by (1).

Using now the method of temperature Green functions ($^{10-12}$), we represent $G^{\alpha\alpha'}(0,0)$ in the form

$$G^{\alpha\alpha'}(0,0) = \frac{\langle S(\beta)u^{\alpha}(0)u^{\alpha'}(0)\rangle}{\langle S(\beta)\rangle}, \tag{4}$$

where

$$S(\beta) = T\exp\left\{-\int_0^{\beta} H_1(z)\,dz\right\},$$

$$H_1(z) = \exp(zH_0)H_1(0)\exp(-zH_0), \qquad H_0(0) = \mu\frac{p^2}{2M}.$$

To compute (4), consider the function

$$G^{\alpha\alpha'}(t,t') = \frac{\langle S(\beta)Tu^{\alpha}(t)u^{\alpha'}(t')\rangle}{\langle S(\beta)\rangle} = \frac{\langle Tu_0^{\alpha}(t)u_0^{\alpha'}(t')S(\beta)\rangle}{\langle S(\beta)\rangle},$$

where $u^{\alpha}(t)$ is defined by (3), $u_0(t) = \exp(tH_0)u(0)\exp(-tH_0)$. It is easy to verify that $G^{\alpha\alpha'}(t,t') = G^{\alpha\alpha'}(t-t')$. Further using Wick' s theorem generalized to the case $T \neq 0$ ($^{10-12}$), we represent the expression for $G^{\alpha\alpha'}(t,t')$ in the form

$$G^{\alpha\alpha'}(t,t') = G_0^{\alpha\alpha'}(t,t') - \frac{\mu}{M}\int_0^{\beta} dz_1 Q^{\alpha\gamma_1}(t,z_1)D^{\alpha'\gamma_1}(t',z_1). \tag{5}$$

Here

$$G_0^{\alpha\alpha'}(t,t') = \langle Tu_0^{\alpha}(t)u_0^{\alpha'}(t')\rangle, \qquad Q^{\alpha\gamma_1} = \langle Tu_0^{\alpha}(t)p_0^{\gamma_1}(z_1)\rangle, \qquad \gamma_1 = 1,2,3,$$

and the function $D^{\alpha'\gamma_1}(t',z_1)$ is determined by the equation

$$D^{\alpha'\gamma_1}(t',z_1) = Q^{\alpha'\gamma_1}(t',z_1) - \frac{\mu}{M}\int_0^\beta dz_2 K^{\gamma_1\gamma_2}(z_1,z_2) D^{\alpha'\gamma_2}(t',z_2), \qquad (6)$$

with

$$K^{\gamma_1\gamma_2}(z_1,z_2) = \langle T p_0^{\gamma_1}(z_1) p_0^{\gamma_2}(z_2)\rangle, \qquad p_0^{\gamma}(z) = \exp(zH_0)p^{\gamma}(0)\exp(-zH_0).$$

It is easy to verify that

$$F^{ik}(x,x') = F^{ik}(x-x'),$$

where $F^{ik} = G^{\alpha\alpha'}$, $G_0^{\alpha\alpha'}$, $K^{\gamma_1\gamma_2}$, $Q^{\alpha\gamma_1}$, and for $x - x' < 0$

$$F^{ik}(x-x') = F^{ik}(x+\beta-x'). \qquad (7)$$

Extending $F^{ik}(x-x')$ periodically over the whole line ([11]), we obtain that relation (7) holds for all $x - x'$. This makes it possible in expressions (5) and (6) to replace the lower limit by $-\beta$, placing a factor 1/2 before the integral. Expanding further all functions in (6) in a Fourier series, we express the Fourier coefficients of the function $D^{\alpha'\gamma_1}(t',z_1)$ through the Fourier coefficients of the functions $Q^{\alpha'\gamma_1}(t',z_1)$, $K^{\gamma_1\gamma_2}(z_1,z_2)$, which are easily calculated if the solutions of the dynamical problem of the ideal solvent lattice are known, i.e. ω_s and $\mathbf{v}_j(s)$. Knowing, in this way, $D^{\alpha'\gamma_1}$, from equation (5) one can determine the function $G^{\alpha\alpha'}(t,t')$, which for $t,t' = 0$ determines the Mössbauer factor.

Since, in practice, the solutions of the dynamical problem for the ideal lattice are unknown, to estimate the Mössbauer factor of an impurity atom we shall consider a model monatomic crystal in which ω_s depends

only on $\mathbf{f}$. Then the Fourier coefficient for $D^{\alpha'\gamma_1}(t',z_1)$ is written in the form

$$D_n^{\alpha'\gamma_1} = \frac{\hbar}{\beta N}\left[\sum_f \frac{\varepsilon_n}{\varepsilon_n^2+\varepsilon_f^2}\right] \Bigg/ \left[1 + \frac{\mu}{N}\sum_f \frac{\varepsilon_f^2}{\varepsilon_f^2+\varepsilon_n^2}\right],$$

where $\varepsilon_n = 2\pi n/\beta$, $\varepsilon_f = \hbar\omega_f$. Substituting the value found for $D^{\alpha'\gamma_1}(t',z_1)$ in (5), we compute $G^{\alpha\alpha'}(0,0)$; in doing so

$$\chi^{\alpha}\chi^{\alpha'}G^{\alpha\alpha'}(0,0) = \sum_f q_f^2(2\bar{n}_f+1) + \frac{2\mu R_0}{\beta}\sum_{n=-\infty}^{\infty}\left[\left(\frac{1}{N}\sum_f \frac{\varepsilon_n}{\varepsilon_f^2+\varepsilon_n^2}\right)^2\right] \Bigg/ \left[1+\frac{\mu}{N}\sum_f \frac{\varepsilon_f^2}{\varepsilon_f^2+\varepsilon_n^2}\right], \qquad (8)$$

Fig. 1

Figure 1: Fig. 1

where $R_0 = \chi^2\hbar^2/2M$ is the recoil energy, $q_f = \chi\hbar/\sqrt{2MN\hbar\omega_f}$.

Expression (8) determines the dependence of the Mössbauer factor on the parameter μ for the model under consideration at an arbitrary value of the temperature and can be used for the numerical calculation of the Mössbauer factor. Here, however, we shall restrict ourselves to the case $T \to 0$, assuming a linear dependence of ε_f on $\mathbf{f}$. In this case, for the Mössbauer factor we obtain the expression

$$f = \exp\left\{-\frac{3R}{2\varepsilon_0}\,\varphi(\mu)\right\},$$

where

$$\varphi(\mu) = (1+\mu)^{-1}\left(1 + \frac{12\mu}{\pi}\int_0^{\pi/2} \frac{(\tan x - x)^2(\tan^2 x + 1)\,dx}{\tan^3 x\left[(1+\mu)\tan^3 x - 3\mu(\tan x - x)\right]}\right),$$

$$R = \frac{\chi^2\hbar^2}{2m}, \qquad \varepsilon_0 = \hbar\omega_{\max}.$$

Fig. 1

The dependence of the function $\varphi(\mu)$ on the ratio M/m is presented graphically in Fig. 1*. It follows from the graph that the Mössbauer factor drops sharply as the ratio M/m increases. Thus, in order to increase the intensity of the phononless emission line of γ quanta by an impurity nucleus, it is necessary to place it in crystals with as large an atomic mass as possible (for the same Debye temperature of the crystals). The most favorable will be solvent crystals with a large atomic mass and a high Debye temperature.

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Note: Figure translations are in progress. See original paper for figures.

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