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Abstract

Full Text

PHYSICS

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ON THE THEORY OF DEFECTS FORMED IN SOLIDS BY RECOIL ATOMS UNDER GAMMA IRRADIATION

(Presented by Academician G. V. Kurdyumov, 8 XII 1961)

In the lines of the γ -spectrum emitted by the nuclei of atoms of a solid, two components can be distinguished (^{1,2}): a) one with the natural width Γ_1 and energy equal to the transition energy ε ; b) one with width

$$\Gamma_2 \approx \varepsilon \frac{v}{c} \quad (1)$$

and with an energy shifted relative to ε by the amount

$$W = \frac{\varepsilon^2}{Mc^2}, \quad (2)$$

equal to the recoil energy acquired by the emitting nucleus of mass M ; here v is the mean velocity of the thermal motion of the emitting nucleus, and c is the speed of light; therefore part of the γ -emission events is accompanied by the appearance of recoil atoms with considerable energy. These atoms may produce various kinds of defects in the crystal. Some results for the problem of the formation of defects under the action of recoil atoms can be obtained, without resorting to detailed models of defect formation by atoms with large kinetic energy (³), on the basis of considerations analogous to those developed in (^{4,5}) for the kinetics of processes caused in condensed bodies by the action of fast particles.

The mean duration τ_2 of a single γ -emission event accompanied by the appearance of a recoil atom is

$$\tau_2 \approx \frac{\hbar}{\Gamma_2} \approx \frac{\hbar c}{\varepsilon v}. \quad (3)$$

It follows from this that, for reasonable values of v and ε , the conditions

$$\tau_2 \ll \frac{d}{v} \quad \text{or} \quad \frac{\hbar c}{\varepsilon} \ll d, \quad (4)$$

are satisfied, where d is the interatomic distance and d/v is a time of the order of the period of thermal vibrations of the atoms. Thus, if $v \approx 5 \cdot 10^5$ cm/sec and $\varepsilon \approx 4 \cdot 10^6$ eV, then $\tau_2 \approx 10^{-17}$ sec, ($d/v \approx 10^{-13}$ sec). Therefore one may consider that the emission of a γ -quantum and the emergence of a recoil atom with energy W occur practically instantaneously. The latter usually greatly exceeds the threshold energies (per particle) $E_1 \gg kT$, $E_2 \gg kT$, etc., of various activation processes possible in solids: vacancy formation, the melting process, and so on. For example, at $\varepsilon \approx 4 \cdot 10^6$ eV, $M \approx 5 \cdot 10^{-23}$ g, $W \approx 550$ eV ($E \approx 0.5 \div 3$ eV). The instantaneous release of recoil energy leads to the sudden appearance, in a small volume of the crystal, of a very high energy density, which gradually dissipates among the densely arranged interacting particles of the solid.

However, during a short interval of time immediately after the act of γ -quantum emission, the local energy density in some volume surrounding the emitter is so large that nonlinear effects

in the motion of the particles of such a region can no longer be regarded as small perturbations. At the same time, they can play a significant role in the formation of defects. For example, individual particles located in a region of increased energy density may much more easily acquire energy sufficient to pass over a potential barrier of height $E \gg kT$. After the local energy density has decreased appreciably and the probability of defect formation has become relatively small, further dissipation of energy will occur in accordance with the laws of thermal conductivity usual for the given solid.* Therefore the process of dissipation of the energy W may be divided into two stages. The first covers the time τ from the moment of emission of the γ -quantum to the moment when the local energy density in the vicinity of the emitter has noticeably decreased owing to the distribution of this energy in some volume Q . During the time τ , when the local energy density is significant, the processes of defect formation occur most intensively. To estimate Q and τ we shall use a criterion analogous to that applied in ^(4,5) to the dissipation of the energy transferred to the substance by a fast particle: we shall assume that the number of defects in the volume Q becomes quasistationary only when the volume energy density decreases to values satisfying the condition

$$(W/n) \approx (Wd^3/Q\beta) \lesssim E_{\min}, \quad (5)$$

which will subsequently cause a considerable decrease in the probability of defect formation in the vicinity of the emitter; here E_{\min} is the smallest of the threshold values of the energies E_1, E_2 , etc.; n is the number of degrees of freedom in the volume Q ; β is their number in the volume d^3 . The second stage, which is of no interest to us, consists in the subsequent dissipation of the energy W from the volume Q by thermal conduction.

Relation (5) makes it possible to estimate the lower bound of the volume Q corresponding to the quasistationary number of defects formed with threshold

energies $E_{\min}, E_1 > E_{\min}$, etc. From (2) and (5) we obtain for the linear dimensions of the volume Q :

$$l \approx Q^{1/3} \approx d (\varepsilon^2 / \beta M c^2 E_{\min})^{1/3}. \quad (6)$$

The minimum value of the corresponding time scale is equal to

$$\tau_{\min} \approx (l/v_0) \approx (d/v_0) (\varepsilon^2 / \beta M c^2 E_{\min})^{1/3}, \quad (7)$$

where v_0 is the mean velocity of energy transfer in the system. But this value of τ_{\min} is apparently underestimated, since at a high local energy density in the volume Q the mean free path l_f for energy transfer will be small. Therefore the values of τ will apparently be close to the maximum value:

$$\tau_{\max} \approx \frac{l^2}{\chi_c} \approx \frac{l^2}{v_0 d} \approx \frac{d}{v_0} \left(\frac{\varepsilon^2}{\beta M c^2 E} \right)^{2/3}, \quad (8)$$

where it is assumed that $l_f \approx d$ and the local thermal diffusivity is $\chi_c \approx d v_0$. Because of the complexity of the processes of energy redistribution among many interacting particles at high energy density, a more precise determination of Q , l , and τ appears difficult. Therefore Q (or l) should be regarded as an adjustable parameter of the method, considered below, for calculating the rates of defect formation. However, the relations obtained above allow one to assert that the volume Q contains a large number of degrees of freedom.** This provides a basis, when calculating the num-

* Over the time interval during which the local energy density is large, the local conditions of energy transfer may differ substantially from those usual for the given solid.

** For $\varepsilon \approx 4 \cdot 10^6$ eV, $M = 5 \cdot 10^{-23}$ g, $\beta = 3$, $E_{\min} = 0.5$ eV, $d = 3 \cdot 10^{-8}$ cm we obtain $n \gtrsim 1200$, $Q \gtrsim 400 d^3 \sim 10^{-20}$ cm³, $l \gtrsim 10^{-7}$ cm.

of the defects formed at the first stage of the energy distribution W , make use of a statistical description of the volume Q , and introduce the local entropies $S(U_{ef})$ and $S(U_{ef} - E)$ of the volume Q , corresponding to the energies $U_{ef} = U + W$ and $U_{ef} - E$, where U is the energy of the volume Q immediately before emission of the γ -quantum. Then the probability of formation, in the volume Q during the time τ , of a defect with threshold energy E (provided that U and W are given) is equal to

$$P \left(\frac{E}{U_{ef}} \right) = \exp \left[\frac{S(U_{ef} - E) - S(U_{ef})}{k} \right]. \quad (9)$$

The probability of the same event, irrespective of the value of U , is equal to

$$P(W) = \int f(U)P\left(\frac{E}{U_{ef}}\right) dU \quad (10)$$

and is a function of W (since U_{ef} depends on W),

$$f(U) = \frac{1}{\sqrt{2\pi\alpha^2}} \exp\left[-\frac{(\Delta U)^2}{2\alpha^2}\right], \quad (11)$$

where $\Delta U = U - \bar{U}$, $\alpha^2 = \overline{(\Delta U)^2}$.

Using (9) and (11), we write (10) in the form

$$P(W) = \frac{1}{\sqrt{2\pi}} \int \exp\left[-\frac{(\Delta U)^2}{2\alpha^2}\right] \exp\left[\frac{S(U_{ef} - E) - S(U_{ef})}{k}\right] \frac{dU}{\alpha}. \quad (12)$$

Expanding $S(U_{ef} - E)$ in a series in powers of E , we obtain

$$\frac{S(U_{ef} - E) - S(U_{ef})}{k} = -\frac{E}{kT(U_{ef})}, \quad (13)$$

where

$$T(U_{ef}) = T(\bar{U} + W + \Delta U) \simeq T_j + \frac{\Delta U}{C} \quad (14)$$

is the local temperature of the region Q , corresponding to the energy U_{ef} ; $T_j = T(\bar{U} + W)$ is the temperature of the same region, corresponding to the energy $\bar{U} + W$; the quantity

$$C = \beta c_0 \frac{Q}{d^3} \simeq C_0 \frac{W}{E_{\min}} \quad (15)$$

represents the heat capacity of the volume Q ; C_0 is the heat capacity calculated per one degree of freedom*. Using the relation

$$\frac{1}{T(U_{ef})} \simeq \frac{1}{T_j(1 + \Delta U/CT_j)} \simeq \frac{1}{T_j} \left(1 - \frac{\Delta U}{kT_j}\right)$$

and equalities (12), (13), and (14), we obtain for the probability $P(W)$ the expression

$$P(W) = \exp\left(\frac{E^2 \alpha^2}{\alpha_j^2 \alpha_j^2}\right) \exp\left(-\frac{E}{CT_j}\right), \quad (16)$$

where $\alpha_j^2 = kCT_j^2$. If C does not depend on T , then

$$T_j = T + \frac{W}{C} = T \left(1 + \frac{W}{CT}\right),$$

$$\Delta T = T_j - T = \frac{W}{C} \simeq \frac{E_{\min}}{k}. \quad (17)$$

* In deriving relations (13) and (14), it was assumed that at least one of the conditions is satisfied: a) C does not depend on T , b) $(E/CT_{ef})^2 \ll 1$ and $(\Delta U/CT_j) \ll 1$.

Taking into account that $\alpha^2 = kCT^2$, we rewrite (16) in the form

$$P(W) = \exp\left[-\frac{E^2}{2\alpha^2(1+W/CT)^4}\right] \exp\left[-\frac{E}{kT(1+W/CT)}\right]. \quad (18)$$

It follows from this that a substantial decrease in the crystal temperature T will have a relatively weak effect on the probability of defect formation by recoil atoms, since the main contribution to T_j is made by ΔT , which is practically independent of T .

The dissipation of the energy W from the volume Q over time intervals exceeding τ leads to a kind of “freezing” of defects that have already formed; their “annealing,” caused by thermal motion, proceeds relatively slowly, especially at low temperatures.

In the calculation presented above it was assumed that the mean time between two successive acts of γ -radiation in the volume Q is significantly greater than τ .

Emission of γ -quanta with the natural line width $\Gamma_1 (\lesssim 5 \cdot 10^{-6} \text{ eV}^{(1)})$ does not lead to the appearance of recoil atoms and defects, since in this case the duration of the γ -emission act $\tau_1 \simeq \hbar/\Gamma_1 (\gtrsim 10^{-10} \text{ sec.})$ considerably exceeds the period of oscillation of the atoms of the crystal ($\simeq 10^{-13} \text{ sec.}$), and during the time τ_1 the emitting atom performs a large number of thermal oscillations.

An analogous consideration can be applied to defect formation and to some other effects caused by recoil nuclei and fragments arising in other nuclear processes occurring in the atoms of the crystal matrix or impurity ⁽³⁾.

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