Collective spontaneous emission by polyatomic systems

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Collective spontaneous emission by a completely excited extended polyatomic system is considered. The time and angular dependences of the emission for a volume of arbitrary shape are investigated. The dynamics of radiative decay and shape of the emission spectrum are obtained in the isotropic case.

1. INTRODUCTION

Collective effects in spontaneous emission from a system of identical two-level quantum-mechanical radiators were investigated for the first time by Dicke,[1] who studied especially thoroughly the case of a radiating system contained within a volume of linear dimensions much smaller than the radiation wavelength.

To determine the possibility of collective emission in the optical region, the problem must be generalized to include systems of large extent. Collective effects in spontaneous emission from two excited atoms separated by an arbitrary distance were considered, taking the Coulomb interaction into account, in[2,3].

Studies of extensive many-atom systems are usually based on the assumption of a predominant emission direction, and are limited to an account of interaction with only a single mode of the electromagnetic field.[4-6] Directionality of the emission can be determined by the geometry of the system's volume[4,6] or by the direction of the exciting pulse.[5] Spontaneous emission from a fully inverted extended many-atom system within a volume of arbitrary shape where a preferential direction is lacking and the radiation initially has noncollective angular and temporal properties, was considered by Ernst and Stehle,[7] who investigated spectral line broadening and the angular correlation of the photons. However, in solving this problem by the Wigner-Weisskopf method several simplifications were employed which led to an incorrect dependence of the collective effects on the volume of the system.

In the present work we consider the same problem, using the perturbation-theoretical diagram method[8] that we applied in[3] to the case of spontaneous emission from two atoms. The temporal and angular characteristics of the radiation are investigated. For the isotropic case the dynamics of the radiative decay process and the shape of the spectrum are determined.

2. COLLECTIVE RADIATION CONSTANTS FOR AN EXTENDED SYSTEM

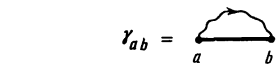
Let us consider a system of N two-level atoms with the resonant transition frequency ω_0, within a volume v of linear dimensions much larger than the wavelength λ. We shall assume an atomic density $n = N/v \gg \lambda^{-3}$; consequently, the system will be quasihomogeneous within the volume.

The operator of atomic interaction with the transverse electromagnetic field is[1)]

$$\hat{H}_I = \sum_{\mathbf{k}} \{f_{\mathbf{k}} \hat{R}_{\mathbf{k}}^{+} \hat{A}_{\mathbf{k}} + \text{h.c.}\},$$
$$f_{\mathbf{k}} = -i\left(\frac{2\pi}{K}\right)^{1/2} \frac{\omega_0}{\sqrt{V}} (\mathbf{de}_{\mathbf{k}}), \quad \hat{R}_{\mathbf{k}}^{+} = \sum_a \hat{R}_a^{+} \exp(i\mathbf{K}\mathbf{r}_a). \tag{1}$$

Here $\hat{R}_a^{+}$ is the excitation operator of the a-th atom, $\hat{A}_{\mathbf{k}}$ is the destruction operator of a photon with the wave vector $\mathbf{K}$ and polarization $\mathbf{e}_{\mathbf{k}}$. The matrix elements of the dipole moment operator $\mathbf{d}$ are assumed to be identical for all atoms. The quantization volume V of the electromagnetic field is assumed to be considerably larger than the volume v of the atomic system.

Collective effects in the spontaneous emission are associated with the virtual exchange of a transverse photon between different atoms. This process is described by off-diagonal matrix elements of the self-energy part:

γ_{ab} =  (2)*

$$= \gamma \int d\mathbf{K}\delta(K-\omega_0)[\mathbf{dK}]^2 \exp(i\mathbf{K}\mathbf{r}_{ab}) \Big/ \int d\mathbf{K}\delta(K-\omega_0)[\mathbf{dK}]^2.$$

The diagonal matrix element γ_{aa} equals the radiation constant γ of an isolated atom.

The problem of determining single excited "superradiant" states consists in diagonalizing the matrix γ_{ab}. For states $N^{-1/2}\hat{R}_{\mathbf{k}}^{+}\Phi_0$, where Φ_0 is the ground state of the atomic system and $K \sim \omega_0$, the matrix $\{\gamma\}$ is approximately diagonal:

$$\gamma_{\mathbf{k}_1\mathbf{k}_2} = \frac{1}{N}\sum_{a,b} \gamma_{ab} \exp(-i\mathbf{K}_1\mathbf{r}_a + i\mathbf{K}_2\mathbf{r}_b)$$
$$= \frac{1}{2\pi\omega_0}\frac{N}{v^2}\int d\mathbf{K}\int d\mathbf{r}_a\, d\mathbf{r}_b \delta(K-\omega_0)[\mathbf{dK}]^2 \exp\{i\mathbf{r}_a(\mathbf{K}-\mathbf{K}_1) - i\mathbf{r}_b(\mathbf{K}-\mathbf{K}_2)\}$$
$$= \frac{(2\pi)^5}{\omega_0}\frac{N}{v^2}\int d\mathbf{K}\delta(K-\omega_0)[\mathbf{dK}]^2 \Delta(\mathbf{K}-\mathbf{K}_1)\Delta^*(\mathbf{K}-\mathbf{K}_2), \tag{3}$$

where

$$\Delta(\mathbf{K}-\mathbf{K}_1) = \frac{1}{(2\pi)^3}\int d\mathbf{r} \exp\{i\mathbf{r}(\mathbf{K}-\mathbf{K}_1)\} \tag{4}$$

is a delta function with dispersion $\sim 1/L$. The diagonal matrix element is

$$\gamma_{\mathbf{k}_1\mathbf{k}_1} = N\gamma(\mathbf{K}_1) = \frac{(2\pi)^2}{\omega_0}\frac{N}{v}\int d\mathbf{K}\,\delta(K-\omega_0)[\mathbf{dK}]^2 \tilde{\Delta}(\mathbf{K}-\mathbf{K}_1), \tag{5}$$
$$\tilde{\Delta}(\mathbf{K}-\mathbf{K}_1) = \frac{(2\pi)^3}{v}|\Delta(\mathbf{K}-\mathbf{K}_1)|^2 \approx \Delta(\mathbf{K}-\mathbf{K}_1). \tag{6}$$

If, as an illustrative example, it is assumed that the volume containing the atoms is a rectangular parallelepiped with edges L_X, L_Y, and L_Z, then

$$\gamma(\mathbf{K}_1) = 2\pi\omega_0^{-1}[\mathbf{dK}_1]^2 S, \tag{7}$$

where S is the area of the section of a rectangular parallelepiped, with edges $1/L_X$, $1/L_Y$, and $1/L_Z$ and centered at the point $\mathbf{K}_1$, by a sphere of radius ω_0. In the general case, by first integrating with respect to the vector $\mathbf{K}$ and limiting ourselves to such $\mathbf{K}$ that $\omega_0 \gg |\mathbf{K}-\mathbf{K}_1| \gg 1/L$, we obtain

$$\gamma(\mathbf{K}_1) = 2\pi\omega_0^{-1}[\mathbf{dK}_1]^2 l_{k_1}/v, \tag{8}$$

where $l_{\mathbf{K}_1}$ is the linear dimension of the system in the direction of $\mathbf{K}_1$, averaged over its volume.

It is seen from (7) or (8) that the maximum value of $\gamma_{\mathbf{K}_1\mathbf{K}_2}$ as a function of $\mathbf{K}_1$ corresponds to the direction of the greatest extent L_{max} of the volume, and is of the order $\gamma n\lambda^2 L_{max}$. In the case of a spherical volume, by averaging $\gamma_{\mathbf{K}_1\mathbf{K}_2}$ over the direction of $\mathbf{K}_1$ we obtain

$$\bar{\gamma}_{\mathbf{k}_1\mathbf{k}_1} \approx \gamma n\lambda^2 L \approx \gamma N\lambda^2/L^2. \tag{9}$$

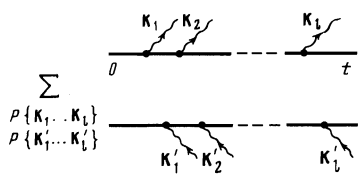
Thus the spatial extent of the system reduces the superradiant constant by the factor $(\lambda/L)^2$ compared with Dicke's problem. The number of single excited superradiant states can be estimated as the ratio between the area of a sphere of radius ω_0 and the cross-sectional area $(1/L)^2$, i.e., as $(L/\lambda)^2$. From the conservation of the trace of the matrix $\{\lambda\}$ we obtain

$$\mathrm{Spur}\{\gamma\} \approx \bar{\gamma}_{\mathbf{kk}}(L/\lambda)^2 \approx N\gamma. \tag{10}$$

This last relation is an approximation, because we have taken into account only the large superradiant constants corresponding to vector magnitudes $K \approx \omega_0$. This diagonalization procedure cannot be used to obtain small radiation constants corresponding to nonresonant K, because the absence of quasi-homogeneity prevents us in the case of $K \sim n^{-1/3}$ from replacing summation over the atoms by integration over the volume of the system.

3. DYNAMICS OF COLLECTIVE SPONTANEOUS EMISSION

Let us assume that all the atoms are initially excited. The probability that, up to a time t, photons with the momenta $\mathbf{K}_1 \ldots \mathbf{K}_l$ will be emitted is represented by the following density matrix element of the system:



(11)

with summation over all permutations P of the wave vectors in the upper and lower lines (the sets $\{\mathbf{K}\}$ and $\{\mathbf{K}'\}$ coincide). Summation over all atoms is performed at each interaction point where a photon line begins.

If Φ_N is the initial state of the system, after the first interaction point the state is

$$\Phi_{\mathbf{k}_1} = \hat{R}_{\mathbf{k}_1}\Phi_N(Z_{\mathbf{k}_1})^{-1/2}, \tag{12}$$

where $Z_{\mathbf{K}_1}$ is the normalization factor: $Z_{\mathbf{K}_1} = \langle \hat{R}_{\mathbf{K}_1} - \Phi_N | \hat{R}_{\mathbf{K}_1} - \Phi_N \rangle$. Following the l-th interaction point we obtain the propagator of the state

$$\Phi_{\mathbf{k}_1\ldots\mathbf{k}_l} = \hat{R}_{\mathbf{k}_l}\ldots\hat{R}_{\mathbf{k}_1}\Phi_N(Z_{\mathbf{k}_1\ldots\mathbf{k}_l})^{-1/2},$$
$$Z_{\mathbf{k}_1\ldots\mathbf{k}_l} = \langle \hat{R}_{\mathbf{k}_l}\ldots\hat{R}_{\mathbf{k}_1}\Phi_N | \hat{R}_{\mathbf{k}_l}\ldots\hat{R}_{\mathbf{k}_1}\Phi_N \rangle. \tag{13}$$

The state $\Phi_{\mathbf{K}_1\ldots\mathbf{K}_l}$, like the state $\hat{R}_{\mathbf{K}}\Phi_0$, approximately diagonalizes the matrix of the self-energy part. We obtain the radiation constant $\gamma_{\mathbf{K}_1\ldots\mathbf{K}_l}$ of this state in the first order of perturbation theory:

$$\gamma_{\mathbf{k}_1\ldots\mathbf{k}_l} = \frac{1}{2\pi\omega_0}\int d\mathbf{K}\delta(K-\omega_0)[d\mathbf{K}]^2 \sum_{\{a_1\ldots a_{l+1}\}} |\langle \Phi_{\mathbf{k}_1\ldots\mathbf{k}_l}|\hat{R}_{\mathbf{k}}^{+}\hat{R}_{a_1}\ldots\hat{R}_{a_{l+1}}\Phi_N\rangle|^2. \tag{14}$$

Calculations outlined in the Appendix show that

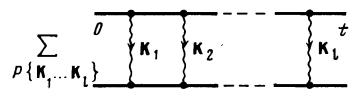
$$\gamma_{\mathbf{k}_1\ldots\mathbf{k}_l} = \int d\mathbf{K}\delta(K-\omega_0)[d\mathbf{K}]^2\frac{(N-l)}{2\pi\omega_0}\left(1+\frac{(2\pi)^3}{v}\sum_{s=1}^{l}\bar{\Delta}(\mathbf{K}-\mathbf{K}_s)\right)$$
$$= (N-l)\left(\gamma + \sum_{s=1}^{l}\gamma(\mathbf{K}_s)\right), \tag{15}$$

where $\gamma(\mathbf{K})$ is the collective radiation constant defined by (5) and (8).

We note that for $\mathbf{K} = \mathbf{K}_{l+1}$ the integrand in (15) coincides, except for a factor $(2\pi)^2/V$, with the squared modulus of the matrix element that corresponds to the $(l+1)$-th interaction point.

Since $\gamma(\mathbf{K}) \sim \gamma(\lambda/L)^2$, collective spontaneous emission begins only after the emission of $(L/\lambda)^2$ photons. This collective emission will have the character of stimulated emission and its angular directionality will be strongly dependent on the geometry of the volume in virtue of the angular function $\gamma(\mathbf{K})$ that was mentioned in Sec. 2.

The probability $W_{\mathbf{K}_1\ldots\mathbf{K}_l}(t)$ that up to the time t the emission of l photons in the directions $\mathbf{K}_1 \ldots \mathbf{K}_l$ ($K_i = \omega_0$) will have occurred can be determined by integrating (11) over the frequencies of these photons. If the width $\Delta\omega$ of the emission spectrum is smaller than L^{-1}, then, in the approximation $\Delta\omega L \ll 1$, by integrating over the photon frequencies we obtain



$$= W_{\mathbf{k}_1\ldots\mathbf{k}_l}(t). \tag{16}$$

Since we know the radiation constants of the propagators between the interaction points, and also the squared moduli of the matrix elements for all the interaction points, by differentiating (16) with respect to time we obtain the following kinetic equation:

$$W_{\mathbf{k}_1\ldots\mathbf{k}_l}(t) = -\gamma_{\mathbf{k}_1\ldots\mathbf{k}_l}W_{\mathbf{k}_1\ldots\mathbf{k}_l}(t) + (N-l+1)$$
$$\times\sum_{i=1}^{l}\left\{\left(\varkappa(\mathbf{K}_i) + \sum_{j\neq i}^{l}\gamma(\mathbf{K}_j)\bar{\delta}(\Omega_i-\Omega_j)\right)W_{\mathbf{k}_1\ldots\mathbf{k}_{i-1}\,\mathbf{k}_{i+1}\ldots\mathbf{k}_l}(t)\right\}, \tag{17}$$

where

$$\gamma(\mathbf{K}_j)\bar{\delta}[\Omega_i-\Omega_j] = \bar{\Delta}(\mathbf{K}_i-\mathbf{K}_j)\frac{(2\pi)^2\omega_0}{v}[d\mathbf{K}_i]^2|_{\mathbf{K}_i=\mathbf{K}_j=\omega_0}. \tag{18}$$

Here $\varkappa(\mathbf{K})$ describes the angular distribution of spontaneous emission from an isolated atom:

$$\varkappa(\mathbf{K}) = \frac{\omega_0}{2\pi}[d\mathbf{K}]^2|_{K=\omega_0}, \qquad \int_{4\pi}\varkappa(\mathbf{K})\,d\Omega = \gamma. \tag{19}$$

From the kinetic equation (17) we derive, in particular, the normalization condition

$$\frac{d}{dt}\sum_{l=0}^{N}W_l(t) = 0, \tag{20}$$

where $W_l(t)$ is the probability that l photons are emitted in any directions:

$$W_l(t) = \frac{1}{l!}\int_{4\pi}d\Omega_1\ldots d\Omega_l W_{\mathbf{k}_1\ldots\mathbf{k}_l}(t). \tag{21}$$

According to (17), a photon is emitted with greater probability in the direction of an already emitted photon, i.e., stimulated emission of photons occurs in "rays" with angular dimensions $\sim\lambda/L$.

The angular distribution of photons after total radiative decay, i.e., for $t \to \infty$, can be obtained by integrating successively with respect to the interaction time points in graph (16). Integrating from right to left, we obtain

$$W_{\mathbf{k}_1\ldots\mathbf{k}_l}(\infty)=\delta_{l,N}\sum_{P\{\mathbf{K}_1\ldots\mathbf{K}_N\}}\left\{\prod_{s=1}^{N}\frac{A_s}{B_s}\right\},$$
$$A_s=\varkappa(\mathbf{K}_s)+\sum_{i=1}^{s-1}\gamma(\mathbf{K}_i)\delta(\Omega_s-\Omega_i),\quad B_s=\gamma+\sum_{i=1}^{s-1}\gamma(\mathbf{K}_i). \tag{22}$$

The dynamics of the emission can be studied considerably more simply in the two limiting cases of a) an isotropic collective radiation constant and b) a maximally anisotropic radiation constant (different from zero for only one direction). The angular dependence of the collective radiation constant is determined by the geometry of the sample and by the angular factor $[\mathbf{dK}]^2$ that is associated with the initial polarization of the atoms. Therefore the isotropic case can be realized, in principle, through the compensation of these two dependences. The maximally anisotropic case occurs for a system whose volume is greatly elongated in one direction.

Taking the collective radiation constant to be isotropic, $\gamma(\mathbf{K})=\Gamma$, we integrate with respect to angles in (17) and obtain the simpler equation

$$\dot{W}_l(t)=-(N-l)(\gamma+l\Gamma)W_l(t)+(N-l+1)(\gamma+(l-1)\Gamma)W_{l-1}(t). \tag{23}$$

When we here take $\Gamma\to\gamma$, which corresponds to Dicke's problem ($L\ll\lambda$), we obtain the decay law that was investigated in[1,9] for a system of small linear dimensions. The same equation will describe the decay law for a greatly elongated sample if Γ is interpreted as an anisotropic collective decay constant. The case of emission in a single direction, which is formally analogous to Dicke's problem, was considered in[6].

Neglecting the dispersion of the distribution $W_l(t)$, we transform the difference equation (23) into a differential equation (compare with[9]):

$$\frac{\partial W(x,t)}{\partial t}=\gamma\frac{\partial}{\partial x}\left[\left\{\frac{N\Gamma}{2\gamma}(1-x^2)+(1+x)\right\}W(x,t)\right] \tag{24}$$

where

$$W(x,t)=W_l(t),\quad x=1-2l/N,\quad -1\leqslant x\leqslant 1.$$

Here x denotes the projection of the classical "energy spin" of the system and characterizes the population of excited states of the atomic system or the relative number of emitted photons.

The solution of (24) for the initial condition $W(x,0)=\delta(x-1)$ is

$$W(x,t)=\delta(x-x(t)), \tag{25}$$

$$x(t)=\left[\frac{\gamma}{N\Gamma}-\left(1+\frac{\gamma}{N\Gamma}\right)\mathrm{th}\left\{\left(\frac{N\Gamma+\gamma}{2}\right)(t-t_0)\right\}\right]. \tag{26}$$

It follows herefrom that both the radiative lifetime and the delay time t_0, which are determined from the initial condition, are of the order $(N\Gamma)^{-1}$. A similar decay law governs the case of highly directional emission.[4-6]

The shape of the spectrum can be determined using a Fourier transformation of the square root of the radiation energy flux:

$$W(\omega)\sim\left|\frac{1}{\sqrt{2\pi}}\int_0^\infty e^{i(\omega-\omega_0)t}[\dot{x}(t)]^{1/2}\,dt\right|^2. \tag{27}$$

With the aid of (26) we obtain the following expression for the normalized spectrum:

$$W(\omega)=\frac{1}{2\pi}\frac{(\gamma+N\Gamma)^2}{4N\Gamma}\left|\int_0^\infty\frac{dt\,e^{i(\omega-\omega_0)t}}{\mathrm{ch}\{{}^1/_2(N\Gamma+\gamma)(t-t_0)\}}\right|^2. \tag{28}$$

The full spectral width $\Delta\omega$ is thus of the order $N\Gamma$, which was obtained by assuming $\Delta\omega\ll L^{-1}$. We may expect that this inequality expresses the limit for the spectral width of collective spontaneous emission. Indeed, the interaction matrix element corresponding to the emission of a photon $\mathbf{K}$ becomes small if the photon momentum is replaced by $\delta K>L^{-1}$. We also note that L/c can be interpreted as the lifetime of radiation from the system and that therefore the dimensions of the system determine the maximal radiation rate.

In order to observe superradiance it is evidently advisable to select dimensions of a system such that L^{-1} exceeds the inhomogeneous broadening and also the Coulomb interaction between the atoms.

Our condition of full inversion at the initial time can be realized when levels associated with a superradiant transition are excited and the uppermost of these levels is populated initially.

APPENDIX: CALCULATION OF THE RADIATION CONSTANT OF THE STATE

The definition (14) of the radiation constant $\gamma_{\mathbf{K}_1\ldots\mathbf{K}_l}$ of the state $\Phi_{\mathbf{k}_1\ldots\mathbf{k}_l}$ can be expanded as follows:

$$\gamma_{\mathbf{k}_1\ldots\mathbf{k}_l}=\frac{1}{2\pi\omega_0}\int d\mathbf{K}\,\delta(K-\omega_0)\,[\mathbf{dK}]^2\times\frac{\langle R^-_{\mathbf{k}_l}\ldots R^-_{\mathbf{k}_1}\Phi_N\,|\,R^+_{\mathbf{K}}R^-_{\mathbf{K}}\,|\,R^-_{\mathbf{k}_l}\ldots R^-_{\mathbf{k}_1}\Phi_N\rangle}{\langle R^-_{\mathbf{k}_l}\ldots R^-_{\mathbf{k}_1}\Phi_N\,|\,R^-_{\mathbf{k}_l}\ldots R^-_{\mathbf{k}_1}\Phi_N\rangle}=\frac{1}{2\pi\omega_0}\int d\mathbf{K}\,\delta(K-\omega_0)\,[\mathbf{dK}]^2\frac{Z_{\mathbf{K}\mathbf{k}_1\ldots\mathbf{k}_l}}{Z_{\mathbf{k}_1\ldots\mathbf{k}_l}}. \tag{29}$$

The quantity $Z_{\mathbf{k}_0\mathbf{k}_1\ldots\mathbf{k}_l}$ (where $\mathbf{K}_0=\mathbf{K}$) in the numerator can easily be put into the form

$$Z_{\mathbf{k}_0\mathbf{k}_1\ldots\mathbf{k}_l}=\sum_{\{a_0\ldots a_l\}}\sum_{P\{a_0\ldots a_l\}}\exp\left(i\sum_{j=0}^{l}\mathbf{K}_j(\mathbf{A}_j-\mathbf{A}_{P_j})\right). \tag{30}$$

We here sum over all sets $a_0\ldots a_l$ of $(l+1)$ atoms and over $(l+1)!$ permutations; $\mathbf{A}_i=\mathbf{r}_a$. It is obvious that $Z_{\mathbf{k}_0\mathbf{k}_1\ldots\mathbf{k}_l}$ is symmetric with respect to interchanges of its arguments. We now distinguish the dependence of $Z_{\mathbf{k}_0\mathbf{k}_1\ldots\mathbf{k}_l}$ on $\mathbf{K}_0$:

$$Z_{\mathbf{k}_0\mathbf{k}_1\ldots\mathbf{k}_l}=\sum_{\{a_1\ldots a_l\}}\sum_{P\{a_1\ldots a_l\}}\left\{\left[\sum_{a_0}\sum_{s=0}^{l}\exp(i(\mathbf{K}_0-\mathbf{K}_s)(\mathbf{A}_0-\mathbf{A}_{P_s}))\right]\times\exp\left(i\sum_{j=1}^{l}\mathbf{K}_j(\mathbf{A}_j-\mathbf{A}_{P_j})\right)\right\}. \tag{31}$$

Calculating the quantity within the square brackets by means of a volume integration, we obtain

$$Z_{\mathbf{k}_0\mathbf{k}_1\ldots\mathbf{k}_l}=(N-l)\left(1+\frac{(2\pi)^3}{v}\sum_{s=1}^{l}\bar{\Delta}(\mathbf{K}_0-\mathbf{K}_s)\right)Z_{\mathbf{k}_1\ldots\mathbf{k}_l}, \tag{32}$$

where

$$\bar{\Delta}(\mathbf{K}_0-\mathbf{K}_s)=\frac{1}{(2\pi)^3v}\int d\mathbf{r}\,d\mathbf{r}'\exp[i(\mathbf{K}_0-\mathbf{K}_s)(\mathbf{r}-\mathbf{r}')]=\frac{(2\pi)^3}{v}|\Delta(\mathbf{K}_0-\mathbf{K}_s)|^2. \tag{33}$$

Substituting (32) in (29), we finally have

$$\gamma_{k_1\ldots k_l}=(N-l)\left(\gamma+\sum_{s=1}^{l}\gamma(\mathbf{K}_s)\right). \tag{34}$$

[1)]We are using a system of units where $\hbar = 1$, $c = 1$.

*$[d\mathbf{K}] \equiv \mathbf{d} \times \mathbf{K}$.

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