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MÖSSBAUER-BORRMANN SUPERRADIANCE

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Résumé - Considérons le comportement cinétique d'un laser pour produire des rayons gamma, construit de noyaux isomériques aux sites de réseau d'un cristal parfait et simple, dont les dimensions et la structure sont choisies pour favoriser l'émission anormale dans le mode de Borrmann qui contient le nombre maximum de réflexions de Bragg. Ceci réduit fortement l'excitation requise pour le lasing. Notre analyse de plusieurs systèmes hypothétiques démontre que la superradiance, plutôt que l'amplification d'émission spontanée, sera le mode d'opération, pour autant que les noyaux isomériques soient pompés très rapidement à un niveau Mossbauer de durée courte et que l'intégrité du cristal soit préservée. Ce résultat justifie la recherche de solution aux problèmes principaux : noyaux candidats, préparation de l'isomère, et transfert du niveau isomérique au niveau lasant.

Abstract - We consider the kinetic behavior of a gamma-ray laser comprising an array of isomeric nuclei located at regular lattice sites in a perfect single crystal of dimensions and structure so chosen as to favor anomalous emission into that Borrmann mode having the maximum possible number of component Bragg-reflected beams, which greatly reduces the excitation requirements. Our analysis of several hypothetical systems shows that superradiance, rather than amplified spontaneous emission, will then be the preferred mode of deexcitation, provided the nuclei can be pumped rapidly to a short-lived Mossbauer level while preserving crystal integrity. This warrants a search for solutions to the major problems : candidate nuclides, preparation of a storage isomer, and interlevel transfer from storage to lasing state.

INTRODUCTION

It is appropriate that a conference on x-ray lasers (i.e., lasers that rely upon exciting *electronic* currents within the atom) take note of the current status of work aimed at achieving lasing at sub-nanometer wavelengths by stimulating the *nuclear* currents of resonant Mossbauer transitions, where the main problem is to provide adequate power for pumping the lasing states without destroying the laser. We have hopes that that difficulty can be overcome by exploiting several phenomena, unique to this problem, that reduce the excitation requirements by many orders of magnitude.

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UNIQUE FEATURES OF NUCLEAR TRANSITIONS

Nuclear transitions cover a much wider range of lifetimes and energies than do inner-shell atomic transitions, and, in contrast with x-ray transitions, which are predominantly electric dipole (E1) in character, magnetic dipole (M1), electric quadrupole (E2) and still higher order transitions are also common. The radiative lifetime of a nuclear state is strongly dependent on the multipole order of the transition by which it decays; "isomers" exist with lifetimes of hours, months or longer.

In solid hosts, nuclear sources emit extremely narrow gamma-ray lines; the Mössbauer effect eliminates Doppler broadening. That single-crystal hosts act as waveguides for x rays, reducing the nonresonant absorption losses (the Borrmann effect) has been known for decades. Less well known is that they are high-quality resonators, selectively enhancing interaction with nuclear transitions of particular multipole order, increasing the gain for a given population inversion, and improving the angular collimation of the output radiation.

KINETICS CONSIDERATIONS

Speculations about the possibility of lasers date back to the earliest days of lasers.[1] Most of the early proposals failed to meet the requirement that the basic conditions for gain must be preserved until lasing can get under way. Proposals that featured amplified spontaneous emission in an elongated body overlooked the peculiar kinetics of a narrow-line system. For example, methods were suggested for eliminating inhomogeneous broadening in long-lived isomers (which have very narrow natural linewidths), so as to increase the gain above the threshold for lasing. Such procedures, however, cannot be completed before inversion has decayed.[1] At the opposite extreme are proposals to use an intense burst of radiation (viz., slow neutrons) to pump a short-lived transition; here, again, inversion would decay before the spontaneous emission is amplified appreciably, unless the gain were very high indeed.[1]

TWO-STEP PUMPING

Today attention is concentrated upon a hybrid system, in which most of the excitation energy is delivered slowly by radiochemical procedures to create a "storage isomer," that is then rapidly transferred to a short-lived state.[2] Besides reducing the pumping energy, this proposal has a major advantage: when population inversion is created suddenly, energy may be released by coherent spontaneous emission ("Dicke superradiance")[3-6] rather than by a (far slower) chain of random stimulated emission events.[6]

RECOILLESS EMISSION

In nuclei bound in cool solid hosts, photon emission (absorption) in transitions between low-lying states may occur without exciting phonons in the solid (Mössbauer effect), so a narrow line of nearly natural width is emitted (absorbed). Two well-known examples are the 14.4-keV gamma ray of ^{57}Fe ; another is the 24-keV Mössbauer transition of 18-ns half-life in the nuclide ^{119}Sn , which is fed from an isomeric level of 293-d half-life. The latter could be a storage level, except that we do not know how or whether transfer can be induced to the Mössbauer level. The transfer operation, the crucial step in such a two-step pumping process, must be rapid, and, to preserve the integrity of the solid host, the energy required must be relatively low compared with the energy of the lasing transition. Conceivably, the effects of crystallinity can sufficiently enhance the interactions of both the gamma rays and the transfer radiation; however, this remains to be established by research. Note that both examples above are of magnetic dipole transitions.

Measured cross sections for recoilless resonant nuclear transitions of submicrosecond lifetimes are comparable with those typical of optical lasers, although their wavelengths are shorter by many orders of magnitude. The linewidths typical of Mössbauer transitions are less than a microvolt; those for x-ray transitions are of the order of a few eV. For example, the resonance cross section for the transition in ^{57}Fe , 5.3 Mb, compares favorably with cross sections for optical laser transitions, and its resonant width is only 16 MHz.

The pumping power required to excite inversion in ^{57}Fe at the threshold for gain, although far less than for an electronic transition of equal energy, is still much too high to preserve the Mössbauer effect after transfer, unless an appropriate nuclide can be found with a small energy difference between storage and transfer levels, that can be pumped by a two-step process.

NUCLEAR SUPERRADIANCE

Consider an assemblage of storage nuclei in a solid host in the form of a long thin cylinder. An unspecified transfer radiation rapidly establishes a state of high gain; then, as in the well studied optical region, [4] superradiance begins to evolve. After a short delay, if gain is not destroyed prematurely, a giant pulse of coherent radiation is emitted along the axis of the cylinder. If above-threshold gain can be established rapidly, while preserving the conditions for Mossbauer and Borrmann effects, the kinetic problem is mitigated.

In amorphous hosts, the gain coefficient is the product of the resonant emission cross section and the inversion density; i.e.,

$$G_0 = n^* \sigma_s = (\gamma^2 f / 2 \pi \Gamma T) (n_2 - wn_1) \quad (1)$$

The net gain is given by G_0 minus the nonresonant absorption coefficient.

Experimental and theoretical studies at MIT have established simple analytical formulas for the main features of strong superradiance: relations among the number of active atoms, the maximum gain-length product, and the delay, width and intensity of the superradiant pulse. [5] At Los Alamos, we have applied these formulas to estimate the performances of hypothetical systems. [6] Table I shows a typical example: ^{57}Fe in a cubic iron host; pulse intensity and width are plotted in Figure 1 as functions of the number of active nuclei. Of course, this nuclide has no storage level. Our purposes are: a) to obtain an order-of-magnitude estimate of the number of storage nuclei that must be transferred to an upper lasing state, and b) to ascertain the relative importance of nuclear and solid-state parameters, as a guide to finding candidates and, ultimately, to designing systems for experimental study.

We find [6] that, even in amorphous hosts, as few as 10^{13} active nuclei in a cylinder of millimeter length and micron diameter should deliver an extremely intense, well collimated pulse carrying a large fraction of the stored energy. The ratio of the energy delivered in the directed beam to that spontaneously radiated is of the order of 10^7 .

THE TWO-BEAM BORRMANN MODE

Moreover, we find that the effects of crystallinity are of crucial importance in gratings, allowing gain to be achieved with inversion densities orders of magnitude less than would be required in an amorphous system.

Monochromatic x-radiation traveling within a perfect single crystal in a direction \underline{k} will be strongly scattered into the direction \underline{k}' if

$$\underline{k} - \underline{k}' = n\underline{q}; \quad (2)$$

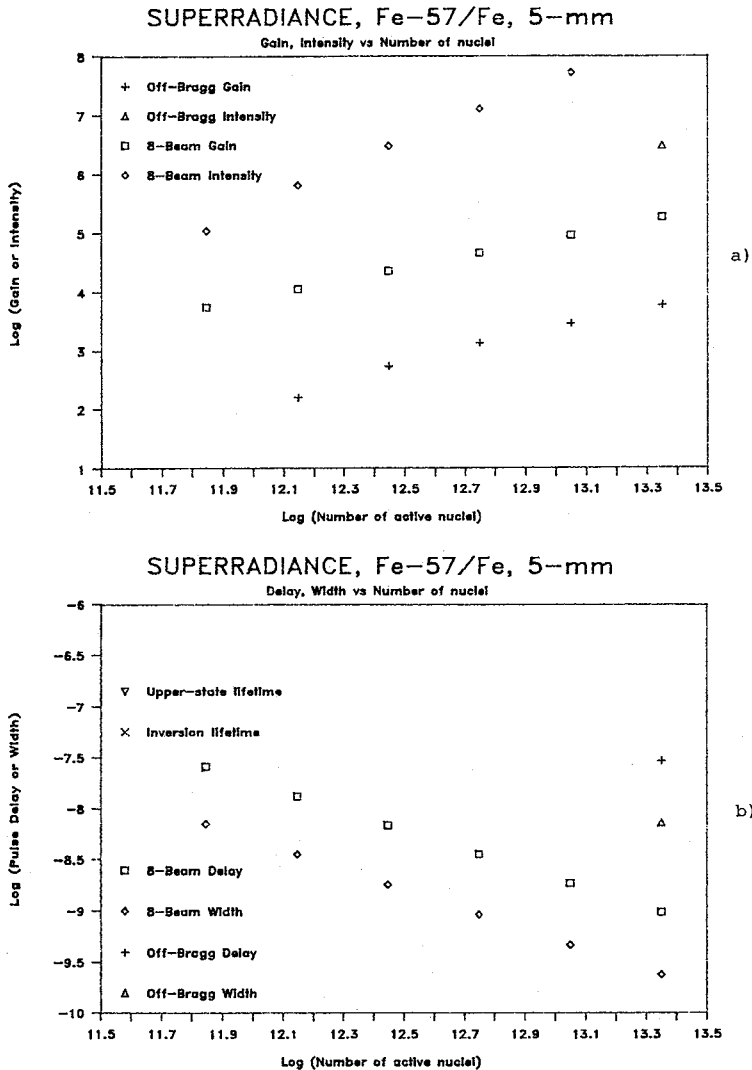


Fig. 1. Estimated behavior of a hypothetical graser system based on the 14.4-keV Mossbauer line of ^{57}Fe in an iron host.

i.e., if the Bragg condition is satisfied for the set of planes within the crystal characterized by the reciprocal lattice vector \underline{g} (See Figure 2). In this case, neither \underline{k} nor \underline{k}' will be eigenmodes of the wave field; rather, the eigenmodes will consist of coherent superpositions of these two waves, with the character of traveling waves in the direction $\underline{k} + \underline{k}'$, and standing waves normal to that direction. Of the two eigenmodes for the case of s-polarized radiation, one (the Borrmann mode) has vanishing electric-field amplitude at each lattice plane, so it does not couple strongly to the atomic electrons and photoelectric absorption in this mode is suppressed. The conjugate ("Anti-Borrmann") mode couples strongly to the electrons and is rapidly attenuated.[7]

Although the *electric* field for the Borrmann mode is zero at the lattice planes, the *magnetic* field and the electric-field *gradients* are not. Thus, while this mode will not couple to electric dipoles (E1) located at regular lattice sites, it will couple strongly to magnetic dipole (M1) and electric quadrupole (E2) transitions in Mossbauer nuclei. Such transitions can feed this mode, generating pairs of highly collimated beams that emerge from the crystal in the Bragg directions. This "anomalous emission" effect, although predicted for the two-beam case a decade ago,[8] still awaits experimental verification.

For Borrmann modes, the gain coefficient (Eq. 1) contains an additional factor, g_N , which measures the coupling of the mode to the nuclear transition. A similar factor, g_a , measures the coupling of the mode to the electrons and appears in the absorption term. Expressions for the coupling factors[9] are given in Table II.

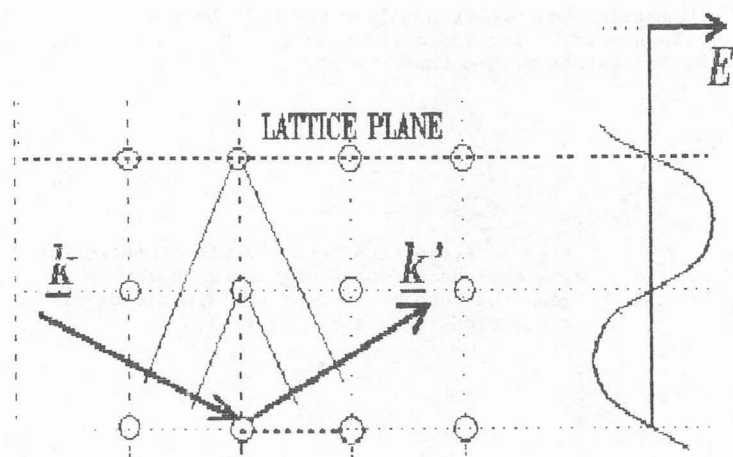


Fig. 2. Two wave Bragg reflection by a set of crystal lattice planes and the variation of electric-field amplitudes with distance from the reflecting planes for the two eigenmodes with s polarization.

MULTIBEAM BORRMANN MODES

It is possible for the Bragg condition to be satisfied simultaneously by several waves in a crystal. As in the two-beam case, radiation-field modes then exist that do not couple to electric dipole transitions, but do couple strongly to transitions of higher multipolarity.[9]

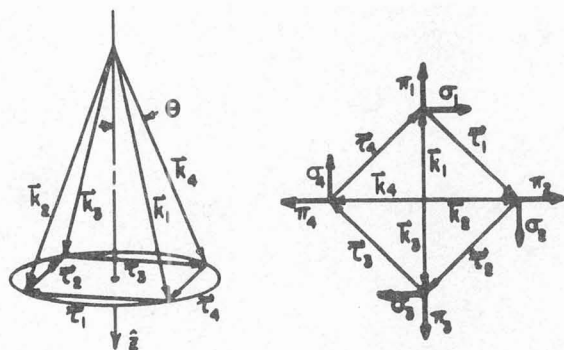


Fig. 3. Wave-propagation, polarization and reciprocal lattice vectors for a four-beam Borrmann mode in a cubic or tetragonal crystal.

A four-beam case which can occur in cubic or tetragonal crystals is shown in Figure 3. The four wave-propagation vectors $\underline{k}_1, \underline{k}_2, \underline{k}_3,$ and \underline{k}_4 lie on the surface of a cone whose base contains the six reciprocal lattice vectors that define the mode. Each wave satisfies the Bragg condition for reflection into each of the other three waves in the mode. Several of the eight eigenmodes in this case are Borrmann modes; i.e., they have zero electric field at the lattice sites. Provided the wave vectors are sufficiently long, modes with 4, 8, 12 and even more beams are possible about fourfold crystal axes, and modes with 3, 6, 12,... beams are possible about three-fold crystal axes.

Figure 4 shows a projection of an eight-beam Borrmann mode involving the 310, 130, $\bar{1}30,$ etc. reflections onto its cone base. All the s-polarizations are excited equally and in phase, so that complete cancellation of the electric field occurs at the lattice sites; the magnetic field, however, is quite large there. This variation of the direction of polarization with azimuth is exactly that of the field emitted by a classical magnetic dipole aligned along the z-axis. This wave mode couples strongly to $M = 0$ magnetic dipole transitions and will be fed automatically by magnetic dipole sources (viz., recoilless nuclear $M1$ transitions) located at or near regular lattice points within the crystal.

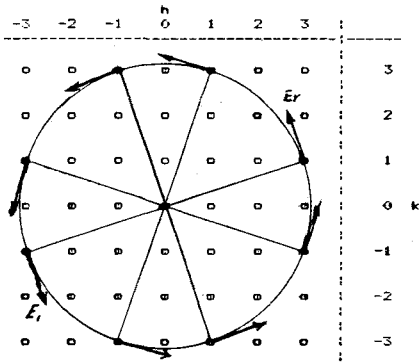


Fig. 4. Projection onto the plane containing the cone base for an eight-beam Borrmann mode that can be excited by magnetic dipole transitions in a cubic crystal.

The coupling factors, $g_n,$ between these modes and the nuclei depend upon both the number of beams and the cone angle (see Table II); the graser body should have its greatest extension in the direction of the mode with the largest value of $g_n;$ either that mode with the largest cone angle or the mode with the most beams (or perhaps both). To compare the relative effects of g_n and $g_a,$ consider the inversion density requirement for a simple example; a steady-state laser containing n active nuclei per unit volume located at lattice sites in a host having negligible nonresonant absorption. Let the inversion density be

$$n^* = n_2 - w n_1 = x n \tag{3}$$

where

$$n = n_2 + n_1. \tag{4}$$

The net gain per unit length is then given by

$$G = g_n \sigma_s n x - g_a \sigma_a n. \tag{5}$$

The threshold for positive gain is

$$x_0 = (g_a/g_n)(\sigma_a/\sigma_s), \tag{6}$$

while the slope of the gain-vs-inversion density curve is

$$dG/dx = g_n \sigma_s n. \quad (7)$$

Both g_n and g_a determine the threshold for positive gain, but, for inversion well above threshold, the nuclear coupling factor, which alone appears in the slope of the gain-vs-inversion relation, will primarily determine the gain.[9]

This is illustrated by Figure 5, which shows the best case found for ^{57}Fe in a cubic Fe lattice--an 8-beam mode with 72-degree cone angle--and Figure 6, which compares gain for this mode and for the off-Bragg case. The nuclear coupling factor in the 8-beam case is 23; the atomic, 2.57. Although this g_a is considerably larger in value than for off-Bragg modes ($g_a = 1$), the enhanced nuclear coupling far outweighs increased photoabsorption. Figure 1 compares the estimated performance for this case with that of the amorphous system, assuming that the same basic relations for superradiance apply in each case. Of course, lacking a storage level, we have no way to pump this transition. These numbers do, however, encourage a search for candidates that can be pumped; they need not have stable lower states.

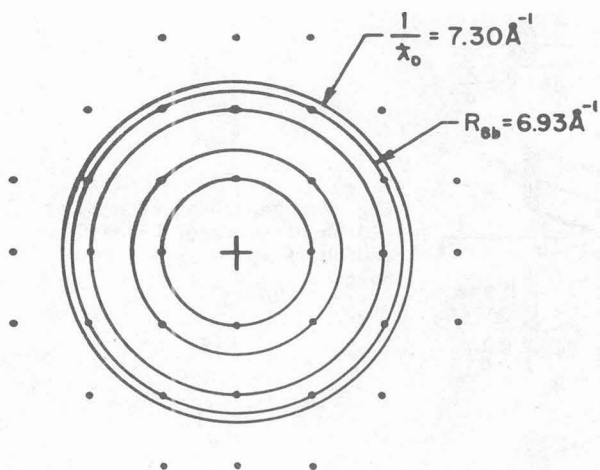
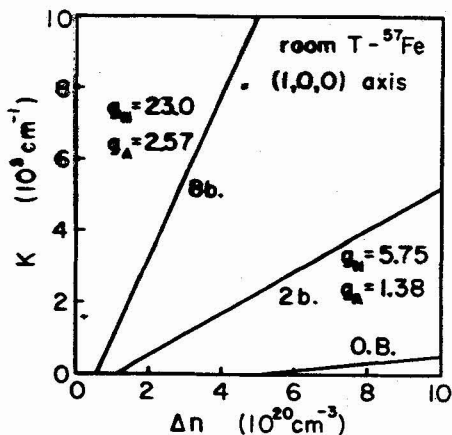


Fig. 5. The eight-beam Borrmann mode for 14.4-keV Mossbauer radiation ($k = 73.0 \text{ nm}^{-1}$) of ^{57}Fe in cubic Fe. The circle of radius 69.3 nm $^{-1}$ passes through eight points of the reciprocal lattice.

A further advantage of multibeam Borrmann modes is illustrated in Figure 7. The angular width of the single output beam from an off-Bragg graser system is determined by the aspect ratio of the host body. In a system that emits into a multibeam Borrmann mode, the emergent radiation is divided among several beams, each having angular divergence determined by the crystal structure (typically 10^{-5} to 10^{-4} rad).

Fig. 6. Gain vs inversion density for ^{57}Fe radiation in Fe, for the 8-beam case of Figure 5, compared with the off-Bragg (amorphous) case.



BORRMANN MODES FOR INTERLEVEL TRANSFER

Considerations of lifetime-vs-multipolarity suggest that the storage and transfer levels will differ by two or more units of angular momentum. We therefore can envisage using Bragg reflections for the transfer radiation to enhance its interaction with the storage nuclei[10] while simultaneously reducing unwanted interactions (assuming that resonant absorption to a real level is the transfer mechanism).

To that end, we propose to incorporate the storage nuclei in the host in the form of a superlattice. It would be helpful for the transfer radiation to propagate at the same direction and velocity (Bragg angle) as as the radiation to be stimulated (Figure 8), but even more beneficial if the superlattice could establish multibeam Borrmann modes for the transfer radiation that couple strongly to the desired nuclear transition, but only weakly to the electronic structure of the host.

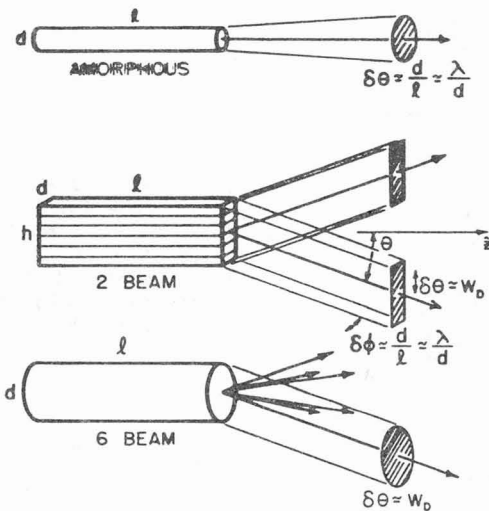


Fig. 7. Comparison of angular widths of emergent beams from amorphous and crystalline hosts.

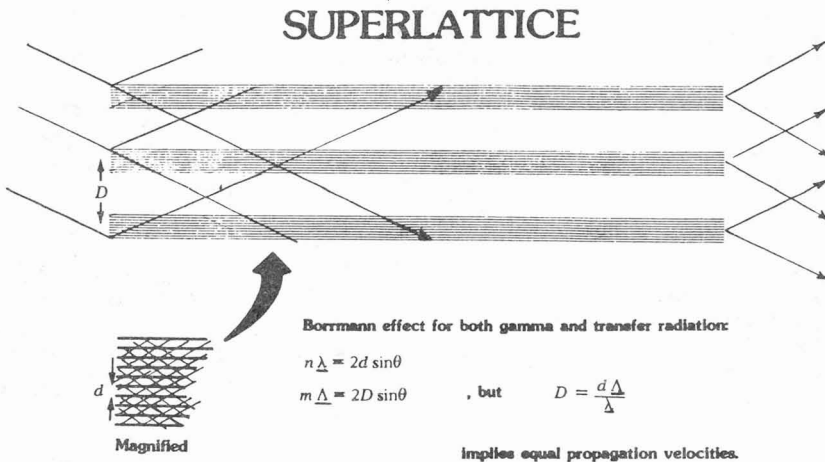


Fig. 8. Concept for a graser body prepared in the form of a superlattice, for which Bragg angles of transfer and stimulated gamma radiation are equal.

T A B L E I

Properties of a hypothetical graser based on ^{57}Fe in a cubic iron host, assuming that an inversion can be established rapidly.

TRANSITION		
Photon energy, keV	14.142	keV
Multipole order	M-1	
Upper-state lifetime	141	ns
Conversion coefficient	8.6	
Nonresonant absorption (Fe)	5.5×10^{-21}	cm^2
HOST		
Debye temperature	470	k
Absorption coefficient	521	cm^{-1}
Fresnel number	1	
Length	0.5	cm
Volume	2.19×10^{-9}	cm^3
8-BEAM BORRMANN MODE		
Lattice constant	2.87×10^{-8}	cm
Cone angle, 8-beam mode	75.05	degrees
Coupling, atomic	1.01	
Coupling, nuclear	27.95	

T A B L E I I

Coupling factors to multibeam Borrmann modes for various multiplicities as functions of cone angle θ and beam multiplicity m .

MULTIPOLE ORDER	COUPLING FACTOR
E-1	$m \langle x^2 \rangle / \lambda^2 (\sin^2 \theta) / \cos \theta$
M-1	$m (\sin^2 \theta) / \cos \theta$
E-2	$m (\sin^2 \theta) / \cos \theta$
M-2	$m (\sin^2 2\theta) / \cos \theta$
E-3	$(5/4) m (\sin^2 2\theta) / \cos \theta$
M-3	$m [(\sin^2 2\theta) / \cos \theta] [15 \cos^4 \theta - 7.5 \cos^2 \theta + 1]$

NOTE: m = Number of beam-pairs feeding the mode*
 θ = Half-angle of the cone containing the associated propagation vectors.

CONCLUDING REMARKS

Conceivably, the pumping power problem for sub-nanometer lasers may be overcome by a combination of features: using nuclear transitions that can be pumped to emit recoillessly by exploiting the strong dependence of lifetime on multipole order, and exploiting multibeam Borrmann effects and anomalous emission in perfect crystals, both for transfer and for superradiant emission.

However, we are not yet in a position to decide whether these measures can suffice to establish feasibility, particularly because low-energy interlevel transfer and anomalous emission have not yet been demonstrated experimentally, nor do we yet have a satisfactory candidate nuclide. These problems involve several distinct disciplines. To that end, it is desirable first to develop a unified analysis of the emission of gamma radiation by sources of arbitrary multipolarity located in real single crystals, including an account of the kinetics of interlevel transfer.

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