Multipole Nature of Elementary Sources of Radiation—Wide-Angle Interference

SIMON FREED AND S. I. WEISSMAN
George Herbert Jones Laboratory, University of Chicago, Chicago, Illinois
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It has been shown possible in wide-angle interference experiments to analyze radiation with respect to its multipole character. The fluorescence from ions of europium in solution consisted of different kinds of radiation in that they possessed different interference properties. From the behavior of the fringe systems, one group of spectral lines was identified as arising from magnetic dipoles while two other groups had their origin in the forced oscillations of electric dipoles. The multipole character of the radiation of a quantum transition was found modified by a change in the microscopic fields about the emitting ions.

TWO beams issuing from a source of light at a wide angle may be brought to give sharp interference fringes provided the dimensions of the source are very small. The observed fringes being the superposition of the fringes from all the elementary centers depend upon the intensities and polarizations of the two diverging rays from each center and these, in turn, are expressions of the structure of the radiation fields characteristic of emitting electric or magnetic dipoles or of poles of higher order. Hence, without prior knowledge of quantum states, one is in position to ascertain the nature of the radiation from the properties of the interference pattern and draw conclusions concerning the possible quantum transitions. Halpern and Doermann[1] have deduced how the visibility of the fringes for each kind of radiation from isotropic point sources varies with the angle of divergence of the beams, with the orientation of polarizing apparatus the beams traverse, and with the experimental arrangement in general.

To illustrate the method and give a rather perspicuous example of some of the factors at work, we shall discuss the wide-angle interference experiment of Selényi[2] which we have modified to a minor degree in the present investigation.

A fluorescent film of fluid in between the plane of a right half-cylinder C and a thin sheet of mica M constitutes the source of radiation. It is excited to fluorescence by light condensed to form a luminous spot on the axis. To obtain distinct fringes without overlapping, that is, to achieve in the fluorescent spot a sufficiently close approximation to the ideal point source, the thickness of the source should be no more than about one-twentieth a wave-length of light, and its diameter about one-twentieth the expected linear separation of the fringes. The separations are greater the thinner the mica, and with mica 0.008 mm thick, the source may be several tenths of a mm in diameter.

The two beams of Fig. 1 diverge from the source 0 at the angle 2θ and are rendered parallel by the total reflection of one of them at the mica-air interface. The fringes originating in the crossing of the pencils of rays at finite angle become visible only after the colors are resolved and viewed in the spectroscope S, the image of the slit for the different colors may be seen crossed with interference fringes. With a solution of fluorescein as the film, a system of fringes appeared having a sinusoidal distribution of in-

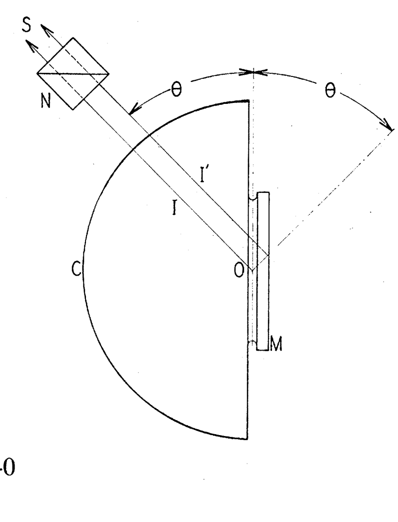


FIG. 1. Schematic representation of apparatus.

[1] O. Halpern and F. W. Doermann, Phys. Rev. **52**, 937 (1937); F. W. Doermann, Phys. Rev. **53**, 420 (1938); F. W. Doermann and O. Halpern, Phys. Rev. **55**, 486 (1939).
[2] P. Selényi, Ann. d. Physik **35**, 444 (1911); Zeits. f. Physik **108**, 401 (1938); Phys. Rev. **56**, 477 (1939).

tensity with black minima when the angle of divergence 2θ was 90° and the Nicol prism N was set to transmit light with electric vector perpendicular to the plane of the two beams I, I'. After the Nicol prism had been turned through ninety degrees, the image of the slit was uniformly illuminated—i.e., the fringes were now absent. This behavior of the light led Selényi to conclude that the fluorescence centers were electric dipoles.

His argument follows: Let it be assumed that the isotropic source of light may be properly represented as the superposition of light from three mutually perpendicular sets of independent oscillating electric dipoles. Consider the radiation from a set of dipoles whose axes are at right angles to the plane of the beams I, I' and have the Nicol prism oriented so that the only radiation transmitted has its electric vector also perpendicular to the plane I, I'. Not only will the direct beam I radiated by this set of dipoles traverse the Nicol prism but also the beam diverging from it at 90° and reflected at the mica-air interface, now the beam I'. The intensities of the two interfering beams will be nearly equal[3] and hence the resulting pattern of fringes may be expected to be completely sinusoidal in its intensity distribution. The two remaining sets of oscillating electric dipoles with axes in the plane I, I' may be imagined so disposed that one set has its axes along the direct beam I and the other at right angles to I. The dipoles along I will not radiate in that direction nor will those at right angles radiate along I' and hence no fringes are to be observed when the Nicol prism is turned to transmit this radiation.

The same result is arrived at by Halpern and Doermann, taking account of the phase relations of the rays from the dipoles in all random orientations. They demonstrate, however, that the identification made by Selényi is not unique. Rays propagated by magnetic quadrupoles at this particular angle of divergence (90°) furnish a fringe system which responds to the orientations of the polarizing prism in the same way as the fringes from electric dipoles. At another angle of divergence, particularly if the choice is 45°, the two kinds of radiation may be distinguished.

[3] The indices of refraction of the glass, mica, and film differ but little and the reflection on the mica surface may be taken as ideal.

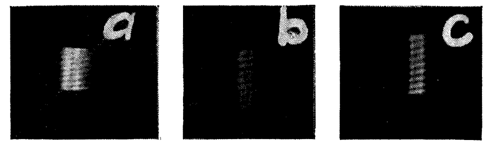


FIG. 2. Wide angle interference fluorescence of the salcylaldehyde compound of europium at 6100A. (a) $\theta=22.5°$, $\chi/2=0°$; (b) $\theta=45°$, unpolarized; (c) $\theta=45°$, $\chi/2=90°$. $\chi/2=90°$ when the electric vector of the polarizing prism is perpendicular to the plane I, I'.

The visibility of the fringes from magnetic quadrupoles is now reduced to zero while that from electric dipoles is scarcely altered from the value it had at the divergence 90°.

We confirmed Selényi's results with a film of fluorescein and examined the fringes at an angle of divergence of 45° also but noted little change in the visibility. The radiation emanates, then, from electric dipoles as Selényi had supposed.

Our spectrograph, with its three prisms, strongly polarized any light entering it, and, as a result, there was but little radiation falling on the photographic plate at one of the required orientations of the Nicol prism N. In order to maintain constant the polarization of the light entering the spectrograph, a quarter-wave plate was attached with a drop of glycerin to the Nicol prism (actually a Glan-Thomson prism) so that the beam was rendered circularly polarized. Another quarter-wave plate was fixed before the slit of the spectrograph at such an orientation that the plane polarized beam issuing from this plate was at the angle of maximum transmission of the spectrograph. The direction of this plane was independent of the orientations of the Glan-Thomson prism.

The ions of the rare earths might be expected to afford opportunity for the study of various kinds of multipole radiation. In the optical transitions, all the electrons contributing to the quantum character of the states remain within the same shell ($4f$) and therefore electric dipole radiation is "forbidden" by the Laporte rule, which is valid for ions in the gaseous state, or, in general, when the environment of the ions possesses a center of inversion. Furthermore, the intensity of the fluorescent radiation is very weak compared with the usual electric dipole radiation from gases or from dyestuffs such as fluorescein. Such feeble intensities are to be expected in radiation from magnetic dipoles, from electric or magnetic quadrupoles, etc.

The absorption and fluorescent spectrum of the ions of europium in crystals and in solution have been analyzed in terms of quantum states and the symmetries of the electric fields about the ions.[4] The effective spectral region for excitation lies between 3500A and 4000A. The fluorescence is unpolarized and may be regarded as emerging from an isotropic source. One group at about 6100A corresponds to a transition from $J=0$ in the upper state to $J=2$ of the basic multiplet $F_{0123456}$. Another group at 5880A originates in the same upper level $J=0$ and ends in the $J=1$ level of the basic multiplet, and a line at 5790A corresponds to the $0\rightarrow 0$ transition.

Europium nitrate in about $2M$ solution of glycerin, not entirely free of water, yielded two groups of lines in fluorescence of sufficient intensity when excited by light from a carbon arc carrying fifty to sixty amperes direct current. The lines were rather broad and it was not found possible to resolve the components of the group representing the decomposition of the basic state by the electrical fields of the environment of the ions. When the observations were made at $\theta=45°$ and the Glan-Thomson prism was set to transmit light with its electric vector perpendicular to the plane I, I', the 6100A group, $J=0\rightarrow J=2$, was crossed with sharp fringes, while the 5880A group, $J=0\rightarrow J=1$, was of uniform intensity along the image of the slit. When the polarizing prism was rotated through 90° the situation was reversed, the 6100A group became uniform in its intensity whereas the 5880A group was crossed with fringes. With θ changed to 22.5°, both groups showed distinct fringes at both orientations of the prism. According to theory, the $0\rightarrow 2$ transition gave rise to electric dipole radiation and the $0\rightarrow 1$ transition to magnetic dipole radiation. The $0\rightarrow 1$ transition was about one-fifth as intense as the $0\rightarrow 2$ transition from this solution of europium. Unfortunately, the $0\rightarrow 0$ transition at 5790A, forbidden completely in a spherically symmetrical environment, was too feeble for these measurements.

However a $0.2M$ solution in benzene of the ammonia-salicylaldehyde compound[5] of europium furnished this line with sufficient intensity (Fig. 2). The other groups also appeared with greater intensity than from the nitrate. The $0\rightarrow 0$ line proved to be electric dipole radiation. We also confirmed the nature of both the $0\rightarrow 2$ and the $0\rightarrow 1$ transitions. However, despite the fact that the fluorescent spectrum from this compound was much sharper and the structure within the groups was resolved, the radiations of the $0\rightarrow 1$ and $0\rightarrow 2$ groups were not as pure in their multipole character as from the nitrate. Especially was this true in the $0\rightarrow 1$ transition where what had appeared as magnetic dipole radiation was now definitely mixed with another species. A change in the environment of the ion had brought about a change in the species of the radiation.

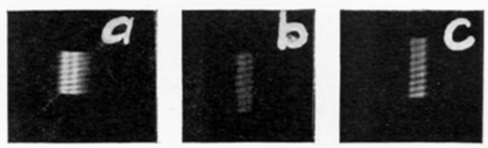
The kinds of radiation revealed in these experiments are in accord with their feeble intensities. The $0\rightarrow 0$ and $0\rightarrow 2$ transitions are excluded as electric dipole radiation in fields of spherical symmetry and only the presence of fields of low symmetry about the ions in solution[6] can "force" the appearance of such radiation, and then with weak intensities. The spherical rotational symmetry must be reduced to C_3 or less before the $0\rightarrow 0$ transition can take place and to tetragonal or less for the $2\rightarrow 0$ transition. Both also require the absence of a center of inversion. The $1\rightarrow 0$ transition is not excluded as magnetic dipole radiation in spherically symmetrical fields but the transition probability is very low. Fields of low symmetry may increase the feeble intensity but it would still be of an entirely different order of magnitude from the strong radiation emitted in spontaneous electric dipole transitions.

We are much indebted to the Penrose Fund of the American Philosophical Society for the support it has given this investigation. We also wish to acknowledge the assistance of Works Projects Administration Official Project 665-54-3-387.

[4] H. Gobrecht, Ann. d. Physik **28**, 670 (1937); S. Freed and S. I. Weissman, J. Chem. Phys. **8**, 878 (1940).

[5] Prepared in the same way as the acetylacetonate of reference 6(b) but was found on analysis to have some ammonia attached to it.

[6] (a) S. Freed and S. I. Weissman, J. Chem. Phys. **6**, 297 (1938); (b) S. Freed, S. I. Weissman and F. E. Fortess, J. Am. Chem. Soc. **63**, 1079 (1941).



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