Superradiant Behavior of Cr\textsuperscript{3+} ions in Ruby Revealed by Whispering Gallery Modes

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We present evidence for collective action of Cr\textsuperscript{3+} ion impurities in a highly doped ruby crystal at microwave frequencies. The cylindrical geometry of the crystal allows for the creation of a superradiant, or “spin-mode” doublet, with spatial structure similar to that of microwave whispering gallery modes (WGMs). This results in a strict criteria of selection rules regarding the interaction of resonant WGMs and spin-modes; namely that only modes with the same wavenumber and azimuthal phase may interact. What results is an avoided level crossing between the two, in which both WGM doublet constituents are seen to interact with the spin resonance. We demonstrate that a four harmonic oscillator model is necessary to accurately describe this result.

Superradiance is an important phenomenon in quantum optics as often the sample under study features separation distances small compared to the wavelength of exciting radiation, $\lambda$. There is renewed interest in these systems for quantum information sciences and to attain new insights into quantum field-theory [1, 2]. Superradiance was initially defined in 1954 by Robert Dicke as the cooperative, spontaneous emission of photons from a collection of atoms [3]. When $N$ atoms are close together compared with $\lambda$, they act like one big atom and decay collectively, in phase with one another. As a result, the atoms radiate their energy $N$ times faster than for incoherent emission. A direct result is the inherent directionality associated with the emitted radiation; the emitted photons travel in the same direction as the exciting photons. This directionality is a result of the timing of the excitations; the atoms at the “front” of the sample are excited first, and those at the back, last, leading to the excitations appearing as spatial phase factors [4]. Superradiance is a consequence of extra coherence in the system, which can be observed in additional ways on top of an increased emission rate.

Superradiance was first observed experimentally in 1973 in the optical regime in HF Gas [5]. It has since been observed in other ultracold atomic gases [6–9], organic semiconductors [10, 11], polymer thin films [12], numerous crystalline systems [13–15], and in artificial atoms [16–18]. Here, we report the observation of superradiance in the microwave regime in a highly doped ruby sample, with relatively high concentrations of Cr\textsuperscript{3+} ions replacing Al\textsuperscript{3+} ions in the crystal lattice.

It is a well known phenomenon that resonant photonic whispering gallery modes (WGMs) in a circular cavity are the result of a linear superposition of clockwise and counter-clockwise circularly polarised waves. Imperfections within a cavity can result in backscattering effects that cause the degeneracy between the two waves to be lifted, by introducing a coupling between them. A WGM will therefore manifest as two orthogonal modes (or doublets) with a difference of Sine and Cosine in the mode’s azimuthal dependence in it’s analytical expression (i.e. a difference of $\pi/2$ in azimuthal phase), henceforth referred to as the “s” and “c” modes. Despite the orthogonality, the coupling produced by these imperfections in the crystal symmetry results in the mode appearing as a doublet. This manifests as a splitting of a single resonant peak into two resonant peaks by a distance equal to two times the coupling value, $\kappa$. In sapphire crystals, the losses of such WGMs are so low, that the bandwidth of these modes is generally less than $2\kappa$ hence the doublet resonance can be resolved.

The Hamiltonian describing such a WGM doublet resonance is

$$H_0 = \sum_k \omega_k \left( a_{k,s}^\dagger a_{k,s} + a_{k,c}^\dagger a_{k,c} \right) + \sum_k \kappa_k \left( a_{k,s} a_{k,c}^\dagger + a_{k,c} a_{k,s}^\dagger \right).$$

Here $\omega_k$ is the angular frequency of a WGM with wavenumber $k$, and $a_{k,s}$, $a_{k,s}^\dagger$, $a_{k,c}$, and $a_{k,c}^\dagger$ are the bosonic raising and lowering operators of the “s” and “c” doublet constituents of this WGM, respectively. The first term in equation (1) represents both modes as simple harmonic oscillators (SHOs), while the second term represents the coupling between them, which produces the mode-splitting and doublet appearance.

A crystal containing dilute concentrations of paramagnetic ion impurities will demonstrate an absorption of energy from these WGMs into the spin angular momentum of the ion’s valence electrons if the frequency of the latter transition is tuned (via the Zeeman effect) to be coincident with that of the former. Only WGMs with magnetic field components perpendicular to the applied DC magnetic field will interact in this fashion. This limits the discussion to WGMs that are polarised with a $\left( H_r, H_\phi, E_z \right)$ field distribution (“WGH” modes), since the applied magnetic field in the described case is aligned with the z-axis of the crystal.

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emitting photons, hereon in referred to as the “spin-mode”, is completely analogous to a photonic WGM, and therefore can exist as a doublet due to backscatterers, in exactly the same way. It will also display the same type of wavenumber orthogonality, doublet orthogonality and a coupling between the two doublet constituents. Spin doublet modes have been previously observed in ferromagnetic YIG samples [20], but never before in a doped sapphire system. In such a scenario, the Hamiltonian describing the interaction between the photonic cavity WGM doublet and the spin-mode doublet would appear as

$$H = H_0 + \sum_k \omega_k \left( \sigma^+_{k,s} \sigma^-_{k,s} + \sigma^+_{k,c} \sigma^-_{k,c} \right) +\sum_k \chi_k \left( \sigma^+_{k,s} \sigma^-_{k,c} + \sigma^+_{k,c} \sigma^-_{k,s} \right),$$

where $\chi$ represents the coupling between the two spin-mode doublet constituents; “$s$” and “$c$”. This Hamiltonian is derived following the treatment of Dicke [3] when describing radiation from a gas of large extent. From eq. (2); a summation over all modes and TLSs, a transition is made to just the former. The selection rules of such a system [3] dictate that only modes with equal wavenumbers, $k$, may interact. In addition to this, the equivalent doublet orthogonality of the spin-modes and WGMs allow for both “$s$”–“$s$” and “$c$”–“$c$” spin-WGM interactions, but not “$s$”–“$c$”. This removes the requirement that one of the spin-mode couplings be set to zero. It is reasonable to assume that the coupling strengths of the two “$s$” modes will be equal to the two “$c$” modes, hence the use of a non-polarisation-specific coupling term, $g_k$. As such, the allowed spin-WGM interactions are described by the third term in eq. (3).

The second expression in (3) represents the spin-mode doublets as two SHOs, while the final term represents the coupling between them, resulting from imperfections in the crystal; a direct analogue of the last term in eq. (1).

Equation (3) is represented diagrammatically in FIG. 1 for a single value of $k$. It describes a scenario of four SHOs with the allowed linear couplings. This is distinctly different from the case described by eq. (2), which would exists as three SHOs [19].

The experimental set up is identical to that described by Farr et al. [21], however we examine WGMs closer to the zero-field splitting levels of the Cr$^{3+}$ ensemble. Typical ESR parameters for Cr$^{3+}$ ions can be find in [22]. In this paper, we deal with the $\Delta m = \pm 1$ transitions; $|−3/2⟩ → |−1/2⟩$ and $|3/2⟩ → |1/2⟩$. Both these transitions have a zero-field frequency of 11.447 GHz, and tune in opposite directions as B field is swept ($\Delta m = \pm 1$ increases in
frequency with an increase in B field, and vice versa) with \( df/dB = \pm g_L \beta \), where \( g_L \) is the Landé g-factor and \( \beta \) is the Bohr magneton.

FIG. 2: Spectroscopy results showing the interaction of the generated and pump WGMs with the \(|+3/2\rangle \rightarrow |+1/2\rangle\) \( \text{Cr}^{3+} \) electron spin transition as magnetic field is swept.

For example, FIG. 2 shows the \( \Delta m = -1 \) transition (in red), as it moves through five distinct WGMs (in black). Each of the data points that make up the black curves represent the position of a resonant peak within a single 8 MHz sweep centred around that particular frequency for that particular magnetic field value. The power incident on the crystal is \( P_{inc} = -60 \) dBm, which corresponds to a photon occupation number on the order of \( 10^7 \). Far from the intersection of the WGMs and the ESR transition, the black curves in FIG. 2 represent the frequency location of the WGMs, however when the ESR is tuned such that a particular WGM is within its bandwidth, the black curves represent hybrid spin-WGMs, and an avoided level crossing (ALC) can be observed, as depicted in the inset figures of FIG. 2.

Equation (2) predicts a gyrotropic response for the ALC of a WGM doublet and ESR, which may be modelled with great accuracy by three SHOs [19]. The ESR spectroscopy results for the ruby crystal in question (FIG. 3 and 4) clearly show an absence of this gyrotropic response. We observe that both components of the WGM doublet interact with the spin transition.

As FIG. 3 demonstrates, there are four asymptotes to which the hybrid modes converge. The two horizontal asymptotes of FIG. 3 are a standard result of the WGM in question \( (f = 9.55 \text{ GHz}) \) existing as a doublet. The vertical asymptotes, which in fact depend on B-field just as the red curve in FIG. 2 (observable if the y-axis scale were broader), confirm the presence of a spin-mode doublet. Their separation is \( 2\chi \). In the general paramagnetic case of equation (2), there would be only one vertical asymptote [19].

The presence of these four asymptotes requires a four SHO model (FIG. 1) to fit the experimental data. Using values of \( g = 67 \) MHz, \( \kappa = 0.43 \) MHz and \( \chi = 76 \) MHz, a fit is produced which is displayed in FIG. 3. To produce good agreement with the model, it is essential that the cross coupling terms \( g_\alpha \) be neglected, or at least be much smaller than the spin-mode couplings \( g \) – consistent with the allowed terms in the third expression in eq. (3).

The requirement for spatial orthogonality of the spin-modes is again confirmed by FIG. 4. Here, we see the same type of ALC as in FIG. 3. However, we also observe the tail end of another ALC originating at a slightly higher frequency enter the frame. It is the hybrid mode of a higher frequency spin-mode and WGM of a different order. As predicted by the selection rules of such a system, these two doublets simply merge; there is no interaction, due to their different wavenumbers.

As WGMs hybridise with a spin-mode (or paramagnetic spin ensembles, for that matter), not only is a frequency shift observable due to the altered magnetic susceptibil-
of the unit cell is that of a trigonal crystal system, the number of impurity ions in the crystal, \( N_t \), where
\[
N_t = \frac{V}{a^3} N_+ \tag{10}
\]
and \( V \) is the total volume of the ruby crystal, one can solve for \( N_T = 5.42 \times 10^{18} \) ions.

Given that there are two \( \text{Al}^{3+} \) ions per unit cell of sapphire that \( \text{Cr}^{3+} \) can potentially replace, and the volume of the unit cell is that of a trigonal crystal system, the total concentration of ion impurities can be calculated as \( N_T \) divided by the total number of potential lattice cites for \( \text{Cr}^{3+} \) ions to take. A concentration of approximately 40 ppm \( \text{Cr}^{3+} \) is calculated. This agrees very well with previously measured values for this same crystal, reported as 34 ppm \([21, 22]\), hence confirming the validity of the four SHO approximation used here, and ergo the conclusions that can be drawn from it.

This concentration is approximately two orders of magnitude larger than the concentration of \( \text{Fe}^{3+} \) impurities in \([19]\) (150 ppb), and the value of \( g \) is also approximately an order of magnitude larger. In addition, the losses associated with the \( \text{Cr}^{3+} \) ESR (\( \Delta \omega = 9 \text{ MHz} \)) are three times less than those associated with the \( \text{Fe}^{3+} \) case (\( \Delta \omega = 27 \text{ MHz} \)) \([19, 23]\). In the present case, \( g > \Delta \omega \), satisfying the conditions for strong coupling, however this is unobservable due to the combined losses of the WGM and ESR, and the non-unity coupling between the microwave pump source and WGM. Large atom-field coupling is required for superradiance as it is this interaction from which the collective action of the ensemble is derived, and large ion concentrations reduce the separation between emitters. This explains why a superradiant ESR, and hence a four SHO model, is observed in the \( \text{Cr}^{3+} \) case and not in the previously reported \( \text{Fe}^{3+} \) case.

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[1] A. Svidzinsky and J.-T. Chang, Phys. Rev. A 77, 043833 (2008).
[2] S. Haroche and J. M. Raimond, Exploring the Quantum: Atoms, Cavities, and Photons (Oxford Univ. Press, Oxford, 2006).
[3] R. H. Dicke, Physical Review 93, 99 (1954).
[4] M. O. Scully and A. A. Svidzinsky, Science 325, 1510 (2009).
[5] N. Skribanowitz, I. P. Herman, J. C. MacGillivray, and M. S. Felt, Phys. Rev. Lett. 30, 309 (1973).
[6] H. Xia, A. A. Svidzinsky, L. Yuan, C. Lu, S. Suckewer, and M. O. Scully, Phys. Rev. Lett. 109, 093604 (2012).
[7] L. E. Sadler, J. M. Higbie, S. R. Leslie, M. Vengalattore, and D. M. Stamper-Kurn, Phys. Rev. Lett. 98, 110401 (2007).
[8] A. Crubellier, S. Liberman, and P. Pillet, Phys. Rev. Lett. 141, 1237 (1978).
[9] M. Gross, C. Fabre, P. Pillet, and S. Haroche, Phys. Rev. Lett. 36, 1035 (1976).
[10] F. Meinardi, M. Cerminara, A. Sassella, R. Bonifacio, and R. Tubino, Phys. Rev. Lett. 91, 247401 (2003).
[11] S.-H. Lim, T. G. Bjorklund, F. C. Spano, and C. J. Bardeen, Phys. Rev. Lett. 92, 107402 (2004).
[12] S. V. Frolov, W. Gellerlmann, M. Ozaki, K. Yoshino, and Z. V. Vardeny, Phys. Rev. Lett. 78, 729 (1997).
[13] C. Greiner, B. Boggs, and T. W. Mossberg, Phys. Rev. Lett. 85, 3793 (2000).
[14] S. Kondo, H. Nakagawa, T. Saito, and H. Asada, Current Applied Physics 4, 439 (2004).
[15] S. Kondo, K. Suzuki, T. Saito, H. Asada, and H. Nakagawa, Phys. Rev. B 70, 205322 (2004).
[16] A. Barenco, D. Deutsch, A. Ekert, and R. Jozsa, Phys. Rev. Lett. 74, 4083 (1995).
[17] M. Bayer, P. Hawrylak, K. Hinzer, S. Fafard, M. Korkusinski, Z. R. Wasilewski, O. Stern, and A. Forchel, Science 291, 451 (2001).
[18] J. A. Mlynek, A. A. Abdumalikov, C. Eichler, and A. Wallraff, Nat Commun 5 (2014).
[19] M. Goryachev, W. G. Farr, D. L. Creedon, and M. E. Tobar, Phys. Rev. B 89, 224407 (2014).
[20] M. Goryachev, W. G. Farr, D. L. Creedon, Y. Fan, M. Kostylev, and M. E. Tobar, Phys. Rev. Applied 2, 054002 (2014).

[21] W. G. Farr, M. Goryachev, D. L. Creedon, and M. E. Tobar, Phys. Rev. B 90, 054409 (2014).
[22] W. G. Farr, D. L. Creedon, M. Goryachev, K. Benmessai, and M. E. Tobar, Phys. Rev. B 88, 224426 (2013).
[23] J. Bourhill, K. Benmessai, M. Goryachev, D. L. Creedon, W. Farr, and M. E. Tobar, Phys. Rev. B 88, 235104 (2013).