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A proof-of-principle experiment of EIT with gamma radiation in $FePSe_3$ single crystal

Hisakazu Muramatsu · S. Nakajo · K. Nakagami · K. Nagata · S. Mochizuki · H. Suzuki

Abstract From a series of Mössbauer measurements performed on a single crystal of $FePSe_3$, we have observed the effect of electromagnetically induced transparency (EIT) in a second nuclear system, after a first experiment using the mineral siderite $FeCO_3$. When the condition of nuclear level crossing was fulfilled, an obvious deficit in absorption was observed at the point of level crossing. This reduced absorption cannot be ascribed to saturation effects because of the specific geometry of the measurement. We conclude that the observed deficit in absorption is due to coherence and interference effects.

Keywords EIT · Electromagnetically induced transparency · Mössbauer effect · Level-crossing

1 Introduction

Electromagnetically induced transparency (EIT) has been demonstrated under several experimental conditions: in continuous wave and pulsed regimes for different wavelengths raging from optics to microwaves [1–8]. Interesting proposals have been discussed so far to obtain lasing for gamma rays by utilizing coherent effects, and the EIT effect via the level-crossing technique at the single-photon level has been theoretically predicted [9–12]. Some features of quantum optics related to inversion less amplification are translated to nuclear systems, but there are several differences between the nuclear and atomic systems (shorter wavelength, weaker

H. Muramatsu (✉) · S. Mochizuki · H. Suzuki
Department of Chemistry, Faculty of Education, Shinshu University, Nagano-shi, Nagano 380-8544, Japan
e-mail: hmurama@shinshu-u.ac.jp

S. Nakajo · K. Nakagami · K. Nagata
Graduate School of Education, Shinshu University, Nagano-shi, Nagano 380-8544, Japan

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nuclear transitions, no coherent gamma-ray sources). Although there are some difficulties connected with nuclear systems, some ideas have been proposed that could be translated to nuclear radiation [13, 14], and they show that nuclear systems offer some advantages compared to the atomic ones because of the possibility of controlled nuclear level mixing phenomena.

We have already reported the first steps towards a proof-of-principle experiment, demonstrating EIT with gamma radiation for the Mössbauer effect in the mineral siderite $FeCO_3$ using the level-crossing technique [15–17]. We have tried to seek other candidates for absorbers possible to use in the same type of experiment. Recently, we have successfully synthesized small flakes of single crystal of $FePSe_3$. In the present paper we will report the second proof-of-principle experiment demonstrating EIT for the Mössbauer effect by using a single crystal of $FePSe_3$. Our attention will be focused on the problem whether $FePSe_3$ can be an appropriate absorber for EIT experiments or not.

2 Experimental

$FePSe_3$ was synthesized by solid phase reaction in a vacuum-shielded tube (Vycol glass tube) containing a mixture of stoichiometric amounts of high-purity elements, for 1 month. The reaction temperature to synthesize was set to be 840°, and the cooling rate was 40 degree/day. Single crystals of $FePSe_3$, shaped like thin foils, were obtained with a dimension of 3 mm × 3 mm. Since the direction of the hyperfine magnetic field is parallel to the principal axis of the electric field gradient (EFG), the *c* axis, all measurements were made with the gamma-ray parallel to the *c* axis, i.e. the direction perpendicular to the ab-plane. The single crystal sample was sandwiched in between two lead collimators with a window of 2 mm in diameter and mounted on a copper sample holder in a cryostat connected to a small He gas filled refrigerator (Cryo-Mini, Iwatani Plantec Co.) in such a way that the ab-plane faces the direction of the gamma-rays at a right angle. Mössbauer spectra were recorded in transmission geometry in the temperature range of 300 K to 16 K, using a single-line source of ^{57}Co (10 mCi) in a rhodium matrix and a conventional constant acceleration transducer. The calibration of the velocity scale for the spectrometer was carried out using an α-Fe foil.

3 Results and discussion

A strong temperature-dependent internal magnetic field parallel to the EFG-axis in the single crystal allows for crossings in the nuclear level structure of ^{57}Fe. Besides the magnetic hyperfine field, the Fe^{2+} nucleus in the crystal is subjected to a large axially symmetric EFG, which results in a well-resolved quadrupole doublet. (See Fig. 1a) Below the Néel temperature (106 K), Mössbauer spectra consist generally of six absorption lines due to its antiferromagnetic behavior. When dealing with a single crystal, however, depending on the orientation of the incident gamma ray with respect to the crystal c-axis, different selection rules apply. The source emits a statistical mixture of left- and right-circularly polarized photons, each allowing for a specific, angle-dependent, Δm transition. In case of the gamma photon being incident

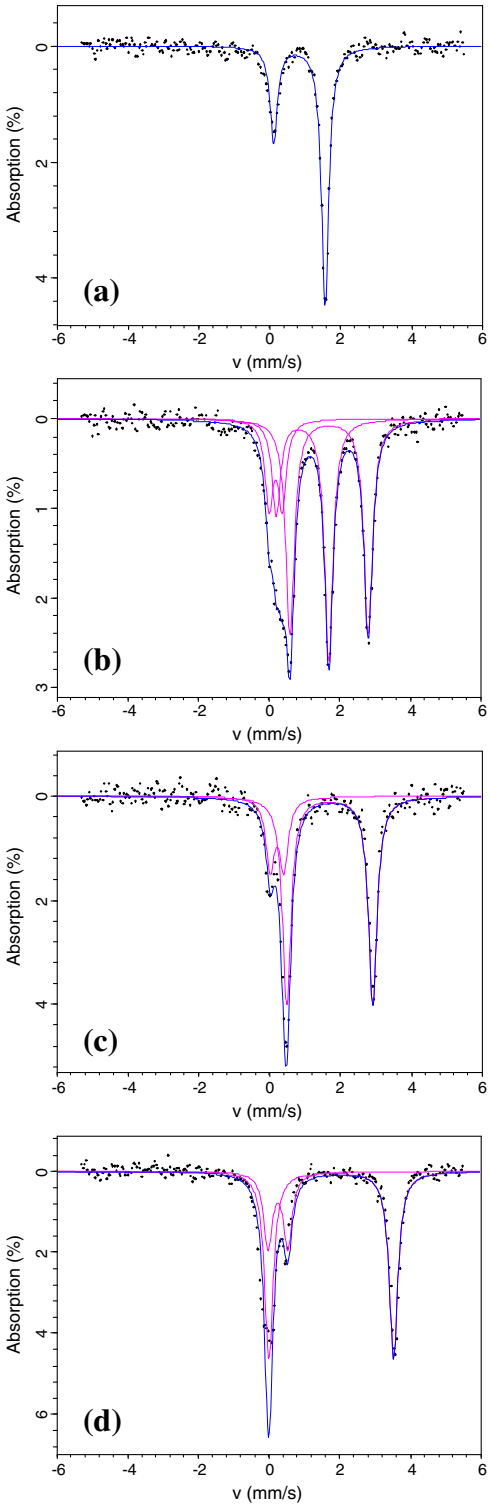

Fig. 1 Mössbauer spectra taken with $FePSe_3$ single crystal at various temperatures. **a** R.T., **b** 106 K, **c** 105 K, **d** 31 K

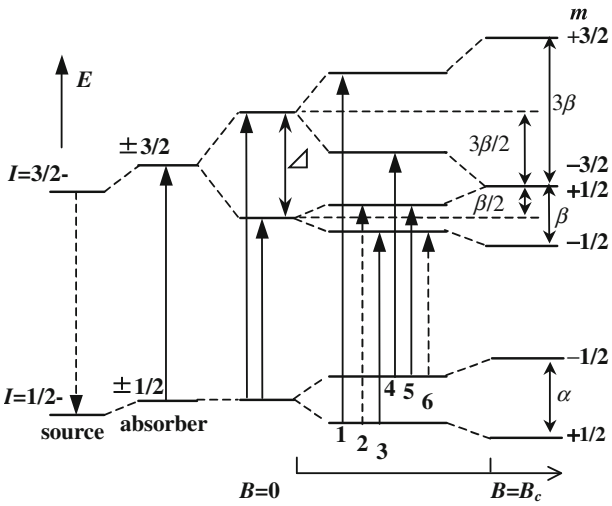

Fig. 2 Energy levels of the system as a function of the magnetic hyperfine field, B_{hf}

perpendicular to the crystal c-axis ($\theta = \pi/2$), each photon can coherently excite three transitions, and then six Mössbauer lines can be observed. On the other hand, when the gamma photon is incident along the crystal c-axis ($\theta = 0$), each photon can only excite one transition. In total, only four lines can be observed. We call this orientation the '*parallel geometry*' hereafter. In the actual measurements, since Mössbauer spectra of the $FePSe_3$ single crystal were taken in parallel geometry, the observed spectra consist of a paramagnetic quadrupole doublet at higher temperatures and four magnetically split lines below the Néel temperature due to its antiferromagnetic behavior. In the present spectral analyses, we decomposed the spectrum into two pairs of doublets. Each doublet (we call it a *component*) has 6 parameters in total, that is, Center Shift (C.S.), Splitting (distance between the two lines in each doublet), Linewidth (w_+), Area (A), Area Ratio (A_-/A_+), and Linewidth Ratio (w_-/w_+). The components are defined as follows; for example, component-1 consists of (+3/2, +1/2) and (−3/2, −1/2) transitions while (+3/2, +1/2) and (−3/2, −1/2) transition are designated to transition 1 and 4, respectively. The component-2, consisting of (+1/2, +1/2) and (−1/2, −1/2) transitions, that is $\Delta m = 0$ transition, vanishes in the present geometry.

As we have already experienced in our previous work, we encountered a curious behavior of the parameters obtained from an analysis under the condition that all parameters were allowed to be free except that Area Ratio (A_-/A_+) and Linewidth Ratio (w_-/w_+) are fixed to be equal to 1 ('*equal intensity and equal width*'). So, in the next step, we have fixed some parameters in such a way that the center-shift value is set to be the expected value from the extrapolation of the higher temperature region and the values of splitting are nearly equal to the expected ones from the shape of the temperature dependence for the hyperfine field (B_{hf}). From the theoretical considerations, some relations between each splitting of two pairs of doublets shown in Fig. 2 were taken into account when some parameters were fixed in the analyses. In this way we could get a set of parameters for the final fitted spectrum.

In Fig. 1 are shown Mössbauer spectra taken with a $FePSe_3$ single crystal absorber at various temperatures. It should be noted that there coexist paramagnetic and

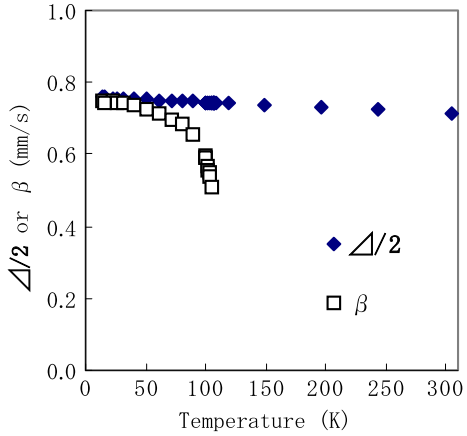

Fig. 3 Temperature dependence of magnetic hyperfine field and one half of the quadrupole splitting

antiferromagnetic phases around the temperature of the phase transition (around 106 K), as shown in Fig. 1b.

As is easily seen from Fig. 2, at the crossing temperature, β should be equal to one half of the quadrupole splitting ($\Delta/2$). As shown in Fig. 3, from the analysis of a series of Mössbauer spectra taken at various temperatures, the values of $\Delta/2$ closely approach β values around 30 K, and then we determine that below 30 K the states $|I = 3/2,\ m = -3/2\rangle$ and $|I = 3/2,\ m = 1/2\rangle$ cross and are mixed, and two lines, 4(−1/2, −3/2) and 5(−1/2, 1/2), merge into one single line. A typical spectrum at this level-crossing region is depicted in Fig. 1d. In this spectrum an obvious deficit in absorption was observed at colliding peaks around 0 mm/s, and this might be explained as an interference of the two transition amplitudes corresponding to the two lines having the same energy.

We have to rule out other possible causes for this reduction in absorption in order to confirm that we have observed EIT. If two absorption lines overlap and a thick absorber is used, the total intensity of the coinciding lines is in general not necessarily the sum of the intensities of the two partner lines due to saturation effects. This is, however, not the case, as is mentioned below. The single line source, ^{57}CoRh, emitted photons having a well-defined circular polarization, either $\sigma = +1$, or $\sigma = -1$, each occurring statistically with the same probability. In general, each photon can induce different Δmtransitions according to the transformation of the photon wavefunction when rotating from the propagation axis to the axis of the hyperfine fields. In the perpendicular geometry, since each photon, whatever its circular polarization, can induce three possible transitions, $\Delta m = 1$, $\Delta m = 0$ and $\Delta m = -1$, some correction for the effective thickness and the total intensity at the sum peaks would be required. In the parallel geometry, only the transitions $\Delta m = \sigma$ are possible and hence, for a particular photon, only one transition can be induced, either $\Delta m = 1$ or $\Delta m = -1$, depending statistically on the circular polarization. Therefore, the effective thicknesses of such a transition and its doublet partner are exactly equal, and the intensity of the sum peak is consequently expected to be the sum of the intensities of the two partners. Therefore, we conclude that the observed deficit in absorption is due to coherence and interference effects.

References

1. Tittonen, I., et al.: Phys. Rev. B **47**, 7840 (1993)
2. Vagizov, F.G.: Hyperfine Interact. **61**, 1359 (1990)
3. Lippmaa, M., et al.: Phys. Rev. B **52**, 10268 (1995)
4. Shvydko, Y.V., et al.: Phys. Rev. Lett. **77**, 3232 (1996)
5. Shvydko, Y.V., et al.: Phys. Rev. Lett. **52**, R711 (1995)
6. Olariu, S., et al.: Europhys. Lett. **37**, 177 (1997)
7. Olariu, S., et al.: Phys. Rev. B **50**, 616 (1994)
8. Kocharovskaya, O., et al.: Phys. Rev. Lett. **82**, 3593 (1999)
9. Coussement, R., et al.: Phys. Rev. Lett. **71**, 1824 (1993)
10. Kocharovskaya, O., et al.: Laser Phys. **9**, 745 (1999)
11. Kolesov, R., et al.: Opt. Commun. **179**, 537 (2000)
12. Shakhmuratov, R., et al.: Opt. Commun. **179**, 525 (2000)
13. Coussement, R., Put, P., Schevenneels, G., Hardeman, F.: Hyperfine Interact. **23**, 273 (1985)
14. Put, P., Coussement, R., Schevenneels, G., Hardeman, F., Berkes, I., Hlimi, B., Marest, G., Sau J., Sayouty, E.H.: Phys. Lett. A **103**, 151 (1984)
15. Coussement, R., et al.: Phys. Rev. Lett. **89**(10), 107601 (2002)
16. Odeurs, J., et al.: Hyperfine Interact. **143**, 97 (2002)
17. Gheysen, S., et al.: J. Mod. Opt. **51**, 2589 (2004)